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TITLE:

Preliminary sensory characterization of the diverse astringency of mono-varietal Italian red wines and correlation of sub-qualities with chemical parameters

AUTHORS:

Paola Piombino[1]*, Elisabetta Pittari[1], Angelita Gambuti[1], Andrea Curioni[2], Simone Giacosa[3], Fulvio Mattivi[4,5], Giuseppina Paola Parpinello[6], Luca Rolle[3], Maurizio Ugliano[7], Luigi Moio[1]

*Corresponding author's e-mail: paola.piombino@unina.it, telephone number: +39 081 2532602

AFFILIATIONS:

[1]Department of Agricultural Sciences, Division of Vine and Wine Sciences, University of Naples Federico II, Viale Italia, 83100 - Avellino, Italy

[2]Department of Agronomy, Food, Natural Resources, Animals and Environment (DAFNAE), University of Padova, Italy

[3]Department of Agricultural, Forestry and Food Sciences, University of Torino, Italy

[4]Center Agriculture Food Environment, University of Trento, San Michele all'Adige, Italy

[5]Fondazione Edmund Mach, Research and Innovation Centre, Department of Food Quality and Nutrition, San Michele all'Adige, Italy

[6]Department of Agricultural and Food Sciences, University of Bologna, Italy

[7]Department of Biotechnology, University of Verona, Italy

AUTHORSHIP DECLARATION:

All authors have contributed significantly, and all authors are in agreement with the manuscript.

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RUNNING TITLE: Astringency diversity of Italian red wines

Preliminary sensory characterization of the diverse astringency of mono-varietal Italian red wines and correlation of sub-qualities with chemical parameters

ABSTRACT

Background and Aims*:* Italy is the richest grape producing country in terms of cultivars. Our aim was to describe the astringency diversity of Italian red wines from 11 varieties (Teroldego, Corvina, Raboso, Nebbiolo, Sangiovese, Sagrantino, Montepulciano, Cannonau, Aglianico, Primitivo, Nerello) and to test correlations between in-mouth sensory variables and chemical parameters.

Methods and Results: A sample sub-set was selected by sorting and assessed on astringency sub-qualities and tastes. Inter-varietal differences were detected for 6 out of 7 sub-qualities: 3 diverse intensities for drying, 2 for harsh, unripe, dynamic, complex and velvet, none for particulate. Discriminant analysis showed that sub-qualities allowed a good discrimination of the wines according to the variety. Well reclassified samples (88%) were considered to develop mono-varietal “Astringency spectra”, profiles describing the balance among sub-qualities. Correlations highlighted that neither phenols nor proanthocyanidins can predict the perception of all astringency nuances.

Conclusions: For some mono-varietal wines, it was possible to identify a pattern of astringency features likely linked to the variety.

Significance of the Study: This work adds insights to the understanding of astringency sub-qualities while enhancing the knowledge about Italian wines. Results may support winemakers awareness on wines from native varieties, and help in building models of astringency.

Keywords: mono-varietal Italian red wines; diversity; astringency sub-qualities, “Astringency spectra”; sensory characterization.

1. INTRODUCTION

According to the OIV Focus (2017), Italy is the grape producing country with the highest number of cultivars. This results from centuries of human selection, which led to a tight cultivar-environment relationship. This rich ampelographic heritage composed nowadays of around 500 cultivars, considering those listed in the Italian National Catalogue of Grapevine Varieties (Lacombe et al. 2011), includes red grapes with very different compositions in terms of polyphenols (Mattivi et al. 2002, Mattivi et al. 2009). The corresponding wines present a wide spectrum of sensory features, including diverse astringency. This means diversified mouth-feel characteristics, as reported in the different Disciplinary Regulations of Italian wines (https://www.politicheagricole.it/). Some of these grapes are used for the production of worldwide renowned wines, such as Chianti or Barolo, which in spite of their richness in tannins and intense mouth-feel, are appreciated by consumers and represent some of the best examples of Italian red wines (Piacenza et al. 2009, de Luca et al 2019). At the end of the last century, there was the renaissance of Italian wines and, at the beginning of the new century, a rising trend of propagation (a parameter evaluating the market interest on cultivars) was observed (Mannini 2004). Nebbiolo changed its yearly nursery production from 300.000 graftings to 1.700.000, Aglianico from 200.000 to 1.000.000, Primitivo from 100.000 to 1.000.000 . Nowadays, there is an interest through Italian varieties also outside the Italian territory. As an example, some white and red (eg. Sangiovese, Montepulciano, Barbera, Lambrusco, Nero d'Avola, etc.) native grapes have been included among the winery grown fruits in several Australian regions (eg. Riverina, Barossa Valley, McLaren Vale, Riveland, King Valley, etc.) (National Vintage Report, 2019).

In light of this wide biodiversity, this rise in high quality products and economic potential, it is quite surprising that the astringency of Italian red wines was never systematically investigated and compared from a sensory point of view. Astringency is of great interest because it represents an intrinsic parameter of red wines that is strictly linked to its perceived quality (Sáenz-Navajas et al. 2011, and references therein). Several Italian wines were studied in terms of chemical composition of polyphenols. Data about their astringency as sensory parameters can only be recovered for some of them in a fragmentary way as results on the impact of viticultural/enological practices on the sensory profile (Boselli et al. 2004, Gerbi et al. 2006, Gambuti et al. 2009, Torchio et al. 2010, Pagliarini et al. 2013, Patrignani et al. 2017). Moreover, data on different cultivars are not comparable because of the methodological/terminology differences (oenology, sensory techniques, phenolic analysis, vocabulary, etc.). This lack is one of the reasons why today it is not really possible to identify specific astringency characters as one typical feature of any Italian wine. Without this knowledge, winemakers are not supported neither by the knowledge of strengths and weakness of a specific grape, nor by a shared sensory model. In the current market, the ability to associate a certain product to specific sensory attributes and territories is often a vehicle to commercial success. Then, a more comprehensive characterization of the astringency of Italian red wines would provide an opportunity to support/consolidate their international image, with positive commercial outcomes. Indeed, the commercial value of a wine is related to its intrinsic (e.g. sensory features) and extrinsic (e.g. geographical origin) characteristics and both of them are drivers for wine purchase and repurchase (Charters and Pettigrew 2007, Mueller et al. 2010, Sáenz-Navajas et al. 2016). Among the different sensory characteristics of red wine, astringency gives a key contribution to its perceived quality, although it is one of the most difficult sensory parameter to characterize and understand, due to the complex mechanisms underpinning its perception (Ployon et al. 2018). The wide complexity of this sensation, has been hierarchized in a vocabulary including 7 categories and 33 terms (Gawel et al. 2000). Some of the 7 categories are basically considered as "unpleasant" (drying, harsh, unripe, dynamic, particulate) and some others as "pleasant" (complex, surface smoothness). Some authors (Vidal et al. 2017) spoke about of a "polarization of astringency" related to terms: those related to soft textures opposite to those related to rough textures and aggressiveness. Our consideration is that less pleasant astringency sensations could positively impact the perceived quality when present in a well-balanced wine. This seems to be supported by the fact that they are often present in premium wines suitable for long ageing. On the other hand, those astringency sensations considered as pleasant, could lead to less appreciated wines if not combined with other descriptors. Vidal et al. (2017) expected that both low and extremely high global astringency intensity could be perceived as indicators of low quality Tannat wines, being the tipicity of this product linked to its astringency. We hypothesize that red wines can differ according to the balance between "strong" and "smooth" sensations defining their astringency. These two terms were already adopted to differentiate wines upon their astringency. Based on the characterization of the intensity and sub-qualities of astringency, different groups of Tannat wines were identified: those characterized by intermediate astringency (described as dry, rough and mouth-coating), those eliciting smooth astringency characteristics (described as velvety, silky and suede), and those characterized by their strong astringency (described as hard, harsh and aggressive) (Vidal t al. 2017). Overall sensory intensity and persistence of red wines are positively correlated with astringency (Peynaud 1987), and therefore to tannins content (Gonzalo-Diago et al. 2013). A relationship between tannins content and wines allocation grade, that is related to market value, has also been described (Mercurio et al. 2010). Several authors studied red wines' astringency through their sub-qualities (Green 1993, Gawel et al. 2001, Francis et al. 2002, Vidal et al. 2004, Ferrer-Gallego et al. 2014, Vidal et al. 2018), showing that astringency is not only complex, but also a time-dependent sensation. Recent studies investigated the alternation and development of astringency sub-qualities over time by approaching this subject through temporal measurements (Guinard et al. 1986, Cadena et al. 2014, Vidal et al. 2016, Kang et al. 2019). They highlighted the importance of addressing astringency through an holistic chemosensory approach including complementary information coming from static and/or temporal sensory assessments and chemical analyses. Some of these papers addressed the characterization of the astringency features of a specific wine through the investigation of astringency sub-qualities and the correlation between these sensory variables and chemical parameters (Vidal et al. 2016).

In a similar manner, but for the first time on a wide set of Italian red wines 100% from native grapes, the main purpose of this work was to study the astringency diversity of red wines from 11 varieties representative of the whole Italian territory: Teroldego, Corvina, Raboso Piave, Nebbiolo, Sangiovese, Sagrantino, Montepulciano, Cannonau, Aglianico, Primitivo and Nerello Mascalese. These varieties are actually used for the production of different wines labelled with Denomination of Origin Controlled (DOC) and Guaranteed (DOCG).

To reach our goal the astringency sub-qualities of an initial set of 111 commercial wines, were investigated by sensory analysis adopting a two-step analytical strategy composed of a sorting task and a sensory assessment through a numerical category scale. Multivariate statistical analyses such as Agglomerative Hierarchical Clustering following Multidimensional Scaling (AHC. MDS), Analysis of Variance (ANOVA), Principal Component Analysis (PCA) and Quadratic Discriminant Analysis (QDA) allowed a step by step definition of a reduced set of representative samples used to develop mono-varietal astringency profiles called "Astringency spectra".

