

# Exceptional long-term durability of Coratina monovarietal extra virgin olive oil evaluated through chemical parameters and oxidative stability test

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**Abstract** – Coratina is a very popular olive cultivar, native of the Apulian region (Italy), but today worldwide cultivated and appreciated. In the present study, durability of Coratina monovarietal extra virgin olive oils (CMOO), produced in the Molise region (Southern Italy), was investigated up to 6-years storage in metal tin cans, under room temperature and darkness. Durability was considered the storage time in which an oil remained food grade. Yearly, the free fatty acids, peroxide value, K232 and K270 indices, fatty acid composition, diacylglycerols, phenolic profile and Rancimat induction time at 130 °C were determined on the oil. Free fatty acids and peroxide value increased linearly over time but never surpassing the European limits set for the EVOO category. Conversely, K232 and K270 exceeded the EVOO limits after five and four years, respectively. A linear decrease of phenolic compounds was observed with direct effect on the oil oxidative stability measured by Rancimat test. In conclusion, despite some signs of aging, the CMOO highlighted a remarkable long durability being food-grade up to 6-years.

**Keywords:** monovarietal extra virgin olive oil / Coratina / shelf life / durability / oil quality

**Résumé** – Durabilité exceptionnelle à long terme de l'huile d'olive extra vierge monovariétale Coratina évaluée par des paramètres chimiques et un test de stabilité oxydative. Coratina est un cultivar d'olive très populaire, originaire de la région des Pouilles (Italie), mais aujourd'hui mondialement cultivé et apprécié. Dans la présente étude, la durabilité des huiles d'olive extra vierges monovariétales Coratina (CMOO), produites dans la région de Molise (Italie du Sud), a été étudiée jusqu'à 6 ans de stockage dans des boîtes métalliques, à température ambiante et à l'obscurité. La durabilité a été considérée comme le temps de stockage pendant lequel une huile reste de qualité alimentaire. Chaque année, les acides gras libres, l'indice de peroxyde, les indices K232 et K270, la composition en acides gras, les diacylglycérols, le profil phénolique et le temps d'induction (test de Rancimat) à 130 °C ont été déterminés sur l'huile. Les acides gras libres et l'indice de peroxyde ont augmenté linéairement au fil du temps, sans jamais dépasser les limites européennes fixées pour la catégorie EVOO. En revanche, les huiles K232 et K270 ont dépassé les limites de l'EVOO après respectivement cinq et quatre ans. Une diminution linéaire des composés phénoliques a été observée avec un effet direct sur la stabilité oxydative de l'huile mesurée par le test Rancimat. En conclusion, malgré certains signes de vieillissement, l'huile CMOO a fait preuve d'une remarquable longévité, étant de qualité alimentaire jusqu'à 6 ans.

**Mots clés :** huile d'olive extra vierge monovariétale / Coratina / durée de conservation / durabilité / qualité de l'huile

## 1 Introduction

Recently, demand of extra virgin olive oil (EVOO) obtained from olives of a single variety (called “monovarietal oil”) is growing rapidly on the market, also promoted by

dedicated events and exhibitions (Rotondi *et al.*, 2013). Moreover, all the EVOO with Protect Designation Origin (PDO) or Protect Geographical Indication (PGI) is based fundamentally on the local genetic biodiversity of olive cultivar. In this perspective, numerous studies are carried out to highlight the distinctive and particular features of specific monovarietal oils (Luna *et al.*, 2006; Rotondi *et al.*, 2010, 2013; Shendi *et al.*, 2020b; Squeo *et al.*, 2021).

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Coratina is an autochthonous olive cultivar of the Apulian region (Southern Italy) growing principally in the province of Bari, especially in the municipalities of Andria, Barletta and Corato (hence its name). Coratina's trees have a medium vigor and inflorescence shaped typically at cluster, hence the vernacular name of “*olivo a raciuoppe of Corato*”, where “*raciuoppe*” is synonymous with cluster (Caruso, 1883; Patanelli, 1937). Coratina's trees are characterized by good adaptability to different pedological and climatic conditions, high and constant productivity and late fruit ripening. Coratina olives are characterized by uneven colour, an average weight of around 4.7 g and good oil content up to 27%.

At the beginning of the twentieth century, the Coratina monovarietal olive oil (CMOO) was reputed only as an Apulian local oil (known also with the name of “Andria's oil”) because commercialized locally as salad oil or in blends with refined oils (Vossen, 2007). Nowadays, Coratina is one of the main known Italian olive varieties and it is permitted in the production of all the POD/PGI Apulian EVOOs, mainly in the POD “Terra di Bari”. Furthermore, over the years, the interest for this cultivar is expanded globally to point that Coratina is today considered one of the most influential olive oil varieties of the World, together with “Picual”, “Koroneiki”, “Arbequina”, “Frantoio”, and “Leccino” (OLEADB, <http://www.oleadb.it>; Zarruk *et al.*, 2009). The growing interest in spreading Coratina cultivar is attested also by numerous studies in which the CMOOs produced in several parts of the world have been characterized (Fig. 1) (Mailer *et al.*, 2010; Benincasa *et al.*, 2011; Verma *et al.*, 2012; Costa, 2014; Dabbou *et al.*, 2015; Hassanein *et al.*, 2016; Irigaray *et al.*, 2016; Bruscatto *et al.*, 2017; Ceci *et al.*, 2017; Fuentes *et al.*, 2018; Yu *et al.*, 2021).

CMOO produced in Italy is characterized by high level of oleic acid and phenolic compounds and, as concern the organoleptic profile, by a marked bitter and pungent taste and a medium-intense level of green fruity, with predominant scent of fresh almond and slight sensations of grass/leaf and artichoke (Oli Monovarietali Italiani, <http://www.olimonoar.itali.it>; Gómez *et al.*, 2007).

Although the terms “shelf life” and “durability” are considered commonly as synonymous, we have deliberately used them with a different meaning in the present study. Specifically, we used the term “shelf-life” to indicate the storage period in which an EVOO maintains all its legal requirements to be marketed into the “extra-virgin” category. Conversely, we used the term “durability” to indicate the storage period in which the oil remains food grade or suitable for consumption, despite the loss of some requirements fixed for its original commercial category. Generally, food is discarded when its shelf life expired. Long oil durability is undoubtedly a valuable feature for both producers and consumers due to the declassified-but-still-edible oil could be recycled instead of be discarded. Therefore, the evaluation of durability meets the growing demand of sustainability and reduction of food waste.

Oxidation is the main process affecting both the EVOO shelf life and durability by causing rancidity, unpleasant smell, formation of toxic compound and nutritional loss (Frankel, 2010). EVOO oxidative stability depends on several factors, such as the oil composition (levels of unsaturated fatty acids

and antioxidants) and the storage conditions (temperature; exposure to air and light; packaging) (Morello *et al.*, 2004; Krichene *et al.*, 2010; Houshia *et al.*, 2019).

Generally, the studies on the EVOO shelf life are based on an observation period of maximum 24 months (Fadda *et al.*, 2012; Ben-Hassine *et al.*, 2013; Shendi *et al.*, 2020a; De Leonardis *et al.*, 2021). In this study, the changes of some chemical parameters and oxidative stability occurred in CMOO during a very long storage period (up to 6-years) were evaluated. We have chosen to package the oil in tin can because these is nowadays one of the containers more currently used for the EVOO. The tin cans were stored at room temperature in darkness by measuring yearly the free fatty acids, peroxide value, K232 and K270 indices, fatty acid composition, diacylglycerols, phenolic profile and Rancimat induction time at 130 °C.

## 2 Materials and methods

### 2.1 Materials and sampling design

The oil samples used for this study were two independent oils, named C1 and C2, produced in October 2015 in the Molise region (Southern Italy). In detail, the C1 and C2 oils have been produced from distinct batch of Coratina olives, harvested and worked in 2-phase olive mills located in the Guglionesi and Campomarino countryside, respectively.

Each oil (3 L) was packaged in 3-liter tin cans, in such a container number as to guarantee an annual triplicate independent sampling. The tin cans were stored in a dark room at environmental temperature (ranging from 18 to 24 °C as derived from an electronic temperature sensor located in storage room). Yearly, for a total of 6 years, in the first days of October, three tin cans for each sample were opened to analyze independently the oil; the residual oil was discarded.

