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DETERMINATION OF THE RAW MATERIAL SOURCE USED IN THE PRODUCTION OF CERAMICS OF THE HATAHARA ARCHAEOLOGICAL SITE, AM.

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ABSTRACT

The archaeological interventions carried out at the Hatahara archaelogical site, located in the central Amazonia, showed the presence of a great amount of ceramic artifacts in this region. As a consequence, several works have been conducted with this archaeological material, searching clear questions on how the ancient societies produced such objects, as well as, the use they did of the environment where they were inserted. Considering that the analysis of the ceramic material showed the simultaneous occurrence of four distinct phases of occupation in the Hatahara site, which, in relation to its pre-colonial composition is as an integral part of a quite complex context, the present work had the purpose of helping the Archaeologists to understand better the development of the societies that occupied this region, with basis on the study of the archaeological ceramics provenance. For this, the chemical characterization was done, with application of the analytical technique by neutron activation analysis (NAA); the elementary concentrations of As, Ba, Ce, Co, Cr, Cs, Eu Fe, Hf, K, La, Lu, Na, Nd, Rb, Sb, Sc, Ta, Tb, Th, U, Yb and Zn were determined in 127 ceramic fragments and in 7 samples of clay, collected next to the Hatahara archaeological site. The data of elementary concentrations were submitted to the multivariate statistical analysis, the techniques of cluster analysis and discriminant analysis. The results showed that a single type of clay was used in the manufacture of a group of 25 ceramic fragments, belonging to the phases Paredão, Manacapuru and Guarita. These results have been added to the archaeological interpretations with regard to the classification of the rescued ceramics fragments, in order to complement them. Therefore, this work supplied some pertinent clarifications that certainly will give support to the reconstruction of human path in the Hatahara archaeological site.

1. INTRODUCTION

Production technology, provenance and use of the ceramics by ancient societies, have been the main issues approached since 1950, when the first works about these materials started to appear [1]. Under this perspective, the physical and chemical techniques constitute essential tools, being constantly used for the study of such aspects.

The contribution of the earth sciences to study archaeological materials, as a representation of a common techniques and approaches of distinct areas of knowledge, allow clear processes that drive social, political, economical aspects, patterns and customs of the people that occupied the archaeological sites bringing, as well as, explanations concerning interactions and exchanges occurred with these people through of the centuries. These studies have created a series of information that aim to complement the archaeological works and help in the reconstruction of human past in these areas [2].

The archaeological ceramics are important tools for the study of the societies lifestyle, which had manufactured them. Buried or on the surface of archaeological sites, the ceramics have remained for several centuries, because, since their production, they have become, virtually, indestructible [1]. These artifacts are of great value for the Archaeology, because they keep characteristics, which reflect patterns of behavior of a group of persons [3].

Clay is the main component of the ceramic. In provenance studies, the elementary concentrations determined in the ceramics and clays are correlated. Due to the fact that these elements are represented for lower concentrations, archaeometric studies were used, because they provide better information [4]. The objective is to verify the chemical similarity between the elementary concentrations in order to determine the raw material source used in the production of the ceramics. In this case, the "Provenance postulate" will be applied, which is the chemical difference between two sources of clay. This difference must be higher between two distinct sources, than within one same source [5].

The Hatahara archaeological site, located in the central Amazonia, has a great amount of ceramics and black land in all its extension; it, also, shows the occurrence of four distinct simultaneous phases of occupation. Such aspects characterize the site, as quite complex in relation to its pre colonial composition, being archaeometric studies relevant for understanding the process formation. Thus, is possible to work, also, with other problems such as the chronology of these occupations, size and their duration [6].

In the present work, the results of the elementary concentrations obtained by analysis of the ceramic fragments were interpreted by multivariate statistical techniques, widely used in Archaeometry [7,8]. The ceramics were grouped according to similarities and dissimilarities derived from the chemical composition data, which indicated the existence of clay deposit used in the production of the ceramics.

