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Statistical analyses of grain-size, geochemical and mineralogical data in core CM92-43, Central Adriatic basin.

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ABSTRACT

A statistical strategy feasible for paleoenviromental studies is described. We present some examples of multivariate methods (principal component, factor and cluster analyses) on chemical, sedimentological and mineralogical data, from a core (CM92-43) collected in the Mid Adriatic Deep (MAD). This core, raised in 250 m water depth, provides a continuous expanded record of the late-Quaternary sea level rise and highstand. Problems related to peculiar characteristiscs of the data set (the presence of unusual sporadic events and the constant sum) are discussed, and some solutions are presented. Specific patterns resulting from the multivariate statistics are compared to independent results, indicating good correspondance to specific climatic events recorded by other proxies (age boundaries, chemostratigraphy, foraminifera ecozones, palynology)

Key words: multivariate statistical analyses, grain size, sediment core, late-Quaternary, Adriatic Sea

1. INTRODUCTION

Multiproxy records and age depth models of core CM92/43 have been extensively described in several papers of this issue (Trincardi *et al.* 1996; Asioli; Langone *et al.*; Artitzegui *et al.*; Lowe *et al.*; this volume all). We present here new data on grain-size and mineralogy, as well as a statistical analysis of this and other data (magnetic minerals, organic matter). The goal of this paper is to show an example of multivariate statistical approach to compositional and geochemical data, in order to highlight patterns of paleoenvironmental variability and compare them to specific events (age boundaries, chemostratigraphy, record of stratigraphic events, foraminifera ecozones and pollen zones) recorded in the core.

Time series obtained from sediment cores present special interpretation difficulties, principally because they record multivariate non-stationary, non-linear processes that are sampled non-uniformly in time (Meeker *et al.* 1996). In this paper we focus on the lithological and chemical records developed from a sediment core. These

particle sizes and chemical species are selectively deposited in the Mid Adriatic Deep (MAD) by a variety of processes where, when sampled, they become accessible through chemical and statistical analyses. Such analyses are complicated by the existence of multiple sources for most chemical species and minerals, and the variety of modes of chemical and biological evolution possible during the marine transport from source to deposition site and, in some instance, post-depositional transformation or diffusion within the sediment matrix.

In this study, a particular strategy has been used for the analysis of a set of subsamples, taken every 10 cm in a marine muddy section that spans approximately the last 16,000 calibrated years (Langone *et al.* 1996, this volume). This core provides the most expanded continuous marine record for the late-Quaternarypost glacial interval in the Adriatic region (Trincardi *et al.* 1996, this volume). Well known techniques of cluster, discriminant and factor analyses were used. More general stratigraphic aspects of the data are presented in comprehensive studies by Asioli 1996; Langone *et al.*; Artitzegui *et al.*; Lowe *et al.* (this volume all) This paper aims at the identification and, if possible, the statistical discrimination and description of the contributions of the different sources composing the record.

2. DESCRIPTION OF THE DATA

2.1. Analytical methods

Grain-size analyses were done on sample pretreated with H_2O_2 to eliminate organic material, sieved at 63 µm and weighed. On the fraction < 63 µm the determination were done by means of a laser Galai CIS 1 instrument, with a specific analytical size intervals of 1 micron. The size distribution of insoluble particles in rain was analysed by Galai Cis 1 technique. The grain size measurement was based on the relationship between the transition time of particles moving in a photodefined zone and their size. A focused laser beam scans an area of 600 µm diameter with a beam size of 1.2 µm; when a particle is detected a photodiode produces a signal proportional to the size of the particle. This device covers a range from 0.5 to 600 µm of particle diameters. Granulometric parameters used here, were expressed in the following intervals: 0.5-1, 1-2, 2-4, 4-8, 8-16, 16-63, >63 µm. A detailed description of the method was made by Molinaroli & De Falco (1995).

X-ray diffraction was used to obtain clay mineral data, and was performed on the sediment fraction $< 2\mu m$. Semi-quantitative calculations of smectite, illite, clorite and kaolinite followed the method suggested by Biscaye (1965). Chemical analyses of Ca and Mg were done with ionic chromatography on the hydrochloric acid leachate of the samples, and then calcite and dolomite fractions were calculated. The assumption is that all the Mg come from dolomite; then the part of calcium related to dolomite is subtracted fom the total Ca, and calcite concentration is calculated.

