NEUTRON ACTIVATION ANALYSIS OF ARCHAEOLOGICAL MATERIALS: AN OVERVIEW OF THE ITN NAA LABORATORY, PORTUGAL

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Instrumental neutron activation analysis (INAA) has been applied to archaeological samples at the Portuguese Research Reactor (RPI) since 1973, and the number of samples analysed has increased steadily since the mid-1990s. Archaeometric applications of INAA at RPI include the analysis of archaeological ceramics and the stones used in monumental architecture (menhirs) and other historical monuments. A programme of analysis also exists for measuring the precision of and contributing to the certified values of geochemical reference samples. In addition, the study of interference factors is performed to obtain better accuracy in the determination of concentrations of some elements (e.g., corrections for spectral interferences from uranium fission products in the determination of barium, rare earth elements and zirconium).

KEYWORDS: INAA, PORTUGUESE RESEARCH REACTOR (ITN), NEUTRON ACTIVATION ANALYSIS, ARCHAEOMETRY, ARCHAEOLOGICAL CERAMICS, PORTUGAL

INTRODUCTION

The Junta de Energia Nuclear (JEN) was established in 1954, and in 1961, the JEN's Sacavém Laboratory was equipped with the fifteenth nuclear reactor built abroad by the United States of America under the 'atoms for peace' programme. The Portuguese research reactor (RPI) at Instituto Tecnológico e Nuclear (ITN) is an open-core pool-type reactor, with a maximum power of 1 MWth. The reactor uses MTR type fuel elements with U–Al alloy, with water as a moderator and for cooling. The RPI is used for chemical characterization of samples of different matrices, primarily in support of geosciences and environmental research. Neutron activation analysis (NAA) has proven to be the most profitable analytical procedure offered by the RPI, and several variants of this procedure are used by the Sacavém laboratory: (1) instrumental neutron activation analysis (INAA) by comparison with multi-elemental patterns (since the mid-1970s); (2) INAA by the K0 factor method, by comparison with a reference monitor (gold) (since the mid-1980s); (3) cyclic activation (CINAA) for short life nuclide analyses, by comparison with mono-elemental patterns (since the mid-1980s); and (4) prompt gamma NAA (since the early 1990s). To date, more than 3000 ceramics, 600 samples of related raw materials (e.g., clays etc.) and 250 stones from prehistoric (menhirs) and historical (churches) monuments and related outcrops (trachytes and granites) have been analysed at ITN since the late 1970s, with increasing usage during the 1990s.

The first NAA experiments at RPI were conducted in 1973 by M. Ângela Gouveia and J. Peixoto Cabral, who developed NAA experiments at RPI and adapted existing software packages, specifically the Gelian and Olive programs (Op De Beek 1972, 1974), to facilitate comparison of NAA results. In the late 1970s, the NAA laboratory at Sacavém began operating

at full capacity. Research conducted during that decade included: (1) the evaluation of the quality of results obtained by chemical analyses through periodic analyses of international reference samples; (2) the application of NAA to samples of different matrices, primarily soils, sediments, rocks, waters, biological samples, aerosols and inorganic compounds; and (3) the application of NAA in several disciplines, including cultural heritage, archaeometry, surface geochemistry and environmental studies. The archaeometry research group at ITN currently is known as the 'Património Cultural e Ciências' (Cultural Heritage and Science Group—CHS); the primary researchers are M. I. Prudêncio and M. I. Dias.

NAA AND ARCHAEOMETRY AT THE SACAVÉM LABORATORY

In the late 1970s, the first results from the laboratory were presented at Portugal's National Physics Conference, using data generated from INAA of ceramic materials (Cabral and Gouveia 1978), by researchers within the 'Métodos Nucleares de Análise' (Nuclear Methods of Analysis) group. The first publications regarding the application of INAA to archaeological materials occurred a few years later (Cabral and Gouveia 1979). In the same year, other works regarding INAA laboratory data and performance also were published (Cabral *et al*. 1979). Since 1970, INAA has been applied to numerous studies of archaeological materials, primarily pottery and raw materials (e.g., Cabral *et al*. 1983, 1988, 1993–4, 1996, 2002; Cabral and Gouveia 1984; Prudêncio 1987; Prudêncio and Cabral 1988; Prudêncio *et al*. 1988, 1989, 2003, 2005; Oliveira *et al*. 1997, 2005; Dias *et al*. 2000a,b, 2001a–c, 2002, 2003a–d, 2005a–d; Prudêncio and Dias 2001; Jorge *et al*. 2005; Waerenborgh *et al*. 2005).

The primary standards used for analyses of ceramics and raw materials during the 1970s and 1980s were those issued by the United States Geological Survey (USGS). Recent analyses have made use of standards from the Institute of Geophysical and Geochemical Prospecting, People's Republic of China (IGGE). Research of interference factors has also been performed to better determine the concentrations of additional elements (Gouveia *et al*. 1987; Martinho *et al*. 1991).

In 1992 our group was renamed 'Química Aplicada às Ciências da Terra e Ambiente' (Chemistry Applied to Earth Sciences and Environment). Researchers dealing with INAA as applied to cultural heritage questions were M. A. Gouveia and M. I. Prudêncio. Prior to this time, most of the archaeometric research at the Sacavém laboratory was devoted to the analysis of ceramics and geological materials (e.g., clays, weathering of rocks etc.). In the latter case, most research was performed with the goal of establishing local and/or regional geochemical backgrounds that would help to define raw material fingerprints for comparison with archaeological ceramics.

Research conducted in the early 1990s included studies of the variation of major and trace elements in stones from prehistoric and historical monuments. Studies included analysis of monuments, raw material outcrops, potential quarries and biological colonization (e.g., lichen and moss) in order to determine the provenance of the stones and to evaluate weathering mechanisms to aid in conservation (Gouveia *et al*. 1993; Prudêncio *et al*. 1995, 1998, 2000; Delgado *et al*. 1996; Figueiredo *et al*. 1996; Prudêncio 1997, 2003; Nasraoui *et al*. 1999, 2001a,b, 2002; Romão *et al*. 2000; Sequeira Braga *et al*. 2000). In all of these archaeometric studies INAA was the primary analytical technique, but it was often complemented by other analytical methods (e.g., Mössbauer spectroscopy, XRF, DRX, PIXE etc.).

In the late 1990s, research groups at the ITN chemistry department were reorganized, and a new group devoted to archaeometry was formed—'Património Cultural e Ciências' (Cultural

Heritage and Science Group—CHS). M. I. Prudêncio served as the head of this group, and M. A. Gouveia and M. I. Dias served as primary researchers. During this time, technicians, students (undergraduate, M.Sc. and Ph.D.) and postdoctoral appointees also contributed significantly to the group's archaeometric research.

In 1999 a collaboration was initiated between ITN and the Portuguese Archaeological Institute (IPA), with yearly project submissions devoted to chemical characterization of ceramic artefacts (by INAA), metal artefacts (by XRF), and the dating of organic and inorganic archaeological contexts (by radiocarbon and luminescence respectively). The genesis of the CHS group, the ITN–IPA collaboration and other interdisciplinary projects have resulted in a huge boost to the application of nuclear analytical techniques for studies of cultural heritage.

Establishment of the luminescence laboratory has further encouraged the use of INAA of archaeological materials. The use of TL–OSL (thermoluminescence–optically stimulated luminescence) to date ceramic artefacts, as well as sediments of archaeological contexts, has occurred since 2002 (Richter *et al*. 2003). Consideration of the concentration of naturally radioactive elements is necessary in the TL–OSL age calculation, and these concentrations are determined by INAA.

AN EXAMPLE OF ARCHAEOLOGICAL RESEARCH INVOLVING NAA AT ITN

Chemical approaches to archaeological materials characterization are used to address questions posed by archaeologists that deal essentially with provenance, technology and use. The foremost objective is to aid archaeologists in understanding the characterisation of material culture, technologies, and the organisation of artefact production. Additionally, such studies can be used to understand resource exploitation strategies and regional and interregional interaction patterns, from chronological and diachronic perspectives. Below, we present a case study of the application of INAA to Roman period ceramics, and we show how such analyses can aid in accomplishing the above-mentioned archaeological objectives.

Analytical methods

Ceramics from Roman kilns were analysed by INAA. Standard reference materials GSD-9 and GSS-1 were used to calibrate data generated for this project. Reference values were obtained from data tabulated by Govindaraju (1994). Aliquots of approximately 1 g each of powdered ceramics and reference materials were dried in an oven at 110°C for 24 hours and stored in a desiccator. Once dried, 200–300 mg of powder was measured into cleaned high-density polyethylene vials.

