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Physicochemical and sensory characteristics of virgin olive oils in relation to cultivar, extraction system and storage conditions

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ABSTRACT

This research was carried out to evaluate the effects of variety, extraction system and storage conditions such as packaging type and temperature variation on the quality of virgin olive oil. Several parameters were studied, namely, quality indices, polyphenols, tocopherols, volatile compounds and sensory properties. Thus, two olive varieties Chemlali (Tunisia) and Coratina (Italy) were selected. The olive oils were extracted by different industrial processes (super press, dual and triple phase decanter) then stored in the established conditions (ambient and refrigerator temperature) in the following packaging materials: clear and dark glass bottles and metal bottles. The oils were analyzed before and after being stored for 9 months. Principal Component Analysis and Graphical Modeling were applied to fully explore the influence of the studied factors. Results revealed that among samples, oils from Coratina cultivar were the richest in γ-tocopherol while Chemlali oils contained the highest amount of Δ-tocopherol. Quality indices namely K232 and K270 values were mainly influenced by the storage date and packaging material. Meanwhile, free acidity and peroxide value were mainly influenced by the extraction system. Concerning tocopherols, α-tocopherol content was mainly influenced by the packaging material, β-tocopherol was mainly affected by the storage date, for γ-tocopherol content the main influencing factor was the cultivar whereas for Δ-tocopherol the main influencing factor was the extraction system. Regarding volatile compounds their amounts were influenced mainly by the storage date, that was influenced by the packaging material, where a considerable decrease was observed after storage which was reflected by the change of sensory characteristics of stored oils: loss of positive attributes fruitiness, bitterness and pungency and onset of defects which were mainly influenced by the storage date (fruity and bitter attributes), packaging material (pungent, rancid and fusty attributes) and extraction system (musty attribute).

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1. Introduction

Tunisia olive groves present several varieties among which Chemlali constitutes the bulk of the Tunisian olive cultivation. Grown in warm coastal and low steppe Chemlali groves occupy nearly 85% of the olive-growing area and contribute over 80% of national production in olive oil. To improve olive oil quality produced in Tunisia, some approaches have been considered among them the introduction of different new cultivars. During the last years, foreign cultivars which showed an excellent olive oil quality and perfect adaptation to modern intensive cultivation conditions in their original site, have been introduced in Tunisian environment by private Tunisian companies for their easy harvesting and more rapid return on investment (Allalout et al., 2009). One of the introduced varieties is the Italian Coratina. This variety is known for its good quality and richness in antioxidants such as polyphenols and tocopherols.

It is well established that olive oil quality is influenced by several factors such as variety, environmental conditions, cultural and harvesting practices and technological processes. Hence, one of the primary causes of loss of olive oil quality is oxidation. The oxidation is an inevitable process that starts after the virgin olive oil has been extracted and leads to a deterioration that always becomes more serious during oil storage.

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Initially lipids are radically oxidized to hydroperoxides, which are odorless and tasteless (Frankel, 1985) and do not account for sensory changes. However, they are susceptible to further oxidation or decomposition into products of secondary reactions, which, conversely, are responsible for typical unpleasant sensory characteristics, identified on their whole as a rancid attribute.

Storage conditions are considered critical variables that influence the quality of olive oil and its shelf life which is attributable to lipid oxidation mechanisms which lead to rancidity (Vacca, Del Caro, Poiana, & Piga, 2006). Glass and metal bottles were used for packaging olive oils. The major function of packaging materials is related to their barrier properties against moisture, oxygen and light. It is known that these reactions are catalyzed by light and heat and are partly slowed down by compounds belonging to the unsaponifiable fraction. Polyphenols and tocopherols are the two main groups of compounds acting as primary antioxidants to inhibit oxidation in virgin olive oils. Many methods for phenol determination from different plants showed a variation of their concentrations which may be due to the exposure to several factors (Figueiredo-González, Simal-Gándara, et al., 2012; Pérez-Lamela, García-Falcón, Simal-Gándara, & Orriols-Fernández, 2007; Rodrigues, Pérez-Gregorio, Garcia-Falcon, & Simal-Gandara, 2009; Rodrigues, Pérez-Gregorio, García-Falcón, Simal-Gándarab, & Almeida, 2011) which could explain the significant difference between phenolic profiles from different plants. Regarding the great importance of the protective effect of virgin olive oil against cardiovascular, cancer, and neurodegenerative diseases, studying functional foods may be of great interest. Virgin olive oil is considered as an exceptional source of polyphenols, equalling all well-known polyphenol-rich plants such as green tea, pomegranate and date *Vitis vinifera* L. red grape varieties (Figueiredo-González, Martínez-Carballo, et al., 2012), onions (Pérez-Gregorio, Regueirob, Simal-Gándara, Rodrigues, & Almeida, 2013) and other plants.

The biological activities of olive oil phenolic compounds have prompted several studies on their potential activity in the prevention of cardiovascular diseases and cancer. However, a decrease in LDL oxidation was demonstrated with a greater ingestion of virgin olive oil phenolic compounds (Covas et al., 2006). In recent years, the anti-inflammatory and vascular protective properties of EVOO polyphenols have been extensively investigated (Fitó et al., 2008; Mc Geer, Mc Geer, & Schwab, 2009). This assumption has reinforced the proposed suggestions on phenolic compounds, which are able to interact with biological systems and act as bioactive molecules; in particular, they are important inhibitors of lipid peroxidation (Jemai, Bouaziz, Feki, El Fki, & Sayadi, 2008) and are believed to be effective through their free radical scavenging and metal-chelating properties. Despite the numerous studies that have shown the powerful antioxidant properties of olive oil, whether the effects really occur and the mechanisms responsible for these beneficial effects are not presently known. Polyphenols and tocopherols mainly act as chain breakers by donating a radical hydrogen to alkylperoxyl radicals formed during the propagation step of lipid oxidation and subsequently forming a stable radical tocopherols constitute the lipophilic antioxidant group and are noted for their effective inhibition of lipid oxidation in all vegetable oils (Velasco & Dobarganes, 2002).

Many studies on oxidation of virgin olive oils were based on accelerated test measurements (Aparicio, Roda, Albi, & Gutierrez, 1999; Baldioli, Servili, Perretti, & Montedoro, 1996; Gutierrez & Fernandez, 2002; Gutierrez, Villafranca, & Castellano, 2002; Krichene et al., 2010). In these studies, good correlations between changes in various components and stability were found. However, the extreme conditions in accelerated test high temperatures and with air bubbled into the oils do not simulate actual storage conditions and may lead to qualitative and quantitative changes to the oil that are not related to real-time storage. The main objective of our research is to investigate in depth how the extraction system and storage conditions affect the quality and composition of olive oil quality of two varieties: the Tunisian Chemlali and the Italian Coratina. To our knowledge, this is the first study of its kind that has applied the graphical modeling for full exploration of factors affecting olive oil physicochemical and sensory characteristics in a real-time shelf life study.

