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Box-Cox transformation on dataset from compositional studies of archaeological potteries

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Abstract In archaeometric data set it was verified by statistical tests that some variables almost never follow a multivariate normal distribution using logarithmic transformation or other. This work presents a multivariate Box-Cox transformation for Mardiás and Roytońs tests for a data set of fifty ceramic fragments from archaeological site Justino, Xingó, Brazil, and one clay sample collected near of the site. The samples were analyzed by instrumental neutron activation analysis, INAA. The study was made using companion to applied regression package from R software and was tested by Hotelling's T^2 statistics.

Keywords Geochemistry · Archaeometry · MVN test · Box-Cox transformation · INAA

Introduction

Archaeological potteries could be seen as a source of insights into people and cultures, since the ceramics enclose messages about the society that manufactures and use, such as its cosmological structures, rituals, trade routes, productive systems and others feature [1]. Technological information of potteries from natural science together classical archaeological approaches have been used to reconstruct ancient cultures [2–4].

A number of different analytical techniques have been applied with varying degrees of success to Archaeometry studies [5]. Although a number of techniques have been employed to characterize potteries, the instrumental neutron activation analysis (INAA) it is one longest and most successful for provenance studies [5, 6].

The formation of compositional groups based on their elemental chemical compositions and the evaluation of their provenances are aims of archaeometry studies of ancient ceramics. For that, it has been applied several multivariate statistical techniques [7]. Many techniques require the assumption of multivariate normality of data, or at least it is considered to be desirable, because the performance of these methods dramatically decrease if the data are not multivariate normal [8].

In this paper was studied the multivariate normality of two compositional groups of ceramic archaeological from sites of the Brazilian Northeast region, which it was generated by INAA. The multivariate normality it was verified by Mardia's test and Royston's H test that can be implemented in the MVN contribution of R Package [9, 10]. This study is also presented an application of the Box-Cox transformation to multivariate normality, based on companion to applied regression package (CAR) from R program, once the logarithmic transformation does not produce the data normalization [11]. For this 50 ceramic fragments were studied by means of INAA and one clay sample.

Methodology

Sample collection

The samples were collected at Justino site, situated in Canindé do São Francisco, about 150 km from Aracaju, Sergipe State, in the Brazilian Northeast (Fig. 1). In order to obtain the results, 50 ceramic fragments (Cemeteries: B

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25 and C 25) were studied using INAA and a clay sample. Chronology studies of ceramist occupation at Justino site, accomplished by thermoluminescence dating, showed chronology amplitude from 5.570 to 1.280 BP [12–14].

Sample preparation

Ceramic powder samples were obtained by cleaning the outer surface and drilling to a depth of 1–2 cm using a tungsten carbide rotary file attached to the end of a flexible shaft, variable speed drill. Depending on the thickness, 3 or 5 holes were drilled as deep into the core of the fragment as possible without drilling through the walls. Clay sample was ground in an agate mortar until a granulometry of 100 mesh was achieved and quartered for the chemical determination by INAA.

Constituent elements in coal fly ash—NIST-SRM-1633b, were used as standards in all analysis. The standard reference material brick clay—NIST-SRM-679 was used to check the analytical quality of the results. The standards and the samples were dried in an oven at 105 °C, the standards for 4 h and samples for 24 h and stored in desiccators until weighing [15–17].

Irradiation

About 100 mg of sediments samples, and NIST-SRM-1633b were weighed in polyethylene bags and wrapped in aluminum foil. Groups of 8 ceramic samples and two reference materials were packed in aluminum foil and irradiated in the swimming pool research reactor, IEA-R1m (IPEN/CNEN-SP) at a thermal neutron flux of about 5×10^{12} cm^{-2} s^{-1} for 8 h.

Gamma spectrometry

Two measurement series were carried out using Ge (hyperpure) detector, model GX 1925 from Canberra, with a resolution of 1.90 keV at the 1332.49 keV gamma peaks of ^{60}Co, with S-100 MCA of Canberra with 8192 channels. As, K, La, Lu, Na, Nd, Sm, U, and Yb were measured after 7 days cooling time and Ba, Ce, Co, Cr, Cs, Eu, Fe, Hf, Rb, Sb, Sc, Ta, Tb, Th, and Zn after 25–30 days. Gamma ray spectra analysis and the concentration were carried out using the Genie 2000 NAA Procedure from Canberra.

