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Chemical description and organoleptic evaluation of Pinot noir wines from different parts of Italy: a three year investigation

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ABSTRACT

Aim: this work gives a chemical description and sensory evaluation of several Pinot noir wines from different parts of Italy. For three subsequent years (2016-2018) the wine samples were submitted for in an Italian annual national Pinot noir competition aiming to define the best Pinot noir red wine from Italy. All of the wine samples were 3-years old (from vinification) at the moment they were analysed and evaluated; they were also registered for the competition the same year they were put on the market.
Methods and results: all the wines were evaluated by a tasting panel composed of oenologists and wine experts, using the overall sensory quality as the descriptor. For the chemical screening, standard oenological chemical parameters (total acidity, colour, alcohol degree, total phenolic content, tannin indexes, etc.) and the content of the most abundant phenolic molecules by means of LC-MS analysis (triple quadrupole with internal standards) were determined. Pinot noir red wines produced from different parts of Italy showed a high variability for most standard wine chemical parameters considered, while the content of most single phenolic constituents was more retained and consistent with data from literature; except for *t*-resveratrol, which was significantly higher in our analyses, and delphinidin-3-glucoside, which was lower. Moreover, changes regarding the corresponding wines from the three vintages were noted. A correlation between the chemical parameters and the tasting panel results was also attempted. The results from a statistical analysis confirmed that alcoholic content, malvidin-3-glucoside and total anthocyanins had the highest positive impact on quality scores, while gallic acid, color tonality and total phenolic content had the highest negative.
Conclusions: our results indicate that most wine producers have a conservative attitude with very slight differences found in the corresponding wines over the three years of investigation. The strong effects of agronomical, winemaking and ageing processes on chemical and sensorial features of Pinot noir red wines from Italy were also clearly shown. Compared to other monovarietal Pinot noir red wines from the same temperate area, single polyphenol content tended to be more retained than most standard chemical parameters.
Significance and impact of the study: an overall quality assessment of a monovarietal wine, with its typicity as the main goal of a sensorial investigation, appears to be different from an objective quality assessment carried out by trained professional personnel using single standardised descriptors. Positive and negative correlations exist between sensorial judgement and chemical parameters, and the multiple linear regression model revealed relationships between the wine score and the set of the most important wine score description parameters.

KEYWORDS

red wine, sensory evaluation, polyphenols, tannins, linear modeling

ABBREVIATIONS

Ha: hectare
TPC: Total Polyphenol Content
UHPLC-MS: Ultra High-Performance Liquid Chromatography-Mass Spectrometry
IAC: ionized anthocyanins content
Abs: absorbance
WS: wine score

Supplementary data can be downloaded through: https://oeno-one.eu/article/view/3098

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INTRODUCTION

Pinot noir is an important international grape variety which accounts for approximately 112,000 ha of world vine area, thus being one of the most widespread and important red grape variety for worldwide wine production (VV.AA. Focus OIV, 2017). In Europe, Italy is the fourth biggest producer of Pinot noir grape (preceded by France, Germany and Switzerland), with around 5046 ha of vine area, of which approximately 464 are in Alto Adige (South Tyrol) and 353 are in the Trentino provinces (Anderson and Aryal, 2013; VV.AA., 2017). Pinot noir vine is known to preferentially grow in relatively cool environments with a significant day-to-night temperature excursion, which hilly and mountain territories can provide (Robinson *et al.*, 2013). However, it is also considered to be very sensitive to the exogenous factors to which it is subjected, so that the wines obtained from different areas and/or following different agronomical and oenological procedures (in other words, from different terroirs) can be very different (Rigaux, 2010; Vaudour, 2005).

While the Pinot noir grape can be blended with other varieties for vinification, it is highly appreciated as a monovarietal grape for the production of both sparkling white (including Champagne) and still red wines. Pinot noir is considered to be an "elegant" red wine due to its well-balanced structure with fine organoleptic qualities (light colour intensity, medium to light body, medium to low phenolic tenor, typical olfactory profile; Robinson *et al.*, 2013), and it is also suitable for mid to long aging (Jaffré *et al.*, 2009). Depending on the growing area, harvest and clone, the wines of Pinot noir can have relatively soft tannins, and it has a characteristic pleasant fruity aroma. In young wines the smell of cherries and raspberries is dominant with a wide range of other fruity aromas. As an aged wine Pinot noir can have aromas of mulch, truffles or other fungi (Robinson *et al.*, 2013).

The profound effects of both genotype and environment on grape composition is well known. In terms of genotype, some specific chemical properties of the Pinot noir grape cultivar are currently well-established in scientific literature. Kennedy *et al.* (2002) reported a higher total proanthocyanidin content in Pinot noir berries (intended as seed + skin tannin, including monomers) compared to Cabernet-Sauvignon from the same area, with a much higher percentage of monomers, especially from seeds. Moreover, a particular composition of anthocyanin pigments with relatively low total content and a lack of acylated products has long been known (Wenzel *et al.*, 1987). When compared to the average of 64 other red grape cultivars, Mattivi *et al.* (2006) reported a much lower amount of total anthocyanins and a slightly lower amount of total flavonols. In terms of environmental and terroir influences, climate parameters (such as temperature and sun irradiation) are known to have an affect on the content of anthocyanins flavan-3-ols and flavonols in red wines, including Pinot noir. For example, sun-exposed Pinot noir grape clusters contain up to 10 times more quercetin glycosides than those that are shade-exposed (Bertamini *et al.*, 1998; Price *et al.*, 1995). Tannin and anthocyanin content and composition vary with different vine vigour (Cortell *et al.*, 2008); as a consequence, both temperature and sun exposure can also have a sensorial-organoleptic impact.

The chemical composition of grapes is obviously reflected in the corresponding wines. Pedri *et al.* (2019) have recently reported the chemical composition of several Pinot noir grapes, musts and wines produced on different sites of Trentino-Alto Adige from 1996 to 2001. All the wines described in their paper were 1-year old monovarietal products, with vines originating from the same clone and rootstock, and with similar harvesting time. Moreover, they did not undergo fining processes on wood supports (barrels and/or chips) during winemaking. The study was conducted to define the features of this wine variety, especially when produced in a particular area and, as far as possible, separately from any other agronomical and oenological variable. Some particular definite features of the wines emerge in this work: for example, total phenolic content (TPC) ranged from 1442 to 2126 mg/L, alcoholic grade from 12.1 to 13.2 % and titrable acidity from 4.3 to 5.0 g/L. Mawdsley *et al.* (2019) recently reported similar values for 1-year old Pinot noir wines from one site in California over three vintages (2016-2018). In contrast, Samoticha *et al.* (2017) reported lower alcoholic grade and higher titrable acidity for 1-year old Pinot noir wines from Poland produced during the 2014 vintage with a comparable maceration period. Such reports confirm that while genotype is crucial, geography, climate and vintage also affect wine chemistry and sensorial quality. Neverthless, values for key chemical parameters in Pinot noir

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wine can be very different from other important Italian and international red wines. Van Leeuw *et al.* (2014) described the phenolic composition of several commercial monovarietal red wines produced in different countries (including Italy) and how Pinot noir wines were an exception. This was because the range of phenolic compounds (flavonols, anthocyanidins, flavan-3-ols, phenolic acids, resveratrol, etc.) was significantly different from all the others, which showed limited differences among each other in most cases; for example, compared to Syrah, Merlot and Sangiovese wines, anthocyanidin and flavonol content was lower, while flavan-3-ol and phenolic acid content was higher. Again, ranges were wide when the different origins were considered. Landrault *et al.* (2001) studied the composition of several red wines from France, and their results show relatively higher content of catechins and acids and lower content of anthocyanins. When compared to Tannat wines, lower anthocyanin content and a lack of acetylated and p-coumarylated derivatives in Pinot noir wines from Uruguay has recently been confirmed (Piccardo *et al.*, 2019). All these data suggest that cultivar genotype is the key factor for the development of most of the chemical and organoleptic features of red wines, especially aroma and phenolic composition, and that mesoclimatic variability can also have a strong impact on the characterisation of terroir and the sensorial characteristics of a wine. Both elements must be considered when investigating the concept of typicity of wines obtained from a single cultivar.

