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Markers of sensory dynamics in phenols-rich virgin olive oils under optimal storage conditions

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ABSTRACT

Early changes in sensory quality of phenols-rich virgin olive oil (VOO) and their relationship with the chemical changes are less studied in the literature. Therefore, the objective of this study was to propose a predictive model of dynamics of sensory changes based on specific chemical markers. The evolution of the sensory quality of phenol-rich VOOs from Tuscan cultivars stored under optimal storage conditions (i.e., absence of light, no O_2 exposure, low temperature) was investigated using a multi-step methodological approach combining sensory (official sensory analysis (so-called Panel Test), Descriptive Analysis and Temporal Dominance of Sensation) and chemical measurements. The sensory map from descriptive data was related to the phenolic and volatile profiles, measured using HPLC-DAD and HS-SPME-GC–MS, respectively. A predictive model of the sensory changes over storage based on chemical compounds was developed. Results showed that very early changes involving phenolic and volatile compounds profiles occur in VOOs stored under optimal storage conditions, which turn in changes in sensory properties evaluated by the official panel test, the descriptive analysis and the temporal dominance of sensation. Furthermore, a chemical marker of sensory dynamics of oils during storage was identified as the ratio between two groups of secoiridoids. The proposed model, supported by the mentioned chemical marker, has the potential of improving the control of sensory changes in phenols-rich virgin olive oils during storage in optimal conditions.

1. Introduction

Virgin olive oils (VOO) are obtained from the fruit of the olive tree solely by mechanical or other physical means under conditions that do not lead to alterations in the oil (Reg EU 1308/2013). The European legislation establishes that the virgin olive oil commercial classification is based on a series of chemical parameters in addition to sensory attributes, which are measured by the so-called Panel Test. According to this classification, extra virgin olive oil (EVOO) is the premium category of virgin olive oil. Only those oils that have the chemical parameters within the established limits, the median of sensory defects equal to 0 and the median of fruity greater than 0 are classified as EVOO (Reg EU 2104/2022).

The above legal classification does not consider the nutraceutical properties and the sensory profile that make the EVOO one of the most successful edible oils around the world. Nutraceutical properties are due to the presence of antioxidant molecules such as tocopherols and hydrophilic phenolic compounds typical of *Olea europaea* L. (Cecchi et al., 2017; López-Biedma et al., 2016). The European Food Safety Agency approved a health claim for EVOO phenolic compounds (EFSA, 2011) based on their proven health-promoting effects (Covas et al., 2006; De La Torre-Carbot et al., 2010).

The wide variety of EVOO sensory profiles (e.g., intensity and quality of sensory attributes) represents a valuable product specification due to its connection with olive biodiversity and producer ability (Monteleone, 2014). Sensory profile is related to both volatile organic compounds (VOCs) and hydrophilic phenolic compounds (Andrewes et al., 2003; Cecchi et al., 2022; Servili et al., 2004). Changes in chemical, nutraceutical and sensory characteristics of EVOOs can occur during storage mainly due to oxidative processes induced by light and oxygen exposure (Castillo-Luna et al., 2021; Cecchi et al., 2019; Krichene et al., 2015; Miho et al., 2020). These detrimental oxidative phenomena affect triglycerides, volatile and phenolic compounds, leading to changes in the sensory profile (i.e., decay of fresh notes and onset of defects) and

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limiting the EVOO shelf-life (Castillo-Luna et al., 2021; Cecchi et al., 2019; COI, 2018aCOI/BPS/Doc. No 1, 2018a; Zanoni, 2014), which is often downgraded to the virgin olive oil (VOO) category (COI, 2018aCOI/BPS/Doc. No 1, 2018a). Both oil chemical characteristics and storage conditions can affect the VOO stability. High hydrophilic phenolic compound contents are positively associated with VOO stability, slowing down the onset of sensory defects and the decay of the fresh notes (Averbuch et al., 2023; Esposto et al., 2020). Avoiding light and O_2 exposure, and a temperature range between 13–25 °C have been defined as optimal storage conditions (COI, 2018aCOI/BPS/Doc. No 1, 2018a). However, the variation of nutraceutical and sensory properties can occur even when the legal quality markers do not significantly change also in oils with high phenolic content and stored in optimal conditions (Cecchi et al., 2019; Klisović et al., 2022).

Early dynamics of sensory properties of phenols-rich VOOs stored in optimal conditions are difficult to depict. The International Olive Council (IOC) standard method for the sensory evaluation of oils (i.e., the Panel Test), mandatory in the European Union, represents an effective method to classify oils as extra virgin, virgin or lampante olive oil. This approach employs trained assessors who utilise specific descriptors and standardised protocols for evaluating the sensory qualities of olive oils. However, this method is not sufficient to describe the sensory properties of extra virgin olive oils (Monteleone, 2014).

Beyond the Panel Test (COI, 2018b(COI/T.20/Doc. No.15/Rev. 10, 2018b)), Descriptive Analysis (DA, Lawless & Heymann, 2010) and Temporal Dominance of Sensations (Pineau et al., 2009) were successfully applied to describe the sensory profile of VOOs. Descriptive Analysis offers a comprehensive static profile of sensory attributes, while TDS provides a dynamic view of how these attributes evolve during tasting. These techniques enable a deeper understanding of the sensory properties of VOOs evaluated both in isolation and in food combinations (Dinnella et al., 2012). However, multiple sensory evaluations of VOO during storage are inherently difficult to plan, conduct and validate, particularly for routine analysis in testing labs in which a great number of samples is daily analysed.

The topic of VOO storability is very current as demonstrated by both the recent document published by the IOC (COI, 2018a(COI/BPS/Doc. No 1, 2018a)) concerning "Best practice guidelines for the storage of olive oils and olive–pomace oils" and the large number of literature manuscripts recently published. However, manuscripts focusing on early changes of phenols-rich VOO stored in optimal storage conditions (i.e., absence of light, no O_2 exposure, low temperature), and including several types of chemical analysis and both static and dynamic sensory analyses are still missing.

In this context, some questions are still unanswered: how long do the sensory properties of VOOs rich in phenolic compounds (e.g., higher than 400 mg/kg) and stored in optimal conditions remain stable? Which are the chemical markers of sensory dynamics in phenols-rich VOOs under optimal storage conditions?

To answer the above research questions, the main objective of this study was to propose a predictive model of dynamics of sensory changes based on specific markers beyond the legal quality parameters. To this aim, the following multi-step methodological approach was adopted working with experimental oils obtained at industrial scale: (i) measuring and mapping sensory similarities and dissimilarities among a set of samples representing fresh (within two weeks from production, T0) phenols-rich VOOs using the conventional Descriptive Analysis coupled with Principal Component Analysis (PCA); (ii) studying the relationships between the obtained sensory map and the chemical composition of oils (i.e., phenolic compounds and VOCs) by means of a Principal Component Regression (PCR) and selecting the chemical variables correlated to the first two dimensions of the sensory space; (iii) developing a classification model of the T0 oils based on the selected chemical parameters to map modifications of oils during the storage (T1, three months-T4, 12 months) in optimal conditions; (iv) studying the relationship between the distance of the stored samples from the centre of the classification model and their sensory properties measured using IOC, DA and TDS methods: this will allow identifying the chemical markers of sensory dynamics in oils under optimal storage conditions.

2. Material and methods

2.1. Samples and experimental plan

The experimental procedure was designed to address variability arising from the factors olive cultivar (2 levels), harvesting date (2 levels), and extraction conditions (3 levels). In total, twelve monovarietal oils were produced at industrial scale in an oil mill (Società Agricola Buonamici, Fiesole, Florence, Italy), from the two main olive cultivars in the area: six oils were obtained from the Frantoio cultivar, and six from the Moraiolo cultivar (coded as F and M, respectively).

For each cultivar, six different samples were produced with olives from two harvesting dates very close to the optimal ripening stage and from three different production trials selecting the processing conditions typically adopted in the productive area: three oils were produced in the second week of November (coded as 1, followed by "a", "b" and "c" for indicating the different production trials on the same date), and three oils were produced in the third week of November (coded as 2, followed by "a", "b" and "c" for indicating the different production trials on the same date). Virgin olive oils were extracted from 300 kg of olive fruits for each trial by processing the olives in the same day of harvesting using a plant (Mori TEM, Tavarnelle Val di Pesa, Florence, Italy) consisting of: i) a cleaning system in turn consisting of a debrancher for removing twigs, stones and other solid residues following by a defoliator, a washer and a drier; ii) a knife crusher; iii) a vertical malaxator working with reduced oxygen content thank to a slight vacuum; iv) a two-phase decanter with a capacity of 1.5 tons and capable of working with no water addition; v) a filtration system consisting of 6 stainless steel cartridges as a pre-filter and a filter with cellulose cardboards. All the processing conditions were carefully selected to limit as much as possible any chemical alterations of the oils. The selected oils were then stored in filled 0.25-L glass bottles, at a temperature ranging from 14 to 16 °C and in the darkness for twelve months.