TOC and magnetic determinations are described elsewhere (Langone *et al.* 1996; Alvisi & Vigliotti 1996, this volume)

3. STATISTICAL STRATEGY

The following steps summarise the strategy employed in the study:

(i) data are transformed in rank, for the representation of true outliers;

(ii) cluster analysis is used to identify some clusters without an "a priori" criterion for subdividing the samples into groups;

(iii) cluster analysis of the variables is also performed to visualize groupings within the variables and their redundance, partly related to the constant sum problem of grain-size and mineralogical data (see discussion below);

(iv) by studying the results of clustering, a stratigraphically meaningful number of groups of samples is choosen;

(v) with an "a priori" criterion, i.e. the results of cluster analysis, discriminant analysis is employed to characterise the groups of samples in relation to the variables that have greater discriminating power;

(vi) factor analysis (*q-mode*) is used to reduce a large number of variables to a few uncorrelated variables (factors);

(vii) a variation diagram of the factors is constructed to follow their relative importance downcore.

This computational and statistical strategy permit to proceed gradually from simple data analysis to more sophisticated pattern recognition.

3.1 Ranks of the data values

The analysis of sediment cores collected on clastic continental margins is not a routine because of two main factors: (i) sampling is not uniform in time (when a sediment core is sampled at uniform depth intervals, differential rates of accumulation and post-depositional compaction result in sampling intervals that are non-uniform in time); and (ii) exogenous factors (stocastic events that are not related to the basin evolution, such as tephra layers originated in nearby regions). The primary goal for studying a sediment core is an understanding of paleoclimate and the processes which played a principle role in its evolution. This analysis is made more difficult when the record includes sporadic and, occasionally, dramatically large events of both climate-related (e.g. unusual river floods) and non-climate origin (e.g. volcanic).

For this reason we used robust procedures that are much more resistant to the presence of outliers. In particular, all calculations in the analysis and multivariate statistics were based only on ranks of the data values: each value is replaced by a number giving its place in the sequence from highest to lowest value or vice-versa (Swan & Sandilands 1995). Ranking is a non-parametric procedure which requires very limited assumptions about the underlying distribution. Furthermore, the rank transformation is a suitable method for the representation of true outliers and whenever standardisation is needed. It is also suitable when there is reason to suspect that the data are not measured on a nice interval scale or do not have the kind of distribution required for many statistic techniques.

Another possible source of error is the fact that grain-size and mineralogical data suffer of the problem of the constant sum, i.e., components of data expressed as percentage are not free to vary independently. As the weight of one component increases, the proportion of one or more other components must decrease, and it is inevitable that induced correlation will result. The principal consequence of closure for geochemistry is that correlation can thus produce misleading results (Hugh 1993). This observation has important implications if we are looking for real geological trends that can be interpreted as paleoenvironmental indicators.

4. RESULTS

Grain size data show that most of the core is composed of mud, the mean fraction > 63 μ m being around 1%. Average median diameter is 3.8 μ m, and the values range from \approx 3 μ m in the Holocene part of the core, to \approx 5 μ m at the bottom of the core (full glacial). Mean clay mineral fraction (<2 μ m) is 25%, and the range is 20-35%, with a decreasing trend toward the bottom of the core (Fig. 1).

Carbonate fraction accounts for $\approx 35\%$, with mean calcite values of 23% and little variations with depth in core (Fig. 1), whereas dolomite values increase significantly, from 6% in the top layers to more than 15% in the lower 3 m of the core (avg. 12%).

Clay minerals show two distinct patterns: (i) smectite and kaolinite both decrease downcore, dramatically the smectite (from 30 to 5%, avg. 19%), and slightly the kaolinite (from 15 to 10%, avg 13%); (ii) illite and clorite show the opposite trend, increasing from 40 to 60% (avg. 49%), and from 14 to 28 % (avg. 19%) respectively (Fig. 1).

Those data essentially confirm the observations on other cores collected in the MAD (Calanchi *et al.* 1996, this volume), where major changes in sediment composition (carbonates, mineral supply) occur in correspondence of the most significant late-glacial to Holocene stratigraphic boundaries.

4.1 Cluster analysis

Cluster analysis is a classification more than a statistical procedure. Clusters are concentrations of points (the points being objects, observations or specimens) in space, and two points in the same cluster tend to be more similar than two points in different clusters. In other words, a group of objects which are classifiable together on numerical grounds will form a cluster of points in multivariate space.

To display the structure in our multivariate data we have used the Cluster analysis in "mode q" (results cluster objects on the basis of values of variables) and in "mode r" (the variables are clustered on the basis of their values in objects). Cluster analysis was performed using Ward's hierarchical agglomerative method and Euclidean distance measure (Ward 1963). The advantages of this technique are: (i) any coherent group will not split among different categories; (ii) the boundaries between clusters fall, by definition, in regions of multivariate space where there are few points; if this sudivision derives from aspects of the geological process, the boundaries would be "natural"; (iii) the methodology readily allows consideration of all variables.

Some disadvantages come from the instability introduced by the addition of observations to the analysis, that is likely to add new clusters and will inevitably redefine old ones.

Figure 2 shows the results of the cluster analysis in "*mode q*", applied to 92 sediments layers (every 10 cm) and 21 variables. The 21 variables consist of grain size intervals (0.5-1, 1-2, 2-4, 4-8, 8-16, 16-63, >63 microns), clay minerals (smectite, illite, caolinite and chlorite), calcite and dolomite, sediment accumulation rate (g cm⁻² yr⁻¹), and seven magnetic variables (Xfd, X, ARM, SIRM, SIRM/ARM, Karm/K, for details see Alvisi & Vigliotti 1996, this volume). These were clustered on the basis of their values in the different samples ("*mode r*"). The dendrogram shows how the different core levels cluster on the basis of their variable values.

Figure 2a shows the results of a cluster analysis performed on all the subsamples of core CM92-43. Samples are defined by 21variables and labelled by their calibrated

age (see Langone et al. 1996, this volume, for calibration procedures). The main result of this kind of representation is that, based on their similarity coefficient all samples fall in four main groups that have a clear stratigraphic meaning. The first cluster includes all samples younger than 6,000 calibrated years and corresponds to the distal high-stand record that rests above the maximum flooding surface (Trincardi et al. 1996, this, volume; Langone et al. 1996, this volume); this group corresponds to zone 1 of planktic forams (Asioli 1996, this volume). The second group includes all samples that are between 11,000 and 6,000 calibrated years and represents the uppermost of the three units that make up the transgressive record (TST) in the Adriatic basin (Trincardi et al. 1996, this volume); this cluster corresponds to zones 2, 3 and 4 of planktic forams (Asioli 1996, this volume). The third main cluster downcore includes the late glacial interval (Younger Dryas, Allerød and Bölling periods) down to about 13,400 calibrated years and corresponds to the middle unit of the TST (Trincardi et al. 1996, this volume; Langone et al. 1996, this volume), or zones 5 and 6 fo planktic forams (Asioli 1996, this volume). It is worth to point out that the base of this interval corresponds to the main spike in δ^{18} O depletion (Ariztegui *et al.* 1996, this volume), possibly associated to the first post-glacial meltwater pulse (Fairbanks 1989; Bard et al., 1995) or, perhaps, to a more local signal of the melting of Alpine glaciers. If this δ^{18} O spike in core CM92-43 is global, its age may not be in exact agreement with the age of the same event in other basins, because it depends on assumptions and possible errors in the age-depth model available for this core interval (see discussion in Langone et al. 1996, this volume). The fourth group of samples corresponds to the lower unit of the TST. This unit records the early, and lower-rate, interval of relative sea-level rise when fluvial sources were closer to the MAD and climatic conditions were the same as during full glacial times and corresponds to zone A of planktic forams (Asioli 1996, this volume). This stratigraphically-older unit is characterized by the highest sediment accumulation rates and therefore includes the highest number of samples (collected at constant intervals of 10 cm downcore) and a much higher degree of variability. In general, it is remarkable how all main clusters are composed of coherent sets of stratigraphycally-homogenous subsamples; furthermore, the way these samples (characterized by physical parameters and grain-size composition) are grouped has a clear stratigraphic meaning and corresponds to the main subdivisions based on totally independent biostratigraphy and high-resolution seismic stratigraphy (Asioli 1996, this volume; Lowe et al. 1996, this volume; Trincardi et al. 1996, this volume).