2. Materials and methods

2.1. Olive samples and oil extraction

Olive fruits (*Olea europaea*) of Coratina varieties were collected in an orchard in Calabria (South of Italy) while olives from Chemlali variety were collected in the orchard in Chaal (60 km away from Sfax in South East of Tunisia) and benefited both from the same cultural practices and the same ripeness index 3.5. We used for this study 900 kg of olives each from the two varieties, the orchard of 10 ha surface with trees planted at 24 × 24 m (17 trees/ha). Twenty marked trees of Chemlali variety were used to carry out this experiment knowing that each tree gave 45 kg of olives whereas for Coratina olives the orchard was of 400 trees/ha from which 30 marked trees were harvested each tree gave 30 kg. After homogenization and cleaning, fruits from both varieties were divided into three portions according to the extraction process used namely pressure system (SP) and centrifugation system with dual-(2P) and triple-phase (3P) decanters.

2.1.1. Pressure system

First, the olives are ground into an olive paste using large millstones. The olive paste generally stays under the stones for 45–50 min. After grinding, the olive paste is spread onto fiber disks, which are stacked on top of each other and then placed into the press. Traditionally, the disks were made of hemp or coconut fiber but nowadays they are made of synthetic fiber, which makes it easier to clean and maintain. These disks are then put on a hydraulic piston, forming a pile. Pressure is applied on the disks, thus compacting the solid phase of the olive paste and percolating the liquid phases (oil and vegetation water). The applied hydraulic pressure can go to 400 atm. The liquids are then separated by a standard process of decantation.

2.1.2. Centrifugation system

2.1.2.1. Triple-phase centrifugation system. Through this procedure, the olives were crushed to a fine paste. This paste was then malaxed for 50 min in order to allow the small olive droplets to coalesce. At this time, the paste was pumped into an industrial decanter to achieve the separation of the different constituents of the paste. During this process water was added to the paste in order to facilitate the separation of the three phases (oil, vegetation water and solids). The decanter is a large capacity-horizontal centrifuge rotating approximately 3000 rpm. The high centrifugal force created allows the phases to be readily separated according to their different densities (solids > vegetation water > oil). Inside the decanter's rotating conical drum is a coil rotating a few rpm (rounds per minute) slower pushing the solid materials out of the system. The separated oil and vegetation water are then rerun through a vertical centrifuge, working around 6000 rpm separating the small quantity of vegetation water still contained in oil and vice versa.

2.1.2.2. Dual-phase centrifugation system. The olives were crushed to a fine paste, then, olive paste was kneaded for 50 min at 22 ± 2 °C. Afterwards, the paste is pumped into a dual-phase industrial decanter where the liquid and solid phases were separated. Finally, the oil was separated from the obtained liquid phase by means of an automated discharge vertical centrifuge. After extraction, the olive oils were stored, in full filled bottles of three kinds of packaging: clear, dark glass and metal bottles, up to 9 months at temperatures of 8 °C and ambient temperature, and analyzed at the beginning of the experiment and again after 9 months of storage. Temperature conditions were chosen to simulate ideal and poor storage.

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The full design consisted of a total of 36 olive oil samples: six evaluated at fresh, and the remaining 30 samples at the two storage temperatures and packaging storage conditions.

2.2. Quality indices

Free fatty acidity, peroxide value (PV), conjugated dienes (K232) and conjugated trienes (K270) were determined by the methods reported in Regulation EEC/2568/91 of the European Union Commission (EEC, 1991).

2.3. Determination of tocopherol composition

Tocopherol composition was measured by the method of Gimeno, Castellote, Lamuela-Raventos, De La Torre, and Lopez-Sabater (2000). The oil sample was diluted in hexane (1:10). Thereafter, 200 μL was transferred to other test tube, where 600 μL of methanol and 200 μL of the internal standard solution (300 μg/mL of α-tocopherol acetate in ethanol) were added. The samples were filtered through a 0.45 μm pore size filter and an aliquot was directly injected into the chromatograph. Separation by HPLC was carried out using a Hewlett-Packard liquid chromatographic system (Waldbronn, Germany) with an HP-1100 pump system and a Rheodyne Model 7725 injector (CA, USA) with a final volume loop of 20 μL. The detector was a HP-1200 multi-array detection system. The column was a supelcosil ODS-2 (150 × 4.5 mm, i.d. 5 μm).

2.4. Volatile compounds

Supelco (Bellefonte, PA) SPME devices coated with polydimethylsiloxane (PDMS, 100 μm) were used to sample the headspace of 2 mL of olive oil inserted into a 5-mL vial and allowed to equilibrate for 30 min. After the equilibration time, the fiber was exposed to the headspace for 50 min at room temperature. Once sampling was finished, the fiber was withdrawn into the needle and transferred to the injection port of the GC–MS system. GC–EI/MS analyses were performed with a Varian (Palo Alto, CA) CP 3800 gas chromatograph equipped with a DB-5 capillary column (30 m × 0.25 mm, 0.25 μm; Agilent, Santa Clara, CA) and a Varian Saturn 2000 ion trap mass detector. Analytical conditions were as follows: injector and transfer line temperatures were 250 and 240 °C, respectively; oven temperature was programmed from 60 to 240 °C at 3 °C/min; helium was used as carrier gas at a flow of 1 mL/min. The identification of the volatile compounds was based on the comparison of the retention times with those of authentic standards, comparing their linear retention indices (LRI) relative to a series of n-hydrocarbons, and on computer matching against commercial (NIST 98 and Adams) and home-made library mass spectra, built from pure substances, components of known oils, and MS literature data (Adams, 1995; Davies, 1990; Jennings & Shibanoto, 1982; Massada, 1976). Moreover, the molecular weights of all the substances identified were confirmed by GC–CI/MS, using methanol as the ionizing gas.

2.5. Phenolic composition

The phenolic fraction was obtained by extraction of a solution of oil in hexane with methanol/water mixture (80:20) 2% between 20, two times. Folin–Ciocalteu reagent and sodium carbonate were added to a suitable aliquot of the combined extracts and absorbances of the solution were measured at 765 nm. Total phenol concentrations are expressed as mg of hydroxytyrosol per kg of oil (Montedoro et al., 1993).

2.6. Sensory analysis

Sensorial evaluation of the oils was performed according to the panel test method by a fully trained analytical taste panel recognized by the International Olive Oil Council (IOOC). A panel test was established using the IOOC standard profile sheet method (COI, 2011).

2.7. Statistical analysis

To point out the relationship between storage factors and analyzed parameters, principal component analysis (PCA) was carried out on oil samples. In a second stage and in order to determine the influence of the factors Extraction System, Variety, Storage, Date and Temperature we use a technique called graphical models that allows us to determine which of these factors is more determinant in the variation of each parameter. All the estimation procedures were done by software called MIM that was implemented by Edwards (2000).

We start with a complete graph where the vertices are the four factor variables Storage, Date, Packaging and Extraction Systems and with a fifth vertex that represents one of the physicochemical parameters. We then perform a backward stepwise selection procedure where the most "non-significant" (corresponding to the greater P-value) edge is suppressed. This procedure stops when all the edges of the graph correspond to P-values less than the 5% level of significance.

3. Results and discussion

The mean values and standard deviations of the studied analytical parameters for the olive varieties Chemlali and Coratina before and after the 9-month storage in the different storage conditions are summarized as supplementary data (Tables A.1 and A.2).

In the following sections we explore in depth the variation of the physico-chemical and sensorial characteristics of the different oil samples with storage and the influence of each factor on the quality of the studied VOOs.

3.1. Principal component analysis

The use of exploratory and classification statistical approaches such as principal component analysis (PCA) can identify patterns in samples and variables contributing to the clustering of samples.