2. STUDY AREA

The Hatahara archaeological site belongs to the district of Iranduba, 30 km southwestern of Manaus, in the region located on the left margin of the Solimões river, next to the merging with the Negro river. [9]. The site is formed by a farm with 160.000m^2 of area, in a fertile valley in the central Amazonia. Corn, rice and beans, represent the grains mostly grown in this area; papaya, banana and mango plantations are also found. As it can be observed in Figure 1, the Hatahara site is situated in the embankment adjacent to the swamps [10].

The Hatahara site is characterized by a great extension along the Solimões river, as well as by the presence of a great amount of ceramic material and black land, which is a type of rich soil in organic material, present in its entire surface. Some structures, called *montículos,* are also found. These structures are distributed in a total amount of thirteen and reach about 1.50m high and 3m of diameter. Since 1999, when the archaeological interventions started, two *montículos* have been excavated. In a vertical perspective in Figure 1 (below), a deeper layer of land, 2.50m, is represented by the *Açutuba* phase, where a few ceramic fragments were found.

Beyond, the second depth was found, with a few ceramics from the *Manacapuru* phase and presenting black land. The third depth excavated is formed by ceramics from the *Paredão* phase, with black land, fauna and flora remains, apart from human burials. The fourth depth is formed by a mix of fragments belonging to the *Paredão* and *Guarita* phases, with black land and fauna remains. The fifth and last depth is formed by few fragments of the *Manacapuru* and *Paredão* and *Guarita* phases, together with a great amount of black land. The archaeological interventions made possible that archaeometric studies could be carried out with a great amount of ceramics rescued of the Hatahara archaeological site [11,12,13].

Figure 1. Aerial vision of the Hatahara archaeological site (photo Neves, 1999; signalizing Grosch, 2005).

3. ANALYTICAL TECHNIQUE

3.1 Neutron activation analysis

The NAA is a nuclear analytical technique and has great applicability in searches that involve historic objects [2]. The analysis uses a small amount of sample, with relatively easy preparation, allowing the determination of several elements simultaneously, with precision and accuracy [14]. The neutron activation analysis is based on the measurement of the induced radioactivity in the sample elements, by means of the irradiation with neutron. This phenomenon occurs due to a nuclear reaction, when a neutron is captured by the target forming a compound nucleus in an excited state. This interaction results in the formation of a radioisotope.

For this work, the particles of interest are the gamma ray, emitted according to the half-life of each radioisotope. Thus, being the energy of gamma rays and half-life characteristic parameters for each radioisotope, it is possible to obtain the qualitative and quantitative determination of the elements present in the samples.

4. STATISTICAL STUDIES

4.1 Cluster analysis

The cluster analysis is a multivariate statistical technique that has, as main objective, to group similar samples in accordance with their characteristics. For the group's formation, it is necessary to consider the proximity between the points, since points that are next represent regions whose samples are similar. In the end of the application phase of the cluster analysis, the groupings can be represented graphically, by means of dendrogram. The visual inspection of the dendrogram allows the identification of the groups formed.

4.2 Discriminant analysis

The discriminant analysis is a multivariate statistical technique that has the objective of discriminate the population and/or classifying objects in populations previously defined. [15]. Thus, the main objectives of this technique are (a) finding functions of the original variables (discriminant functions) that explain samples differences and (b) allowing new samples to be inserted in the populations involved in the analysis. For the application of the discriminant analysis, the populations should be well-defined.

5. EXPERIMENTAL

5.1 Sample preparation

The ceramic fragments were, initially, washed with water using a brush of fine bristles. Then, the external surface of the ceramics was cleaned with tungsten carbide drill bits, with the purpose of preventing any contamination in the analysis, since the fragment collected was dirty. However, the tungsten carbide causes alteration in the elementary concentrations of Co and Ta [19]. In this work, the procedure to obtain the sample by means of tungsten carbide drill bits was utilized, because there are works in the literature reporting the relation of the contamination effect and the homogeneity of the samples to be analyzed. The results showed that, apart from preventing contamination, it is not also necessary to destroy the fragment [14,18].

Then, with the objective of assuring the chemical homogeneity, about 5 holes in different internal parts of each of the fragments were made. It was obtained 500mg in the form of powder from each fragment and this amount of sample was dried in 104ºC stove, for 24 hours [14].

