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Comparison of INAA elemental composition data between Lago Grande and Osvaldo archaeological sites in the central Amazon: a first perspective

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Abstract In this work, 50 ceramic fragments from the Lago Grande and 30 from the Osvaldo archaeological site were compared to assess elemental similarities. The aim is to perform a preliminary comparison between the sites, which are located in the central Amazon, Brazil. The analytical technique employed to obtain the ceramics elemental composition was instrumental neutron activation analysis (INAA). The data set obtained was explored by the multivariate statistical techniques of cluster, principal component and discriminant analysis. The analyzed elements were: Na, Lu, U, Yb, La, Th, Cr, Cs, Sc, Fe, Eu, Ce and Hf. The results showed the existence of at least two compositional groups for Lago Grande and Osvaldo. Each compositional group of Osvaldo archaeological site matches with one group of Lago Grande. Correlated with the archaeological background, the results suggest commercial or cultural exchange in the region, which is an indicative of socio-cultural interactions between those sites.

Keywords INAA · Ceramics · Central Amazon · Multivariate analysis

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Introduction

Instrumental neutron activation analysis (INAA) has been widely used in the elemental characterization of ceramic material in archaeometric studies [1, 2]. By using some physical analytical techniques like INAA in the studies of ceramics, it is possible to interpret and associate its life cycle to the behavior of the people involved, assessing patterns of exchange and trade [3].

In this work, the Lago Grande and Osvaldo archaeological sites were chosen to address some archaeometric and archaeological questions due to the intensive excavation work performed there and because they represent a microcosm of the central Amazon region [4]. One of the important aspects related to the study of sites like Lago Grande and Osvaldo is that they show evidence of large human settlements in areas outside *várzeas* (low and flat land alongside a watercourse). It raised the question whether the higher soil productivity of *várzea* sites was really a necessary condition for sedentarism and populational growth [4], as believed in the traditional model. In this manner, the study of those sites is inserted in the wider frame of the Amazon occupation issue, especially in its central region. Details about the traditional model for the Amazon pre-colonial occupation and some critics may be seen in the literature [5–9].

Ceramics have been classified in groups (phases) in Central Amazon since 1968, when the German archaeologist Peter Hilbert [11], using the method of seriation, divided the ceramic material in three different phases, establishing a chronology for the cultural groups that habited that place before the European contact. Prior studies, related with the Central Amazon Project (CAP), compiled a final chronology for the region with new datings (Table 1).

Table 1 Ceramic chronology for the central Amazon based on ^{14}C dating

Ceramic phase	Period (century)
Açutuba	III BC–III AD
Manacapuru	VI AD–X AD
Paredão	VIII AD–XI AD
Guarita	IX AD–XVI AD

Source [10, 11]

Those ceramic phases were established based on the decoration; form of the vessel, and mostly by the kind of temper used in the paste. The two phases discussed in this work, Manacapuru and Paredão, belongs to an early macro-ceramic complex called Incised Rim tradition, which are characterized by the use of *cauixi´* (a river sponge) as temper, and has a geographical distribution that extends from de Orinoco Basin until the Upper Xingu, in Brazil. They also have other shared general characteristics such as large rims; the presence of plastic decorations (incisions), normally on the rim surfaces; presence of red engobe; among others. Furthermore, some ceramic fragments in those two sites could not be associated to both Manacapuru nor Paredão phases and a new ceramic phase denominated *Açutuba* was suggested by Lima, Neves and Petersen [10].

The sites are located in the confluence region of the Negro and Solimões rivers in the central Amazon, Brazil (Fig. 1). The confluence region is formed by hills and mounts, crossed seasonally by *igarapés* (narrow water channels). The Amazon forest is the original vegetation of the region.

The Lago Grande site is located in one of the spits that extend into the Grande Lake. It was chosen for intensive excavation because it presents high archaeological material density associated with dark earth and well-preserved structures, such as defensive trenches in strategic locations. The ceramic material was found up to 1 m deep [8]. The majority of the material belongs to the Paredão phase, but ceramics from the Manacapuru phase, related to the same tradition, characteristic of other archaeological sites as Osvaldo, were also found in lower proportions. It may be an indicative of socio-economical and cultural exchanges between the groups from the central Amazon region.

The Osvaldo site is a spit of 700 × 250 m, limited by the Limão Lake north and by *igarapés* east and west. Archaeological remains were found up to 1 m deep and in association with dark earths. The ceramics belongs to the Manacapuru phase in its majority. Nevertheless, it presents Paredão ceramic fragments in lower proportions, and it corroborates the social-economical and/or cultural exchange assumption, as in the case of Lago Grande archaeological site. It is believed to present a single component context, characteristic of a unique occupation. This characteristic turns the Osvaldo archaeological site important to study the settlements and the use of space [11].

The confirmation of interactions between Lago Grande and Osvaldo archaeological sites would strengthen the hypothesis related to the existence of regional networks in

Fig. 1 Lago Grande and Osvaldo archaeological sites in the Negro and Solimões rivers confluence region. *Source* Ezilon maps and [4]

the pre-colonial Amazon, which could amplify the possibilities in the use of the space in sites outside *várzeas.* It would justify the population growth and the existence of big settlements mentioned in the historical record of the first European travelers in that region [4–7].

The archeological ceramic fragments from Osvaldo and Lago Grande archaeological sites were provided by the Museum of Archaeology and Ethnology of the University of São Paulo (MAE-USP). The artifacts were excavated during the Central Amazon Program (CAP), an effort to understand the pre-colonial occupation of that region. CAP objectives included studying the size and form of the settlements, identify the density and duration of occupations and the refinement of the confluence region chronology [11].

Experimental

Eighty ceramic fragments (50 from Lago Grande and 30 from Osvaldo) were chosen for elemental analysis. The samples from Lago Grande were excavated in a unit which the archaeologists believe to be associated to a former garbage area with high density of archaeological material [8]. For this reason, it is considered a convenient area to represent the archaeological record of Lago Grande. Furthermore, all the stratigraphic levels are represented. The samples from Osvaldo represent the stratigraphy from the surface down to the levels associated with the highest density of ceramic materials (around 0–40 m) and the presence of anthropogenic soils.

The ceramic fragments selected were analyzed by INAA in order to obtain their elemental composition. The external surface was removed with a fine bristle brush. After this procedure, holes were made on the samples with a tungsten carbide rotary file, attached to a variable speed drill. Around 500 mg of powdered sample was obtained from three to eight holes on the side surface of the ceramic fragment, preventing the drill from crossing over the walls. As the ceramic walls may have surface treatment and decoration, drilling the side surfaces prevent their destruction for future analysis by archaeologists. This powder is then collected, dried for 24 h in an oven at 104 °C and stored in desiccators [12], until it is cold enough for weighing.

Around 120 mg of each sample were weighed in polyethylene involucres and sealed with sealing iron. Each involucre was wrapped in aluminum foil. Groups of up to seven ceramic powdered samples and two reference materials were wrapped in another aluminum foil, in order to group them in a parallel fashion to receive approximately the same neutron flux inside the nuclear reactor. The Standard Reference Material-NIST-SRM 1633b was used as standard for analysis and IAEA Soil 7 for the analytical quality control. The samples were irradiated in the swimming pool research reactor IEA-R1 of the Nuclear and Energy Research Institute (IPEN-CNEN/SP), at a thermal neutron flux of 5×10^{12} n cm^2 s^{-1} for 1 h, or 1.4×10^{12} n cm^2 s^{-1} for 8 h.

The gamma-ray spectrometry was carried out with a hyperpure germanium detector (model GX 2519 from Canberra), with a resolution of 1.90 keV at the 1332 keV gamma peak of ^{60}Co. The spectra were collected by a Canberra S-100 MCA with 8,192 channels. The software Genie-2000 NAA Processing Procedure, developed by Canberra, was used to analyze the gamma-ray spectra.

Two measurement series were carried out. The elements As, K, La, Lu, Na, Nd, Sb, Sm, U and Yb were measured after 7 days of decay. The elements Ba, Ce, Co, Cr, Cs, Eu, Fe, Hf, Rb, Sc, Ta, Tb, Th and Zn were measured after 25–30 days of decay [13].

Statistical treatment

Data pre-treatment

As cited in a previous work [14], the elements employed in the analysis of the Lago Grande archaeological ceramics were Na, Lu, U, Yb, La, Th, Cr, Cs, Sc, Fe, Eu, Ce, Hf and Tb, chosen for their analytical precision (<10%), good accuracy assessed by the quality control and negligible contamination during the experimental procedure. For the Osvaldo archaeological site, the same elemental set was used for the same reasons, and to compare its results with Lago Grande ones. The exception is for Tb, excluded in this work, which has almost 30% of missing values.

Regarding the element Lu, there is one missing value and it was chosen the imputation technique of linear correlation with the element Yb, whose regression coefficient was $R^2 = 0.9361$, to estimate its concentration.

It is recommended to rescale the data before applying multivariate techniques, as principal component analysis (PCA) and discriminant analysis (DA). One of the main reasons is that many multivariate techniques rely on the assumption of normality of the data. In archaeometric studies, the elemental concentrations are frequently logarithmically transformed, based on the assumption that the trace elements have a natural lognormal distribution [15–17]. In the present work, the elemental concentrations were rescaled to base-10 logarithms.

Many multivariate algorithms are sensitive to outliers and it is convenient to identify and remove them before analysis [18, 19]. The procedure employed to analyze the existence of outliers was based on the Mahalanobis distance using the Wilks critical value. The quadratic Mahalanobis distance matrix between each sample coordinates vector and the mean vector was calculated by a routine developed in *Scilab* software. The values were compared with the critical value of Wilks [20].

Multivariate statistical analysis

A cluster analysis was carried out to pre-classify the samples. Three methods were employed: Ward's method, average linkage and *k*-means. The software used was *Statistica* version 8.

The Ward's method was employed because it tends to form groups with high internal homogeneity and takes account of the cluster structure [21]. The allocation of one element in a group by Ward's method is performed so that a measure of internal homogeneity is minimized. This measure is based on the partition of the total sum of squares related to a variance analysis [22]. The similarity measure employed was the squared Euclidean distances matrix between the samples.

The average linkage method is another approach used frequently in archaeological pottery studies and also takes the cluster structure into account. The average linkage method uses the average of distances among the elements of each group as the measure of distance between groups [22]. The comparison of the latter two methods of clustering is advised by Baxter [21].

In hierarchical cluster analyses, the allocation of individuals inside groups is not necessarily optimal. When one an object is placed in a group, it remains there until the end of the clustering. The *k*-means method can improve the classification according to the cluster criterion used. The *k*-means is a partition method where the number of groups must be chosen a priori in order to reduce the partition possibilities, making the method more efficient in terms of the computing effort needed. The criterion of the grouping quality used by *k*-means is based on the minimization of the partition of the total sum of squares in a variance analysis.

After the clustering procedure, a PCA was carried out in order to verify the data structure. It was used the covariance matrix instead of the correlation matrix, because with the latter the results are more difficult to interpret [21]. One possible reason for that is the dilution of the differences with respect to those variables that best discriminate the clusters.

A canonical discriminant analysis was carried out in order to assess how reasonable was the classification of objects by the clustering algorithms. The standardized canonical discriminant functions were calculated.

Results

Table 2 presents the arithmetic means for the elemental concentrations for both archaeological sites.

The analysis of the Lago Grande data set has been already performed in a previous work [14], but it was repeated with the extraction of the element Tb, which cannot be used for the data from Osvaldo. It was obtained basically the same results, with two compositional groups identified, represented here in the principal component space (Fig. 2) and in the discriminant graph (represented by groups 1 and 2 in Fig. 4).

