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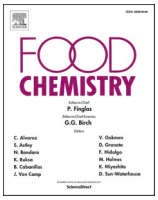

Modeling grape taste and mouthfeel from chemical composition

Sara Ferrero-del-Teso [a], Alejandro Suárez [a], Chelo Ferreira [b], Daniele Perenzoni [c], Panagiotis Arapitsas [c], Fulvio Mattivi [c,d], Vicente Ferreira [b], Purificación Fernández-Zurbano [a], María-Pilar Sáenz-Navajas [a,*]

[a] Instituto de Ciencias de la Vid y del Vino (ICVV) (Universidad de La Rioja-CSIC-Gobierno de La Rioja), Carretera de Burgos Km. 6, Finca La Grajera, 26007 Logroño, La Rioja, Spain
[b] Laboratorio de Análisis del Aroma y Enología (LAAE), Department of Analytical Chemistry, Universidad de Zaragoza, Instituto Agroalimentario de Aragón (IA2) (UNIZAR-CITA), c/ Pedro Cerbuna 12, 50009 Zaragoza, Spain
[c] Research and Innovation Centre, Food Quality and Nutrition Department, and Technology Transfer Centre, Fondazione Edmund Mach, via E. Mach 1, 38010 San Michele all'Adige, Italy
[d] University of Trento, Department of Cellular, Computational and Integrative Biology, CIBIO, 38122 Trento, Italy

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ABSTRACT

This research aims at predicting sensory properties generated by the phenolic fraction (PF) of grapes from chemical composition. Thirty-one grape extracts of different grape lots were obtained by maceration of grapes in hydroalcoholic solution; afterward they were submitted to solid phase extraction. The recovered PFs were reconstituted in a wine model. Subsequently the wine models, containing the PFs, were sensory (taste, mouthfeel) and chemically characterized.

Significant sensory differences among the 31 PFs were identified. Sensory variables were predicted from chemical parameters by PLS-regression. Tannin activity and concentration along with mean degree of polymerization were found to be good predictors of dryness, while the concentration of large polymeric pigments seems to be involved in the "sticky" percept and flavonols in the "bitter" taste. Four fully validated PLS-models predicting sensory properties from chemical variables were obtained. Two out of the three sensory dimensions could be satisfactorily modeled. These results increase knowledge about grape properties and proposes the measurement of chemical variables to infer grape quality.

1. Introduction

Perceived intrinsic quality of wine is driven by volatile and non-volatile compounds involved in the formation of aroma, taste, mouthfeel and color (Sáenz-Navajas et al., 2015). Wine aroma is the result of aroma and aroma precursors in the raw grape material, the action of microorganisms during alcoholic and malolactic fermentation, and aging. Wine taste, mouthfeel, and color are driven principally by phenolic compounds present in grapes and their interaction with other wine components (e.g., polysaccharides, acids, alcohol or aroma among others). Grape phenolic compounds are extracted mainly from skins and seeds during the maceration and fermentation processes. The underlying aromatic and phenolic composition of the grape berry has an important impact on the final intrinsic quality of wines. Grape growers, winemakers, and researchers seek to infer wine quality from both the sensory and the chemical properties of wine grapes. For this purpose, classical oenological measures are traditionally employed in addition to berry-tasting with the specific aims of evaluating both the evolution of grape properties during ripening and the grapes potential with which to elaborate wines. The determination of chemical parameters is usually limited to a reduced number of variables (total acidity, color intensity, total polyphenolic index or pH among others), which does not produce an overall quality potential classification of the grape lot. The sensory assessment of grapes is not a generalized practice probably because the proposed berry sensory assessment method (Mantilla et al., 2010; Rousseau & Delteil, 2000) has two main limitations that make the generation of reliable results difficult. The first is the use of a reduced number of panelists to carry out the sensory evaluation and second the lack of grape representativeness, because generally, in each evaluation one expert analyses a relatively reduced number of berries. These limitations, related to the sensory characterization of grapes, could be overcome by extracting the main sensory-active compounds of grapes,

* Corresponding author.
E-mail address: mpsaenz@icvv.es (M.-P. Sáenz-Navajas).

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mainly aroma, aroma precursors and phenolic compounds from a representative sample of grapes and characterize their sensory attributes with a sufficient number of panelists.

Recently, a study published by Alegre et al. (2020) shows a promising strategy for evaluating grape quality by focusing on the study of the aroma potential of wine grapes. The authors subjected reconstituted polyphenolic and aromatic fractions of grapes to accelerated hydrolysis in strict anoxia which yielded strong and differentiated aromas. This approach has proven useful for characterizing the potential aroma of grapes and thus, the evaluation of the potential aroma quality of grapes. The present research is focused on taste and mouthfeel induced by the phenolic fraction. Taste quality refers to the classical percepts of sweetness, sourness, bitterness, saltiness and umami which are the sensations that occur when the receptors present in the taste buds of the oral cavity are activated. Mouthfeel is related to tactile sensations generated in the oral cavity by the activation of the trigeminal nerve. The mechanisms modulating mouthfeel are the least understood; the study of "astringency" and its sub-qualities elicited by wine phenolic compounds have been scarcely found in the literature (Gawel, Oberholster, & Francis, 2000; Piombino et al., 2020; Sáenz-Navajas et al., 2017).

Little is known about the sensory properties elicited by the phenolic fraction of grape berries and their relationship with chemical variables. This knowledge would be valuable for inferring grape quality. Despite the large amount of instrumental chemical strategies currently available, perceived taste and mouthfeel cannot be predicted from chemical composition and consequently, these percepts can only be measured by sensory evaluation strategies. In this context, the present work aims at identifying chemical markers that allocate the inference of sensory properties driven by the phenolic fraction of grapes. One of the main challenges is to differentiate the phenolic fraction of grapes based on their mouthfeel properties. This could be attributed to the lack of reference materials illustrating these sensory characteristics. New strategies have been developed and successfully applied for describing wine mouthfeel. These strategies are alternative to classical descriptive analysis and do not require consensus among participants and thus do not need reference materials (Ares et al., 2014; Valentin et al., 2012; Varela & Ares, 2012). These strategies include non-verbal and verbal approaches. Among the first, similarity based methods such as "free sorting task" (Chollet, Valentin, & Abdi, 2014) have shown to be interesting approaches that highlight the most salient sensory differences among wines in terms of mouthfeel and taste perceptions (Sáenz-Navajas, Ferrero-del-Teso, Jeffery, Ferreira, & Fernández-Zurbano, 2020). Verbal-based strategies such as "rate-all-that-apply" (RATA) or its variant *rate-k-attributes* have been successfully applied to differentiate red wines in terms of mouthfeel and taste by wine experts without previous training (Sáenz-Navajas et al., 2020). In this verbal-based strategy, the list of specific attributes of each product is a main concern to discriminate among samples. To this end, a relatively ample list of mouthfeel and taste properties elicited by phenolic fractions was recently developed (Sáenz-Navajas et al., 2017) and its use in discrimination trials was confirmed to be valuable with a wide range of wines (Sáenz-Navajas et al., 2020) and phenolic fractions (Ferrero-del-Teso et al., 2020). However, the efficacy of this strategy to discriminate phenolic fractions directly from grapes has remained unevaluated.