Statistical interpretation

Outlier detection

The outlier detection in multivariate space is an important task in statistical interpretation of archaeometry data because its presence may interfere in statistical interpretation of data, especially in provenance studies [3]. Generally, the atypical samples in a data set of elemental chemical composition does not need to be necessarily high (or low) in relation to all values of a variable, and thus attempts to identify these samples with classical univariate methods commonly fail. In order to resolve this difficulty, several methods to identify outlier on multidimensional data set have been proposed based on covariance matrix and mean vector, such as the methods which use Mahalanobis distance [18]. This statistical method often uses classical Mahalanobis distance (MD) to detect if an observation is far from the center of the data distribution and if MD is larger than critical value the sample is candidate to be outlier. However, masking effects decrease the MD of an outlier, on the other hand, swamping effects increase the MD of non-outlying observation [19].

In this context, the minimum covariance determinant estimator (MCD) is frequently used as robust estimator for location and covariance, where MCD is determined by a subset of observation of size k, generally $k \approx 0.75n$ (n is a number of samples), which minimizes the determinant of the sample covariance matrix. The choice of k determines

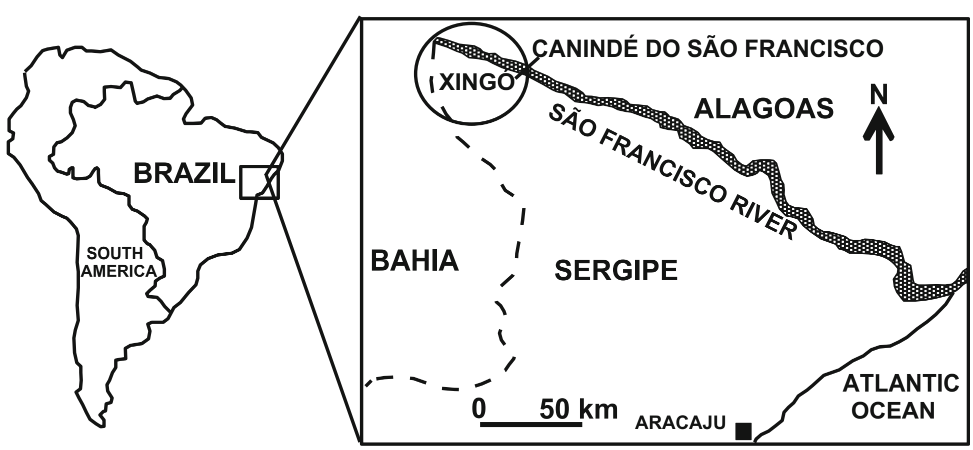


Fig. 1 A localization map of the study area

the robustness of the estimator. Using robust estimators of location and scatter in Eq. (1) results in so-called robust distances (RDs) and if $RD^2 > \chi^2_{p,0.975}$ the sample can be declared a candidate outlier [20]. In this work, for outlier detection by RD analysis the "*mvoutlier*" package running under R environment were used [21–24].

Multivariate normality tests

The assumption of multivariate normality (MVN) is required in many multivariate statistical procedures to Archaeometry studies, such as linear discriminant analysis (LDA) and multivariate analysis of variance (MANOVA) [25, 26]. In provenance studies of archaeological potteries as well as geochemistry researches, this basic requirement is still neglected [27]. Many archaeometric studies have utilized a lognormal statistical distribution for interpretation of chemical composition of archaeological potteries. But, frequently some elements are not lognormally distributed [3, 4]. Thus, it is necessary to check the assumption of multivariate normality to statistical interpretation.

In assessing multivariate normality, a number of test procedure have been proposed, which most of them are based either on generalization of the univariate test statistics [28, 29]. Unfortunately, there is no uniformly most power test and it is recommended to perform several tests to assess MVN [30]. In this work, two tests were applied to assess the MVN of data which it was obtained by INAA: Mardia's MVN test and Royston's test, which are implemented in R [9].

