



Determination of Effects Commercial Antioxidant and Essential Oil Additives on Some Physico-Chemical Properties of Olive Oil

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Abstract

In this study, the effects of several spice essential oil and some constituents on the oxidative stability of olive oil at 0.1% level of essential oils at 60 °C was determined. Free fatty acid values of olive oil with different additives along at first-fourth weeks were changed between 1.61-2.01 %, 1.81-2.17 %, 1.67-2.23 %, 1.79-2.62 % respectively. Peroxide values of olive oil with different additives along at first-fourth weeks were changed between 11.98-15.10 meq O₂/kg, 15.48-19.64 meq O₂/kg, 18.22-27.50 meq O₂/kg, 18.70-39.60 meq O₂/kg respectively. Viscosity values of olive oil with different additives along at first- fourth weeks were changed between 39.90-55.45 m.Pas, 53.35-59.60 m.Pas, 33.10-54.70 m.Pas, 34.00-50.80 m.Pas respectively. The weakest antioxidant effect was determined in sater oil. Thujene exhibited the highest antioxidant effect, followed by eucalyptol, ocimene, myrtle-white, BHA (butylated hydroxyanisole), fennel and savory-sater essential oil respectively. Fatty acid compositions of olive oils had been partly affected from essential oil (0.1%) and some corresponding constituents (0.01%). Total amount of fatty acids changed between 96.86 % to 99.99 %. The most effected acids were linoleic acid, followed oleic and linolenic acids.

Keywords: Essential oil, BHA, antioxidant effect, peroxide value, viscosity, virgin olive oil.

Zeytinyağının Bazı Fiziko-Kimyasal Özellikleri Üzerine, Ticari Antioksidan ve Esansiyel Yağ Bileşenlerinin Etkilerinin Belirlenmesi

Öz

Bu çalışmada, 60°C de birkaç baharat esansiyel yağının ve bazı bileşenlerin % 0.1 seviyesinde, zeytinyağının oksidatif stabilitesi üzerindeki antioksidan aktivitesi belirlenmiştir. Araştırma sonuçlarına göre, zeytinyağına farklı ilaveler ile yağın serbest yağ asitliği değerleri, birinci haftadan dördüncü haftaya kadar sırasıyla % 1.61-2.01, % 1.81-2.17, % 1.67-2.23, % 1.79-2.62 arasında değişmiştir. Peroksit sayısı değerleri, birinci haftadan dördüncü haftaya kadar sırasıyla 11.98-15.10 meq O₂/kg, 15.48-19.64 meq O₂/kg, 18.22-27.50 meq O₂/kg, 18.70-39.60 meq O₂/kg arasında değişmiştir. Viskozite değerleri, 39.90-55.45 m.Pas, 53.35-59.60 m.Pas, 33.10-54.70 m.Pas, 34.00-50.80 m.Pas arasında değişmiştir. Araştırmanın ilk haftasında yağın peroksit değerleri 11.98 meq O₂ / kg ile 15.10 meq O₂ / kg (p<0.05) arasında değişmiştir. En zayıf etki, sater yağında belirlenmiştir. Thujene en yüksek antioksidan etkiyi göstermiş olup, ardından okaliptol, ocimene, mersin beyazı, BHA (bütillenmiş hidroksianisol) ve rezene izlemiştir. Zeytinyağlarının yağ asidi bileşimleri, esansansiyel yağlardan (% 0.1) ve buna karşılık gelen bazı bileşenlerden (% 0.01) kısmen etkilenmiştir. Zeytin yağının toplam yağ asidi miktarı % 96.86 ile % 99.99 arasında değişmiştir. En çok etkilenen yağ asitleri linoleik asit, ardından oleik ve linolenik asitler olmuştur.

Anahtar kelimeler: Esansiyel yağ, BHA, antioksidan etki, peroksit değeri, sızma zeytin yağı, viskozite.