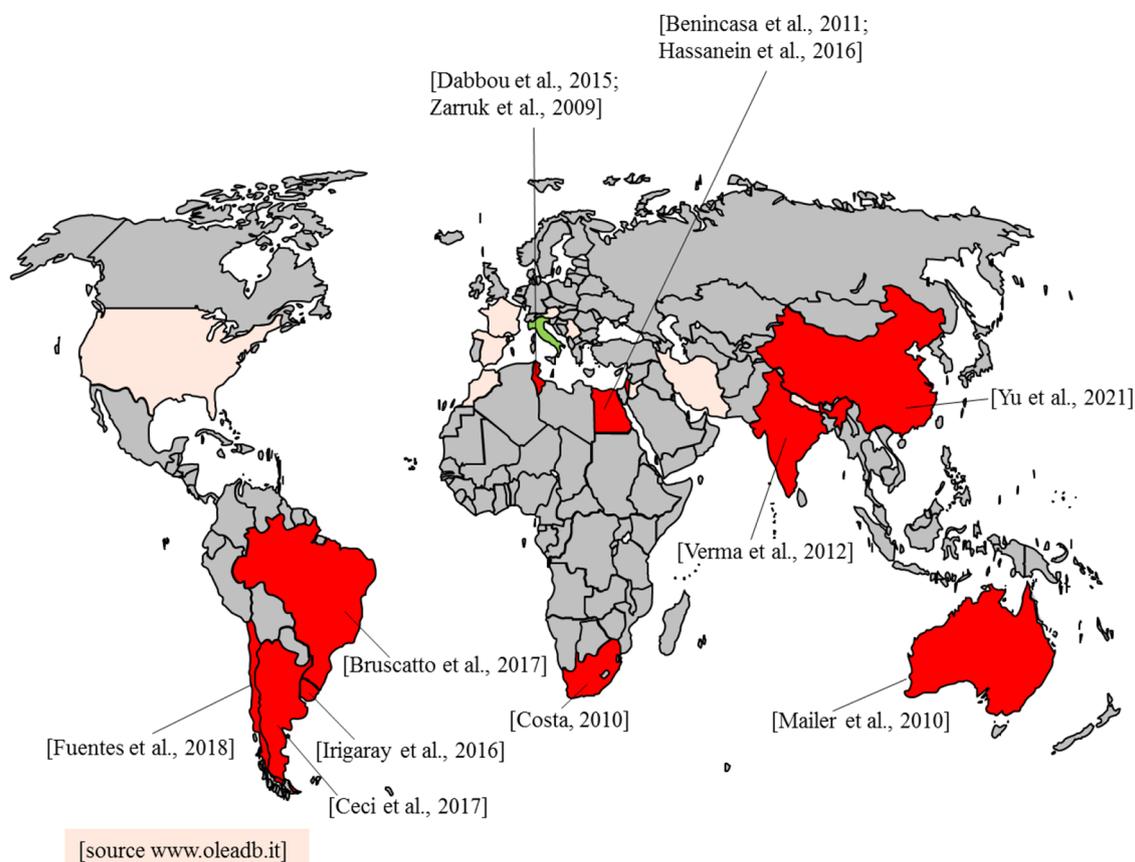
All reagents used were of analytical grade or HPLC grade. The gallic acid and hydroxytyrosol standards were purchased from Sigma-Aldrich Co (St. Louis, MO, USA).

### 2.2 Analytical methods

Free fatty acids (FFA, as percentage of oleic acid), peroxide value (PV, as meq O<sub>2</sub>/kg) and the spectrophotometric K232 and K270 indices (as absorbance of 1%, w/v, oil-iso-octane solution measured at 232 nm and 270 nm) were determined by the methods described by the Regulation EEC/2568/91 of the European Union Commission [EEC 2568/91].

Percentage of fatty acid composition and diacylglycerols were determined by gas chromatography TRACE 1300 (Thermo Fisher Scientific SpA, Rodano, MI, Italy) equipped with a flame ionization detector. Alltech EC-1000 FFAP (Alltech, USA) (30 m × 0.32 mm i.d.; 0.25 μm film) and Restek Rtx-65TG (30 m × 0.32 mm i.d.; 0.10 μm film) capillary columns were used for the analysis of fatty acids and diacylglycerols, respectively. The preparation of samples and the instrumental operative conditions were carried out according to the method described by our previous report (De Leonardis *et al.*, 2017).

Phenolic compounds were three times extracted by dissolving about 6 g oil in a mixture of 80% methanol-water (v/v)



**Fig. 1.** Countries in which there is evidence from the literature of the production of Coratina extra virgin olive oil.

(De Leonardis *et al.*, 2021). At the end of extraction, the obtained dried phenolic extract was dissolved in 2 mL of 80% methanol-water (v/v) solvent and filtered by a syringe filter (0.45  $\mu\text{m}$  PTFE filter). Total phenol (TP) was measured spectrophotometrically with an Evolution<sup>TM</sup> 201/220 UV-Visible Spectrophotometer (Thermo Fisher Scientific SpA, Rodano, MI, Italy) by using the Folin–Ciocalteu reagent and quantifying phenols through a gallic acid standard calibration curve (mg/kg oil as gallic acid equivalent, GAE). Phenolic profile was determined according to the International Olive Oil Council method (IOC, 2009) according the HPLC method described by our previous report (De Leonardis *et al.*, 2021). The quantification of phenolic compounds was carried out at 280 nm by using an external hydroxytyrosol (Hy) standard calibration curve by expressing the concentration as mg/kg oil as hydroxytyrosol equivalent (HyE).

According to Macciola *et al.* (2020) the oxidative stability was measured as the induction time (hour) determined by the Rancimat Instrument Mod.730 (Methrom, Herisau, Switzerland) at 130 °C and 20 L/h air flow.

### 2.3 Statistical analysis

Statistical analysis was performed by the software IBM SPSS statistics for Windows version 26 (IBM Corp., Armonk, NY, USA). Generally, the data are presented as the mean  $\pm$  standard deviation (SD) of three independent determinations and analyzed by one-way analysis of variance (ANOVA) and

Tukey's test by highlighting the difference statistically significant with  $p \leq 0.05$ .