Main feature of Figure 2b is the clear subdivision of most of the variables into two groups, mainly related to fine (< 4μ m) and coarse (8-32 µm) grain-sizes. The method does not allow to map these groups as clusters, and could also be (better) investigated by PCA method. Two reasons explain the two groups: (i) specific surface area (SSA), i.e., some variables tend to be positively correlated with smaller particles (with a higher SSA), particularly TOC (Total Organic Carbon), X, ARM, smectite; and, (ii) hydrodynamics, i.e., the smaller particles deposit in the MAD (mainly during the Holocene), distance, and at that time also the distance of the sources was different. Therefore coarse grain-size means riverine input closer to the MAD and certain minerals (dolomite, kaolinite) and magnetic signatures more concentrated than later on.

4.2 Discriminant analysis

Discriminant analysis (DA) is used to distinguish statistically between two or more predefined groups of samples on the basis of multiple variables. The analysis contains tests for establishing the rate of success for discriminating variables when they are combined into a discriminating function. It contains criteria for controlling a stepwise selection of variables according to their discriminating power. These computations are described by Nie *et al.* (1975).

Here we have used the 4 groups of samples obtained by clustering (Fig. 2a), and the results of DA for the 92 samples are shown in figure 3, where the discriminant scores have been plotted for the three discriminant functions as a scattergram.

Table 1 summarises the results obtained. The variables with greater discriminating power are:

(i) ARM, dolomite, Karm/K, SIRM/ARM and smectite for function 1, that separates groups 1 and 2 (Holocene) from 3 and 4 (late Glacial);

(ii) SIRM, fraction >63 μ m and kaolinite for function 2, that separates group 3 (YD and BA) from 4;

(iii) X, accumulation rate and size range16-63 μ m, for function 3, that distinguishes group 1 (HST) from 2.

4.3 Principal component and Factor analysis

Principal Component Analysis (PCA) and Factor Analysis (FA) are suitable for finding the directions of maximum variance of the data, using these to ordinate data in one, two or three dimensions and interpreting them as factors influencing the data.

PCA is a technique for finding linear compounds of correlated variables, called principal components (PCs), that have two useful properties:

- 1. In general, most of the total variance of the p variables in a data set can be accounted for by a comparatively small number of k of the new variables
- 2. They are uncorrelated, which facilitates examinations and analysis of the data

FA is a technique for reducing a large number of variables to a few uncorrelated variables (Factors) so that a variation diagram may contain information about a large number of variables instead of the usual two or three. The method is well described in Le Maitre (1982) and Swan & Sandilands (1995). An original set of variables is transformed into a new set of variables called principal component co-ordinate (PCs). We can therefore represent the total variation in the data set by a small number (q) of geological factors, each manifested in a degree of correlation in the data scatter. In FA, we only find q vectors (whereas in PCA the number of PCs equals the number of variable, p), and it is found that the variance in those q directions can be maximised by rotating the axes away from the basic PC eigenvectors solution. Here we used the VARIMAX method, where orthogonality of the axes is preserved, and rotation of the axis system is attempted to account for as much variance as possible. The FA method has been criticised on the grounds that there is no clear criterion for the number of factors and for the search of rotation, to maximise the variance. Therefore subjectivity can result, and FA can be more of an interactive modelling exercise.

The time series derived from the sediment core chemical concentrations are not independent. Rather, the chemical signals are transported to the seabottom by water masses which contains chemical concentrations representative of their sources, mode of generation, and chemical reactivities. Furthermore, deposition processes occurring at a particular time may affect different chemical species in similar ways to introduce further dependencies among the individual records. Nevertheless, the "dissection" of sediment core chemistry provides records of temporal evolution of individual chemical and mineralogical elements rather than of the compounds of the water masses in which they were transported to the deposition sites. Therefore, considerable climatological information could be gained from an analysis which explores the variation and covariation of the individual series in order to determine relationships which reflect the various geochemical modes of production and transport. Such procedures attempt to "reassociate" chemical compounds and granulometric characteristics of particles which are, by necessity, disassociated by the analysis.

Q-mode factor analysis is used in our multivariate data-set (Joreskog *et al.*, 1976). Ranks of data values are used as input data. Principal Components as extraction method and Varimax normalised as Factor rotation. The results of the Q-FA set are presented in Table 2, where the composition of the factors in terms of loading of variables is listed; and in figure 4 where normalised factor scores are plotted with calibrated ages downcore. Three factors explained almost 93% of the total variability.

The description of the factor scores along the depth (age) in the core is as follows:

Factor 1 (F1, 42.5% of variance) is interpretable as the "Holocene" factor. In F1 variables that positively correlate with particles $< 4\mu m$ (with a higher specific surface area) are smectite, TOC, calcite, Xfd and ARM. The factor scores are maxima (avg. 85%) in the last 8,000 years, then decline between 8,000 and ,000 (50-75%), raise again, and decrease dramatically in the YD period, to get the minima of less than 10% at $\approx 14,500$.

Factor 2 (F2, 41.9%) is the "Glacial" factor, also interpretable as coarse grainsize. Variables that covary with the 4-16 μ m particles are dolomite, illite, sediment accumulation rate, X, SIRM. F2 scores are on average 10% in the period 0-10,000, then it starts increasing in importance with a break in YD cold event, and reaches its maxima after 14,500 (avg. 80%).

Factor 3 (F3, 8.5% of variance) represents the "Perturbing" factor. It is probably related to oceanographic instability and changes in supply fluxes (e.g. Mfs, melt waters, sapropel). The most important variables in F3 are 8-32 μ m size interval, kaolinite, clorite and, with negative sign, X and SIRM. The highest scores of F3 are in the Late glacial (avg. 20%) and secondarily in the periods of 8,000-9,000 and around 5,500 calendar years.

The first two factors represent the main forcing functions in our model, and are somewhat inversely correlated. In any case, it seems that most of the variability in our data set is highly correlated to the grain size. Several concurrent components can be responsible for the distributions of different % of factors 1 and 2 in the different core intervals. Among these factors the most critical are: (i) the distance of the riverine supply (Po River and others); (ii) the increasing water depth; (iii) the enhanced connection with the Mediterranean; and (iv) the onset of cyclonic gyre and increased wind fetch. The main physical oceanographic features seem to drive most of the granulometric distribution, as well as chemical and mineralogical variables.

All proxies available for core CM92-43 show that the time interval between 10,000 and 14,000 calibrated years records the highest degree of short-term paleoenvironmental variability (Fig. 4; see Asioli, this volume; Lowe *et al.* 1996, this volume). In particular, an abrupt shift toward warm surface water is marked by the entrance of planktic forams in the basin about 14,200 calibrated years ago (Asioli 1996, this volume); several short-term oscillations from warmer to cooler water temperatures characterise the transition to the Younger Dryas cold event (Asioli 1996, this volume). Similar short-term oscillations affect also the relative weight of Factors 1 and 2. These oscillations are superposed on a general trend characterised by the gradual decrease in the load of Factor 1 and the concurrent increase of the importance of Factor 2. Factor 3 represents less than 10% of the variance before 13.400 and after 10.500 calibrated years ago; the significance of Factor 3 is greatest

between these two dates and accounts for as much as 40% of the variance in the lower portion of the Younger Dryas interval. Significantly, this interval is also characterised by a revertance of the relative importance of Factor 1 (related to the relatively-coarser sediment fractions) possibly in response to increased sediment fluxes induced by changed climatic conditions (Trincardi *et al.* 1996, this volume; Langone *et al.*, 1996, this volume).

In general, the three factors used in this analysis show markedly different relative importance within the main stratigraphic intervals of core CM92-43. These factors describe most of the variability derived by 21 variables related to physical properties of the sediment. However, the boundaries between intervals characterised by different loads of the three factors match the main subdivisions based on planktic foraminifera and pollen spectra (Asioli 1996, this volume; Lowe *et al.* 1996, this volume). Factor 3, in particular, is most important during times when the other biostratigraphic variables indicate rapid oceanographic and climatic turnovers.

5. CONCLUSIONS

The statistical methods used here are well known. They are commonly used when the interplay of many sources of variation do not allow an objective and comprehensive synthesis for the description of physical phenomena.