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1. Introduction

Olive oil has an important place in vegetable oils with its unique flavor (Erinç ve Kıralan, 2008). As well known, natural olive oil is obtained mechanically by pressing, centrifuging and filtering the olive fruits. In order to prevent oxidative degradation of this natural olive oil, it is necessary to store the oil in acid-inert equipment and at low temperature in such a way that it eliminates or minimizes contact with air and light and metal ions. (Türkan, 2008). Recently, the important of spices and herbs as natural antioxidants in foods is increasing. These effects are caused by antioxidant components in the content of the plant, such as flavonoids, essential oil components, plant phenolics (Nilsson et al., 2005; Tawaha et al., 2007; Salluca et al., 2008; Temitope et al., 2010; Rice-Evans et al., 1996; Özcan and Al-Juhaimi, 2011; Rice-Evans et al., 1997). Bioactive components of spices such as curcumin, zingerone, allicin are good antioxidant sources for lipid peroxidation (Nuutila et al., 2003; Noguchi et al., 1994). The use of synthetic antioxidants butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) and tert-butyl hydroquinone (TBHQ) due to their potential health risks and toxicity is increasingly restricted (Moure et al., 2001). As far as our literature survey could as certain, scarce information was available on the antioxidant effect of both spice essential oils and some corresponding constituents on virgin olive oil. The objective of present study was to investigate the antioxidant effects of the essential oils and some their corresponding compounds on olive oil.

2. Material and Method

2.1. Material

The antioxidant effects were determined for all the spices and herbs (fennel bitter; myrtle-white, rosemary, oregano, laurel, basil, myrtle-black, mint, sage, savory sater in Table 1.) and five constituents (carvacrol, eucalyptol, ocimene, thujene and thymol) using peroxidation assay in model system. Five constituents (carvacrol, eucalyptol, ocimene, thujene and thymol) were provided 90-98% purities from by Sigma-Aldrich Corporation company (Merck). The olive oil was obtained from the North-West region of Saudi Arabia that commercial virgin olive oil. This oil did not contain food additive. BHA was used as a standard antioxidant for a comparison. The storage condition was carried out in a dark glass bottle in a cool and moisture-free environment for 4 weeks.

2.2. Method

2.2.1. Extraction of the essential oil

After dried and ground spices (about 100 g for each) were subjected to hydrodistillation for 3 h at 60°C added average water 1:2 ratio using a Clevenger-type apparatus, the oils were dried over anhydrous sodium sulfate.

2.2.2. Physico-chemical analysis

All analyses (viscosity, free fatty acidity, peroxide number) were applied according to the AOCS (1990). The 0.01% BHA, 0.1% essential oil and 0.01% constituents were added directly into olive oil, and a solution was obtained by manual homogenisation (15 °C) for about 5 minutes. A control sample was prepared without addition of any antioxidant.

2.2.3. Determination of fatty acids

The fatty acid methyl esters were identified by comparing the retention time of the samples and appropriate fatty acids methyl esters standards Hışıl (1988). About 20 mg of sodium hydrogen sulphate (monohydrate, extra pure; Merck, Darmstadt, Germany) was added in samples and after centrifugation at 4500 rpm for 10 min, the top n-heptane phase was injected in a Gas Chromatography (Shimadzu GC-2010) equipped with a flame ionising detector (FID). The silica capillary column (RTX-2330, 100 m x 0.25 mm i.d.; film thickness 0.20 micrometer).

Working condition of GC, as follows;

Temperature

Column : 180°C

Enjector : 200°C

Dedector : 200°C

Flow

Carrier gas (N₂) : 30 ml/min.

Combustible gas (H₂) : 28 ml/min.

Dry air: 220 ml/min.

Printer: Chromatopac CR 6A (Shimadzu)

Enjection volume: 1µl

2.2.4. Statistical Analyses

A complete randomized split plot block design was used, and analysis of variance (ANOVA) was performed by using JMP version 9.0 (SAS Inst. Inc., Cary, N.C.U.S.A). All analyses were carried out triplicate and the results are mean±standard deviation (MSTAT C) of independent spice and constituents (Püskülcü and İkiz, 1989).

Table 1. Plants used in experiment

| Used plants | Name | Family | Used parts |
|---|---------------|-----------|-----------------|
| <i>Foeniculum vulgare</i> L. ssp <i>piperitum</i> | Bitter fennel | Apiaceae | Fruit |
| <i>Myrtus communis</i> L. (with white fruit) | Myrtl | Myrtaceae | Leaves |
| <i>Myrtus communis</i> L.(with black fruit) | Myrtl | Myrtaceae | Leaves |
| <i>Rosmarinus officinalis</i> L. | Rosemary | Lamiaceae | Leaves +Flowers |
| <i>Origanum minutiflorum</i> L. | Oregano | Lamiaceae | Leaves +Flowers |
| <i>Laurus nobilis</i> L. | Laurel | Lauraceae | Leaves +Flowers |
| <i>Ocimum basilicum</i> L. | Basil | