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Discrimination of gossans using principal components analysis of standardized data

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ABSTRACT

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The results of a study involving geology, geochemistry, mineralogy and exploratory statistical analysis of data for a set of gossans located in the south of Portugal are presented.

The objective of the case study is to demonstrate how to complement data concerning the mineralogy and geochemistry of gossans with a multivariate statistical method – Principal Components Analysis of Standardized Data – in order to discriminate gossans according to their geological environment.

Moreover, a test was carried out in order to assess how summary measures for characteristic variables of each gossan compare (in this case, means versus robust estimates as medians).

INTRODUCTION

A geological, mineralogical and geochemical research programme was carried out in Alentejo, Southern Portugal, by the Portuguese Geological Survey, in order to detect sulphide ores.

During this programme, exposed gossans situated in three main Hercynian fold belts were studied by mineralogical and geochemical methods.

The results of this study and its articulation through the application of Principal Components Analysis of Standardized Data are presented. The objective is to discriminate between gossans, based on mean and median values of the distribution of 19 elements, and to select variables associated with economic mineralization.

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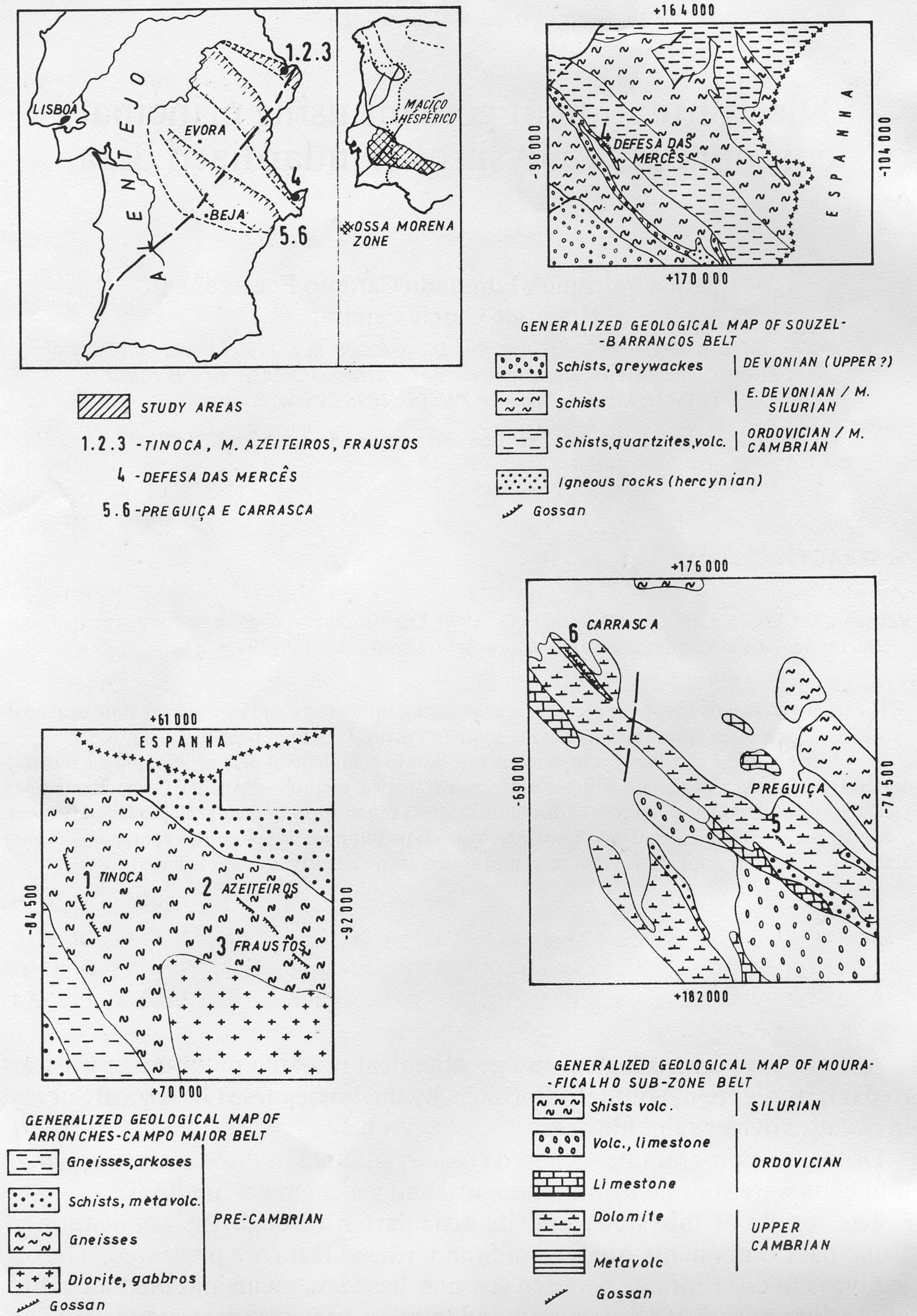


Fig. 1. Paleogeographic and geotectonic units of Portugal. Generalized geological map showing location of gossans.

GEOLOGY, PHYSIOGRAPHY AND MINERALIZATION

The study areas are situated in Alentejo, Southern Portugal (Fig. 1). The Alentejo province is a peneplain with residual rugged hills of low relief (Feio, 1951), some of them of tectonic origin.

A Mediterranean climate prevails in the zone, with an average annual rainfall of 550–650 mm.

Numerous economic concentrations are found in the zone. The most characteristic are polymetalic sulphides of Pb, Zn and Cu, occurring in Hercynian fold belts (Carvalhosa, 1965; Thadeu, 1965; Carvalho et al., 1971; Goinhas, 1971, 1981; Soares de Andrade, 1966, 1969, 1983).

Gossans are located in a paleogeographic and geotectonic unit of the Maciço Hispérico, the Ossa Morena zone (Lotze, 1945). Precambrian, Early Cambrian, Ordovician, Silurian and Devonian formations (Fig. 1) are present in the Ossa Morena zone (Nery-Delgado, 1908; Teixeira, 1951; Soares de Andrade, 1966, 1969, 1983; Gonçalves and Coelho, 1970; Carvalho et al., 1971; Carvalhosa, 1971; Carvalho, 1972; Bernard and Soler, 1974; Oliveira et al., 1985; Oliveira and Almeida, 1986).

The gossans studied in this paper are situated in three main Hercynian fold belts (cf. Fig. 1):

(a) Tinoca (1), Monte Azeiteiros (2) and Fraustus (3) are located in the Arronches–Campo Maior belt, where stratiform copper mineralization is found, related to Precambrian gneissic formations.

(b) Defesa das Merces (4) is located in the Sousel–Barrancos belt. In this area, copper mineralization occurs in Hercynian acid subvolcanic rocks and associated breccias, intruding Silurian formations. The mineralization (chalcopyrite, pyrite, pyrrhotite) fills the cement of subvolcanic breccias or is dessiminated in Silurian schists.

(c) Preguiça (5) and Carrasca (6) are located in the Moura–Ficalho subzone belt. The Zn–Pb Preguiça mineralization is associated with certain horizons of the Early Cambrian dolomites. Carrasca, a gossan of unknown origin, seems to be related to intermediate acid metavolcanics associated with dolomitic formations.

The multivariate method, to be applied in the sequel, is aimed at testing the following environmental classification of gossans, suggested by geological evidence: carbonate environmental formation for Preguiça and mainly silicate for the other gossans.

SAMPLING AND ANALYTICAL TECHNIQUES

A total of 146 samples were collected at the 6 base-metal gossan sites (Fig. 1). In well-exposed gossans, a 2-kg aggregate of sample chips was collected from individual 1-m^2 sites, along several traverses. The distance between traverses was adjusted according to outcrop size and regularity. In areas of poor exposure, available outcrops were sampled across and along strike. The number of gossan samples examined by petrographic, geochemical, mineralogical and electron microprobe techniques represents approximatively 72% of the total samples collected.

Geochemical samples were crushed, split and ground in an agate disc mill to less than 200 mesh (74 μm). The samples were analysed for Cu, Pb, Zn, Mn, Co, Ni, Ag, Mo, As, Cr, P, B, V, Ba, Sb, and Cd by DCP emission spectrometry following an $HF–HNO_3–HClO_4$ digestion. Iron was analyzed by titrimetry and Ca and Mg by AAS following an $HF–HClO_4$ dissolution. Analytical precision, defined as the percent relative variation at the 95% level, ranges from 8 to 15%.