In order to find homogeneous groups of olive oil samples according to the studied factors, PCA was applied to the physicochemical variables.

The PCA results are graphically displayed using two plots. In the first ones, the sample scores are plotted to show the relationship between the samples (Fig. 1A, B, C, and D); in the second (Fig. 2), the loadings of the original measured variables on principal components are plotted to aid the interpretation of components in terms of the original variables. The two plots can be interpreted together.

The first two PCAs explained 48.96% of variance (Fig. 2). The first PC accounted for 32.44% of the total variance and was highly correlated with K270, K232, PV and acidity values. The second PCA (17.51%) showed positive correlation with Δ-tocopherol.

Fig. 1A shows that, independently of the rest of the studied factors, the samples of each variety were clustered together along the PC2. The Chemlali samples are characterized by higher amounts of Δ-tocopherol in comparison to those of Coratina olive oil samples.

In Fig. 1B, the obtained PCA shows the presence of two main groups clustered according to the storage date. Fresh olive oil samples are characterized by high amounts of α-tocopherol and low PV, K232 and K270 values in contrast to the stored olive oil samples. In a recent work, Dabbou, Gharbi, Dabbou, Brahmi, and Nakbi (2011) have also found that PV, K232 and K270 increase with storage. Similar results were also found by Rababah, Feng, Yang, Eriefej, and Al-omoush (2011) for Jordanian olive varieties.

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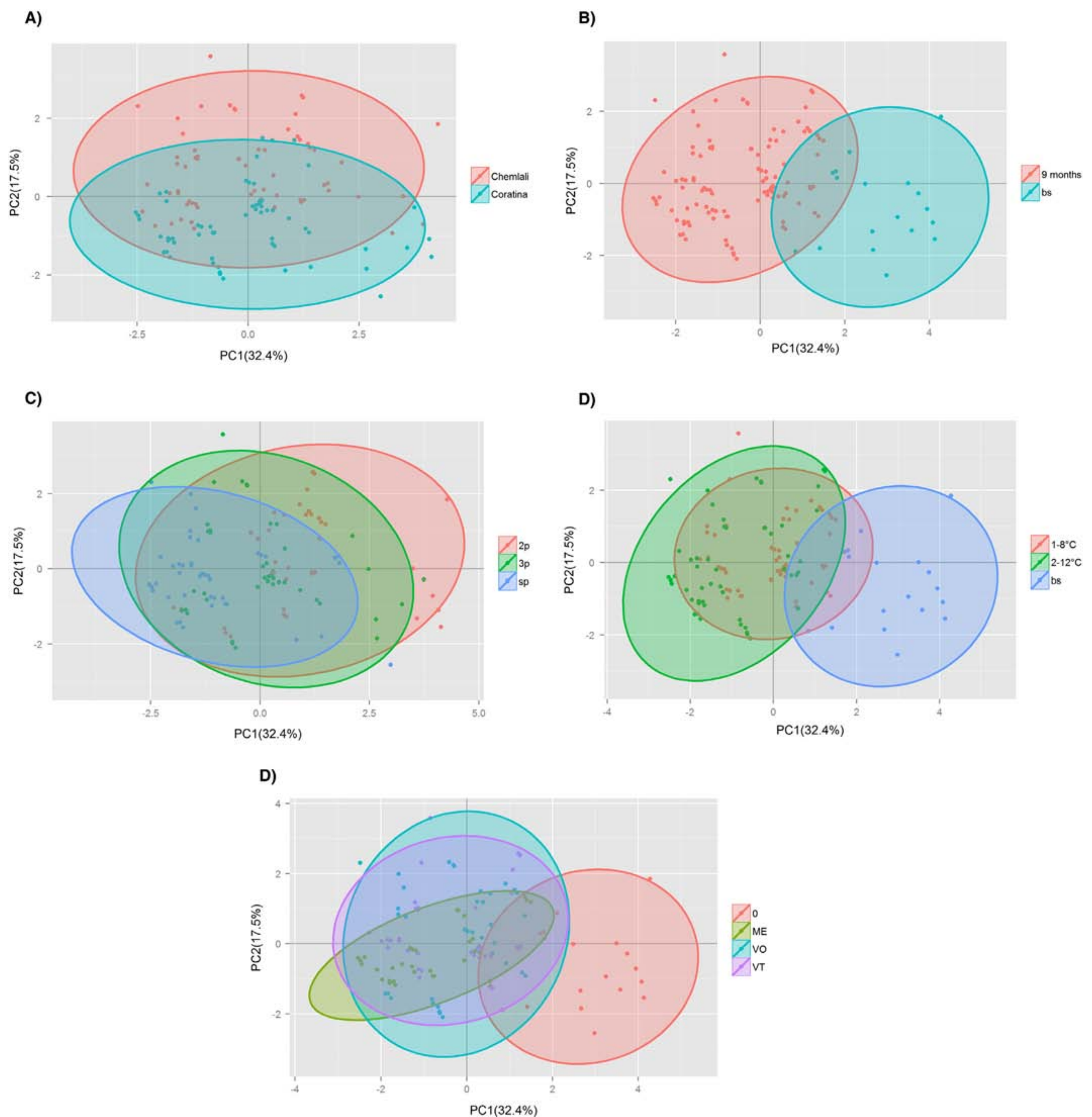

Fig. 1. A. Scores plot of PCA applied to the data set of analytical variables obtained from different studied varieties. 1 (Chemlali), 2 (Coratina). B. Scores plot of PCA applied to the data set of analytical variables obtained from different studied periods. 0 (before storage), 2 (after 9 months of storage). C. Scores plot of PCA applied to the data set of analytical variables obtained from different extraction processes. 1 (SP), 2 (2P), 3 (3P). D. Scores plot of PCA applied to the data set of variables according to the studied temperatures. 0 (Fresh VOOs), 1 (Fridge temperature), 2 (Room temperature). E. Scores plot of PCA applied to the data set of variables according to the used packaging. 0 (Fresh VOOs), 1 (Clear glass bottles), 2 (Dark glass bottles) 3 (Metallic bottles).

The application of PCA to the samples on the basis of the extraction system employed does not show a good separation among VOOs especially for the 2-phase and 3-phase centrifugation system (Fig. 1C).

According to the temperature storage variation, the observation of Fig. 1D allows one to distinguish the presence of three main groups. The first one consists of the fresh VOOs and the second one is formed by the VOO samples stored under refrigeration temperature and the last one composed of the VOOs stored under room temperature conditions. Examining the loading plot of the variables, we can conclude that the K232 and K270 values increase with the storage under higher temperature.

Three kinds of packaging were used during the storage (clear, dark glass bottles and metallic packaging). Fig. 1E shows that VOO samples from the different packaging were aggregated together around axis's origins. No important separation was observed especially between clear and dark glass samples. Oils stored in metallic packaging have higher PV.