In this context, the ability to differentiate the phenolic fraction of different grape lots based on their sensory properties following alternative sensory descriptive methods such as sorting task or rate-k-attributes method was the first hypothesis of the present study. The second hypothesis was the ability to predict sensory differences elicited by grape phenolic fractions from chemical measurements. To test these hypotheses a wide range of Garnacha Tinta and Tempranillo Tinto grapes harvested at different maturation points from distinct geographic origins were selected in order to represent a large variability in their chemical composition. Tempranillo Tinto typically presents higher levels of phenolic compounds than Granacha Tinta (Santesteban, Miranda, & Royo, 2011); the variability endowed by the range of harvests should also provide a wide array of taste and mouthfeel sensory attributes. The phenolic fraction (PF) of 31 grape batches (15 for Tempranillo Tinto and 16 for Garnacha Tinta) was obtained following the strategy proposed by Alegre et al. (2020). Reconstituted PFs were sensory evaluated following both verbal and non-verbal strategies and sensory descriptors derived from the verbal task were predicted by PLS-regression models from chemical variables.

2. Material and methods

2.1. Samples

2.1.1. Grapes

During the 2017 harvest, two different varieties, Tempranillo Tinto and Garnacha Tinta from different regions (DO Ribera del Duero and DOCa Rioja for Tempranillo Tinto and DOCa Rioja and DO Somontano for Garnacha Tinta) were harvested by hand from distinct blocks in different dates. For the Tempranillo Tinto variety, a total of 15 samples from five different blocks and in three different weeks were collected. For Garnacha Tinta, 16 samples from eight blocks were harvested in two different dates, each separated by one week (Table S1 and Table S2 of the Appendix B in Supporting Information are detailed lists of the samples).

2.1.2. Preparation of grape extracts

For each sample, ten kilograms of grapes were first destemmed, and 8.5 Kg of the destemmed grapes were macerated with 5 g/hL of potassium metabisulfite and ethanol 15 % (p/p). After one week of maceration at 5 °C samples were pressed with a hydraulic wine press of 15 Kg of capacity and stored at 5 °C in the dark. Two weeks later, when the solids were precipitated, samples were bottled. For each grape batch, eight 750 mL-bottles were obtained as average (corresponding to a yield of 70%).

2.1.3. Preparation of phenolic fractions (PF)

A volume of 750 mL of each grape extract was centrifuged at 4500 rpm, 10 °C for 20 min after which were separated from the alcohol in a rotary evaporator system (8 mbar, 28 °C, 30 min). The resulting dealcoholized extracts (containing no more than 2% ethanol) were passed through a 10g C18 prepared cartridge (Waters-Sep Pak-C18 35 cc). For cartridge conditioning, methanol followed by milli-Q water with 2% ethanol were employed. Then, the whole sample was loaded, and washed with milli-Q water pH 3.5 to remove sugars, amino acids, acids and ions. Cartridges were finally dried by letting air pass through them and phenolic fractions (PF) were recovered with 100 mL of ethanol as described by Alegre et al. (2020).

2.1.4. Preparation of samples for sensory analysis

PFs (coming from 750 mL of grape extract and eluted with 100 mL of ethanol in the SPE system) were twice concentrated by rotatory evaporator system (i.e., the resulting PFs were 15 times concentrated). Then, PFs were reconstituted to their original volume in a solution prepared with mineral water, 1 g L^{-1} of tartaric acid reaching 7% of ethanol concentration and pH was adjusted to 3.7.

The level of ethanol in the hydroalcoholic model solution (7%) was selected in preliminary bench top tastings. It corresponds to the minimal level of ethanol able to induce the lowest "burning" and "hot" effect that is able to mask other sensations.

2.2. Chemical analysis

Chemical information is detailed in Appendix A of the Supplementary material.

2.2.1. Conventional oenological parameters

The concentrated PFs were reconstituted in a 3.7 pH solution prepared with 5 g L^{-1} tartaric acid, milli-Q water, and hydroalcoholic solution (12%, v/v).

Total polyphenol index (TPI) was estimated as absorbance at 280 nm (Ribéreau-Gayon, 1970) and color intensity (CI) as the sum of absorbance at 420, 520 and 620 nm (Glories, 1984).

2.2.2. Analysis of anthocyanin-derived pigments

Determination of monomeric (MP), small polymeric pigments (SPP), and large polymeric pigments (LPP) in wines and fractions was carried out as described by Harbertson, Picciotto, & Adams (2003).

2.2.3. Characterization of tannins

Tannin activity was calculated as the specific enthalpy of interaction between tannins and a hydrophobic surface (polystyrene divinylbenzene HPLC column), as proposed by Revelette, Barak, and Kennedy (2014). The concentration of tannin and pigmented tannins were also determined, and were reported in (−)-epicatechin equivalents. Detailed information of the chromatographic conditions employed can be found in Appendix A of the Supplementary material.

2.2.4. Mean degree of polymerization by phloroglucinol reaction

The protocol was followed according to Arapitsas et al. (2021). Detailed information about the method is provided in Appendix A of the Supplementary material and chromatograms illustrated in Fig. S1 of Appendix B.

The mean degree of polymerization (mDP) was calculated as the ratio between total units (extension + terminal) to terminal units (calculated as the difference between monomers before and after the phloroglucionolysis nucleophilic reaction).

2.2.5. UHPLC-MS/MS quantification of low molecular weight polyphenols

The quantification of anthocyanins was carried out using the method described by Arapitsas et al. (2012). All the compounds were quantified as equivalents of malvidin-3-*O*-glucoside.

For the determination of flavanols, flavonols and hydroxycinnamic acids the method described by Vrhovsek et al. (2012) was employed.

Chromatographic conditions of both methods are detailed in Appendix A of the Supplementary material and chromatograms illustrated in Figs. S2 and S3 of Appendix B.

2.3. Sensory characterization

Two different sensory strategies were followed: sorting task (non-verbal) and rate-k-attributes (verbal). Both tasks were carried out by 21 wine experts from Rioja area, Spain (18 women and 3 men, ranging in age from 26 to 55, with an average age of 39). They were all established winemakers with extended experience in wine production and tasting. Samples were served in normalized dark approved wine glasses (German Institute for Normalization, DIN) labeled with 3-digit random codes, in a randomized distinct order of presentation for each participant. Samples were served at room temperature and evaluated in a ventilated, air-conditioned tasting room (approximately 20 °C).