The mineralogy of each sample was determined by XRD using Cu k-radiation and by reflected light microscopy. An EDS-electron microprobe study was also carried out as an aid to mineral identification.

Geochemical and microprobe analyses were conducted in the laboratory of Direcção Geral de Geologia e Minas.

MINERALOGY OF GOSSANS

The composition of the primary sulphide ore and the progressive geochemical changes during supergene alteration play a fundamental role on the development of gossans. The sequence of sulphide oxidation and the geochemical environment in the oxide zone control, to a large extent, the geochemical development of base-metal gossans (Andrew, 1980).

In such a complex geochemical environment, it is not expected that a single factor, such as the sulphide-oxide profile, can account for the ultimate characteristics of a gossan. In fact, a complex interplay of factors, such as composition, buffering qualities of gangue and groundwater composition, must be considered as responsible for gossan formation (Andrew, 1977).

Arronches–Campo Maior belt

Gossans overlying the Arronches–Campo Maior belt ores are associated with stratiform Cu deposits, with minor occurrences of Zn and Pb. The mineralogical composition of gossans is mainly goethite and hematite, whilst the accessory components are magnetite, barite, gahnite, pyrite, calcopyrite, malachite and quartz. Barite and gahnite were not detected in the Fraustus and Monte Azeiteiros gossans, but only at Tinoca.

Fe hydroxides (goethite and probably hydrogoethite) are poorly crystallyzed. The original sulphide texture is partially obliterated, but some replica textures ("boxworks", massive pseudomorphs) and residues are observed.

Hematite is associated with goethite and results mainly from magnetite alteration (martitization).

Sousel–Barrancos belt

Gossans in Defesa das Mercês have an unknown origin. Disseminated Cu mineralization occurs in the area. Its mineralogical composition is mainly goethite and hematite, whilst the accessory minerals are psilomelane, pyrolusite, pyrite and quartz.

Fe hydroxides (goethite and probably hydrogoethite) are poorly crystallized. Hematite, less abundant than goethite, occurs mainly in zoned textures, associated with the latter.

Moura–Ficalho belt

The gossan overlying Preguiça is associated with a polymetallic sulphide ore. The mineralogical composition of the gossan is mainly hematite, goethite, smithsonite, calcite and dolomite, whilst the accessory mineralogical components are lepidocrocite, Mn oxides, pyrite, sphalerite and calamine. Hematite is more abundant than goethite, often forming microcrystalline aggregates and rhythmic banding. According to Wilhelm and Kasakevitch (1979), this hematite–goethite association, with occurrence of lepidocrocite, supports development from a carbonate environment.

The Carrasca gossan is of unknown origin. It is associated with acid metavolcanics. The mineralogical components of the gossans are mainly hematite, goethite, magnetite and Mn oxides, whilst the accessory mineralogical components are barite and quartz. The most abundant Fe oxide is hematite, often associated with goethite, which shows colomorphous and fibrous forms.

STATISTICAL ANALYSIS OF DATA

The geochemical data set consists on a series of 6 matrices (one for each gossan) of n samples ($n=22$ for Tinoca, $n=11$ for Monte Azeiteiros, $n=10$ for Fraustus, $n=17$ for Defesa das Mêrces, $n=21$ for Preguiça, $n=21$ for Carrasca) and p variables ($p=19$, concentrations in Fe, Ca, Mg, Ba, P, Cu, Cr, Ag, B, Zn, Sb, Pb, Ni, V, Mn, Mo, As, Co, Cd).

Univariate data analysis

In order to visualize and summarize the available data in each gossan, box plots of variables were constructed, in the frame of Exploratory Data Analysis (Tukey, 1977).

For each variable, the following univariate statistical measures were calcu-

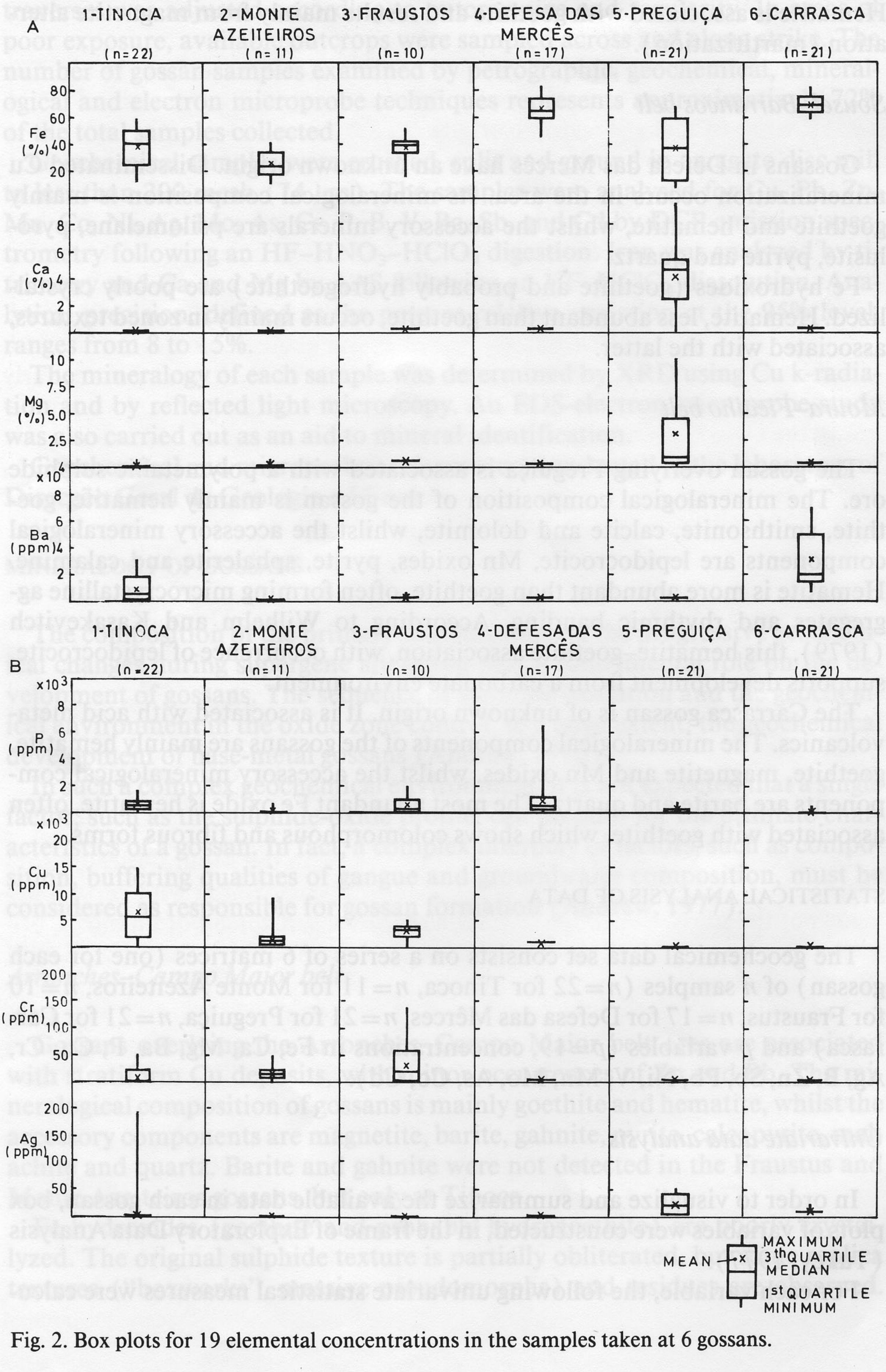
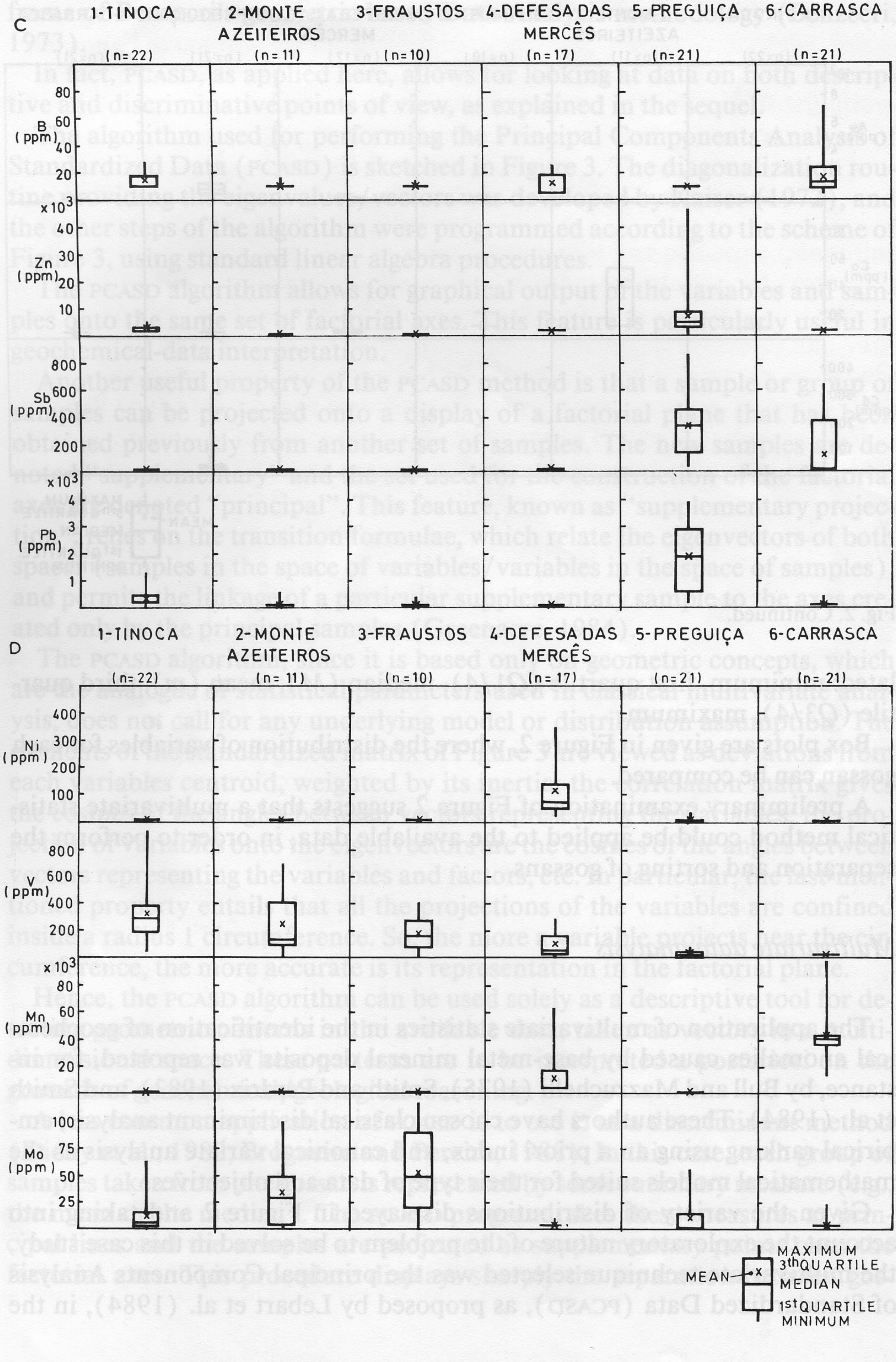


