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Multiple Discriminant Analysis in the Analytical Differentiation of Venetian White Wines. 4. Application to Several Vintage Years and Comparison with the *k* Nearest-Neighbor Classification

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Fifty-three samples of the Venetian white wines Soave Classico, Prosecco di Conegliano-Valdobbiadene, and Verduzzo del Piave, collected from vintages of 1980 and 1981, were analyzed for the parameters considered in previous papers. Results confirm previous findings on the variability of products through the years. Multiple discriminant analysis was applied to all the available data in our file [i.e., referred to samples of 1977, 1980, and 1981 Soave, 1977, 1979, and 1980 Prosecco, and 1977 and 1981 Verduzzo wines]. The apparent error rate was about 12.3%. The expected actual error rate was estimated both by the jackknife procedure and the learning set/test set partition method. Results were almost coincident (about 18%). A definite overlap between the Prosecco and the Verduzzo areas in the discriminant space was revealed. Results obtained by the *k* nearest-neighbor classification show that the two methods of pattern recognition have, in this case, practically the same classification power.

Previously, we reported on the applicability of the statistical method of multiple discriminant analysis to the problem of differentiation of three Venetian white wines (Soave Classico, Prosecco di Conegliano-Valdobbiadene, and Verduzzo del Piave) by means of the following analytical parameters: sodium, potassium, calcium, magnesium, chloride, pH, titratable acidity (TA), phosphorus, ash content (AC), and alkalinity of the ashes (AA) (Moret et al., 1980; Scarponi et al., 1981). Also, it was shown that, owing to the possible high variability of the products through the years, data from several vintage years need to be processed in order to obtain conclusive results (Scarponi et al., 1982).

Continuing this program, in this paper we report the results of the analytical measurements mentioned above and performed on samples obtained from 1980 and 1981 vintage years. The multiple discriminant analysis was repeated by using all the data, and the classification results, together with the estimate of the expected actual error rate, are reported. Moreover, the data set was processed by the reference, nonparametric classification rule of the *k* nearest neighbors (kNN). Results obtained by the two pattern recognition methods are compared.

EXPERIMENTAL SECTION

Collection and Analysis of Samples. Eight and ten samples of wine Soave, vintage of 1980 and 1981, respec-

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Table I. Analytical Results

parameter	wine Soave, 1980 vintage[c]	wine Soave, 1981 vintage[d]	wine Prosecco, 1980 vintage[e]	wine Verduzzo, 1981 vintage[f]
	mean values ±95% CI[a] (±95% TI[b])			
sodium, mg/L × 10^{-1}	1.5 ± 0.8 (±2.9)	1.7 ± 0.5 (±2.2)	1.1 ± 0.4 (±1.7)	2.6 ± 0.7 (±3.5)
potassium, mg/L × 10^{-2}	3.8 ± 0.8 (±2.9)	5.4 ± 0.5 (±1.9)	8.7 ± 1.0 (±4.5)	8.5 ± 0.6 (±3.0)
calcium, mg/L × 10^{-1}	8.6 ± 1.6 (±5.8)	8.8 ± 1.6 (±6.3)	11.8 ± 0.7 (±3.0)	9.2 ± 0.8 (±4.3)
magnesium, mg/L × 10^{-1}	8.3 ± 1.2 (±4.7)	8.6 ± 1.1 (±4.2)	9.4 ± 1.0 (±4.3)	7.7 ± 0.6 (±2.9)
chloride, mg/L × 10^{-1}	1.0 ± 0.3 (±1.2)	1.7 ± 0.3 (±1.3)	2.3 ± 0.3 (±1.3)	3.6 ± 0.6 (±3.0)
pH	3.20 ± 0.10 (±0.35)	3.25 ± 0.06 (±0.22)	3.32 ± 0.07 (±0.29)	3.44 ± 0.05 (±0.26)
titratable acidity, g of tartaric acid/L	6.4 ± 0.6 (±2.2)	6.2 ± 0.4 (±1.7)	6.9 ± 1.0 (±4.3)	6.0 ± 0.3 (±1.6)
phosphorus, mg of P_2O_5/L × 10^{-2}	1.6 ± 0.2 (±0.7)	2.1 ± 0.3 (±1.0)	2.3 ± 0.3 (±1.4)	1.7 ± 0.2 (±1.0)
ash content, g/L	1.5 ± 0.1 (±0.5)	1.5 ± 0.2 (±0.6)	2.3 ± 0.2 (±0.9)	2.2 ± 0.2 (±0.8)
alkalinity of the ashes, mequiv/L × 10^{-1}	1.6 ± 0.1 (±0.4)	1.6 ± 0.1 (±0.5)	2.7 ± 0.2 (±0.9)	2.1 ± 0.2 (±0.8)