Valentin *et al.* (2016) investigated colour as a driver for quality judgement of Pinot noir wines from New Zealand. From a literature review on wine evaluation and quality assessment, they discussed how perceived quality is usually related to wine attributes other than colour, including abstract wine concepts such as typicity and complexity, and to hedonic aspects such as likability. In particular, the authors underline the lack of published data and address how wine professionals, rather than trained panelists using descriptive ratings, judge wine quality either within a winery or the wider wine industry (e.g., as part of a jury at wine competitions), which is how they assess overall wine quality precisely. Results from sensory evaluations of a wine and its overall quality assessment can therefore be highly subjective and aleatory unless precise guidelines are provided. Official guidelines produce standardised descriptors for wine evaluation. However, some of these parameters (like limpidity, persistence and effervescence) have a more objective and clear definition compared to others (like quality, general impression and typicity) which retain a more indefinite and subjective character (OIV, 2009). Moreover, the different organoleptic and sensorial attributes that are usually considered during the evaluation of red wines (aroma, alcoholic grade, acidity, tannin quantity and character, colour) may not necessarily lead to the assessment of "genuine" (i.e., absolute) quality, especially if the determination of typicity of a monovarietal wine is the main goal of a sensorial investigation.

The aim of this work was to assess the best Italian Pinot noir red wines, which feature the highest qualitative organoleptic profile within the typical characteristics of the variety, among distinct batches deriving from a national competition which occurred during three consecutive vintages (2016-2018), and including wines with different geographical origins (regions and/or terroirs). Sensory evaluations and chemical analyses (standard wine parameters, tannin amount and characterisation, and single phenolics content) were performed, and the resulting data were evaluated in order to investigate the putative correlation between the two.

MATERIALS AND METHODS

1. Chemicals and reagents

Delphinidin-3-glucoside chloride (≥ 95 %), malvidin-3-glucoside chloride (≥ 95 %), isorhamnetin (≥ 99 %), myricetin (≥ 99 %), quercetin (≥ 99 %) and t-resveratrol (≥ 99 %) were purchased from Extrasynthese (Genay, FR). (-)-epicatechin (≥ 99 %), (-)-gallocatechin (≥ 90 %), astilbin (≥ 90 %), caftaric acid (≥ 90 %), coutaric acid (≥ 65 %), petunidin-3-glucoside chloride (≥ 95 %), procyanidin B1 (≥ 90 %), procyanidin B2 (≥ 95 %), procyanidin C1 (≥ 90 %), quercetin-3-glucuronide (≥ 90 %) and taxifolin (≥ 99 %) were from Phytolab (Vestenbergsgreuth, DE). (+)-catechin (≥ 99 %) and caffeic acid (≥ 98 %) were from Sigma-Aldrich (St. Louis, US). Gallic acid (≥ 99 %) was from Roth (Karlsruhe, DE), vanillin (99 %) and $FeCl_3$ (anhydrous, flushed with N2) were from Acros (Morris Plains, US). Grape seed extract solutions (95 % proanthocyanidins content) were from ArdaNatura (Piacenza, IT).

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Sulphuric acid (95-97 %) was from J.T.Baker (Phillipsburg, US). Hydrochloric acid (≈36 % - 12N) was from Fisher (Loughborough, GB). *n*-butanol (HPLC grade) was from Chemlab (Zedelgem, BE). methanol (gradient grade) was from VWR (Fontenay-sous-Bois, FR). Acetonitrile (LC-MS grade) was from Panreac (Barcelona, ES) and formic acid (LC-MS grade) was from Merck (Darmstadt, DE). Ultrapure deionized water was from Millipore MilliQ apparatus (Burlington, US).

2. Sampling

Wine samples were provided from wineries for an Italian annual national Pinot noir competition which took place in three consecutive years (2016-2018) in the dedicated sensory room at the Laimburg Research Centre (see "Sensory Evaluation" section for details). The geographical origins of the samples are shown in Figure 1. Further details (product specifications and values for all chemical analyses and sensory evaluation of all wines in each vintage) are listed in Supplementary Table 1 (ST-1). All wines were 3-years old as from winemaking at the time they were evaluated and analysed. Most wines were registered for the competition the same year they were released on the market. Shortly before tasting, samples of the same wine (3 to 5 regular labeled market bottles) were opened and pooled in a wine jar to ensure homogeneity, and three aliquots (0.2 L each) were immediately transferred to dark glass bottles, saturated with N_2 to avoid excessive oxidation, closed with screw cap and stored at +4 °C until chemical analyses were performed (standard wine parameters, tannin-proanthocyanidin analysis, LC-MS analysis). All the rest was for the sensory evaluation.

3. Sensory evaluation

The sensory evaluation took place on 07/04/2016 (2013 vintage), 06/04/2017 (2014 vintage) and 12/04/2018 (2015 vintage) using a tasting panel consisting of 20 commissions, each composed of two judges. All the judges were familiar with red wine evaluation, being professional oenologists or journalists for food press and possessing medium to good knowledge of wine tasting and wine evaluation. Wines were submitted to each commission in a randomised way and sequence. To avoid saturation of mouthfeel and tasting capability, each commission evaluated 50 wines, so that each wine was considered sufficiently detailed in evaluation after submission to 12-14 different commissions (depending on total number of samples). No particular descriptors were selected or suggested for the analytical evaluation of each wine: the panelists were asked to examine the organoleptic profile of wine samples, using their knowledge and experience to evaluate the positive and negative sensations received by their nose, mouth and eyes in relation to the typical paradigmatic features of Pinot noir wines. The aim of the evaluation was not to provide a detailed scientific description of the wines, but to assess their overall quality. For harmonisation among the members of the panel, three blind tastings of randomly chosen registered wines were performed before the beginning, followed by open discussion. On consensus, wines were given an overall quality score by each commission, with remarks based on a one hundred-point scale as follows:

- < 59: bad, defective, unacceptable wine;



FIGURE 1. Origin of Pinot noir wine samples from the Italian territory in the three years of examination.

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- 60 - 69: wine having no particular defects, but with some lacks (excessive tannins, acidity, etc.);
- 70 - 79: average quality wine, linear, with no remarkable attributes;
- 80 - 89: fine, neat wine, above the average, with positive expressions in smell and taste;
- 90 - 100: very good to excellent wine, with outstanding complexity and varietal character.

The ability of each commission to recognise the same wine and discriminate different wines was evaluated by blind tasting five samples in two replicates, and F values were calculated (Kobler, 2008). Only F values belonging to 95 % confidence interval were considered for acceptance of scores from a given commission, otherwise all scores from that commission were not considered. The final score for each wine (wine score, WS) was calculated as the median of all scores from the different commissions that evaluated a given wine.

4. Standard wine analysis

4.1. Sample preparation

Wine samples were filtered using a syringe filter (5.0 µm pore size) prior to analyses. No further treatment (including dilution) were performed except when specifically indicated.

4.2. Fourier-Transform Infra-red (FT-IR) analysis

Analyses of the standard wine parameters were conducted on WineScan™ SO2 Auto instrument (Foss, Hillerod, Denmark). Wavelengths in the mid-infrared spectrum (2400 nm–10000 nm) were used for our method, and the following parameters were determined: alcohol content (% v/v), reducing sugars (g/L), pH, total acidity (g/L), volatile acidity (g/L), total dry extract (g/L), glycerol (g/L), methanol (% v/v as anhydrous ethanol), malic acid (g/L) and lactic acid (g/L). Both internal and external control of the results were performed daily and monthly respectively, using standard reference samples from the laboratory and a proficiency test from 106 other laboratories. Instrument control and data collection were performed with FOSS Integrator software (version 1.7.8, Hillerod, Denmark).

4.3. Colorimetric analysis

Colorimetric analyses were performed using an automatic Hyperlab Plus UV/VIS spectrophotometer with thermostatic cuvette holder (Steroglass, S. Martino in Campo, Italy). Total polyphenol content (TPC) and ionised anthocyanins content (IAC) were determined using specific reaction kits (Steroglass) and absorbance (Abs) readings at 620 nm and 520 nm respectively. The two assays were based on the Folin-Ciocalteau (F-C) reaction and ionisation of anthocyanins in acidic conditions respectively. For TPC determination, the samples were diluted 1:4 and the results multiplied by the dilution factor and expressed as gallic acid equivalents. For IAC, the results were expressed as anthocyanins using a proprietary calculation based on averages from HPLC analyses. Wine colour was also objectively evaluated by measuring the absorbances at three different wavelengths (420 nm, 520 nm and 620 nm). Colour features of wines are expressed as intensity (the sum of Abs 420, Abs 520 and Abs 620) and tonality (ratio of Abs 420 and Abs 520). Instrument control and data collection were performed with Steroglass Hi software (version 0.44.5, S. Martino in Campo, Italy).

