

Multivariate Data Analysis in Classification of Musts and Wines of the Same Variety According to Vintage Year

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Multivariate analysis has been used to discriminate among different vintage years (1986, 1987, 1988) of Chardonnay musts and wines from 31 growing areas in a relatively small geographical region of Trentino (Italy). Musts have been analyzed as to their content of amino acids and wines with respect to amino acids, volatile compounds (i.e., alcohols, esters, amides, acids, and others), and metal ions. Principal component analysis has been used as a display technique. To identify the most significant parameters for discrimination, an appropriate stepwise procedure for feature selection was carried out before linear discriminant analysis was applied, which led to quite satisfactory classification and prediction rates. Biochemical and oenological arguments have been proposed to account for the significance of the selected sets of variables. On the other hand, unsatisfactory results were obtained by univariate approach based upon calculation of Fisher weights.

INTRODUCTION

A great deal of work has been done on the application of pattern recognition techniques in discriminating wines of different varieties on the basis of objective analytical-chemical parameters (Etievant et al., 1988, 1989; Larrechi et al., 1988; Marais et al., 1981; Moret et al., 1984; Rapp and Güntert, 1985; Scarponi et al., 1982; Schreier et al., 1976). A few papers dealing with the characterization of musts and wines obtained from grapes of the same variety, harvested in quite different geographical areas, have also been published (Marais et al., 1981; Rapp et al., 1985; Rapp and Hastrich, 1978; van Rooyen et al., 1985). On the other hand, as to the classification of wines of the same variety and geographical origin on the basis of their vintage year, we found in the literature only a very few papers. An early paper reports on the results of a univariate approach to the discrimination between two different vintages (Scarponi et al., 1982); in a recent paper (Heymann and Noble, 1989) the canonical variate analysis based on wine descriptive variables tested at the same time led in some cases to good separation among three vintage years. Furthermore, data relative to the different amino acids in musts for sparkling wines—Chardonnay included—(Millary et al., 1986), as well as nitrogen content under various forms (Betz and Schmitt, 1980, 1982), for different varieties in two different zones, for two consecutive years with marked changes in climate characteristics, have been considered.

The scarce attention paid to this problem led us to the study reported in the present paper, taking into account that the vintage year is acknowledged to be a basic point of reference in defining quality and peculiarities of a given product. On the one hand, the characteristics of grapes, musts, and wines of the same variety and coming from the same place mainly reflect the whole of meteorological

factors typical of a given year ["millésime" (Ribereau-Gayon et al., 1975)]; on the other hand, they should be more or less efficiently recognized by a suitable set of objective chemical variables.

In view of these considerations, we have tested the capability of multivariate techniques in discriminating the vintage year (1986, 1987, 1988) of musts and relevant wines from Chardonnay grapes grown in different zones of Trentino (Italy). As to the musts, we have chosen to carry out the procedure using exhaustive amino acid maps (23 compounds plus ammonia). With regard to the wines, the discrimination has been based on their content of 20 amino acids (plus ammonia), 38 different volatile compounds (alcohols, esters, amides, acids, and others), and 4 metal ions.

An additional goal of our work was the identification of the most significant chemical variables for this kind of classification problems. When possible, biochemical and oenological reasons have been given for the selection of a set of discriminating variables.

EXPERIMENTAL PROCEDURES

Characteristics of the Samples and Analytical Tests. Thirty-one Chardonnay (130 SMA and standard clones with various rootstocks) vineyards in different zones of Trentino, submitted to similar agronomical practices, have been considered. In the three vintage years examined, no particular irregularity in climate could be evidenced and the wines produced were at good and comparable levels of quality. Agronomical procedures were as much as possible similar from one case to another, with respect to controlled fertilization and pruning; the vineyards were irrigated only when strictly necessary. The soil was in any case of calcareous origin. Two quintals of grapes for each vineyard, picked at the ripening time suitable to obtain sparkling wines, were made into wine in the experimental cellar of Istituto Agrario Provinciale, San Michele all'Adige, Italy, under strictly reproducible conditions. Harvested grapes were cooled for 12 h at 4 °C and then submitted to pneumatic automatic pressing to the same yield in must (70% w/w). The resulting must, added with 70 mg/L SO₂ and 300 mg/L bentonite, was static settled and fermented by a selected *Saccharomyces cerevisiae* strain (Ep

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111 from the yeast collection of IAP) between 18 and 21 °C. The decanted wines were filtered after 2 weeks, added with 30 mg/L SO₂, and stored under similar conditions (from 12 to 15 °C). All samples were analyzed between March and May in the year following that of the vintage.