Panelists were instructed to put the sample in the mouth and to gently distribute it during five seconds throughout the oral cavity (as a mouthwash) to reach the entire surface of the mouth (including the mouth wall, gums, back palate and tongue). After expectorating the sample, panelists had to wait one minute before rating the sample. After each sample, they were required to follow a mandatory rinsing protocol with mineral water and pectin (1 g L^{-1}) before tasting the next sample (Colonna, Adams, & Noble, 2004). Although samples were odorless, they were instructed not to smell samples in an orthonasal manner.

Participants were informed that samples had been prepared in the laboratory and were not commercial wines. They were also required to sign a consent form prior to undertaking the sensory testing. They were neither informed about the objective of the study nor paid for their participation.

2.3.1. Sorting task

Two sorting task sessions (30 min each) were held on the same day (separated by at least 15 min), devoted to Tempranillo Tinto and Garnacha Tinta Garnacha sample sets, respectively.

In each session, participants were simultaneously presented with all samples (15 for Tempranillo Tinto and 16 for Garnacha Tinta) and were asked to sort them on the table according to similarities in the sensations perceived in mouth (mouthfeel and taste). Participants could form as many groups as they wished (minimum of two groups) and put as many samples as they wanted in each group (groups could be formed by only one sample). After that, they were asked to note the three-digit codes of the samples belonging to each group on a paper sheet and were asked to describe the groups they formed with their own words (maximum of three terms per group).

2.3.2. Rate-k-attributes

Panelists attended two sessions (35 min each) held on different days; one for each variety. Each session was split into two parts separated by an imposed pause of 15 min. Samples were characterized following a rate-k-attribute method with a list of 23 taste and mouthfeel related attributes (Table S3 of the Appendix B in Supporting Information) that had been previously developed (Sáenz-Navajas et al., 2017). Participants were asked to taste and rate the intensity of a maximum of five attributes appearing in each sample on a 7-point scale (1 = not intense; 7 = very intense). Attributes that were not rated were allocated a value of zero when collecting data. To avoid bias due to order of presentation, attributes on the list appeared in a distinct randomized order for each participant.

2.4. Data analysis

2.4.1. Sorting task

For each participant, results were encoded in an individual similarity matrix (wines × wines) with each cell indicating weather two wines were put in different groups or in the same group (0 and 1, respectively). These individual matrices were summed across participants; the resulting co-occurrence matrix represents the global similarity matrix, where larger numbers indicate higher similarity between samples and the main diagonal accounts for the number of participants. The resulting co-occurrence matrix was submitted to a non-parametric MDS analysis in order to obtain a spatial representation of the samples. Hierarchical cluster analysis with the Ward criterion was performed on all the MDS dimensions.

Terms derived from the description of the groups were analyzed. First an initial list was built with all the terms elicited by participants. This list was reduced by omitting adverbs and words with hedonic or emotional character. Then, a lemmatization process was performed; words sharing the same lemma or root were grouped. Finally, a triangulation process was followed individually by three experienced researchers to achieve a final consensual list of terms. Terms belonging to the same semantic category were grouped, the frequency of citation of each consensual term was calculated, and only those cited by at least three panelists (15% of the panel) were considered. Chi-square ($\chi 2$) test was applied for calculating significance differences ($P < 0.05$) among clusters. In addition, two-way ANOVAs (panelists as the random and cluster as the fixed factors) were calculated with the scores of the 23 terms obtained by rate-K-attribute method to get an alternative characterization of clusters derived from the sorting task. For significant attributes ($P < 0.05$), pair-wise comparison test (Fisher test) was applied (5% risk).

2.4.2. Rate-k-attributes

To find discriminate attributes a two-way ANOVA (panelists as the

random and samples as the fixed factors) was calculated for each of the 23 attributes of the list. Then, for discriminate attributes, pair-wise comparison test (Fisher test) was applied (5% risk) for significant effects. A principal component analysis (PCA) based on the correlation matrix was carried out with the mean intensity scores (n = 21) of the significant attributes. A hierarchical cluster analysis (HCA) with the Ward criteria was finally applied to all dimensions derived from the PCA. To identify the attributes defining clusters, a two-way ANOVA with the scores of attributes was calculated with panelists as the random factor and cluster as the fixed factor. For significant attributes ($P < 0.05$), pair-wise comparison test (Fisher test) was applied (5% risk).

2.4.3. Comparison of sensory strategies

The degree of similarity between the two sensory spaces derived from the sorting task and the rate-k-attributes was calculated employing an RV coefficient (Robert & Escoufier, 1976) and the Pearson correlation coefficients between the dimensions of both sensory spaces.

2.4.4. Relationship between chemical and sensory variables

To establish relationships between mouthfeel related attributes and chemical variables, the 31 samples (15 Tempranillo Tinto and 16 Garnacha Tinta) were considered.

Firstly, to find discriminate attributes among the sample set, two-way ANOVAs (panelists as the random and samples as the fixed factors) were calculated for each of the 23 attributes of the list. Then, for discriminate attributes, pair-wise comparison test (Fisher test) was applied (5% risk) for significant effects. On the first three PCs a PCA was carried out with the mean intensity scores (n = 31) of the significant sensory attributes with VARIMAX rotation. Rotation eases the interpretation of results by maximizing high- and low-value factor loadings and minimizing intermediate values. Further Pearson correlation coefficients (r) and their significance were calculated between the significant mouthfeel related attributes among samples (n = 6), PCA dimensions (n = 3), and the calculated chemical variables (n = 45).

The six sensory attributes and the three PCA sensory dimensions were predicted by regressing calibration from chemical variables by PLS-regression attending to the following model:

$$Y = XB + F$$

where for a sample size n ($n = 31$), $X_{(31,45)}$ represents the input matrix, $Y_{(31,6)}$ the output matrix with the chemical variables, $B_{(45,6)}$ is the matrix of regression coefficients and $F_{(31,6)}$ the matrix of residuals. Single response models are analyzed. Then, single Y – variable Partial Least Square regression method is used for every sensory variable and the 45 chemical variables (X).

Input variables X have been filtered applying a 7 points window Stavizki-Golay smoothing; and, they have been standardized to comparable noise levels. Likewise, sensory variables $y_{i;1\leq i\leq 6}$ have been standardized.

Variable selection has not been considered to avoid the problem of overfitting. The model was validated using full cross validation.

All the analyses have been carried out with Unscrambler X 10.5.1, Matlab R2018a, R 4.0 and XLStat v2018.

3. Results and discussion

3.1. Sensory characterization of Tempranillo Tinto and Garnacha Tinta sample sets

The first objective of the present research was to evaluate the capacity of the proposed chemosensory strategy to differentiate among distinct sample sets. Therefore, 15 and 16 phenolic fractions of the Tempranillo Tinto and Garnacha Tinta sample sets, respectively, were obtained and characterized with the two sensory strategies.

3.1.1. Tempranillo Tinto sample set

In the sorting task carried out with the 15 Tempranillo Tinto samples, participants formed 2 to 8 groups; 5 on average. Two samples were grouped together a maximum of nine times (43% of participants). Fig. 1a presents the dendrogram derived from the cluster analysis calculated with all the MDS dimensions that the sorting task data yielded. Four main groups were identified containing three, four, two and six samples, respectively. Nine attributes including “dry” (62% of maximum citations for a given wine), “bitter” (52%), “astringent” (52%) and “sour” (43%), followed by “sweet” (19%), “watery” (19%), “persistent” (14%), “sticky” (14%) and “coarse” (14%) were among the most cited to describe the groups. Based on the highest frequency of citation for each cluster, cluster 1 was mainly described as “dry” (48%) and “bitter” (41%), cluster 2 as “dry” (50%), cluster 3 as “dry” (40%) and “astringent” (40%), and cluster 4 was mainly characterized as “astringent” (44%) and “sour” (31%). These results suggest that the most salient attributes of the set of Tempranillo Tinto PFs are “dry”, “bitter”, “astringent” and “sour”. However, no significant effect was observed among clusters according to Chi-square (χ^2) test. Similarly, no significant effect of cluster was found for any of the attributes derived from the rate-k-method. Clusters obtained from MDS-HCA illustrate important sensory differences among the Tempranillo Tinto PFs herein studied. However, the specific attributes explaining such significant differences could not be identified. This, firstly suggests that differences among samples could not be verbalized in the description step of the sorting task and secondly, that the list of terms of the rate-k-attributes method did not contain the specific term differentiating among samples.

Regarding the results derived from rate-k-attributes methodology, ANOVA results showed six significant attributes to differ among clusters: “coarse” ($F = 7.35$; $P < 0.0001$), “dusty” ($F = 6.50$; $P < 0.001$), “burning” ($F = 6.23$; $P < 0.001$), “bitter” ($F = 4.56$; $P < 0.005$) and “fleshy” ($F = 3.91$; $P < 0.05$), as well as “sticky” ($F = 2.54$; $P < 0.1$) when relaxing the criteria for significance. Further cluster analysis calculated on all PCA dimensions (PCA computed with the six significant terms) showed four main clusters of samples (Fig. 1b) with different sensory properties. Cluster 1, formed by two samples was mainly described as “sticky” (average = 1.26) and “dusty” (average = 0.98), cluster 2, formed by three samples, presented the lowest score value for the term “bitter” (average = 1.36) (Fig. S4 of Appendix B of the Supplementary material). Cluster 3, including two samples, was significantly characterized by “burning” (average 0.76) and reached the highest score for “bitter” (average = 2.43). Cluster 4, which is formed by eight samples, presented the lowest scores for the attributes “coarse” and “fleshy”.

The two sensory spaces obtained by two different sensory strategies were compared. Therefore, MDS dimensions obtained from sorting task and the PCs derived from sensory characterization by rate-k-attributes method were employed to determine the RV coefficient. The RV coefficient was found to be 0.398 ($P > 0.1$), which indicates that the configurations of the two sensory spaces were different. A possible explanation for this result is that these two approaches, verbal (rate-k-attributes) and non-verbal (sorting task), induce participants to adopt different strategies when characterizing samples. In the sorting task panelists follow a holistic strategy in which overall and most salient differences among samples (Valentin, Chollet, Nestrud, & Abdi, 2017) are identified. While the rate-k-attribute methodology, which follows an analytical approach, is able to identify subtler and specific sensory differences among the sample set (i.e., “coarse”, “dusty” “burning”, “bitter”, “fleshy” and “sticky”).

3.1.2. Garnacha Tinta sample set

Based on the sorting task, the 16 Garnacha Tinta samples were grouped into 3 to 8 groups, 5 on average, similar to the Tempranillo Tinto sample set; illustrating the sensory variability associated with Garnacha Tinta PFs. Any two samples were grouped together a maximum of 10 times (48% of participants). As Fig. 2a shows, four main groups were identified. In this case, cluster 1 was formed by six samples,

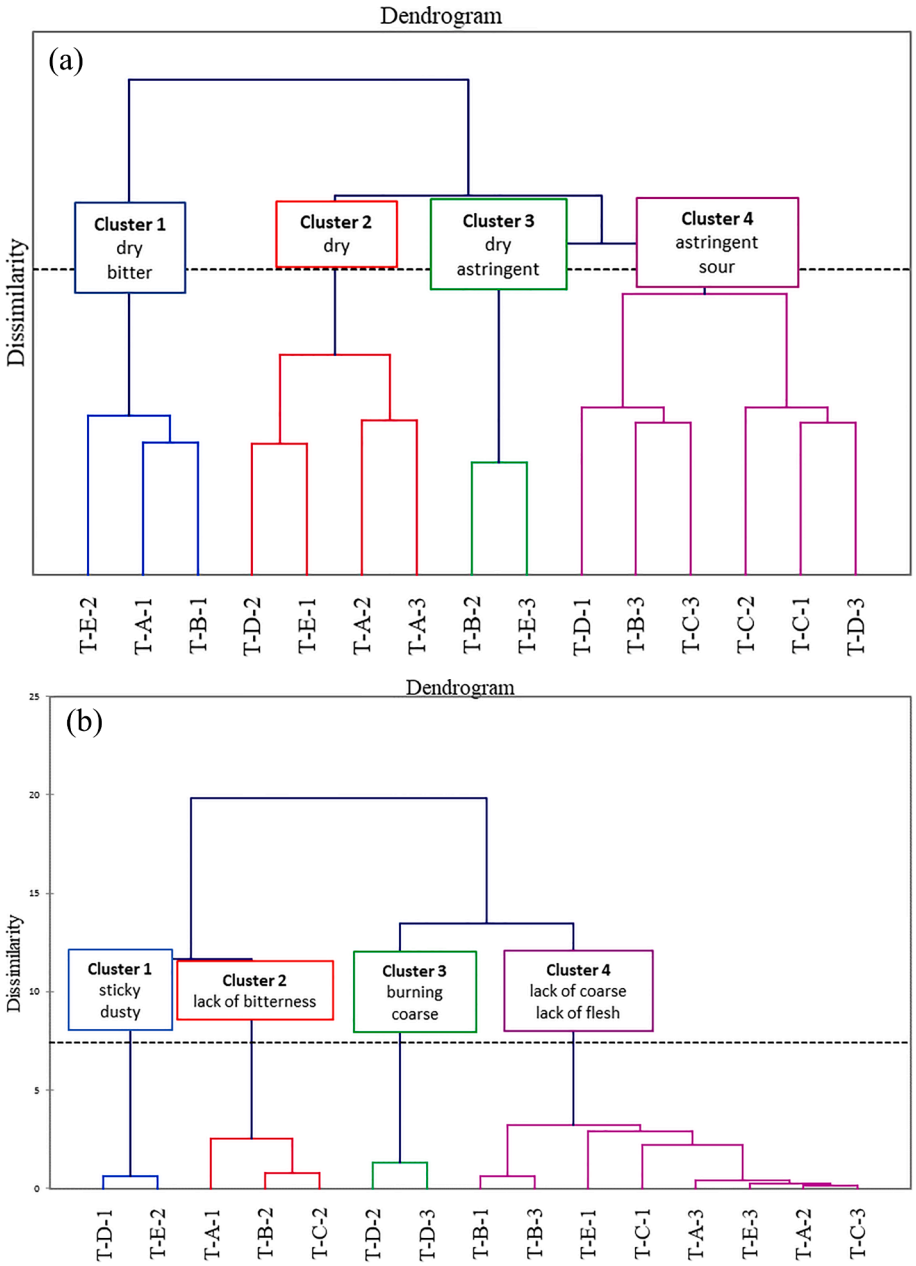

Fig. 1. Dendrogram of the polyphenolic fractions of Tempranillo Tinto derived from the sensory descriptions of samples by a) sorting task and b) rate-k-attribute methods. Attributes describing clusters refer to a) terms with highest average frequency of citation-FC-calculated with individual FC of wines which belong to each cluster and b) terms with highest average scores calculated with wines that belong to each cluster.

cluster 2 and 3 were built by three and five samples, respectively, and cluster 4 was composed of only two samples. The analysis of the attributes employed to describe the groups led to a final consensual list of 8 attributes including "dry" (62% of maximum citations for a given wine), "bitter" (52%), "sour" (48%), "astringent" (38%), and "watery" (24%), followed by "sticky" (19%), "sweet" (19%) and "alcoholic" (14%). Based on the highest frequency of citation for each cluster, all clusters were mainly described with the attributes "bitter" and "dry": 44% and 41% of citations for cluster 1, 41% and 51% for cluster 2, 43% and 48% for cluster 3, and 38% and 55% for cluster 4. Notably, cluster 2 reached the highest frequency of citation with "sour" (40%) and cluster 4 with "astringency" (31%). However, there was no significant attribute that differed among clusters according to chi-square (χ^2) tests. Distinctly, three significant attributes evaluated by rate-k-attribute method appeared to be significant among clusters derived from sorting task: "dry on the tongue" ($F = 3.956$; $P < 0.05$), "fleshy" ($F = 4.648$; $P < 0.05$), and "bitter" ($F = 2.467$; $P < 0.1$) (Fig. S5 in Appendix B of the Supporting Information). Cluster 3, which included five samples, was significantly characterized by "bitter" (average 2.05). Cluster 4, which was formed by two samples, presented the highest score for the attribute "dry on tongue" (average 1.26) and cluster 2, formed by three samples, was significantly characterized by "fleshy (average 0.37).

In regards to the results derived from rate-k-attributes, as occurred for the Tempranillo Tinto samples, three out of 23 attributes presented the highest scores among the 16 PFs studied: "dry" (max = 4.00), "bitter" (max = 2.33) and "dry on palate" (max = 2.14).

Fig. 2b illustrates the four clusters obtained from the hierarchical cluster analysis calculated on all the PCA dimensions. Even if this option (two clusters) was the most natural partition of the tree diagram, we chose the partition containing a total of four clusters as it permitted more precise descriptions of samples belonging to each of the groups. The ANOVA results identified significant differences among clusters for

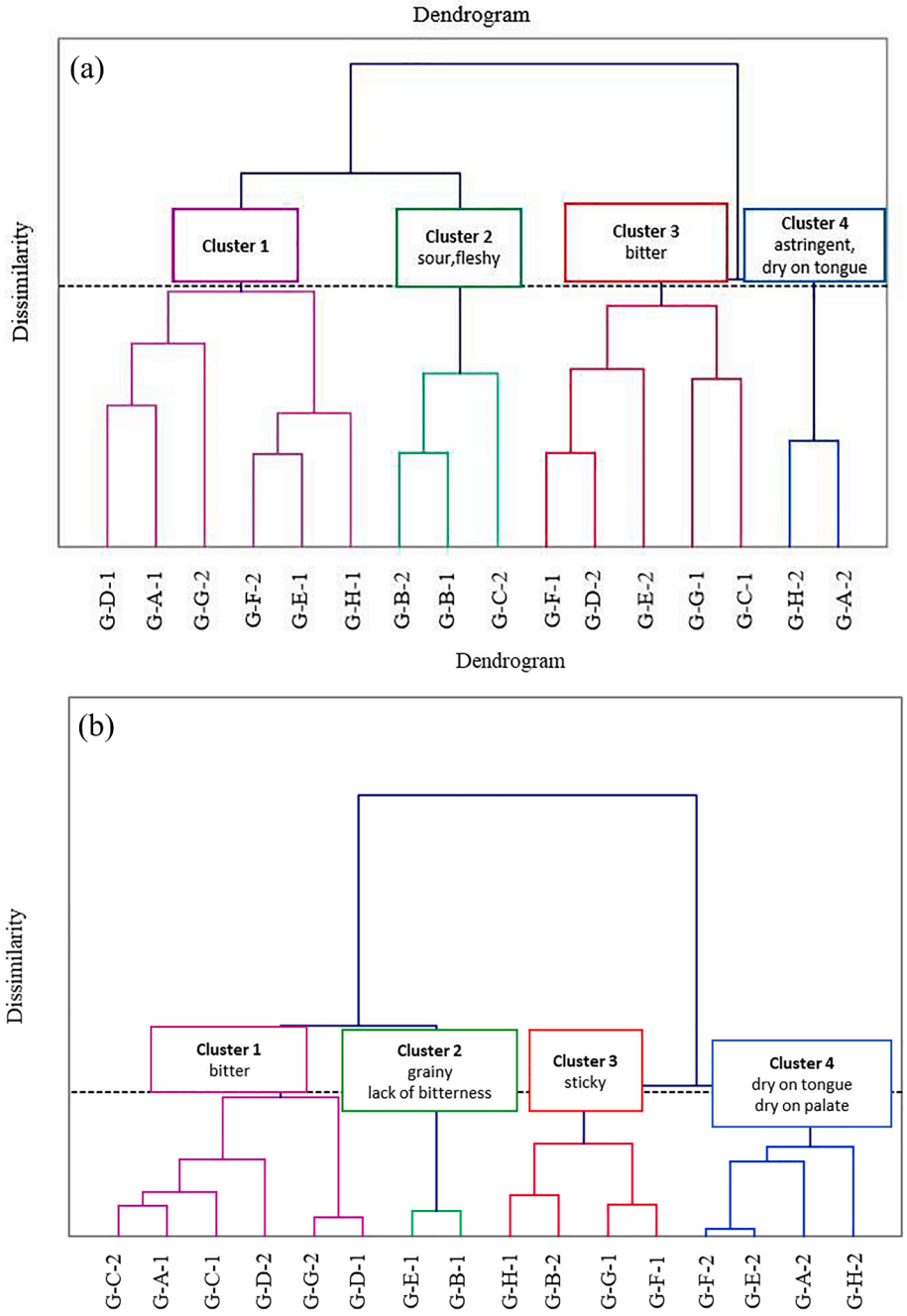

Fig. 2. Dendrogram of the polyphenolic fractions of Garnacha Tinta derived from the sensory descriptions of samples by a) sorting task and b) rate-k-attribute method. Attributes describing clusters refer to a) terms with highest average frequency of citation-FC-calculated with individual FC of wines which belong to each cluster and b) terms with highest average scores calculated with wines that belong to each cluster.