5. Determination of flavan-3-ols and tannins content

5.1. Sample preparation

Wine samples were centrifuged (20800 x g, 10 min at +4 °C) prior to analyses.

5.2. Total flavan-3-ols content (monomeric to oligomeric proanthocyanidins, including low-weight condensed tannins)

The vanillin assay was applied for the determination of catechins and proanthocyanidins-condensed tannins (Sun *et al.*, 1998). In a 2.0 ml plastic tube with screw cap, 20 µL of wine was mixed with 180 µl MeOH, 500 µL of sulphuric acid (25 % v/v in methanol) and 500 µL of vanillin 1 % (w/v) in methanol (sample) or with 500 µL of methanol (blank). Both sample and blank tubes were incubated for 30 minutes at 30 °C. Absorbance was read in a 2 ml Suprasil quartz cuvette (1,0 cm optic path) immediately after incubation; the difference in absorbance at 500 nm between the sample and the blank was compared to a calibration curve obtained from (+)-catechin solutions (0-250 mg/L in MeOH, 200 µL each level solution instead of 20 + 180 µL as for samples), and the results were expressed as mg catechin equivalents per L of wine. The analyses were

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conducted in duplicate and the means reported as a final value.

5.3. Total condensed tannins content (oligomeric to polimeric proanthocyanidins, all-range condensed tannin)

Since the vanillin assay can underestimate the presence of highly-polymerised proanthocyanidins (condensed tannins), total condensed tannin concentration was also measured using the method of Porter *et al.* (1986), as described with modification by Hagermann (2002). In a 15-ml glass vial with screw cap, 50 µL of sample was mixed with 950 µL MeOH and 6.0 ml of 5 % (v/v) HCl (12 N) in n-BuOH, then added with 200 µL of Fe(III)Cl_3 solution (1 % in HCl 2 N). An aliquot of 1,5 mL was immediately transferred in a quartz cuvette and read at 550 nm, for blank evaluation. Next, tightly closed vials were incubated at 110 °C for 50 min on a heat block plate; after cooling at room temperature, the samples were read at 550 nm. Absorbance was read in a 2 ml Suprasil quartz cuvette (1.0 cm optic path). The difference in absorbance between sample and blank was compared to a calibration curve obtained from grape seed extract solutions (95 % proanthocyanidins content; standard solutions 0-250 mg/L in MeOH, 1.0 ml each level solution instead of 50 + 950 µL MeOH as for samples), and results were expressed as mg grape seed extract equivalents per L of wine. The analyses were conducted in duplicate and the means reported as a final value.

6. Analysis of single polyphenols with LC-MS

5.1. Sample preparation

Wine samples were centrifuged (20800 x g, 10 min at +4 °C) prior to analyses.

5.2. UHPLC-MS/MS analysis

Identification and quantification of single polyphenols was performed on a UHPLC-DAD-MS/MS system (Thermo Scientific, Waltham, USA) consisting of an Accela 1250 quaternary pump, an autosampler, a column oven and an Accela PDA detector coupled with a TSQ Quantum Access Max triple quadrupole system equipped with a heated electrospray ionisation (HESI) ion source. The analysis was based on an LC–MS/MS method described for the analysis of Sangiovese wines, with slight modification (Arapitsas *et al.*, 2012). Briefly, polyphenols were separated on a Hypersil Gold C18 column (50 x 2.1 mm, 1.9 µm particle size, Thermo Fisher Scientific, USA) with pre-column 0.2 µm filter, using a linear gradient elution with solvent A (formic acid 2.5 % in deionised ultrapure water) and solvent B (formic acid 2.5 % in acetonitrile) as follows: 0-1 min 2.5 % (B), 5-6.5 min 16.5 % (B), 7.5 min 23.5 % (B), 10.5 min 55.0 % (B), 11-12.5 min 95.0 % (B), 13-16 min: 2.5 % (B). Flow rate was 0.5 mL/min and column thermostated at 40 °C. Injection volume was 2 µL. All compounds were detected and quantified in selected reaction monitoring (SRM) mode using at least one quantifier and one qualifier transition, which were previously determined by direct infusion of standard solutions and by comparison with the literature. The source conditions were as follows: voltage (both positive and negative) 1500 V, vaporiser temperature 450 °C, capillary temperature 150 °C, sheath gas 60 (arbitrary unit, Arb), auxiliary gas 20 (Arb). Retention time and MS/MS detection parameters for the polyphenols analysed in this study are summarised in Supplementary Table 2 (ST-2). All operations were controlled by Thermo Xcalibur Version 2.2 software (Thermo Scientific, Waltham, USA).

5.3. Quantitation and method validation

For quantitative analysis of Pinot noir wine samples, an external calibration curve was built for each analyte using standard solutions. Wine samples were first diluted 1:10 in deionised water, then all samples and calibration solutions were added with internal standards before analysis. Each sample was prepared in triplicate (technical replicates). 100 µL of sample or standard solution were diluted with 90 µl of deionized ultrapure water and added with 10 µL of an IS mix containing cyanidin-3-galactoside (4 mg/L), (-)-epigallocatechin gallate (20 mg/L), and quercetin-4-glucoside (20 mg/L). The internal standards used for each compound are included in ST-2. All samples and standards were injected as a single replicate, and a quality control (QC), prepared by pooling all the samples, was injected every 12th analysis to control the absence of chromatographic drift. Quantitative determination was obtained by comparing the analite/internal standard ratio in samples and the external calibration curves, which were obtained by plotting the analyte/internal standard response (area ratio, average of triplicate) against the concentration that was injected (µg/mL). Coutaric acid values

were corrected because of the purity of the standard available (≥65 %). Regression factor (r^2) was calculated by means of least-square analysis for linearity evaluation. Calibration data are included in ST-1. The lowest limit for the linear range (instrumental limit of quantification, ILOQ) was established as the lowest standard providing a quantifiable signal (i.e., 10 times signal-to-noise ratio). The intraday precision was expressed as the Coefficient of Variation (CV %) of 6 analyses of the QC in the same day, while the interday precision was also expressed as the CV % of 6 QC samples analysed on three consecutive days. Official guidelines were followed when validating the method. (Magnusson, 2014).

6. Statistical analysis

6.1. Exploratory analyses

All parameters were visualized in box-plots, and means, standard deviations and ranges were considered for each vintage separately. To evaluate statistical differences for all the parameters among the three vintages, Kruskal-Wallis and Wilcoxon tests were performed (the latter being specific for values measured in only two vintages). Pearson´s correlations coefficients were calculated between all indicators (standard chemical analyses, total proanthocyanidins and total tannins analyses, LC-MS analyses and score values).

6.2. Linear modelling

The final aim was to describe the wine score using all available chemical parameters. In order to have a model which explains relationships between wine score and the important chemical parameters, multiple linear regression analysis was used.

The equation for the overall regression model becomes:

$$WS = b0 + b1X1 + b2X2 + \ldots + bn*Xn + e$$

where WS is wine score and b0 is intercept (constant term), b1 to bn are the estimated regression coefficients, X1 to Xn variables (wine compounds) and n are number of variables. A step-wise selection approach was used, starting with the full model and then eliminating parameters according to Akaike's information criterion (Hastie et Pregibon, 1992; Venables and Ripley, 2002).

All statistical analyses were carried out using Excel® software (Microsoft, US) and the statistical software R (VV.AA., 2019).

RESULTS AND DISCUSSION

1. General aspects on samples and sensory evaluation

As previously stated, the primary aim of this work was to assess a ranking based on results from sensory evaluations of the overall quality of the organoleptic profiles of Pinot noir wines produced in Italy, taking into account the typical features of this red wine. Furthermore, a chemical description of the wines registered for the competition is reported, highlighting their most characteristic features and tentatively correlating them with the sensory evaluation and scores.