Furthermore, the wide diversity in polyphenols and astringency features of Italian red wines, was exploited as an opportunity to investigate the relationship between specific compositional and in-mouth sensorial parameters. For this purpose, the correlations between specific sensory variables (single astringency sub-qualities, and tastes) and some chemical parameters concerning polyphenols measured with different methods, macromolecules and base chemical parameters, were tested. Only some of these results were presented in this paper.

MATERIALS and METHODS

2.1. Wine samples

111 Italian red wines, 100% mono-varietal, vinified in 2016 from 11 Italian grape varieties harvested in the corresponding main geographical areas of production (12 regions), were sampled from the commercial wineries where they were produced. For that reason, oenological parameters varied. The set of wines was composed of: 11 Teroldego Rotaliano (from Trentino-Alto Adige: TER), 7 Corvina (from Veneto: COR), 9 Raboso Piave (from Veneto: RAB), 13 Nebbiolo (from Piemonte: NEB), 19 Sangiovese (12 from Romagna: SAR; 7 from Toscana: SAT), 10 Sagrantino di Montefalco (from Umbria: SAG), 9 Montepulciano (from Abruzzo: MON), 9 Cannonau (from Sardegna: CAN), 10 Aglianico (from Campania: AGL), 11 Primitivo (from Puglia: PRI), and 3 Nerello Mascalese (from Sicilia: NER). Wines were fermented in stainless steel vats, in commercial scale, at wineries among the most representative in each area of production, and sampled before MLF and before wood ageing. All samples were protected with 50 mg/L of free SO_2 before bottling, and bottles were closed with a Select Green 500 cork type (Nomacorc, France) prior to storage at constant cellar temperature (12 ± 2°C) until the analyses.

2.2. Experiment 1: wines selection

This step was carried out to select the most representative wines belonging to each grape variety and to have first rough indications about the astringency features of the different wines.

2.2.1. Sorting task

2.2.1.1. Panel

The jury was composed of 14 people (7 M, 7 F; 22-49 years) recruited among students and staff members from the University of Naples Federico II, Department of Agricultural Sciences, Division of Vine and Wine Sciences. They were selected on the basis of their interest, availability and ability in recognizing oral stimuli. They all were expert wine tasters and had several previous experiences in performing sensory tests on wine. The study protocol has been approved by the Ethics Committee of University of

Naples Federico II. All participants were volunteers and before participating in the study they signed an informed consent form defining type of research, voluntary participation and agreement to sip and spit reference solutions and wines. All data were collected anonymously.

2.2.1.2. Panel training (phase 1: familiarization with in-mouth sensations)

In order to familiarize with the astringency vocabulary, judges were provided with a list of 7 terms defining the diverse astringency categories (designated hereinafter as "sub-qualities") of red wine as described at the first level of the "Mouthfeel wheel" (Gawel et al. 2000): drying, harsh, unripe, dynamic, particulate, complex and surface smoothness. Assessors were provided with a sheet with the Italian translation of the definitions reported by Gawel et al. (2000). After the theoretical introduction, 9 different taste/mouthfeel references were presented to the jury in order to develop a consensual list of terms describing the oral sensations elicited by each standard (Tables 1 and 2). The same references were employed to exercise the jury to recognize and discriminate the different oral sensations and also to help in the use of terms consistently to the corresponding definitions. The references (20 mL in covered disposable plastic cups) were presented in water and in table red wine. A five year old Pinot Noir was used as reference for the surface smoothness (Cliff et al. 2007). Tannic acid and four commercial tannins based products were used as sensory references for astringency and its sub-qualities (Table 1). Preliminary intra-lab tests were carried out to choose concentrations. The association of terms to these references was obtained by asking the assessors to take a sip (15 mL), to move the sample (15s) while wetting the whole mouth and then record the most intense sensations. Only descriptors cited at least by 85% of the jury, were matched to the terms as reported in Table 1 and considered as consensually associated to the corresponding sensory reference. A discussion on the perceived sensations was made at the end of each tasting session in order to agree on a common definition (Table 2). Relationships and redundancies among the terms were discussed. At the end of the training, it was consensually decided that the terms "Surface smoothness" and "Particulate" were to be intended as "Velvet" and "Powdery" astringent sensations, respectively. To help in memorization and consistent use of terms, as well as to prevent overlapping, a consensus was found on simplified descriptions for the terms. They were schematized as reported in Table 2 and a sheet with the simplified descriptions was attached to the wall of each individual booth during all the subsequent sessions. The first session was considered as introductive, so that only data collected from the 2nd and 3rd training sessions were employed to calculate the frequency of citations for matching standards with descriptor/s and to test panellists' performances.

2.2.1.3. Panel training (phase 2: familiarization with sorting)

Assessors were introduced to the sorting procedure. For this purpose, 8 red wines (30 mL in covered ISO wine glasses) from different varieties were presented. Judges were asked to introduce the sample into their mouth, focus on the perception of astringency and sort samples according to their similarities in astringency sub-qualities on which they were trained. Panellists were asked to label each group with the dominant sub-quality/s perceived among the seven on which they were trained. Judges were allowed to make as many groups of similar samples as possible and groups of single samples were permitted. Between two samples, assessors were asked to rinse the mouth by drinking bottled still water (Evian), to eat some apple slices, then drink a second time and finally wait at least 30 s before the subsequent evaluation. At the end, it was checked if the definitions of terms needed to be refined in this context of wines representative of the sample set under investigation. After discussion, no changes were made and the consensus was confirmed on all the definitions reported in Table 2. During the discussion judges were also asked about the roughness/aggressiveness of the different sensations: drying, harsh, dynamic, unripe and particulate were mostly perceived as strong/aggressive while complex and velvet as smooth/not aggressive.

2.2.1.4. Samples analysis

Wines were evaluated by sorting according to an intra-varietal experimental design meaning that all the wines from a given variety were sorted in the same session. In this way, an intra-varietal sorting was performed in order to investigate similarities and dissimilarities among wines belonging to the same variety (from 7 Corvina to 13 Nebbiolo). Due to the limited number of samples (only 3), Nerello Mascalese was not included in this first intra-varietal experimental step so that a total of 108 samples were analysed by sorting. Judges attended a total of 11 sessions corresponding to the number of mono-varietal wines (Sangiovese wines were divided into two sessions according to the geographical origin). The evaluation procedure was the same of the training (section 2.1.2.3.). Assessors were asked to group samples according to similarities in their astringency sub-qualities and label the groups. Thirteen samples, corresponding to the maximum number of wines sampled within a mono-varietal wine, were evaluated during each session. When less than 13 wines were available, "fake" samples were obtained by blending available wines of the same variety; data about these samples were not considered. 30 mL Samples were presented according to a randomized arrangement in covered ISO approved wine glasses labelled with three-digit random codes. All wines were served at room temperature (21 ± 1°C) and were evaluated in individual booths.

2.3. Experiment 2: wines sensory assessment

This step was aimed to obtain a sensory descriptive assessment of in-mouth features (tastes and astringency sub-qualities) of a reduced number of wine samples selected as the most representative within each mono-varietal wine.

2.3.1. Wine samples

A set of 77 wines was analysed: 74 (5 SAT and 5 SAR; 8 TER; 7 NEB, RAB, CAN, SAG, MON, COR, PRI and AGL) were selected according to the results of the sorting and 3 were the Nerello Mascalese (NER) wines.

2.3.2. Descriptive analysis

2.3.2.1. Panel training

The nine taste/mouthfeel references reported in Table 1 were presented to the jury in order to train them to score the intensity of different in-mouth sensations on the following numerical category scale: 1= very low, 2 = low, 3 = medium, 4 = high, and 5 = very high, with half values allowed. Materials and serving conditions were the same as above (section 2.2.1.2.).

In order to familiarize the jury with the evaluation procedure, 9 samples (3 RAB, 3 SAG and 3 TER) were tested prior to the analytical sessions (in duplication), as run-through. The procedure and the conditions were the same as described above (section 2.2.1.2.). Data were employed to test panellists' performances.

2.3.2.2. Sample analysis

The 77 wines were analysed in terms of astringency and taste by using the terms reported in the Table 2 and scoring the intensity of the perceived descriptors on the scale applied during the training (section 2.3.2.1)

The sensory assessment was performed according to an inter-varietal experimental design meaning that 11 wines corresponding to the 11 mono-varietal wines were evaluated during each of the 7 sessions. 25 mL of each sample were served as previously described (section 2.2.1.4). Panellists were asked to taste each sample by focusing on astringency by paying attention not only to the most intense sensation but also to that/those catching their attention the most during the tasting time, describing and scoring the diverse sensations by using the 7 terms corresponding to the different sub-qualities, and finally by scoring taste sensations (sweet, acid, bitter). Judges were informed that, based on data from training sessions, at least 3 of the astringency descriptors were expected higher than the minimum value on the scale, but no limitations were imposed. Judges were asked to rinse their mouth between two samples as reported above (section 2.2.1.3.).

2.4. Wine chemical analyses

Ethanol, reducing sugars, volatile acidity and titratable acidity were measured according to the methods OIV (2015). pH was determined by potentiometry (InoLab 730 pH meter, WTW, Germany). Total phenols by Folin-Ciocalteu assay were measured as previously described (Singleton et al. 1999). The proanthocyanidins content was determined after acid hydrolysis with warming (Bate–Smith reaction) using a ferrous salt ($FeSO_4$) as catalyst (Di Stefano et al. 1989, Torchio et al. 2010). Analyses were performed in triplicate.

