

# Study of Exchange Networks between two Amazon Archaeological Sites by INAA

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## Abstract

Intensive archaeological research is trying to answer questions regarding the pre-colonial occupation in the Amazon region. A critical issue is whether environmental constraints would impose limitations for the emergence of social complexity and sedentarism among the Amazon forest Indians. To contribute to this discussion, instrumental neutron activation analysis (INAA) was used to determine the concentration of 24 chemical elements in pottery shards from Lago Grande and Osvaldo archaeological sites, in Central Amazon. The multidimensional data were analyzed by cluster and principal component analysis for grouping samples of similar elemental composition and graphical visualization. The multivariate patterns combined with ceramic phase distribution among the chemical groups were interpreted as correlated to potential exchange networks driven by three mechanisms: trade, exogamic marriage and territorial integration.

**Keywords:** Archaeometry; Amazonian Pottery; Neutron Activation Analysis; Amazonian Archaeology.

## 1. Introduction

One of the main objectives in Archaeology is the study of ancient human groups by their material vestiges. As it is not possible to observe the human behavior of those societies directly, the analysis of those materials vestiges is essential [1]. In many parts of the world, pottery shards are the most common type of artifact due to their high resistance to most physical-chemical soil conditions. Their macroscopic characteristics as decoration, color, form and function make them useful cultural and chronological indicators in Archaeology [2].

On the other hand, the microscopic characteristics of pottery shards, as their elemental chemical composition, can be used to infer their origin and possible trade routes, location of sources of raw materials for the ceramic production, production methods, among other aspects [3].

Trace elements, like the lanthanides and some transition metals, are important in physical-chemical characterization of archaeological ceramics due to their geochemical properties. They are found typically dispersed in nature, and their concentration profiles depends on the igneous rock formation and contribute to differentiate between ceramic materials made of different clays, which are formed by intemperism of the original rocks [4, 5], as expressed in the *provenance postulate* [6]. One of the most employed techniques for elemental characterization of ceramics is neutron activation analysis [7, 8], which is suitable in terms of analytical accuracy and precision for trace elements quantification and needs only  $\approx 100$  mg of sample for adequate analysis. This is

important in research designs in which sample destructiveness is a factor to be minimized.

In this interdisciplinary work, instrumental neutron activation analysis (INAA) was applied for quantitative determination of trace and some micro-elements in ceramic material from Central Amazon, in the confluence region of Negro and Solimões rivers in Brazil. The sites selected for sampling were Lago Grande and Osvaldo, which represent a microcosm of the region that is suitable to test archaeological hypotheses. The material analyzed was excavated under the context of Central Amazon Project (PAC), coordinated by Prof. Eduardo Góes Neves, from the University of São Paulo, in cooperation with the Federal University of Amazon.

## 2. Archaeological Framework

By the end of the 15<sup>th</sup> century, the Amazon basin was densely occupied by different indigenous people. The first European travelers in the region wrote about the existence of large settlements, occupied by thousand of peoples and interconnected by roads in extensive commercial networks and regional political federations [9]. That picture contrasts with the current ethnographic situation, characterized by sparse occupations with low demographic density, in which the activities are organized by age and genre structures.

The perspective of the environmental determinism is that the Amazonian pre-colonial occupations were similar to the current ethnographic picture. In this way, it would not be possible the existence of complex human organizations in the tropical forest, due to natural limitations that would stimulate an intense mobility of its inhabitants [10, 11]. Under this perspective, the high degree of material vestiges in the archaeological record would be the result of multiple reoccupations.

One of the alternative perspectives arguments in favor of large settlements in the pre-colonial period, called *cacicados*, which would be based in a hierarchic organization with a center of power of regional reach. In this way, this critical view tries to diminish the role of environmental difficulties as a limiting factor for the emergence of complex societies, which contrasts with the current ethnographic picture. The decrease and dispersion of populations would be linked the European colonization, which brought sickness, war and slavery to those people [11, 12].

Ceramics have been classified in groups (phases) in Central Amazon since 1968, by German archaeologist Peter Hilbert [13], establishing a chronology for the cultural groups that lived in that place prior to the European contact. The refined chronology based on <sup>14</sup>C dating may be found in Table 1.

Table 1 – Ceramic chronology for the central Amazon based on <sup>14</sup>C dating. Source: [13]

Ceramic Phase	Period (century)
Açutuba	III B.C. – III A.D.
Manacapuru	VI A.D. – X A.D.
Paredão	VIII A.D. – XI A.D.
Guarita	IX A.D. – XVI A.D.

