STUDY OF CROSS-MODAL INTERACTIONS THROUGH SENSORY AND CHEMICAL CHARACTERISTICS OF ITALIAN RED WINES

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Introduction

Wine is a hedonistic product and, more so today than in the past, its consumption and preference are sensitive to its quality, flavour and sensory characteristics (Chironi et al., 2013; Vecchio et al., 2019). Flavour results from the integration of all sensations perceived in the mouth and in the nose cavities, including olfactory (orthonasal and retronasal), tastes, and other oral sensations involving tactile and trigeminal perceptions (Prescott, 2012; Small & Prescott, 2005). During tasting, flavour perception is significantly affected by the interactions among sensory stimuli (Noble, 1996). Among all the sensory stimuli, odour and astringency are important characteristics in defining the complexity and quality of red wines and represent the two main intrinsic drivers of red wine consumers' purchasing decisions (Charters & Pettigrew, 2007; Peynaud, 1987; Sáenz-Navajas et al., 2016).

Around one thousand Volatile Organic Compounds (VOCs) have been identified in wines, ranging from hundreds of mg/L to the µg/L or ng/L level (Li, 2006), however only some of them are involved in wine olfactory complexity (Guth, 1997).

Astringency is a complex sensation mediated by both tactile and trigeminal receptors located in the mouth (Bate-Smith, 1954; Chen & Engelen, 2012; Jiang et al., 2014; Schöbel et al., 2014). It is commonly accepted that astringency stems from the interaction between tannins and flavan-3-ols with salivary proteins (Bate-Smith, 1973; Kallithraka et al., 1998; Soares et al., 2011; Soares et al., 2017); however, the chemical and sensory aspects behind the elicitation of the different sensations ascribable to astringency and characterizing different red wine styles are still unknown. Due to its complexity, astringency has been described by 33 different terms, grouped in seven categories, among which two terms are frequently referred to smoother astringency characteristics (i.e., complex and surface smoothness), while the other five usually describe stronger stimuli (i.e., drying, harsh, unripe, dynamic, and particulate) (Gawel et al., 2000).

In the literature, the interactions between non-volatile and volatile wine fractions, and between the sensory stimuli elicited by their constituents, are broadly reported to influence wine chemical and sensory characteristics. Orthonasal and retronasal olfactory perceptions have been reported to be strongly influenced by wine chemical components, such as polyphenols, due to their effects on aroma release (Pittari et al., 2021). At the same way, astringency perception has been reported to be strongly influenced by wine chemical properties (i.e., pH, acidity, ethanol, and polysaccharides) (Fontoin et al., 2008; Hort & Hollowood, 2004; Watrelot et al., 2018). Some works addressed the study of multimodal interactions (i.e., aroma–aroma, aroma–taste, taste–astringency and aroma–

astringency) and their sensory impact (De-la Fuente-Blanco et al., 2016; Sáenz-Navajas et al., 2012). Notwithstanding the unclear mechanisms at the base of those interactions, it is known that they impact wine sensory perception and quality (Cameleyre et al., 2018; Sereni et al., 2016). Hence, studying the cross-modal interactions of wine odour–mouthfeel stimuli is a subject of interest for wine researchers and producers to understand consumers' perceptions and choices.

Cross-modal sensory interactions have been explored in food products and beverages, mostly in model matrices (Poinot et al., 2013). In the case of wines, a limited number of works focused on wine-like solutions, and, even fewer, on real wine matrices, showing, moreover, contradictory results. In an early study (Sáenz-Navajas et al., 2010), applying a construction/deconstruction method, authors suggested that the addition of volatile fruity extracts from a Chardonnay white wine to the deodorized non-volatile extracts of a red wine decreased astringency and bitterness and increased sweet perception. Vice versa, the substitution of a white wine volatile matrix with a red wine one, caused an increase in astringency perception and a decrease in sweetness. In a subsequent experiment (Sáenz-Navajas et al., 2018), it has been demonstrated that the green mouthfeel character of red wines is positively correlated with vegetal aromas and negatively correlated with woody, ripe fruit, and oxidized ones. Moreover, some authors showed that in astringent model solutions for the presence of 2 g/L of catechin or epicatechin, the addition of specific VOCs with fruity, leather, and smoked notes (due to isoamyl acetate, ethyl hexanoate, damascenone, 4-ethylphenol, and 4-ethylguaiacol) increased the astringency persistence and intensity, underlining the relations between aromas and astringency (Ferrer-Gallego et al., 2014). Nonetheless, results from a very recent work (Sáenz-Navajas et al., 2020), conducted with and without nose clips, reported that except for the oily mouthfeel attribute, the perception of aromas did not have an impact on the other palate sensations of red wines, including numerous astringency descriptors (i.e., dry, sticky, dusty, grainy, sandy, coarse, fleshy, mouthcoating, silky and, gummy). Those results support a previous study (De-la-Fuente-Blanco et al., 2017), which concluded that aroma–astringency interactions were quantitatively not relevant in determining the astringency intensity levels of red wines, regardless of consumers' expertise level. By contrast, bitterness increased with animal aromas in the novice group.