8 out of the 23 attributes evaluated, "dry" ($F = 9.577$; $P < 0.0001$), "bitter" ($F = 6.705$; $P < 0.001$), "dry on tongue" ($F = 6.363$; $P < 0.001$), "coarse" ($F = 5.445$; $P < 0.005$), "watery" ($F = 3.827$; $P < 0.05$), "grainy" ($F = 3.097$; $P < 0.05$), "sticky" ($F = 2.851$; $P < 0.05$) and "dry on palate" ($F = 2.480$; $P < 0.1$) when relaxing the criteria for significance (Fig. S6 of Appendix B of the Supplementary material).

Clusters 1 and 2 (formed by six and two samples, respectively) present the lowest scores for "dry", "dry on tongue", "sticky" and "coarse", yet the highest for "watery". Cluster 2 presents the lowest value for the attribute "bitter" (average = 0.86), and the highest for "grainy" (average = 0.26). Clusters 3 and 4 (formed by four samples each) presented the highest scores for the "dry" and "coarse" attributes. Cluster 3 was characterized by "sticky" (average = 0.96); while, cluster 4 was mainly described as "dry on tongue" (average = 1.12) and "dry on palate" (average = 1.67).

As in the Tempranillo Tinto sample set, when comparing the two sensory spaces derived from sorting and rate-k-attributes, differences are found according to the RV coefficient, which was not found to be significant ($RV = 0.420$; $P > 0.1$). This is consistent with the fact that although the sorting task approach can overcome the difficulties in verbalizing mouthfeel features of wine reported by various authors (Piombino et al., 2020; Sáenz-Navajas et al., 2017), only evident sensory differences among the studied samples can be identified. Distinctly, panelists follow an analytical strategy under the rate-k-attribute, in which more specific and subtle differences can be identified.

Overall, these results confirm our first hypothesis related to the

appropriateness of the proposed chemosensory strategy to identify intra-varietal sensory variability associated with phenolic fractions of grapes. The RATA methodology was more efficient in identifying subtle and specific sensory differences among PFs derived from the same variety.

3.2. Relationships between mouthfeel properties and chemical measurements

The main objective was to establish relationships and predict mouthfeel attributes from chemical variables of grape phenolic fractions. Therefore, the sensory data derived from the rate-k-attributes method and chemical variables for the two varieties were pooled together (total of 31 samples) in order to increase the robustness of the statistical tests.

3.2.1. Sensory dimensions and significant taste and mouthfeel properties

Six significant attributes among the 31 samples differed based on ANOVA results; "dry" ($F = 2.382$; $P < 0.0001$), "coarse" ($F = 2.277$; $P = 0.0002$) "bitter" ($F = 1.538$; $P = 0.035$), "dry on tongue" ($F = 1.485$; $P = 0.048$), "sticky" ($F = 1.477$; $P = 0.050$) and "watery" ($F = 1.374$; $P = 0.090$) when relaxing the criteria for significance. Fig. 3 shows the PCA calculated with these significantly different attributes. The first three dimensions, which explain 70% of the original variance, are considered significant according to Kaiser criterion (eigen value > 1). These three dimensions were rotated with VARIMAX algorithm to facilitate the interpretation of the results. The first dimension, after rotation (D1), explains 27% of the variance and is mainly positively contributed by the attributes "dry on tongue" (43% of contribution, $r = 0.842$) and negatively by "watery" (30%, $r = -0.700$). The second dimension, D2, presents 19 % of the variance, and is mainly formed by "bitter" (59%, $r = 0.824$) and "sticky" (37%, $r = 0.650$). The third dimension explains 24% of the variance; it is mainly built by the attributes "coarse" (54%, $r = 0.881$) and "dry" (31%, $r = 0.673$). These results identify the presence of three main independent and non-correlated mouthfeel and taste dimensions defining the sensory space of PFs.

3.2.2. Correlation between taste and mouthfeel dimensions and chemical parameters: correlation coefficients and PLS-modeling

Table 1 presents the correlation coefficients (r) between sensory attributes and dimensions and chemical variables. Table 2 contains the models that satisfactorily predict the sensory parameters (4 out of 9). Validated models explain more than 60% of original variance by full-cross validation which corresponds to high correlation coefficients (r) between predicted and measured values of at least 0.77. Explained variances by calibration, reach values at least of 80% ($r > 0.90$). Fig. 4 lists the chemical variables included in models and the sign and magnitude of their coefficients following a color code. Interestingly, two out of three independent, non-correlated sensory dimensions identified (D1 and D2 of the PCA calculated with Varimax rotation) could be successfully modeled.

The highest significant linear correlations ($P < 0.0001$) (Table 1) were found between two chemical variables (tannin activity and tannin concentration) and the attribute "dry" ($r = 0.68$ for both variables). The tannin activity was measured as the interaction of tannins with a hydrophobic surface (Barak & Kennedy, 2013). Interestingly, "dry on tongue" was also positively correlated with both chemical variables ($r = 0.55$ for tannin activity, $r = 0.50$ for tannin concentration), but to a lesser extent than for "dry". This is well in line with the PLS-models obtained for both the "dry" attribute and the PCA dimension D1 (related to dry on the tongue). Both models highlight the importance of tannin activity because this chemical variable presents the highest positive correlation coefficients, followed by tannin concentration (Fig. 4). These results partly differ from previous studies in which no significant correlation between the tannin concentration or tannin activity and wine dryness perception was found (Watrelot, Byrnes, Heymann, & Kennedy, 2016). However, the results observed in the present research with grape PFs are well in line with other studies carried out in our laboratory with red wines, where linear (Ferrero-del-Teso et al., 2019) and non-linear (Sáenz-Navajas et al., 2019) relationships between tannin activity and the global "astringency" attribute were reported.

Regarding the mean degree of polymerization of tannins (mDP), significant positive correlations were found with "dry" ($r = 0.62$) and "dry on tongue" ($r = 0.49$). Accordingly, the coefficient for mDP is very high for the D1 model, and especially for "dry". Thus, this data is in agreement with other studies (Arnold, Noble, & Singleton, 1980; Gawel, 1998; Peleg, Gacon, Schlich, & Noble, 1999; Vidal et al., 2003) where an increase in mDP resulted in an increase in the perceived overall astringency. A positive correlation was found between %PD and "watery" which is in line with the high positive correlation coefficient observed in the PLS-model. This could be explained in terms of decreases in the perception of astringency with higher %PD shown in previous research (Chira, Schmauch, Saucier, Fabre, & Teissedre, 2009; Lisjak et al., 2020; Vidal et al., 2003).