The competition was organised by the Research area Enology of the Laimburg Research Centre, and both tasting evaluations and chemical analyses took place on the Laimburg site (Vadena, BZ). This event aimed to identify and promote the best and most typical Italian red wine from Pinot noir grape. It was thus initially intended as an event with commercial and/or marketing implications other than a mere scientific description. It was decided that a jury which did not include specifically trained personnel would be more appropriate for this purpose, in order to give an objective evaluation corresponding to consumer feelings.

The number of wines registered for the competition was 72 in 2016, 56 in 2017 and 83 in 2018, and they were produced in the regions of Trentino-Alto Adige, Veneto, Friuli-Venezia Giulia, Valle d´Aosta, Piemonte, Lombardia, Toscana and Sicilia. Twenty-five of the same wines were submitted in all three consecutive years of competition, among which one was from Piemonte and the remaining 24 were all from Trentino Alto-Adige region. The highest percentage (around 75 to 85 %) of Pinot noir wines from all three vintages was notably produced in Trentino Alto Adige (Figure 1). In fact, the production of Pinot noir vines and wines from this region - adhering in most part to "Controlled Designation of Origin" (CDO) product specification - is relatively high when compared to the whole Italian territory, probably due to a positive combination of grape attitude (genotype and phenotype), pedoclimatic features for its development and historical tradition.

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2. Standard wine analysis and scores

Different approaches were used for the chemical characterisation of Pinot noir wine samples in this work. Automatised FT-IR instrumentals using mid-infrared spectrum (such as WineScan™ SO2 Auto) are widely used in routine analyses of standard parameters for wines, musts and juices, since fast and reliable results can be obtained for most of the key components required (Vilanova *et al.*, 2017; Whitener *et al.*, 2017; Jouanneau *et al.*, 2012). Spectrophotometric/colorimetric assays are also used to obtain information about important wine composition parameters in a reliable way; for example, total phenolic and ionisable anthocyanins content (Aleixandre-Tudo *et al.*, 2017). TPC is usually determined using a Folin-Ciocalteau reagent, which oxidises hydroxyl groups of polyphenols present in the wine sample in a highly alkaline medium. Ionisable anthocyanins can be determined due to their capacity to get ionised in an acidic medium. With this method, it is possible to determine the anthocyanins in an ionised form, but not those polymerised with tannic substances. Colour features of wines, together with vanillin and BuOH-HCl assays performed for proanthocyanidins-tannin evaluations, are also based on the colorimetric quantitative determination of reaction products.

The values obtained for standard wine parameters, total proanthocyanidin- tannin assays, and sensory evaluation over three years are summarised in Supplemetary Table 1 (ST-1).

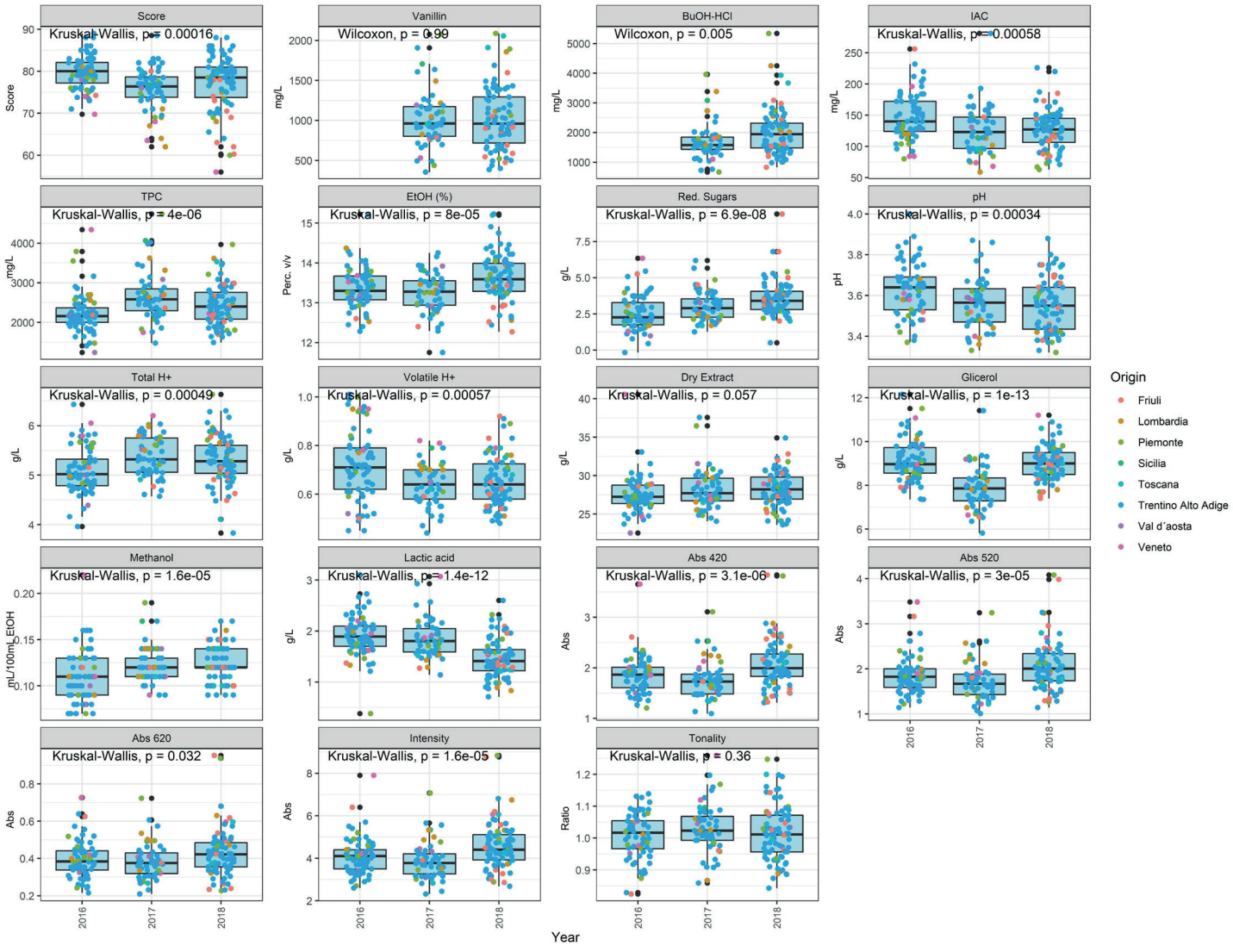


FIGURE 2. Year-to-year comparison of measured parameters from standard chemical analyses. Groups (vintages) are considered statistically different when $p<0.05$.

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Standard chemical parameters and sensory evaluation scores in the three vintages are summarised as box-plots in Figure 2. The trend followed by the indicators over the three years is also visible. It can first be noted that the evaluation scores and all the parameters that were taken into account showed significant differences between the three vintages ($p<0.05$), with only the exceptions of vanillin assay (vintage 2016 was lacking), dry extract residue and colour tonality. This means that a significant vintage effect exists in the period of investigation, which could be mainly due to different climate conditions affecting grape development and, to a lesser extent, to number/origin of samples. Glycerol, lactic acid, reducing sugars and TPC showed the highest differentiation and most chemical parameters showed high variability, as demonstrated by the wide range of values obtained in comparison with data reported for monovarietal Pinot noir wines. Overall quality, indicated by the score and expressed as an average ± standard deviation, was highest in 2016 (79.69 ± 3.97) and lowest in 2017 (75.82 ± 5.15), with 2018 being intermediate (77.07 ± 6.71). However, wines from Trentino-Alto Adige earned higher scores in all three vintages compared to the rest of Italy (80.42 ± 3.77 vs. 76.35 ± 3.11 in 2016; 77.32 ± 3.78 vs. 71.32 ± 6.17 in 2017; 79.20 ± 5.19 vs. 71.18 ± 7.01 in 2018), even if lower representativity of the other Italian regions has implications at a statistical level. The spread existing between Trentino-Alto Adige and the rest of Italy, especially for vintages 2017 and 2018, can be considered as a valid indicator of a better expression of Pinot noir wine and its typical characteristics from this terroir. A similar pattern to score along the three vintages is proper of chemical parameters such as IAC and glycerol. Similar development is displayed by EtOH, colour intensity, absorbances (420, 520, 620 nm) and volatile acidity, while an opposite trend is shown by TPC and total acidity. A relationship between certain chemical parameters of wine samples and their organoleptic quality is thus evident in the first instance.

