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Influence of Different Filtration Systems on Chemical and Volatile Characteristics of Virgin Olive Oil

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Filtration, which eliminates suspended solids and moisture, plays a critical role in stabilising virgin olive oil (VOO) before storage. However, the effects of different filtration systems on the chemical and volatile characteristics of VOO are not well understood. This study compared the processing effects of gravity-driven and mechanical filtration systems on the oil quality and volatile properties of VOO. Chemical analyses, measuring oil quality parameters and total phenolic and pigment contents, showed higher acidic parameter values and a decrease in both total phenolic and pigment contents in oils processed with either gravity-driven or mechanical filtration systems than in unfiltered oil. This suggests that these filtration processes negatively affect the initial quality of VOO. The initial oxidation level of the mechanically filtered oil was lower than that of the oil processed using gravity-driven filtration. Electronic nose analysis, which is used to examine volatile compounds, showed that the concentrations of important volatiles with sizes less than or equal to C6, which result in the olive oil aroma, were significantly decreased by gravity-driven filtration but not by mechanical filtration. In addition, the oxidative stability test showed that both filtered oils had higher stability than that of unfiltered oil, exhibiting a positive impact on the final quality of VOO. These results suggest that mechanical filtration should be performed before storage.

1. Introduction

Olive oil, which is a core component of the Mediterranean diet, is now consumed worldwide because of its beneficial properties for human health (Preedy and Watson, 2010). Olive oil contains very high levels of monounsaturated fat and a moderate amount of vitamins A and E. Some health benefits have been associated with antioxidants, including phenolic compounds (Parkinson and Cicerale, 2016). The health claims of “olive oil polyphenols” by the European Commission Regulation (EEC, 2012) refer to the effect that phenolic compounds can protect blood lipids against oxidative stress (Tsimidou and Boskou, 2015). Extra virgin olive oil (EVOO) is the highest grade of virgin olive oil (VOO) that contains 0.80% free fatty acids (FFAs), a peroxide value (PV) of 20 meqO_2/kg, and K270 value of 0.22, which are all within the threshold values; EVOO also has a superior taste, some fruitiness, and no defined sensory defects (IOC, 2019). The characteristic aroma, taste, and colour of EVOO can be distinguished from those of other vegetable oils. The colour of EVOO is mainly related to the presence of chlorophyll and carotenoid pigments, which are responsible for producing green and yellow colouration, respectively. It plays an important role in consumer acceptance (Nielsen et al., 1998). The unique and delicate flavours of EVOO are related to the presence of several volatile compounds. Previous studies reported that aldehydes, alcohols, esters, ketones, and other compounds have been identified in good-quality olive oil (Kalua et al., 2007). These volatile compounds contribute to the distinctive organoleptic characteristics that make EVOO unique.

VOO is produced only by physical means, including washing and crushing the olive fruits, malaxing the olive paste, centrifuging to separate the oil from the olive debris and water, and decanting the oil. Suspended solids and moisture can remain in VOO after extraction and promote oil deterioration. The filtration process, which removes solids and moisture from VOO before storage, plays a critical role in contributing to the transparency, quality, and shelf-life of VOO and may have a positive, negative, or neutral impact on its stability, phenolic

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compounds, pigments, and sensory parameters (Fregapane et al., 2006). In general, the two main types of filtration systems are driven by gravity or mechanical vacuum/suction. Gravity-driven filtration is more convenient for removing suspended matter in VOO without the need for a mechanical system, allowing implementation at a low price. Although mechanical filtration is also more commonly used for this purpose because it aims to process VOO quickly and in large volumes, mechanical filtration systems have disadvantages in terms of cost. Knowledge of the differences between gravity-driven and mechanically driven filtration systems for oil treatment is limited and the impacts of the different filtration systems on the chemical and volatile characteristics of VOO are not well understood. Hence, the main objective of this study was to compare the effects of different filtration systems on the oil quality, volatile property, and oxidative stability of VOO.