The analysis has shown that a multivariate statistical strategy highlights physical paleoenvironmental factors in core CM92/43 and independently confirm the reliability of stratigraphic subdivisions based on biological factors (Asioli 1996, this volume). Furthermore, the statistical approach represents a tool for quantitative characterisation.

While more research is needed to relate all important granulometric and chemical parameters available (and other ones not yet determined), it is useful to employ statistical techniques for the definition of magnetic parameters. Additional parameters as well as appropriate sampling schemes for a more detailed study are being suggested by the analysis.

If compared with the model constrained by foraminifera ecozones and chronostratigraphy, geochemical and sedimentological patterns gave similar information with respect to one or more "forcing functions" (factors) responsible of the variations of the parameters with time (and therefore depth in core).

The conclusions related to factor analysis could be better explained if oxygen isotopes and biological data (in particular planktic and benthic foraminifera) were added to the multivariate analysis. Time-trend analysis of short-term variability will also be possible if adequate numbers of samples were added, according to the different sedimentation rates, in order to reach uniformity in sampling intervals.

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FIGURE CAPTIONS

Fig. 1 - Plot of median grain-size, clay fraction ($<2\mu m$), , calcite, dolomite (upper), and clay minerals (lower), versus calibrated age in core CM92/43.

Fig. 2 - Dendrogram produced by clustering 92 samples (a), and 21 variables (b) from core CM92/43. The numbers in the column to the left correspond to the calibrated ages. The values plotted on the horizontal axis are the % of similarity. The numbers (1 to 4) in the left dendrogram (a) are the four clusters identified by the classification.

Fig. 3 - Discriminant score 3D scatterplot for the three discriminant functions obtained: classification of the four groups of samples obtained by cluster analysis (see Fig. 2a). Discriminant variables are: dolomite, $<2\mu$ m, smectite, ARM, SIRM/ARM, KARM/K for Function 1; >63 μ m, SIRM for Function 2; sedimentation rate, X for Function 3.

Fig. 4 - Downcore plot of normalised components for the three factors (F1, F2, F3) extracted by the Q-Mode Factor analysis. mfs = maximum flooding surface: 5,000-6,000 (Trincardi *et al.* 1996, this volume); S1 = sapropel: 7,800-9,000; YD = Younger Dryas cold event: 11,500-12,700 years.

Table 1- Group means of the discriminating elements for each function (dolomite, <2µm, smectite, ARM, SIRM/ARM, KARM/K for Function 1; >63mm, SIRM for Function 2; sedimentation rate, X for Function 3

GROUP	dolomite	<2µm	Smectite	ARM	SIRM/ARM	KARM//K	>63µm	SIRM	sed. rate	Х
1	7	. 33	29	54	10	0.05	1.6	553	0.03	30
2	9	29	30	44	8	0.06	1.1	348	0.02	18
3	11	25	18	27	12	0.04	0.4	311	0.07	19
4	16	21	9	13	56	0.01	1.1	661	0.05	28

Table 2. Q-Mode Factor score matrix of data for 92 samples. The elements with loading >1.00 (bold) are those that characterise the factors and are discussed in the text.

	F 1	F 2	F 3
calcite	1.20	-0.10	0.89
dolomite	-0.29	1.53	0.67
0.5-1µm	1.51	0.04	-0.29
1.0-2.0 μm	1.56	0.07	-0.48
2.0-4.0 µm	1.48	0.29	-0.87
4.0-8.0 μm	-0.04	1.35	0.47
8.0-16.0 μm	-0.31	1.21	1.59
16-32 μm	-0.12	0.99	1.62
>63 µm	1.23	0.58	-0.98
sed. rate	0.03	1.22	0.60
smectite	1.45	-0.23	0.59
illite	-0.16	1.66	-0.01
kaolinite	0.80	0.05	1.62
clorite	-0.22	1.29	1.11
TOC	1.33	-0.26	0.95
Xfd	1.37	0.17	-0.24
Х	0.61	1.50	-1.71
ARM	1.46	-0.21	0.49
SIRM	0.44	1.58	-1.47
SIRM/ARM	-0.10	1.74	-0.36
KARM/K	1.33	-0.35	1.19



FIG. 1