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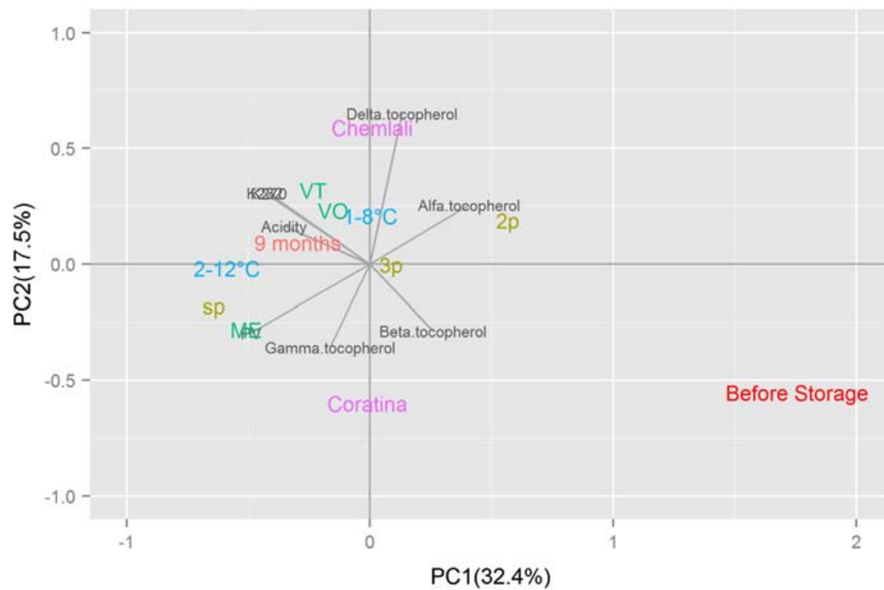

Fig. 2. Loading plot of PCA applied to the studied variables.

Another PCA was applied to simultaneously compare the effect of variety, storage, packaging material and extraction system on the physicochemical characteristics of the analyzed oils (Figs. 3 and 4).

Figs. 3 and 4 show that Coratina oils are characterized by higher amounts of 2 methyl butanal, cis 3 hexen 1 ol, cis 3 hexenyl acetate, hexyl acetate, cis-2-pentenol, 2-pentanone, and 3-pentanone and it is noteworthy for its higher content of phenolic compounds, while Chemlali samples were richer in pentanol, trans-3-hexenol, octanal, hexanal and penten-3-ol. This variation of aromatic composition between varieties is in agreement with previous work which reported that genetic characteristics fix the contents of the different enzymes and are therefore responsible for the qualitative composition of volatile compounds (Angerosa, Basti, Vito, & Lanza, 1999).

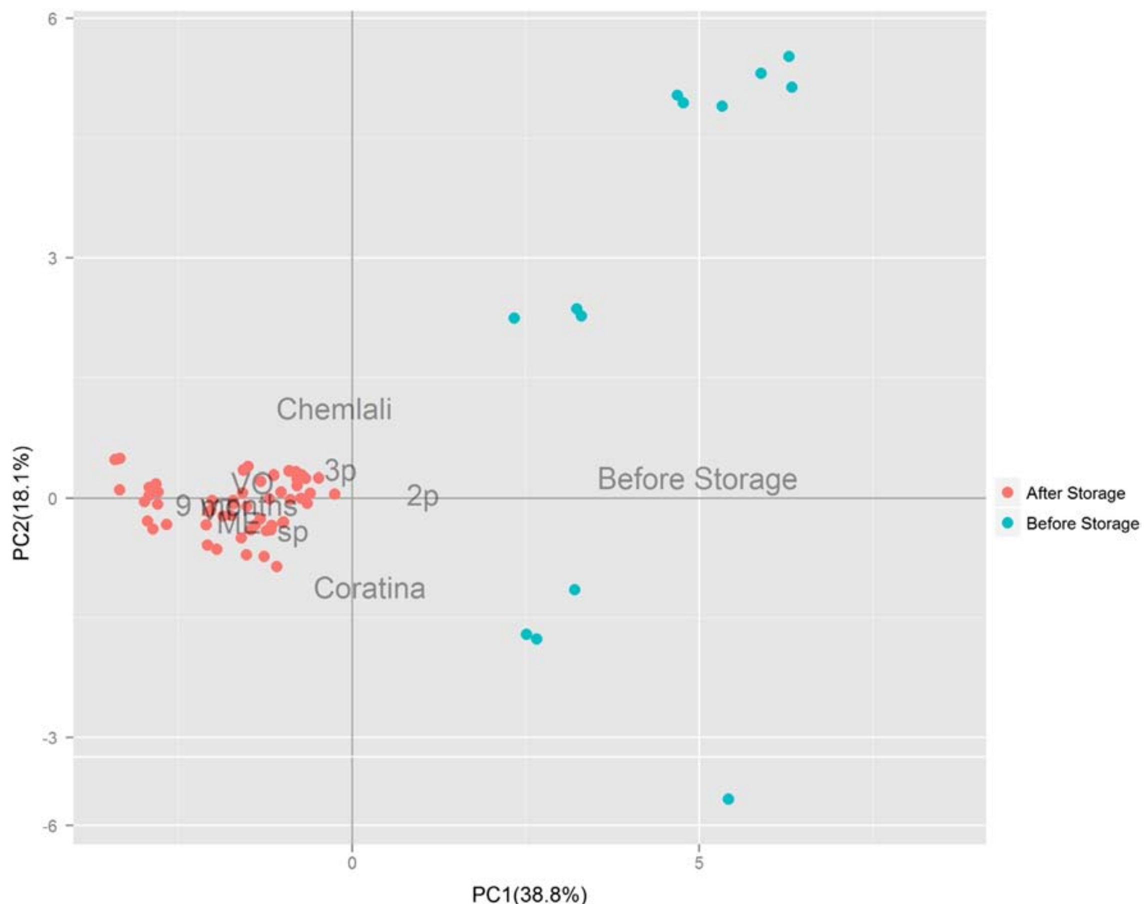

Fig. 3. PCA applied to the data set according to all factors under study.

Fig. 4. Loading plot of PCA applied to the studied traits.

Before storage olive oil samples presented positive attributes especially Coratina oils were more pungent and bitter, both varieties had similar degree of fruitiness. After storage, olive oil samples lost their sensory characteristics of bitterness and pungency. In virgin olive oils, these sensations are related to the presence of phenolic compounds (Servili & Montedoro, 2002). During storage they undergo qualitative and quantitative modifications due to decomposition and oxidation reactions (Morelló, Motilva, Tovar, & Romero, 2004). The total phenol content decreases and, consequently, the typical bitter taste and pungent note of fresh EVOO decrease in intensity. This result is consistent with that reported by Dabbou et al. (2011) who found that polyphenol amounts decrease with storage.

Regardless of the packaging used, all stored VOOs were clustered together and negatively correlated with aromas. Their amounts decrease considerably over storage time. No important difference was observed at the end of storage time between oils obtained by different extraction systems. Oils extracted with 2P system were slightly correlated with the aroma compound 2-methyl butanal.

3.2. Graphical modeling

In this section, we propose a probabilistic model in order to fully understand the impact of variety, extraction system and storage conditions on the physicochemical characteristics of virgin olive oils. In this part the storage temperature variation was not considered.

Tables 1.A–1.E show the estimated mixed graphical obtained from the data. This later family of models can be used for discrete and continuous variables. They combine at the same time log-linear models for discrete variables with graphical Gaussian models for continuous variables

Table 1.A
Variables linked to extraction system factor.