2. Experimental

2.1 Materials

Olive fruits of the Olivière cultivar were harvested and processed at Clermont-l'Hérault (France). VOO was obtained using industrial processors. Fifty litres of unfiltered VOO was filtered once through a filter paper (CKP V12, Cordenons, S.p.A., Milano, Italy) by gravity-driven filtration in an open system or mechanical filtration with up to 28 psi pressure using a filtration machine (model EUR 20, MORI-TEM Srl, Tavarnelle, Florence, Italy). The oil samples were then stored in dark, closed bottles at 17 °C. Each experimental condition included three independent sets of samples.

2.2 Analytical procedures

An OxiTester (CDR, Ginestra Fiorentina, Italy) (Kamvissis et al., 2008) was used to measure the level of FFAs, PV, K270 value, and total phenolic content of the oil samples. Preliminary confirmation of the FFAs, determined using the OxiTester method, was performed by comparing the results of the oil samples over a wide range of values with those from the official analysis method (Gucci et al., 2012; Kishimoto, 2019). Oil samples were added to pre-filled cuvettes for analysis. The volume of oil used was 2.5 μL for measuring FFAs, 1–2.5 μL for PVs, 10 μL for K270 values, and 10 μL for total phenolic content.

The chlorophyll and carotenoid pigment contents, expressed as mg/kg of oil, were determined using a UV-1700 spectrophotometer (Shimadzu, Kyoto, Japan) following the method described by Mínguez-Mosquera et al. (1991) and Kishimoto (2019). One gram of the oil sample was dissolved in 10 mL isooctane. Absorbance was then detected at 670 nm for chlorophylls and 470 nm for carotenoids. The contents were calculated using the following equation:

$$\text{Chlorophylls (mg/kg)} = \frac{(A_{670} \times 10^6)}{(613 \times 100 \times d)} \tag{1}$$

$$\text{Carotenoids (mg/kg)} = \frac{(A_{470} \times 10^6)}{(2000 \times 100 \times d)} \tag{2}$$

where A is the absorbance and d is the path length of the cells (1 cm).

2.3 Flash gas chromatography electronic nose analysis of volatile compounds in VOO

To analyse the volatile compounds in the oil samples, a HERACLES II electronic nose (e-nose) (Alpha MOS, Toulouse, France) (Kishimoto, 2018) was used to examine the headspace (gas mixture) prepared in a temperature-controlled vial. This instrument was equipped with parallel nonpolar (MXT-5; 10 m length × 180 μm diameter) and polar (MXT-WAX; 10 m length × 180 μm diameter) columns to produce two chromatograms simultaneously. An HS100 auto-sampler (CTC Analysis AG; Zwingen, Switzerland) was used to automate sample incubation and injection. An alkane mixture (from *n*-hexane to *n*-hexadecane) was used to convert the retention times into Kovats indices for calibration. An aliquot of oil (2.0 g) was placed in a 20-mL vial and sealed with a magnetic cap. The vial was placed in the auto-sampler, which was then placed in a HERACLES II shaker oven and incubated for 15 min at 60 °C while shaking at 500 rpm. A syringe was used to obtain a 5 mL headspace sample, which was injected into the gas chromatograph. The oven temperature was initially 40 °C (held for 10 s) and then increased to 250 °C at 1.5 °C/s and held at this temperature for 60 s. The total separation time was 120 s. Hydrogen gas was used as the carrier gas. Data were acquired and processed using AlphaSoft software v2020 (Alpha MOS). An ArochemBase module was used to identify the volatile compounds.

2.4 Accelerated oxidative stability test of VOOs

To evaluate the oxidative stability of VOOs, the oil samples (10 mL) were placed in 10 mL glass bottles and then incubated at 40 °C for 3 months. For each set of experiments, an equal amount of oil (100 µL) was taken from the same bottle and analysed monthly.