2.5. Data Analysis

In order to visualize groupings of wine samples due to astringency similarities analysed by sorting, Multidimensional Scaling (MDS) analysis followed by Agglomerative Hierarchical Clustering (AHC) analysis were performed and the co-occurrence similarity matrices were considered. As previously reported (Sáenz-Navajas et al. 2012, and references therein), for each assessor, results were organized under an individual similarity matrix (wines x wines): 1 corresponded to a couple of wines put into the same group while 0 was for two wines put in different groups. The sum of the individual matrices across judges, was merged into a co-occurrence matrix representing the global similarity matrix where the higher the number the higher the similarity between samples. This method assumes that samples frequently grouped together were perceived as more similar compared to those sorted into different groups. The proximity matrix (Euclidean distances between the products) was the base for the MDS analysis (SMACOF algorithm). The quality of fit was measured by the stress value (from 0 = perfect fit to 1 = worst fit). As previously reported and applied, a value bellow 0.2 can be considered as a good agreement between the initial and final configurations, so that this stress value was adopted as criterion to select the number of dimensions for the MDS spaces. Coordinates of samples in the retained MDS configurations were submitted to a HCA with the Ward criterion. We applied the automatic truncation option, which is based on the entropy and tries to create homogeneous groups. HCA was helpful for the interpretation of MDS maps allowing the identification of wines belonging to each cluster. We arbitrary decided to select at least 7 samples of each mono-varietal wine. In this way at least 50% of each mono-varietal sample set was selected, indeed the most numerous set of wines was composed of 13 NEB.Data from the descriptive sensory assessment were analysed by one-way ANOVA (wine was the factor and judges were considered as random factor), and the mean intensities for each astringency sub-quality were compared (intra- and inter-varietal) by a Tukey post-hoc test ($p< 0.05$).

A Principal Component Analysis (PCA) was applied to the original in-mouth variables (astringency sub-qualities and tastes) constituted by the sensory scores. Sensory data referring to astringency-sub qualities were also computed as the geometric mean of frequency and mean intensity (Mean Sensory Modified Frequency: MF) as described by Dravnieks (1982): $MF=(F * I)^{1/2}$, where F is the frequency of citation expressed as a percentage of the maximum frequency of citation (i.e. total number of judges) and I is the mean intensity expressed as a percentage of the maximum rate.

Quadratic Discriminant Analysis (QDA) was used to classify the wines assuming the variety as qualitative dependent variable and MF of the astringency sub-qualities as quantitative explanatory variables (inequality of covariance matrices tested by Box test; Jarque-Bera normality test; $\alpha = 0.05$). The classes weight correction was applied because the number of observations for the various classes for the dependent variables was not uniform. The classification functions were used to determine which class (variety) an observation (wine) is to be assigned to using values taken for the various explanatory variables. An observation was than assigned to the class with the highest classification function. Only wines that, after cross-validation, resulted well-classified to the corresponding grape variety, were further considered to develop mono-varietal astringency patterns. In order to satisfy the assumption that the number of explanatory variables (six) was lower than each sample size, NER samples (only 3) were not included in the discriminant analysis.

Pearson correlation analysis ($p<0.05$) was applied across the whole set of wines (sample size = 77) for the computation of correlations between the intensity of astringency sub-qualities and in-mouth sensory variables or chemical parameters.

Performance of the trained judges was tested by three-way ANOVA (Tukey, p<0.05) with interactions of assessor*session, assessor*sample, sample*session (Vidal et al. 2016).

Data elaboration was performed by XLStat (version 2018.7), an add-in software package for Microsoft Excel (Addinsoft Corp., Paris, France).

3. RESULTS

3.1. Wines selection

Basic compositional data of the wine samples were shown in Table 3. The ranges of these parameters were large, thus astringency differences were expected in the set of sampled wines. Data from the sorting performed according to astringency similarities, were analysed by AHC after MDS. According to the dendrograms (Figure sm1), within each mono-varietal wine, samples resulted clustered into three groups represented on three (Sangiovese, Sagrantino, Raboso, Primitivo, Nebbiolo, Corvina) or four (Aglianico, Montepulciano, Cannonau, Teroldego) dimensions on the MDS spaces (not shown).

From these results, we selected samples from each wine type according to the following criteria: the most similar couple of wines, couples including the central object of each cluster, at least three wines from the most homogeneous cluster (lowest within-class variable) when larger than two objects, at least one sample (central object) belonging to each cluster (excluding clusters composed of one sample). When necessary, distances from the MDS output were adopted as additional criteria to select at least 50% of samples from each variety. In this manner we reduced the number of samples belonging to each mono-varietal wine by preserving the representativeness in terms of intra-varietal similarities and diversities. The final set of 77 selected wines was then composed of: 10 Sangiovese (5 from Romagna and 5 from Toscana), 8 Teroldego, 7 Nebbiolo, Aglianico, Primitivo, Montepulciano, Cannonau, Raboso Piave, Corvina and Sagrantino, plus 3 Nerello Mascalese.

3.2. Wines description and discrimination

In the box-plots (Figure 1), the 11 mono-varietal wines were compared with respect to each astringency sub-quality. Several differences emerged for 6 out of 7 sub-qualities. According to the significance ($p<0.05$) reported on the top of each box, only some of these differences were significant.

Three main levels of drying intensity were identified: Nebbiolo and Sagrantino showed the highest mean intensities, followed by Raboso, Primitivo and Nerello Mascalese, and then by Corvina. Two further intermediate levels corresponded to the drying intensity of the other wines. For the harsh, Sagrantino and Corvina wines represented the two opposite, showing the highest and the lowest values, respectively. Some significant differences were detected among the other wines, except for Sangiovese and Nerello.

. For unripe, the highest mean intensity was associated to Raboso, in contrast to Sangiovese, Nebbiolo and Nerello which were the less unripe and significantly different from Corvina, Montepulciano was not different according to its unripe character. Astringency of Sagrantino was perceived as the most dynamic while Teroldego, Primitivo, Montepulciano and Corvina , the less. For dynamic no differences emerged for all the other wines. Cannonau and Primitivo were different from Nebbiolo that was the less complex. Corvina, was opposite to Nebbiolo with the highset and the lowest values for surface smoothness, respectively. Raboso and Primitivo were more velvet than Nebbiolo, while Sangiovese less than Corvina. Finally, the 11 mono-varietal wines did not resulted significantly different according to the sub-quality particulate, and therefore, this sub-quality was not considered for the subsequent analyses.

Figure 2 shows the PCA where all in-mouth sensory variables (a) and observations (b) were plotted on the first two components representing 58.81% of the variance. The astringency sub-qualities and the bitter taste are mostly represented on PC1, while the contrast between acid and sweet tastes is represented on PC2. The variables positively correlated ($p<0.0001$) to each other are: dynamic with drying ($R^2 = 0.565$), harsh with bitter ($R^2 = 0.771$), acid with unripe ($R^2 = 0.593$), surface smoothness with complex and sweet ($R^2 = 0.283$ and $R^2 = 0.256$, respectively). Drying and dynamic were negatively correlated ($p<0.0001$) to surface smoothness ($R^2 = -0.642$ and $R^2 = -0.463$, respectively). Compared to unripe, harsh showed an opposite correlation to acid taste ($R^2 = -0.577$). Most of Sangiovese, Nebbiolo and Sagrantino wines show the largest squared cosines to positive values of the first factor, where the variables drying and dynamic, harsh and bitter are well projected. On the other side of the first factor, in the space where the best represented variables are acid, surface smoothness and unripe, different wines showed the largest squared cosines, mainly Corvina and Raboso. Along the second factor, some Raboso, Aglianico and Montepulciano wines were linked to the acid taste, opposite to Cannonau, Primitivo and Teroldego linked to the sweet. A wide intra-varietal diversity results for Aglianico wines, which occupy the most diversified positions in the PCA space.

Figure 3 shows the output of the QDA. The goal was to test if the mono-varietal wines could be discriminated and clustered only according to their astringency sub-qualities (MF values). As previously applied on olfactory and in-mouth descriptors (Lelièvre et al. 2008), the MF method was applied because it takes into account both types of values produced by assessors: the frequency of citation of a sensory term and the intensity assigned to it. In this way we properly considered cases in which a term has been used frequently but with low scores, and cases in which the same descriptor has been poorly cited but with high scores. The loading plot (Figure 3a) represents the contribution of each astringency sub-quality to the discrimination. On the first two factors 82.09% of the variance is represented: F1 carried the majority of the differentiation of the samples (65.57%) with the sub-qualities dynamic, drying and harsh opposite to unripe and surface smoothness. The first three resulted correlated on the positive semi-axis (R=0.616, R=0.888, R=0.767, respectively), while the two latter on the negative one (R=0.830, R=0.731, respectively). F2 was negatively correlated to complex. The representation of centroids and corresponding confidence ellipses on the factor axes (Figure 3b) showed that some mono-varietal wines were better discriminable than others according to their astringency sub-qualities. Raboso and Corvina were mainly distinguishable for their unripe astringency, with a velvet character in the latter. Nebbiolo, Sagrantino and Sangiovese were mostly discriminated for their strong astringency components (drying, dynamic, harsh) while the remaining wines were mostly in the middle of the map showing overlapping confidence ellipses.

For each observation (wine sample), the probability to belong to each group (mono-varietal wine) was computed, and each wine was reclassified into the group for which the probability of belonging was the greatest. According to the confusion matrix, 88% of the wines were correctly reclassified: Corvina, Raboso, Nebbiolo, Sagrantino and Sangiovese samples were 100% correctly matched to the corresponding variety, followed by Cannonau and Primitivo (85.71%), Teroldego (75.00%), Aglianico (71.43%) and Montepulciano (57.14%).

Only the wines correctly reclassified were taken into account to develop, for each of the corresponding 10 mono-varietal wines, a graphical representation of their astringency features. For each mono-varietal wine, the astringency sub-quality with the highest MF (mean value over the wines retained in the analysis) was considered as 100 and the MFs of the 5 remaining sub-qualities were normalized with respect to it. In this manner, as for a typical mass spectrum, we obtained a histogram corresponding to the

"Astringency spectrum" of a given mono-varietal wine where, the 6 sub-qualities were conceived as "Fragments" of the whole astringency of that wine (Figure 4). Being the abundance of each astringency sub-quality plotted by computing its occurrence relative to the most important sub-quality detected in that mono-varietal wine, we obtained normalized profiles that allowed us to compare the average relative contribution of each sub-quality to the astringency, within each of the diverse mono-varietal wines. The patterns resulted different from each other, 8 wines were dominated by the drying astringency (Figures 4a,b,c,d,e,f,h,i), 2 by the complex (Figures 4l and 4m) and 1 by the unripe (Figure 4g).

3.3.Correlations

Pearson correlations ($p<0.05$) were computed to test, across the different mono-varietal wines, the association between variables describing in-mouth sensations (astringency sub-quality: A; taste sensation: T), and a set of chemical variables concerning polyphenols (PPh), and wine base chemical parameters (BCP) (mean of triplicate repetitions). Figure 5 represents the map of the correlations (correlation coefficients were detailed as supplementary material in Table sm1). At least one significant correlation was found for each variable and in most cases with a p-value <0.0001.

The PPh variables, total phenols and total proanthocyanidins, were: 1) highly ($p<0.0001$) positively correlated to drying (R^2 = 0.558 and 0.708, respectively), harsh (R^2 = 0.479 and 0.475) and dynamic (R^2 = 0.468 and 0.583); 2) weakly negatively correlated to unripe (R^2 = 0.304 and 0.365) and surface smoothness (R^2 = -0.408 and -0.433); 3) not correlated to complex. Among sweet, acid and bitter tastes, only the two latter showed some weak correlations with PPh parameters.

Also some correlations between BCP and in-mouth variables emerged but only those between pH and acidity (R^2 = -0.562) or bitterness (R^2 = 0.497) resulted the strongest ($p<0.0001$). The volatile acidity resulted positively correlated with harsh (R^2 = 0.444), bitter (R^2 = 0.405) and drying (R^2 = 0.311), and negatively to acid (R^2 = -0.290) and complex (R^2 = -0.265).

3. DISCUSSION

3.1. Wines description and discrimination

From this study we obtained sensory profiles describing the balance among astringent sensations elicited by an extensive sample set of mono-varietal Italian red wines representing different styles of astringency.

Several studies focusing on molecules known to be responsible for astringency, have been conducted on Italian red wines/grapes (Mattivi et al. 2002, Mattivi et al. 2009) but, for the first time, the astringency diversity of Italian red wines, has been systematically investigated and compared from a sensory perspective. Like in previous studies on red wine astringency (Vidal et al. 2016, Ferrer-Gallego et al. 2016), this study was carried out in full perceptual conditions (all senses). This allowed to assess wine astringency in conditions similar to that occurring during wine consumption, when cross-modal sensory interactions can occur. By merging the results reported through this study it seems possible to state that even if an intra-varietal diversity was detected, it was possible to identify a pattern of astringency features common to wines from a given grape variety. Indeed, referring to the box-plots (Figure 1), we could gather that the shorter the box, the lower the variability of that sub-quality in that wine type. This suggests a wine feature that has been perceived in a similar manner in all samples by all judges, and therefore likely linkable to the grape variety (e.g. strong unripe in Raboso and Corvina; very low dynamic in Teroldego, Corvina and Primitivo; absence of velvety character in Nebbiolo and Sagrantino). This result points out that these astringency features could be linked to the grape variety.

The detection of single wines or groups with different levels of intensity for the various astringency sub-qualities testifies the inter-varietal astringency diversity. The 11 mono-varietal wines were differentiated at least for 3 different levels of intensity for drying, 2 for harsh, unripe, dynamic, complex and velvet, while none for particulate. This indicates that judges showed a good understanding of what the different sub-qualities are, and that the 11 wines were distinguishable mostly according to the drying astringency sensation . The lack of significant differences among wines regarding the term particulate (here intended as powdery), is in agreement with latest results obtained by applying the modified progressive profiling, a dynamic sensory method (Kang et al. 2019). The study reports that, differently from the other sub-qualities, the graininess, which was defined as a sensation of particulate matter on the mouth surface, resulted a variable not useful to discriminate the astringency of 13 red wines.

The PCA performed on sensory intensities, highlighted correlations between the 6 astringency sub-qualities and tastes (Figure 2). Some of these correlations (eg. harsh and bitter, unripe and acid) suggest that judges correctly used the sub-qualities descriptors according to their definitions (Table 2). Taste variables occupied three distinct parts on the map. Also the 6 astringency sub-qualities were well projected on three distinct areas of the chart, each of them close to a taste variable. The unripe astringency resulted not correlated to none of the other sub-qualities, suggesting a different "nature" of this sub-quality compared to the others. The PCA found that in-mouth sensations of Sagrantino, Nebbiolo and Sangiovese were perceived as similar, and mainly associated to strong astringency sub-qualities and bitter taste. The other wines resulted spread on the opposite side of the chart sharing some common characteristics. The outputs of the QDA (Figure 3), showed that only some of the 11 mono-varietal wines were discriminable from others due to their astringency features. Corvina and Raboso were discriminable to the other mono-varietal wines and similar to each other, mostly for their unripe character. The discriminability of Nebbiolo, Sagrantino and Sangiovese was highlighted. All the other wines were not well discriminable according to their astringency features. This could be due to a higher degree of intra-varietal variability or to a more balanced contribution of the diverse astringency sensations. Each mono-varietal wine showed a unique pattern among the six astringency sub-qualities. The "Astringency spectra" (Figure 4) of the mono-varietal wines that were 100% correctly reclassified (Corvina, Raboso, Nebbiolo, Sagrantino and Sangiovese), can be considered as more reliable than the others. The future assessment of a larger and new distinct representative set of the same mono-varietal wines could be useful to validate the astringency profiles that were developed in this study. According to the dominant sub-quality, three groups of wines can be distinguished: those dominated by the drying character, a couple dominated by the complex sub-quality, and the one dominated by an unripe astringency, namely Corvina. The "Astringency spectra" of Sagrantino (Figure 4d) and Sangiovese from Romagna (Figure 4b) were similar as relative contribution of drying, harsh and complex while different mainly for that of surface smoothness and dynamic: the first was rather important in Sangiovese from Romagna and the second almost absent in Sagrantino. This lack of surface smoothness was also detected in Nebbiolo wines (Figure 4c). In the scientific literature we did not find sensory data on Sagrantino wines, however our results seem in line with previous chemical results. A study that measured the amount, the localization and the extractability of flavan-3-ols and anthocyanins in 25 high-quality red grapes, classified Sagrantino grapes as the richest in extractable polyphenols and proanthocyanidins (Mattivi et al. 2002). Moving to Nebbiolo, it produces wines with high acidity and tannic when young, so that they require long ageing to reach a balance between acidity, astringency, full body and aroma complexity (Asproudi et al. 2015). Barbaresco wines (100% made with Nebbiolo grapes) are often characterized by light colour and high roughness (Gerbi et al. 2006). From a chemical point of view, Nebbiolo grapes are known to be poor in anthocyanins and rich in proanthocyanidins (Mattivi et al. 2002, Locatelli et al. 2016). Astringency is reported as an important sensory descriptor of SAR wines (Pagliarini et al. 2013, Laureati et al. 2014, Patrignani et al. 2017), which showed the lowest level of copigmentation compared to the other wines (Versari et al. 2007). This could correspond to a higher astringency as a consequence of a poor inclusion of some astringent monomeric components into copigmentation stacks (Boulton 2001, Alvarez et al. 2009, Escribano-Bailón and Santos-Buelga 2012). Moreover, in the last years, unbalanced Sangiovese wines with excessive alcohol and astringency, have been related to climate change (Filippetti et al. 2015). The rising temperature during ripening can negatively affect the acidity content and the synthesis of polyphenols provoking the rise of sugar accumulation leading to excessive alcohol. Due to the importance of Sangiovese grapes and wines (the principal Italian red variety), this issue is of impact also taking into account the enhancing role of increased ethanol on astringency (Noble 1999) and, the high maximal values we observed both for the total proanthocyanidins as well as for ethanol (Table 3). For the first time, our results compared Sangiovese wines from the two main areas of production showing different astringency features. Compared to SAR (Figure 4b), the "Astringency spectrum" of SAT (Figure 4a) was different for a higher relative contribution of the complex sub-quality and an importantly lower impact of the harsh and dynamic components (mean intensities were significantly different; Tukey: $p<0.05$). Unripe characterized the profile of Raboso wines (Figure 4h). Raboso Piave grapes are known to have high acidity and unbalanced polyphenols with predominant low molecular flavanols (catechin), leading to astringent wines not easy to drink if the grape maturity, the winemaking and the ageing are not well managed (Mattivi et al. 2006, Corso et al. 2013). For Aglianico (Figure 4i), the pattern showed a balanced contribution of the different sub-qualities other than drying. High release and astringency of seed tannins compared to other grapes were detected in Aglianico. Studies on winemaking and ageing optimization to smooth the astringency and balance the sourness, two sensations characterizing young Aglianico wines, were carried out (Mattivi et al. 2002, Gambuti et al. 2009). In Montepulciano (Figure 4f) the important contributors harsh and unripe were counterbalanced by surface smoothness and complex. Only 57% of our Montepulciano samples were correctly reclassified to the corresponding mono-varietal wine and for this reason the resulting "Astringency spectrum" was the least reliable compared to the others. Cannonau (genetically the same variety as Grenache) was one of the two wines showing the dominance of the complex (Figure 4m); follow an important relative contribution of strong sub-qualities (drying, harsh, unripe) but also a good occurrence of surface smoothness. In a comparison with a large number of Italian varieties (Mattivi et al. 2002), Cannonau exhibited a medium or low-medium level of polyphenols having less than 40% of the catechins and proanthocyanidins reactive to vanillin located in the seeds, and the content of extractable proanthocyanidins in the seeds not exceeding 35%. In Primitivo wines the most important astringency sub-qualities resulted drying and complex, with a good relative contribution of surface smoothness (Figure 4e). Primitivo wines, rich in colour intensity but scarce in tannins content, commonly reach high alcohol levels and have a ruby-purple colour, with a sensory profile showing a good balance between astringency, body and pleasantness (Suriano et al. 2016, Trani et al. 2016). The "Astringency spectrum" of Corvina wines (Figure 3g) resulted the only one dominated by an unripe astringency and, at the same time, by the highest relative contribution of surface smoothness compared to the other wines. This astringency profile fits in with previous knowledge about Corvina grapes, indeed it is reported as characterized by a low tannin content and a green flavour (herbaceous/balsamic) that has been correlated to high concentration of hexanols (Paronetto and Dellaglio 2011) and cyclic terpenes (Slaghenaufi and Ugliano 2018). Moreover, even if blended with other grapes, it gives the wine a powerful structure but surprising smoothness (Paronetto and Dellaglio 2011). Finally, Teroldego is generally characterized by a very intense ruby colour and smooth in the mouth. Compared to other grapes, Teroldego resulted the richest in extractable antocyanins, showing an average content of extractable proanthocyanidins, with a low percentage from the seeds (Mattivi et al. 2002). Like Cannonau, its "Astringency spectrum" (Figure 4l) was dominated by the complex. This, together with a good surface smoothness, contrasts with the important contribution of drying and unripe with a net result, in terms of astringency, that suggest a soft mouthfeel.