Therefore, the effect of aroma modulation on astringency and taste perception remains an unclear subject that needs to be explored further, ideally in real wines showing different sensory characteristics and matrix composition. In a recent study, investigating the in-mouth sensory characteristics of 11 single-cultivar Italian red wines, we found that the 11 wine types showed diverse astringency patterns characterized by a different balance among the six astringency sub-qualities (drying, harsh, unripe, dynamic, complex, and surface smoothness) (Piombino et al., 2020). Moreover, testing the correlations between sensory (including the six astringency sub-qualities) and chemical parameters, results partially supported the hypothesis that olfactory cues related to wine VOCs might play a role in modulating the perception of some astringency sub-qualities. The exploration of this aspect is of interest and useful for oenologists to manage and control wine quality and to better comprehend consumer preferences/acceptance. Indeed, integrative brain processes, such as cross-modal interactions, could explain why it is difficult to find a direct correlation between specific compounds or chemical structures and astringency sensations that are of great interest for research and production.

For this reason, the main aims of this study were: (i) to investigate both odour–astringency (single sub-qualities) and odour–taste cross-modal sensory interactions in a wide set of real wine matrices, exploiting the sensory diversity of 10 single-cultivar Italian red wines; (ii) to test and compare the correlations between sensory (odour descriptors, astringency sub-qualities, and tastes) and chemical compositional parameters (total phenols, proanthocyanidins, ethanol, reducing sugars, pH, titratable acidity, volatile acidity) both in the presence and in the absence of VOCs. To do this, a sample set of 74 wines was assessed considering two types of evaluation conditions: whole wines (WWs) and corresponding deodorized wines (DWs), meaning wines with or without odorants, respectively. To exclude olfactory perceptions, a deodorization procedure was applied to make subjects comfortable with the sensory test and to simulate, as much as possible, the same breathing conditions experienced during a 'normal' wine tasting. Unlike previous methods applied for wine deodorization to study astringency or aroma (Lytra et al., 2012; Muñoz-González et al., 2014; Rodríguez-Bencomo et al., 2011; Sáenz-Navajas et al., 2010a; Sáenz-Navajas et al., 2010b), a new deodorization procedure was optimized to avoid the use of solvents and obtain representative deodorized wines that could be safely tasted by judges.

Materials and methods

1. Wine Samples

Seventy-four 100% single-varietal Italian red wines, all vintage 2016, harvested from 10 Italian grape varieties were sampled from the producers. The wine-set included: 10 Sangiovese (Romagna and Toscana), 8 Teroldego Rotaliano (Trentino-Alto Adige), 7 Corvina (Veneto), Raboso Piave (Veneto), Nebbiolo (Piemonte), Sagrantino (Umbria), Montepulciano (Abruzzo), Cannonau (Sardegna), Aglianico (Campania), and Primitivo (Puglia). Samples were selected from the most representative cellars of each production area, fermented in stainless steel vats at commercial scale, and sampled before malolactic fermentation and oak barrels ageing. Before bottling, all samples were protected with 50 mg/L of free SO_2 before bottling; bottles were closed with a Select Green 500 cork type (Nomacorc, Revisaltes, France) and stored at controlled cellar temperature (12 ± 2 °C) until analyses.

2. Sensory Analysis

Since the aim of the present research was to investigate the impact of the olfactory stimuli on astringency and taste perceptions during a red wine tasting, the 74 wine samples (whole wines: WWs) and the corresponding 74 deodorized wines (DWs) were characterized in terms of odour, astringency and taste properties using a descriptive sensory assessment on a nine-point numerical category scale (1 = very low, 2 = low, 3 = medium, 4 = high, and 5 = very high, with half values allowed).

2.1. Panel

Fourteen subjects (7 M and 7 F, 22-49 years old), recruited among students and researchers (Department of Agricultural Sciences, Division of Vine and Wine Sciences, University of Naples Federico II), participated to the study. They were selected based on their interest, availability, and ability to recognize olfactory and oral stimuli. They were all expert wine tasters and had previous experience in performing sensory tests on wine. All procedures were conducted in accordance with the ethical standards of the institutional and/or national research committee and with the 1964 Helsinki declaration and its later amendments or comparable ethical standards.

2.2. Procedure

2.2.1. Panel training

Judges' selection and familiarization with 10 in-mouth sensations (seven astringency sub-qualities: drying, harsh, unripe, dynamic, particulate/powder, complex, and surface smoothness/velvet; and three tastes: sweetness, sourness, and bitterness) and with olfactory stimuli representative of different odour families and wine volatiles (fruity, dehydrated fruits, dried fruits: nuts, floral, vegetal, spicy, toasted, woody, earthy, alcoholic, and off-odours: phenolic, sulphurous, cork taint, maderised/oxidised) were performed according to procedures and standard materials previously reported (Piombino et al., 2020; Pittari et al., 2020).

At the end of each training session, the perceived sensations were discussed with the participants to prevent overlapping and redundancies among terms and to help their memorization. Finally, to familiarize the jury with the application of the procedure to real wines as well as to test panellists' performances, 10 commercial wines (selected among samples under investigation) were assessed (two replicates) using the same evaluation procedure as run-through prior to the real analytical sessions. Subjects were provided with water and required to wait at least 15 s between each sample.

2.2.2. Sensory assessment

WWs and DWs were analysed by descriptive sensory assessment using the same vocabulary and the nine-point numerical category scale employed during the training. The 74 WWs + 74 DWs (two replicates), were assessed during 15 sessions, each session split into 2 sub-sessions with an imposed break of 15 min, and the evaluations of WWs or corresponding DWs were performed in each sub-session. All participants evaluated the 74 WWs by first smelling and scoring odours intensities, and then by tasting for astringency sub-qualities and tastes according to the procedure previously described (Piombino et al., 2020). The same tasting procedure was repeated on DWs in a separated sub-session.
Subjects were not informed about the nature of the samples. For each sample, 25 mL were served in covered glasses (ISO, 1997) coded with three-digits and presented in a randomized order. Wines were served at room temperature (21 ± 1 °C) and evaluated in individual booths (ISO, 2007).