Fig. 2. Box plots for 19 elemental concentrations in the samples taken at 6 gossans.


C
1-TINOCA
2-MONTE AZEITEIROS
3-FRAUSTOS
4-DEFESA DAS MERCÊS
5-PREGUIÇA
6-CARRASCA
(n=22)
(n=11)
(n=10)
(n=17)
(n=21)
(n=21)
B (ppm)
Zn (ppm)
x10³
Sb (ppm)
Pb (ppm)
x10³
D
1-TINOCA
2-MONTE AZEITEIROS
3-FRAUSTOS
4-DEFESA DAS MERCÊS
5-PREGUIÇA
6-CARRASCA
(n=22)
(n=11)
(n=10)
(n=17)
(n=21)
(n=21)
Ni (ppm)
V (ppm)
x10³
Mn (ppm)
Mo (ppm)
MAXIMUM
3thQUARTILE
MEAN
MEDIAN
1stQUARTILE
MINIMUM

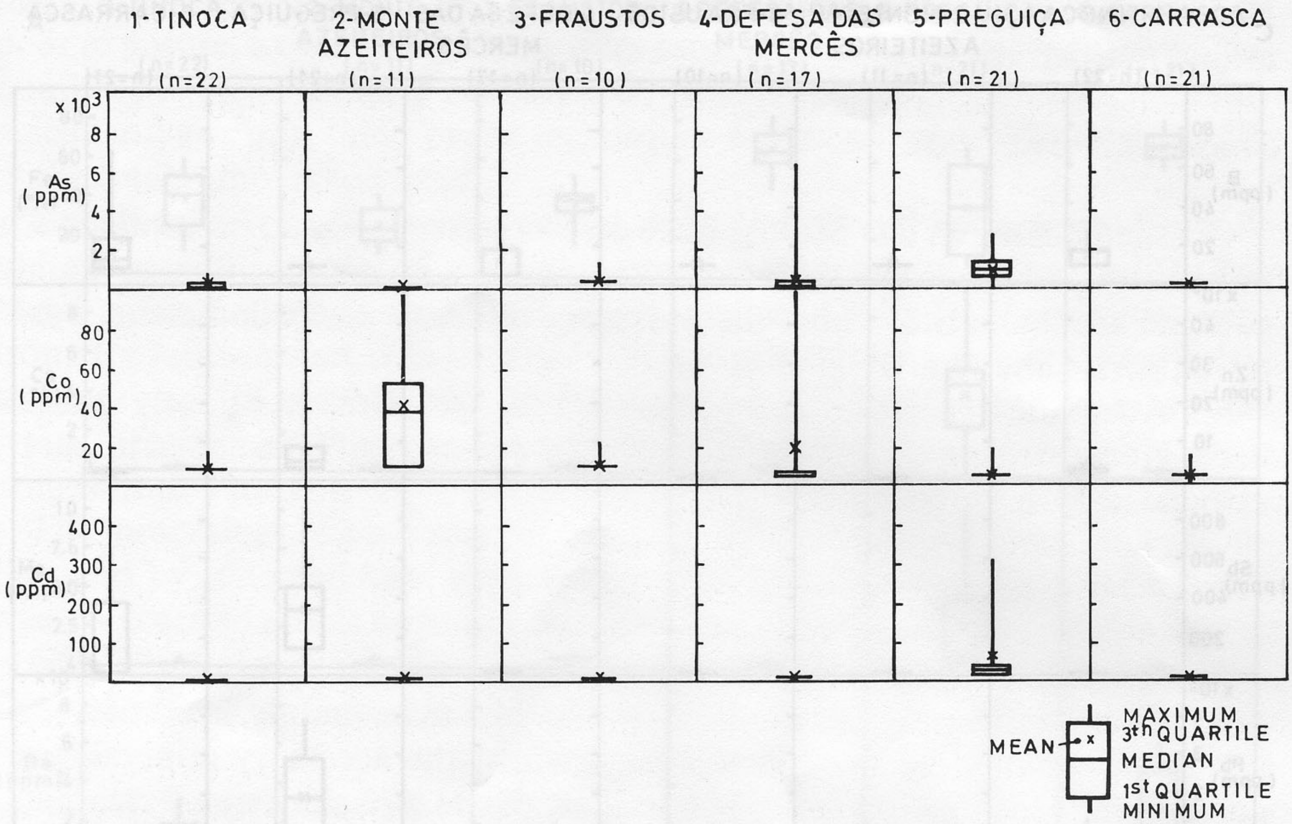


Fig. 2. Continued.

lated: minimum, first quartile ($Q1/4$), median (M), mean (m), third quartile ($Q3/4$), maximum.

Box plots are given in Figure 2, where the distribution of variables for each gossan can be compared.

A preliminary examination of Figure 2 suggests that a multivariate statistical method could be applied to the available data, in order to perform the separation and sorting of gossans.

Multivariate data analysis

The application of multivariate statistics in the identification of geochemical anomalies caused by base-metal mineral deposits was reported, for instance, by Bull and Mazzuchelli (1975), Smith and Perdrix (1983), and Smith et al. (1984). These authors have chosen classical discriminant analysis, empirical ranking using an a priori index, and canonical variate analysis as the mathematical models suited for their type of data and objectives.

Given the variety of distributions displayed in Figure 2 and taking into account the exploratory nature of the problem to be solved in this case study, the multivariate technique selected was the principal Components Analysis of Standardized Data (PCASD), as proposed by Lebart et al. (1984), in the frame of Benzecri's geometric based Data Analysis methodology (Benzecri, 1973).

In fact, PCASD, as applied here, allows for looking at data on both descriptive and discriminative points of view, as explained in the sequel.

The algorithm used for performing the Principal Components Analysis of Standardized Data (PCASD) is sketched in Figure 3. The diagonalization routine providing the eigenvalues/vectors was developed by Kaiser (1972), and the other steps of the algorithm were programmed according to the scheme of Figure 3, using standard linear algebra procedures.

The PCASD algorithm allows for graphical output of the variables and samples onto the same set of factorial axes. This feature is particularly useful in geochemical-data interpretation.

Another useful property of the PCASD method is that a sample or group of samples can be projected onto a display of a factorial plane that has been obtained previously from another set of samples. The new samples are denoted "supplementary" and the set used for the construction of the factorial axes are denoted "principal". This feature, known as "supplementary projection", relies on the transition formulae, which relate the eigenvectors of both spaces (samples in the space of variables/variables in the space of samples), and permits the linkage of a particular supplementary sample to the axes created only by the principal samples (Greenacre, 1984).

The PCASD algorithm, since it is based only on geometric concepts, which are the analogue of statistical parameters used in classical multivariate analysis, does not call for any underlying model or distribution assumption. The elements of the standardized matrix of Figure 3 are viewed as deviations from each variables centroid, weighted by its inertia; the correlation matrix gives the cosines of the angles between vectors representing the variables; the projection of variables onto the eigenvectors are the cosines of the angles between vectors representing the variables and factors, etc. In particular, the last-mentioned property entails that all the projections of the variables are confined inside a radius 1 circumference. So, the more a variable projects near the circumference, the more accurate is its representation in the factorial plane.

Hence, the PCASD algorithm can be used solely as a descriptive tool for detecting geometric patterns in the available data, taken as vectors in a multidimensional space. These patterns are to be interpreted a posteriori on the grounds of geochemical/geological reasoning.

A less common application of PCASD is to use it as a discriminant method (Diday et al., 1982; Brogueira and Pereira, 1988). In this case, each group of samples taken from one gossan is represented by some summary measure (e.g., the mean or the median). The PCASD program takes these measures as principal lines and the samples are projected as supplementary points onto the factorial axes. This procedure displays sharply the separation between gos-

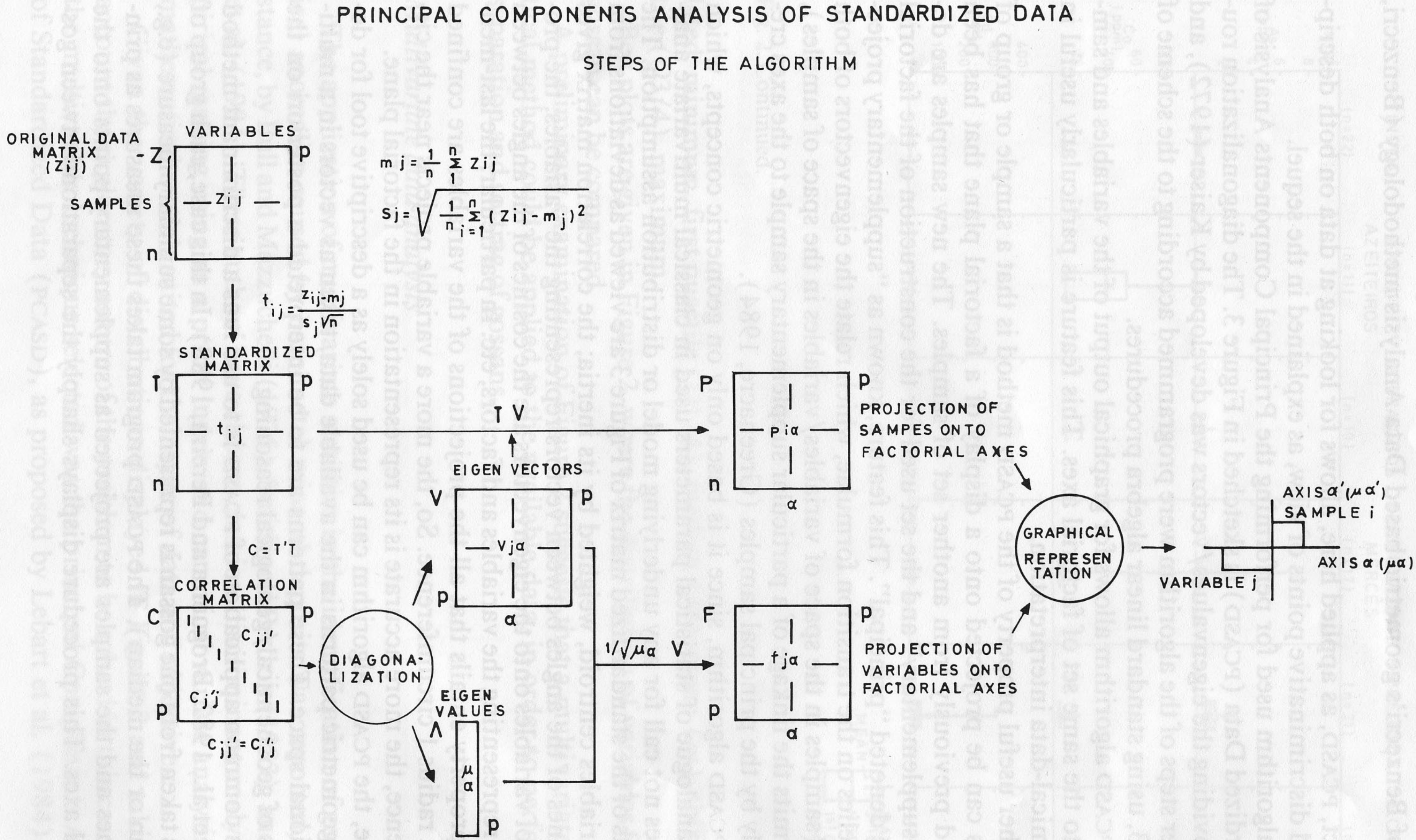


Fig. 3. Scheme of the algorithm of the Principal Components Analysis of Standardized Data technique.

sans, emphasizing their differences and showing which variables are to be interpreted as controlling the discrimination.

When PCASD is applied, inputing as principal lines some summary measure of each gossan, this technique is a powerful tool for achieving a distribution-free geometric-based discrimination.

Results of PCASD based on global data

A first application of PCASD as a descriptive tool was performed, inputing a matrix of 102 principal lines (samples), 6 supplementary lines (centroids of each gossan) and 19 columns (concentrations). The results of this experiment are given in Figure 4, where projections of samples, centroids and variables are displayed in the plane defined by axes 1 and 2, which were constructed on the basis of the whole set of unstructured samples. Even though a clear separation of Preguiça gossan is apparent in Figure 4, it was decided to repeat the analysis using as input a matrix containing 6 principal lines (the