Chemical and sensory analysis were carried out within two weeks after production (T0), and after 3 (T1), 6 (T2), 9 (T3) and 12 months (T4). All analyses were performed in triplicate, using three different bottles for each sample.

2.2. Legal quality parameters

The chemical legal quality characteristics of the oils such as free acidity, peroxide value ad UV spectrophotometric indices (i.e., K_{232}, K_{270}, ΔK) were determined following official analytical methods (Reg EU 2104/2022).

2.3. Analysis of phenolic compounds

Analysis of phenolic compounds was performed according to the official method established by the International Olive Council (COI, 2022(COI/T.20/Doc. No.29/Rev. 2, 2022)). Briefly, 2 g of oil sample were extracted with 6 mL of MeOH:H_2O 80:20 solution including 5 mL of pure extractive solution and 1 mL of Internal Standard solution (i.e., syringic acid 0.015 mg/mL in MeOH:H_2O 80:20). The obtained phenolic extracts were analysed using a HP 1200 liquid chromatography coupled with a Diode Array Detector (Agilent Technology, California, USA). Molecules were separated in a LiChrospher 100 endcapped RP-18 column (5 μm, 250 mm × 4.6 mm id) after injecting 20 μL of extract. Solvents (H_2O (pH 2.0 with H_3PO_4), MeOH and Acetonitrile) were eluted following the gradient described in the method, and chromatograms were acquired at 280 nm. Both total phenolic compounds and each single phenolic compound content were determined following the Internal Standard method, using syringic acid as the internal standard

and tyrosol as the reference for the response factor, therefore expressing the results as mg_{tyr}/Kg_{oil}.

2.4. HS-SPME-GC–MS analysis of volatile organic compounds

The Volatile Organic Compounds (VOCs) linked to both sensory defects and positive attributes (i.e., the VOCs derived from the lipoxygenase pathway, LOX) according to the literature (Cecchi et al., 2021) were analysed in agreement with a method previously described and based on Head Space-Solid Phase Micro Extraction for VOCs preconcentration, Gas Chromatography for their separation and Mass Spectrometry for their detection (i.e., HS-SPME-GC–MS technique) (Guerrini et al., 2020; Vichi et al., 2003). Briefly, 4.3 g of oil samples and 0.1 g of internal standard solution (i.e., 4-methyl-2-pentanol 75 mg/kg in refined olive oil free from interfering VOCs) were weighted into a 20-mL screw cap vial. After sample equilibration (5 min), VOCs were extracted onto a 1-cm SPME fibre (50/30 μm, DVB/CAR/PDMS, Supelco) for 40 min at 40 °C, and then desorbed for 2 min at 260 °C in the injection port of the gas-chromatograph (splitless mode, 260 °C), which was a Trace GC coupled with a Trace DSQ Thermo Finnigan instrument (Thermo Fisher Scientific, Illkirch, France). VOCs were then separated in a ZB-FFAP capillary column (30 m × 0.25 mm, 0.25 μm DF, Zebron). The column temperature stayed at 36 °C for the first 10 min, then increased up to 156 °C at 4 °C/min, and then up to 220 °C at 10 °C/min, with a final stay at 220 °C for 1 min. Ion source and transfer line temperature were both 250 °C. Mass detector acquired in scan mode in the range 30——330 Th, applying an IE energy of 70 eV.

For quantitative analysis, a calibration curve was built for each VOC to be quantitated using the commercial standard of that molecule and normalising areas by mean of the internal standard (i.e., 4-methyl-2-pentanol), which was added in the same amount in all the vials with samples and external standard solutions. Results were expressed as mg_{VOC}/kg_{oil}.

2.5. Sensory evaluation

Sensory analysis was carried out following three different approaches: 1) the official method proposed by the International Olive Council (i.e., the IOC **Panel Test**); 2) the Descriptive Analysis (**DA**); 3) the Temporal Dominance of Sensations (**TDS**).

– Official IOC method: sensory analysis according to the official IOC method was performed by a professional panel acknowledged by the Italian Ministry of Agricultural Policies (MASAF): briefly, the panel consisted of a panel leader and a minimum of 8 trained assessors. Samples were presented to assessors in blue glass glasses (15 mL). Each sample was smelt and tasted by the assessors, which marked the intensity of defects (i.e., *rancid*, *fusty/muddy sediment*, *musty/humid/earthy*, *winey/vinegary*, *metallic*, other) and positive attributes (i.e., *ripe fruity*, *green fruity*, *bitter*, *pungency*) on a 10-cm unstructured line scale (COI, 2018b(COI/T.20/Doc. No.15/Rev. 10, 2018b)). Data were acquired on paper profile sheets. The median values were considered for virgin olive oil commercial classification purpose according to the EU regulation. For each storage time a mean value of the median of the oils was computed for statistical analyses.

– Descriptive Analysis, DA (Lawless & Heymann, 2010): thirteen participants(5 men, mean age 30), very familiar with extra virgin olive oil were recruited at the Sensory Laboratory of the University of Florence. The term generation sessions ended with a consensus list of twenty sensory descriptors in the categories of aroma (*green olive*, *ripe olive*, *artichoke*, *grassy*, *tomato leaf*, *almond*), flavour (*green olive*, *ripe olive*, *artichoke*, *grassy*, *tomato leaf*, *almond*, *green fruity*, *rancid*, *bitter*), and chemesthetic/tactile sensations (*peppery*, *pungency*, *viscous*, *astringent*). To train the assessors in evaluating the descriptors, standards were prepared in accordance with Monteleone et al. (2014). All standards were prepared to induce a moderate intensity, corresponding to the central point of the nine-point scale. Assessors participated in three evaluation sessions at each storage time (T0-T4). Six samples were evaluated per session presented in two subsets of three samples each. Oils were evaluated in triplicate. Samples were presented in blue glass glasses (15 mL) with three-digit codes. The presentation's order of samples was balanced to control for first order and carry over effects. Assessors were asked to smell the sample to evaluate its aroma, then to taste the sample in the mouth to evaluate its *viscosity* and, after 8 s, *bitterness* and *astringency*; after further 12 s, assessors were asked to evaluate *pungency* (described as the sensation perceived mainly in the throat and typical of oils obtained from green olives, especially during the early stages of the harvest) and *peppery* sensation. There was a 15-min break between subsets. After each sample, participants rinsed their mouths with distilled water for 30 s, had some plain crackers for 30 s and finally rinsed their mouths with water for a further 30 s. Intensity of each sensation was evaluated on a 9-point scale anchored from "extremely weak/absent" to "extremely strong". Data were expressed as mean values.

– Temporal Dominance of Sensation, TDS (Pineau et al., 2009). Twelve participants (two women, mean age 29 y.o.) were trained and took part in the dynamic evaluations of oils during storage. Assessors participated in two sessions for generating a list of attributes describing the dominant sensations in oils. In the first session, the concept of dominance was explained to the assessors as "the attribute associated with the sensation catching the attention of the assessors at a given time, not necessarily being the one with the highest intensity" (Bruzzone et al., 2013). For term generation, assessors were asked to indicate the dominant sensations during oil tasting and describe their temporal evolution. After a common discussion, the consensus was reached on a list of eight attributes (*ripe olive*, *green olive*, *grassy*, *artichoke*, *astringency*, *pungency*, *rancid* and *bitterness*). Two sessions were run to train assessors to the use of the computer system for TDS data acquisition. Assessors were trained to click on the "Start'' button as soon as the sample was in their mouth and to immediately start the selection of dominant attributes. They were also told that not all the attributes have necessarily to be selected as dominant and, at the same time, that a given attribute can be selected as dominant several times during the evaluation. At each storage time (T0-T4), four evaluation sessions were run. In each session the six samples were evaluated. This means that a total of 48 evaluation for each oil were run (i.e., 12 assessors × four replicates; one replicate for each session). Presentation order and rinsing procedure were the same as described for DA evaluation. The order of attributes was randomised between participants but was always the same for a given assessor. Samples (3.5 ml) were presented in a test tube identified by a three-digit code. Assessors were instructed to pour the whole test tube content in a spoon, hold the sample in their mouth and immediately start the evaluation. After 8 s, assessors were prompted by a screen signal to swallow the samples and to continue the evaluation. The total duration of each evaluation of each sample was 60 s.

DA and TDS evaluations were performed under red light and FIZZ System software (version 2.47B, Biosystèmes, Courtenon, France) was used for data acquisition.