Those ceramic phases were defined based on the decoration, form of the vessel, and mostly on 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, that is characterized by the use of *cauixi* (a river sponge) as temper, and has a geographical distribution that extends from de Orinoco Basin up to

the Upper Xingú, in Brazil. They also have other shared general characteristics such as large rims; the presence of plastic decorations (incisions) frequently on the rim surfaces; and presence of red engobe.

The sites Lago Grande and Osvaldo are located in the confluence region of the Negro and Solimões rivers in the Central Amazon, Brazil (Figure 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.



Figure 1 – Lago Grande and Osvaldo archaeological sites in the Negro and Solimões rivers confluence region. Source: Ezilon maps and [14].

The Lago Grande site has around 500 x 400 m and is located in one of the spits that extends 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 [15]. Most part of the material belongs to Paredão ceramic phase, but Manacapuru ceramics, related to the same tradition and characteristic of other archaeological sites like Osvaldo, were also found in lower proportions.

The Osvaldo site is a spit of 700 x 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. Most of the ceramic material belongs to the Manacapuru phase. Nevertheless, it presents Paredão ceramics in lower proportion. It is believed to present a single component context, characteristic of a unique occupation. It turns Osvaldo important to study settlement patterns and the use of space [13].

Lago Grande and Osvaldo are located around 9 km of each other, which turns them interesting to study potential exchange networks. Previous studies [13-16] identified pottery shards which do not belong the main ceramic phase predominant in both sites. Manacapuru ceramics in Lago Grande and Paredão ceramics in Osvaldo could have an external origin, and be a correlate of an exchange network as they were contemporary for some time. That potential network could be part of a broader context related to a regional system of interaction, which have already been observed in other regions of Amazon, but has not been intensively investigated in its central region. Those

systems would be characterized by common ideology and cultural aspects, extensive exchange networks, exogamic marriages and inter-communitarian alliances.

The study of Lago Grande and Osvaldo is interesting for theories which try to contest the ecological determinism as those sites are located in regions that are not alongside the watercourse of Negro and Solimões rivers, which would be the best region to get natural resources for life. They contain dark earth in the archaeological record, which may be a correlate that the former inhabitants practiced agriculture and fishing. In this way, they would overcome the natural difficulties imposed by the tropical forest environment by a combination of activities to obtain food and other resources [14]. That dynamics could create the conditions for the emergence of social complexity and sedentarism in the region [11, 17, 18].

### **3. Materials and Methods**

The approach adopted in this work comprises the sampling of ceramic material from Lago Grande and Osvaldo archaeological sites, with unknown origin of raw materials. INAA was carried out for the determination of the elemental concentration profiles. Due to the number of variables generated (24 elements), multivariate pattern recognition and reduction techniques were applied for grouping samples with similar elemental composition, seeking for geochemical and archaeological evidences to justify the definition of such chemical groups.

#### **3.1. Instrumental Neutron Activation Analysis (INAA)**

Approximately 200 ceramic fragments were chosen for elemental analysis. The samples from Lago Grande were excavated in three regions with a high density of archaeological vestiges [15], which many archaeologists believe to be associated to reuse of materials for house elevation or funerary purposes. For this reason, it is considered a convenient area to represent the archaeological record of Lago Grande. Furthermore, all the stratigraphic levels were represented. The samples from Osvaldo represent the stratigraphy from the surface down to the levels associated with the highest density of ceramic materials (concentrated in 0-40 cm) and the presence of dark anthropogenic soils, sampled in a large area of the site.

The lateral external surfaces of pottery shards, without decoration, were removed with a tungsten carbide sharpening rotary file. After this procedure, holes were made on the samples with a tungsten carbide cutter rotary file (diameters of 1.5 and 2.2 mm), attached to a variable speed drill. Around 500 mg of powdered sample were obtained from 3 to 8 holes on the side surface of the ceramic fragment, preventing the drill from crossing over the walls. That powder was then collected, dried for 24 hours in an oven at 104 °C and stored in desiccators for posterior weighing [19, 20].

Around 120 mg of each sample were weighed in polyethylene involucre and sealed with sealing iron. Each involucre was wrapped in aluminum foil. Groups of up to 7 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 [21]. The samples were irradiated in the swimming pool research reactor IEA – R1 of the Nuclear and Energy Research Institute, in São Paulo, at a thermal neutron flux of  $1.2\text{--}1.6 \cdot 10^{12} \text{ n.cm}^2.\text{s}^{-1}$  for 8 hours, for most of the samples.

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 1332.5 keV gamma peak of  $^{60}\text{Co}$ . The spectra were collected by a Canberra S-100 MCA plate with 8192 channels. The software Genie-2000 Gamma Acquisition & Analysis, v. 3.1.a, 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 5-7 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.

### 3.2. Multivariate Statistical Analysis

All the concentrations were reported in  $\mu\text{g.g}^{-1}$  prior to any transformation. It is recommended to rescale the data before applying multivariate analysis to compensate differences of scale in the elemental concentrations. It contributes to homogenize the weights of the variables in multivariate analysis [22]. It might also improve the condition of normality, assumed in many multivariate techniques. In archaeometric studies, the elemental concentrations are frequently logarithmically transformed based on the assumption that trace elements have a lognormal distribution in many geochemical contexts [23, 24, 25], although formal tests should be performed prior to further conclusions about data distribution. In this work, the elemental concentrations were rescaled to base-10 logarithms.

Many multivariate methods are sensitive to outliers and it is convenient to identify and remove them before analysis. 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 for identification of multivariate outliers [26].

Cluster analysis (CA) for the log-transformed data was carried out as implemented in the *MURRAP* software, according to Eq. (1). In this hierarchical clustering method, the measure of distance between groups to be minimized is the average of the distances of their elements:

$$d[G_1, G_2] = \sum_{j \in G_1} \sum_{k \in G_2} \frac{d_{jk}^2}{g_1 g_2} \quad (1)$$

where  $g_1$  and  $g_2$  are the number of samples in clusters 1 and 2 and  $d_{jk}^2$  is the Squared-mean Euclidean distance (SMED) between the logarithms of concentrations, given by (adapted from 23):

$$d_{jk}^2 = \frac{1}{\min(p_j, p_k)} \sum_{i=1}^{\min(p_j, p_k)} [\log(C_{ij}) - \log(C_{ik})]^2 \quad (2)$$

where  $p_j$  and  $p_k$  are the number of variables (chemical elements) available for samples  $j$  and  $k$ , and  $C_{ij}$  is the concentration of element  $i$  in the sample  $j$ . The advantage of this metric is that eliminates problems regarding missing values, as it averages over only the elements with concentrations higher than the detection limit for INAA. It prevents from estimation of such values or elimination of samples.

Visual inspection of dendrograms from cluster analysis is a useful method for the identification of preliminary groups, working properly for discrimination of groups

of high hypersphericity and multidimensionally well separated. However, it can fail in data sets with high correlation between variables. To minimize this problem, the groups defined in the clustering procedure were refined using the Mahalanobis distances of samples to the group centroids and associated to F distribution to calculate the pertinence probabilities to each group [23].

The principal component analysis (PCA) was carried out for dimensionality reduction and visualization of the groups defined in CA. This reduction is obtained by the transformation of the original variables in a new set of uncorrelated ones, the principal components, extracted in such way to maximize the system variance explained by the first few components. PCA was carried out with the covariance matrix [27], as the data set was already log-transformed to compensate scale differences.

The softwares used for the statistical treatment were *MURRAP* 8.6, developed at the MURR Archaeometry Laboratory, *R Statistical Package* 2.13.0, and *Scilab* 5.2.2.

#### 4. Results and Discussion

The analytical quality control of the results was carried out by irradiating the standard reference material IAEA–Soil–7 as an unknown sample within the batches of ceramic powder samples. The details can be found in [28]. Fifteen elements were selected according the analytical quality parameters ( $s < 15\%$  and Bias  $< 10\%$  for approved elements and Bias  $< 25\%$  for warning elements): As, K, La, Lu, Na, Yb, Ce, Co, Cr, Cs, Eu, Fe, Hf, Sc and Th. All those elements presented coefficients of variation lower than 15% and only K presented that value higher than 10% (14.4%). Uranium fission by fast neutrons were not necessary for the data set, as 82% of its concentration in pottery shards were lower than  $5 \mu\text{g}\cdot\text{g}^{-1}$ , a value suggested as reference for those corrections [23]. Additionally, interferences due to the plastic involucres could be discarded according to INAA results, since all the elements of interest presented concentrations lower than the detection limit. The element Co was removed from multivariate analysis due to evidences of contamination in the sample preparation using a tungsten carbide rotary file [28, 29].

Among the elements selected, only Na, K and Fe were not measured in concentrations in the order of  $\mu\text{g}\cdot\text{g}^{-1}$  or lower in the ceramic data set. The descriptive parameters of the data set used in this work are presented in Table 2.

By analyzing Table 2, it is possible to observe a general tendency of lower coefficients of variation for the clay sources near Osvaldo site. This is an expected result considering that antiplastic materials (like sand, crushed shards and shells) may be added to the paste in the ceramic production, and even different sources of clay might be selected. In the case of Lago Grande and Osvaldo, archaeological analyses found that the main temper used was cauxi, a fresh water sponge [13]. Crushed shards and *cariapé*, a material extracted from trees, could also be found as secondary sources of antiplastic materials in Paredão and Manacapuru ceramics.

Table 2 – Descriptive parameters for the elemental concentration data set of pottery shards from Lago Grande and Osvaldo in Central Amazon.

Element	n	Lago Grande				Osvaldo				Fired Osvaldo clays			
		Min	Max	Mean	$\sigma$ (%)	Min	Max	Mean	$\sigma$ (%)	Min	Max	Mean	$\sigma$ (%)
As	176	2.4	17.3	8.2	39.6	0.5	18.6	5.7	56.9	5.8	13.6	9.9	24.3
K (mg/g)	142	2.0	30.6	11.2	45.9	3.2	21.9	10.0	41.5	3.7	10.9	6.5	45.8
La	181	23	127	42	27	21	86	42	25	76	92	82	6
Lu (ng/g)	180	151	1303	481	26	284	884	498	21	703	923	810	11
Na (mg/g)	181	0.3	6.3	2.1	71.6	0.3	5.0	1.8	63.6	0.3	0.8	0.5	37.4
Yb	181	1.4	8.1	3.0	25.5	2.1	5.9	3.1	20.2	4.6	5.9	5.1	11.1
Ce	177	42	298	88	38	30	161	82	32	91	161	143	15
Co	177	2	47	10	59	3	121	12	129	2	4	3	30
Cr	177	36.4	108.9	67.7	18.4	33.2	100.3	63.4	18.5	57.9	125.9	97.9	19.8
Cs	175	1.3	19.1	9.3	36.7	3.2	17.6	8.9	34.7	8.4	16.5	10.7	27.5
Eu	176	0.4	3.4	1.3	32.0	0.4	2.8	1.3	30.6	1.8	2.4	2.1	8.4
Fe (mg/g)	177	13.2	60.4	37.9	24.6	23.4	48.6	34.1	18.2	37.8	64.3	49.8	18.9
Hf	177	2.9	17.3	6.8	35.1	3.0	18.8	7.9	39.9	15.9	24.9	21.6	13.2
Sc	177	7.6	26.4	15.6	18.6	9.8	24.9	15.1	20.5	14.5	19.8	17.7	12.4
Th	177	7.6	23.7	14.5	16.6	8.6	24.1	15.0	20.3	18.0	36.2	30.7	18.3

Ratios between concentrations of chemical elements in *cauixi* and pottery shards were calculated combining INAA results. The ratios varied between 3% for sodium and 26% for iron. Iron can be found as a cover of silica spicules mainly as  $\text{Fe}_2\text{O}_3$  of fluvial origin [30].

Approximate estimations of the area fraction of spicules in optical microscopy in relation to the remaining components in the ceramic paste were always below 20%. Assuming proportion in area equal to proportion in volume, a mass density of  $2 \text{ g.cm}^{-3}$  for *cauixi* [31] and  $1.3 \text{ g.cm}^{-3}$  for the pottery shards, the mass fraction of *cauixi* in the samples was estimated as lower than 30% for the 17 samples observed in the optical microscope. Ignoring any volatility of the chemical elements associated to the biogenic silica during the ancient firing by former inhabitants of Lago Grande and Osvaldo, the maximum dilution in the elemental concentration in pottery would be always lower than 22.2% for iron (the least affected element) and lower than 29.1% for sodium (the most affected element).

From the previous estimations, it would be inferred that all the chemical elements concentrations should present patterns of dilution due to the insertion of temper during ceramic manufacture. Nonetheless, it was observed strong patterns of chemical enrichments for the elements sodium ( $\approx 300\%$ ) and potassium ( $\approx 60\%$ ) in the pottery shards, which was considered an indication of diagenetic effects.

The criteria to choose the chemical elements selected for analysis among the total 24 elements analyzed by INAA were acceptance in the quality control analysis, a minimum value for missing values (10%) and inexistence of interferences. In this sense, fourteen elements were selected for CA and PCA: As, K, La, Lu, Na, Yb, Ce, Cr, Cs, Eu, Fe, Hf, Sc and Th. Potassium had around 20% missing values, but it was maintained in the analyzes to investigate evidences of enrichment due to diagenetic effects.