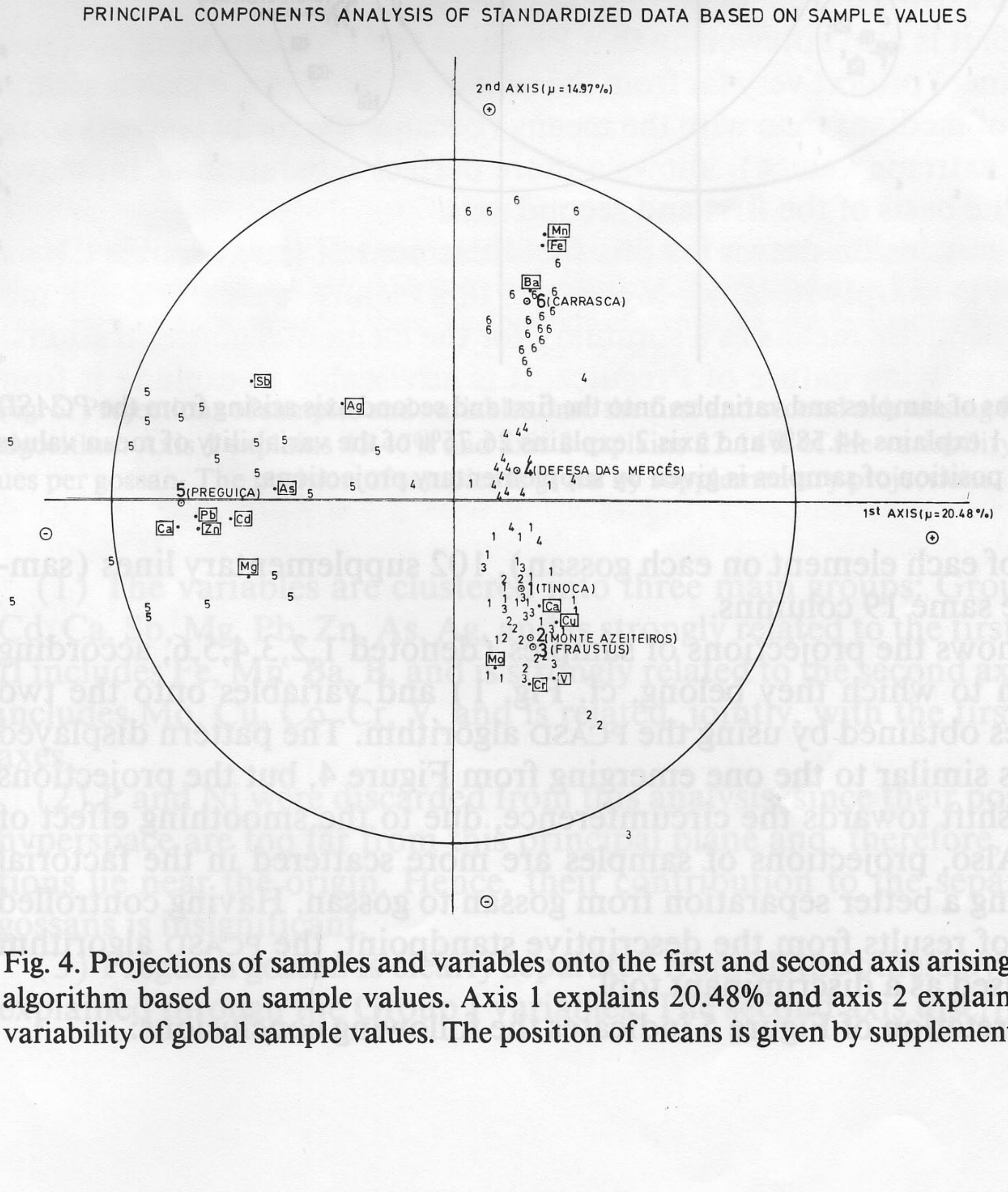


Fig. 4. Projections of samples and variables onto the first and second axis arising from the PCASD algorithm based on sample values. Axis 1 explains 20.48% and axis 2 explains 14.97% of the variability of global sample values. The position of means is given by supplementary projections.

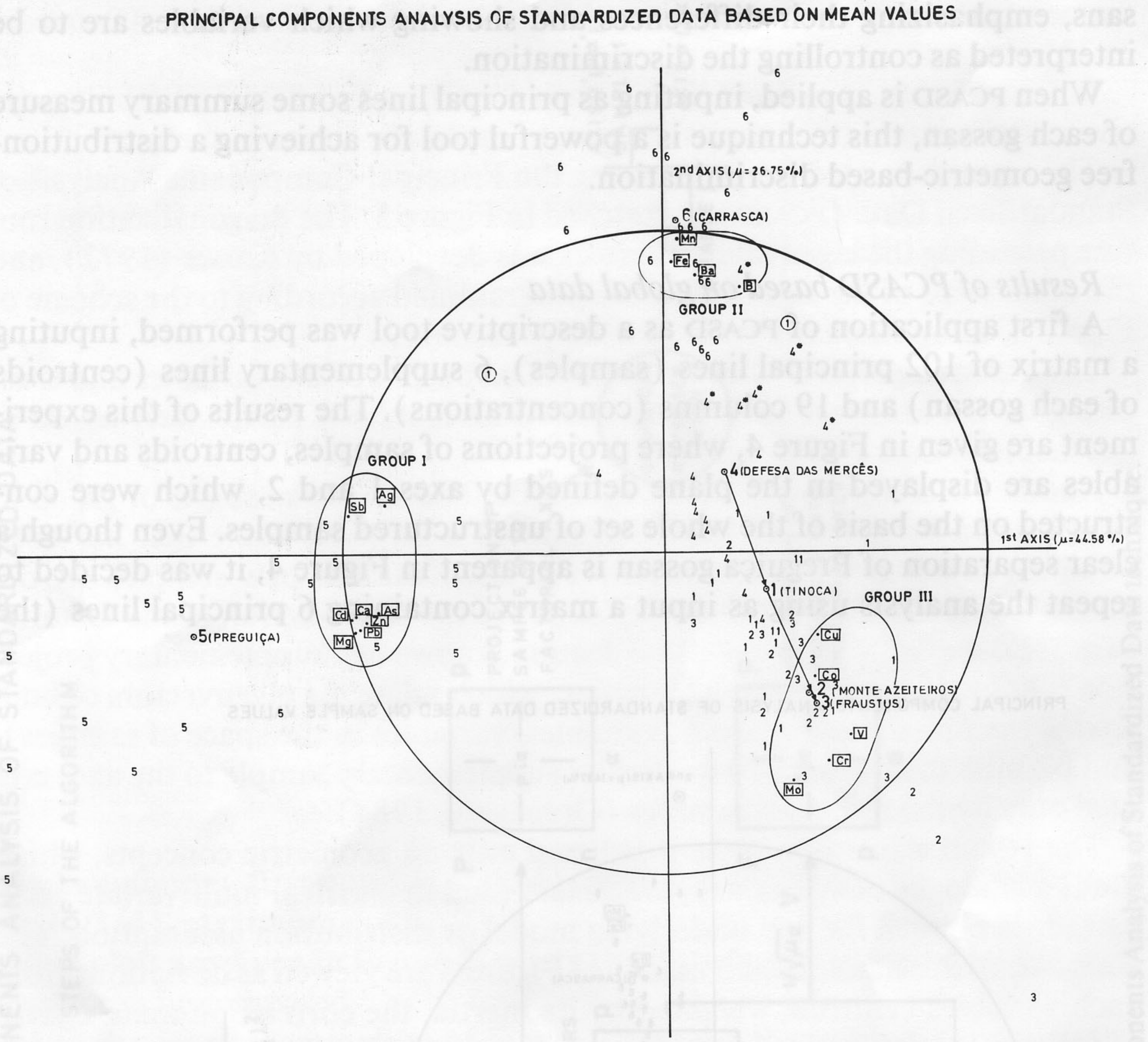


Fig. 5. Projections of samples and variables onto the first and second axis arising from the *PCASD* algorithm. Axis 1 explains 44.58% and axis 2 explains 26.75% of the variability of mean values per gossan. The position of samples is given by supplementary projections.

mean value of each element on each gossan), 102 supplementary lines (samples) and the same 19 columns.

Figure 5 shows the projections of samples (denoted 1,2,3,4,5,6, according to the gossan to which they belong, cf. Fig. 1) and variables onto the two principal axes obtained by using the PCASD algorithm. The pattern displayed in Figure 5 is similar to the one emerging from Figure 4, but the projections of variables shift towards the circumference, due to the smoothing effect of the means. Also, projections of samples are more scattered in the factorial plane, allowing a better separation from gossan to gossan. Having controlled the stability of results from the descriptive standpoint, the PCASD algorithm can now be used as a discriminant tool.