2.5 Statistical analysis

Data are presented as the mean ± standard deviation of three replicates. The statistical significance of differences between two groups was analysed using Student's *t*-test in Microsoft Excel. Tukey–Kramer test in Microsoft Excel was used to identify significant differences among the means of multiple groups. The data were analysed by one-way analysis of variance followed by Tukey–Kramer test in Microsoft Excel. Differences were considered statistically significant at $p < 0.05$.

3. Results and Discussion

3.1 Effects of different filtration systems on chemical characteristics of VOOs

This study examined the effects of different filtration systems on VOO quality parameters. Figure 1 shows the main quality indices for olive oil as set by the International Olive Council (2019) in unfiltered and different filtered oils. FFA levels in both filtered oils increased compared with those in the unfiltered oil, indicating that both filtration processes lead to oil oxidation. PV, which is an index of the primary oxidation products of olive oil, also increased in both filtered oils. The level of FFAs and PVs in mechanically filtered oil were lower than those in oil processed using gravity-driven filtration. The difference in oil oxidation levels between the different filtration systems may occur because gravity-driven filtration requires more processing time than that required in mechanical filtration, increasing oxygen exposure time due to operation in an open system. In contrast, the spectrophotometric index K270, an index of secondary oxidation products (Malheiro et al., 2009), was higher in mechanically filtered oils than in unfiltered oils but not in oils processed using gravity-driven filtration. Kishimoto and Kashiwagi (2019) previously reported this gravity-driven filtration effect on VOO.

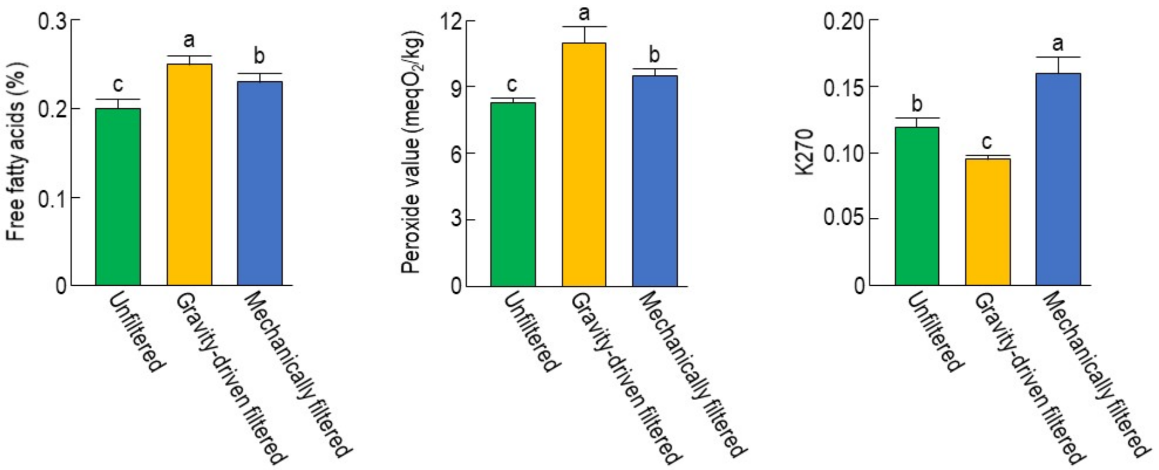

Figure 1: Oil quality assessment of VOO processed by different types of filtration. $^{a\text{-}c}$*Mean values with different letters are significantly different (p < 0.05).*

Figure 2 shows the total phenolic, carotenoid, and chlorophyll pigment contents in unfiltered oil and different filtered oils. The total phenolic content in the filtered oils was lower than that in the unfiltered oil. The carotenoid and chlorophyll pigment contents were also slightly lower in both filtered oils than in unfiltered oil. There was no difference in the phenolic and pigment contents between the different filtration systems. Taken together, although both filtration processes led to the oxidation of VOO, the mechanical filtration system may lead to milder oxidation of VOO than that by the gravity-driven filtration system.