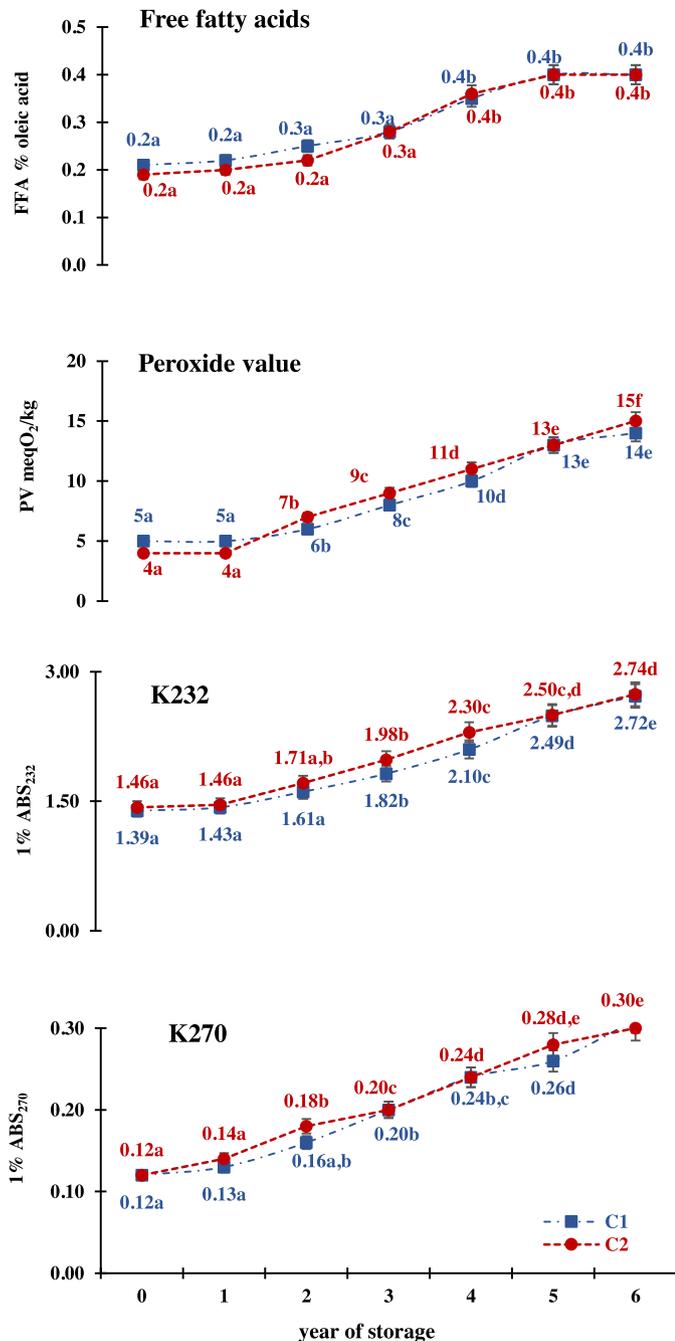
## 3 Results and discussion

### 3.1 Basic qualitative parameters

Commonly, a shelf life of 12–18 months from the bottling is recommended for the EVOO (De Leonardis *et al.*, 2021). Actually, the effective end of the shelf life is signed by the overcoming of some legal indices fixed for each commercial category of olive oil. The principal quality limits for the EVOO commercial category are: free fatty acids  $\leq 0.8$  g oleic acid/100 g oil; peroxide value  $\leq 20$  meq  $\text{O}_2$ /kg; UV indices K232  $\leq 2.50$  and K270  $\leq 0.22$ ; fruity median  $> 0$  and defect median = 0 by Panel Test [EEC 2568/91; EEC 1348/2013]. All these parameters have been monitored in this study, with exception of the Panel Test that unfortunately was not made. However, we can refer to Baiano *et al.* (2014) that found in CMOO significant loss of median fruity and presence of defect after 7-years storage in dark glass bottles.

The changes of free fatty acids (FFA), peroxide value (PV), K232 and K270 UV-indices observed during the storage of the CMOOs are given in Figure 2.

At the beginning of the study, both the oils had basic qualitative parameters very similar and well below the above-mentioned EVOO category limits. This high initial quality was



**Fig. 2.** Changes of the basic quality indices of the Coratina monovarietal extra virgin olive oils monitored yearly during the storage period (six years) Data represent mean and SD (n = 3) Values with different letters for each oil sample are significantly different at  $P \leq 0.05$ .

a crucial requirement for the purposes of this study and indicated that both the oils had been obtained from olives of good quality and through a good performing extraction process (Gambacorta *et al.*, 2010).

Evolution of the basic indicator during the storage period followed a similar increasing trend between the oil samples. By extrapolating only the data relative to FFA and PV, both the

oils could be classified as “extra-virgin-olive-oil” up to the sixth year of storage. In both C1 and C2 samples, the free fatty acids content (Fig. 2) was doubled at 6-years storage going from 0.2 to 0.4%. By looking better, acidity remained substantially unchanged until the third year of storage, showing a progressive slow increasing in the following years. In the same period of observation, the PV (Fig. 2) reached the final value of 14 and 15 meq O<sub>2</sub>/kg in C1 and C2, respectively. Thus, starting from the third year the PV in both oils increased progressively although remaining below the limit of 20 meq O<sub>2</sub>/kg up to the sixth year.