All the wines showed very low values for residue malic acid (<0.01 g/L in most wines), meaning that they all underwent malolactic fermentation. It is interesting to note that in all three vintages certain parameters, like ethanol content and TPC, had different values to the typical averages and/or ranges for monovarietal Pinot noir wines from other European terroirs (Pedri *et al.*, 2019; Stój *et al.*, 2019; Van Leeuw *et al.*, 2014). In fact, results reported in this work indicate higher average values for TPC (2224 ± 497 in 2016, 2663 ± 613 in 2017, 2444 ± 507 in 2018), ethanol content (13.36 ± 0.50 in 2016, 13.24 ± 0.50 in 2017, 13.64 ± 0.59 in 2018) and total titrable acidity (5.10 ± 0.46 in 2016, 5.37 ± 0.40 in 2017, 5.31 ± 0.48 in 2018). These higher average values might also be a consequence of global warming occurring in the past decade. Moreover, it is common practice to "correct" some features during vinification, such as colour, aroma and body by adding hexogenous tannins and/or performing wood-based aging processes. These procedures can be performed both to exalt typical features of a wine and to render products more "stable" and/or "balanced", thus responding to general consumer tastes, sometimes at the expense of a wine's typicity. With the same goals in mind, it is fair to assume that single winemakers may choose not to develop Pinot noir as a single-variety product, but will add, for example, sweeter more coloured and higher-bodied blendings; such treatments can reflect the style of a winemaker and become his/her trademark. Depending on the procedures adopted for such corrections during winemaking and the blending components used - all complying with the protocol restrictions for production quality - some parameters may be deeply affected (e.g., colour, TPC and tannin indexes) while others (e.g., total acidity) may not.

The variability of some parameters is also valid when only Pinot noir wines from Trentino-Alto Adige are considered. As an example, the total polyphenolic content (TPC) for the corresponding 25 wines examined over three consecutive vintages from that area, with values ranging from 1500 to 4000 mg/L, are shown in Figure 3. High variability in some chemical parameters is evident, despite the similarities in microclimatic and pedological features, evoked by the same area of origin (Trentino Alto-Adige), and the strict protocol regarding the production of wines (COD), traceable for the whole batch considered (see ST-1). As previously mentioned, monovarietal Pinot noir wines from the same region had different values for such parameters (Pedri *et al.*, 2019). This is likely due to variations in agronomical (especially harvesting time), winemaking and ageing processes, which in turn can cause big discrepancies in both the chemical and organoleptic profile of Pinot noir, even when

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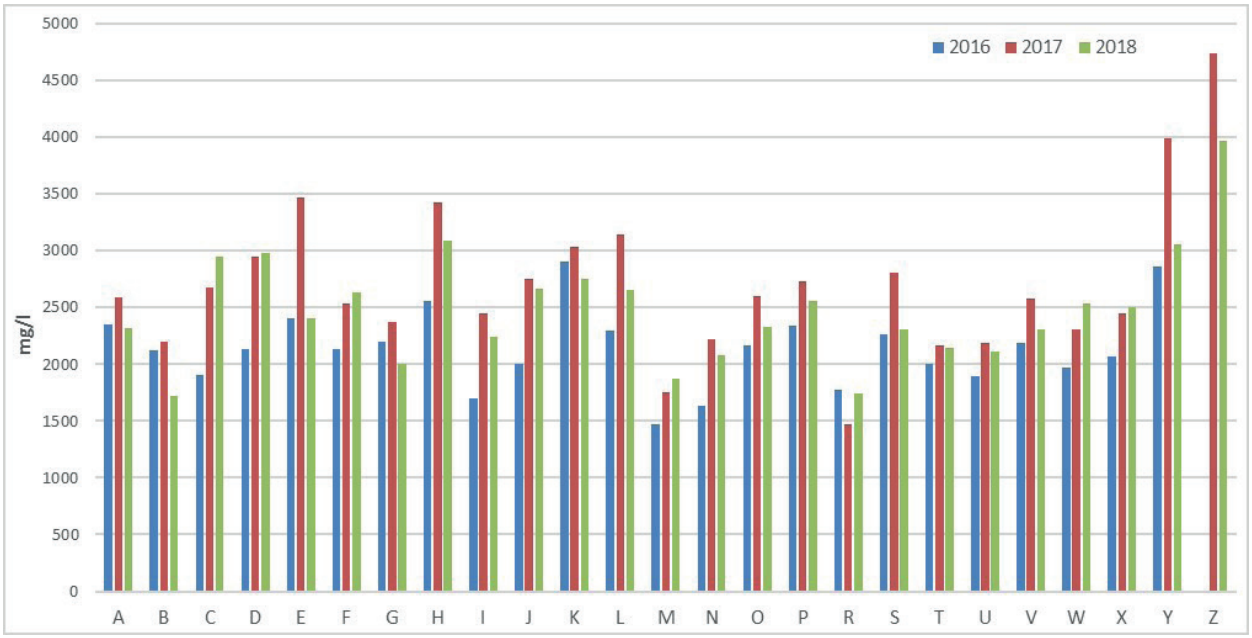


FIGURE 3. Total phenolic content (TPC) in corresponding Pinot noir wine samples from Trentino-Alto Adige in the three vintages (samples are listed as A–Z; see ST-1, row W).

from the same mesoclimatic area. To a lesser extent, it could reflect the highly sensitive and mutable character of Pinot noir vine as previously mentioned.

As can be inferred from Figure 3, TPC are highly retained over the three-year period, with a small "vintage" effect for most of them. This is probably the consequence of the winemakers' conservative attitude and pursuit of the preferred "style" of their own products, trying to maintain it over the years for better characterisation and recognisability; in other words, to promote their terroir.

Nevertheless, these factors do not result in harmonious results and scores. For example, "M" sample obtained 77 points in 2017 and 64 points in 2018, while "Y", having more than double the content of TPC, obtained 77 points in 2017 and 79.75 points in 2018. The multiplicity of factors involved in red wine production processes, the vintage effect and the complexity of red wine (in terms of organoleptic attributes to be evaluated) are thus all confirmed and likely to have affected this variability.

3. Flavan-3-ols and tannins content in Pinot noir wines and scores

Vanillin and BuOH-HCl assays are based on spectrophotometric determinations and were used in this work for proanthocyanidins and total tannins quantitation respectively in Pinot noir wine samples from 2014 and 2015 vintages (analysed in 2017 and 2018 respectively). Value ranges were 355 to 2087 and 668 to 5341 mg/L, with averages of 1024 and 1879 mg/L respectively (tab. ST-1). Rigo *et al.* (2000) previously reported 416 to 1741 mg/L for proanthocyanidins, and 669 to 2180 mg/L total tannin content in monovarietal 4 to 5 year-old Pinot noir wines from Trentino-Alto Adige (aged in bottle) using the same methods (ethanol was used instead of *n*-butanol for acid-catalyzed total tannins assay). The authors suggest that such wide ranges could be explained by blending with varieties having high proanthocyanidin content and/or oenological processes involving addition of proanthocyanidin-based tannins. They propose an index of condensation as a vanillin/BuOH-HCl assays ratio, with values ranging from 0.62 to 1.04; our values ranged from 0.44 to 0.84 in 2017 and from 0.34 to 0.69 in 2018.

The mechanism of reactions leading to the coloured products to be measured is different in the two assays. The vanillin reaction is based on adduct formation with flanan-3-ols C6 and/or C8 positions, depending on the presence of interflavanic linkages or other substituent groups, and the reaction yield is maximum for monomers and oligomers, becoming lower as proanthocyanidins become highly polymerised

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(Sun *et al.*, 1998). The reaction with BuOH-HCl leads to coloured products after the cleavage of unit-unit linkage, so that the reaction yield is null for monomers, increasing with the increasing polymerisation (Hagerman, 2002).