Mardia's MVN test is based on p-multivariate extensions of skewness ($\hat{\gamma}_{1,p}$) and kurtosis ($\hat{\gamma}_{2,p}$) and it is available in a number of the general purpose statistical software package [9]. Here, the skewness ($\hat{\gamma}_{1,p}$) and kurtosis ($\hat{\gamma}_{2,p}$) measures as follows:

$$\hat{\gamma}_{1,p} = \frac{1}{n^2}\sum_{i,j} m_{ij}^3 \quad \text{and} \quad \hat{\gamma}_{2,p} = \frac{1}{n}\sum_{i,j} m_i^2 \tag{1}$$

where m_i^2 is Mahalanobis squared Distance. In this paper, the multivariate skewness and kurtosis has been calculated as well theirs corresponding statistical significance (5 %), by means of use of MVN package, which is implemented in R [9].

The Royston's test is based on the well know omnibus test for testing univariate normality [30]. The test it was performed using R program, with significance of 5 %. This H statistic is approximately distributed as a χ^2_ξ and it can be obtained:

$$H = \xi \frac{\sum_j R_j}{p} \tag{2}$$

where ξ is the equivalente degrees of freedom and R_j is related to average and variance, and it can be calculated according:

Table 1 Composition mean (*M*) and coefficient of variation (CV %) for pottery Cemeteries B and C

	*Cemetery B ($n = 19$) M (CV)	*Cemetery C ($n = 18$) M (CV)
Cr	22.56 (72.31)	27.94 (52.29)
Cs	5.72 (40.17)	4.23 (60.89)
Eu	2.49 (22.03)	1.75 (50.61)
Fe %	5.31 (22.88)	3.89 (37.88)
Hf	7.59 (19.88)	7.22 (36.18)
La	53.54 (24.18)	44.95 (40.44)
Lu	0.64 (33.22)	0.43 (39.44)
Na %	2.08 (12.79)	1.77 (23.65)
Sc	15.63 (22.81)	11.35 (43.44)
Th	11.94 (37.78)	13.14 (53.58)
Yb	4.09 (34.48)	3.06 (47.50)

Values in ppm unless indicated

* Compositional groups without outliers

$$R_j = \left\{\Phi^{-1}\left[\Phi(-Z_j)/2\right]\right\}^2 \tag{3}$$

where Z_j denote the value obtained after applying the normalizing transformation, with $1 \leq j \leq p$ and Φ denotes the standard normal in the cumulative distribution function (cdf) [30].

Multivariate Box-Cox transformation

The logarithmic transformation of the data is the type most used in archaeometry studies to provenance determining of potteries on based to elemental chemical composition [31].

Box-Cox transformation is considered a family of transformation for $X > 0$, in univariate data set, where t, for $\lambda > 0$ and $Y(\lambda) = \log_e^X$, $\lambda = 0$ [32]. The coefficient λ can be estimated by using the maximum likelihood method [33]. The Box-Cox transformation to multivariate data and its significance test for $\hat{\lambda} = (\lambda_1, \ldots, \lambda_p) = (1, \ldots, 1)$, considering that p is the number of variable, which can be defined by

$$Y_{ij}(\lambda_j) = \begin{cases} \dfrac{X_{ij}^{\lambda_{ij}} - 1}{\lambda_{ij}}, & \text{if } \lambda > 0 \\ \log_e^{X_{ij}}, & \text{if } \lambda = 0 \end{cases} \tag{4}$$

In this work, we used the maximum likelihood approach to select a transformation of a multivariate response for normality, by using, "*CAR*" R package [34].

Linear discriminant analysis

One of the most widely used multivariate procedures to data interpretation in geological samples is discriminant analysis [35]. Mathematically, the discrimination of