The term "sticky" ($r = 0.58$) as well as the second dimension (D2 contributed mainly by the attributes "sticky" and "bitter") of the PCA ($r = 0.40$) are correlated with the level of large polymeric pigments (LPP), which present high positive correlation coefficients in the D2 of the PLS-model (Fig. 4). Besides this, low molecular weight anthocyanins

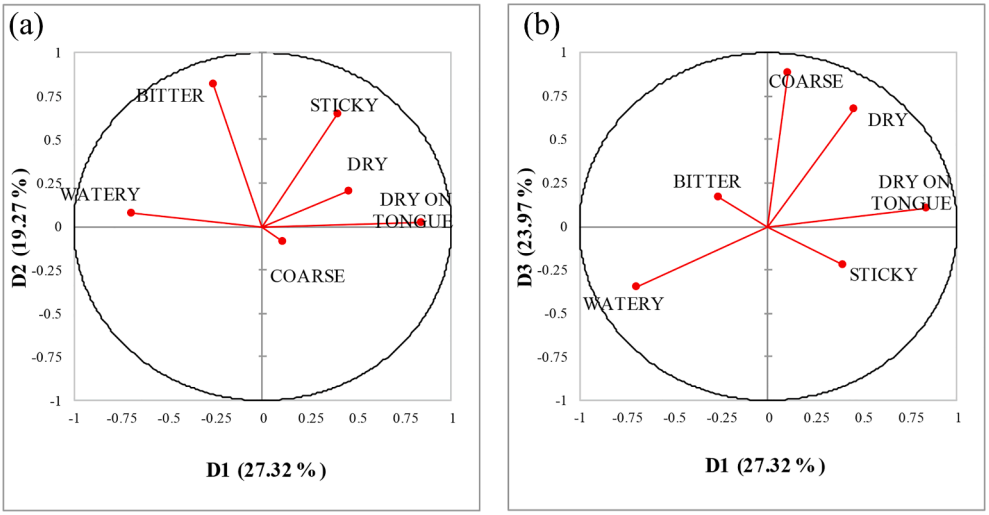

Fig. 3. PCA plots with the projection of rotated dimensions a) D1–D2 and b) D1-D3 performed for the significant terms of the 31 samples.

Table 1
Pearson correlation coefficients (r) calculated between chemical variables,[1] significant taste and mouthfeel attributes and sensory dimensions (i.e., principal components derived from PCA of 31 samples). Significant correlations ($P < 0.05$) are marked in bold.

	Sensory attribute						Sensory dimensions		
	Bitter	Dry	Dry on tongue	Sticky	Coarse	Watery	D1[2]	D2[3]	D3[4]
Tannin characterization									
TA	0.05	**0.68**	**0.55**	0.25	0.2	−0.27	**0.54**	0.23	0.28
Tannin concentration	0.24	**0.68**	**0.50**	0.23	0.15	−0.12	**0.40**	**0.37**	0.29
Pigmented tannin	0.20	**0.57**	0.34	0.10	0.20	−0.09	0.24	0.25	0.34
mDp	0.1	**0.62**	**0.49**	0.15	0.19	−0.20	**0.42**	0.21	0.30
%PD	0.02	−0.21	−0.04	0.03	−0.23	**0.47**	−0.19	0.07	−0.28
%G	−0.34	−0.13	−0.18	−0.13	0.19	−0.26	0.00	**−0.38**	0.09
Conventional oenological parameters									
TPI	0.21	**0.40**	0.17	0.17	0.19	−0.04	0.12	0.28	0.27
IC	0.31	**0.53**	0.29	0.19	0.11	−0.03	0.20	**0.40**	0.26
Anthocyanin-derived pigments									
MP	0.18	**0.54**	0.34	0.21	0.15	−0.09	0.28	0.30	0.25
LPP	0.10	0.27	0.25	**0.58**	−0.07	−0.27	**0.46**	**0.40**	−0.11
Anthocyanins									
Acylated	0.29	**0.48**	0.21	0.08	0.18	−0.05	0.11	0.31	0.33
Glycosylated	0.01	**0.51**	0.01	−0.04	0.3	−0.07	0.02	0.03	**0.44**
Vitisine A	−0.36	−0.02	0.00	−0.25	0.14	−0.25	0.09	**−0.44**	0.11
Flavonols									
Kaempferol	**0.43**	0.28	0.15	0.18	0.04	0.07	0.03	**0.45**	0.16
Myricetin	**0.47**	0.34	0.19	0.25	−0.1	0.00	0.12	**0.54**	0.07
Laricitrin	**0.39**	0.12	0.03	0.30	−0.12	−0.18	0.12	**0.46**	−0.03

[1] **TA**: Tannin activity (-J/mol), **Tannin concentration** (mg/L), **Pigments concentration** (mg/L), **TPI**: Total polyphenol index (a.u.), **IC**: Colour intensity (a.u.), **MP**: Monomeric pigments (a.u.), **LPP**: Large polymeric pigments (a.u.), **mDp**: mean degree of polymerization, **%PD**: percentage of prodelphinidins, **%G** percentage of galloylation, **Acylated** (mg/L): Delphinidin 3-(6″-acetyl)-glucoside, Cyanidin 3-(6″-acetyl)-glucoside, Petunidin 3-(6″-acetyl)-glucoside, Peonidin 3-(6″-acetyl)-glucoside, Malvidin 3-(6″-acetyl)-glucoside, Delphinidin 3-(6″-p-coumaroyl)-glucoside, Cyanidin 3-(6″-p-coumaroyl)-glucoside, Petunidin 3-(6″-p-coumaroyl)-glucoside, Peonidin 3-(6″-p-coumaroyl)-glucoside, Malvidin 3-(6″-p-coumaroyl)-glucoside, **Glycosylated** (mg/L):): Delphinidin 3-glucoside, Cyanidin 3-glucoside, Petunidin 3-glucoside, Peonidin 3-glucoside, Pelargonidin 3-glucoside, Malvidin 3-glucoside, Delphinidin 3,5-diglucoside, Cyanidin 3,5-diglucoside, Petunidin 3,5-diglucoside, Peonidin 3,5-diglucoside, Malvidin 3,5-diglucoside, **Vitisine A** (mg/L), **Kaempferol** (mg/L), **Myricetin** (mg/L), **Laricitrin** (mg/L).
[2] Mainly built by dry on tongue and watery.
[3] Mainly built by bitter and sticky.
[4] Mainly built by dry and coarse.

Table 2
Variables successfully modeled in the set by PLS regression, % of explained variance by full cross validation (and the % of explained variance), the number of PLSs included in each model and the root mean squared error of prediction.