[a] CI = confidence interval. [b] TI = tolerance interval; coverage 90% (Kendall and Stuart, 1979; Mandel, 1978). [c] 8 samples. [d] 10 samples. [e] 14 samples. [f] 21 samples.

tively, fourteen samples of wine Prosecco, vintage of 1980, and twenty-one samples of wine Verduzzo, vintage of 1981, were collected in the production areas from lots for which genuineness and tipicality were guaranteed and certified by the Italian D.O.C. (Denominazione di Origine Controllata—Certified Brand of Origin) brand (*Gazz. Uff. Repub. Ital.*, 1968, 1969, 1971, 1976).

Analytical measurements were performed according to the methods previously reported (Moret et al., 1980; Scarponi et al., 1982).

Before the analyses of new samples were made, reproducibility of measurements over time was checked reanalyzing some of the earlier samples. In this way differences due to possible analytical bias were prevented.

Data Set. In addition to samples collected and analyzed in this work, those studied in previous papers (Moret et al., 1980; Scarponi et al., 1982) were considered too. Then the data set was constituted by the following wine samples: 1977, 1980, and 1981 Soave; 1977, 1979, and 1980 Prosecco; 1977 and 1981 Verduzzo. This data set was used for the statistical analyses throughout.

Discriminant Analysis. Discriminant analysis [see, e.g., Lachenbruch (1975) and Chatfield and Collins (1980)] was performed by using all the data in our file (see above) and by grouping data obtained from samples of the same type of wine collected in the different vintage years. Concentration units of the chemical parameters were the following: mg/L for sodium, potassium, calcium, magnesium and chloride; g of tartaric acid/L for titratable acidity; g/L for ash content; mg of P_2O_5/L for phosphorus; mequiv/L for alkalinity of the ashes.

As in previous papers, calculations were performed by using the DISCRIMINANT subprogram of the SPSS (Version 8.3) statistical package for the computer (Nie et al., 1975; Hull and Nie, 1981). Both direct and stepwise methods (the last performed according to several criteria defined as Wilks, Mahal, Maxminf, Minresid, and Rao) were used. The optimization procedures, as obtained by excluding from the analysis one, two, ..., and eight parameters, respectively, beginning from the less important one in the selection orders of parameters, were performed and classification results were compared. The apparent classification error rates and the estimate of the actual error rate by the jackknife procedure were calculated as previously reported (Scarponi et al., 1982).

A more correct estimate of the actual error rate was calculated by the learning set/test set partition method Lachenbruch, 1975). In this method the data set is split in two parts, defined as the training set and the test set. Then the test samples are classified on the basis of the discriminant and classification functions as obtained with the training samples, and the pertinent error rate is computed. A total of 60, 70, 80, 90, and 95% of cases were randomly sampled for the training set by the SELECT procedure of SPSS (Hull and Nie, 1981). In each case analysis was repeated 20 times and the results were averaged.

***k* Nearest-Neighbor Classification.** Among the nonparametric methods used for supervised pattern recognition in analytical chemistry, the kNN classification has received more and more attention in the last decade (Kowalski and Bender, 1972; Kowalski, 1975; Saxberg et al., 1978; Duewer et al., 1978; Albano et al., 1978; Massart et al., 1978, Sjostrom and Kowalski, 1979; Varmuza, 1980; Coomans and Massart, 1982a–c). The statistical foundation of this method is well established (Cover and Hart, 1967; Cover, 1968; Peterson, 1970; Kowalski and Bender, 1972), and it has been suggested as "a reference with which other more sophisticated procedures may be compared" (Cover, 1968; Kowalski and Bender, 1972).

The method consists in the computation of the Euclidean distance in the multidimensional space between each couple of points, in order to obtain a distance matrix. Then an unknown pattern is classified according to the majority vote of its *k* nearest neighbors. In the case of ties, the closer neighbors get the greater weight. Calculations with *k* equal to 1, 3, and 5, respectively, were performed by using the KNN subroutine of the SIMCA package (Wold, 1976, 1977).

RESULTS AND DISCUSSION

Analytical Results. Table I gives the results of the analytical determinations performed on the collected samples. Mean values, 95% confidence intervals, and 95% tolerance intervals, coverage 90% (Kendall and Stuart, 1979; Mandel, 1978), are reported. In addition, Table II gives the results obtained by grouping data of the same type of wine as obtained from the different vintage years. The comparison of the present results with previous findings confirms the high variability of the products through the years.

Analysis of Variance and Correlation. Results of the one-way analysis of variance test for equality of group means on a single variable are reported in Table III. Data relative to samples of the same type of wine have been grouped. It can be noted that the order of importance of the variables is slightly changed with respect to the results previously reported (Scarponi et al., 1981, 1982). No-

Table II. Results Obtained by Grouping Data of the Same Type of Wine

parameter	wine Soave[c] (1977, 1980, and 1981 vintages)	wine Prosecco[d] (1977, 1979, and 1980 vintages)	wine Verduzzo[e] (1977 and 1981 vintages)
	mean values ± 95% CI[a] (± 95% TI[b])		
sodium, mg/L × 10^{-1}	1.3 ± 0.3 (±1.7)	1.4 ± 0.3 (±1.9)	2.9 ± 0.6 (±3.9)
potassium, mg/L × 10^{-2}	5.6 ± 0.6 (±3.8)	7.9 ± 0.4 (±3.0)	8.9 ± 0.6 (±3.9)
calcium, mg/L × 10^{-1}	9.0 ± 0.7 (±4.1)	12.0 ± 0.5 (±3.6)	10.5 ± 1.0 (±5.8)
magnesium, mg/L × 10^{-1}	8.3 ± 0.5 (±3.2)	9.4 ± 0.7 (±4.9)	8.9 ± 0.9 (±5.2)
chloride, mg/L × 10^{-1}	1.2 ± 0.2 (±1.1)	2.2 ± 0.3 (±2.1)	3.3 ± 0.5 (±2.8)
pH	3.16 ± 0.05 (±0.27)	3.32 ± 0.04 (±0.26)	3.45 ± 0.04 (±0.26)
titratable acidity, g of tartaric acid/L	6.4 ± 0.2 (±1.4)	6.4 ± 0.4 (±2.6)	6.0 ± 0.2 (±1.4)
phosphorus, mg of P_2O_5/L × 10^{-2}	2.8 ± 0.5 (±2.8)	2.5 ± 0.3 (±2.3)	2.8 ± 0.5 (±3.1)
ash content, g/L	1.5 ± 0.1 (±0.4)	2.1 ± 0.1 (±0.8)	2.3 ± 0.1 (±0.8)
alkalinity of the ashes, mequiv/L × 10^{-1}	1.7 ± 0.1 (±0.3)	2.4 ± 0.1 (±0.9)	2.3 ± 0.2 (±0.4)

[a] CI = confidence interval. [b] TI = tolerance interval; coverage 90% (Kendall and Stuart, 1979; Mandel, 1978). [c] 32 samples. [d] 47 samples. [e] 35 samples.

Table III. Univariate F Ratios (One-Way Analysis of Variance Test for Equality of Group Means on a Single Discriminating Variable) with 2 and 111 Degrees of Freedom

parameter	F ratio	significance
AC	52.2	<0.01
pH	46.6	<0.01
AA	36.4	<0.01
Cl	36.2	<0.01
K	33.1	<0.01
Ca	18.3	<0.01
Na	17.5	<0.01
TA	2.1	0.13
Mg	2.0	0.14
P	0.7	0.48

Table IV. Discriminating Power of the Discriminant Functions Obtained Using the Direct Method

Eigenvalues Associated with Functions			
discriminant function	eigenvalue	rel %	canonical correlation
1	2.07	68.3	0.821
2	0.96	31.7	0.700

Change in Wilks' Λ				
functions derived	Λ	χ^2	df[a]	significance
0	0.166	191.1	20	<0.01
1	0.510	71.8	9	<0.01

[a] df = degrees of freedom.

ticeable are the increase in importance of the parameter AC with respect to pH and the reduction of discriminating power obtained for parameter P. From the correlation matrix obtained from all the available data (not reported here), correlations previously found are confirmed.