	Extraction system			Variance
	sp	2p	3p	
Free acidity (%C18:1)	0.745	0.465	0.45	.011
PV (Meq O_2/kg)	18.064	11.471	13.872	8.892
Δ-Tocopherol (ppm)	1.147	1.265	1.217	.02
Musty	.546	0	.054	.24
	Graph			

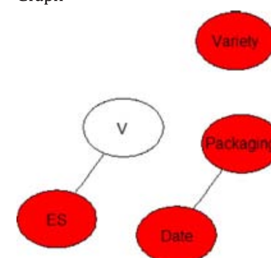

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Table 1.B
Variables linked to storage date factor.

	Date		Variance
	t = 0	9 months	
K232	1.94	2.543	0.148
β-Tocopherol (ppm)	13.275	2.642	54.076
2-Methyl butanal (ppm)	0.806	0.046	0.095
2-Pentanone + 3-Petanone (ppm)	12.988	0.108	8.249
Hexanal (ppm)	39.204	0.162	213.965
1 Penten-3-ol (ppm)	27.682	0.036	54.594
1 Pentanol (ppm)	4.921	0.069	5.056
Hexyl acetate (ppm)	2.737	0.01	2.513
Octanal (ppm)	8.558	0.002	31.8
Cis-3-hexenyl acetate (ppm)	12.716	0.062	17.584
Cis-2-penten-1-ol (ppm)	5.342	0.031	8.281
Trans-3-hexen-1-ol (ppm)	24.68	0.271	197.437
Cis-3-hexen-1-ol (ppm)	51.438	0.193	674.444
Fruity	3.867	1.995	0.82
Bitter	2.55	1.152	0.489
	Graph		

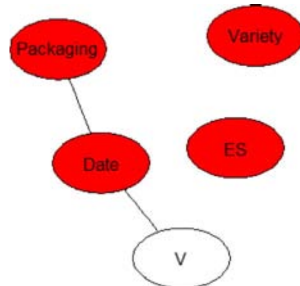


Table 1.C
Variables linked to packaging factor.

	Packaging				Variance
	t = 0	P1	P2	P3	
Fusty	0	1.05	0.272	1.044	0.608
Rancid	0	1.622	0.344	1.972	0.557
Pungent	2.194	0.928	0.772	1.511	0.524
	Graph				

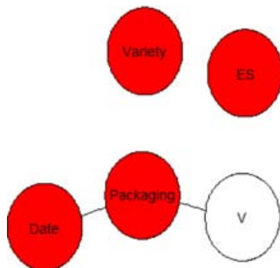


Table 1.D
Variables linked to varietal factor.

	Variety		Variance
	Chemlali	Coratina	
γ-Tocopherol (ppm)	6.937	9.674	6.713
	Graph		

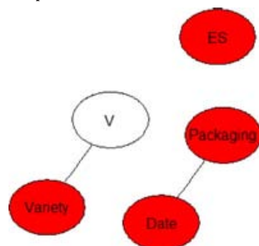
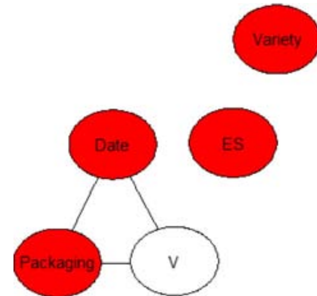


Table 1.E
Variables linked to packaging and date factors.

	Packaging-date				Variance
	t = 0	9 months			
		P1	P2	P3	
α-Tocopherol (ppm)	187.43	103.676	132.716	95.063	2758.371
K270	0.137	0.253	0.271	0.238	0.002
	Graph				



(see Lauritzen, 1996). We have used in this paper homogeneous mixed interaction models since we have made the assumption that the variances do not depend on the discrete variables (factors). These models are a generalization of ANOVA and MANOVA. The used underlying joint distribution is the multivariate conditional-Gaussian distribution (see Edwards, 2000 for details). The main advantage of these models is that they can have firstly a graphical representation and secondly the model can be

Table 2.A
Table explaining the graphical model for the variables linked to extraction system factor.

Variable	Edge added	Statistic test	DF	P	
Free acidity (%C18:1)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ec]	101.7064	2	P < 0.0001	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vc]	6.2298	1	0.0126	+
	[ab]	80.9763	3	P < 0.0001	+
	[ac]	15.7	1	0.0001	+
	[bc]	16.1831	3	0.001	+
PV (Meq O_2/kg)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ed]	65.5149	2	P < 0.0001	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vd]	2.2998	1	0.1294	
	[ab]	80.9763	3	P < 0.0001	+
	[ad]	12.6633	1	0.0004	+
	[bd]	39.3219	3	P < 0.0001	+
Δ-Tocopherol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ej]	13.1961	2	0.0014	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vj]	2.6618	1	0.1028	
	[ab]	80.9763	3	P < 0.0001	+
	[aj]	1.4318	1	0.2315	
	[bj]	7.5975	3	0.0551	
Musty	[EM]	25.7995	2	P < 0.0001	+
	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[MV]	15.1033	1	0.0001	+
	[Ma]	5.6733	1	0.0172	+
	[Mb]	7.4037	3	0.0601	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[ab]	80.9763	3	P < 0.0001	+

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Table 2.B
Table explaining the graphical model for variables linked to storage date factor.

Variable	Edge added	Statistic test	DF	P	
K232	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ee]	3.2627	2	0.1957	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Ve]	16.7941	1	P < 0.0001	+
	[ab]	80.9763	3	P < 0.0001	+
	[ae]	44.3573	1	P < 0.0001	+
	[be]	48.5541	3	P < 0.0001	+
	[bk]	83.6216	3	P < 0.0001	+
β-Tocopherol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ei]	3.3545	2	0.1869	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vi]	5.1309	1	0.0235	+
	[ab]	80.9763	3	P < 0.0001	+
	[ai]	39.0227	1	P < 0.0001	+
	[bi]	39.0834	3	P < 0.0001	+
	[bl]	37.8219	3	P < 0.0001	+
Hexanal (ppm)	[EH]	0.118	2	0.9427	
	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[HV]	10.7596	1	0.001	+
	[Ha]	91.961	1	P < 0.0001	+
	[Hb]	91.9615	3	P < 0.0001	+
	[Va]	0	1	P < 0.0001	
	[Vb]	0	3	P < 0.0001	
	[ab]	80.9763	3	P < 0.0001	+
1-Penten-3-ol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Em]	0.7591	2	0.6842	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vm]	5.3578	1	0.0206	+
	[ab]	80.9763	3	P < 0.0001	+
	[am]	131.8301	1	P < 0.0001	+
	[bm]	131.8301	3	P < 0.0001	+
Hexyl acetate (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Eh]	5.0989	2	0.0781	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vh]	9.3544	1	0.0022	+
	[ab]	80.9763	3	P < 0.0001	+
	[ah]	51.0646	1	P < 0.0001	+
	[bh]	51.0646	3	P < 0.0001	+
Octanal (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Eo]	4.6383	2	0.0984	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vo]	10.6855	1	0.0011	+
	[ab]	80.9763	3	P < 0.0001	+
	[ao]	42.1164	1	P < 0.0001	+
	[bo]	42.1164	3	P < 0.0001	+
Cis-3-hexenyl acetate (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ep]	4.9125	2	0.0858	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vp]	3.7811	1	0.0518	
	[ab]	80.9763	3	P < 0.0001	+
	[ap]	105.759	1	P < 0.0001	+
	[bp]	105.7597	3	P < 0.0001	+
Cis-2 penten-1-ol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
Cis-2 penten-1-ol (ppm)	[Eq]	3.28	2	0.194	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vq]	11.9927	1	0.0005	+
	[ab]	80.9763	3	P < 0.0001	+
	[aq]	56.6036	1	P < 0.0001	+
	[bq]	56.6051	3	P < 0.0001	+
Trans-3-hexen-1-ol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Et]	2.64	2	0.2671	
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vt]	13.0238	1	0.0003	+
	[ab]	80.9763	3	P < 0.0001	+
	[at]	51.798	1	P < 0.0001	+
	[bt]	51.799	3	P < 0.0001	+
Cis-3-hexen-1-ol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Er]	16.1475	2	0.0003	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vr]	0.92	1	0.3375	
	[ab]	80.9763	3	P < 0.0001	+
	[ar]	62.2547	1	P < 0.0001	+
	[br]	62.2549	3	P < 0.0001	+
Fruity	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Es]	19.135	2	0.0001	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vs]	0.806	1	0.3693	
	[ab]	80.9763	3	P < 0.0001	+
	[as]	66.3365	1	P < 0.0001	+
	[bs]	66.545	3	P < 0.0001	+
Bitter	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Eu]	6.9273	2	0.0313	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vu]	3.2536	1	0.0713	
	[ab]	80.9763	3	P < 0.0001	+
	[au]	63.4188	1	P < 0.0001	+
	[bu]	67.1092	3	P < 0.0001	+