2.6. Ethics & standards requirements

The study was conducted according to the principles established in the Declaration of Helsinki for medical research involving humans and was subject to ethical standards that promoted and ensured respect for all human participants and protected their health and rights. In line with national regulations, given that the research was not medical, the research protocol was not submitted for approval to an ethical committee. The researchers involved in the study followed the code of Ethics & Standards for Sensory Project Managers developed by the Italian Sensory Science Society. Written informed consent was obtained from all participants according to the GDPR (General Data Protection Regulation) 2016/679. Participants were able to withdraw from the study at any time without giving a reason. The products tested were safe for

consumption. Participants received a gift card payment to motivate their participation in the study.

2.7. Statistical analysis

TDS data.

Panel performance and differences between products for attribute dominance were assessed following the procedure proposed by Dinnella et al., (2013), after a visual inspection of the curves, the first 60 s of evaluation were split into two intervals (0–15 s and 16–60 s). For each time period, the binomial data (0 = attribute not selected as dominant; 1 = attribute selected as dominant) were summarised and the frequency values of attribute dominance by each subject were computed. Frequency values were then submitted to a mixed model ANOVA with assessor, product, and interaction effects. The assessor effect and its interaction with the product were considered as random factors. The ANOVAs were performed using the XLSTAT software version 2018.1 (Addinsoft, Long Island, NY, USA).

Perceptual maps.

The differences among the samples from the Descriptive Analysis were studied by means of a Principal Component Analysis (PCA) computed on the panel averages of each significant attribute ($p < 0.05$) arising from Two Ways ANOVA model (sample and assessors, with assessors as random effect) on intensity data. Samples were included as dummy variables (down-weighted in the data matrix) to improve the visual interpretation (Martens & Martens, 2001). The full cross validation was computed to validate the interpretation of the first two components.

To investigate the relationship between sensory and chemical data, a Principal Component Regression (PCR) model was computed. For this purpose, panel averages of each sensory descriptor discriminating significantly among samples at T0 were used as the X matrix, while phenolic and VOCs as the Y matrix. The full cross validation was computed to validate the interpretation of the first two components.

Classification model.

A classification method was computed to explore variations in the phenolic and volatile compounds selected from the PCR described above and measured in the experimental oils at varied storage time adopting the SIMCA (Soft Independent Modeling of Class Analogy) approach. SIMCA is based on making a PCA model on phenolic compounds and VOCs of experimental oils at T0 (training set − class model). The distance from the centre of the class model (dM) of T1, T2, T3 and T4 oil samples were then assumed as an index of the chemical modification of the oils. A One Way ANOVA (factor: storage time, T0 −T4) was run on dM values of the six selected oil samples.

The relationship between dM and sensory (IOC, DA and TDS) and chemical compounds were investigated by computing Pearson correlation coefficients.

Sensory and chemical marker map.

A final PCA was computed on sensory data from the IOC, DA and TDS sensory methods and identified phenolic markers measured in all oil samples at all storage times. Data were standardised and a full cross validation was computed to validate the interpretation of the first two components.

All multivariate analyses were carried out using the software package The Unscrambler V.11 (Camo Analytics, Norway).

3. Results

The values of the analyzed legal chemical parameters of the oils were within the range of EVOOs, as reported in the sup.Table S1. In particular, free acidity was in the range 0.19–0.27 %, peroxide value was in the range 3.6–6.2 meq_{O2}/kg_{oil}, K_{232} was in the range 1.63–1.87, and K_{270} was in the range 0.15–0.17. All these values were much lower than the legal ones (Reg EU 2104/2022). Concerning the sensory analysis by the Panel Test, the samples showed medians of *pungency* (range 4.2–5.8) greater than that of *bitterness* (range 3.5–4.6), with median of *fruity* ranging 3.7–4.4. The assessors also perceived slight *rancidity* in 4 out of the 12 samples, with a mean of the medians of 0.53; the very slight intensity of *rancidity* at time 0 was also confirmed by data from DA, and they depict cases already reported in the specialised bibliography (Aparicio-Ruiz et al., 2019): oils with defects at a "very slight intensity" are difficult to be qualified with certainty as EVOOs or VOOs. Considering the compliance of chemical parameters with EVOO classification, the high content of phenolic compounds and tocopherols, the *rancid* median values from IOC panel very close to 0 and the absence of an increasing trend of the median values with the storage time, the experimental oils were assumed to be border-line samples that, according to the EU legislation, are classified within the VOO category.

Concerning phenolic compounds, the oils showed high contents ranging 446–733 mg/kg, which means 2- to 3-fold greater than the minimum amount requested by the EFSA health claim (EFSA, 2011); on average, the values were greater for the Moraiolo cultivar (i.e., 687 mg/kg) than for the Frantoio cultivar (i.e., 612 mg/kg). Furthermore, all the oils showed very low levels of free tyrosol (1–2 mg/kg) and hydroxytyrosol (0–2 mg/kg) indicating a phenolic profile typical of fresh oils at low hydrolytic level (Brenes et al., 2001; Breschi et al., 2022; Mulinacci et al., 2013). Finally, tocopherols varied in the range 249–350 mg/kg, with values greater for the Frantoio cultivar (i.e., average 293 mg/kg) than for the Moraiolo cultivar (i.e., average 258 mg/kg).

A descriptive sensory analysis was then performed, and six oils were selected among the 12 experimental samples to represent the whole range of similarities and differences, as following explained in detail. Details of samples' sensory properties are provided as supplementary materials (Tables S2a-c).

3.1. Step 1: Measuring and mapping sensory similarities and dissimilarities among a set of samples representing fresh (within two weeks from production, T0) phenols-rich VOOs using the conventional descriptive analysis coupled with Principal component analysis (PCA)

A perceptual map describing sensory differences among the twelve oils at T0 was derived from a PCA computed on the mean intensities from DA. The attributes *almond* (odour by mouth) and *ripe fruitiness* were not included in the PCA model since only significant attributes resulting from the Two-Ways ANOVA models computed for each descriptor were considered (Table S2 a-c). The variables *almond* (odour by nose), *ripe olive* (perceived both by nose and by mouth) were not included too, since their mean intensities across samples varied always between 1 ("extremely weak") and 2 ("very weak") without reaching the latter value. Thus, a first PCA model was computed on fifteen out of twenty sensory descriptors. To maximise the explained variance of the model after two PCs', the attributes weakly related to the first two components (*artichoke* by nose, *viscosity, astringency, peppery*) were excluded from the computation of the final model (Fig. 1). The first component of the PCA model explains the 44 % of systematic variability among samples and separates oils according to the olive variety. "Moraiolo" samples sitting at the left of the map are described as more intense in all "green" flavour notes such as *grassy*, *green olive* and *tomato leaf* in opposition to the "Frantoio" oils falling at the right of the map. Samples' differences along the second component (23 % of explained variance) are mainly due to their *artichoke* flavour, which decreases when moving from the bottom to the top of the map in opposition to *bitterness*. Although in general *rancid* mean intensities from DA were low, systematic small variations of this attribute in the oils are positively correlated to both components. The attribute *pungency* contributed less to discriminating samples.

A subset of samples that effectively encompassed the entire spectrum of sensory differences among the oils was selected to study sensory changes of oils under optimal storage conditions. Samples were selected following a visual inspection based on the two principal components. Samples M1c and F2a were selected to represent the main variations observed along the first component. Additionally, samples F1c and M2c

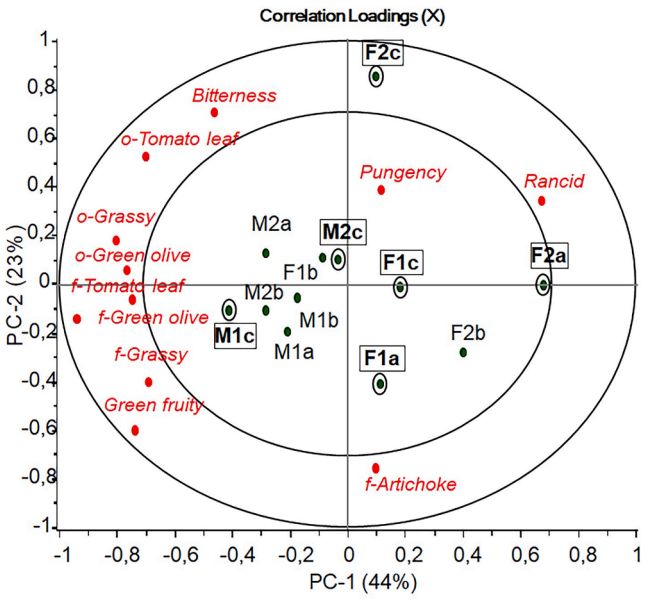

Fig. 1. PCA on sensory descriptive data of fresh oils (T0): score and correlation loading plot. Sample codes: F = Frantoio, M = Moraiolo: 1–2 harvest time, a-c = production trials. In italics sensory attributes. Samples were included as dummy variables. Outer and inner circles on the map represent 100 % and 50 % explained variance, respectively. The bold font and the circled points represent the selected samples.

were chosen to consider the whole variability of sensory properties of oils along that component. Finally, samples F1a and F2c were selected to represent the primary variation observed in the second principal component. As a result, the following samples were retained for testing under ideal storage conditions: M1c, M2c, F1a, F1c, F2a, and F2c.