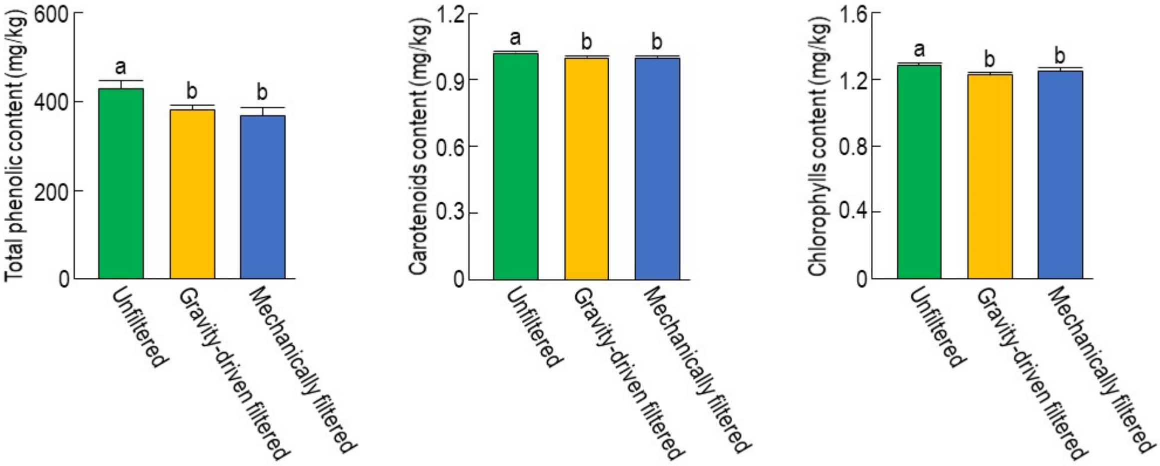

Figure 2: Total phenolic and pigment contents of VOOs processed by different types of filtration. ^{a,b}Mean values with different letters are significantly different ($p < 0.05$).

3.2 Comparison between volatile profiles from unfiltered and different types of filtered VOO

To evaluate the volatile compounds in the unfiltered and different filtered oils, an e-nose analysis was performed. Figure 3 shows a representative gas chromatogram of the headspace gases obtained from the unfiltered oil using the MXT-5 column. Twenty-two peaks were detected in the oil.

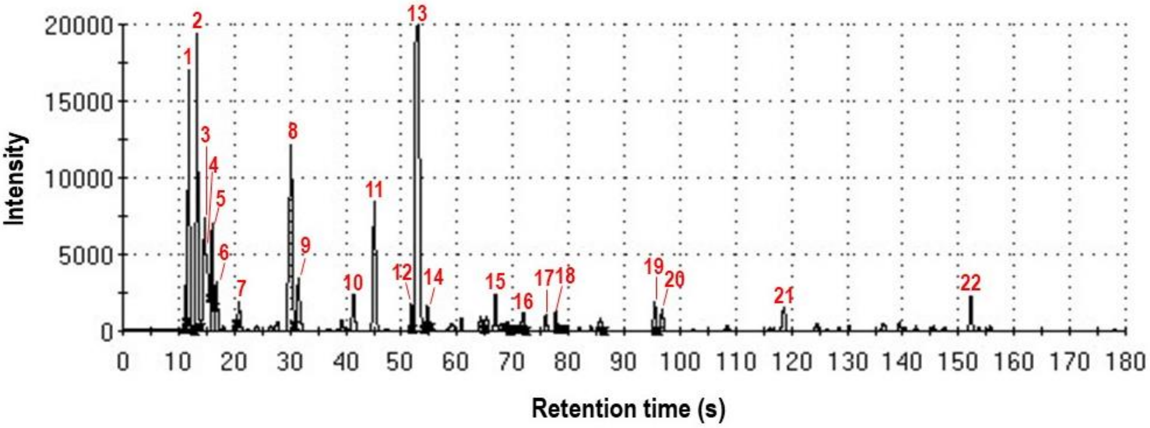

Figure 3: Raw e-nose data of volatile compounds in unfiltered VOO. The panel shows chromatogram from the MXT-5 column. The numbers show the peak numbers that were detected for unfiltered VOO.