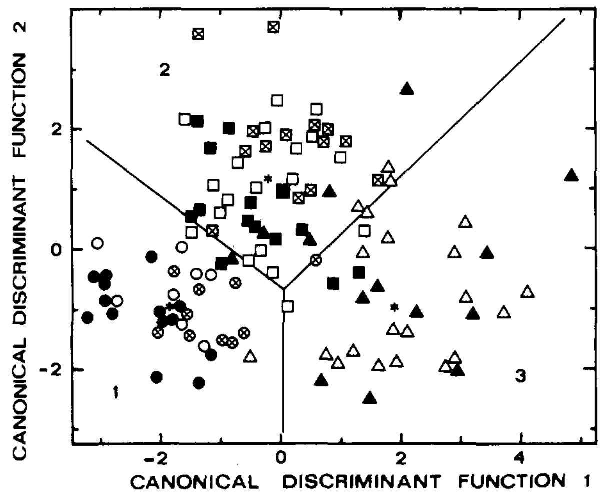

Figure 1. Graphical representation of wine groups in the discriminant plane. Area 1, Soave 1977 (●), 1980 (○), and 1981 (⊗); area 2, Prosecco 1977 (■), 1979 (□), and 1980 (⊠); area 3, Verduzzo 1977 (▲) and 1981 (△). (*) Group centroids.

Discriminant Analysis. *Direct Method.* Table IV reports the necessary information for judging how many discriminant functions obtained by the direct method should be derived. From the values reported (relative percentage of the eigenvalue associated with the second function, 31.7; Wilks' Λ computed after the first function is derived, 0.510), one can see that it is useful to derive the second (and last) discriminant function. Consequently, the remaining computations are based on the two functions.

Tables V and VI and Figure 1 show the results of the multiple discriminant analysis performed by the direct method. From the standardized canonical discriminant

Table V. Discriminant and Classification Function Coefficients Obtained with the Direct Method

variable	standardized funct 1	standardized funct 2	unstandardized funct 1	unstandardized funct 2	wine Soave	wine Prosecco	wine Verduzzo
	canonical discriminant function coefficients				classification function coefficients		
Na	0.279	−0.422	2.19 × 10^{-2}	−3.30 × 10^{-2}	5.13 × 10^{-1}	4.79 × 10^{-1}	5.85 × 10^{-1}
K	0.325	−0.250	1.91 × 10^{-3}	−1.47 × 10^{-3}	6.61 × 10^{-2}	6.61 × 10^{-2}	7.28 × 10^{-2}
Ca	0.030	0.565	1.37 × 10^{-3}	2.59 × 10^{-2}	1.17 × 10^{-1}	1.73 × 10^{-1}	1.29 × 10^{-1}
Mg	−0.158	−0.018	−7.04 × 10^{-3}	−7.89 × 10^{-4}	−2.33 × 10^{-1}	−2.46 × 10^{-1}	−2.59 × 10^{-1}
Cl	0.413	0.200	3.98 × 10^{-2}	1.92 × 10^{-2}	1.43 × 10^{-1}	2.47 × 10^{-1}	2.94 × 10^{-1}
pH	0.614	−0.290	4.88	−2.31	5.41 × 10^{2}	5.44 × 10^{2}	5.58 × 10^{2}
TA	0.297	−0.043	3.10 × 10^{-1}	−4.45 × 10^{-2}	4.15 × 10^{1}	4.19 × 10^{1}	4.26 × 10^{1}
P	−0.017	−0.600	−1.28 × 10^{-4}	−4.59 × 10^{-3}	5.63 × 10^{-2}	4.65 × 10^{-2}	5.46 × 10^{-2}
AC	0.636	−0.499	1.86	−1.46	−1.14 × 10^{2}	−1.14 × 10^{2}	−1.08 × 10^{2}
AA	−0.606	1.400	−1.50 × 10^{-1}	3.46 × 10^{-1}	−2.04	−1.56	−2.50
constant			−2.07 × 10^{1}	3.10	−9.11 × 10^{2}	−9.37 × 10^{2}	−9.87 × 10^{2}

Table VI. Classification Results: Direct Method

actual group	no. of samples	predicted group membership: Soave	Prosecco	Verduzzo	apparent correct classification, %[a]
Soave	32	31	0	1	96.9
Prosecco	47	2	40	5	85.1
Verduzzo	35	2	4	29	82.9

[a] Total apparent correct classification = 87.7%.

Table VII. Selection Order of Variables in the Stepwise Methods

selection order	selection methods: Wilks and Minresid	Mahal[a]	Rao
1	AC	pH	AC
2	AA	Cl	Cl
3	Cl	Ca	AA
4	P	Na	pH
5	Ca	P	Na
6	Na	AA	P
7	pH	AC	Ca
8	K	Mg	K
9	TA	TA	TA
10	Mg	K	Mg

[a] Selection order for method Maxminf is equal to that of method Mahal with the exception of the sequence Mg, TA, which is reversed.