3.2. Step 2: Studying the relationships between the obtained sensory map and the chemical composition of oils (i.e., phenolic compounds and VOCs) by means of a Principal component Regression (PCR) and selecting the chemical variables correlated to the first two dimensions of the sensory space

Table S3 reports the phenolic and volatile compounds measured at T0 on the twelve experimental oils. Two independent PCRs, one for phenolic compounds and one for VOCs, were computed. PCRs allowed the selection of seven phenolic compounds and thirteen VOCs correlated with the largest sensory differences among fresh oils (T0). The correlation loading plot in Fig. 2 shows that the first dimension of the sensory space is positively correlated to oxidised aldehydic and hydroxylic form of ligstroside aglycone (*p*-HPEA-EAOHox), di-aldehydic form of ligstroside aglycone (*p*-HPEA-EA), oleuropein aglycone (3,4-DHPEA-EAagl), oxidised di-aldehydic form of decarboxymethyloleuropein aglycone (3,4-DHPEA-EDAox). These compounds are correlated to all the "green" descriptors of the flavour of the oils falling on the right of the map. On the contrary, the di-aldehydic form of decarboxymethylligstroside aglycone (*p*-HPEA-EDA), aldehydic and hydroxylic form of oleuropein aglycone (3,4-DHPEA-EAOH), and aldehydic and hydroxylic form of ligstroside aglycone (*p*-HPEA-EAOH) are negatively correlated to the first dimension. They fall in the area of the map where the oils with less intense "green" descriptors and a higher *rancidity* are located. Similarly, Fig. 3 shows the relationship of the first dimension of the sensory map from the descriptive analysis and the VOCs. In particular, (*Z*)-3-hexenyl acetate, (*Z*)-3-hexenol, (*Z*)-2-pentenol, (*E*)-2-pentenal, heptanal, octanal, and phenol are positively correlated to the "green" descriptors at the right of the map, in contrast to the VOCs ethyl acetate, 3-methylbutanal, 2-methylbutanal, 2- and 3-methylbutanol, propanol and 2-octanone, which are located on the left.

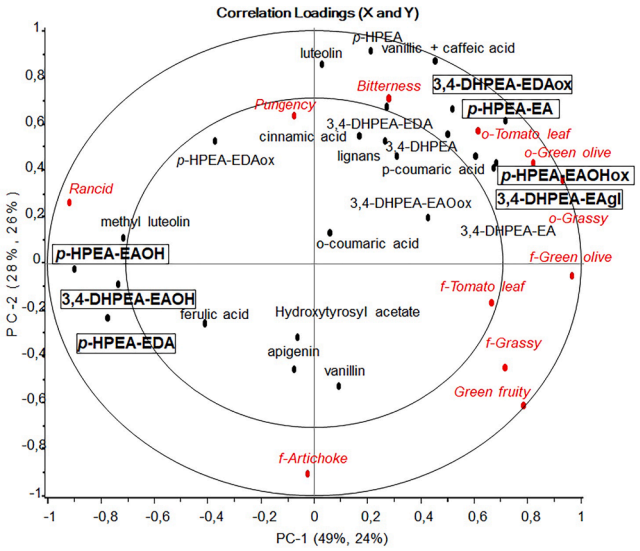

Fig. 2. Principal Component Regression (PCR): Sensory Descriptive data (X matrix) *vs* Phenolic compound data (Y matrix). Score and correlation loading plot. Sample codes: F = Frantoio, M = Moraiolo: 1–2 harvest time, a-c = production trials. In italics sensory attributes. Outer and inner circles on the map represent 100 % and 50 % explained variance respectively. In bold variables strongly correlated to the first dimension of the map.

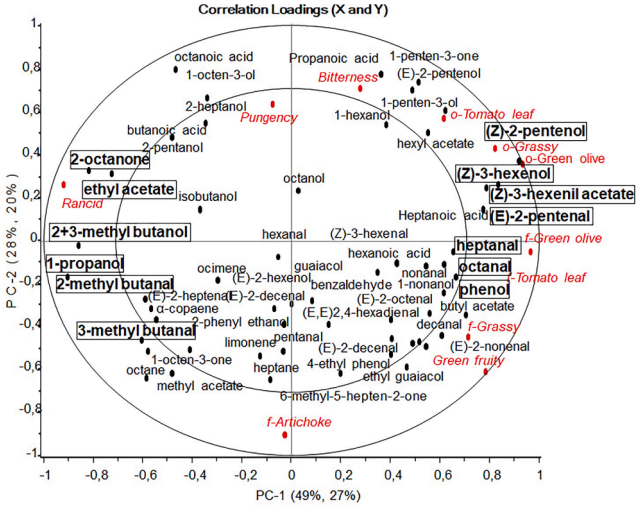

Fig. 3. Principal Component Regression (PCR): Sensory descriptive data (X matrix) *vs* Volatile compound data (Y matrix). Score and correlation loading plot. Sample codes: F = Frantoio, M = Moraiolo: 1–2 harvest time, a-c = production trials. In italics sensory attributes. Outer and inner circles on the map represent 100 % and 50 % explained variance respectively. In bold variables strongly correlated to the first dimension of the map.

3.3. Step 3: Developing a classification model of the T0 oils based on the selected chemical parameters to map modifications of oils during the storage (T1, three months-T4, 12 months) in optimal conditions

Both phenolic and volatile compounds measured on T0 samples and selected in the previous step 2 were used to build up a PCA classification model. The same variables were also measured over storage (T1-T4) in the selected samples (step 1). Results are illustrated in Fig. 4. The model oils (T0) are located bottom left of the figure. Changes in the amount of phenolic and volatile compounds during storage determine an increase of the distance (dM) from the centre of the PCA classification model at T0. It can be noticed that none of the samples at T1 is classified within

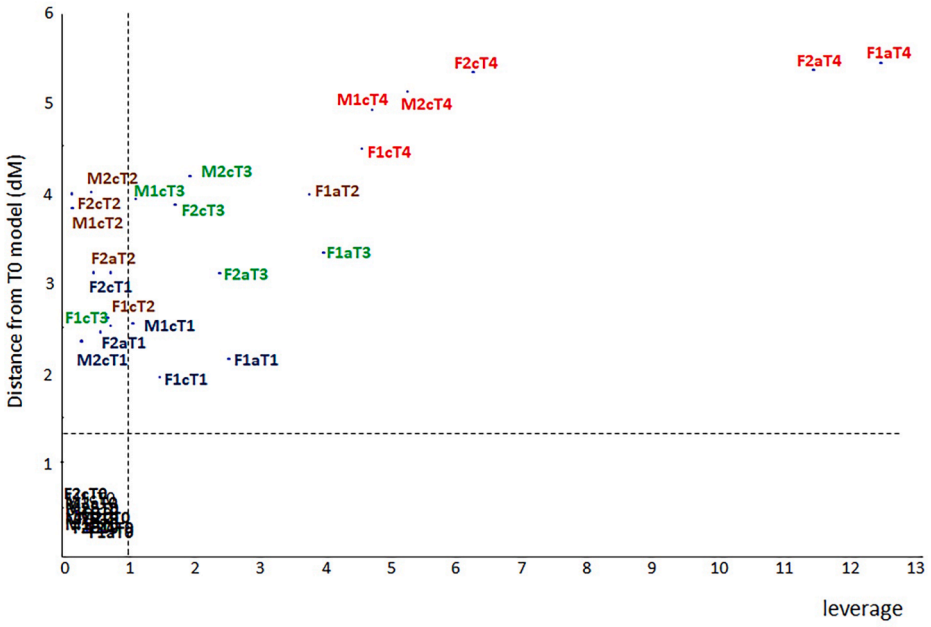

Fig. 4. PCA classification model (SIMCA): distance from the centre of the model (dM) and leverage values. Training samples included twelve samples at T0. Test samples included 6 oils for each storage time. Sample codes: F = Frantoio, M = Moraiolo: 1–2 harvest time, a-c = production trials. T0-T4 storage time.

the original (T0) model and that, when the storage time increases from T1 to T4, dM increases with it. To better show the effect of the storage time under optimal condition, a One-Way ANOVA was run on dM values of the six selected oil samples at all storage times (T0 −T4). Significant variations were observed in dM values during storage time ($F_{4,25} = 82.5$; $p < 0.0001$). Mean dM values steeply increased passing from T0 (dM = 0.44) to T2 (dM = 3.52). A further steep increase of the values was evident passing from T3 (dM = 3.4) to T4 (dM = 5).