The interpretation of Figure 5 indicates the following conclusions:

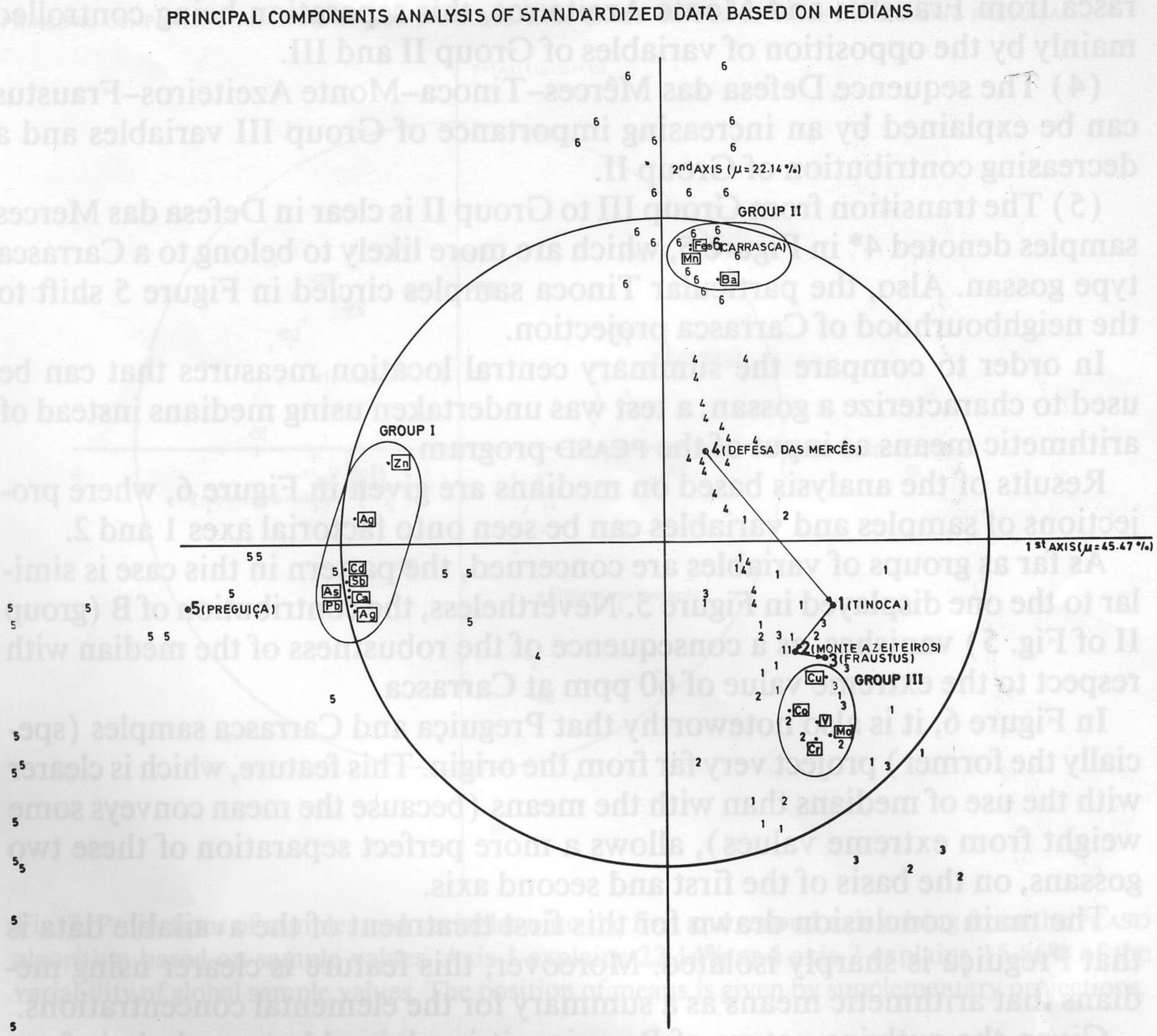


Fig. 6. Projections of samples and variables onto the first and second axis arising from the *PCASD* algorithm. Axis 1 explains 45.47% and axis 2 explains 22.14% of the variability of median values per gossan. The position of samples is given by supplementary projections.

(1) The variables are clustered into three main groups: Group I includes Cd, Ca, Sb, Mg, Pb, Zn, As, Ag, and is strongly related to the first axis. Group II includes Fe, Mn, Ba, B, and is strongly related to the second axis. Group III includes Mo, Cu, Co, Cr, V, and is related, jointly, with the first and second axes.

(2) P and Ni were discarded from this analysis, since their positions in the hyperspace are too far from this principal plane and, therefore, their projections lie near the origin. Hence, their contribution to the separation of the gossans is insignificant.

(3) Preguiça gossan is clearly separated from all the others by the first axis, explained through the Group I variables. The second axis discriminates Car-

rasca from Fraustus and Monte Azeiteiros, this separation being controlled mainly by the opposition of variables of Group II and III.

(4) The sequence Defesa das Mêrces–Tinoca–Monte Azeiteiros–Fraustus can be explained by an increasing importance of Group III variables and a decreasing contribution of Group II.

(5) The transition from Group III to Group II is clear in Defesa das Merces samples denoted 4* in Figure 5, which are more likely to belong to a Carrasca type gossan. Also, the particular Tinoca samples circled in Figure 5 shift to the neighbourhood of Carrasca projection.

In order to compare the summary central location measures that can be used to characterize a gossan, a test was undertaken using medians instead of arithmetic means as input of the PCASD program.

Results of the analysis based on medians are given in Figure 6, where projections of samples and variables can be seen onto factorial axes 1 and 2.

As far as groups of variables are concerned, the pattern in this case is similar to the one displayed in Figure 5. Nevertheless, the contribution of B (group II of Fig. 5) vanishes, as a consequence of the robustness of the median with respect to the extreme value of 60 ppm at Carrasca.

In Figure 6, it is also noteworthy that Preguiça and Carrasca samples (specially the former) project very far from the origin. This feature, which is clearer with the use of medians than with the means (because the mean conveys some weight from extreme values), allows a more perfect separation of these two gossans, on the basis of the first and second axis.

The main conclusion drawn for this first treatment of the available data is that Preguiça is sharply isolated. Moreover, this feature is clearer using medians that arithmetic means as a summary for the elemental concentrations.

Given the outlying nature of Preguiça, it is adviseable to exclude it from the data analysis, in order to focus on the more subtle differences between the other gossans.

Results of PCASD excluding Preguiça

Results of PCASD based on the set of samples excluding Preguiça are given in Figure 7. The pattern displayed in Figure 7 is different from the one obtained by the global analysis (Fig. 4). In fact, Axis 1 separates the group Tinoca+Fraustus+Monte Azeiteiros from Carrasca, the latter being associated with Sb, Mn, Ca, Cd, Fe and Ba. The former group is explained mainly by Zn, Pb, Cu, V, Cr and Mo. The second axis isolates Defesa das Mêrces, which is controlled mainly by As and Ni (P, Mg and B were discarded).

In Figure 8 a similar pattern was obtained inputing the means of each gossan as principal lines to the PCASD program. The major differences from Figure 7 is that variables are scattered around the circumference and the Tinoca is isolated by axis 2.

PRINCIPAL COMPONENTS ANALYSIS OF STANDARDIZED DATA BASED ON SAMPLE VALUES (EXCLUDING FREGUIÇA)