3. Deodorization and Reconstitution of Wines

Drawing from methods previously reported in literature (Lytra et al., 2012; Rodríguez-Bencomo et al., 2011), a new rapid (~2 h) deodorization procedure was optimized to obtain representative and safe deodorized wines (DWs). WWs were deodorized during the two days preceding the date fixed for the corresponding session of assessment. Wines were deodorized one by one (two replicates) as follows: 360 mL of wine were weighed and treated in an ultrasound bath (Transsonic 460 H, Elma, Germany) with water as processing liquid, working at a fixed frequency of 35 KHz, and maintained at a controlled low temperature of 20 °C for 30 min. The samples were then evaporated at 30 °C under reduced pressure (Rotavapor R-210, Büchi, Switzerland). The process was stopped when the samples reached a weight loss of ~95% (~90 min). As the deodorization procedure stopped, the samples were weighed and reconstituted, one by one, at the initial weight by adding distilled water and food-grade ethanol at a proper concentration to reach the initial alcohol degree (%v/v) of the wine. DWs were then stored at (12 ± 2 °C) until the analysis. Any visual differences between reconstituted wine and real wine were ascertained on a subset of samples randomly chosen within each grape variety, by means of discriminant analysis [triangle test (ISO, 2004)]: differences resulted not significant (α=0.01). This test, along with an informal check to verify the absence of off-odours and off-tastes, was conducted internally at the laboratory. The efficacy of the deodorization was confirmed by Gas-Cromatography/Mass Spectrometry (GC-MS) analysis (Genovese et al., 2005) of the volatile fraction of the wines prior and after deodorization–reconstitution. Different methods for VOCs isolation were applied for the check: pre-concentration by SPME and liquid–liquid extraction as previously reported (Piombino et al., 2010; Piombino et al., 2020).

4. Chemical Analysis

Ethanol, reducing sugars, volatile acidity (VA), and titratable acidity (TA) were measured according to the Organisation Internationale de la Vigne et du Vin (OIV) methods (OIV, 2015). pH was determined by potentiometry (InoLab 730 pH meter, WTW, Weilheim in Oberbayern, Germany). Total phenols were measured by Folin–Ciocalteu assay (Singleton et al., 1999). The concentration of proanthocyanidins 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).

5. Data Analysis

Two Principal Component Analyses (PCA) were carried out on the correlation matrices (Pearson, $p<0.05$) of the mean intensities of each grape variety for significant in-mouth sensations and odours, for sensory characterizing WWs samples.
A three-way ANOVA (judges as random factor, grape variety and perception modality as fixed factors; Tukey, $p<0.05$) with interactions (grape variety*perception modality) was computed to test the discrimination effect of in-mouth descriptors and to evaluate the impact of the perception modality (with and without VOCs) on astringency sub-qualities and tastes perception across the 74 red wines. A two-way ANOVA (judges as random factor and wine variety as fixed factor; Tukey, $p<0.05$) was also computed to test the discrimination effect of olfactory descriptors across the 74 wine samples. To test the impact of olfactory cues on the perception of the astringency sub-qualities in the 10 wine types, other two-way ANOVAs (judges as random factor and wine as fixed factor; Tukey, $p < 0.05$

and 0.1) were performed on the intensity scores of astringency sub-qualities in WWs and corresponding DWs of each wine type.

Pearson correlation analyses ($p<0.05$) were applied to the whole set of wines (sample size: 74) for the computation of correlations between specific odour descriptors and in-mouth sensory variables, and between these latter for WWs or DWs and chemical parameters.

Performance of the trained judges was tested by a three-way ANOVA (Tukey, $p<0.05$) with three interactions: assessor*session, assessor*sample, sample*session [56].

Data was processed with XLStat (version 2019.6), an add-in software package for Microsoft EXCEL (Addinsoft, Paris, France).

Results and discussion

1. Olfactory/in-Mouth Cross-Modal Interactions

The main aim of this study was to investigate the impact of olfactory cues on tastes and astringency sub-qualities during red wine tasting. In order to account for the wide sensory diversity that different red wines can show, the experiments were carried out on 74 wines selected among the 111 Italian red wines (11 grape varieties), whose astringency has been recently studied (Piombino et al., 2020). As a first step, we tested the sensory diversity of the 74 wines produced with 10 grape varieties. The sensory features of the 10 single-varietal wines are shown in two separated PCAs computed on the mean intensities of oral (astringency and taste) characteristics (Figure 1a) and olfactory attributes (Figure 1b), respectively. The first biplot (Figure 1a) accounts for more than 74% of the variance, while the second (Figure 1b) for around 73%. The charts show the sensory diversity of the 10 wine types.