To compare the 22 peaks shown in Figure 3 among the unfiltered and different filtered oils, the relative ratios of the peak area of both filtered to unfiltered oils were measured (Figure 4). The peak areas of the mechanically filtered oil were not significantly different from those of the unfiltered oil, indicating that there was no change in the concentrations of volatile compounds in VOO processed by mechanical filtration. Conversely, the relative ratios of peak areas 1–11 of oil processed using gravity-driven filtration were significantly lower than those of mechanically filtered oil. Peak 11, eluted at 45 s, was identified as hexanal (Kishimoto, 2021), a major volatile component of olive oil (Kiritsakis, 1998), indicating that peaks 1–11 were attributed to volatiles with sizes less than or equal to C6 (Kishimoto and Kashiwagi, 2019). Given the distinctive aroma of olive oil is attributed to C5 and C6 volatile compounds, consisting of aldehydes, alcohols, ketones, hydrocarbons, furans, and esters (Kalua et al., 2007), gravity-driven filtration can reduce olive oil aroma. A notable decrease in the concentrations of smaller-sized volatiles by gravity-driven filtration was observed, consistent with the findings of a previous report (Kishimoto and Kashiwagi, 2019). As volatility decreases with an increase in molecular weight (Bradley, 1954), smaller volatiles tend to be lost under gravity-driven filtration, which unlike mechanical filtration, is conducted in an open system, increasing the exposure time to air. These results suggest that processing with gravity-driven filtration leads to the loss of important volatile aroma compounds in VOO and that mechanical filtration can prevent this loss.