Conversely, taking into consideration only the UV indices, both the oils surpassed the K232 limit set for the EVOO category (2.50) in the fifth year of storage, while the K270 limit (0.22) was passed in the year before. Notoriously, K232 measure the formation of conjugated dienes, while K270 the secondary oxidation products. Several factor may affect the evolution of these indices in the oil, such as the exposure to air, sunlight and heat (Houshia *et al.*, 2019). It is reasonable suppose that the packaging chosen in this study could have affected the K232 and K270 increasing. Indeed, Baiano *et al.* (2014) found a slower increasing of K232 and K270 by using dark glass bottles. Moreover, the tin cans may not be completely filled for practical (difficulty in pouring the oil when the container is too full) and physical (variation of the oil volume with the temperature) issues, so leaving unavoidable headspace air on the oil (De Leonardis *et al.*, 2021). To be more precise, the used tin cans (TC) have dimensions of  $15 \times 9.2 \times 26$  (height) corresponding to a potential volume of 3,588 cm<sup>3</sup> and a real capacity of 3,000 cm<sup>3</sup> (as declared by the manufacturer); therefore, the headspace air on the oil could be estimated of 0.588 cm<sup>3</sup>. Moreover, in this work, the tin can headspace was not filled with nitrogen as the commercial companies of olive oil do usually.

By summarizing data of Figure 2, durability of CMOO must be considered a remarkable outcome despite the static presence of air in the container, and also by considering the very long storage of this study. To our knowledge, there are no in the literature studies conducted under conditions similar to ours and for such a long time, apart from Baiano *et al.* (2014) that found comparable outcomes for CMOO stored in 1-L dark glass bottles.

### 3.2 Fatty acid composition

In Table 1, the fatty acid composition of the studied CMOOs determined at the beginning and at the end of the storage period (6-years) is given.

Fatty acid profile of both the CMOOs was characterized by three more abundant fatty acids, namely the oleic acid, with level up to 78%, followed by the palmitic acid, around 10%, and the linoleic acid, around 6.5%. Moreover, linolenic acid was found around 0.6%. According to the literature, the above-mentioned fatty acid profile was typically that of CMOOs produced in Italy (Rotondi *et al.*, 2010; <http://www.wolimono-varietali.it>; Baiano *et al.*, 2014; Deiana *et al.*, 2019). Similar fatty acid composition was found in CMOOs produced in Chile (Fuentes *et al.*, 2018), whereas, higher percentage of the palmitic and linoleic acids and lower level of oleic acid have been found in CMOOs produced in other countries due to the

**Table 1.** Fatty acid composition of the Coratina monovarietal extra virgin olive oils at the beginning and end of the storage period. Data represent the mean and SD (n = 3). No statistically significant differences were found by ANOVA analysis.

Fatty acid (%)	Time zero %	C1		C2	
		At 6 years-storage	Time zero	At 6 years-storage	
C16:0	10.60 ± 0.30	10.15 ± 0.29	10.48 ± 0.30	10.66 ± 0.27	
C16:1	0.44 ± 0.08	0.42 ± 0.07	0.47 ± 0.05	0.48 ± 0.06	
C17:0	0.05 ± 0.01	0.04 ± 0.00	0.04 ± 0.00	0.04 ± 0.00	
C17:1	0.07 ± 0.00	0.06 ± 0.00	0.07 ± 0.00	0.07 ± 0.00	
C18:0	2.28 ± 0.12	2.19 ± 0.19	2.21 ± 0.20	2.00 ± 0.17	
C18:1	78.24 ± 1.25	78.98 ± 1.08	78.60 ± 1.66	78.20 ± 1.34	
C18:2	6.64 ± 0.77	6.57 ± 0.65	6.51 ± 0.59	7.14 ± 0.91	
C18:3	0.66 ± 0.21	0.63 ± 0.18	0.65 ± 0.20	0.57 ± 0.15	
C20:0	0.41 ± 0.10	0.39 ± 0.09	0.40 ± 0.07	0.34 ± 0.06	
C20:1	0.42 ± 0.12	0.39 ± 0.08	0.39 ± 0.07	0.35 ± 0.08	
C22:0	0.12 ± 0.06	0.11 ± 0.05	0.12 ± 0.02	0.09 ± 0.01	
C24:0	0.06 ± 0.00	0.05 ± 0.00	0.05 ± 0.00	0.04 ± 0.00	

influence of different bioclimatic and geographical conditions (Mailer *et al.*, 2010; Hassanein *et al.*, 2016; Bruscatto *et al.*, 2017).

As data of Table 1 show, fatty acid composition of both the oils unchanged in 6-years of storages confirming the stable nutritional value of the CMOO. This conclusion was confirmed also by the ANOVA analysis due to no significant differences emerged. In particular, it should be noted that there was no significant decrease in linolenic acid that was the fatty acid at a higher unsaturation level and so more susceptible to oxidation.