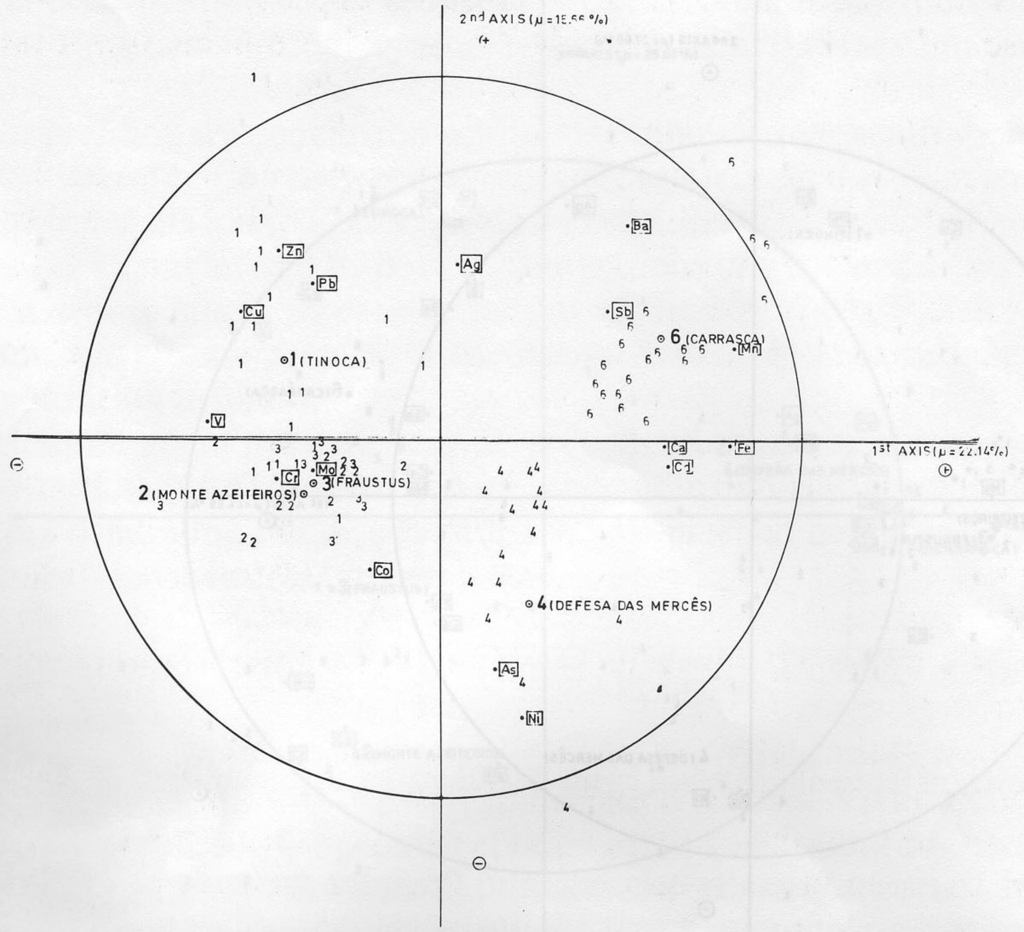


Fig. 7. Projections of samples and variables onto the first and second axis arising from the PCASD algorithm based on sample values. Axis 1 explains 22.14% and axis 2 explains 15.66% of the variability of global sample values. The position of means is given by supplementary projections.

In Figure 8, the circular counter-clockwise sequence of variables and gossans is as follows:

(a) Ag, Zn, Pb, Cu – linked to the positive second axis and to samples 1 (Tinoca), exception made to samples 1*, which are Monte Azeiteiros type;

(b) V – linked both to the first and second axes, and controlling, jointly, Tinoca and Monte Azeiteiros+Fraustus;

(c) Cr, Mo – linked to the negative first axis, and controlling strongly Monte Azeiteiros+Fraustus;

(d) Co – linked both to the first and second axes, and making the transition from Monte Azeiteiros+Fraustus to Defesa das Mêrces;

(e) As, Ni – linked to the negative second axis, and controlling strongly Defesa das Mêrces;

(f) Cd, Fe, Mn, Ca, Sb, Ba, B – linked to the negative first axis, and controlling Carrasca.

Figure 9 shows the results obtained using medians instead of means as the

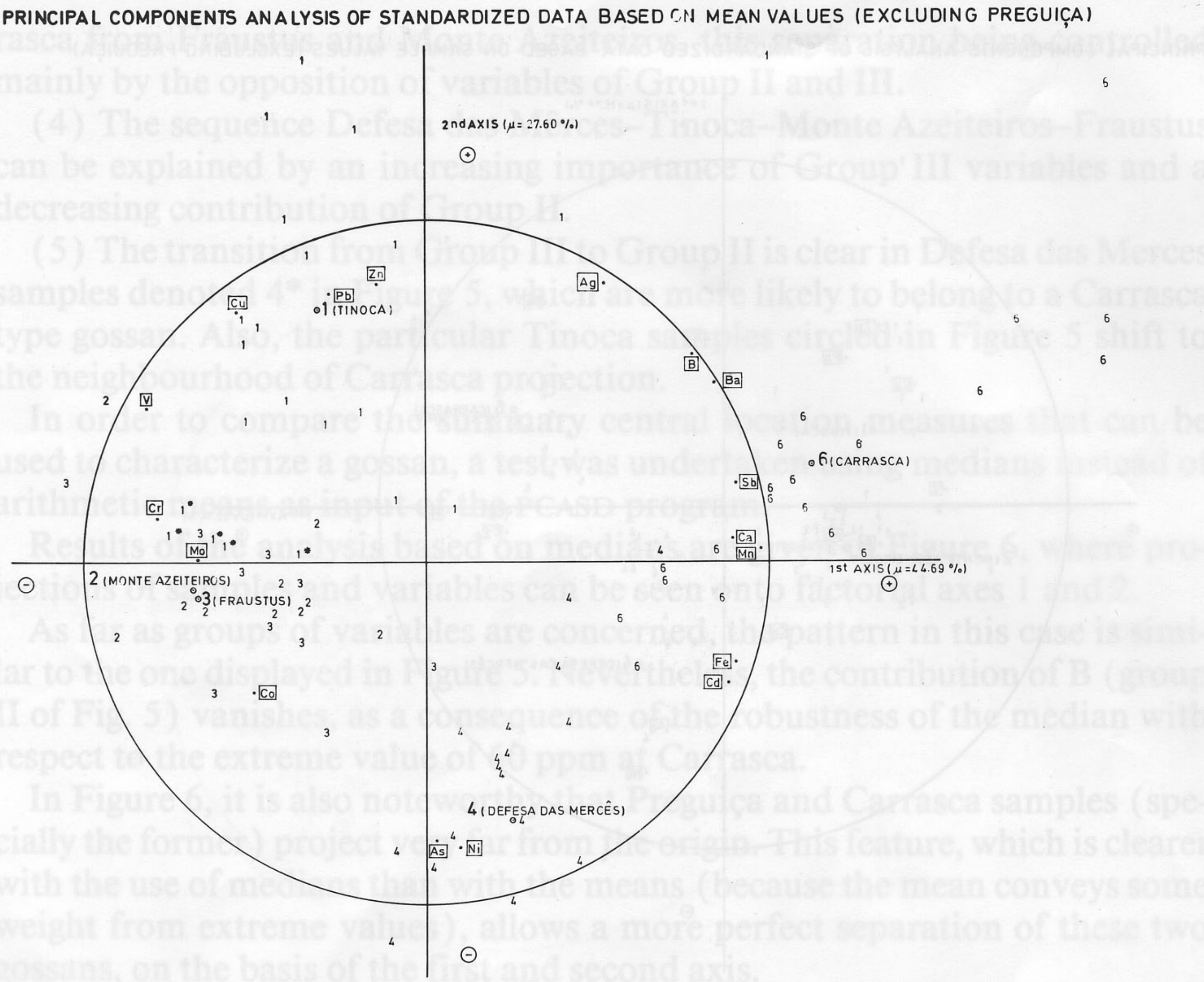


Fig. 8. Projections of samples and variables onto the first and second axis arising from the PCASD algorithm, excluding Preguiça. Axis 1 explains 44.69% and axis 2 explains 27.60% of the variability of mean values per gossan. The position of samples is given by supplementary projections.s

principal lines of the input matrix. The variables B, Ni and As are disregarded in this case, since they project near the origin.

The pattern displayed in Figure 9 can be interpreted as follows:

(a) The contribution of Ni and As, which was important for the means, vanishes when medians are used. This feature explains how Defesa das Mêrces shifts towards Carrasca.

(b) The contrary contribution of P and Mg is apparent, in the case of medians, sharply separating Tinoca (P influence) from Monte Azeiteiros (Mg influence);

(c) The first axis separates Tinoca+Fraustus+Monte Azeiteiros from Carrasca+Defesa das Mêrces. Furthermore, Defesa das Mêrces separates clearly from Carrasca, since the former projects onto the first axis in the interval 0.2/0.6 and all Carrasca samples have a score greater than 0.9. The