Both data sets show a high correlation with the TPC value in 2018 (Figure 4), so that the hypothesis of trivial results and/or artifacts can be discarded (the correlation coefficient values for the 2017 vintage were also >0.85; data not shown). Only 7 samples out of 83 show higher values for the BuOH-HCl assay than for the TPC assay in 2018, and 0 out of 56 in 2017. This is probably due to a particular composition in total phenolics and/or degree of polymerisation of condensed tannins, including the large extent of adduct formation with anthocyanins or other molecules, which prevent the Folin-Ciocalteau reagent from actingto its full potential.

The Vanillin assay had a higher correlation with the TPC assay than with the BuOH-HCl assay, indicating that flavan-3-ols comprise mostly monomers and small oligomers. The correlation between these two assays was high. An average degree of polymerisation around low-medium values (2 to 7), to which both assays give high reaction yield, can be invoked as the main reason for these results and for the indexes of condensation obtained. Oenological condensed tannins used during winemaking would also show a degree of polymerisation consistent with such values. Moreover, the grape seed extract used for the calibration of the BuOH-HCl assay probably shows a similar distribution in degree of polymerisation to most wine samples. These results are coherent with relatively young wines not yet subjected to aging, which would yield a higher average degree of polymerisation together with lower content of monomers and small oligomers.

4. LC-MS analysis

The values of single polyphenol content from the LC-MS analyses are discussed in this section (indicated as mean ± standard deviation, both mg/L). The complete listing is available in Supplementary Table 1 (ST-1). All these compounds are widely known to be main constituents of red wines and have been selected for having been previously reported as the most abundant polyphenols in Pinot noir wines (Samoticha *et al.*, 2017; Van Leeuw *et al.*, 2014; Kemp *et al.*, 2011). Data for these parameters are graphically summarised in Figure 5, allowing a comparison of the data spread for the three vintages. The comparison of parameters can be achieved by considering the scale of expression of the values. Again, the Kruskal-Wallis tests were performed to check the differences between parameters measured over three different years. For discussion, compounds have been grouped into their main classes (catechins and oligomeric procyanidins, anthocyanins, phenolic acids, flavonols, resveratrol) and are graphically represented for the three years in Supplementary Figures 1 to 5 (Figures S1 - 5). It should be highlighted that the parameters considered for the LC-MS analyses also showed significant differences in the three

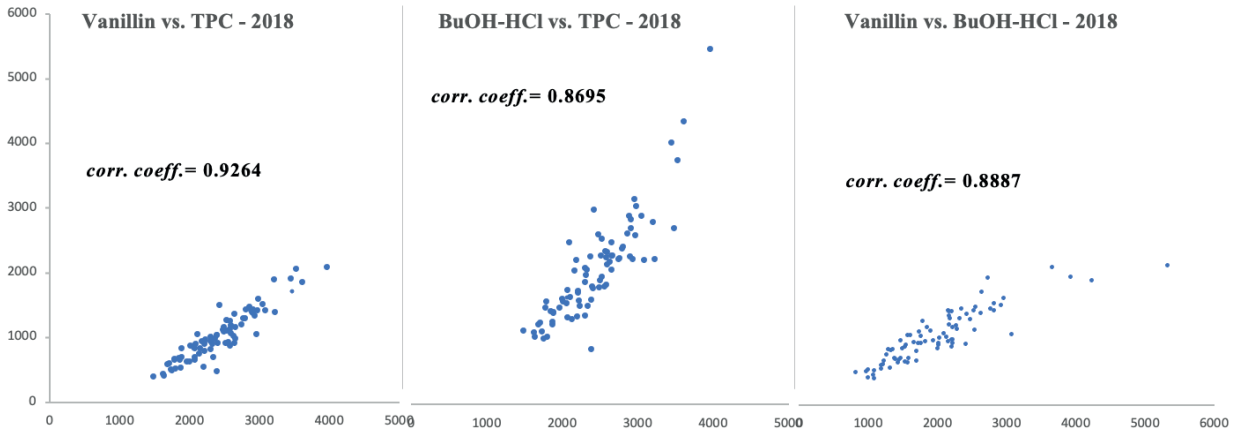


FIGURE 4. Correlation between values from vanillin, BuOH-HCl and TPC assays in 2018.
Values are expressed as mg/L on both axes.

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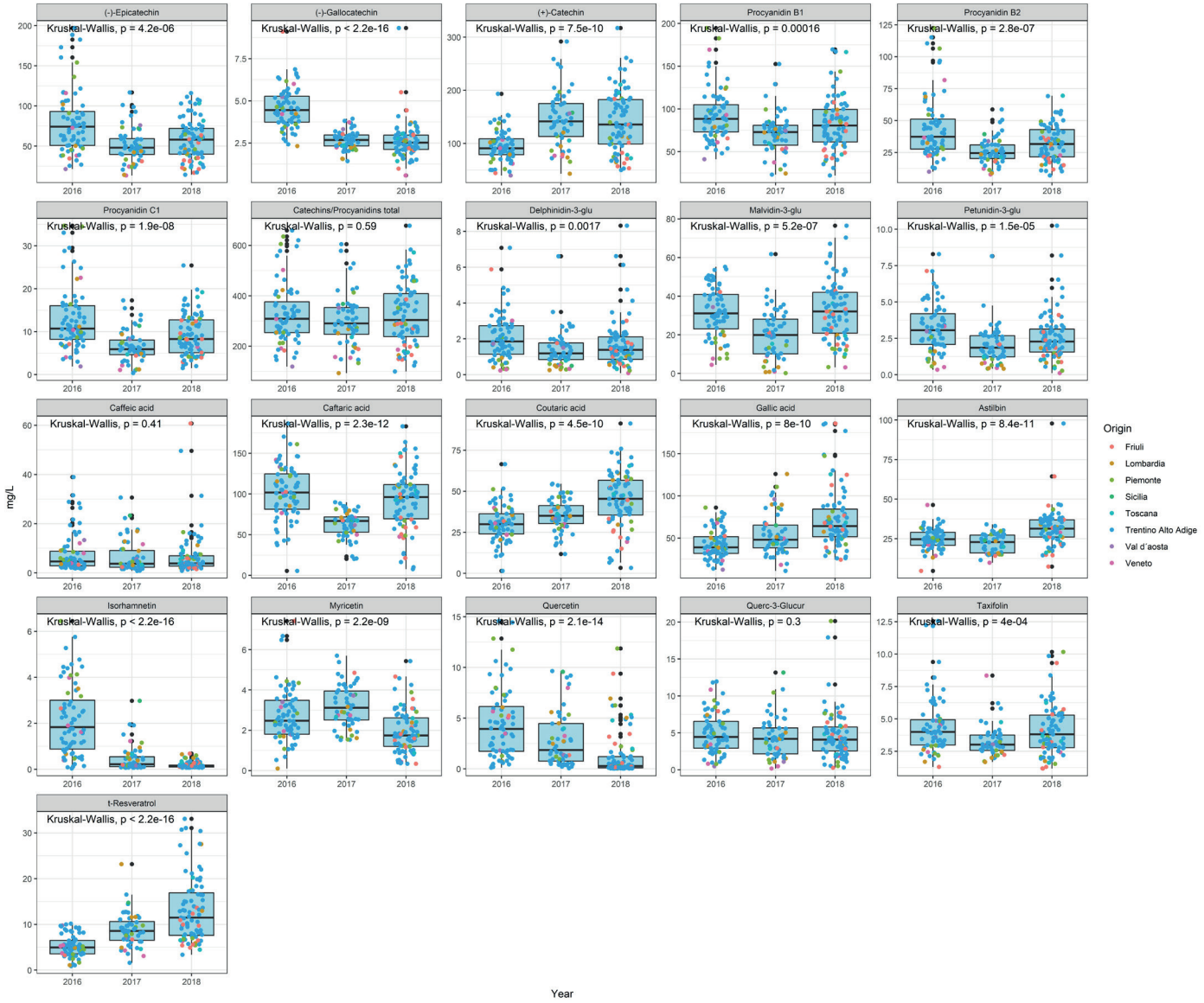


FIGURE 5. Year-to-year of measured parameters from LC-MS analyses for Pinot noir wine samples. Groups (vintages) are considered statistically different when $p < 0.05$ as expressed as mg/L

vintages ($p<0.05$), with the only exceptions being caffeic acid and total catechin/procyanidins. As previously noted for scores and results from standard chemical analyses, a vintage effect exists in the investigated period.