4.2 Correlations

The significant correlations highlighted between sensory and chemical variables (Figure 5, Table sm1) were tested across the 11 different mono-varietal wines. Total phenols and proanthocyanidins were positively correlated to drying, harsh and dynamic while only negative correlations coefficients emerged between surface smoothness, unripe and complex, and a weak significance was detected for the first two only. This result suggests that none of the two PPh variables tested, are able to predict/measure the perception of astringency in all its possible nuances. The fact that at least some aspects of astringency could be connected to aroma compounds could partially impact on this result. Indeed, being unripe and complex two astringency sub-qualities including a retronasal olfactory sensation (Gawel et al. 2000), the volatile composition of the wine could play a significant role on their perception. The absence of correlations between unripe and PPh parameters supports the idea of a multi-dimensional nature of this sensory variable and appears consistent with previous findings. Indeed, in a chemo-sensory study aimed to characterize the fractions driving different mouthfeel properties in red wines, only the category unripe was not included in the final list of terms generated to describe the in-mouth sensations elicited during the tasting of the different odourless fractions (Sáenz-Navajas et al. 2017). The same authors tried to understand the involvement of VOCs modulating the perception of the green character of red wine astringency (Sáenz-Navajas et al. 2018). No specific aroma compounds were identified but high levels of fusel alcohols were observed and the involvement of interactions between isoamyl alcohol and anthocyanin-derivative fractions and/or tannins was suggested. Among the sensorial and chemical parameters considered in this study, total proanthocyanidins showed the highest correlation coefficient. This is in accord with several studies that linked tannin concentration not only to the overall astringency but also to some sub-qualities describing “aggressive” sensations (dry, pucker, chalk) and, in accord with us, to the decrease of smooth sensations (surface smoothness, silky, velvet) (Vidal et al. 2004, Preys et al. 2006, Vidal et al. 2018). A positive correlation was also found between the intensity of dry measured by modified progressive profiling and total tannin concentration (Kang et al. 2019). Among BCP parameters, ethanol showed a negative correlation with acid and positive with bitter and this is coherent with bibliography, indeed ethanol tends to increase bitterness perception (Fischer and Noble 1994, Vidal et al. 2004, Sokolowsky and Fischer 2012) and suppress sourness (Williams 1972, Gonzalo-Diago et al. 2014). Ethanol was positively correlated with drying and harsh while negatively with unripe and surface smoothness. It has been reported that ethanol decreases protein-tannin interactions and this has been linked to a decrease of the overall intensity of astringency (Waterhouse et al. 2016, and references therein), while our result refers to drying that is a specific sub-quality. This result seems in line with a very recent study (Saenz-Navajas et al. 2020), where the authors found a positive correlation (even if not significant) between ethanol and dry. According to its definition (Gawel et al. 2000), the drying sub-quality corresponds to a lack of lubrication with dehydration, and ethanol is a dehydrating agent. It is reported that ethanol is astringent at high concentrations, due to denaturation and precipitation of salivary proteins (Waterhouse et al. 2016, and references therein). In our work, we tested the correlations across the whole set of wines that, according to data reported in Table 3, includes samples with high alcohol content. A negative correlation between pH an acid taste was observed, and the pH was also weakly positively correlated to harsh and bitter, in line with the definition of harsh. Some studies reported about the influence of pH and ethanol on the different astringency sub-qualities (Gawel et al. 2014, Kang et al. 2019). The trends that we observed for unripe seem in line with previous findings. It has been reported (De Miglio et al. 2002) that the unripe was rated more intensely as ethanol concentration decreased and as pH values lowered. It was suggested that the driving force of these effects could be the impact of ethanol and pH on the perceived acidity and this seems coherent with the definition of unripe.

The titratable acidity confirmed exactly the same correlations detected for pH but with opposite trends. The weak correlations between volatile acidity and in-mouth variables could be linked to the maceration conditions during winemaking. Indeed conditions enhancing polyphenols extraction if combined with the ethanol developed and the limited nutrient status, can stress yeast and even bacteria and may lead to a rise in volatile acidity. A recent paper identified volatile acidity among the top five predictive variables for drying and mouth-coating astringency sub-qualities in Tannat wines (Vidal et al. 2018).

According to our results, harsh and unripe were the sub-qualities that can be affected the most by BCP, while drying and even more dynamic (no correlations with BCP) seem to be driven by the polyphenols composition. Also complex and surface smoothness, the two sub-qualities describing smooth astringency, resulted poorly correlated to BCP. The lack of correlations between complex and PPh supports the hypothesis that other factors, likely olfactory cues, could play an important role on its perception but specific investigations are necessary.

CONCLUSIONS

Overall, this work gives a first picture of the diverse astringency of red wines from Italian native grapes, including some mono-varietal products that have never been investigated before on their astringency. Furthermore, a contribution to the knowledge about the influence of chemical composition on the perception of astringency sub-qualities, is given.

The 11 mono-varietal wines were differentiated at least for 3 different levels of intensity for drying, 2 for harsh, unripe, dynamic, complex and velvet, while none for particulate. Despite the detected intra-varietal variability, which was expected due to viticultural and oenological differences in commercial wine production, recurrent astringency features were found within wines from a given variety: intense unripe in Corvina and Raboso; very low dynamic in Teroldego, Primitivo, Corvina and Montepulciano; no velvety in Sagrantino and Nebbiolo. All samples were produced in the same vintage and had no contact with wood, therefore it seems reasonable to think that these recurrent features can be essentially referred to the astringency of the grape varieties.

The "Astringency spectra", sensory patterns describing the relative balance among six astringency sub-qualities of the mono-varietal wines, were different from each other. Further experiments are necessary to validate these profiles on other wines produced from the same varieties, and in limited perceptual conditions in order to evaluate the impact of cross-modal sensory interactions.

The correlation study conducted over a set of very different wines, confirmed the positive correlation between total proanthocyanidins and astringency, highlighted that neither total phenols nor total proanthocyanidins were able to measure/predict the perception of astringency in all its nuances, and suggested that the diverse astringency sub-qualities could be affected in different manners by the chemical parameters, such as ethanol or pH.

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Table 1. References and corresponding consensual descriptors, used to train the assessors in recognizing and distinguishing among the different in-mouth sensations (tastes and astringency sub-qualities)

References	Concentration (g/L)*	**Descriptors***	Producers
Fructose	2	Sweet	J.T. Baker (Avantor; Radnor, PA, U.S.A.)
Tartaric Acid	4	Sour	Chem-Lab (Eernegem, West-Vlaanderen, Belgium)
Caffeine	2	Bitter	ACEF (Piacenza, Italy)
Tannic Acid	2	Astringt	J.T. Baker (Avantor; Radnor, PA, U.S.A.)
Tannin VR Color (Catechin and ellagic tannins formulation)	4	Drying and Harsh	Laffort (Bordeaux, France)
Tannin VR Grape (Proanthocyanidic tannins extracted from grape skin and seeds)	2	Particulate (as Powdery) and Unripe	Laffort (Bordeaux, France)
Tannin plus (Tannins formulation)	4	Complex and Drying	Laffort (Bordeaux, France)
Tannin Galalcool (Gallic tannins from gallnuts in granulated form)	2	Unripe	Laffort (Bordeaux, France)
Red wine (Pinot noir 5 years old)	-	Surface Smoothness (as Velvet)	St. Michael Eppan (Trentino Alto Adige, Italy)

* both in distilled water and in table red wine (pH=3.2; ethanol =12.5 % v/v; titratable acidity=7.7 g tartaric acid/L; residual sugars=1.5 g/L; total anthocyanins=36 mg/L ; BSA reactive tannins=112 mg/L)