All samples were irradiated at RPI, Sacavém. Samples and standards were bundled together and irradiated at a thermal flux of 3.34×10^{12} n cm^{-2} s^{-1}, $\Phi_{epi}/\Phi_{th} = 1.4\%$, $\Phi_{th}/\Phi_{fast} = 12.1$, for 7 hours. The bundles were rotated continuously during irradiation to ensure that all samples received the same neutron exposure. Iron (Fe) flux monitors were irradiated with the samples to allow corrections due to variation in neutron flux. The long irradiation permitted quantification of the following elements: Na, K, Fe, Sc, Cr, Co, Zn, Ga, As, Br, Rb, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Th and U. Short irradiations were also performed to generate data for Mn and Dy, using a 2-minute irradiation at a flux of 4.4×10^{12} n cm^{-2} s^{-1}. Corrections were carried out for spectral interference from uranium fission products in the determination of barium, REEs and zirconium (Gouveia *et al*. 1987; Martinho *et al*. 1991). In general, precision was less than 5%.

Data were manipulated using a variety of statistical methods including Statistica (StatSoft, Inc. 2003), to identify distinct chemical groupings of ceramics. Cluster analysis was done using the absolute concentration of the chemical elements as variables. Specifically, the clustering method using a linkage based on unweighted pair-group average (UPGMA) and, as a distance measure, the similarity coefficients were calculated using the Euclidean mean to identify outliers and the Pearson correlation coefficient to evaluate correlation between samples. Discriminant, factor, principal component and classification analyses were also performed on the data.

Roman ceramics: Tagus and Sado kilns

Several Roman ceramic production centres have been identified in the Tagus and Sado basins of Portugal. The main sites identified for the Tagus basin are Muge, Garrocheira, Porto dos Cacos and Quinta do Rouxinol, and for the Sado basin, Barrosinha, Bugio, Abul, Herdade do Pinheiro and Zambujalinho. Amphorae produced in Porto dos Cacos, Quinta do Rouxinol and Herdade do Pinheiro were analysed and compared with amphorae data from Correeiros, a consumption centre in Lisbon (Fig. 1). In total, 355 ceramic samples were analysed.

Archaeological work at Porto dos Cacos (PC) revealed the site to be a true industrial complex, specializing in the manufacture of domestic pottery and amphorae used to store salted-fish products (Amphorae Classes 20/21 = Dressel 14, 22 = Almagro 50 and 23 = Almagro 51c) and probably for wine (Lusitana 3 and Lusitana 9). Apart from this formal diversity, production apparently spanned a long period of time, beginning in the middle of the first century AD and continuing up until the fifth century AD (Raposo 1990; Raposo and Duarte 1996).

At Quinta do Rouxinol (QR) archaeological work revealed two kilns and the possible remains of a third (Duarte 1990). QR produced amphorae of Classes 22 = Almagro 50 and 23 = Almagro 51c and Lusitana 9 and a diversity of domestic pottery between the middle of the second century AD and at least the end of the fourth century AD.

At Herdade do Pinheiro (HP) eight kilns were identified, with different shapes and diverse ceramic productions between the first and fifth centuries AD. In the first phase great circular kilns occurred, with the production of Classes 20/21 = Dressel 14 and common ware, in addition to square kilns for production of building materials. The second phase, from the second half of the third century AD until the beginning of the fifth century AD, produced mainly Class 23 = Almagro 51c (Mayet and Silva 1998).

Archaeological investigations at Correeiros (CR) revealed an urban riverside settlement with strong industrial, handicraft and trading characteristics (Bugalhão 2001). At the time of the Roman Empire (between the first and fifth centuries AD), most of the study area was occupied by fish-salting industries and supporting facilities. Several amphorae samples were collected from CR, mainly Classes 22 = Almagro 50 and 23 = Almagro 51c and form Lusitana 9, although Classes 20/21 = Dressel 14 and Lusitana 3 were also present.