The samples of clay were ground in agate mortar and put through a 100 to 200 mesh sieve, in order to obtain a powder fine enough for the trace and ultra trace element analysis [16,17].

5.2 Analytical procedure

127 samples of ceramic fragments and 7 clay samples were analyzed. For this, 120mg in powder of each clay sample were collected. As for ceramic samples, 120mg in powder were collected from 500mg, previously obtained from each of the fragments. This material was weighed in polyethylene packaging and sealed with iron, with the same amount of Standard Reference Material NIST-SRM 1633b, used as standard. These packaging were mingled with sheets of aluminum paper.

After this procedure, the samples and standard were irradiated for one hour at the IEA-R1 reactor of IPEN-CNEN/SP, under a thermal neutrons flow of 8.92 x $10^{12}cm^{-2}s^{-1}$. Two countings were carried out: As, K, La, Lu, Na, Nd, Sb, Sm, U and Yb were determined after seven days of decay. After 25-30 days of decay, Ba, Ce, Co, Cr, Cs, Eu, Fe, Hf, Rb, Sc, Ta, Tb, Th and Zn were determined.

6. RESULTS AND DISCUSSION

Initially, the concentrations of As, Ba, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Na, Nd, Rb, Sb, Sc, Sm, Ta, Tb, Th, U, Yb e Zn were determined in 8 samples of the Reference Material IAEA-Soil 7 Trace elements in soil, with the aim of studying the precision and accuracy for each of the elements. Some statistical studies were applied to the data, such as mean and standard deviation determination, mean confidence interval, precision and accuracy. The results demonstrated that most elements showed a precision ≤ 10%. This precision is considered by several authors [18], appropriate for the choice of chemical elements for studies of archaeological objects chemical characterization, using multivariate statistical analysis.

Co and Ta, although showed precision less that 10%, were eliminated, due the contamination by means of tungsten carbide during the sample preparation [19]. The Zn also was eliminated because it suffered interference in the gamma ray spectra of the ^{46}Sc. Although As, Ba, Nd, Rb, Sb showed a good precision, previous studies showed that are not reliable elements to insert in the database, due to the great dispersion in the ceramics concentrations. Therefore, the elements used were Ce, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Na, Sc, Tb, Th, U and Yb.

Following, the concentrations of the ceramics and clay samples were transformed in $\log_{10}$. This transformation before applying multivariate statistical techniques is a usual procedure in archaeometric studies and there are two reasons for this: the first is explained by the fact that a normal logarithmical distribution of the elements exists. The other is the difference magnitude between elements, which it was found in percentage and trace level. After this, the detection of the outliers was done by means of *Mahalanobis* distance, D_i^2, a method used when numerous variables are determined [20]. Considering a database with *n* samples and *p* measured variables, the distance (Di^2) is calculated by:

$$D_i^2 = \left(X_i - \overline{X}\right)^T S^{-1} \left(X_i - \overline{X}\right) \tag{1}$$

where,

$$S = \sum_{i=1}^{n} \left(X_i - \overline{X}\right) \left(X_i - \overline{X}\right)^T / n - 1 \tag{2}$$

S represents the covariance-matrix;

X_i is the observation of interest;

$\bar{X}$ is the vector of the average;

T is the relocated matrix

For each sample in the variables set, D_i^2 was calculated using the lambda Wilks criterion as critical value [21]. The samples with values overtaking the critical value were eliminated and the same procedure was repeated for the remaining samples. The procedure finished when the samples showed values of *Mahalanobis* distance lower than the critical value. By means of this procedure, seven sample outliers were found and removed from the data set.

Due to the limited number of pages in this work, it was not possible to show the results of elementary concentrations for 127 samples of ceramic fragments and 7 clay samples, as well as for values of *Mahalanobis* distance and outliers removal.

6.1. Cluster analysis

After the elimination of the samples outliers, the 120 ceramic samples results were submitted to cluster analysis using Ward´s method and Euclidean distance. The dendrogram (Figure 2) show the similarity between ceramic samples and the groups formed. The samples were separated in five groups, showing similarity in the chemical composition between the samples gathered in each group. This allows concluding that five distinct raw material sources were used in the production of the ceramic artifacts of the Hatahara archaeological site.