Table VIII. Optimization Procedure and Comparison of the Various Selection Methods

no. of included variables[a]	apparent correct classification, %, for selection method: Wilks and Minresid	Mahal and Maxminf	Rao
9	86.8	86.8	86.8
8	85.1	85.1	85.1
7	86.0	86.0	86.0
6	86.0	82.5	83.3
5	85.1	81.6	83.3
4	82.5	80.7	82.5
3	82.5	79.0	82.5
2	79.8	71.1	71.1

[a] On the basis of the selection orders given in Table VII.

function coefficients, it can be seen that the most significant parameters for the differentiation are pH, AC, and AA, in the first function, and AA, P, and Ca, in the second one, respectively. Conversely, parameters such as Mg, TA, and K give low contributions to both functions.

As regards the classification obtained with the new functions it is to be noted that (i) samples of the same wine collected in different vintage years are well grouped together in the discriminant space (see Figure 1), (ii) a definite overlap exists between the Prosecco and Verduzzo areas, and (iii) the classification error (apparent value 12.3%) reflects this incomplete separation of the two wines. Such results strongly suggest that, in order to improve the classification, parameters with more discriminating power need to be sought out.

Stepwise Methods and Optimization. Tables VII and VIII give the results obtained for the order of selection of variables in stepwise methods and for the optimization procedure, respectively. It can be noted that, contrary to previous findings, classification cannot be improved by exclusion of any variable from analysis. However, it is possible to delete up to five or eight variables without an increase of the apparent error rate above 15 and 20%, respectively (methods Wilks and Minresid).

Referring to the results previously published (Scarponi et al., 1981, 1982), one can note that both the order of entry of variables, in the stepwise methods, and the relative importance of variables in the discriminant functions, in the direct method, are changed. Obviously, the variability of the chemical parameters from year to year is responsible for this change. Possibly stable functions will be obtained when samples from many more vintage years will be considered.

Estimate of the Expected Actual Error Rate. The results of this estimate obtained by the learning set/test set partition method are reported in Figure 2. It can be seen that results are not significantly affected by the percentage of cases grouped in the training set (60–95% inclusive). On the average, the expected actual error rate is estimated about 17.5%. Being 18.4%, the error estimate obtained by the jackknife procedure (see Table IX), it can be concluded that the two methods are at present almost coincident.

***k* Nearest-Neighbor Rule.** Results of the kNN classification, with *k* equal to 1, 3, and 5, are reported in Table IX. For comparison purposes the results of the jackknife procedure in discriminant analysis are reported also. It can be observed that performances of the two methods are

Table IX. Results of the kNN Analysis: Comparison with Discriminant Analysis (Jackknife Procedure)

method	actual group	no. of samples	predicted group membership: Soave	Prosecco	Verduzzo	correct classification, %
1 nearest neighbor	Soave	32	30	1	1	93.8
	Prosecco	47	3	38	6	80.9
	Verduzzo	35	2	7	26	74.3
						total: 82.5
3 nearest neighbor	Soave	32	30	1	1	93.8
	Prosecco	47	3	41	3	87.2
	Verduzzo	35	2	7	26	74.3
						total: 85.1
5 nearest neighbor	Soave	32	30	1	1	93.8
	Prosecco	47	2	43	2	91.5
	Verduzzo	35	3	6	26	74.3
						total: 86.8
discriminant analysis (jackknife procedure)	Soave	32	31	0	1	96.9
	Prosecco	47	5	37	5	78.7
	Verduzzo	35	3	7	25	71.4
						total: 81.6

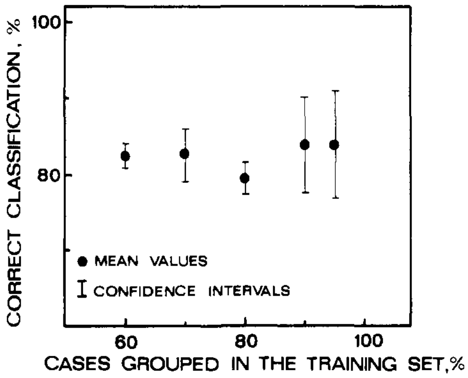

Figure 2. Percentage of correct classification of cases grouped in the test set against the percentage of cases grouped in the training set.

practically coincident. In the kNN rule, the increment in the correct classification obtained when one passes from $k = 1$ to $k = 5$ is possibly due to the larger sample size of Prosecco wine with respect to Soave and Verduzzo [see Coomans and Massart (1982a)].

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