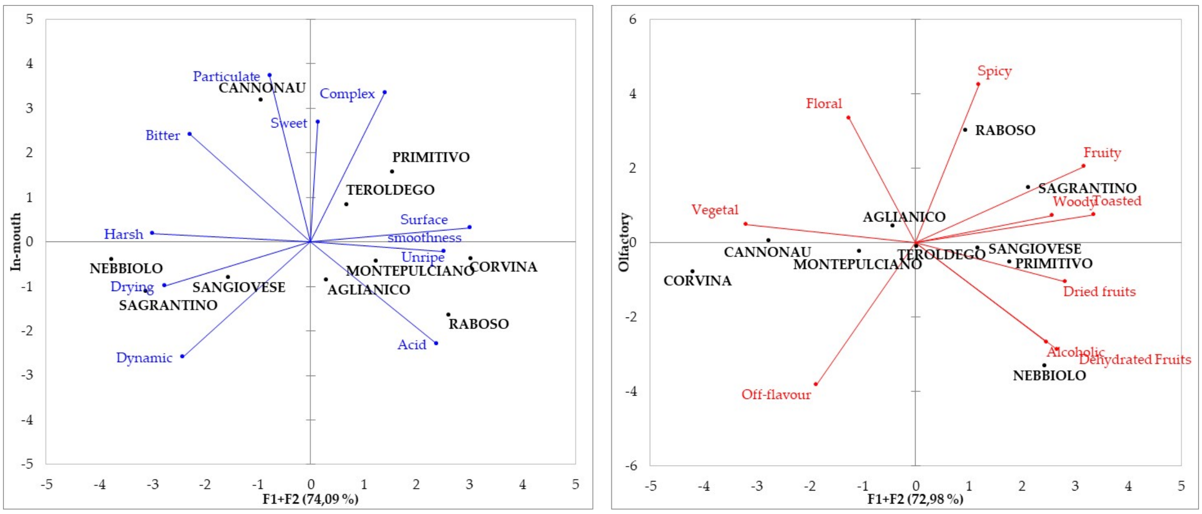

Figure 1. Principal component analysis (PCA) plots carried out on the correlation matrices (Pearson, p<0.05) of the mean intensities over the 10 single-varietal wines rated by the 14 judges for significant: (a) in-mouth (astringency and tastes) characteristics and (b) olfactory attributes.

In Figure 1a, Corvina and Raboso show the largest squared cosines to positive values of F1, where the variables surface smoothness, unripe and acid taste are well projected. Montepulciano and Aglianico occupy the same area but show lower squared cosines. On the opposite side of F1, Nebbiolo, Sagrantino and Sangiovese are all well correlated to harsh, drying, and dynamic astringency. Particulate, complex, and sweet sensations are well represented on positive F2 and correlated with Cannonau, while Primitivo and Teroldego are mostly correlated with complex and smooth astringency. Figure 1b shows that the sample set was representative of wines with different olfactory characteristics. F1 represents the contrast between wines with dominant vegetal odours, mainly Corvina and Cannonau, and those presenting different notes: fruity, toasted, and woody odours such as Sagrantino; dried fruits like Primitivo; dehydrated fruits and alcoholic notes like

Nebbiolo. On F2, opposite to off-flavours, there are wines with spicy and floral odours, such as Raboso and Aglianico, respectively. This latter wine along with Montepulciano, Sangiovese and Teroldego, has low squared cosines suggesting a lower and/or more balanced contribution of different odours.
The discrimination effect of oral and olfactory descriptors among the 74 wines was tested by ANOVA and results are reported in Tables 1 and 2, respectively.

Except for particulate/powder astringency, all the other nine in-mouth descriptors showed significant effects for the fixed factor grape variety (Table 1). Eight out of 11 olfactory descriptors resulted significantly different depending on the grape variety (Table 2): dried fruits (nuts) and woody were not significant and so it was the earthy descriptor, which was not considered for further analyses—including the PCA reported in Figure 1b—due to the lack of significance of its model. These first results confirm an inter-varietal sensory diversity of the 10 monovarietal wines. This diversity represents the assumption for the investigation of cross-modal sensory interactions between olfactory cues and in-mouth sensations during red wine tasting.

Oral descriptor	Model		Grape variety		Perception modality		Perception modality * Grape variety	
	F	P	F	P	F	P	F	P
Drying	15,488	**< 0,0001**	14,557	**< 0,0001**	0,191	0,662	1,438	0,157
Harsh	10,697	**< 0,0001**	11,253	**< 0,0001**	6,534	**0,011**	0,575	0,836
Unripe	11,541	**< 0,0001**	6,744	**< 0,0001**	11,293	**0,001**	2,046	**0,026**
Dynamic	10,241	**< 0,0001**	16,396	**< 0,0001**	11,001	**0,001**	1,976	**0,032**
Particulate/powder	5,858	**< 0,0001**	1,064	0,387	2,567	0,109	0,891	0,541
Complex	12,593	**< 0,0001**	3,658	**< 0,0001**	54,233	**< 0,0001**	1,368	0,189
Surface smoothness/velvet	7,881	**< 0,0001**	10,517	**< 0,0001**	4,313	**0,038**	0,807	0,622
Sweet	6,277	**< 0,0001**	5,112	**< 0,0001**	8,710	**0,003**	0,397	0,948
Acid	6,913	**< 0,0001**	16,876	**< 0,0001**	0,002	0,963	0,911	0,522
Bitter	7,915	**< 0,0001**	10,126	**< 0,0001**	13,342	**0,000**	1,149	0,321

Table 1. Three-way ANOVA computed to test the discrimination effect of in-mouth descriptors and to evaluate the impact of the perception modality (with and without odours, WWs and DWs respectively) on oral sensory perception of the 74 samples. In bold significant differences (Tukey, P<0.05).