** agreed definitions are reported in Table 2

***consensual association frequency ≥ 85%

Table 2. Definitions of the terms considered to assess astringency

Terms	Agreed definitions	Simplified definitions
Astringency*	Oral tactile sensation mainly characterized by dryness and roughness	
Drying **	Lack of lubrication and dehydration feeling in the mouth	No lubrication+dehydration
Harsh**	Unbalanced in-mouth sensation of dryness, roughness (irregularities and lack of smoothness) and bitterness	Astringency+roughness+bitterness (combined and aggressive/excessive)
Dynamic**	Sensations impacting on fluidity of oral movement	Lack of fluidity
Particulate (as Powdery)**	Oral sensation associated with the touch of powdery matter	Powdery at touch
Unripe **	Unbalanced in-mouth sensation of astringency, sowarness and green aroma	Astringency+Acid+Herbaceous (combined and aggressive/excessive)
Surface Smoothness (as Velvet)**	Oral texture sensation associated with the touch of velvet	Velvet at touch
Complex **	Balanced in-mouth sensation of smooth astringency, acidity and retronasal stimulation	Astringent+Acid+Flavored (combined and not aggressive/eccessive)

* as defined by Vidal et al. (2016)

**agreed definitions elaborated by starting from those reported by Gawel et al. (2000)

Table 3. Oenological parameters determined in the 111 mono-varietal Italian red wines

Parameter	Mean	Minimum	Maximum
Ethanol [% v/v]	13.9	11.4	16.6
Reducing sugars [g/L]	2.6	1.0	20.1
Titratable acidity [g tartaric acid/L]	5.7	4.0	10.0
pH	3.6	3.1	4.1
Total phenols (Folin-Ciocalteu) [mg (+)-catechin/L]	2341	704	5449
Total proanthocyanidins [mg cyanidin chloride/L]	3373	628	6312

Table sm1. Correlation coefficients (Pearson) between in-mouth and chemical variables represented in Figure 5

Variables			In-Mouth								
			Astringency						Taste		
			Drying	Harsh	Unripe	Dynamic	Complex	Surface smoothness	Sweet	Acid	Bitter
Chemical	PPh	**Total phenols (Folin-Ciocalteu) [mg/L]**	**0,558**	**0,479**	**-0,304**	**0,468**	-0,159	**-0,408**	-0,079	**-0,347**	**0,425**
		Total proanthocyanidins ([mg cyanidin chloride/L]	**0,708**	**0,475**	**-0,365**	**0,583**	-0,225	**-0,433**	-0,052	**-0,296**	**0,409**
	BCP	**Ethanol [% v/v]**	**0,363**	**0,396**	**-0,416**	0,179	0,202	**-0,275**	0,171	**-0,421**	**0,278**
		Reducing sugars [g/L]	0,036	-0,010	-0,052	0,013	0,229	0,040	**0,387**	-0,089	-0,093
		pH	0,074	**0,434**	**-0,368**	-0,019	0,056	-0,082	0,031	**-0,562**	**0,497**
		Titratable acidity [g tartaric acid/L]	0,011	**-0,284**	**0,276**	0,020	0,150	0,049	-0,033	**0,451**	**-0,363**
		Volatile acidity [g acetic acid/L]	**0,311**	**0,444**	-0,195	0,172	**-0,265**	-0,134	-0,103	**-0,290**	**0,405**

Values in bold are different from 0 with a significance level $p < 0.05$ (in gray $p<0.0001$)
PPh: PolyPhenols; BCP: Base Chemical Parameters

FIGURE LEGENDS

Figure 1. Box-plots describing inter-varietal diversity of each astringency sub-quality in the 11 mono-varietal Italian red wines investigated (red crosses: means; central horizontal bars: medians; lower/upper limit of the box: first/third quartile; points above/below the whiskers' upper/lower bounds: outliers; box plot's horizontal width: no statistical meaning). Letters reported on the top of each box-plot refer to significant differences tested by ANOVA (Tukey, $p<0.05$; Drying: $F=11.254$, $P<0.0001$; Harsh: $F= 4.655$, $P<0.0001$;Unripe: $F= 5.594$, $P<0.0001$; Complex: $F= 3.346$; $P<0.0001$; Dynamic: $F= 5.943$, $P<0.0001$; Particulate: $F= 0.562$, $P= 0.846$; Surface smoothness: $F= 4.209$, $P<0.0001$).

Figure 2. Principal Component Analysis (PCA) plots (a: variables; b: observations) calculated on intensity scores (TER: Teroldego; COR: Corvina; RAB: Raboso Piave; NEB: Nebbiolo SAN: Sangiovese; SAG: Sagrantino; MON: Montepulciano; CAN: Cannonau; AGL: Aglianico; PRI: Primitivo; points size with Cos^2).

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Figure 3. Quadratic Discriminant Analysis (QDA) computed using MF of astringency sub-qualities (drying, harsh, unripe, dynamic, complex and surface smoothness) as quantitative explanatory variables. (a) Vectors show astringency sub-qualities contributing to the overall variance between mono-varietal wines. (b) Ellipses show 95% confidence intervals for each mono-varietal wine around the corresponding centroids (TER: Teroldego; COR: Corvina; RAB: Raboso Piave; NEB: Nebbiolo SAN: Sangiovese; SAG: Sagrantino; MON: Montepulciano; CAN: Cannonau; AGL: Aglianico; PRI: Primitivo).

Figure 4. "Astringency spectra" developed for the mono-varietal wines.

Figure 5. Map of the correlations (Pearson) between in-mouth and chemical variables (A: astringency sub-qialities; T: tastes; PPh: polyphenols; BCP: basic chemical parameters). Corresponding p-values are reported in Table sm1.

Figure sm1. Dendrograms obtained by Agglomerative Hierarchical Clustering (AHC) performed on data from the sorting test, and used for wine selection (in red: selected samples; in bold: central objects of each cluster).

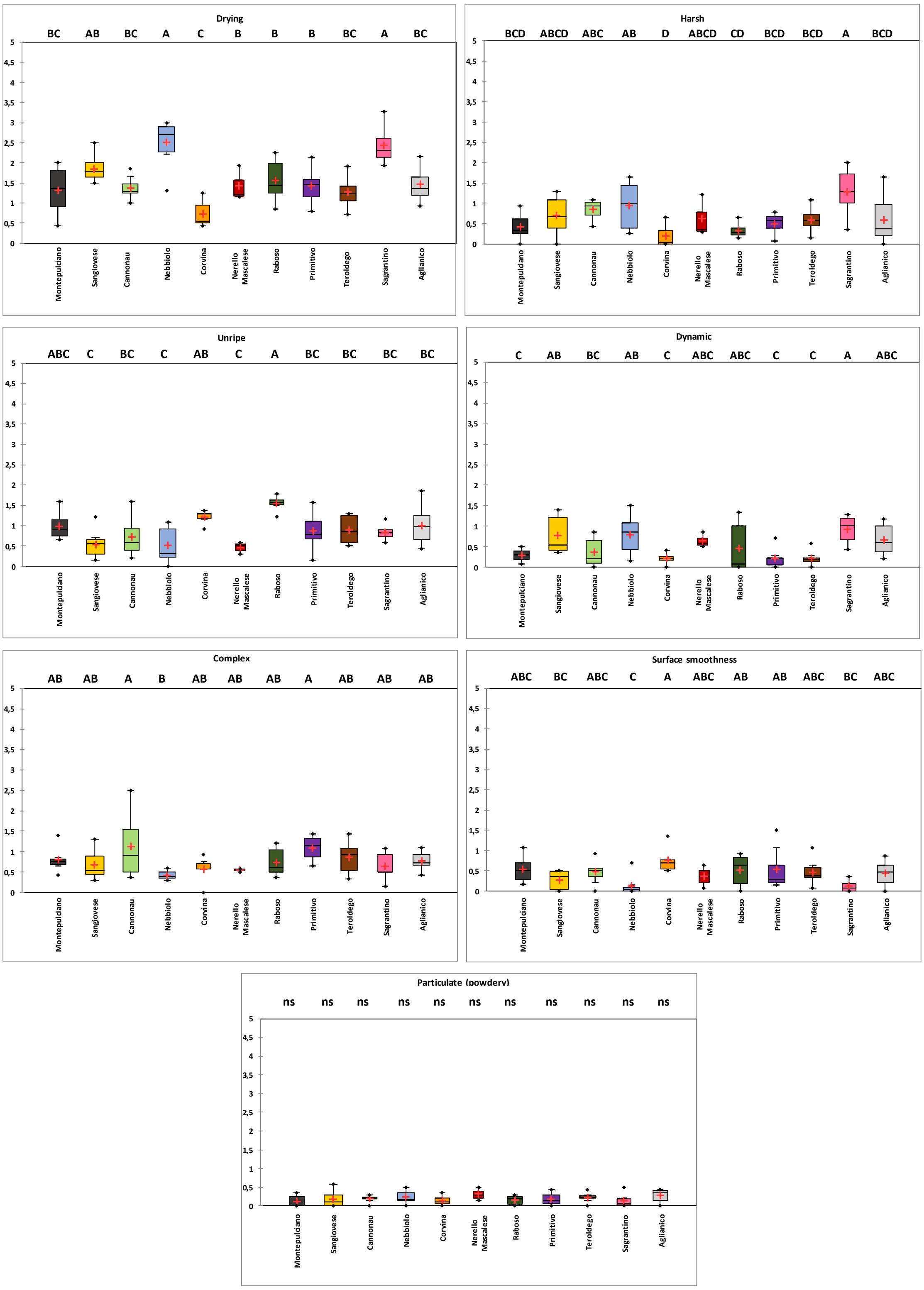
Drying
BC AB BC A C B B B BC A BC
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Harsh
BCD ABCD ABC AB D ABCD CD BCD BCD A BCD
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Unripe
ABC C BC C AB C A BC BC BC BC
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Dynamic
C AB BC AB C ABC ABC C C A ABC
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Complex
AB AB A B AB AB AB A AB AB AB
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Surface smoothness
ABC BC ABC C A ABC AB AB ABC BC ABC
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico
Particulate (powdery)
ns ns ns ns ns ns ns ns ns ns ns
Montepulciano Sangiovese Cannonau Nebbiolo Corvina Nerello Mascalese Raboso Primitivo Teroldego Sagrantino Aglianico

Figure 1.