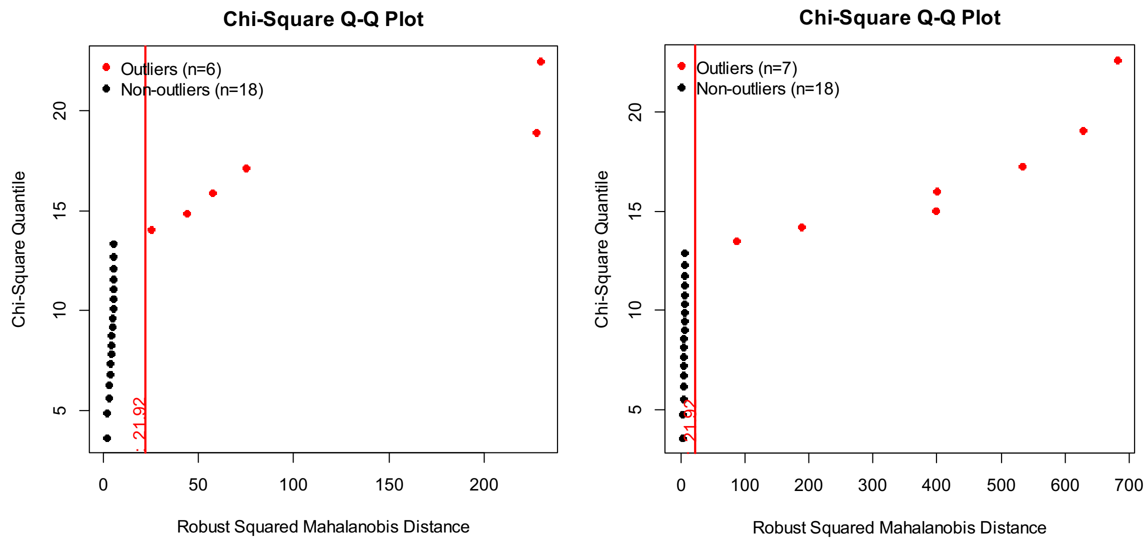


Fig. 2 Multivariate outlier detection to Justino data (*left* Cemetery B and *right* Cemetery C)

Table 2 The results of $\hat{\lambda} = (\lambda_1, \ldots, \lambda_p)$ for MVN transformation of Justino data

Variable	Cemetery B		Cemetery C	
	$\hat{\lambda}_j$	p value[a]	$\hat{\lambda}_j$	p value[a]
Cr	−1.141	0.442	−0.704	0.102
Cs	−0.712		−0.042	
Eu	2.011		−0.523	
Fe %	0.200		0.194	
Hf	0.200		0.136	
La	−3.332		0.493	
Lu	−0.672		0.385	
Na %	1.254		1.007	
Sc	1.650		0.338	
Th	−0.278		0.387	
Yb	0.464		0.263	

[a] Likelihood ratio statistics about transformation parameters

Table 3 p values of MVN tests before and after transforming to Justino data set

Procedure	Cemetery B		Cemetery C	
	Before	After	Before	After
Mardia's test*	0.107	0.074	0.238	0.063
Mardia's test**	0.238	0.979	0.907	0.994
Royston's test	3.151e−07	0.053	0.0007	0.225

* p value to kurtosis and ** p value to skewness

samples is determined using the linear combination of variables that provides the maximum difference between two or more predetermined groups [36, 37].

Results and discussion

To evaluate the analytical process and to establish the chemical elements which can be used in the data interpretation, the elemental concentrations for reference material brick clay—NIST-SRM-679 were statistically compared with the data found in our laboratory. The precision of several elements (La, Th, Sc, Fe, Eu, Ce, Zn, Hf, and Co) was around 5 % or less and it was comparable with that obtained by other authors. For the elements determined with precision of 10 % or more (Nd, Rb, Sm, Ba, Sb, Ta, and Tb) are also in agreement with those found in the the literature [38]. The interference ^{235}U of fission in the determination of La, Ce and Nd was negligible because U concentrations did not exceed 5 ppm and the rare earth concentrations were not very low.

In this work all the elements with precision better 10 % were considered for interpretation of the results (Na, Lu, Yb, La, Th, Cr, Cs, Sc, Fe, Eu, Ce, Zn, Co, Ta and Hf) [39]. Zn presented RSD better than 10 % but was excluded from the data set because its determination suffers strong gamma ray interferences of ^{46}Sc. The elements Co and Ta were eliminated because their concentrations can be affected by tungsten carbides drills [40] even though their precision was smaller than 10 %. The K and Sb were better than 10 %, however they were excluded because they presented 15 % of missing values. Ce was removed from dataset because samples showed high variability, which can be related with characteristics of ionic exchange of the Ce with elements present in the clays [41]. Based on these screening criteria 11 elements: Na, Lu, Yb, La, Th, Cr, Cs, Sc, Fe, Eu and Hf were used in the subsequent data analyses. Mean and variation coefficients are presented in Table 1.