A chemical study to differentiate amphorae found in QR according to its various typologies was attempted (Cabral *et al.* 1993–4), but no differentiation was established. Later studies (Dias *et al.* 2001c) were able to identify two main raw material sources for the production of Class 22 = Almagro 50, Class 23 = Almagro 51c and Lusitana 9. Chemical differentiations between amphorae typologies of PC were also established (Cabral *et al.* 1996). Additionally, the analysis identified chemical differences between amphorae from PC and QR (Cabral *et al.* 1993–4; Raposo *et al.* 2004).
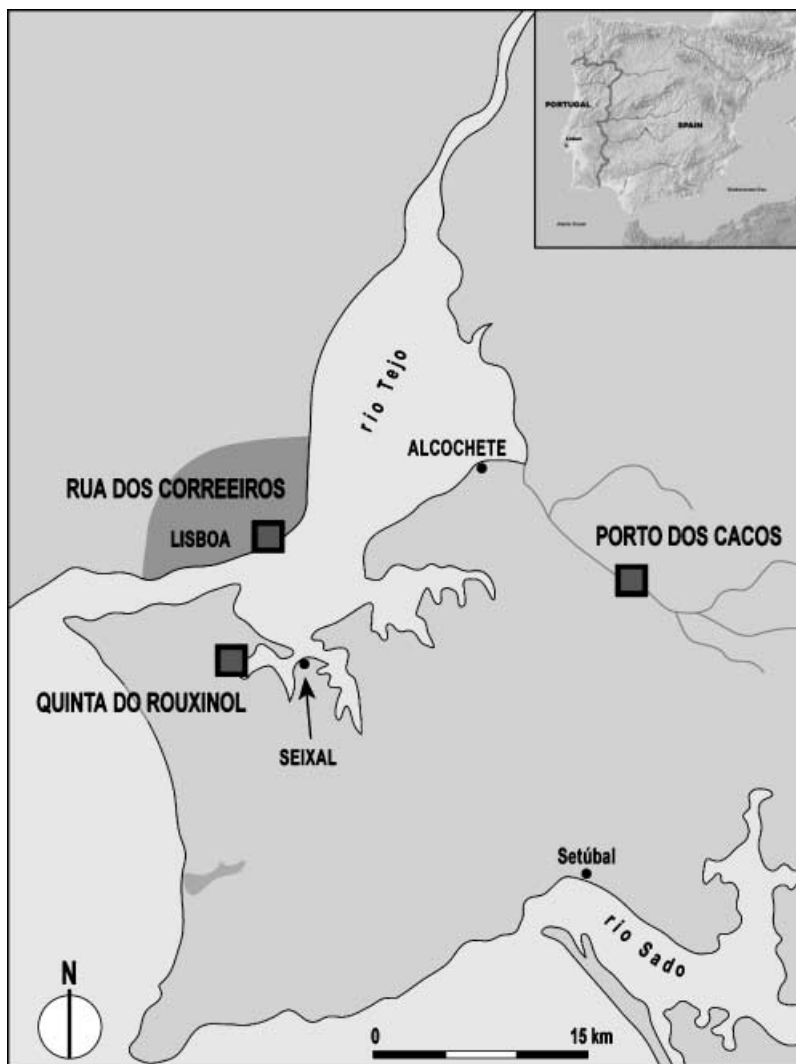
Figure 1 *Roman amphorae from the Tagus production centres.*

Production centres using clay materials of similar geological environments, such as the Tagus and Sado basins, are not easily distinguishable, and mineralogical and textural approaches are not useful for differentiating different production sites (Mayet *et al*. 1996).

A preliminary chemical differentiation based on cluster and factorial analyses found difficulty in distinguishing ceramics from Roman archaeological sites in the Tagus and Sado basins, due to widespread compositional similarities within these regions. Significant compositional differences were identified within the ceramic assemblages of these sites, which might be explained by the exploitation of a wider extraction area or by the addition of varying amounts of temper. Discriminant function analysis and relations between chemical elements were useful tools in that differentiation, and pointed to an important distinction between the
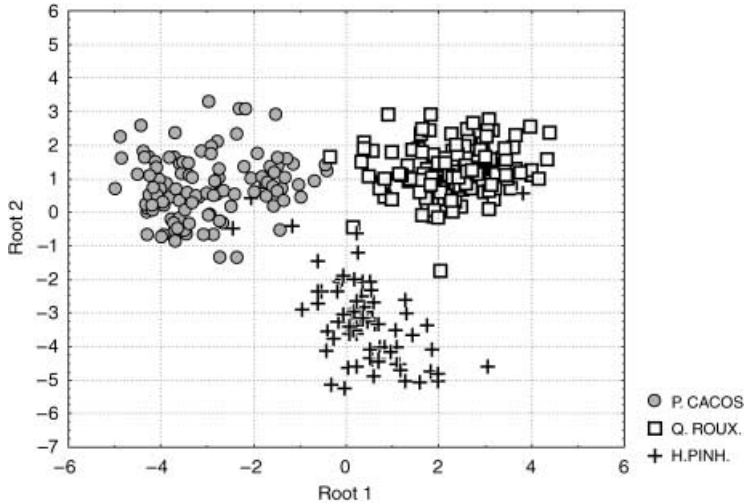
Figure 2   *The identification of multiple groups using discriminant analysis.*

Table 1   *The classification matrix*

Group	Percentage correct	PC p = 0.35161	QR p = 0.41613	HP p = 0.23226
PC	100	109	0	0
QR	98.45	0	127	2
HP	93.06	3	2	67
Total	97.74	112	129	69

studied sites, by the use of different clay raw materials, between and within the two basins (Tagus and Sado) (Dias *et al*. 2003b; Prudêncio *et al*. 2003).