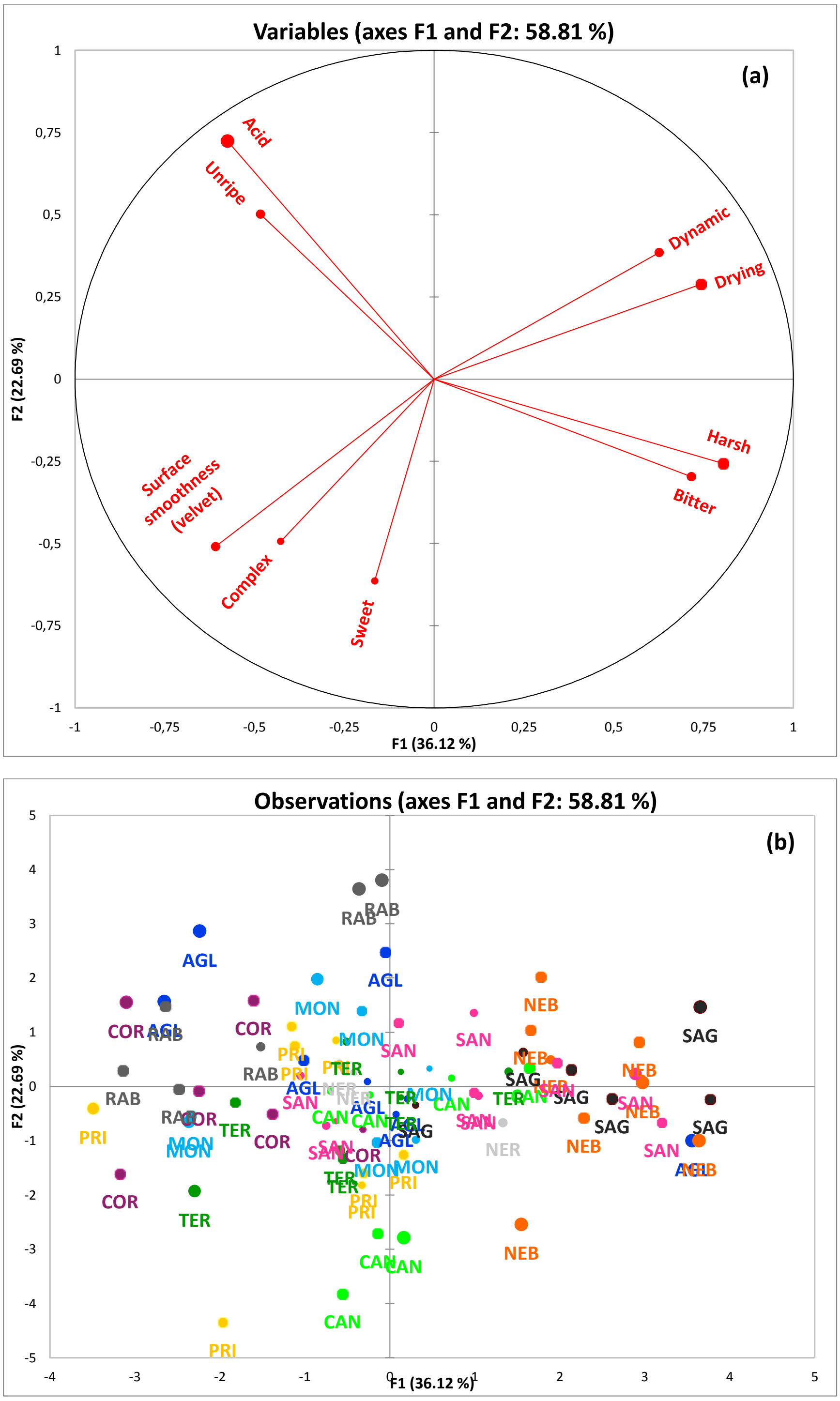
Variables (axes F1 and F2: 58.81 %)
(a)
Acid
Unripe
Dynamic
Drying
Harsh
Bitter
Surface smoothness (velvet)
Complex
Sweet
F1 (36.12 %)
F2 (22.69 %)
Observations (axes F1 and F2: 58.81 %)
(b)
F1 (36.12 %)
F2 (22.69 %)

Figure 2

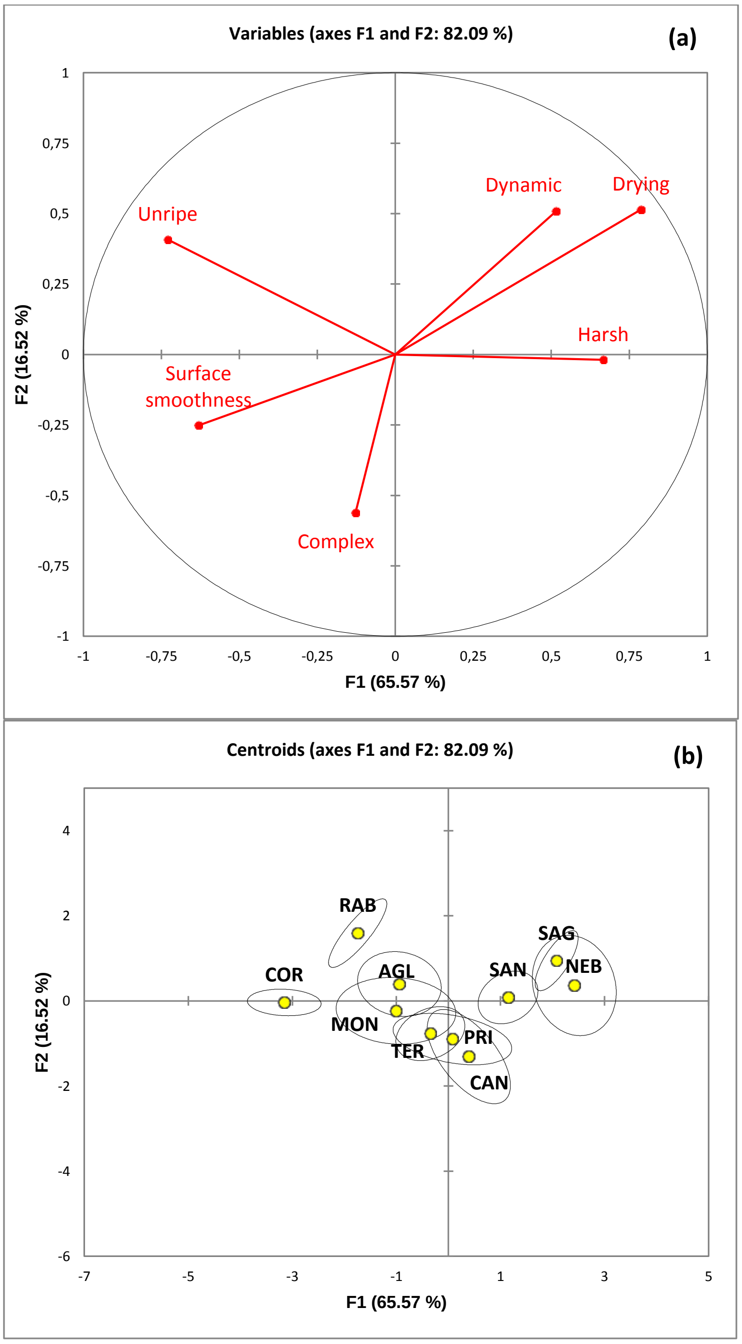
Variables (axes F1 and F2: 82.09 %)
(a)
Dynamic
Drying
Unripe
Harsh
Surface
smoothness
Complex
F2 (16.52 %)
F1 (65.57 %)
1
0,75
0,5
0,25
0
-0,25
-0,5
-0,75
-1
-1
-0,75
-0,5
-0,25
0
0,25
0,5
0,75
1
Centroids (axes F1 and F2: 82.09 %)
(b)
RAB
SAG
COR
AGL
SAN
NEB
MON
PRI
TER
CAN
F2 (16.52 %)
F1 (65.57 %)
4
2
0
-2
-4
-6
-7
-5
-3
-1
1
3
5