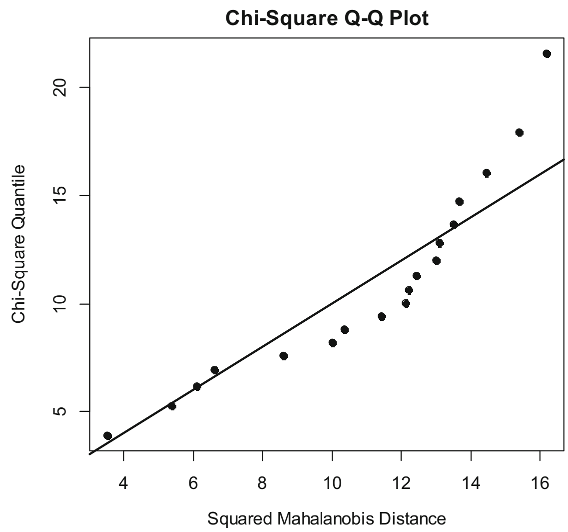
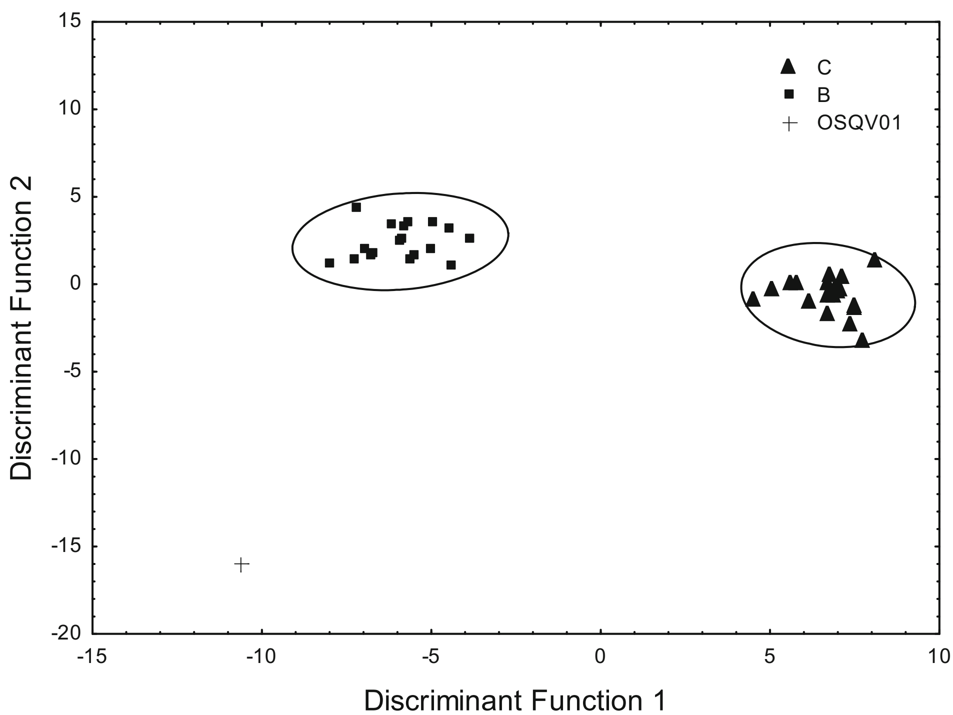


Fig. 3 Chi Square QQ-plot for Cemeteries B (*left*) and C (*right*)

Fig. 4 Linear discriminant analysis of the archaeological pottery samples from Cemeteries *B* and *C* from Justino site. Ellipses represent 95 % confidence level



To identify multivariate outliers on data set, we have utilized the Robust Mahalanobis distance (RD), with $k = 0.75$ [20]. They were found 13 samples that are far away and they are those samples outlying in data set. The Fig. 2 shows the Chi Square plot, where we can identify all outliers in Justino data, six samples in cemetery B and seven in cemetery C. In the Fig. 2, vertical axes indicate the cutoff value $\sqrt{\chi^2_{11;0.975}}$, and if the data set was not contaminated, then all points would lie near the cutoff lines. This approach is defined in the MVN package [9].