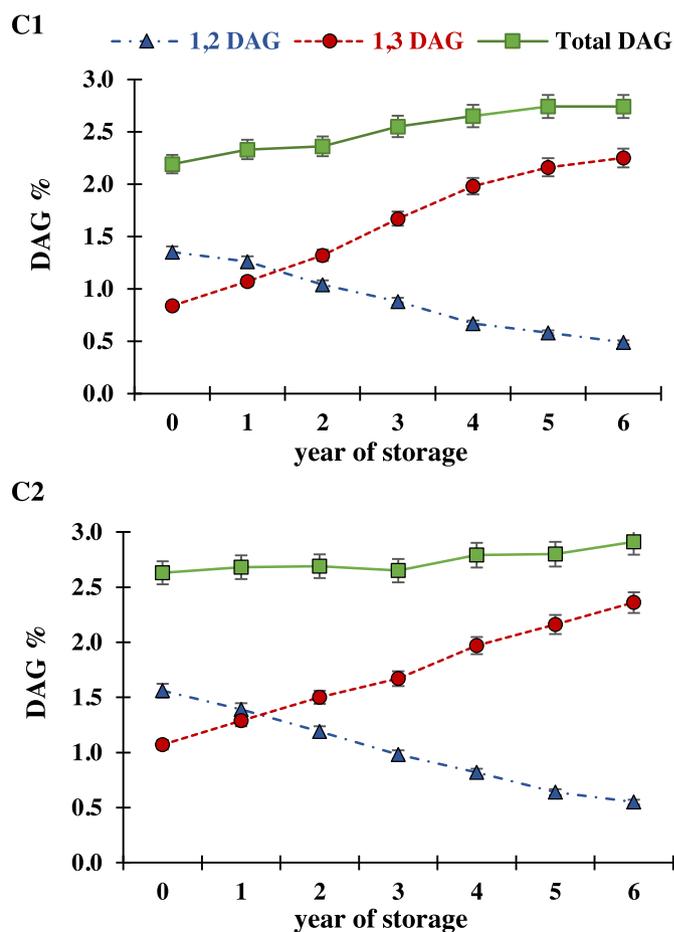
### 3.3 Diacylglycerols

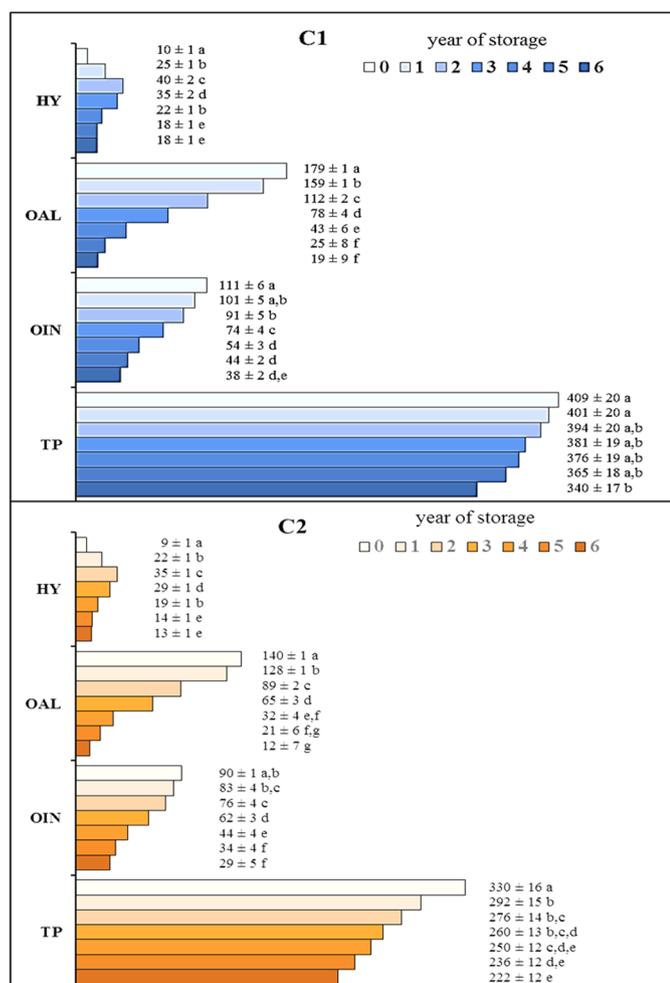
Diacylglycerols (DAG) showed significant variations in the oils during the storage, as it is shown in Figure 3.

Initially, total DAG content was 2.2 and 2.6% in the C1 and C2, respectively. Generally, total DAG are present in EVOO in amounts of 1–3%, as the sum of the 1,2 DAG and 1,3 DAG isomers. The 1,2 DAG derive from the biosynthesis of triacylglycerols, while the 1,3 DAG are generated by the enzymatic or chemical hydrolysis that may occurs before, during and after the oil extraction process (Amelotti *et al.*, 1989).