In short, changes in both phenolic and volatile compounds under optimal storage conditions start very early and their entity increases with time.

The results of the correlation between sample dM and the concentration of each chemical compound are reported in Table 1. Most of the phenolic compounds were significantly correlated to dM either positively or negatively. The sum of *p*-HPEA-EAOHox, *p*-HPEA-EA, 3,4-DHPEA-EAagl and 3,4-DHPEA-EDAox, which were positively correlated to the first dimension of PCR in Fig. 2, significantly decreased during storage (i.e., F PC + 1 = -0.95), and then they showed a relevant decrease of oil freshness. Instead, the sum of *p*-HPEA-EDA, 3,4-DHPEA-EAOH and *p*-HPEA-EAOH, which were negatively correlated to the first dimension of PCR in Fig. 2, significantly increased during storage (i.e., F PC −1 = 0.95). Storage changes of VOCs correlated both positively and negatively to the first dimension of PCR in Fig. 3 were not significant (i.e., HS PC + 1 = 0.57; HS PC-1 = -0,87).

Table 1
Correlation between the selected chemical variables and the distance from the centre of the model (dM).

Phenolic compounds	r	Volatile compounds	r
p-HPEA-EDA	−0.96**	ethyl acetate	−0.79
3,4-DHPEA-EAOH	0.92*	2-methyl butanal	0.05
p-HPEA-EAOH	0.59	3-methyl butanal	−0.91*
3,4-DHPEA-EDAox	−0.97**	1-propanol	−0.79
3,4-DHPEA-EAagl	−0.65	(*E*)-2-pentenal	0.78
p-HPEA-EA	−0.95**	heptanal	0.95**
p-HPEA-EAOHox	0.19	2 + 3-methyl butanol	−0.77
		2-octanone	0.23
F PC −1	0.95**	octanal	0.10
F PC + 1	−0.95**	(*Z*)-3-hexenyl acetate	0.32
		(*Z*)-2-pentenol	0.16
		(*Z*)-3-hexenol	0.34
		phenol	0.00
			−0.87
		HS PC −1	
		HS PC + 1	0.57

* significance ≥ 90 %; ** significance ≥ 95 %.
F PC −1 indicates the sum of phenols correlated to the 1st negative component.
F PC + 1 indicates the sum of phenols correlated to the 1st positive component.
HS PC −1 indicates the sum of volatile compounds correlated to the 1st negative component.
HS PC + 1 indicates the sum of volatile compounds correlated to the 1st positive component.

3.4. Step 4: Studying the relationship between the distance of the stored samples from the centre of the classification model and their sensory properties measured using IOC, DA and TDS methods: This will allow identifying the chemical markers of sensory dynamics in oils under optimal storage conditions

Table 2 reports data across samples of sensory measurements from IOC, DA and TDS methods from T0 to T4. Significant storage time effects were found for most of the IOC attributes. The intensities of *green fruitiness* started diminishing from T2 to reach the minimum at T4. On the contrary, the intensity of *bitterness* increased from T0 to T2 and decreased from T3 to T4. A similar trend was observed for *pungency*, while no significant changes were reported for *ripe fruitiness* (the value at T1 appears to be an outlier) and *rancid*. Concerning *rancidity*, it is worth noting that the mean of the medians at t0 is 0.52. This value even decreased till t2 to a mean value of 0.19. This trend is certainly due to a very slight intensity in some samples that are then difficult to classify with certainty as defective or not as reported above. Data from the DA method depict a similar trend for the "green" notes (i.e., *green odour* and *green fruity*), whose intensities tended to diminish reaching the minimum values in T4. No significant changes were observed for *bitterness* and *pungency* while the attribute *rancid* significantly increased from T3.

Finally, the influence of storage time based on data from the TDS method was investigated. In fresh oils evaluated at T0, the odours by mouth dominated the dynamic profile of the flavour of oils, whereas, during the second evaluation period (16–60 s), *bitterness*, *pungency* and *astringency* characterised the dominance curve of the oils (e.g., sample MC2, Fig. 5a). However, during storage, the dominance profile of oils

Table 2
Mean intensity (IOC panel test and DA) and dominance frequency (TDS) values of the sensory variables evaluated over storage (T0-T4), *F* and *p* values (in bold when < 0.05) from one-way ANOVA. On each row, different letters indicate significant differences among values for $p \leq 0.05$ (Fisher LSD post hoc test*). In the last column, the correlation between the sensory variables and the distance from the center of the model (dM) is also reported.

Method	Attribute	T0		T1		T2		T3		T4		F	p	r
IOC Panel test	Green fruity	3.39	a	2.95	a	1.78	b	2.15	ab	0.71	c	8.82	**≤ 0.001**	**−0.96****
	Ripe fruity	2.98	ab	0.49	c	3.33	a	2.37	b	2.53	ab	9.66	**≤ 0.001**	0.05
	Bitterness	3.38	b	3.92	ab	4.19	a	3.20	b	1.96	c	8.42	**≤ 0.001**	−0.48
	Pungency	4.77	a	4.70	a	5.29	a	4.87	a	2.66	b	9.41	**≤ 0.001**	−0.62
	Rancid	0.52		0.23		0.19		0.65		1.50		1.57	0.214	–
DA	Green odour	11.57	a	10.52	bc	10.97	ab	10.58	bc	10.00	c	3.19	**0.03**	−0.87
	Green fruity	5.07	a	4.31	b	5.28	a	4.19	b	3.82	c	16.82	**≤ 0.001**	−0.57
	Bitterness	4.44	b	5.28	a	5.06	a	5.01	a	4.90	ab	2.43	0.074	0.51
	Pungency	4.96		4.78		4.71		4.85		4.53		0.88	0.492	–
	Rancid	1.58	b	1.57	b	1.74	b	2.43	a	2.35	a	7.55	**≤ 0.001**	0.84
TDS 0–15 s	Green notes	0.43	a	0.42	a	0.39	a	0.32	b	0.24	c	11.80	**≤ 0.001**	−0.85
	Bitterness	0.11	c	0.15	c	0.20	b	0.20	b	0.25	a	9.05	**≤ 0.001**	**0.99*****
	Rancid	0.01	d	0.03	d	0.06	c	0.10	b	0.14	a	25.90	**≤ 0.001**	**0.92***
TDS 16–60 s	Bitterness	0.18	b	0.28	a	0.30	a	0.27	a	0.32	a	2.67	0.056	**0.95****
	Pungency	0.42		0.35		0.42		0.43		0.40		1.65	0.192	–
	Astringent	0.13	a	0.10	b	0.06	c	0.08	bc	0.10	b	8.01	**≤ 0.001**	−0.62
	Rancid	0.02	c	0.04	bc	0.03	bc	0.05	b	0.09	a	7.22	**≤ 0.001**	0.85

* Significance of mean comparisons for bitterness DA and bitterness 16–60 TDS were computed by Fisher LSD post hoc test with $p \leq 0.07$.
In the last column, * significance ≥ 90 %; ** significance ≥ 95 %; *** significance ≥ 99 % (in bold).
(1) The correlation was computed relating the sum of the mean intensities of "green" aroma descriptors green olive, tomato leaf and grassy to dM.
(2) The correlation was computed relating the sum of the dominance frequencies of "green" flavor descriptors green olive, tomato leaf, artichoke and grassy to dM.

dramatically changed (Fig. 5b). *Bitterness* dominated the dynamic profile of oils since the first evaluation period. Table 2 shows that during the first fifteen seconds of the dynamic evaluation of oils, the frequency of dominance selection of "green" flavour notes progressively decreased from T0 to T4. In contrast to this lowering, the frequency of dominance selection of *bitterness* increased with time storage. Thus, the *bitterness* dominated the flavour perception of oils during three-six months of storage in optimal conditions as a consequence of the lowering of intensities and dominance of "green" notes.