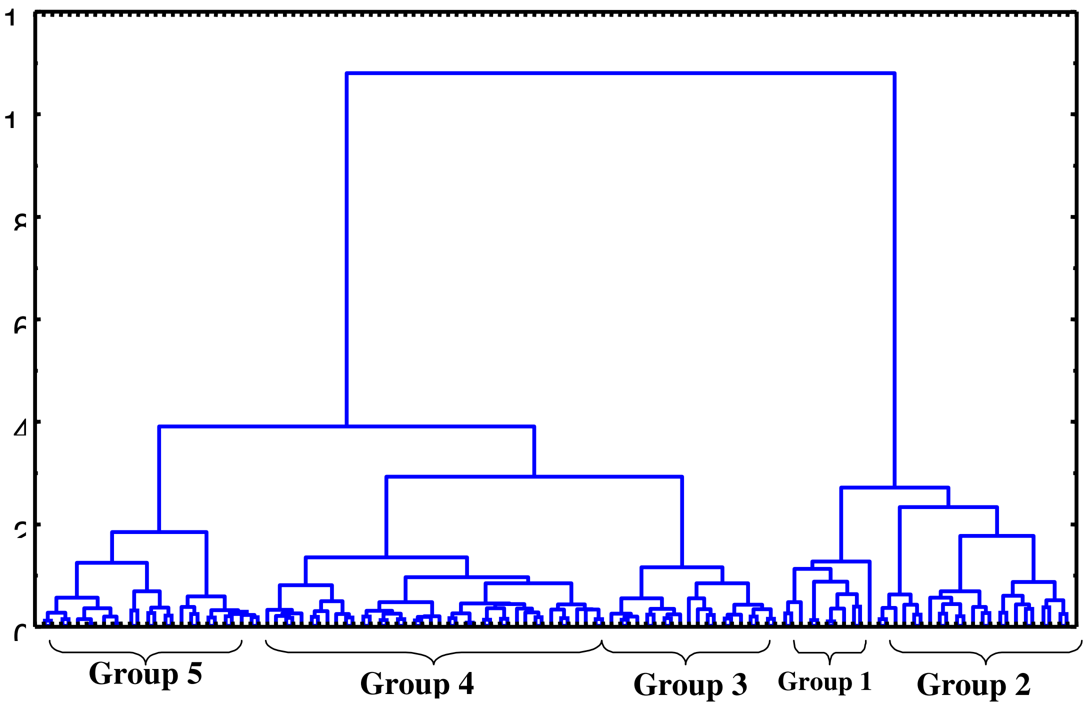

Figure 2. Dendrogram of the ceramics sample using Euclidean distance and Ward´s method.

6.2 Discriminant analysis

After knowing how many groups are involved, the discriminant analysis was applied to the ceramic samples. The discriminant function 1 vs discriminant function 2 in Figure 3 shows the five groups with the samples belonging to each group. It is possible to see that no occurred variation in relation to the number of groups formed by dendrogram.

As it can be seen in Figure 3, the ceramics related to the *Açutuba* phase is missing. This phase is the more ancient occupation of the site, from 300 B.C. to 360 A.D. This phase is not associated to the formation of black land in the region.

The groups of ceramics 1 e 3 are represented by ceramics belonging to the *Manacapuru* phase. Its period of occupation in the region extended from the century V to IX. The formation of these groups revealed only the presence of ceramics related to this phase, but, it was used a different source of clay for each group of these ceramics. The most common typological characteristics in the ceramics of groups 1 and 3 are simple rim**,** parallel and curved incisions.

By means of Figure 3, it is possible to see that a larger amount of ceramics exists in the groups where the presence of ceramics of the *Paredão* phase is predominant (groups 2, 4 e 5). This occurs by the fact that there is a larger amount of ceramics of the *Paredão* phase, than ceramics of other phases.