Table 2 Elemental description for Lago Grande and Osvaldo archaeological sites

	Lago Grande				Osvaldo			
Element	Minimum	Maximum	Average	Relative SD (%)	Minimum	Maximum	Average	Relative SD (%)
Na (%)	0.01	0.60	0.20	70.13	0.04	0.46	0.18	64.27
Lu	0.30	0.70	0.47	20.99	0.31	0.88	0.50	20.86
U	2.40	8.70	4.00	26.03	1.87	5.79	4.03	19.51
Yb	2.00	4.60	2.99	18.76	2.05	5.90	3.14	22.83
La	24.90	61.00	42.54	18.95	29.93	58.29	41.35	17.70
Th	7.60	18.60	13.76	13.82	10.93	21.40	16.01	17.82
Cr	36.40	87.70	64.40	16.44	43.05	100.27	67.03	18.98
Cs	1.30	16.20	9.25	37.02	3.23	17.62	8.60	38.88
Sc	7.60	19.80	14.70	18.11	10.06	24.89	16.36	21.02
Fe (%)	1.30	4.80	3.46	22.94	2.36	4.86	3.68	16.81
Eu	0.65	2.00	1.28	26.70	0.39	1.85	1.23	28.51
Ce	47.60	119.90	80.61	20.95	29.79	160.74	83.47	34.69
Hf	3.40	17.30	6.93	38.59	2.97	18.70	8.01	39.09
Tb	0.30	1.20	0.77	33.74	–	–	–	–

Units are in ppm, unless indicated

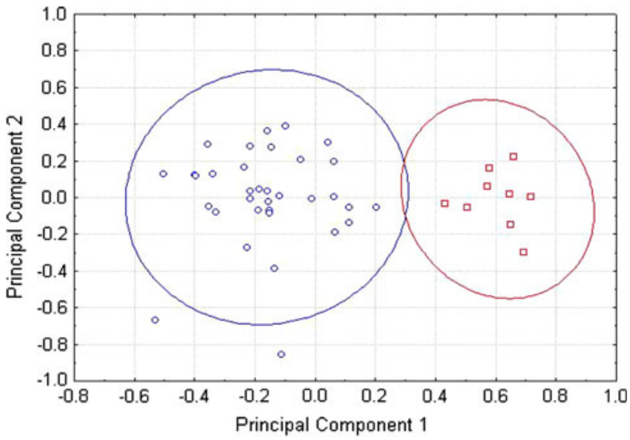

Fig. 2 PCA for ceramic fragments from Lago Grande archaeological site based on [14], without Tb. The ellipses represent the 95% of confidence interval and around 73% of the system variance is represented

It was applied the Wilk's lambda critical value to identify outliers in the data set of Osvaldo archaeological site. No outliers were identified.

Regarding the cluster analysis for Osvaldo data set, the results do not match among the three methods only for two samples. Using the criterion of cutting the dendrogram at the largest variation in the linkage distance, two compositional groups were obtained (represented by groups 3 and 4 in the discriminant analysis of Fig. 4).

The plot of the two first principal components obtained by the PCA of Osvaldo data set is presented in Fig. 3. They represent 69% of the total system variance.

The discriminant analyzes for Lago Grande and Osvaldo archaeological sites are combined in Fig. 4.

The next step was to analyze all the data together in the cluster analysis to assess if there was any difference in the results, when compared to discriminant analysis based on the separate clusters. No changes were observed in the compositional group classification.

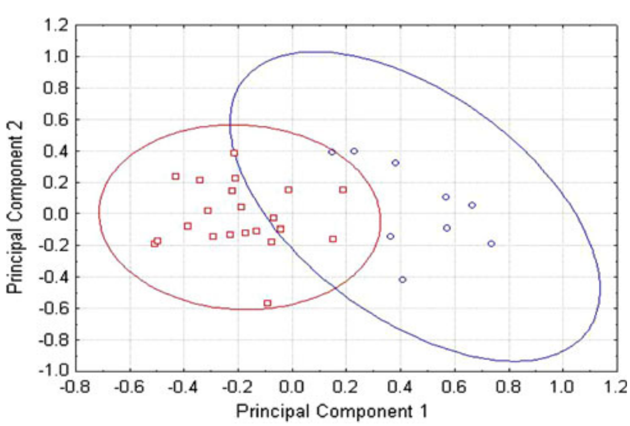

Fig. 3 PCA for Osvaldo archaeological site. The ellipses represent the 95% of confidence interval and around 70% of the system variance is represented

Discussion

The plot of the first two principal components for Lago Grande (Fig. 2) and Osvaldo (Fig. 3) shows that the cluster classification of samples yielded plausible results, with two compositional groups represented for each site.