The identification of multiple groups using discriminant analysis followed by a canonical correlation analysis made it possible to visualize how those functions discriminate between the several production centres studied (Fig. 2). Both functions discriminate between groups. The classification matrix (Table 1) shows the number of correctly and incorrectly classified samples: 98% of all cases are classified correctly in this study; 100% of PC amphorae are correctly classified and they are completely separate from the others; 98% of QR amphorae are also correctly classified, and 93% of HP amphorae are correctly classified. For QR and HP, there are a few ceramics that point to a different provenance. When the Mahalanobis distance was computed, all of the PC ceramics were correctly classified, while five HP ceramics and two QR amphorae were considered misclassified. Computing the posterior probabilities, in which a case will be classified into the group for which it has the highest posterior classification probability, again those misclassified cases are mentioned, as well as their predicted classification. So, for the QR cases, both were reclassified as HP amphorae; for HP, two cases were reclassified as QR production, and three HP as PC production. This enhances the difficulty in differentiating ceramic production centres within basins and the exchange of pottery.
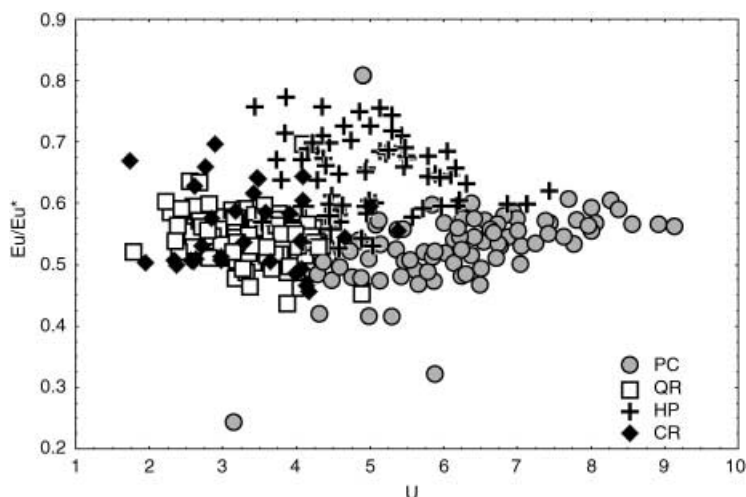
Figure 3   *A plot of Eu/Eu* for the Tagus (PC, QR) and Sado (HP) basins. Roman production centres and for the Lisbon consumption centre (CR).*

The use of geochemical fingerprints became very important for a clear distinction within and between production centres, enhancing which elements have more weight to the established differentiation. A plot of Eu/Eu* versus U differentiates the three production centres in the Tagus and Sado basins (Fig. 3). The U versus Fe and U versus Fe/Sc plots are the best chemical elements that discriminate QR pottery from pottery produced at other sites.

In summary, the chemical compositions that contribute best for differentiating the pottery discussed above are as follows:

• HP amphorae are recognized by lower concentrations of Na, with increased concentrations of Cs and heavy REEs.

• PC amphorae contain the highest concentrations of Na, Co, As and U.

• QR amphorae contain the lowest concentrations of U, and the highest concentrations of Fe.

The results suggest that there are important differences between ceramic samples, consisting of variation in the concentrations of a few elements, with REE concentrations differentiating ceramics between basins, and concentrations of U differentiating ceramics within the Tagus basin.

Preliminary comparison of the amphorae from all production centres with the amphorae from the CR consumption centre suggests a good similarity with QR. This similarity is based on concentrations of U, Eu and Th. As noted above, these elements were considered the best fingerprints for QR production differentiation.

In conclusion, one can infer that, in the case of the Tagus and Sado basins, the concentrations of REE, Eu, U, Th, Sc and Fe, among the studied elements, were found to be the best fingerprints to differentiate the three production sites. These same elements also helped to establish the provenance of the CR amphorae—the QR production centre (Fig. 3).

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