Olfactory descriptor	Model		Grape variety	
	F	P	F	P
Fruity	11,779	**< 0,0001**	2,663	**0,003**
Dehydrated fruits	5,621	**< 0,0001**	3,674	**< 0,0001**
Dried fruits (nuts)	2,836	**< 0,0001**	1,824	0,052
Floral	13,841	**< 0,0001**	3,787	**< 0,0001**
Vegetal	4,757	**< 0,0001**	6,862	**< 0,0001**
Spicy	6,549	**< 0,0001**	2,478	**0,006**
Toasted	4,975	**< 0,0001**	2,450	**0,007**
Woody	6,406	**< 0,0001**	1,166	0,310
Earthy	1,903	0,006	1,679	0,081
Alcoholic	2,680	**< 0,0001**	1,883	**0,044**
Off-odors	5,766	**< 0,0001**	4,508	**< 0,0001**

Table 2. Two-way ANOVA computed to test the discrimination effect of olfactory descriptors among the 74 samples. In bold significant differences (Tukey, P<0.05).

The histograms in Figure 2 illustrate how the astringency sensory profile of each of the 10 single-varietal wines changed after deodorization. We worked under the assumption of representative deodorized samples not only because our procedure has been developed from previous ones (Lytra et al., 2012; Rodríguez-Bencomo et al., 2011). We also considered that rotary evaporation at low temperature (30 °C) represents a method largely applied during the preparative steps for polyphenols analysis in several food matrices (including wine) by several methods (Stalikas, 2007).

The ANOVA highlights several significant differences ($p<0.05$, $p<0.1$) in mean intensities of perceived sub-qualities assessed in WWs and corresponding DWs. At least one significant variation resulted for each wine type. Sagrantino's astringency was impacted the most after deodorization, with four astringency sub-qualities (harsh, dynamic, complex, and particulate) whose mean intensity significantly varied in the absence of olfactory cues. Three significant variations were detected for both Sangiovese (unripe, complex, and surface smoothness) and Aglianico (unripe, complex, and drying) and two for Nebbiolo (unripe and drying) and Primitivo (dynamic and complex). The astringency of the remaining wines was less affected by the absence of VOCs, where significant variations were detected only for one sub-quality, namely complex for Raboso, Cannonau and Teroldego, and unripe for Corvina.

Figure 2. Histograms illustrating the impact of the deodorization on astringency sub-quality profile of each of the 10 single-varietal wines. Significant differences assessed in WWs (dark red) and corresponding DWs (light red) are marked with asterisks (p<0.1, ** p<0.05).*

Two sub-qualities were the most frequently impacted by the deodorization: complex was perceived as significantly less intense in 8 out of 10 wine types (Raboso, Sangrantino, Sangiovese, Aglianico, Primitivo, Cannonau, Teroldego and Montepulciano) and unripe in four (Nebbiolo, Corvina, Sangiovese and Aglianico). This is not surprising because both these astringency sub-qualities correspond to sensations including not only oral but also retronasal olfactory perceptions. Indeed, based on the original definitions (Gawel et al., 2000), our jury developed and used consensual definitions as previously reported (Piombino et al., 2020): complex was intended as a balanced in-mouth sensation of smooth astringency, acidity and retronasal stimulation; unripe corresponded to an unbalanced in-mouth sensation of astringency, acidity, and green aroma.

The direction of the variation was always the same for all sub-qualities across all monovarietal wines, except for two terms: drying that slightly varied ($p<0.1$) in Nebbiolo and Aglianico but in opposite direction; and unripe that increased ($p<0.05$) for deodorized Nebbiolo, Sangiovese and Aglianico, while decreased ($p<0.05$) for deodorized Corvina. This result could be linked to the strong vegetal odours detected in these wines (Figure 1b), in line with previously reported high concentration of VOCs, such as cyclic terpenes and hexanols, characteristic of Corvina wines and responsible for its vegetal/herbaceous/balsamic character (Paronetto et al., 2011; Slaghenaufi & Ugliano, 2018).

A recent study (Sáenz-Navajas et al., 2018), aimed to identify chemical compounds driving green character in red wines, concluded that it is a multivariate character associated to both aroma and mouthfeel descriptors such as vegetal, astringency, green and dry tannins. Based on this knowledge, our hypothesis is that the strong vegetal odours of Corvina can enhance the perception of unripe astringency. This synergic/additive effect could be the reason why, unlike Nebbiolo, Sangiovese and Aglianico (Figure 2b,e,f) that were not characterized by vegetal odours (Figure 1b), in Corvina the unripe astringency was perceived more intense in WWs (Figure 2c). This hypothesis seems to be supported by a similar trend detected in Cannonau (Figure 2h) which, like Corvina, was strongly characterized by vegetal odours (Figure 1b). In order to get a more general result, an ANOVA ($p<0.05$) was applied across the whole set of 74 wines belonging to the 10 different grape varieties, to evaluate the impact of the perception modality (presence or absence of VOCs) and of the interaction “perception modality*grape variety” on in-mouth sensations assessed in WWs and DWs. Results reported in Table 1 show that the perception of 7 (harsh, unripe, dynamic, complex, surface smoothness, sweet, and bitter) out of 10 in-mouth sensations was significantly affected by odours. Complex astringency is the most impacted by olfactory cues ($p<0.0001$) while both the unripe and dynamic sub-qualities were significantly affected by the interaction “perception modality*grape variety”. The variation of mean intensities (across 74 wines) of each astringency sub-quality and taste sensation during DWs tasting compared to corresponding WWs is represented in Figure 3.

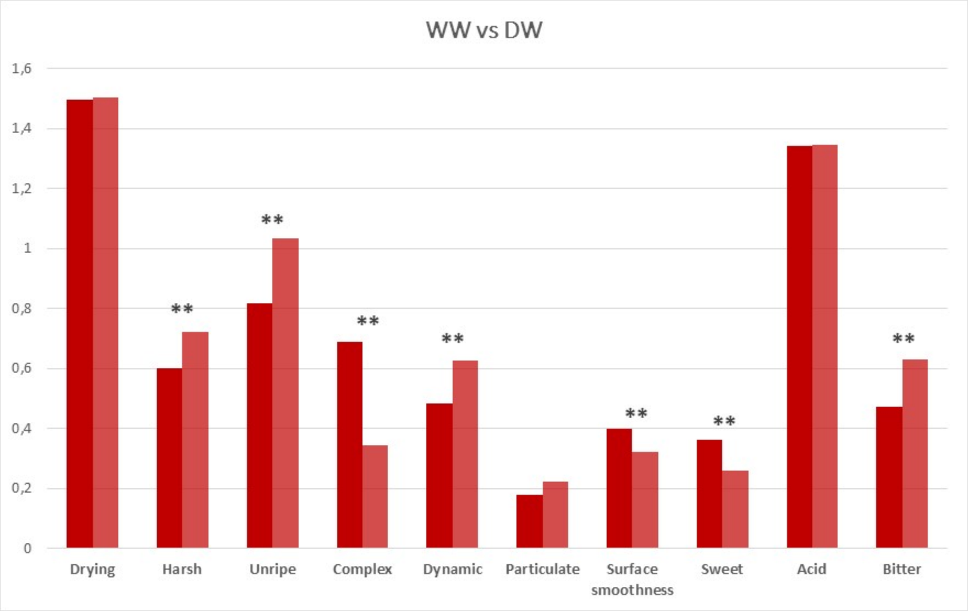

Figure 3. Variation of mean intensities (across 74 wines) of each astringency sub-quality and taste sensation during DWs tasting compared to corresponding WWs. Significant differences assessed in WWs (dark red) and corresponding DWs (light red) are marked with asterisks (p<0.1, ** p<0.05).*

Except for drying and particulate, the other astringency sub-qualities (harsh, unripe, dynamic) were perceived stronger in DWs. This suggests that olfactory perception can smooth these mouthfeel sensations previously described as “strong astringency sensations” (Piombino et al., 2020; Vidal et al., 2017).

On the contrary, complex, and surface smoothness/velvet decreased in DWs, suggesting that olfactory cues can enhance smoother aspects of astringency. The lack of impact on particulate astringency could be because wines were not discriminable according to this astringency feature. The perception of drying astringency that, based on results from consumer studies (Vidal et al., 2015), is assumed to be the basic astringent sensation because the most easily associated to the global term astringency, was not significantly impacted by olfactory cues. A similar result has been already reported (Sáenz-Navajas et al., 2020). Moving to taste sensations, it can be observed that the perception of olfactory stimuli significantly impacted bitterness and sweetness. Bitterness increased in the absence of VOCs, in accordance with previous data (Sáenz-Navajas et al., 2020), whereas perceived sweetness decreased. Those results seem to confirm the ones of an earlier study (Sáenz-Navajas et al., 2012), in which wine sweetness and bitterness perceptions were found to be significantly impacted by aromas. Moreover, previous findings on the effect of aromas on cider tastes showed that, overall, aromas significantly modulated sweetness perception for ciders with a sugar content of around 35–40 g/L (Symoneaux et al., 2015). Although the residual sugar content of our samples was 1 to 20 g/L (Table 3), our results are in line with the mentioned work. Sourness sensation did not show significant differences between WWs and DWs, meaning that the perception of olfactory stimuli did not influence this taste attribute.

According to Figure 1b, the large set of wines showed a wide array of sensory characteristics matching the large range of basic compositional data reported in Table 3.

Parameter	Mean	Minimum	Maximum
Ethanol (% *v*/*v*)	13.89	11.42	16.62
Reducing sugars (g/L)	2.64	1.1	20.1
Titratable acidity (g tartaric acid/L)	5.75	3.99	9.99
pH	3.55	3.07	4.1
Total phenols (Folin-Ciocalteu) (mg (+)-catechin/L)	2354.46	703.59	5448.55
Proanthocyanidins (mg cyanidin chloride/L)	3364.8	627.75	6312.37

Table 3. Oenological parameters determined in the 74 Italian red wines.

Thanks to this diversity, we tried to go deeper into our investigation on cross-modal interactions in red wine tasting, by performing a Pearson correlation analysis to statistically test the relationships between specific olfactory notes and single astringency sub-qualities and tastes. Results report a total of 21 significant ($p<0.05$) correlations, 17 significant correlations between odours and astringency sub-qualities, and 4 between odours and tastes. However, in most cases the computed r value is very low and likely linked to a casual effect. For each astringency sub-quality, one to four significant correlations to olfactory descriptors were found. Fruity was slightly correlated with the complex astringency ($r = 0.308$). In a previous study (Sáenz-Navajas et al., 2010b), it was observed that the addition of a fruity aroma extract coming from a Chardonnay white wine caused a significant decrease in the perception of the global astringency in different red wine matrices. Lately, this output was not confirmed (Sáenz-Navajas et al., 2020). The descriptor dehydrated fruit positively correlated with drying ($r = 0.459$; $p<0.0001$) and harsh astringency ($r = 0.286$), while negatively correlated with surface smoothness ($r = -0.341$). This could suggest that these three sub-qualities are parts of one unique sensation, where smoothness complements strong sensations such as drying and harsh. A similar consideration was recently reported for silky and dry mouthfeel descriptors (Sáenz-Navajas et al., 2020). Dried fruit was the only odour descriptor never correlated with in-mouth sensory variables. Floral aromas showed very weak relationships: positive with the complex sensation ($r = 0.275$) and negative with harsh ($r = -0.284$). Vegetal odours were the only ones related to four sub-qualities. The correlation with unripe ($r = 0.385$; $p<0.0001$) and surface smoothness ($r = 0.237$) astringency was positive while the correlation with drying ($r = - 0.340$) and dynamic ($r = -0.291$) was negative. Spicy only correlated with complex ($r = 0.462$; $p<0.0001$) but it had the largest coefficient both within the whole dataset and, compared to the other odours correlated to this sub-quality: fruity and floral positively and off-flavour ($r = -0.307$) negatively.