After that, it was studied the multivariate normality (MVN) by using two tests: Mardia's test and Royston's test. Table 3 and Fig. 3 are shown the results found, where you can identify the violation of multivariate normality. As can be seen in Table 3, it was rejected the null hypothesis H_0 that states the original data is normal, using a significance level of 5 %. Assuming multivariate non-normality for the data, the multivariate Box-Cox technique was applied to data set [34]. The lambda values are presented in Table 2, and it was computed by using CAR package [34].

The results from in Table 3 showed the effects of multivariate normality using Mardia's and Royston's test as a guide to see the effects of the joint transformation towards multivariate normality. We see in Table 3 that the jointly transformed data now passes the MVN tests, since the p values for two MVN tests after multivariate Box-Cox transformation were substantially greater. Thus, the multivariate Box-Cox transformation approach applied allowed obtaining a multivariate database to statistical interpretation by LDA (Fig. 3).

As seen from Table 3, Mardia's test indicate multivariate normality, but according Royston's test Cemeteries B and C data does not appear to follow a multivariate normality before Box-Cox transformation. One can clearly see that different MVN tests may come up different results, MVN assumption was rejected by Royston's test, but Mardia's test did not reject at a significance level 0.05. In such cases, examining MVN plots, such as QQ-plot, can be useful in order to reach a more reliable decision [9].

After multivariate Box-Cox transformation, a linear discriminant analysis (LDA) of 38 samples it was applied to confirm the existence of two different compositional groups. The Fig. 4 illustrates that the pottery samples from each cemetery B and C from Justino site are chemically homogeneous, since they concentrate in an ellipse with a confidence level of 95 %. These results indicate that the raw materials used to manufacture the potteries were distinct. In Table 1, the differences in Na, La, Lu, Sc and Fe are more prominent, considering the coefficient of variance. It is possible to see that Cemetery B is chemically different from Cemetery C, which suggests that a different paste was used.

According to the variability presented for two Cemeteries B and C in the Table 1, it can be seen that ceramic paste from Cemetery C is more heterogeneous than the one from Cemetery B. Cemetery C was once occupied by an incipient ceramist group, establishing a change from hunter—collector group to ceramist group because of this the variability of the pottery chemical composition in this Cemetery could be the consequence of the technological evolution on pottery manufacturing during this period, indicating an experimentation process in ceramics production. Nevertheless, during the period of Cemetery C occupation, expert craftsmen have detained the know-how to produce potteries with some quality control mainly, the ceramic paste homogeneity.

To confirm results from LDA, Hotelling's T^2 test was performed using the R package [42, 43]. Hotelling's T^2 test, testing the null hypothesis that vectors of mean of observation are the same between two groups ($H_o : \hat{\mu}_1 = \hat{\mu}_2$, where $\hat{\mu}_i = (\mathrm{Na}_i, \mathrm{Lu}_i, \mathrm{Yb}_i, \mathrm{La}_i, \mathrm{Th}_i, \mathrm{Cr}_i, \mathrm{Cs}_i, \mathrm{Sc}_i, \mathrm{Fe}_i, \mathrm{Eu}_i, \mathrm{Hf}_i)$. For data set, Hotelling's T^2 test approach rejected the null hypothesis for Cemeteries B and C, indicating that two cemeteries have potteries chemically distinguishable ($T^2 = 6.39$, p value < 0.05).

Conclusions

This paper showed the multivariate Box-Cox transformation as a potential best practice in statistical interpretation for provenance studies in order to assess the partitioning patterns of data to justify the application of some multivariate techniques, such as LDA and MANOVA. Two statistical tests were used to asssess the multivariate normality: Mardia and Royston's. The Royston's test has rejected the hypothesis in favor of multivariate normality to original data. However, the outline approach to find the optimal for a multivariate Box-Cox and it was applied to Justino data and we have demonstrated the use of multivariate Box-Cox transformation in achieving MVN.

The INAA of pottery from Justino site was successful in identifying distinct compositional groups from pottery chemical composition. The LDA and Hotelling's multivariate statistical procedure from elemental concentration of potteries and clay showed that the sources of raw material for two archaeological sites studied are different or was changed by ancient potters during its production.

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