The content of the different amino acids, both in musts and in wines, was determined with an automatic amino acid analyzer (C. Erba) according to the procedure reported in the literature (Margheri et al., 1982). Some of the volatile compounds in wines, namely methanol, acetaldehyde, ethyl acetate, and higher alcohols, were analyzed on distillate from wine at pH 7, directly by gas chromatography on a packed column [2 m, 2 mm i.d., Carbowax C 60/80 mesh, 0.2% CW1500 (Supelco)] (Gabri and Salvagiotto, 1980). The other volatile compounds were analyzed on WCOT capillary columns (Hewlett-Packard HP-20M, 50 m, 0.32 mm i.d., $d_f = 0.3 \mu\text{m}$; and J&W DB-Wax, 30 m; 0.32 mm i.d., $d_f = 0.5 \mu\text{m}$) after continuous extraction (10 h) by *n*-pentane-dichloromethane (2:1 v/v) (Drawert and Rapp, 1968; Margheri and Versini, 1979). In the former case a C. Erba Model Fractovap 2400 T and in the latter case a C. Erba Model HRGC 5300 chromatograph was used.

Sodium and potassium ions were determined by flame photometry (flame photometer, Turner Model 510) and calcium and magnesium by atomic absorption technique (atomic absorption spectrophotometer, Perkin-Elmer Model 400); in any case conventional procedures were followed (Gazzetta Ufficiale della Repubblica Italiana, 1986).

Processing of Data and Statistical Techniques. Statistical treatment of data (Massart et al., 1978, 1988; Sharaf et al., 1986) has been carried out by using the package PARVUS (Forina et al., 1988). Raw data have been autoscaled in advance; i.e., we subtracted from each variable value the average of all the values for that variable and then divided each of such modified values by the standard deviation of that variable. In other words, our modified variables had an average of zero and a standard deviation of unity. The following techniques have been proved to be most useful.

Principal Component Analysis (PCA). This technique identifies, in the hyperspace of the variables, the directions on which the most part of information is retained, by that reducing the dimensionality of the system. By projecting the objects of the data set onto the space of the first few components, it is possible to demonstrate differences among the various objects, determining at the same time which variables are principally involved.

Fisher Weights. The Fisher weight is a measure of the capability of a given variable to discriminate between two categories. It is proportional to the ratio of intercategory to intracategory variances: it is, hence, higher, the higher the distance between the mean values of the considered categories is, and it is lower, the more scattered the objects within the categories are.

Linear Discriminant Analysis (LDA). Among the different multivariate techniques, the relatively simple LDA was in our case very effective, so that we did not find it profitable to use more complex methods. LDA makes the hypothesis that the distribution is multivariate normal and that the covariance matrix of each category (dispersion of the category) is not significantly different from one case to another. The Mahalanobis distances of each object from the centroids of the categories are computed, the object resulting assigned to the category with lowest distance. The delimiter between two categories is a linear function, i.e., a straight line in the case of two independent variables, a plane for three variables, etc. To check the predictive ability of LDA, the following method has been used: an evaluation set, made of 20% of the objects, randomly selected, has been used to verify the classification rules obtained by the training set; this procedure has been repeated 100 times, and the prediction rate has been computed as the percentage of correct predictions.

RESULTS AND DISCUSSION

Musts. Seventy-three objects (i.e., 73 musts, different from one another with respect to either the vintage year or the growing area) have been considered, each one being described by 24 variables (23 amino acids plus ammonia): variable 1, hydroxyproline; 2, aspartic acid; 3, threonine;

Table I. Data Set Musts: Fisher Weights^a

variable	1986-1987	1986-1988	1987-1988
6	2.231	1.778	0.028
2	1.266	1.552	0.062
8	0.444	0.147	1.335
13	0.349	1.137	0.139
1	1.231	0.102	0.272

^a Only variables having at least one Fisher weight >1 are reported.

4, serine; 5, asparagine; 6, glutamic acid; 7, glutamine; 8, proline; 9, glycine; 10, alanine; 11, citrulline; 12, valine; 13, cysteine; 14, methionine; 15, isoleucine; 16, leucine; 17, tyrosine; 18, phenylalanine; 19, γ -aminobutanoic acid; 20, ammonia; 21, ornithine; 22, lysine; 23, histidine; 24, arginine. Quantities like total sugar, total malic and tartaric acid, as well as total acidity and pH, whose values are quite commonly determined in musts, have not been considered by us to focus our attention on the amino acid map. Preliminary tests have proved that the consideration of these variables alone leads to definitely poor results. On the other hand, malic acid and, to a minor extent, pH, seem to be significant variables when considered in a data set which also includes amino acids (Seeber et al., work in progress).