written using model formulae that are similar to the model formulae for log-linear models and for graphical Gaussian models. In Tables 1.A–1.E we have used when representing these variable red circles for discrete variables and white circles for continuous variables. In Tables 2.A–2.E we give the explanation for the graphical models. The edge indicates a significant ($P < 0.05$) relation between the concerned pair of variables. We have used here a forward stepwise procedure to estimate the mixed graphical model. We start by the main effect model, i.e., the model without any interaction and we add in a stepwise procedure the most significant edge ($P < 0.05$). The procedure stops when no further edge can be added. The hypothesis statistical test used is based on the deviance between the two nested models (see Edwards, 2000).

The factors: variety, storage date, packaging and extraction system showed significant effect on free acidity, $P < 0.05$ (Table 1.A). Only the effect of extraction system was plotted in the chain graph because its P value was the lowest and it carries out the most important effect.

The observation of mean values (Table 1.A) shows that VOOs obtained by SP system presented the highest free acidity values. As reported by Torres and Maestri (2006a,b), during the SP process, the oils were extracted with the vegetable water (aqueous phase plus solid wastes) and remain together until they are separated by decanting, which may favor the hydrolysis of triglycerides, resulting in an increase of free fatty acid level. Peroxide value was also mainly influenced by the extraction

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Table 2.C
Table explaining the graphical model for variables linked to packaging factor.

Variable	Edge added	Statistic test	DF	P	
Fusty	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ew]	7.6816	2	0.0215	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vw]	3.6772	1	0.0552	
	[ab]	80.9763	3	P < 0.0001	+
	[aw]	18.613	1	P < 0.0001	+
	[bw]	36.1314	3	P < 0.0001	+
Pungent	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ev]	9.6134	2	0.0082	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vv]	18.8522	1	P < 0.0001	+
	[ab]	80.9763	3	P < 0.0001	+
	[av]	39.2749	1	P < 0.0001	+
	[bv]	53.9161	3	P < 0.0001	+
Rancid	[ER]	0.2032	2	0.9034	
	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[RV]	9.8408	1	0.0017	+
	[Ra]	35.595	1	P < 0.0001	+
	[Rb]	87.9461	3	P < 0.0001	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[ab]	80.9763	3	P < 0.0001	+

procedure adopted. In fact, the highest mean values of this parameter were registered for olive oils obtained by super press which reached 18.06 Meq O_2/kg.

The conjugated diene (K232) and triene (K270), which are formed during autoxidation of oil, increased above the threshold value over storage time as expected. This result is consistent with that reported by Dabbou et al. (2011). The storage duration and packaging material were the most influencing factors being the K270 influenced the most by the packaging material. Dark-glass-bottled oils had the highest mean value of K270 (0.271).

Regarding tocopherols, results show that α-tocopherol amount decreases after the 9-month storage and depending on the packaging material. Alpha-tocopherol amount was influenced most by the storage date and packaging material; their corresponding P values were the lowest and their statistic test values were the highest (80.9763 and 45.2257, respectively). The fat soluble α-tocopherol is the predominant representative of vitamin E in virgin olive oil. It was reported that the main changes in the concentrations of this compound are associated with the higher oxygen levels in the empty portion of the glass bottles and that α-tocopherol is the first molecule to be oxidized, whereas squalene and o-diphenols are protected in the first months due to the presence of α-tocopherol (Rastrelli, Passi, Ippolito, Vacca, & De Simone, 2002). According to our results, dark-glass bottle seems to be the best packaging material for α-tocopherol preservation during storage. Concerning β-tocopherol amounts, they decreased dramatically after 9 months of storage and the storage duration was the most affecting factor, whereas, for γ-tocopherol content the varietal factor seems to be the most prominent. As our results show, Coratina VOO is richer in γ-tocopherol than Chemlali. On the other side, the extraction system was responsible for Δ-tocopherol variations, olives extracted by 2P and 3P systems gave oils richer in Δ-tocopherol than those of super press.

Table 2.D
Table explaining the graphical model for variables linked to varietal factor.

Variable	Edge added	Statistic test	DF	P	
γ-Tocopherol (ppm)	[EG]	1.8539	2	0.3958	
	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[GV]	25.574	1	P < 0.0001	+
	[Ga]	16.7579	1	P < 0.0001	+
	[Gb]	17.2159	3	0.0006	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[ab]	80.9763	3	P < 0.0001	+

Table 2.E
Table explaining the graphical model for variables linked to packaging and date factors.

Variable	Edge added	Statistic test	DF	P	
K270	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Ef]	17.4417	2	0.0002	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vf]	1.4802	1	0.2237	
	[ab]	80.9763	3	P < 0.0001	+
	[af]	91.6831	1	P < 0.0001	+
	[bf]	98.5159	3	P < 0.0001	+
α-Tocopherol (ppm)	[EV]	0	2	1	
	[Ea]	0	2	1	
	[Eb]	0	6	1	
	[Eg]	6.4484	2	0.0398	+
	[Va]	0	1	1	
	[Vb]	0	3	1	
	[Vg]	27.9057	1	P < 0.0001	+
	[ab]	80.9763	3	P < 0.0001	+
	[ag]	37.7309	1	P < 0.0001	+
	[bg]	45.2257	3	P < 0.0001	+

The aroma of olive oil is attributed to aldehydes, alcohols, esters, hydrocarbons, ketones, furans and, probably, others as yet unidentified volatile compounds. The major volatile compounds reported in virgin olive oils are the C6 and the C5 volatile compounds (Kalua et al., 2007). The C6 and C5 volatile compounds are enzymatically produced from polyunsaturated fatty acids through the so called lipoxygenase (LOX) pathway. The quali-quantitative profiles of these compounds depend on the level and activity of each enzyme involved in this LOX pathway (Angerosa & Basti, 2001; Aparicio & Morales, 1998).