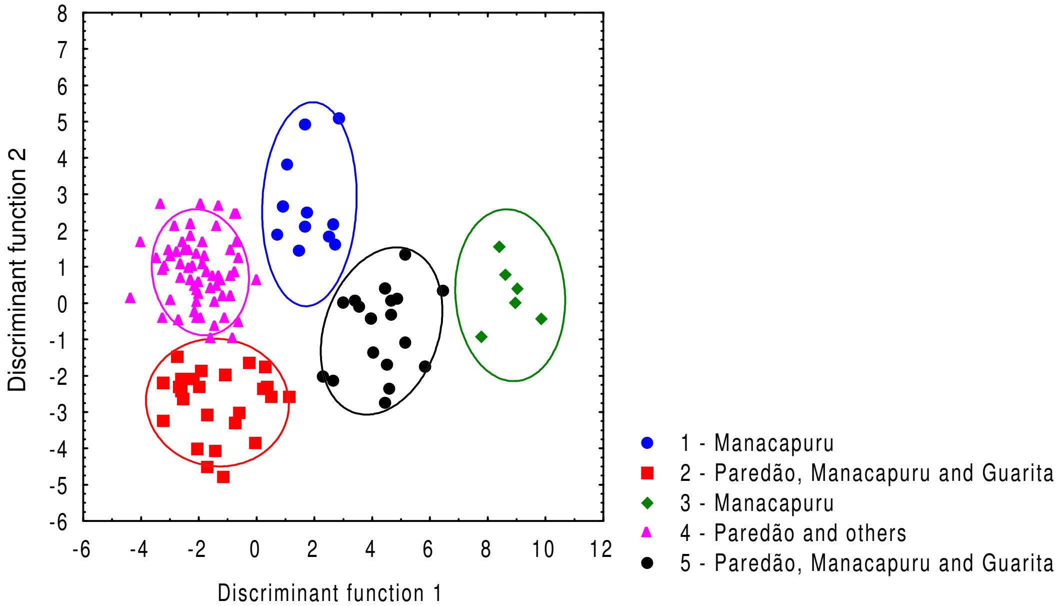

Figure 3. Discriminant function 1 vs discriminant function 2 for the ceramic samples.

The *Paredão* phase, whose occupation extended from the final of the century VII to century XI, is represented by ceramics characterized by vessels and bottles, some of them with pedestals and funerary urns. The decoration is produced with red painting in spiral and anthropomorphic figures**.**

The most recent occupation in the central Amazonia is represented by the *Guarita* phase, which extends through the centuries X and XVI. Ceramics of the *Guarita* phase are disperse for archaeological sites and found in all central Amazonia, being characterized by polychrome figures (red and black) and anthropomorphic funerary urns. Ceramics associated to the traditional polychrome of the Amazonia were produced by means of complex techniques of decoration and are associated to funerary urns identified in sites, generally, of large pasture and of long occupation [22]. Group 5 is represented by ceramics that correspond to 50% of the *Guarita* phase, 25% of the *Paredão* phase and 25% of the *Manacapuru* phase. A unique source of clay was used for the production of the artifacts that make up group 5.

The results confirm the way which the ancient societies occupied the Hatahara archaeological site. The occurrence of ceramics in the same group, with distinct typological characteristics, referring to distinct phases of occupation, with the same raw material source, shows the simultaneous occurrence of societies that developed in the site, as well as, the interaction between their patterns of behavior and own aspects.

6.3 Determination of the raw material source of the ceramic artifacts

The clay samples were analyzed with the purpose of studying the raw material source used in the production of the ceramic artifacts. For this, seven samples of clay next to the Hatahara site were collected, with five samples collected in the margin of the Solimões river, in front of Iranduba, 6km far from the site. The other two samples of clay were collected next to Lago Grande site, approximately 9km far from the Hatahara site. Figure 4 shows the discriminant function 1 vs discriminant function 2, for the seven samples of clay, along with the samples of ceramics.

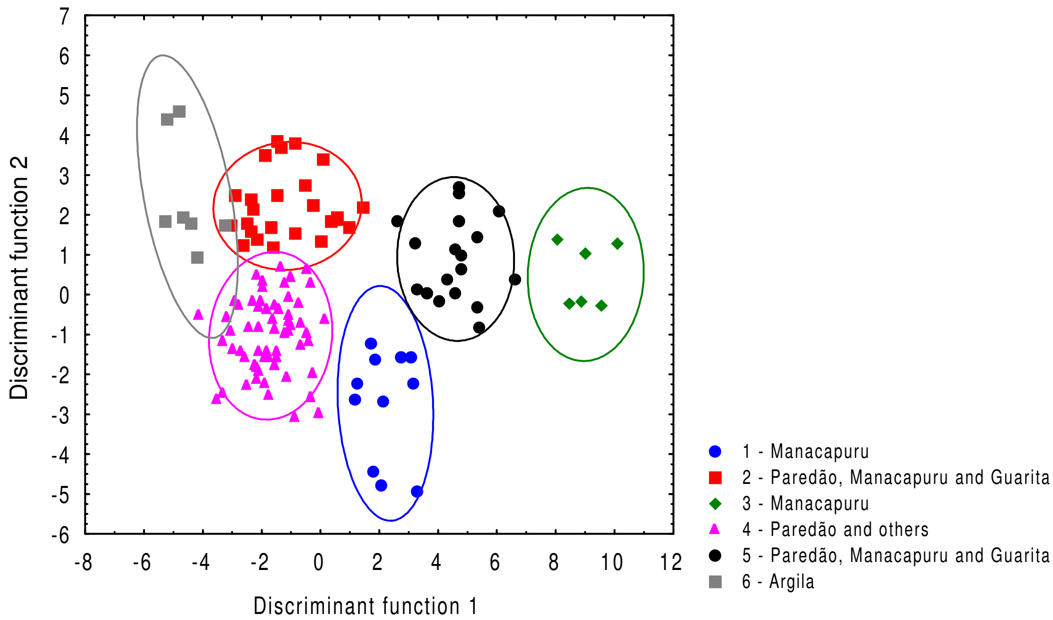

Figure 4. Discriminant function 1 vs discriminant function 2 for the ceramic and clay samples.

As shown in Figure 4, group 6, represented by the group of clay samples, is quite near group 2, formed by 50% of ceramics from the *Paredão* phase and 50% divided equally between ceramics from the *Manacapuru* and *Guarita* phases. It is possible to see that a unique sample of the group 6 is part of group 2 of ceramics, and this allow to stating that such clay was used as raw material source for the production of the ceramics from group 2, due to the similarity that exists between the chemical compositions of this ceramics and clay.

The sample clay used in the production of the artifacts of the group 2 was collected 6km southwestern of the Hatahara site, in the Island of Patience, in the form of embankment in the margins of the main channel. The clay of this place is inserted in deposits of the alluvial plain of the Solimões river [23]. The aspect of the clay deposited in this place, its form of exposition and the proximity of this deposit with the Hatahara site, certainly facilitated the collection of the raw material by the communities, in that period.

7. CONCLUSIONS

The method of neutron activation analysis was extremely important for the determination of several chemical elements, found in lower concentrations in the archaeological ceramics. The results obtained in this work allow concluding that the analytical method is appropriate for this study, showing good precision and accuracy.

In 127 samples of ceramic fragments of the Hatahara archaeological site, the elementary concentrations of Ce, Cr, Cs, Eu, Fe, Hf, K, La, Lu, Na, Sc, Tb, Th, U e Yb were used. The interpretation of these data, by means of multivariate statistical methods, supplied information concerning studies of raw material sources used in the production of the ceramic artifacts found, as well as, confirmed archaeological interpretations as to the distinct simultaneous phases of occupation.

It was possible to identify, in the group of the clay samples, the existence of a unique sample as the raw material source used in the production of the group with 25 ceramic fragments of the *Paredão*, *Manacapuru* e *Guarita* phases.

The present work contributed with the archaeological studies carried out in the Hatahara site. It was possible to obtain some explanations that certainly will support the reconstruction of the human passage in the Hatahara archaeological site, in the region of central Amazonia.

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