Since each variable shows an almost normal distribution on the samples, no transformation of the data has been performed, apart from the cited autoscaling.

A univariate evaluation of the discriminant power of the different variables has been performed by computing the Fisher weights (see Table I). The results obtained suggest that a similar approach is completely inadequate, glutamic acid being the only variable that assumes a relatively high value, but only in discriminating within one pair of years.

To perform PCA, the eigenvectors of the generalized covariance matrix were computed; four of them (79.3% of the total variance) were significant according to double cross validation (Wold, 1978). Eigenvectors 1 and 2, explaining 45.5% and 17% of the total variance, respectively, do not contain any useful information to discriminate among the years, while the projection on the plane of eigenvectors 3 and 4 (9.9% and 6.8% of the overall variance, respectively) shows a satisfactory separation among vintages (Figure 1): eigenvector 3 discriminates 1986 from 1987 and 1988, which are separated from each other by eigenvector 4. The fact that the information related to the vintage year can be found not on the very first eigenvectors suggests that, at least in what concerns the composition in amino acids, factors attributable to different local pedological and climatic characteristics (Schrader et al., 1976), as well as to differences in canopy orientation, can be greater sources of variability than those related to the seasonal differences, as already evidenced for some other qualitative parameters of musts (Crusius, 1982).

The use of LDA as a classification technique has been suggested and made possible by the similarity among the covariance matrices of the three categories. A proper stepwise feature selection procedure (F to enter = 6) (Jenrich, 1960) has been performed as a pretreatment of data devoted to identify the variables with highest discriminant ability and hence to decrease the number of variables to be used in processing the data, eliminating those variables whose information, though possibly significant for other objectives (e.g., determination of the geographic origin) does not contribute to our purpose. Six variables were selected, according to the following order: glutamic acid, aspartic acid, proline, leucine, alanine, and serine. It