These relationships are based only on a statistical approach and, as already stated, the low r values, suggest a casual effect. However, the three largest and significant correlations ($p<0.0001$) that were found—spicy and complex, dehydrated fruits and drying, vegetal and unripe—seems to be confirmed from a cognitive point of view. Indeed, according to Figure 1b, Raboso were the spiciest wines and after deodorization their astringency was perceived as significantly ($p<0.05$) less complex (Figure 2a), confirming the significant and positive correlation previously reported ($r = 0.462$). Nebbiolo was characterized by dehydrated fruits odours (Figure 1b) and the average astringency of deodorized Nebbiolo was perceived as less drying ($p<0.1$), in line with the computed positive correlation ($r = 0.459$). Finally, in accordance with the positive significant correlation ($r = 0.385$) between vegetal odours and unripe astringency, in Corvina wines, which were strongly characterized by vegetal notes (Figure 1b), the unripe astringency was perceived significantly ($p<0.05$) less intense in DWs compared to WWs (Figure 2c). A similar finding (even if not significant) was observed for Cannonau, which was the only other monovarietal wine associated with vegetal odours (Figure 1b). The green character has been negatively correlated to consumers' preference of red wines, resulting intensified vegetal notes, and masked by woody aromas (Sáenz-Navajas et al., 2018). Our results support both these conclusions: woody odours were significantly ($p<0.05$) correlated with unripe astringency, even if with a small negative correlation coefficient ($r = -0.257$). Moreover, alcoholic notes were negatively correlated ($r = -0.340$) with unripe astringency. These results are interesting and need to be verified by further experiments. Few significant correlations were detected between olfactory characteristics and taste sensations and, also in this case, the r values were very low. Sweet taste did not correlate to any odour, while sourness was positively correlated to floral and bitterness showed a low negative correlation with floral and a positive correlation with the dehydrated fruits and off-flavour. This latter descriptor was intended as inclusive of different kinds of wine off-odours (phenolic, sulphurous, cork taint, maderised/oxidised); however, the most cited off-odour was the phenolic/stable/animal taint (data not shown). For this reason, the positive correlation highlighted between bitterness and off-flavour seems to support previous results (De-la-Fuente-Blanco et al., 2017), according to which bitterness was enhanced by animal aromas.

Overall, our findings suggest that during red wine tasting, odour–oral cross-modal interactions could modulate the perception of specific astringency sub-qualities and tastes. Specific olfactory characteristics such as spicy, dehydrated fruits and vegetal odours, could drive this modulation effect for complex, drying and unripe sub-qualities, and this should be further explored by specific experiments.

2. Olfactory Cues and Correlations between Sensory and Chemical Variables

In red wine astringency research, one of the biggest challenges is to find analytical methods able to predict the perceived astringency. Several studies investigated the correlation between astringency as a sensory parameter and measurements essentially based on compositional/metabolomic (Hufnagel & Hofmann, 2008), spectrophotometric (e.g., 280 and 230 nm) (Boulet et al., 2016), and precipitation techniques (Ferrer-Gallego et al., 2012). Thanks to these studies and to those investigating how other wine components (e.g., ethanol, pH, etc.) can influence astringency perception, our knowledge about this sensory stimulus has greatly expanded. However, most of these studies tested the correlation between chemicals and the overall astringency but did not pay attention to the different sub-qualities of this attribute.

According to our recent results (Piombino et al., 2020), and a few further studies addressing this subject (Sáenz-Navajas et al., 2020; Vidal et al., 2018), the current analytical methods are not able to predict astringency in all its sensory nuances, and their predictive power varies depending on the parameter/method applied. We argue that odour–oral cross-modal interactions can affect the correlations between chemical and sensory parameters, thus interfering with the estimation of their predictive power. To test this hypothesis, we computed Pearson correlations between sensory (astringency sub-qualities, and tastes) and chemical compositional parameters (total phenols, total proanthocyanidins, ethanol, reducing sugars, pH, titratable acidity, volatile acidity) across the 74 whole wines (WWs) and the corresponding deodorized wines (DWs). In this way, we were able to compare the correlations under two different tasting conditions: in the presence and in the absence of VOCs. This comparison is reported in Tables 4 and 5, where several significant correlations ($p<0.05$, $p<0.0001$) were found. In most cases, the magnitude of the correlation coefficients between chemical and sensory parameters increased with wine deodorization.