The relationship between sensory data from IOC, DA and TDS methodologies and the distance from the classification model of experimental oils during storage (T0-T4) was investigated. Results are reported in Table 2. "Green" descriptors from any method were always negatively correlated with dM. On the contrary, *rancid* from both DA and TDS description was positively correlated with dM, *bitterness* and *pungency* from IOC method were weakly and negatively correlated to dM. Coherently with outcomes of the TDS analysis, the dominances of *bitterness* (both at 0–15 and 16–60 s) in stored oils resulted positively correlated with the dM. In summary, main changes in sensory properties of oils during storage evaluated by IOC, DA and TDS were correlated with the distance from the classification model centre computed in step 3, thus confirming the goodness of the prediction model based on chemical parameters.

To obtain a comprehensive picture of the sensory changes of the experimental oils during optimal storage conditions, a final PCA was computed on IOC, DA and TDS data collected at all five storage times (T0-T4). To link the description of sensory dynamics during storage with the chemical composition of the samples, the sum of the amounts of phenolic compounds previously coded as PH-PC1 + and PH-PC1- were included in the model. In fact, these two groups of phenolic compounds were strongly and significantly correlated with the dMs of oils during storage. Results are summarised in the biplot of Figure S1. Along the first component, oils are separated and grouped according to their storage time with the T0, T1 and T2 samples mainly on the right of the map in opposition to samples T4 and T3 sitting on the left. The latter are the oils less intense in all "green" IOC and DA descriptors and more intense in the DA attribute *rancid.* The same sensation dominates their dynamic sensory properties evaluated by TDS. In opposition, the oils on the right have a more complex profile, rich in "green" notes evaluated by any methodology and are more intense in *pungency*. It can be noted that the *bitterness* dominance 0–15 TDS is strongly and negatively correlated to *green fruity* IOC, while *bitterness* dominance 16–60 TDS is positively correlated to *bitterness* DA. Finally, PH-PC1 + and PH-PC1- were strongly correlated with the first dimension, positively and negatively respectively. Along the second component, samples are separated in relation to their *bitterness* intensity and/or dominance and the amount of PH-PC1 + .

Results confirm the potential of these phenolic compounds as chemical markers of the sensory dynamics of the oils during storage in optimal conditions. The graph in Fig. 6 relates expected changes in sensory properties of oils on the basis of the variation of PH-PC1 + and PH-PC1- with storage time. The concentration of the two groups of phenols remained stable during the first 3 months of storage (T0-T1), with PH-PC1 + higher than PH-PC1-. In this period, small changes of the sensory properties occur. The strength of "green" descriptors evaluated by means IOC and DA diminished on average by 10 % of the initial intensities measured at T0 across the experimental oils. In the same interval time, the dominance of *bitterness* increases. From three to six months (T1-T2), important chemical changes occurred, and the ratio between PH-PC1 + and PH-PC1- concentrations reversed. These changes were associated with the decrease of "green" notes, both intensities (35 % less than T0) and dominance, and with the increase of *bitterness* dominance. From six to twelve months (T2-T3) the % of PH-PC1 + strongly decreased from the initial level (50 % less than T0), in contrast to the increase of PH-PC1- (30 % more than T0) and this associated with important and negative changes in the sensory profile of oils. In fact, in this period, *rancid* intensity (DA) increased on average by 50 % across oils from T0 along with the significant increase of the dominance of the defect. Finally, when both the decrease of PH-PC1 + and the increase of PH-PC1- reached 50 % with respect to T0, the intensity of *bitterness* and *pungency* (IOC) decreased by around 50 % while *rancid* intensity (DA) increased to the same extent.

4. Discussion

The present study proposes an approach to identify chemical markers of the dynamics of sensory changes of phenols-rich VOOs during storage in optimal conditions. Avoiding light and oxygen exposure, filtering oil, keeping temperature between 13–25 °C, are conditions known to prevent relevant chemical changes of the legal quality parameters of EVOOs (COI, 2018a(COI/BPS/Doc. No 1, 2018a)). However, little is known about the effect of early variations of phenolic and volatile compounds

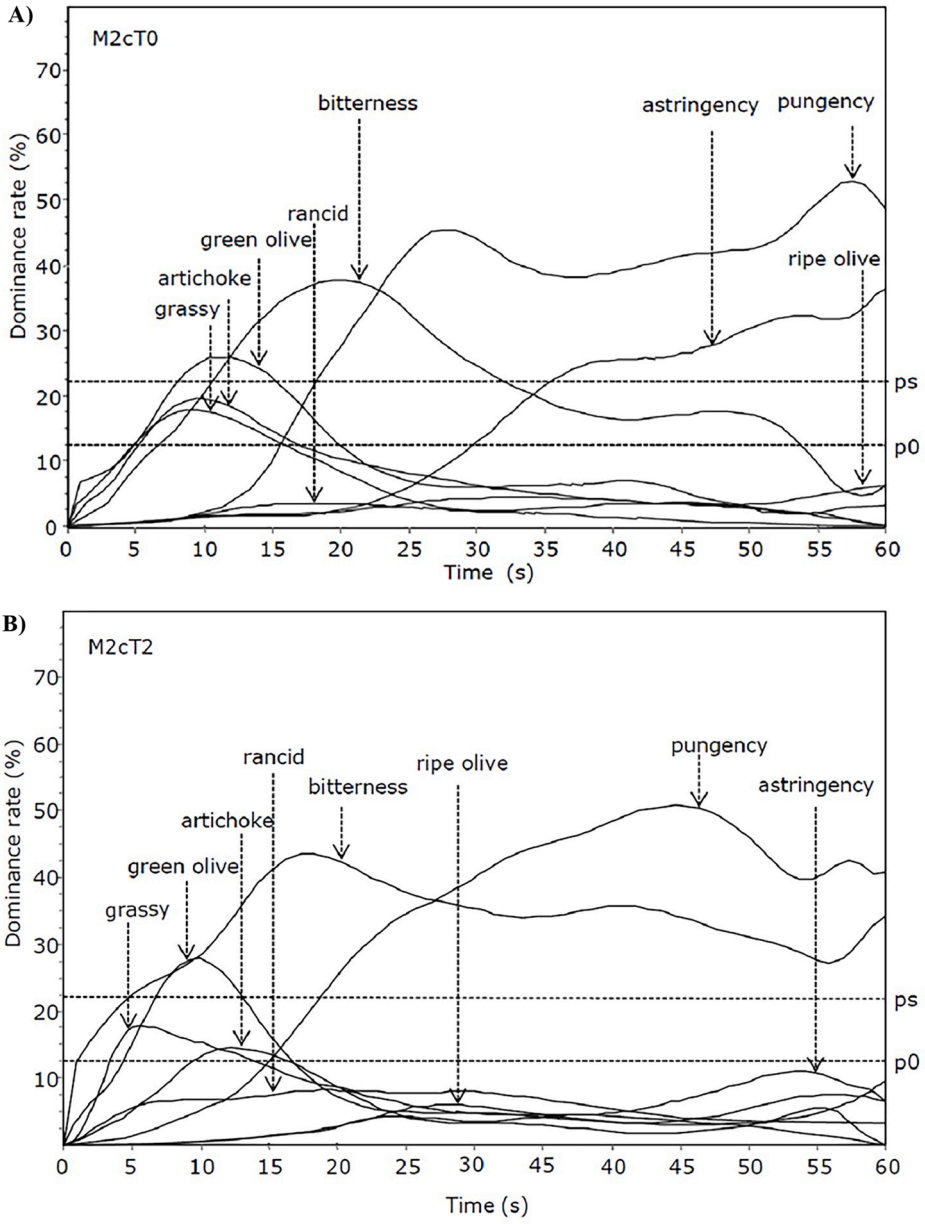

Fig. 5. A-b Temporal Dominance of Sensations data: influence of storage time on dominance curves of sensations in oils. Oil sample M2c TDS profile as described by a trained panel at T0 (a) and at T2 (b) after six months. p0 and ps correspond to chance and 95% significance levels of dominance frequency, respectively.

during storage and their impact on the dynamics of sensory modifications of oils. A high phenolic content is assumed to reinforce the oil stability against the oxidative damage and the onset of sensory defects (Bendini et al., 2007; Frankel, 1996), but their role in preserving attributes that connote fresh EVOOs sensory profile remains unclear. Then, two research questions appear relevant in this scenario: the temporal stability of sensory properties and the chemical markers of sensory properties in VOOs under optimal storage conditions. Answering the first question would allow researchers to re-think optimal storage conditions of high-quality products, whereas identifying chemical markers would be useful to both estimate and monitor the potential and actual stability of the sensory properties of oils.

Proposing models capable of predicting the sensory properties of EVOOs and how they change over time is one of the topics of the literature concerning virgin olive oil (Averbuch et al., 2023; Esposto et al., 2020). For example, models to predict the *bitterness* intensity from phenols content were proposed with conflicting results (Beltrán et al., 2007; Favati et al., 2013). Recently, Lobo-Prieto et al. (2020) studied long-term variation of VOO sensory characteristics using the chemicals that most varied over time, while Kottaridi et al. (2023) proposed mathematical models capable of predicting the intensity of *fruitiness*, *bitterness* and *pungency* using chemical parameters. However, the generalisation of the results is always debatable due to a limited sample in terms of olive oil varieties, geographic locations, and harvest years.

It is well-known that the interplay of a number of factors such as the environmental conditions in the orchard, the olive variety and olive ripening and integrity, the oil extraction processing and the operating conditions during oil storage and distribution, deeply affect chemical and sensory properties of VOOs and their evolution during oil storage (Angerosa et al., 2004; Clodoveo et al., 2014; Servili et al., 2004; COI, 2018a(COI/BPS/Doc. No 1, 2018a); Trapani et al., 2017). It is also known that monovarietal oils are characterised by very specific phenolic profiles that contribute to the peculiar sensory properties of fresh oils and can specifically influence the dynamic of their sensory modifications, even when optimal conditions are set up (Campestre et al., 2017; Esposto et al., 2020). Considering this extremely varied universe, the

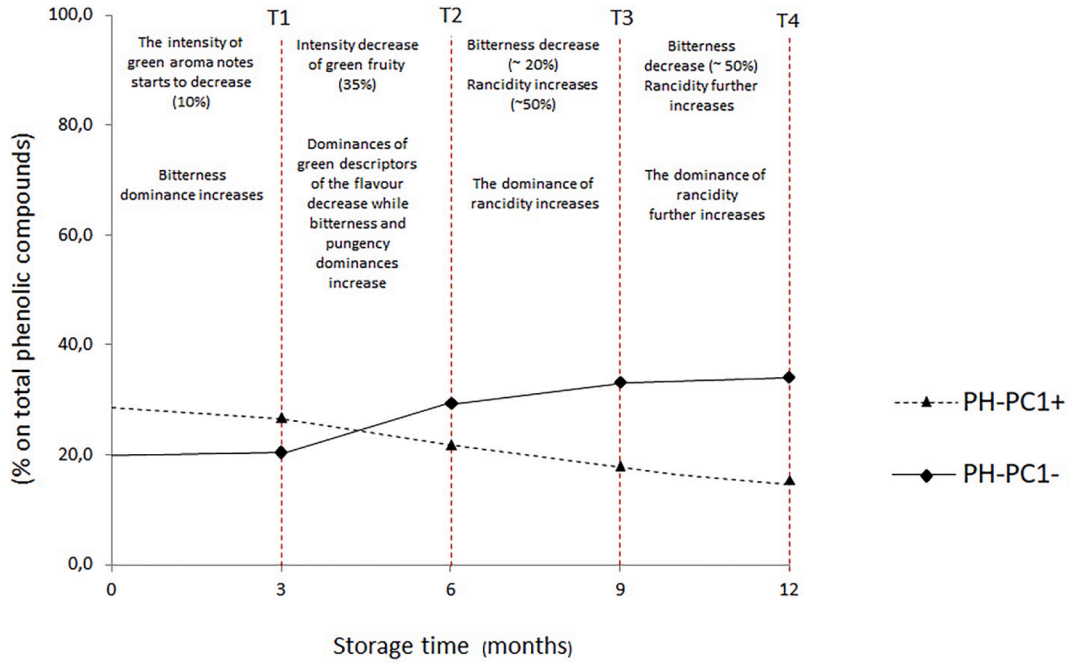

Fig. 6. Changes in sensory properties of oils during storage in relation to the mean variation % of phenolic compounds: PH-PC1+ (percentage of the sum of *p*-HPEA-EAOHox, *p*-HPEA-EA, 3,4-DHPEA-EAagl, 3,4-DHPEA-EDAox on total phenolic content) and PH-PC1- (percentage of the sum of *p*-HPEA-EDA, 3,4-DHPEA-EAOH and *p*-HPEA-EAOH, on total phenolic content).

identification of markers of sensory dynamics in extra virgin olive oil under optimal storage conditions with a general validity is not an achievable goal. On the contrary, when a specific productive context is of interest (given olive varieties and operational conditions in a given physical environment), the identification of chemical parameters related to changes of sensory properties would enable the oil sensory quality monitoring during early storage in optimal conditions.

In the present study, productive conditions were clearly set up in terms of geographic origin, olive variety and ripening, and operational conditions of both extraction process and storage. Furthermore, oils were produced at industrial scale considering the existing variability by batches. Thus, the relevance of the obtained results should be valued in relation to the general validity of the adopted methodological approach and to the relevance of the findings for the selected context.

4.1. The description of the sensory properties of VOOs during storage: Beyond the IOC method

The mandatory application of IOC sensory procedure in Europe contributed to the improvement of the quality of commercial EVOOs in the last approx. 30 years (Barbieri et al., 2020). The need of making easier the application of the method and overcome the evidence of discrepancies among panels in qualifying virgin olive oils induced worldwide researchers to develop chemical and statistical approaches to support the work of tasting Panels (Cecchi et al., 2019b; Quintanilla-Casas et al., 2020). However, the diffused tendency of using the IOC method even when checking for the presence/absence of defects is not the goal of the sensory evaluation is not appropriate. This is the case of many studies on the measurement of sensory modification of EVOOs during storage. The methodologies applied in the present research showed that other methods, more suitable to depict the complexity of the sensory properties of samples, are needed to facilitate the study of the relationships between sensory and chemical properties in both fresh and stored oils. Our results confirm that descriptive analysis allows for a more detailed description of the aroma and flavour of the oils in comparison to the IOC Panel Test (Monteleone, 2014). While the latter consisted in the evaluation of four positive attributes only, the DA sensory profile of experimental oils of this study was described by twenty attributes, fifteen of which significantly and clearly discriminated amongst the oils. Furthermore, the panel means of discriminating attributes were used to compute a robust PCA space (more than 65 % of explained variance after the first two PCs) on which the phenolic and volatile composition of oils was regressed allowing for the selection of the chemicals related to the sensory properties of oils. Perceptual maps depict similarities and differences among samples and allows the experimenter to select the minimum number of samples that retains the main difference among all samples ((Næs et al., 2011)). In our case, 6 oils retained the variability in the descriptive sensory data of the original 12 samples. Thus, coupling DA with Perceptual maps allows the researchers to work on a reduced number of samples making more efficient the study of sensory changes during oil storage.

The evolution of the dominance of the attributes measured by TDS in oils during the storage revealed that a reduction in intensity of the "green" descriptors of the flavour resulted in an increase of *bitterness* dominance. In other terms, the *bitter* connotation of oils rich in phenols after three-six months of storage in optimal conditions is even more pronounced than the one in fresh oils, catching the attention more compared to the other sensory properties. These changes in sensory properties during storage cannot be caught by the intensity evaluation of descriptors but require a method such as the TDS that allows assessing the evolution of the dominance of the sensations during tasting. The *bitterness* of temporal evolution of the flavour during storage could also result in a negative impact on the acceptance of products. Overall, coupling static and dynamic sensory evaluation methods improves the description of the dynamics of sensory properties of oil during storage in optimal conditions.

Of course, the application of these methods to study the temporal evolution of the sensory properties of olive oils during storage should remain strictly voluntary. They should be of interest for those companies producing high quality EVOOs and interested in using the sensory quality and distinctiveness of their oils as key element of their brand identity. Furthermore, their application should be aimed at setting up models to predict sensory changes by chemical markers possibly on a minimum number of samples still representing the sensory variation of

interest.

4.2. The relationship between sensory properties and chemical composition in fresh oils

The regression of chemical compounds on the intensity of sensory attributes showed clear association between specific phenolic and volatile compounds with oil sensory properties. Among the thirteen selected VOCs, those originating from the LOX pathway are all positively correlated with "green" descriptors (Fig. 3), confirming the positive association between volatile aldehydes and alcohols from the LOX pathway and *cut grass*, *green fruity*, *green olive* oil flavour and aroma (Nardella et al., 2023). The VOCs positively correlated with "green" descriptors other than those from the LOX pathways are still of oxidative origin (e.g., heptanal, octanal), indicating that, within the selected VOCs, almost all those positively correlated with "green" descriptors (the only exception is phenol) are of oxidative origin (enzymatic as in the case of the LOX VOCs, auto-oxidative as in the case of heptanal and octanal). On the other side, the VOCs negatively correlated with "green" descriptors (i.e., 2 + 3-methylbutanol, 2-octanone, 2-methylbutanal, 3-methylbutanal, ethyl acetate, 1-propanol) are all originating from microbial activities (Angerosa et al., 2004; Cecchi et al., 2021; Guerrini et al., 2020). These data suggest that in the fresh oils of this study, the presence of these volatile molecules is negatively correlated with the sensory freshness of the samples.