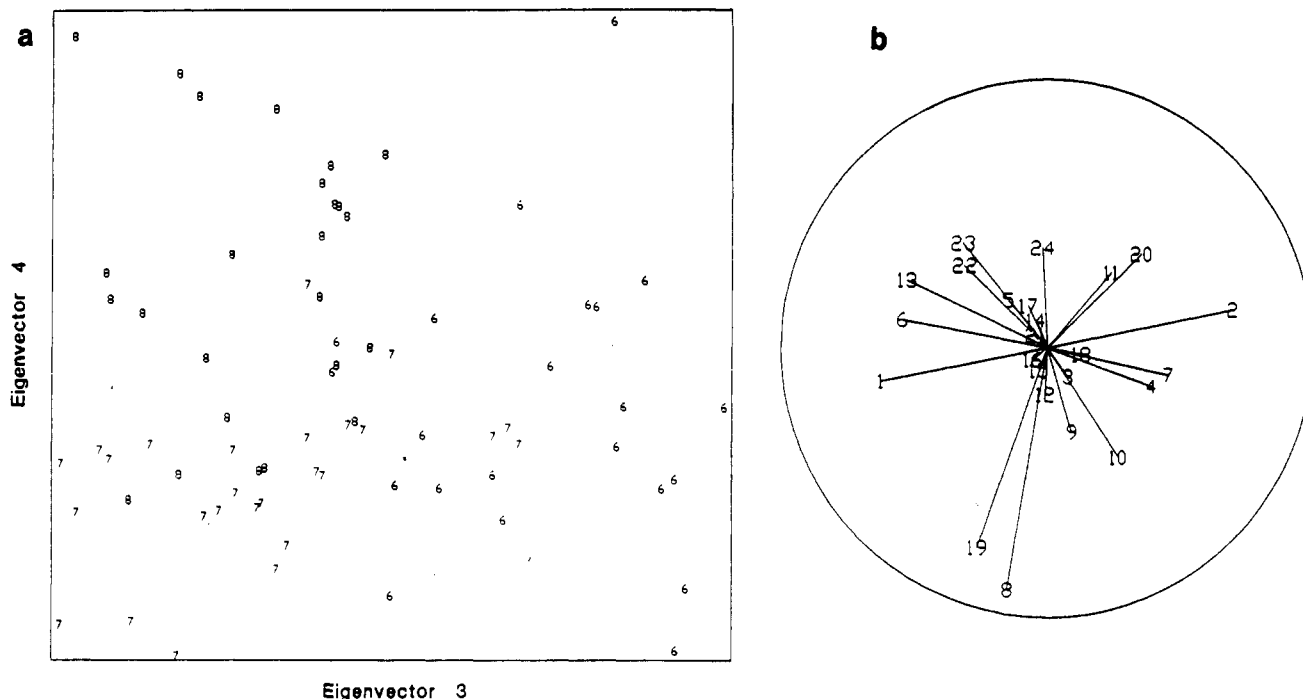


Figure 1. Data set musts: (a) scores (last digit of the year reported: 6 = 1986; 7 = 1987; 8 = 1988); (b) loadings (variable index reported) on eigenvectors 3 and 4.

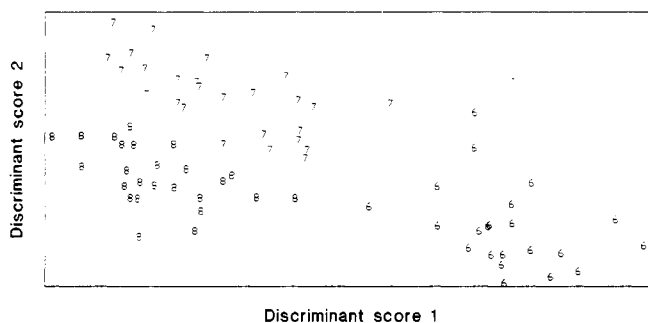


Figure 2. Data set musts. Plot of discriminant score 1 vs discriminant score 2 (last digit of the year reported).

Table II. Data Set Musts: LDA Prediction Matrix^a (Six Variables)

year	1986	1987	1988
1986	96.4	3.4	0.2
1987	3.0	94.5	2.4
1988	0.0	0.8	99.2

^a Mean value: 96.7%.

is interesting to note that alanine, aspartic acid, glutamic acid, and proline are understood to have a partly common biosynthetic origin (Lehninger, 1975; Ribereau-Gayon et al., 1975, pp 161-72). Figure 2 shows the discriminant scores computed by LDA using only the six above-mentioned variables. Perfect classification and very high prediction rates (Massart et al., 1988, Chapter 23) have been obtained, as reported in Table II. From the data in this table, as well as from Figure 2, no significant overlap between 1986 and 1988 vintages is evident, while 1987 exhibits a bit lower correct prediction rate, its characteristics being intermediate between those of 1986 and those of 1988.

Wines. The data set relative to wines consisted of 76 objects, characterized by 63 variables, i.e., 4 metal ions, 20 amino acids (plus ammonia), and 38 volatile compounds: variable 1, sodium; 2, potassium; 3, calcium; 4, magnesium; 5, aspartic acid; 6, threonine; 7, serine; 8, asparagine; 9,

Table III. Data Set Wines: Fisher Weights^a

variable	1986-1987	1986-1988	1987-1988
43	12.092	1.694	2.470
3	0.279	1.625	7.251
26	5.131	2.366	1.128
45	4.141	3.030	0.049
2	0.102	1.741	4.908
41	2.053	0.001	2.474
44	1.625	2.215	0.279
53	0.043	1.209	2.784
55	1.703	2.269	0.034
35	0.067	0.979	2.620
36	0.198	0.304	2.363
47	1.589	0.346	0.905

^a Only variables having at least one Fisher weight >1 are reported.

glutamic acid; 10, glutamine; 11, proline; 12, glycine; 13, alanine; 14, valine; 15, methionine; 16, isoleucine; 17, leucine; 18, tyrosine; 19, phenylalanine; 20, γ -aminobutanoic acid; 21, ammonia; 22, ornithine; 23, lysine; 24, histidine; 25, arginine; 26, methanol; 27, 1-propanol; 28, 2-methyl-1-propanol; 29, 2-methyl-1-butanol; 30, 3-methyl-1-butanol; 31, 27 + 28 + 29 + 30; 32, acetaldehyde; 33, ethyl acetate; 34, isoamyl acetate; 35, hexyl acetate; 36, 2-phenylethyl acetate + isobutyl acetate + 34 + 35; 37, ethyl butanoate; 38, ethyl hexanoate; 39, ethyl octanoate; 40, ethyl decanoate; 41, γ -butyrolactone; 42, ethyl 4-hydroxybutanoate; 43, 4-carbomethoxy- γ -butyrolactone; 44, 1-hexanol; 45, *trans*-3-hexen-1-ol; 46, *cis*-3-hexen-1-ol; 47, benzyl alcohol; 48, 2-phenylethanol; 49, 3-ethoxy-1-propanol; 50, 3-(methylthio)-1-propanol; 51, 1,3-propanediol monoacetate; 52, 4-vinylphenol; 53, 4-vinylguaiacol; 54, butanoic acid; 55, 2-methylpropanoic acid + 2- and 3-methylbutanoic acids + 54; 56, hexanoic acid; 57, octanoic acid; 58, decanoic acid; 59, dodecanoic acid; 60, 56 + 57 + 58 + 59; 61, *N*-(2-methylbutyl)acetamide; 62, *N*-(3-methylbutyl)acetamide; 63, *N*-[3-(methylthio)propyl]acetamide.