However, whether major or minor, volatile compounds are crucial for virgin olive oil. Among olive oil aromas eleven mostly belong to C5–C6 compounds responsible for positive attributes in high quality olive were quantified in this study. The chain graph model revealed that all volatile compounds in analyzed olive oils were affected by storage; regardless of the cultivar and the extraction system, all the detected aroma compounds substantially decrease over storage time which was by its turn influenced by the packaging material.

It was reported that the storage of either the fruit or oil produces volatile compounds that are responsible for off-flavor (Kiritsakis, 1998; Koprivnjak, Procida, & Zelinotti, 2000). The contribution of each volatile compound to the whole aroma and flavor is related to their concentration in the oil with respect to their sensory threshold (Morales, Rios, & Aparicio, 1997). Bendini, Cerretani, Salvador, Fregapane, and Lercker (2010) concluded that *E*-2-heptenal, nonanal and 2-decenal are the most frequently used volatile markers of oxidation of virgin olive oil during storage. These three compounds are characterized by low odor threshold (5, 150 and 100 μg kg^{-1}, respectively) and by negative off-flavors.

Evaluation of the sensory quality of virgin olive oils involves perception of both favorable and unfavorable sensory attributes, with evaluation of sensory defects being used to classify oils into various grades (Kalua et al., 2007). The International Olive Oil Council has developed a specific vocabulary for virgin olive oil sensory descriptors. Fruity, bitter and pungent are considered as positive attributes whereas the common

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defect attributes are fusty, musty-humid, muddy-sediment, winey-vinegary, metallic and rancid.

Perceived sensory attributes usually arise from the influence and interaction of several volatile compounds, rather than the action of a single compound. Sensory evaluation detects oxidative deterioration before changes are observed in these other parameters, and this emphasizes the importance of volatile compounds in detecting early stages of olive oil deterioration (Vichi, Pizzale, Conte, Buxaderas, & Lopez-Tamames, 2003). In our study, the consequent result of the aromas changes is the change of sensory characteristics of stored olive oil. Mean values of positive attributes fruity, bitter and pungent decreased under the studied storage conditions (date and packaging) whereas defect attributes rose up. Although the oils stored in metallic packaging retained more the pungent attribute, they showed more rancidity than those stored in glass bottles.

The clear glass and metallic bottle packaging seems to enhance the apparition of fusty attribute in comparison to the dark glass bottle packaging. Regarding musty attribute, as shown in the table, this defect is mainly due to the extraction process specifically for the super press system where it appears clearly.

4. Conclusions

From the present work, we can conclude that the genetic factor, the extraction system and the storage conditions influence significantly the physicochemical and sensorial characteristics of virgin olive oil. Coratina oils are characterized by higher amounts of 2-methyl-butanal, cis-3-hexen-1-ol, cis-3-hexenyl acetate, hexyl acetate, cis-2-pentenol, 2-pentanone, and 3-pentanone and it is noteworthy for its higher content of phenolic compounds, while Chemlali samples were richer in pentanol, trans-3-hexenol, octanal, hexanal and penten-3-ol. After storage the total phenol content decreases and, consequently, the typical bitter taste and pungent note of fresh EVOO decrease in intensity. Irrespective of the packaging used, all stored VOOs showed a considerable decrease in aromatic compound amounts.

The graphic modeling proved to be a powerful statistical technique for understanding the impact of the various factors under study on the physicochemical characteristics of virgin olive oils and determining the most influencing one. The extraction system was the determining factor for parameters such as free acidity, PV, musty attribute and Δ-tocopherol. The genetic factor was the main factor influencing the γ-tocopherol content. The storage date was the dominant factor influencing K232, β-tocopherol, volatile compounds, fruity and bitter attributes. K270 and α-tocopherol were influenced mainly by the storage date and packaging material type. The packaging material was the dominant factor that influenced pungent, rancid, and fusty attributes.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.foodres.2013.09.007.

References

Adams, R. P. (1995). *Identification of essential oil components by gas chromatography–mass spectroscopy*. Carol Stream, IL: Allured Publishing Company.

Allalout, A., Krichen, D., Methenni, K., Taamalli, A., Oueslati, I., Daoud, D., et al. (2009). Characterization of virgin olive oil from Super Intensive Spanish and Greek varieties grown in northern Tunisia. *Scientia Horticulturae*, *120*, 77–83.

Angerosa, F., & Basti, C. (2001). Olive oil volatile compounds from the lipoxygenase pathway in relation to fruit ripeness. *Italian Journal of Food Science*, *13*, 421–428.

Angerosa, F., Basti, C., Vito, R., & Lanza, B. (1999). Effect of fruit stone removal on the production of virgin olive oil volatile compounds. *Food Chemistry*, *67*, 295–299.

Aparicio, R., & Morales, M. T. (1998). Characterization of olive ripeness by green aroma compounds of virgin olive oil. *Journal of Agricultural and Food Chemistry*, *46*, 1116–1122.

Aparicio, R., Roda, L., Albi, M.A., & Gutierrez, F. (1999). Effect of various compounds on virgin olive oil stability measured by Rancimat. *Journal of Agricultural and Food Chemistry*, *47*, 4150–4155.

Baldioli, M., Servili, M., Perretti, G., & Montedoro, G. F. (1996). Antioxidant activity of tocopherols and phenolic compounds of virgin olive oil. *Journal of the American Oil Chemists' Society*, *73*, 1589–1593.

Bendini, A., Cerretani, L., Salvador, M.D., Fregapane, G., & Lercker, G. (2010). Stability of the sensory quality of virgin olive oil during storage: An overview. *Italian Food & Beverage Technology*, *LX*, 5–18.

COI Conseil Oleicole International (2011). *Analyse sensorielle de l'huile d'olive: Méthode d'évaluation organoleptique de l'huile d'olive vierge.* (COI/T.20/Doc. no 15/Rev.4 Novembre 2011).

Covas, M. I., de la Torre, K., Farre-Albaladejo, M., Kaikkonen, J., Fito, M., Lopez-Sabater, C., et al. (2006). Postprandial LDL phenolic content and LDL oxidation are modulated by olive oil phenolic compounds in humans. *Free Radical Biology & Medicine*, *40*, 608–616.

Dabbou, S., Gharbi, I., Dabbou, S., Brahmi, F., & Nakbi, A. (2011). Impact of packaging material and storage time on olive oil quality. *African Journal of Biotechnology*, *10*, 16937–16947.

Davies, N. W. (1990). Gas chromatographic retention indices of monoterpenes and sesquiterpenes on methyl silicone and carbowax 20 M phases. *Journal of Chromatography*, *503*, 1–24.

Edwards, D.M. (2000). *Introduction to graphical modelling* (2nd ed.)New York: Springer, 333.

European Economic Community, European Union Regulation EEC/2568/91 (1991). On the characteristics of olive oils and their analytical methods and later modification. *Official Journal of European Communities*, *L248*, 1–82.

Figueiredo-González, M., Martínez-Carballo, E., Cancho-Grande, B., Santiago, J. L., Martínez, M. C., & Simal-Gándara, J. (2012a). Pattern recognition of three *Vitis vinifera* L. red grapes varieties based on anthocyanin and flavonol fingerprints, with correlations between their biosynthesis pathways. *Food Chemistry*, *130*, 9–19.