4.1. Catechins-procyanidins

The monomer (+)-catechin is the most abundant of the flavan-3-ols in the three vintages, except for vintage 2016 (mg/L: 94.4 ± 28.9 in 2016, 144.7 ± 51.7 in 2017, 150.0 ± 85.0 in 2018), when the values for procyanidin B1 were almost comparable (mg/L: 92.8 ± 30.3 in 2016, 71.3 ± 23.4 in 2017, 83.2 ± 31.3 in 2018), and the other main monomer (-)-epicatechin was slightly lower (mg/L: 79.5 ± 38.9 in 2016, 51.2 ± 21.7 in 2017, 57.9 ± 25.7 in 2018). In order of abundance, (+)-catechin is followed by procyanidin B1, (-)-epicatechin, procyanidin B2, procyanidin C1 and (-)-gallocatechin, a pattern which remains the same during the whole period. The content ratio of single monomers is consistent with previously reported data (Kemp *et al.*, 2011), while that of dimers and trimers could not be found in the literature.

The Vanillin assay showed stronger correlation with the sum of flavan-3-ol values from the LC-

MS (monomers + dimers + trimer) than from the BuOH-HCl assay (correlation coefficients are 0.6774 and 0.3259, respectively in 2017; 0.5873 and 0.3562 respectively in 2018). Since a relatively high content of flavan-3-ols monomers and oligomers has already been reported in Pinot noir wines (Van Leeuw *et al.*, 2014), which can also be seen in our data (the different reactivity of vanillin and BuOH-HCl assays with monomers and oligomers has already been discussed), an explanation for these correlation values can be found, further validating our data sets.

4.2. Acids

As previously reported by other authors, gallic and caftaric acid are usually the most abundant acids in red wines, even if their ratios can be variable (Van Leeuw *et al.*, 2014). Our results confirmed this evidence: caftaric acid was the most abundant in all three vintages (mg/L: 102.3 ± 33.6 in 2016, 62.9 ± 16.3 in 2017, 93.2 ± 36.1 in 2018), with gallic acid having similar values in 2017 (mg/L: 54.8 ± 24.9), slightly lower ones in 2018 (mg/L: 73.1 ± 36.5) and less than half in 2016 (mg/L: 42.9 ± 17.1). Coutaric acid and caffeic acid came next in terms of abundance.

4.3. Anthocyanins

Malvidin-3-glucoside was by far the most abundant anthocyanin in 2016, 2017 and 2018 (mg/L: 31.6 ± 12.8 in 2016, 19.8 ± 12.6 in 2017, 32.9 ± 16.1 in 2018), followed in order by petunidin-3-glucoside and delphinidin-3-glucoside. Ranges were similar in 2016 and 2018, with slightly lower values in 2017 for all three compounds. While being slightly lower in 2017, relative ratios were also similar in 2016 and 2018, with malvidin-3-glucoside having a ratio around 8- and 10-fold that of P-3-glu and D-3-glu respectively. The absolute values and their ratios are consistent with previously released data on free anthocyanidin content (determined after hydrolysis of glycosides) except for delphinidin-3-glucoside content, which is 70 to 80 % lower in our investigation (Van Leeuw *et al.*, 2014).

4.4. Flavonols and dihydroflavonols

Two glycosides, taxifolin-3-rhamnoside (astilbin) and quercetin-3-glucoside, together with the four aglycones, taxifolin, quercetin, isorhamnetin and myricetin were chosen as representatives of the flavonol and dihydroflavonol subfamilies to be quantified in the Pinot noir samples. Astilbin was found to be by far the most abundant (mg/L: 24.9 ± 6.5 in 2016, 22.1 ± 6.0 in 2017, 31.7 ± 11.4 in 2018), followed by quercetin-3-glucuronide, quercetin, taxifolin, myricetin and isorhamnetin (in that order) in both 2016 and 2017. In 2018, the pattern was similar, but quercetin and taxifolin were lower in content than myricetin. The ranges of values were very similar over the three years for all the six compounds investigated. Similar values for myricetin, quercetin and isorhamnetin in 2018 have previously been reported in Pinot noir wine from Trentino-Alto Adige (Baroň and Kumšta, 2013).

4.5. Stilbenes

Trans-resveratrol was considered as a representative of the stilbene subclass of polyphenols. Its range and average content in the three batches was lowest in 2016 and highest in 2018, with 2017 being intermediate (mg/L: 5.3 ± 2.4 in 2016, 8.8 ± 3.7 in 2017, 13.2 ± 7.1 in 2018).

Stervbo *et al.* (2007) reviewed levels of *t*-resveratrol in monovarietal red wines and discussed how it was found to vary greatly between varieties and regions of origin. Among all the samples examined, the highest average level of *t*-resveratrol was found in wines made from Pinot noir grown in France (5.4 mg/L), followed by wines made from Spanish and Italian Pinot noir (5.1 and 4.8 mg/L respectively). This shows that wines of the Pinot noir variety do indeed contain the highest average levels of *t*-resveratrol. Similar values were also reported for Italian Pinot noir by Van Leeuw *et al.* (2014). Our results only partially confirm these data, with a comparable average value obtained in the 2016 vintage and much higher values obtained in 2017 and 2018. Cool and humid conditions for vine growth are related to higher levels of *t*-resveratrol (Kolouchová-Hanzlíková *et al.*, 2004), but vinification techniques (such as double maceration) are also concomitant with high levels of *t*-resveratrol (Alonso *et al.*, 2002). The high values obtained in our investigation could be related to both these factors, including relation with vintages. Since *t*-resveratrol can undoubtedly be considered as one of the most promising polyphenolic compounds in terms of its antioxidant activity, this particular aspect of

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Italian Pinot noir wines could be highly interesting from a nutraceutical point of view.

5. Correlation between chemical parameters and sensory evaluation

Pearson´s correlation coefficients between all indicators (standard chemical analyses, LC-MS analyses and score values) were computed and are graphically represented as a heat map (Figure 6). Results from the tannin assays were not included since vintage 2016 was missing.

The list of all correlation coefficients between score and all other parameters is available in Supplementary Table 3 (ST-3). Their values ranged from +0.37 and -0.32. Higher positive correlation appears between score and alcohol content (EtOH %), malvidin-3-glucoside, IAC, petunidin-3-glucoside, caftaric acid, delphinidin-3-glucoside, astilbin and glycerol. Negative correlation is highest between score and gallic acid, colour tonality, TPC, Abs 420, methanol and colour intensity. Obviously, some of these characteristics (e.g., Abs 420 and colour tonality) are highly correlated with one another. Moreover, it is a fair assumption that some of these parameters are more objectively recognisable since they can directly and strongly affect tactile, gustative and visual sensations during sensory evaluation (e.g., glycerol, alcohol and anthocyanin-colour respectively); therefore, it is not surprising or accidental that they show higher correlation with overall quality value, especially if typical characteristics of a wine are reflected.