Table III reports the results obtained by univariate approach through evaluation of the Fisher weights for the different possible pairs of years. In this case the data look better than those for musts. However, except for 4-carb-

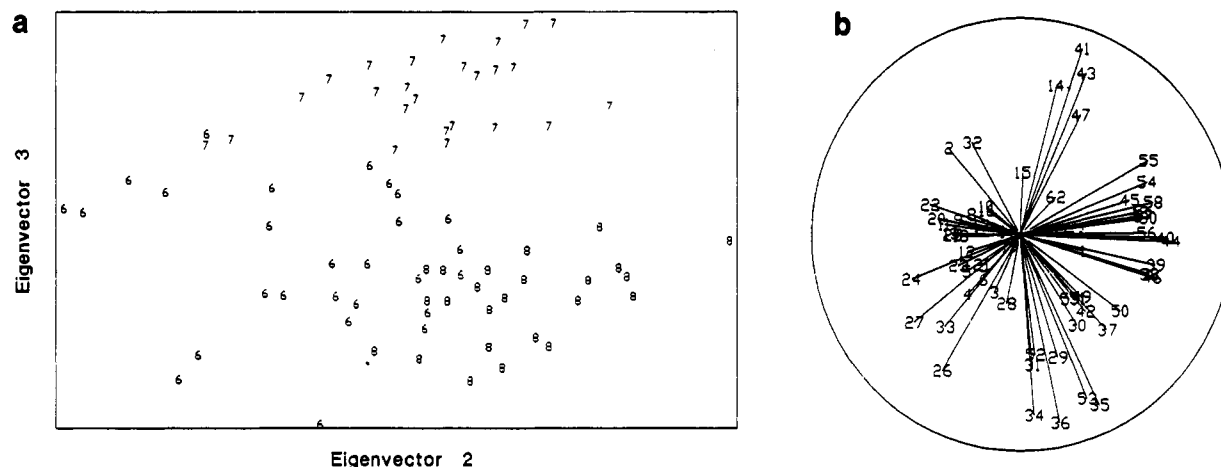


Figure 3. Data set wines: (a) scores (last digit of the year reported); (b) loadings (variable index reported) on eigenvectors 2 and 3.

ethoxy- γ -butyrolactone, acceptable results are obtained in any case only in the discrimination within two of the three pairs of categories, the examined variable being completely unsatisfactory in the third case. Fisher weights can only be useful for a first rough indication about the variables which could be used in a suitable classification procedure. As a matter of fact, proper multivariate criteria for feature selection are much more effective and sound.

Also in this case the eigenvectors of the generalized covariance matrix have been computed. Five eigenvectors (64.7% of the total variance) turned out to be significant according to double cross validation. A satisfactory separation among the different vintages is evident on the plane defined by eigenvectors 2 and 3, as Figure 3a shows: eigenvector 3 discriminates 1987 from 1986 and 1988, which are in their turn well separated by eigenvector 2. Eigenvector 1 (24.9% of variance) is mainly composed by amino acids, whose variability should be ascribed to factors other than those attributable to different vintages, e.g., to not easily rationalizable differences in metabolism by yeasts, implying differences in consumption, modification, production, and final extraction of amino acids, with consequent deep changes in their original map (Ribereau-Gayon et al., 1975, pp 307-18; Margheri et al., 1986). By comparison of the mean values of amino acids in musts with those in the relevant wines, for any year limited increments in proline content are evidenced, in accord with previous findings for musts with rather high nitrogen content (Di Stefano, 1980).