Eleven multivariate outliers were found among 177 analyzed samples with no missing values in the elemental concentrations. The reasons for the existence of outliers

can be improper analytical determination, but it is not the only possible explanation. No clear patterns were identified for the outliers. Problems in the analytical determination for three of the outliers were identified. The remaining eight samples classified as outlier were tagged with a “Warning” and maintained in the multivariate analysis, as they might represent different chemical groups underrepresented in relation to the dominant ones, diagenetic effects or intentional modification in the ceramic production. In the case where they did not yield useful information, they were eliminated.

Cluster analysis for the log-transformed data of the fourteen chemical elements selected was performed in order to identify archaeologically meaningful chemical groups. It was defined that the minimum cut in the dendrogram should differentiate the Osvaldo clay samples, as the univariate inspection has already indicated significant differences in the concentrations of the chemical elements (Table 2). Further cuts in lower linkage distances were analyzed looking for archaeological correlations that would justify the definition of more chemical groups. In the absence of such correlations, the minimum cut was maintained.

In a first approach, the data from the two archaeological sites were analyzed together to assess general patterns and to verify *a posteriori* whether the provenance would be a factor responsible for the separation of chemical groups. This first approach assisted in the identification of diagenetic effects affecting the elements Na, K, Cs, Hf and As. However, no clear evidences of correlation between the chemical groups and archaeological factors as site provenance, ceramic phase, depth or kind of ceramic object were identified.

PCA analysis assisted in the verification of possible diagenetic effects that could be active in a hydrological dynamic region as the Solimões and Negro rivers confluence in Brazil. Sodium was identified as an element strongly altered in the depositional context, dominating the weight coefficients in the first component by an order of magnitude with respect to the majority of the elements. It was assessed that it would lead to a spurious partition of the samples in chemical groups with no archaeological significance. The absence of calcareous sources of clays in the region and lower concentrations of sodium in the cauxi samples ( $\approx 3\%$  of the concentration in pottery) with respect to the pottery shards led to an alternative geochemical hypothesis for the enrichment of sodium in the pottery shards along the depositional time.

There are evidences in the literature that a change in the sodium profile is possible in ceramic materials fired at low temperatures [32-34]. The geochemical hypothesis for the enrichment of sodium in the clay samples would result from the availability of sodium in the confluence region due to the intemperism of feldspars to kaolinite of sediments from Andean and sub-Andean regions, linked to the cycles of sediment deposition on the flooding plains of Negro and Solimões rivers [35, 36]. Rain water could be another important source of sodium in the rivers of the region [37].

In this way, sodium was eliminated from the analyses due to its dominance in PCA and evidences of diagenetic effects that could obscure culturally driven patterns of interest. Arguments similar to the analysis of sodium can be extended for the element potassium, but its enrichment was one order of magnitude lower. Relative dominance of that element in PCA with no clear reasons, associated to geochemical evidences in the literature that a change in its profile is possible in archaeological ceramics [32], also led to the exclusion of potassium from the analyses.

Potential diagenetic effects were also identified for the elements Cs, As and Hf, although in a lower magnitude than the enrichment of sodium and potassium. The evidences for such effects were low correlation coefficients, parameters of dispersion of the variation matrix and geochemical evidence in the literature [33, 38, 39]. After the



exclusion of sodium and potassium, it was verified that hafnium dominated the first and fourth components, responsible for 54% of the system variance. Cesium and arsenium dominated the second and third component, responsible for 33% of the system variance. The change in the chemical groups observed after the extraction of those three elements in the PCA might indicate geochemical patterns affected by post-depositional effects that could obscure archaeological and cultural correlation of INAA results.

After the first approach analyzing all the data combined in the PCA, the chemical elements selected for the exploration of chemical groups of archaeological relevance were La, Lu, Yb, Ce, Cr, Eu, Fe, Sc and Th.

The data from Lago Grande and Osvaldo were analyzed separately in a second approach. Samples which were the last to cluster to the mass of the data, evident outliers in the clustering procedure, were removed in order to increase the resolution of the dendrograms in terms of linkage distance. For the  $\log_{10}$ -transformed data, two chemical groups were identified for each archaeological site. The largest group (A) for Lago Grande contained 68 samples and the smallest one (B) contained 36 samples. The largest group (A) for Osvaldo contained 38 samples and the smallest one (B) contained 17 samples.

Following a first visualization of results, a refinement of the cluster classification was undertaken in order to take the correlation structure of data into account. The reasons to exclude samples were location outside the 95% confidence region of ellipses, pertinence probability lower than 1% to any of the groups [4], and samples without analytical traceability associated to outlying values of standard reference material IAEA–Soil–7. The projection of the refined classification of samples on the two first principal components is showed in Fig. 2.