Variables	Astringency													
	Drying		Harsh		Unripe		Dynamic		Complex		Surface		Particulate	
	WW	DW	WW	DW	WW	DW	WW	DW	WW	DW	WW	DW	WW	DW
Total phenols (Folin-Ciocalteu) [mg/L]	**0,469**	**0,622**	**0,284**	**0,506**	-0,189	0,166	**0,240**	**0,599**	-0,170	**-0,375**	**-0,292**	**-0,414**	**0,238**	**0,318**
Total proanthocyanidins [mg/L]	**0,561**	**0,703**	**0,297**	**0,577**	**-0,279**	0,110	**0,304**	**0,737**	-0,207	**-0,427**	**-0,304**	**-0,569**	0,163	**0,295**
Ethanol [% v/v]	**0,394**	**0,476**	**0,262**	**0,396**	**-0,264**	-0,137	0,094	**0,461**	0,016	-0,051	-0,178	-0,171	0,069	0,129
Reducing sugars [g/L]	-0,013	-0,014	-0,015	-0,165	0,059	0,055	-0,057	-0,017	0,206	0,125	0,043	0,196	0,109	-0,055
pH	-0,010	-0,010	**0,335**	**0,466**	**-0,274**	**-0,376**	-0,023	0,166	0,024	0,165	-0,071	0,106	0,134	0,055
Titratable acidity [g tartaric acid/L]	0,084	0,163	**-0,248**	**-0,313**	**0,258**	**0,493**	0,080	-0,066	-0,041	-0,186	0,033	-0,197	-0,032	0,025
Volatile acidity [g acetic acid/L]	0,193	**0,361**	0,201	**0,447**	-0,067	-0,158	0,215	**0,413**	-0,156	-0,198	-0,056	-0,165	0,051	0,086

Table 4. Correlation coefficients (Pearson) between astringency and chemical variables. Comparison between WW and DW. In bold significant differences (Tukey, p<0.05) (grey: p<0.0001).

On the one hand, the absence of VOCs led to greater positive correlations between drying, harsh, dynamic sub-qualities and total polyphenols, total proanthocyanidins, ethanol, and volatile acidity. On the other hand, the negative correlations of complex and surface smoothness with total phenols and proanthocyanidins were stronger for DWs. As an example, for DWs, total proanthocyanidins showed the greatest positive correlation coefficients with drying and dynamic, which increased from 0.571 to 0.703, and from 0.304 to 0.737, respectively, when compared to WWs. The correlations between volatile acidity and drying, harsh and dynamic became significant for DWs but not for WWs. All these results confirm previous findings on correlations between sensory and chemical parameters (Piombino et al., 2020; Sáenz-Navajas et al., 2020; Vidal et al., 2018) and show the impact of cross-modal oral/olfactory sensory interactions on red wine perception. Among the considered sub-qualities, in DW, unripe became the only one not correlated with chemical parameters linked to polyphenols. These results support the idea that the unripe astringency is a multisensory feeling greatly impacted by VOCs perceptions. As for unripe, also the complex sub-quality is defined as a mouthfeel including aroma sensations. However, unlike unripe, the magnitude of correlation coefficients with total phenols and proanthocyanidins became significant in DW even if with low r values.

The correlations that were detected in WWs between tastes and chemical parameters (Table 5) were confirmed and reinforced in DWs. The only correlation that was not significant in WWs and became slightly significant in DWs was the one between reducing sugars and sweetness (from 0.099 to 0.595). This suggests that the overall aroma might modulate the perception of sweetness in red wine, but further investigation is necessary. The significant positive correlation between pH and bitterness was stronger in DWs.

Variables	Taste					
	Sweet		Acid		Bitter	
	WW	DW	WW	DW	WW	DW
Total phenols (Folin-Ciocalteu) [mg/L]	-0,043	-0,118	-0,089	-0,179	0,168	**0,471**
Total proanthocyanidins [mg/L]	-0,067	-0,163	-0,102	-0,189	0,198	**0,498**
Ethanol [% v/v]	0,036	0,173	-0,210	**-0,331**	0,167	**0,327**
Reducing sugars [g/L]	0,099	**0,595**	-0,016	-0,079	0,019	-0,161
pH	-0,022	0,135	**-0,508**	**-0,656**	**0,371**	**0,529**
Titratable acidity [g tartaric acid/L]	-0,058	-0,115	**0,459**	**0,621**	**-0,276**	**-0,424**
Volatile acidity [g acetic acid/L]	-0,089	0,032	0,000	**-0,359**	0,145	**0,435**

Table 5. Correlation coefficients (Pearson) between taste and chemical variables. Comparison between WW and DW. In bold significant differences (Tukey, $p < 0.05$) (grey: $p < 0.0001$).

Among all the mentioned significant correlations, only a few can be considered good correlations (r > ±0.7). According to these, we can conclude that total proanthocyanidins is the better predictive chemical parameter for both drying and dynamic astringency, but the estimation of its predictive power is strongly affected by olfactory–oral cross-modal interactions.

To the best of our knowledge, this is the first time that this kind of comparison has been done. In our opinion, this approach, if applied to a wider variety of chemical parameters, could be helpful to research aimed at understanding which compounds and structures are related to different mouthfeel sensations. Results confirm the importance of cross-modal interactions on red wine perception and can help to optimize the current predictive analytical parameters/methods. In a perspective of precision oenology, these results could be helpful in the management of wine astringency during winemaking. Only a few and recent reports focus on the impact of the odour stimuli on the perception of single sub-qualities rather than overall astringency, and no experiment was ever carried out on very diverse Italian red wines (Arapitsas et al., 2020; Parpinello et al., 2019; Piombino et al., 2020).

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