The discriminant analysis based on the cluster classification of samples (Fig. 4) shows the existence of at least two compositional groups for Lago Grande and Osvaldo archaeological sites, and their match yields two compositional groups in total.

In the Osvaldo ceramic fragments, there are three samples classified as Paredão material (characteristic of Lago Grande) by the archaeologists. Statistically, it was not possible to correlate them to a specific compositional group in the cluster analysis. There are two samples in group 4 and one in group 3 (Fig. 4). For a better characterization, more samples need to be analyzed.

Similarly, for the Lago Grande archaeological site there are some Manacapuru fragments (characteristic from Osvaldo). It is still not possible to establish their distribution without the necessary archaeological analysis.

The match of most of the Osvaldo fragments elemental composition with Lago Grande ones was an unexpected result that needs to be further analyzed, because most of them were classified as Manacapuru ceramics.

According to intrasite and intersite comparison, the fact that there are at least two compositional groups for each site and their match in the discriminant function space

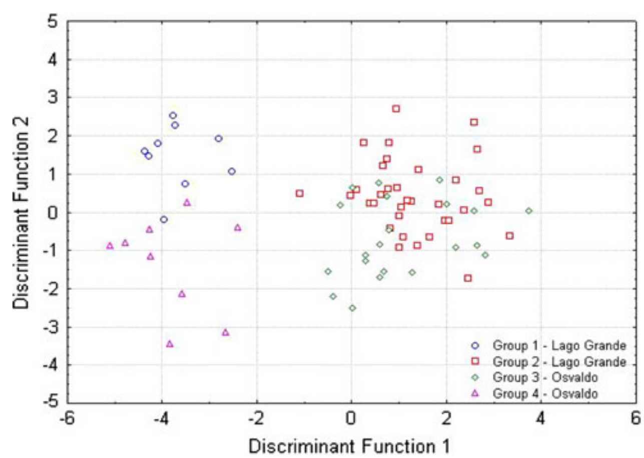

Fig. 4 Canonical discriminant analysis for the Lago Grande and Osvaldo archaeological sites. The group assignment was performed separately for each site for comparison purposes. It was considered the existence of two compositional groups in total: one formed by groups 1 and 4, and the other by groups 2 and 3

(Fig. 4) suggests that the former site inhabitants might have exchanged ceramics in a commercial or cultural relation or that there were two clay sources being used for both groups. These potential scenarios were figured out based on the archaeological background for the central Amazon region [4, 6–8, 11], and must be further explored in the future archaeometric analysis together with the clay samples. The archaeologists suggest [5] that the question regarding the commercial or cultural relation could be a condition of existence of a long-term occupation, as identified in the excavations of Lago Grande archaeological site, together with pre-Columbian forms of agriculture in interfluvial uplands within Amazonia. It contrasts with the short-term occupations documented ethnographically and gives an additional basis to further consider relations between Osvaldo and Lago Grande.

Another evidence of relation between the archaeological sites in this work is the existence of lakes connected by *igarapés* (little water channels) along the Negro and Solimões confluence region (Fig. 1). In the flooding periods, the lakes around which Lago Grande and Osvaldo are located are linked by the *igarapés*, and it gives a transportation means for possible commercial or cultural relations to happen.

The next steps will be to analyze the potential commercial exchange between the sites, to characterize the cultural phases of the Lago Grande ceramic fragments and include the clay sources in the analysis.

Conclusion

The combination of the associated cluster and discriminant analysis proved to yield useful results for the elemental concentration data sets used in this work. Until the present moment, two different compositional groups can be assigned to Lago Grande and Osvaldo archaeological sites, with their match in the discriminant function space yielding two compositional groups in total. It may be an indicative of commercial or cultural exchange, corroborated by the archaeological assumptions for the region in the pre-Columbian times. If demonstrated, it can be considered as an indicative of socio-cultural interactions in the central Amazon region, which may have implications for the archaeological theories regarding the pre-colonial occupation of the central Amazon.

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