Concerning phenolic compounds, it should be noted that tyrosol and hydroxytyrosol are not related to the sensory space of the oils. These compounds are known to be abundant in the phenolic profile of aged or lower quality oil. This evidence confirms that oils at T0 are only slightly differentiated in terms of freshness of the phenolic profile.

The differences in "green" note intensities of the oils are instead correlated to two groups of phenols, in both of which are present derivatives of oleuropein (bearing hydroxytyrosol moiety) and of ligstroside (bearing tyrosol moiety). The group positively correlated to higher "green" notes includes hydroxylated forms such as *p*-HPEA-EAOHox and 3,4-DHPEA-EDAox, a dialdehydic form (i.e., *p*-HPEA-EA) and an aglycone form of oleuropein (i.e., 3,4-DHPEA-Eaagl); the group of phenols negatively correlated to "green" sensory attributes includes two aldehydic and hydroxylic forms such as 3,4-DHPEA-EAOH and *p*-HPEA-EAOH and a dialdehydic decarboxymethyl form (i.e., *p*-HPEA-EDA).

4.3. The predictive model of the sensory dynamics during storage

The variations of the phenolic composition of oils during storage is significantly correlated to the distance of the samples from the centre of the model, indicating that alterations in specific phenols characterising the fresh oils' composition can be used to predict changes in oil sensory properties over time. The observed relative variations of the phenolic compounds over time can be related to several complex phenomena such as the equilibriums among the different single molecules (Rovellini & Cortesi, 2002) and the antioxidant activity exerted by hydrophilic phenolic compounds in non-polar lipid matrices such as EVOO (Bendini et al., 2007; Frankel, 1996). For example, the decrease of 3,4-DHPEA-EAagl and 3,4-DHPEA-EDAox can be attributed to their high antioxidant activity due to the presence of the *o*-diphenol moiety, while the increase of 3,4-DHPEA-EAOH and *p*-HPEA-EAOH can be explained considering that these two molecules are at equilibrium with several other species from which they can originate over storage (Servili et al., 2004). Variations of VOCs appear less systematically linked to the distance from the model centre, with significant positive and negative correlations only found for heptanal and 3-methylbutanal, respectively. Heptanal increase could be related to the oxidation of some fatty acids such as linoleic and oleic acid during storage (Cecchi et al., 2019a; Luna et al., 2006; Morales et al., 1997) and its increase over storage is in agreement with previous literature (Lobo-Prieto et al., 2020). 3-Methylbutanal originates from microbial activity on the amino acid leucine during the early stages of oil production (Angerosa et al., 2004) and losses due to its volatility can explain its decrease along storage. It appears evident that changes in specific phenols characterising fresh oils composition can be used to predict oil sensory dynamics. Overall results from the present study clearly indicate that, when phenols-rich VOOs are stored in optimal conditions, chemical markers of sensory changes are different than those usually considered to track quality deterioration and defect onset (e.g., increase of free tyrosol and hydroxytyrosol in the phenolic fraction, specific volatile markers of sensory defects) (Brenes et al., 2001; Castillo-Luna et al., 2021; Cecchi et al., 2017; Lobo-Prieto et al., 2020). The point raised by the present manuscript is that monitoring early changes in sensory properties during storage requires measuring the modifications of the chemicals associated with the sensory descriptors of fresh oils (T0), which are mainly phenolic compounds belonging to the class of secoiridoids.

Of course, it cannot be excluded that phenolic or volatile molecules different than those considered to build up the model are related to sensory dynamics during storage. It could be for example the case of new formation molecules such as the aldehydes nonanal and (*E*)-2-heptenal usually correlated with the increase of rancidity over storage (Cecchi et al., 2019a). However, according to the approach adopted in the prediction model proposed here, the measurement of compounds known to be associated with sensory defects would not allow depicting early changes in sensory properties of fresh oils.

4.4. Chemical markers of sensory dynamics in oils under optimal storage conditions

Our results support the potential use of phenolic compounds as markers of sensory variations in oils during storage (Castillo-Luna et al., 2021; Korifi et al., 2016; Lerma-García et al., 2009). In our study, a decrease of the sum of *p*-HPEA-EAOHox, *p*-HPEA-EA, 3,4-DHPEa-EAagl, 3,4-DHPEA-EDAox (all together coded as PH-PC1 +) and a contemporary increase of the sum of *p*-HPEA-EDA, 3,4-DHPEA-EAOH and *p*-HPEA-EAOH, (all together coded as PH-PC1-) are directly related to gradual and progressive changes of the intensity and dominance of sensory descriptors of oil freshness. Percentage variations of the two phenolic groups are inversely related: when the former group decreases, the latter group increases. They also indicate that, in the optimal storage conditions applied in this research, variations in sensory characteristics are more pronounced than variations in the chemical profile (in particular, the volatile profile).

The ratio PH-PC1+/PH-PC1- could be adopted as a marker of the dynamics of sensory modification of oils during storage. When the ratio is greater than 1, minor sensory changes occur, and they are limited to a little decrease of the intensities of "green" descriptors along with an initial increase of the dominance of *bitterness*. When the ratio is 1, these changes are much more pronounced, and this value should be considered a cut-off passed which the sensory stability of fresh oils is lost. Finally, when the ratio is lower than 1, severe changes occur, including the perception of defects at non-negligible extent.

The index is based on the ratio between concentrations rather than being built on concentration ranges. Thus, its use can be generalised in similar contexts (similar phenolic profile of oils). The instrumental method to measure the phenolic compounds can be assumed as routinary; this aspect facilitates the possibility of including the analysis as a quality control parameter during storage. The index can also be used to define better conditions of oil storage. The validity of phenols identified as chemical markers of sensory dynamics is strictly related to the specific context of the oil experimental set considered in the present study and further studies, on different contests, adopting the same methodology are needed to verify the possibility of generalising the validity of the marker.

5. Conclusions

While there is a notable trend towards oils with high phenolic content due to their recognized nutritional benefits, it is crucial to acknowledge that even when stored under optimal conditions, these oils may undergo an early reduction in the perception of pleasurable sensations (e.g., green and fruity notes) determining the increase of the *bitterness* dominance in addition to the well-known increase in sensory defects like *rancidity*. To monitor these changes, it is crucial to explore early variations in phenolic and volatile compounds to understand the temporal stability of sensory properties. In the present study, the integration of static and dynamic sensory methods such as Descriptive Analysis (DA) and Temporal Dominance of Sensations (TDS) proved to be useful to obtain a more nuanced and comprehensive understanding of how sensory attributes evolve during storage. This approach also shed light on the correlation between sensory properties and chemical composition of fresh oils, highlighting the substantial associations between particular phenolic and volatile compounds and sensory attributes. The introduced predictive model illustrated how alterations in the phenolic composition of oils are linked to evolving sensory properties during storage. The suggested ratio (PH-PC1+/PH-PC1-) emerges as a potential marker for the dynamics of sensory modification, providing a practical tool for quality control throughout the storage period.

These findings underscore the paramount importance of meticulous planning in the production of phenols-rich oils, encompassing the careful selection of appropriate cultivars and the optimization of the extraction process. The ultimate objective should be to produce phenolic-rich oils without compromising desirable sensory attributes, in order to fully align with consumer preferences and expectations.

Ethical Statement

The study was conducted according to the principles established in the Declaration of Helsinki for medical research involving humans and was subject to ethical standards that promoted and ensured respect for all human subjects and protected their health and rights. In line with national regulations, given that the research was not medical, the research protocol was not submitted for approval to an ethical committee. The researchers involved in the study followed the code of Ethics & Standards for Sensory Project Managers developed by the Italian Sensory Science Society. Written informed consent was obtained from all participants according to the GDPR (General Data Protection Regulation) 2016/679. Participants were able to withdraw from the study at any time without giving a reason. The products tested were safe for consumption. Subjects received a gift card payment to motivate their participation in the study.

CRediT authorship contribution statement

Lapo Pierguidi: Formal analysis, Validation, Visualization, Writing – review & editing. **Lorenzo Cecchi:** Conceptualization, Formal analysis, Investigation, Methodology, Writing – original draft. **Caterina Dinnella:** Conceptualization, Formal analysis, Methodology, Writing – review & editing. **Bruno Zanoni:** Writing – review & editing, Conceptualization, Funding acquisition, Project administration. **Sara Spinelli:** Writing – review & editing, Methodology, Conceptualization. **Marzia Migliorini:** Supervision, Project administration, Funding acquisition. **Erminio Monteleone:** Writing – review & editing, Validation, Supervision, Methodology, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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The authors declare no conflict of interest.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodres.2024.114438.

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