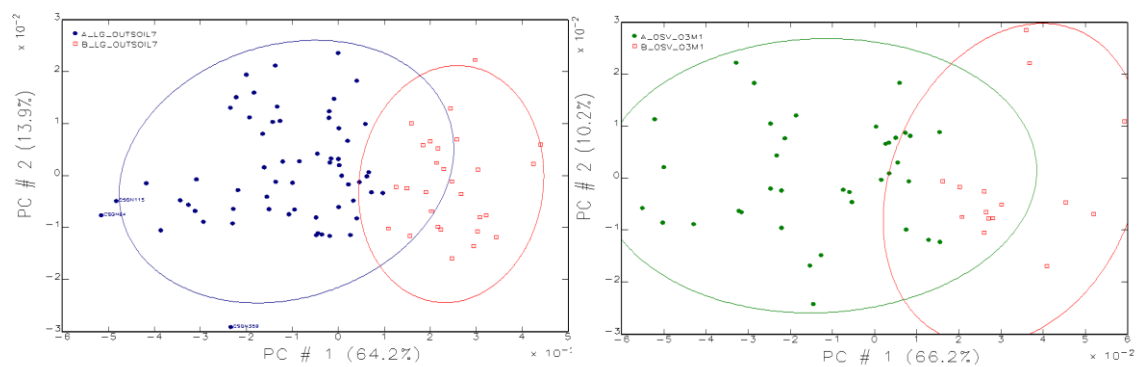


Figure 2 – PCA scores of elemental concentration data from Lago Grande (left) and Osvaldo (right) archaeological pottery after the refinement of the classification in cluster analysis. Ellipses represent the 95% confidence region.

The two groups defined for each site presented a separation according to what is expected from physical-chemical analyzes of pottery from similar geological environment, in intraregional context, with some degree of superposition [32].

Table 3 shows means and standard deviations for the chemical groups defined for Lago Grande and Osvaldo by CA and PCA. Fig. 3 shows the projection of data on the two first principal components of the refined data for both sites combined.

Table 3 – Mean concentrations of chemical elements in the chemical groups of pottery defined for Lago Grande and Osvaldo archaeological sites. Concentrations are in  $\mu\text{g}\cdot\text{g}^{-1}$ .

Element	Lago Grande		Oswaldo	
	A (n=61)	B (n=28)	A (n=37)	B (n=15)
Sc	14.8 ± 1.9	18.6 ± 2.1	13.8 ± 2.0	18.7 ± 2.3
Cr	64.3 ± 8.3	79.1 ± 10.2	59.0 ± 7.4	75.3 ± 9.6
Fe	36543 ± 7991	44197 ± 6505	32432 ± 5723	39165 ± 3728
La	38 ± 5	50 ± 5	38 ± 4	47 ± 7
Ce	75 ± 11	104 ± 17	71 ± 12	107 ± 24
Eu	1.1 ± 0.2	1.7 ± 0.2	1.1 ± 0.2	1.5 ± 0.2
Yb	2.7 ± 0.4	3.5 ± 0.4	2.9 ± 0.4	3.3 ± 0.4
Lu	0.45 ± 0.07	0.56 ± 0.07	0.47 ± 0.07	0.55 ± 0.06
Th	13.7 ± 1.5	16.6 ± 2.0	13.8 ± 2.2	17.2 ± 2.4

The parameter *n* represents the number of samples in each group.

In Table 3, it is possible to observe that the concentrations of chemical elements in the group B are higher than in A for both sites. In fact, in Fig. 3 it was observed a significant superposition between groups A for Lago Grande, the groups with the lowest elemental concentrations. The same can be stated for groups B, the groups with the highest elemental concentrations. The similarities in the concentrations and their dispersion for groups A and B in Table 3 is reflected in the superposition of the ellipses of chemical groups observed in Fig. 3.