As shown in Figure 3b, the highest values for the loadings of eigenvector 2 (16.0% of variance) are proper to fatty acids (variables 54-60) and to some alcohols and esters. Lactones (variables 41 and 43), some acetates (variables 34-36), and 4-vinylguaiacol (variable 53) exhibit the highest loadings for eigenvector 3 (10.2% of variance).

Since the covariance matrices of the three categories are very similar to one another, LDA was also in this case the technique chosen for classification. The proper feature selection procedure (F to enter = 15) picked up five variables, in the following order: calcium, potassium, methanol, 4-carbomethoxy- γ -butyrolactone, and 4-vinylguaiacol. Figure 4 shows the discriminant scores obtained by LDA with the five selected variables. As in the case of musts, perfect classification and very high prediction ability were achieved (see Table IV).

Particularly interesting is the selection of variables whose content in wines cannot be ascribed to glycolytic processes. As to calcium and potassium, taking into account that wine-making was carried out in perfectly

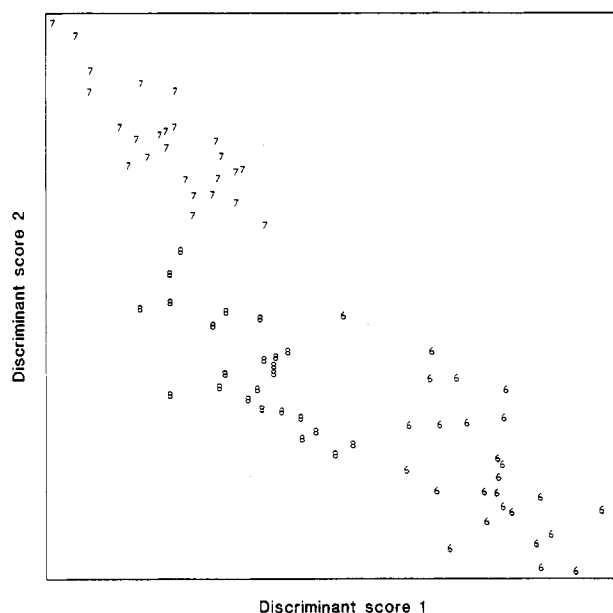


Figure 4. Data set wines. Plot of discriminant score 1 vs discriminant score 2 (last digit of the year reported).

Table IV. Data Set Wines: LDA Prediction Matrix^a (Five Variables)

year	1986	1987	1988
1986	94.5	1.2	4.3
1987	0.0	100.0	0.0
1988	0.0	0.0	100.0

^a Mean value: 97.2%

reproduced conditions and that the absence of any contamination from the outer environment was carefully checked, they suggest a different salification map for the organic acids (Iland and Coombe, 1988; Possner and Kliever, 1985) as a function of the vintage year. The validity of the methanol variable for this kind of classification is supported by the fact that it is not a fermentation product. Differences attributable to unequal pressing of grapes must be disregarded, taking into account that in this case a concomitant and positive change in 1-hexanol content should be found (Bayonove et al., 1976); on the contrary, the relative amounts of these two alcohols vary from one year to another. Different relative contents of methanol and 1-hexanol for different vintages have been recently evidenced in wines for production of cognac distillates (Cantagrel et al., 1990). As to the methanol, differences among vintages have also been evidenced in

experimental wines mainly of *Vitis Labrusca* variety (Lee et al., 1975). The quantity of 4-carbetoxy- γ -butyrolactone should depend on that of its likely precursor, namely 2-hydroxyglutaric acid, whose content is a function both of the starting amount in must and of the changes occurring during the fermentation process (Dittrich, 1987). The variability of 4-vinylguaiacol could be attributed to different quantities of ferulic acid in musts (Albagnac, 1975).

As a final consideration about the kind of approach followed by us for the treatment of data, a simple comparison of the results obtained on univariate and multivariate scale, with both musts and wines, should now be sufficient to affirm that such complex systems can be adequately studied only through multivariate methods.

Additional work is in progress in our laboratories on this, as well as on similar data sets. In a first study we apply multivariate analysis to fit a set of categories defined on the basis of microclimatic differences within the relatively small geographical area considered. The aim is that of identifying the variables that are more strictly connected with different conditions in local temperature, exposure to sunshine, rainfall, etc. In a second study, the most significant variables with respect to discrimination among different categories are identified within each single class of chemical compounds to find the most significant species of the different classes.

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Supplementary Material Available: Tables reporting the data of the set of variables considered for musts and wines, summarized in terms of mean value, standard deviation, and coefficient of variation (standard deviation/mean \times 100) (4 pages). Ordering information is given on any current masthead page.

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