Figure 3

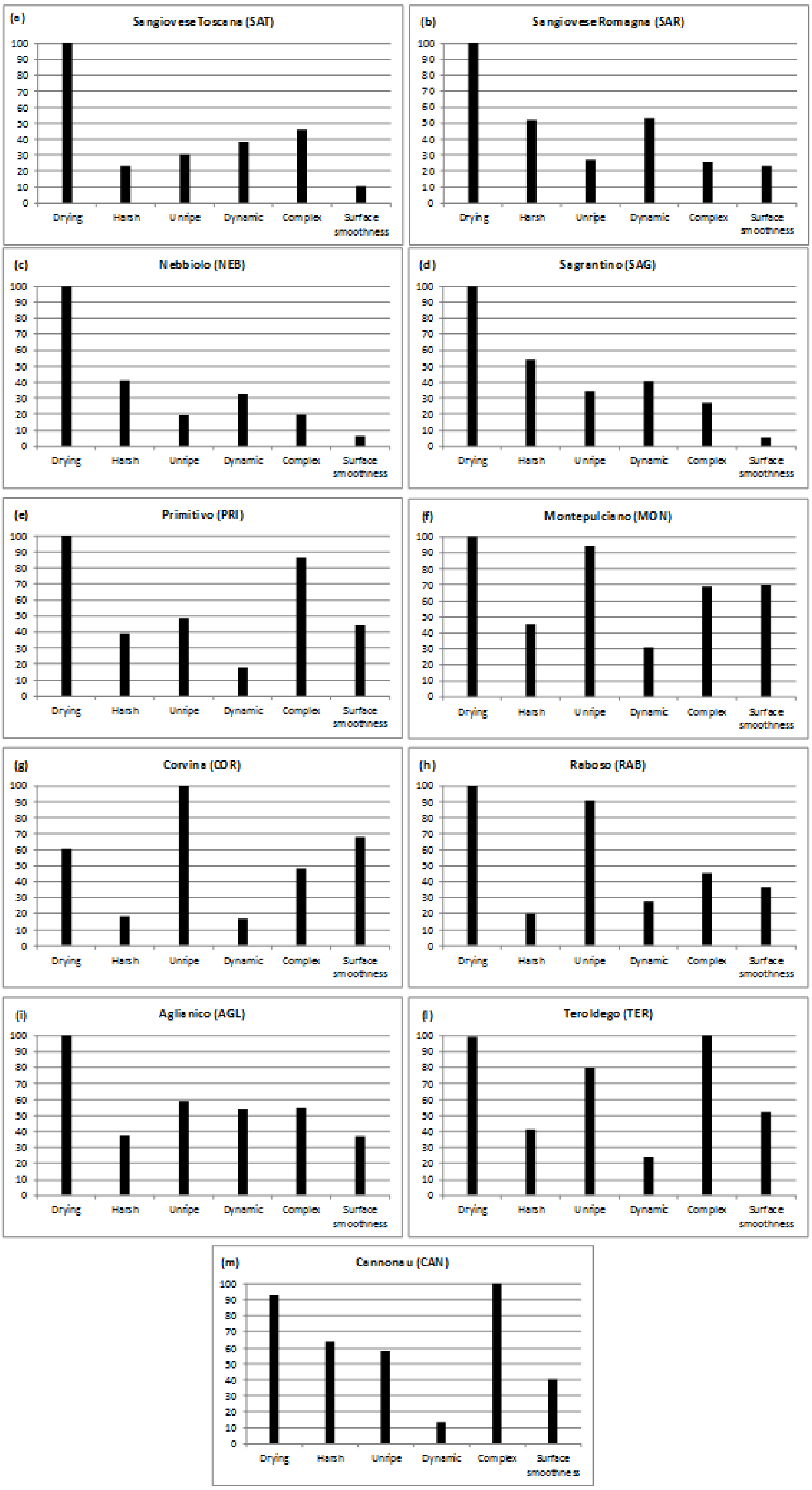
(a) Sangiovese Toscana (SAT)
(b) Sangiovese Romagna (SAR)
(c) Nebbiolo (NEB)
(d) Sagrantino (SAG)
(e) Primitivo (PRI)
(f) Montepulciano (MON)
(g) Corvina (COR)
(h) Raboso (RAB)
(i) Aglianico (AGL)
(l) Teroldego (TER)
(m) Cannonau (CAN)
0
10
20
30
40
50
60
70
80
90
100
Drying
Harsh
Unripe
Dynamic
Complex
Surface smoothness

Figure 4

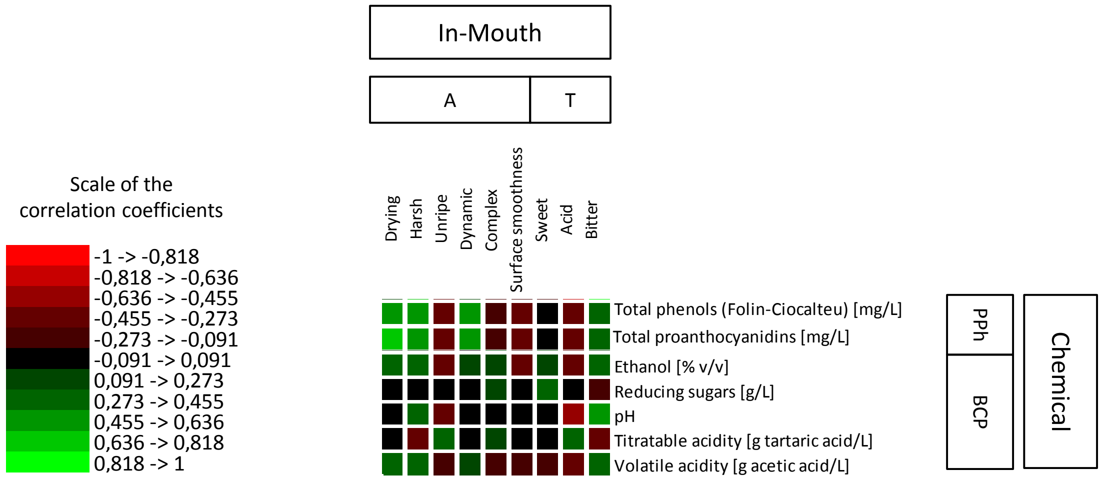
In-Mouth
A
T
Scale of the correlation coefficients
-1 -> -0,818
-0,818 -> -0,636
-0,636 -> -0,455
-0,455 -> -0,273
-0,273 -> -0,091
-0,091 -> 0,091
0,091 -> 0,273
0,273 -> 0,455
0,455 -> 0,636
0,636 -> 0,818
0,818 -> 1
Drying
Harsh
Unripe
Dynamic
Complex
Surface smoothness
Sweet
Acid
Bitter
Total phenols (Folin-Ciocalteu) [mg/L]
Total proanthocyanidins [mg/L]
Ethanol [% v/v]
Reducing sugars [g/L]
pH
Titratable acidity [g tartaric acid/L]
Volatile acidity [g acetic acid/L]
PPh
BCP
Chemical

Figure 5

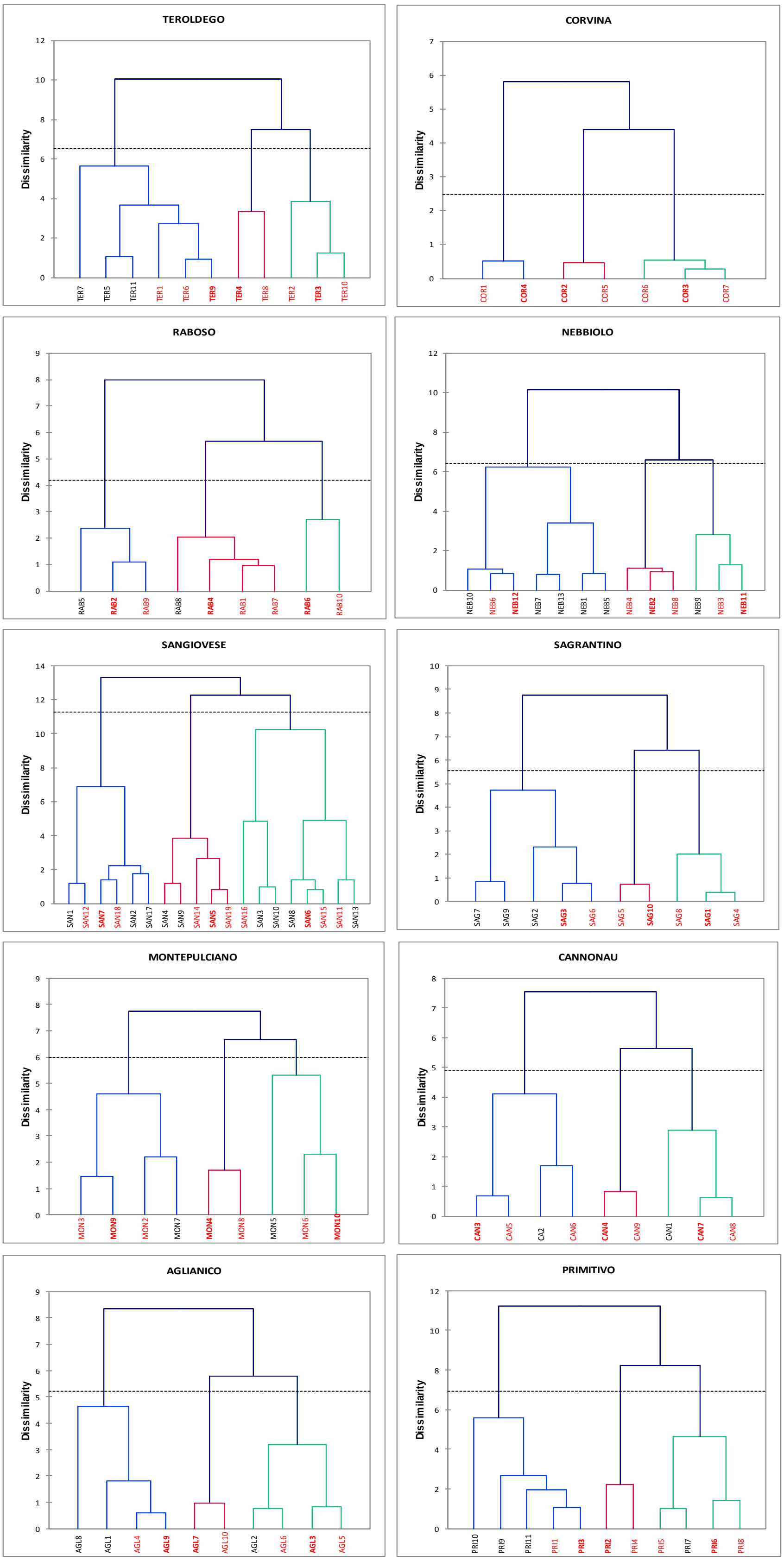
TEROLDEGO
CORVINA
RABOSO
NEBBIOLO
SANGIOVESE
SAGRANTINO
MONTEPULCIANO
CANNONAU
AGLIANICO
PRIMITIVO
Dissimilarity

Figure sm1