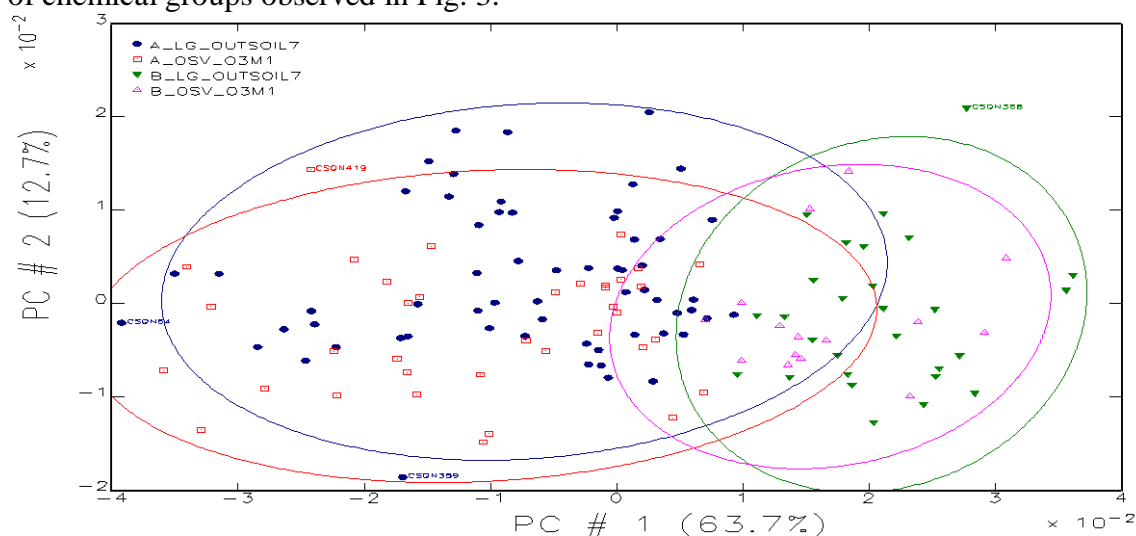


Figure 3 – PCA scores of elemental concentration data from Lago Grande and Oswaldo archaeological pottery combined after classification refinement. Ellipses represent the 95% confidence region.

An indirect validation of the chemical groups defined can be done by the analysis of the coefficients of variation (*cv*). For Lago Grande, the *cv* values varied between 11% and 20% for group A (exception for iron with *cv* = 22%), and between 11% and 15% for group B (exception for cerium with *cv* = 17%). For Oswaldo, the *cv* values varied between 11% and 20% for group A, and varied between 10% and 14% (exception for cerium with *cv* = 23%) for group B.

Harbottle [40] suggested that analytically well determined groups frequently would present coefficients of variation between 15% and 20%. Bishop [32] compared many archaeometric studies throughout the world and observed that chemical groups

properly determined would present coefficients of variation between 10% and 15%. The *cv* values for groups A of Lago Grande and Osvaldo match the variation suggested by Harbottle, and the *cv* values for groups B match the interval suggested by Bishop.

The abundance criterion cannot be applied in this work [22], as the sampling was driven to increase the number of samples regarding the probably exogenous ceramic phase at each site.

## 5. Some archaeological implications

Ideally, the chemical groups defined in the previous session would be correlate of only one ceramic paste recipe. However, there are many constraints to this direct approach.

The first factor of complexity is the possibility of combination of different clay sources to produce a ceramic artifact. Chromatic analyzes of Portocarrero [14] and Lima [13] indicated that the probability of combination of clays is high for the ceramic materials studied in this work. This variability may be driven by the different performance criteria required for objects in different uses by people.

Based on the classification of ceramic phases for each pottery shards by the archaeologists, a histogram of their distribution in the chemical groups defined in the multivariate analysis was plotted in Fig. 4.

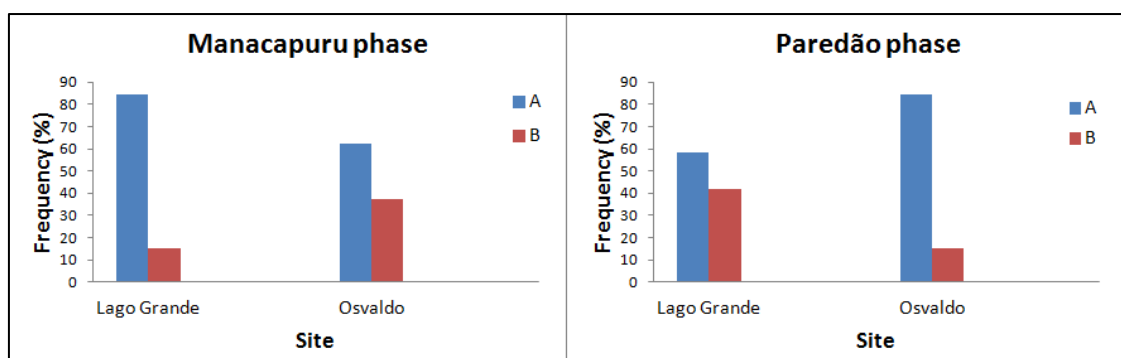


Figure 4 – Distribution of Paredão and Manacapuru pottery shards in the chemical groups of Lago Grande and Osvaldo sites.