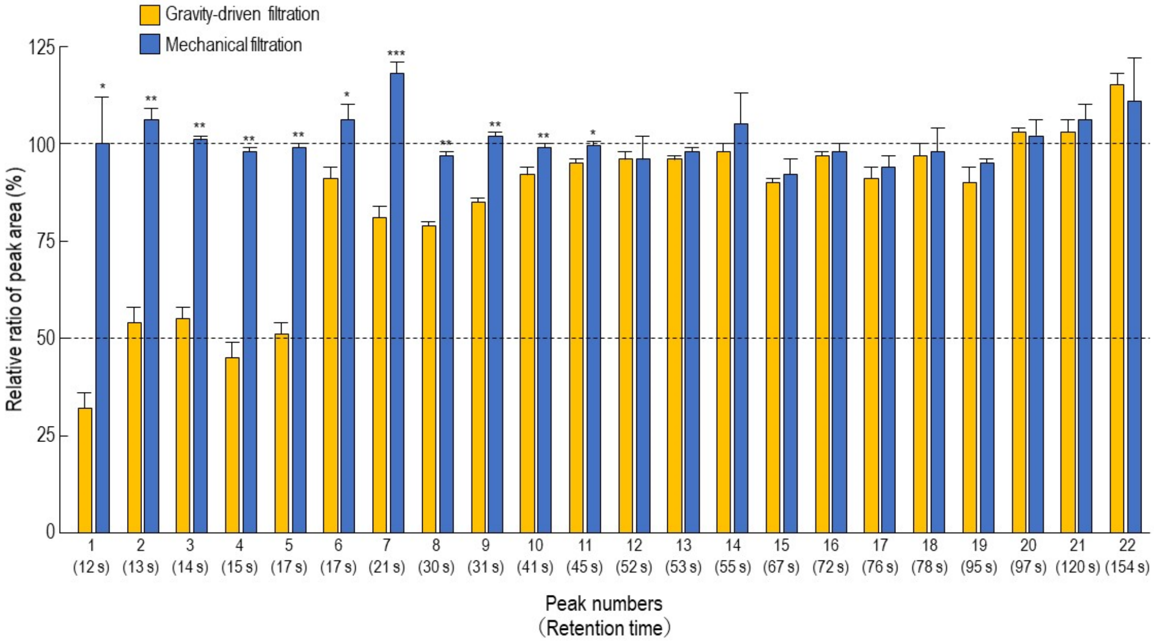

*Figure 4: Relative ratio of peak areas of different filtered VOOs to unfiltered VOO. *p < 0.05; **p < 0.01; ***p < 0.001.*

3.3 Comparison between oxidative stabilities of unfiltered and different filtered VOOs

To compare the stability of unfiltered VOO and the different filtered VOOs, accelerated oxidative stability testing of the oil samples was conducted. Figure 5 shows the changes in the FFA levels, PVs, and K270 value of the oil samples. Both filtered VOOs had higher stability than that of unfiltered VOOs, and there was no difference between gravity-driven and mechanical filtration systems. These results suggest that filtration processes have a positive impact on oil stability.

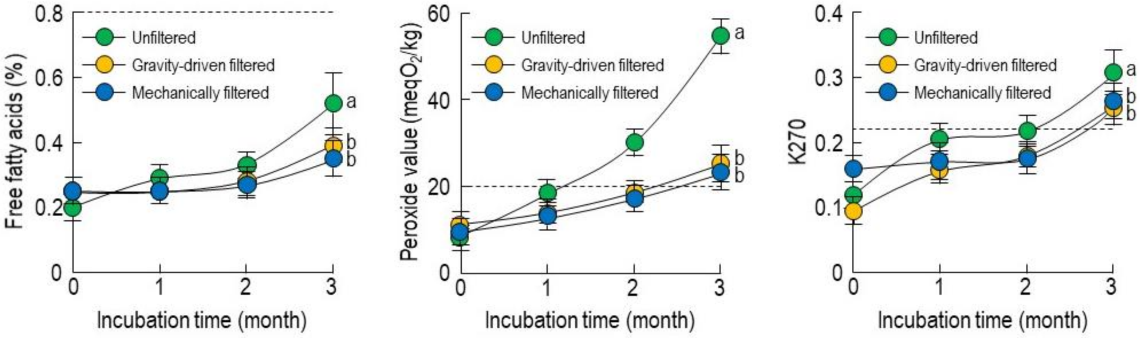

Figure 5: Accelerated oxidative stability testing of unfiltered and the different filtered VOOs. ^{a,b}Mean values with different letters are significantly different (p < 0.05). Dash lines: legal thresholds (the IOC regulation).

4. Conclusions

Chemical analyses demonstrated that compared with unfiltered oil, the initial oxidation level of VOO could be increased by processing either with gravity-driven or mechanical filtration. A comparative study between the different filtration systems showed that mechanical filtration was better than gravity-driven filtration in terms of oil quality. In addition, e-nose analysis detected 22 volatiles in the unfiltered oil, 11 of which were significantly decreased in oil processed with gravity-driven filtration but not in that processed with mechanical filtration. In particular, the volatiles with a size of C6 or less, which were significantly decreased by processing with gravity-driven filtration, may include C5–C6 volatiles such as aldehydes, alcohols, ketones, hydrocarbons, furans, and esters, which contribute to the olive oil aroma. Oxidative stability analysis showed that both filtered oils had higher stability than that of unfiltered oils. These results suggest that different types of filtration can negatively impact the initial quality of VOO yet positively impact the final product, and conducting mechanical filtration can achieve a longer shelf life. This indicates that mechanical driven filtration is better in order to remove particles that causes deterioration of oil quality from VOOs without loss of chemical and characteristics of the oils.

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