Variable	% explained variance P (number of PLSs) [% explained variance C]	RMSEP [RMSE C][1]
Dry	64% (3) [82%]	0.63 [0.4]
Watery	66% (3) [86%]	0.58 [0.38]
D1 (dry on the tongue-watery)	66% (3) [81%]	0.67 [0.43]
D2 (dry/coarse)	63% (2) [80%]	0.57 [0.4]

[1] RMSE is given in z-units for a normal distribution. Given that 99.7% of normal values are between $z = -3$ and $z = 3$, a RMSE of 0.6 represents around 10% of the range.

measured by both Harbertson and Adams assay (MP, $r = 0.54$) as well as glycosylated ($r = 0.51$) and acylated ($r = 0.48$) anthocyanins, measured by chromatography, show significant positive correlations with "dry". While the highest coefficient in the PLS-models was that of the condensed anthocyanins (mainly dimers of anthocyanin-flavanol), which contribute significantly to the "dry" sensation. The high positive coefficients of MP presented by pigmented tannins and acylated anthocyanins in the D2 dimension model was a notable outcome. These results suggest the sensory importance of anthocyanins and anthocyanin-derived pigments in taste and mouthfeel sensations; they reinforce the results recently reported by other authors (Ferrer-Gallego et al., 2015; Ferrero-del-Teso et al., 2020; Paissoni et al., 2018).

Bitter perception is related to phenolics with low molecular weights such as flavonol, aglycones, and monomeric flavanols (Preys et al., 2006; Sáenz-Navajas et al., 2010). Results of this research are well in line with the previous research in the literature as significant positive correlations between flavonols such as kaempferol, myricetin, and laricitrin and the "bitter" attribute ($r = 0.43$, $r = 0.47$ and $r = 0.39$, respectively) were observed. Similarly, the model for the D2 dimension, to which bitterness greatly contributes, shows positive, high correlation coefficients for flavonols.

These results partly confirm our second hypothesis related to the relationships between taste and mouthfeel properties and chemical variables. Two (mainly D1 and D2) out of the three sensory dimensions representing taste and mouthfeel variability among the studied PFAs presented high and significant relationships with some of the chemical variables measured. Both sensory dimensions related to dryness and sticky/bitterness could be satisfactory modeled from the chemical variables studied. The exception was the third dimension (D3), in which "coarse" is the main contributor, from grape fractions. The individual term "coarse" presented no significant correlation with chemical variables; neither it, nor D3 could be satisfactorily modeled. It cannot be ruled out that this percept is driven by other molecules present in wine such as aroma or mannoproteins or their sensory or physical interactions with polyphenols which were not examined in the present research.

4. Conclusions

The current research presents a new chemosensory strategy for characterizing the sensory properties of phenolic fractions of grapes. This approach has shown to be efficient in differentiating grape phenolic fractions based on mouthfeel and taste properties; both inter- and intra-varietal.

The non-verbal and holistic sensory task, i.e., sorting task, highlighted the salient sensory differences among samples. While the more

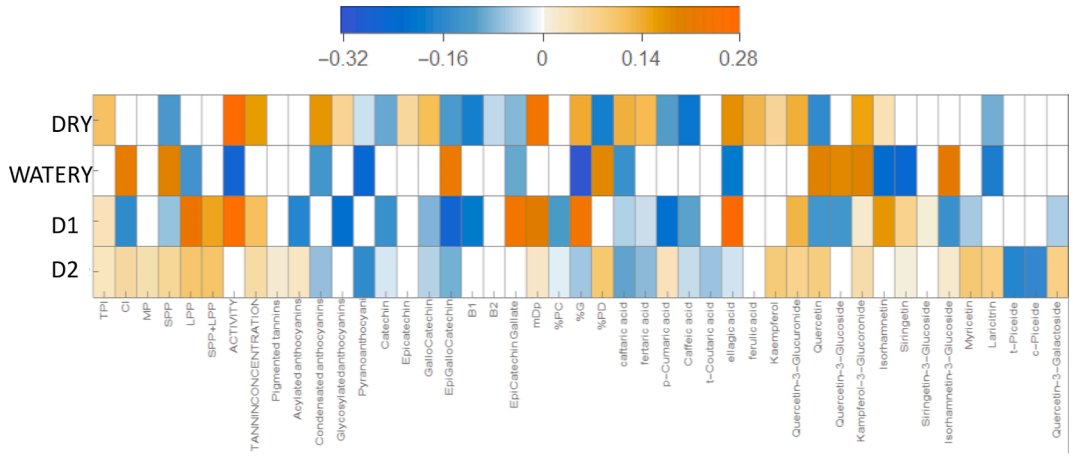

Fig. 4. Map with regression coefficients of variables included in validated PLS-models predicting sensory attributes or dimensions from chemical variables.

specific descriptors and subtle differences varying among Tempranillo Tinto ("coarse", "dusty", "burning", "bitter", "fleshy", "sticky") or Garnacha Tinta ("dry", "bitter", "dry on tongue", "coarse", "watery", "grainy", "sticky", "dry on palate") PFs could be identified by the rate-k-attribute methodology; a methodology which follows an analytical verbal strategy.

Three distinct, independent, non-correlated, sensory dimensions could be identified for the overall sample set: 1) "dry on tongue/watery", 2) "sticky/bitter" and 3) "dry/coarse".

Significant correlations and very satisfactory PLS models could be built to predict sensory variables from chemical parameters. Tannin activity and tannin concentration along with mDP of tannins proved to be good predictors of PF perceived dryness. Flavonols could have a good prediction power for the "bitter" attribute and the "sticky/bitter" dimension. In addition, low molecular weight anthocyanins seem to be involved in the formation of the "dry" attribute, whereas large polymeric pigments in the "sticky" attribute and the "sticky/bitter" dimension.

Distinctly, the "coarse" dimension could not be modeled which suggests that there are other macromolecules involved in the formation of this percept. Examination of the "coarse" dimension should be tackled in future research.

With these results, grape properties and intrinsic quality could be inferred with the measurement of chemical variables. This approach provides an interesting tool to assess grape quality.

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CRediT authorship contribution statement

Sara Ferrero-del-Teso: Investigation, Writing – original draft, Writing – review & editing. **Alejandro Suárez:** . **Chelo Ferreira:** Investigation. **Daniele Perenzoni:** Investigation. **Panagiotis Arapitsas:** Investigation, Methodology, Writing – review & editing. **Fulvio Mattivi:** Project administration, Writing – review & editing. **Vicente Ferreira:** Conceptualization, Project administration, Writing – review & editing, Funding acquisition. **Purificación Fernández-Zurbano:** Conceptualization, Project administration, Writing – review & editing, Funding acquisition. **María-Pilar Sáenz-Navajas:** Conceptualization, Investigation, Methodology, Writing – review & editing, Formal analysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2021.131168.

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