Some hypotheses can be drawn from Fig. 4. The Manacapuru samples concentrate in groups A for Osvaldo and in a higher degree for Lago Grande. The Paredão samples concentrate in group A for Lago Grande and in a higher degree in Osvaldo. The concentration of the pottery shards representing both ceramic phases in the groups A, would indicate that the combination of raw materials, or recipes, represented by such groups would be the preferred by ceramists from both sites.

The fact that the distribution of Manacapuru and Paredão ceramics in the chemical groups were not the same for both sites in Fig. 4 have two potential implications regarding trade assumption. First, this might indicate a change in the selection of raw materials (mainly clay and antiplastics) for the ceramics produced especially for trade between Lago Grande and Osvaldo. A second possibility would be that exogenous ceramics would enter both sites not only by direct trade between Lago Grande and Osvaldo, but also by trade with other Manacapuru and Paredão contemporary sites. The archaeological evidences for such an assumption are the higher time span for Lago Grande occupation when compared to Osvaldo, regional networks identified in other regions of Amazon [16], and the existence of other contemporary sites which have also produced Manacapuru and Paredão ceramics. Furthermore,

preliminary comparisons of data from Lago Grande and Osvaldo with Hatahara and Açutuba, other important archaeological sites in Central Amazon, indicated a partial geochemical superposition among samples of pottery shards analyzed by INAA [28]. This might indicate a larger exchange network.

More generally, three mechanisms would be inferred for exchange networks in Central Amazon: trade, exogamic marriage and territorial integration. Trade would be a means of reinforce social and political integration in the region by commercial relations. On the other hand, ethnographic research identified the practice of exogamic marriages in current times [41]. In this modality, women would carry gifts from their original village, and/or continuing producing ceramics in an “old fashion”, looking for raw materials and means of production practiced prior to the marriage. The identification of hybrid ceramic pieces in Lago Grande would corroborate this mechanism (Lima, 2008). The last mechanism of territorial integration would imply that the ancient inhabitants of Lago Grande, Osvaldo and other contemporary sites used common sources of raw materials. There are evidences in the ethnographic literature for the region that those three mechanisms could have occurred [13, 16]. In this sense, the multivariate geochemical patterns identified in this work would reflect complex and multimodal means of social and cultural interaction in Central Amazon.

The existence of lakes connected by *igarapés* (little water channels) along the Negro and Solimões confluence region, connecting the lakes around which Lago Grande and Osvaldo are located in flooding periods, would facilitate the contact between those peoples. Archaeological research suggests [17] that the existence of exchange networks would be a condition for the existence of long-term occupations, as identified in archaeological record of Lago Grande, and evidences of pre-Columbian forms of agriculture in interfluvial uplands within Amazonia. It contrasts with the short-term occupations documented ethnographically and gives an additional basis for theories which minimize the role of ecological constraints for the emergence of social complexity and sedentarism in the region.

## 6. Conclusion

The application of multivariate statistical analysis to elemental concentration data, obtained by INAA, was carried out with samples from Lago Grande and Osvaldo, two important and representative archaeological sites in Central Amazon. Regarding analytical and geochemical concerns, it is not advisable to use the chemical elements Na, K, Cs, As and Hf for statistical analysis of ceramics from the region, if one seeks for archaeologically meaningful groups. They were presented evidences for diagenetic effects and possible geochemical mechanisms that would drive those post-depositional alterations. Furthermore, the methodological approach adopted yielded useful results for the archaeological context considered in this work.

The combination of cluster and principal component analysis was used for the definition of two chemical groups of pottery shards for each site. Their pairwise superposition in the two first principal components indicated geochemical similarities that may be archaeologically interpreted. The results might be correlate of complex and multimodal mechanisms of interaction, like trade, exogamic marriage and territorial integration. In any case, it generated evidences of socio-cultural interactions in Central Amazon, which may have implications for the archaeological theories regarding the pre-colonial Amazonian occupation which try to minimize the role of environmental constraints for the emergence of social complexity in the region.

Definition of more chemical groups was tried for the data set used in this work. However, there were problems of sample size for some groups. Additionally, no obvious archaeological correlations were found for such a higher resolution in the clustering procedure. Sampling expansion of ceramic artifacts from Central Amazon would help to work in a higher geochemical resolution and seek for further archaeological evidences and relations that would be useful to Amazonian Archaeology.

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