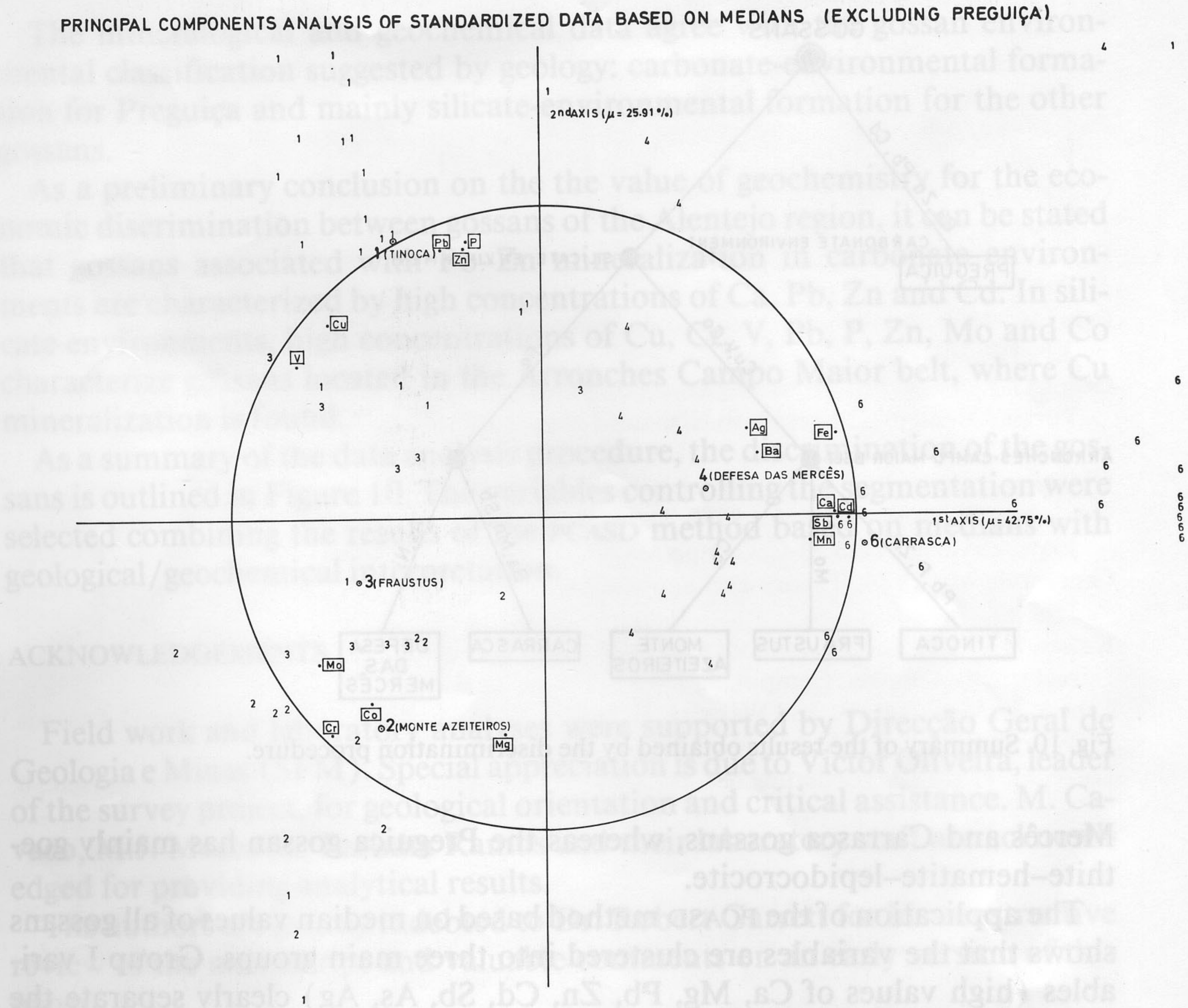


Fig. 9. Projections of samples and variables onto the first and second axis arising from the PCASD algorithm, excluding Preguiça. Axis 1 explains 42.75% and axis 2 explains 25.91% of the variability of median values per gossan. The position of samples is given by supplementary projections.

variables controlling this discrimination are mainly Sb and Cd (projection onto the first axis of 0.97), Ca (0.93) and Mn (0.85).

(d) Separation from Tinoca to Monte Azeiteiros can be seen along the second axis (Fraustus takes an intermediate position). P, Zn and Pb are linked to Tinoca (projections onto the second axis of 0.88, 0.86, 0.86, respectively) and Mg, Cr and Co control, to a lower level, Monte Azeiteiros (projections of −0.69, −0.69, −0.60, respectively).

CONCLUSIONS

The mineralogical study shows that goethite and hematite are the principal iron oxide minerals of the Tinoca, Fraustus, Monte Azeiteiros, Defesa das

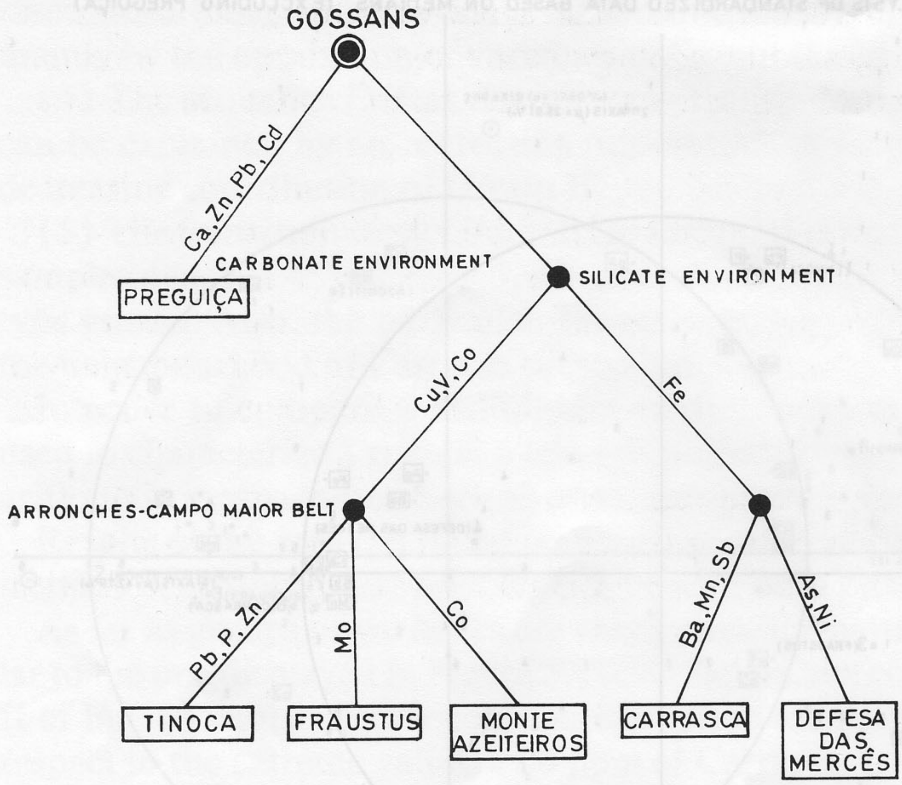


Fig. 10. Summary of the results obtained by the discrimination procedure.

Mercês and Carrasca gossans, whereas the Preguiça gossan has mainly goethite–hematite–lepidocrocite.

The application of the PCASD method based on median values of all gossans shows that the variables are clustered into three main groups. Group I variables (high values of Ca, Mg, Pb, Zn, Cd, Sb, As, Ag) clearly separate the Preguiça gossan (carbonate environment with Pb–Zn mineralization) from all the others (mainly silicate environments with Cu mineralization).

Excluding Preguiça, the application of the PCASD method based on mean values to the silicate-environment gossans shows that the variables are not clustered, but scattered around the correlation 1 circumference. The use of medians instead of means permits the separation of Tinoca+Fraustus +Monte Azeiteiros from Carrasca+Defesa das Mercês. This separation is controlled by the following variables: Ca, Fe, Mn, Sb and Ag. Tinoca+Fraustus+Monte Azeiteiros belong to a clearly alumino-silicate environment, whilst Carrasca+Defesa das Mercês probably represent an intermediate type. Furthermore, the Carrasca gossan, overlying acid metavolcanics with adjacent Cambrian dolomitic lithology, is separated by Ca, Mn, Sb, Cd from Defesa das Mercês gossan, located in Hercynian acid sub-volcanic breccias with carbonate cement and Silurian schists. The separation of Monte Azeiteiros from Tinoca, with Fraustus in an intermediate position, us supported by an increasing importance of Pb, Zn, P and Cu (in the Tinoca sense), and can be explained by underlying sulfide enrichment.

The mineralogical and geochemical data agree with the gossan environmental classification suggested by geology: carbonate-environmental formation for Preguiça and mainly silicate-environmental formation for the other gossans.

As a preliminary conclusion on the the value of geochemistry for the economic discrimination between gossans of the Alentejo region, it can be stated that gossans associated with Pb–Zn mineralization in carbonate environments are characterized by high concentrations of Ca, Pb, Zn and Cd. In silicate environments, high concentrations of Cu, Cr, V, Pb, P, Zn, Mo and Co characterize gossans located in the Arronches Campo Maior belt, where Cu mineralization is found.

As a summary of the data analysis procedure, the discrimination of the gossans is outlined in Figure 10. The variables controlling the segmentation were selected combining the results of the PCASD method based on medians with geological/geochemical interpretation.

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