Figueiredo-González, M., Simal-Gándara, J., Boso Alonso, S., Martínez Rodríguez, M. C., Santiago Blanco, J. L., & Cancho-Grande, B. (2012b). Flavonoids in Gran Negro berries collected from shoulders and tips within the cluster, and comparison with Brancellao and Mouratón varieties. *Food Chemistry*, *133*, 806–815.

Fitó, M., Cladellas, M., de la Torre, R., Martí, J., Muñoz, D., Schröder, H., et al. (2008). *European Journal of Clinical Nutrition*, *62*, 570–574.

Frankel, E. N. (1985). Chemistry of autoxidation: Mechanism, products and flavor significance. In D. B. Min, & T. H. Smouse (Eds.), *Flavor chemistry of fats and oils* (pp. 1–37). Champaign, IL (USA): AOCS Press.

Gimeno, E., Castellote, A. I., Lamuela-Raventos, R. M., De La Torre, M. C., & Lopez-Sabater, M. C. (2000). Rapid determination of vitamin E in vegetable oils by reversed-phase high-performance liquid chromatography. *Journal of Chromatography A*, *881*, 251–254.

Gutierrez, F., & Fernandez, J. L. (2002). Determinant parameters and components in the storage of virgin olive oil. Prediction of storage time beyond which the oil is no longer of “extra” quality. *Journal of Agricultural and Food Chemistry*, *50*, 571–577.

Gutierrez, F., Villafranca, M. J., & Castellano, J. M. (2002). Changes in the main components and quality indices of virgin olive oil during oxidation. *Journal of the American Oil Chemists' Society*, *79*, 669–676.

Jemai, H., Bouaziz, M., Feki, I., El Fki, A., & Sayadi, S. (2008). Hypolipidimic and antioxidant activities of oleuropein and its hydrolysis derivative-rich extracts from Chemlali olive leaves. *Chemical and Biological Interactions*, *176*, 88–98.

Jennings, W., & Shibanoto, T. (1982). *Qualitative analysis of flavor and fragrance volatiles by glass capillary chromatography*. New York: Academic Press.

Kalua, C. M., Allen, M. S., Bedgood, D. R., Bishop, A. G., Prenzler, P. D., & Robards, K. (2007). Olive oil volatile compounds, flavour development and quality: A critical review. *Food Chemistry*, *100*, 273–286.

Kiritsakis, A. K. (1998). Flavor components of olive oil — A review. *Journal of the American Oil Chemists' Society*, *75*, 673–681.

Koprivnjak, O., Procida, G., & Zelinotti, T. (2000). Changes in the volatile components of virgin olive oil during fruit storage in aqueous media. *Food Chemistry*, *70*(3), 377–384.

Krichene, D., Allalout, A., Mancebo-campos, V., Salvador, M.D., Zarrouk, M., & Fregapane, G. (2010). Stability of virgin olive oil and behaviour of its natural antioxidants under medium temperature accelerated storage conditions. *Food Chemistry*, *121*, 171–177.

Lauritzen, S. L. (1996). *Graphical models*. Oxford: Clarendon Press (298 pp.).

Massada, Y. (1976). *Analysis of essential oils by gas chromatography and mass spectrometry*. New York: Journal Wiley and Sons.

Mc Geer, P. L., Mc Geer, E. G., & Schwab, C. (2009). Inflammatory processes exacerbate degenerative neurological disorders. *Current Hypotheses and Research in Alzheimer's Disease*, *4*, 117–124.

Montedoro, G. F., Servili, M., Baldioli, M., Selvaggini, R., Maniati, E., & Macchioni, A. (1993). Simple and hydrolyzable compounds in virgin olive oil. 3: Spectroscopic characterization of the secoiridoids derivatives. *Journal of Agricultural and Food Chemistry*, *41*, 2228–2234.

Morales, M. T., Rios, J. J., & Aparicio, R. (1997). Changes in the volatile composition of virgin olive oil during oxidation: Flavors and off-flavors. *Journal of Agricultural and Food Chemistry*, *45*, 2666–2673.

Morelló, J. R., Motilva, M. J., Tovar, M. J., & Romero, M. P. (2004). Changes in commercial virgin olive oil (cv Arbequina) during storage, with special emphasis on the phenolic fraction. *Food Chemistry*, *85*, 357–364.

Pérez-Gregorio, M. R., Regueirob, J., Simal-Gándara, J., Rodrigues, A. S., & Almeida, D. P. F. (2013). Increasing the added-value of onions as a source of antioxidant flavonoids: A critical review. *Critical Reviews in Food Science and Nutrition*. http://dx.doi.org/10.1080/10408398.2011.624283.

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Pérez-Lamela, C., García-Falcón, M. S., Simal-Gándara, J., & Orriols-Fernández, I. (2007). Influence of grape variety, vine system and enological treatments on the colour stability of young red wines. *Food Chemistry*, *101*, 601–606.

Rababah, T. M., Feng, H., Yang, W., Eriefej, K., & Al-omoush, M. (2011). Effects of type of packaging material on physicochemical and sensory properties of olive oil. *International Journal of Agricultural & Biological Engineering*, *4*, 66–72.

Rastrelli, L., Passi, S., Ippolito, F., Vacca, G., & De Simone, F. (2002). Rate of degradation of alpha-tocopherol, squalene, phenolics, and polyunsaturated fatty acids in olive oil during different storage conditions. *Journal of Agricultural and Food Chemistry*, *50*, 5566.

Rodrigues, A. S., Pérez-Gregorio, M. R., Garcia-Falcon, M. S., & Simal-Gandara, J. (2009). Effect of curing and cooking on flavonols and anthocyanins in traditional varieties of onion bulbs. *Food Research International*, *42*, 1331–1336.

Rodrigues, A. S., Pérez-Gregorio, M. R., García-Falcón, M. S., Simal-Gándarab, J., & Almeida, D. P. F. (2011). Effect of meteorological conditions on antioxidant flavonoids in Portuguese cultivars of white and red onions. *Food Chemistry*, *124*, 303–308.

Servili, M., & Montedoro, J. F. (2002). Contribution of phenolic compounds to virgin olive oil quality. *European Journal of Lipid Science and Technology*, *104*, 602–613.

Torres, M. M., & Maestri, D.M. (2006a). Chemical composition of Arbequina virgin olive oil in relation to extraction and storage conditions. *Journal of the Science of Food and Agriculture*, *86*, 2311–2317.

Torres, M. M., & Maestri, D.M. (2006b). The effects of genotype and extraction methods on chemical composition of virgin olive oils from Traslasierra Valley (Córdoba, Argentina). *Food Chemistry*, *96*, 507–511.

Vacca, V., Del Caro, A., Poiana, M., & Piga, A. (2006). Effect of storage period and exposure conditions on the quality of Bosana extra-virgin olive oil. *Journal of Food Quality*, *29*(2), 139–150.

Velasco, J., & Dobarganes, C. (2002). Oxidative stability of virgin olive oil. *European Journal of Lipid Science and Technology*, *104*, 661–676.

Vichi, S., Pizzale, L., Conte, L. S., Buxaderas, S., & Lopez-Tamames, E. (2003). Solid-phase microextraction in the analysis of virgin olive oil volatile fraction: Characterization of virgin olive oils from two distinct geographical areas of northern Italy. *Journal of Agricultural and Food Chemistry*, *51*, 6572–6577.

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