A general slight increasing of DAG, although never statistically significant, was observed during the 6-years storage as result of a very slow hydrolytic degradation process, correlated certainly with the FFA increase (Fig. 2). The observed low hydrolytic alteration was certainly another remarkable outcome proving the outstanding durability of CMOO.

As the graphs of Figure 3 show, at beginning of the storage, the 1,2 DAG isomers were on average up to 70% on the total DAG in both the oils, while an isomerization of the 1,2 DAG in 1,3 DAG occurred during the oil storage. Specifically, a significant 1,3 DAG increase was observed already in the

**Fig. 3.** Changes of total and isomer diacylglycerols (DAG) content of the monovarietal Coratina extra virgin olive oils monitored yearly during the storage period. Data represent the mean and DS (n = 3).

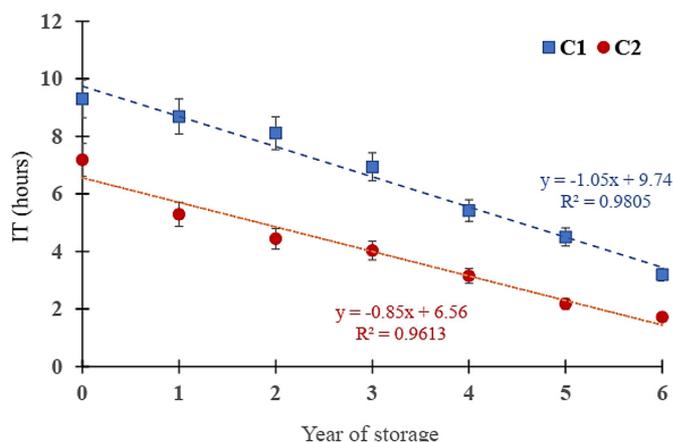


**Fig. 4.** Phenol compound changes occurred in the Coratina monovarietal extra virgin olive oils during the storage period. Labels represent the mean and SD ( $n = 3$ ). Values with different letters for each category of compounds are significantly different at  $P \leq 0.05$ . Hy: hydroxytyrosol; OAL: oleocanthal; OIN: oleacin; TP: total phenols.

second year of storage. It is known that 1,2-DAG isomers turn into the thermodynamically more stable 1,3-DAG isomers during the EVOO storage (Cossignani *et al.*, 2007). Therefore, the formation of 1,3 DAG or the variation of 1,3/1,2-DAG ratio could be used as an index of the EVOO age (Fronimaki *et al.*, 2002). However, there are no evidences of correlated effects between the increasing of 1,3/1,2-DAG ratio and the sensory or nutritional characteristics of the oil (Amelotti *et al.*, 1989; Fronimaki *et al.*, 2002; Cossignani *et al.*, 2007).

### 3.4 Phenolic fraction

Commonly, EVOO total phenols range between 50 and 1,000 mg/kg depending on the cultivar, fruit's ripeness, oil extraction process and oil storage conditions. Coratina olive cultivar is generally characterized by a high content of polyphenols composed mainly from secoroides, enclosed



**Fig. 5.** Induction time (hours) of the Coratina monovarietal extra virgin olive oil samples measured yearly by Rancimat test (130 °C) up to 6-years storage. Data represent the mean and SD ( $n = 3$ ) The drawn line is the linear regression line.

oleacin

(3,4-DHPEA-EDA, dialdehydic form of decarboxymethyl oleuropein aglycon), oleocanthal (p-HPEA-EDA, dialdehydic form of decarboxymethyl ligstroside aglycon); aglycon form of oleuropein (3,4-DHPEA-EA) and ligstroside (p-HPEA-EA); lignans, especially acetoxypinoresinol; phenolic alcohols as hydroxytyrosol and tyrosol (Caponio *et al.*, 2001).

Unfortunately, an accurate comparison of the phenolic profile of the CMOOs produced around the world is very problematic due to the different method of extraction, identification and unit of measurement used by the researchers to quantify the phenolic compounds. However, based on literature data, the CMOOs produced in the Mediterranean regions have a polyphenol content ranging usually from 300 to 600 mg/kg GAE, which is however still higher than that of CMOOs produced in non-Mediterranean countries (Mailer *et al.*, 2010; Benincasa *et al.*, 2011; Verma *et al.*, 2012; Costa, 2014; Dabbou *et al.*, 2015; Hassanein *et al.*, 2016; Irigaray *et al.*, 2016; Bruscatto *et al.*, 2017; Ceci *et al.*, 2017; Fuentes *et al.*, 2018; Yu *et al.*, 2021).

Changes of the total phenols (TP) and of the determined single phenols, occurred in the oils during the 6-years of storage, are presented in Figure 4. Total phenols are expressed as gallic acid equivalent (GAE), while the concentrations of hydroxytyrosol (Hy), oleacin (OIN) and oleocanthal (OAL) are as Hy equivalent (HyE).

At the beginning of the storage, C1 showed a polyphenol content higher than C2. Specifically, TP were 409 and 330 mg/kg in C1 and C2, respectively. It is well recognized that OIN and OAL are among the major phenolic compounds present in the EVOO; these two compounds had been largely studied for their biological and sensory properties (Mateos *et al.*, 2004; Michel *et al.*, 2012; Ben-Hassine *et al.*, 2013). Both the oils samples had a concentration of OAL higher than OIN. Specifically, OIN was 111 and 90 mg/kg and OAL was 179 and 140 mg/kg in C1 and C2, respectively. Finally, at zero time, Hy and Ty had been found in traces.

There is strong evidence that the phenol compounds of EVOO change during the oil storage caused by a series of

reactions, such as oxidation; hydrolysis of complex phenols with the consequent increase of low molecular phenols, especially Hy and Ty; increase of the dialdehydic forms of decarboxymethyl oleuropein aglycone (DAFOA) and decarboxymethyl ligstroside aglycone (DAFLA) (Clodoveo *et al.*, 2007; Carrasco-Pancorbo *et al.*, 2007; Krichene *et al.*, 2010; Lozano-Sánchez *et al.*, 2013; Shendi *et al.*, 2018).

In the present study, a progressive linear diminution of TP, OIN and OAL were observed in C1 and C2. TP were reduced at the sixth year by 17% and 33% in C1 and C2, respectively. Over the long term, also a significant OAL reduction was observed. Specifically, after 6-years of storage, OAL was reduced by 89 and 91% in C1 and C2, while OIN decreased by about 66% in the same oils, respectively. OIN decomposition was expected because this compounds, being an *o*-diphenol, is more active as antioxidant and so, more easily oxidizable than OAL. Moreover, formation of Hy was evident as results of the OIN hydrolysis. Specifically, Hy increased progressively in the in the first two years and then slowly decay (Fig. 4), similarly to what was observed by Baiano *et al.* (2014). Conversely, tyrosol formation was never detected despite the drastic depletion of OAL. Thus, the observed depletion of OAL on the time needs of further investigation to understand its degradation mechanism.

### 3.5 Oxidative stability test

Rancimat test is a popular accelerated method to determine the oxidative stability of oils/fats in standardized conditions, which are a constant temperature (variable from 50 to 220 °C) and air flow (up to 20 L/h) (Macciola *et al.*, 2020). Under these stressing conditions, high amounts of volatile compounds are generated and dissolved into a distilled water vessel where the water conductivity is constantly measured. At the end, the induction period (IP) (expressed in hours) is determined as the moment in which a rapid increase of water conductivity occurs.

The induction time (IT) of the CMOOs measured yearly is given in Figure 5.

At the beginning, the IT of both oil samples was significantly higher than that found in other cultivar oil or generic EVOO (Macciola *et al.*, 2020). However, IT of C1 (9.30 h) was found higher than that of C2 (7.18 h). A significant and progressive IT decrease was observed during the long storage. Specifically, the IT was reduced at the end of storage (six years) by 65.7 and 76.0% in C1 and C2, respectively. Observed depletion of thermal oxidative stability during the storage is a relevant information for a correct recycling of long-stored oil. According to the literature (Lerma-Garcia *et al.*, 2009; Krichene *et al.*, 2010; Macciola *et al.*, 2020), a close linear correlation between phenol and IT changes emerged, as can be seen by comparing the data in Figures 3 and 4. Thus, the key role of phenols on the EVOO thermal oxidative stability was confirmed, while the role of other antioxidant compounds, such as tocopherols, although not determined in this work, should be less relevant according to what reported by Mateos *et al.* (2003).

## 4 Conclusions

Although Coratina monovarietal extra virgin olive oil (CMOO) has been largely studied in the literature, new results concerning its durability are reported in the present study.

By considering only the free fatty acids and peroxide value, it was observed that CMOO stored in tin cans, under environmental temperature and darkness, could retain in the “extra virgin olive oil” category up to 6 years. Nevertheless, some aging signs were evident since the previous years. One of these was the 1,3/1,2-DAG isomerization starting from the first year of storage. Furthermore, the K232 and K270 indices surpassed the EVOO legal limits at the fifth and fourth years, respectively, causing the oil declassing at the “virgin olive oil” category. Finally, a progressive and slow loss of phenolic substances and thermal oxidative stability were observed during the long storage period with consequent partial decline of functional, antioxidant and sensory properties of the oils. Conversely, fatty acid composition unchanged during all storage period.

In conclusion, in this study, it emerged a remarkable long durability of CMOO which could be probably also improved with a more appropriate packaging system. Comparison with similar studies of the literature has been difficult because the containers, environmental conditions and duration of oil storage were different from ours. Given that the aim of this study was not to question the principle that it is preferable to consume the EVOO in the first months from the production (when its nutritional and organoleptic potential is highest), long oil durability is undoubtedly a positive feature for the Coratina oil valorisation, and also for new possible recycled forms of declassified-but-still-edible oils, such as blending or refining. From this viewpoint, long oil durability is of sure scientific and practical interest by meeting the growing demand of sustainability and reduction of food waste.

## Conflicts of interest

The authors declare no conflict of interest.

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