In particular, regarding single vintages, correlation coefficients between score and ethanol content were 0.470 in 2016, 0.339 in 2017 and 0.407 in 2018 (data not shown), being lowest in vintage 2017, which exhibited lowest ethanol contents. Such correlation of overall quality with alcohol content has already been noticed for Pinot noir wines by other authors (Jaffré *et al.*, 2009). As previously mentioned,

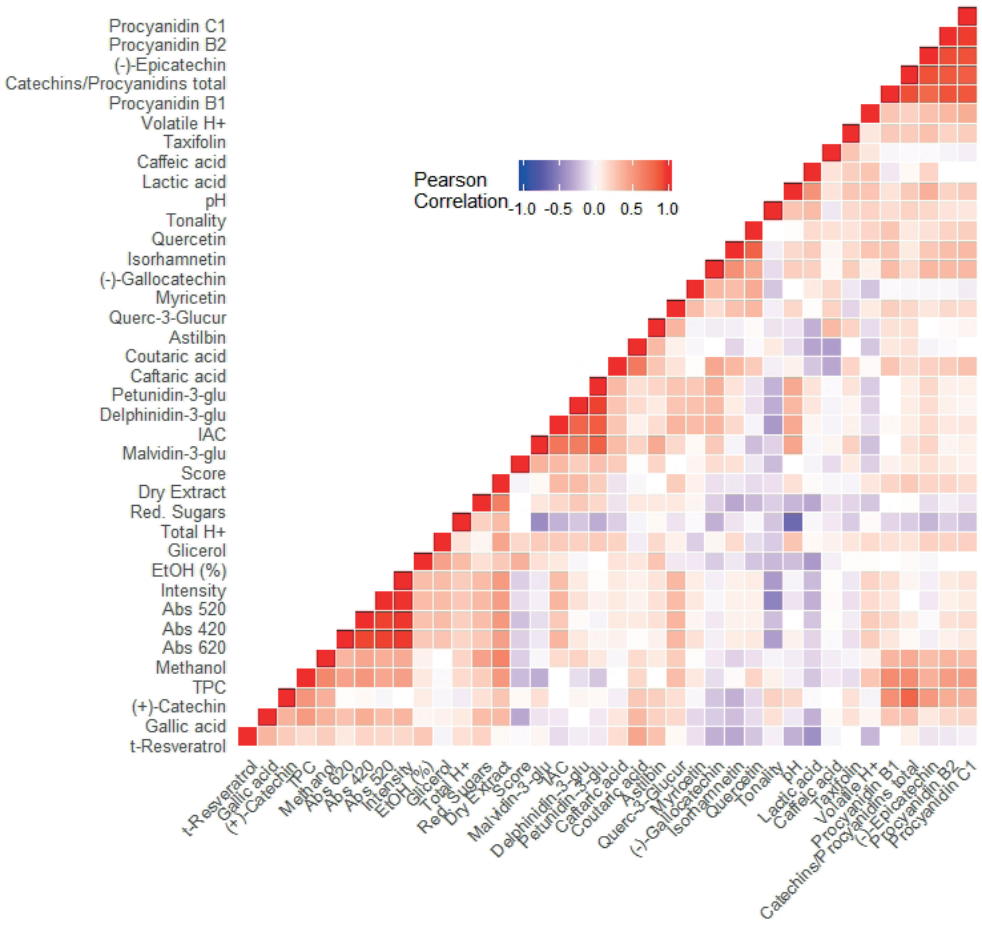


FIGURE 6. Pearson's correlation matrix - between all the chemical parameters and sensory evaluation score.

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the score represents the summary of different sensorial features as one number only; for this reason, weak correlations between such a general parameter and single characteristics of the wine are to be expected.

6. Linear modeling

To describe relationships between wine score and wine chemical parameters, the multiple linear regression is used. To avoid multicolinearity some highly correlated variables are excluded from the model and are represented by a single one from that the same group. In fact, because highly correlated parameters give the same information in models, it is possible to use one to represent the others in the same group. Thus, for example, "Abs 420", "Abs 520", "Abs 620" and "Intensity" are highly correlated, so Abs 420", "Abs 520" and "Abs 620" are discarded and "Intensity" is used as the representative one. "Quercetin" and "Isorhamnetin" are highly correlated, so "Quercetin" is used. "Catechins/procyanidins total", "(+)-Catechin", "Procyanidin B1", "Procyanidin B2", "Procyanidin C1", and "(-)-Epicatechin" are highly correlated, so "catechins/procyanidins total" is used. "Malvidin-3-glu" and "Petunidin-3-glu" are highly correlated, so "Malvidin-3-glu" is used. In the final model, 13 representative parameters have been selected to describe a wine score from the full set of parameters (Table 1).

Fitting linear models to the wine score led to a significant model (p value<2.2e-16, r^2=0.5), showing that wine score is influenced by multiple chemical features.

Parameters with a positive estimated value have a positive effect on wine score, whereas parameters with negative estimates decrease the wine score. As expected, parameters with the highest positive correlation (EtOH and IAC) and negative correlation (methanol, TPC and tonality) with the score are selected in the linear model. Moreover, it can be confirmed that wine quality, which is represented by wine score only, is not derived from just one component, but it is affected by several, as we can see from the multiple regression model.

CONCLUSIONS

A chemical description and sensory evaluation of Pinot noir red wines from different parts of Italy were performed in three consecutive years (2016-2018). All wines were 3-years old from production and were registered for the annual Italian Pinot nir competition taking place in the corresponding year. The purpose of the competition was to assess the best Pinot noir red wine from Italian territories in terms of overall quality. This was achieved using a tasting panel composed of experts in wine evaluation instead of specifically trained personnel. The panel applied their knowledge and experience to judge quality, taking into account the typical characteristics of Pinot noir wines and following

TABLE 1. Multiple linear regression model output with the selected chemical parameters for wine score description.

Term	Estimate	Std. error	t-statistic	p.value
(Intercept)	33.2383	10.0025	3.3230	0.0010617
IAC	0.0781	0.0169	4.6261	0.0000067
TPC	-0.0023	0.0009	-2.4930	0.0134892
EtOH (%)	3.3696	0.5908	5.7031	0.0000000
Red. Sugars	0.6229	0.2837	2.1957	0.0292801
Total H+	2.1168	0.7963	2.6582	0.0085016
Methanol	-25.4953	18.3567	-1.3889	0.1664358
Intensity	-2.1510	0.4496	-4.7846	0.0000034
Tonality	-10.2616	4.6519	-2.2059	0.0285467
Catechins/Procyanidins total	0.0149	0.0035	4.3060	0.0000262
Delphinidin-3-glu	-1.1976	0.4493	-2.6657	0.0083214
Caftaric acid	0.0253	0.0091	2.7770	0.0060157
Gallic acid	-0.0340	0.0118	-2.8803	0.0044125
Querc-3-Glucur	0.1629	0.1134	1.4365	0.1524520

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a general reference scheme provided at the beginning. Chemical analyses consisted of the determination of standard wine parameters, colorimetric analyses (including total polyphenols and tannin quantification) and single polyphenols using LC-MS apparatus. Finally, a putative correlation between scores and results from chemical analyses was investigated.

Most of the Pinot noir wines were from Trentino-Alto Adige, followed by Piemonte, Lombardia, Veneto and Friuli Venezia-Giulia in that order. A total of 25 wines (of which 24 from Trentino-Alto Adige) were examined in all three consecutive vintages and showed only very slight differences over the years, revealing that most producers had a conservative attitude to winemaking. Nevertheless, the most representative standard chemical parameters (like total phenolic content, total anthocyanins, total proanthocyanidins and tannins, alcohol, residue sugars, etc.) showed a high variability among these wines, which was even higher in the whole batches. It was thus demonstrated that both agronomical and winemaking processes (including eventual blending, wood treatments and aging) have strong effects on the chemical composition of single Pinot noir red wines originating from both the same terroir and different parts of Italy. In terms of the whole Italian territory, the average overall quality scores resulting from the sensory evaluation showed limited variations in the three vintages considered, but wines from Trentino-Alto Adige always obtained higher scores than those from the rest of Italy. This therefore suggests a higher affinity for the pedoclimatic features of this region and a consequent expression of typical organoleptic characteristics. Compared to monovarietal Pinot noir wines described in literature, average values obtained in this investigation were higher for standard wine parameters like TPC, total titrable acidity and alcohol content. Meanwhile, single phenolic constituents showed contents as being consistent with previously published data on Pinot noir red wines from the same temperate climatic area, except for *t*-resveratrol, for which we obtained higher values in our analyses, and delphinidin-3-glucoside, for which we obtained lower values. With respect to vintages, a significant variability of most parameters, including scores, was also observed. Moreover, the correlation between the overall quality evaluation scores and all the determined single chemical parameters was investigated: alcohol content, total anthocyanins from the colorimetric assay, single anthocyanins from the LC-MS analysis, caftaric acid and glycerol content had the strongest positive correlation, while gallic acid content, colour tonality and total phenolic content had the strongest negative correlation with sensory evaluation scores. Finally, a multiple linear regression model was applied to select the most informative set of parameters for the description of the wine score, where total anthocyanins, ethanol, reducing sugars, total acidity, catechins/procyanidins total, caftaric acid and quercetin-3-glucuronide have a positive contribution; while total phenolic content, methanol, color intensity, color tonality, delphinidin-3-glucoside and gallic acid have a negative contribution to overall quality wine score.

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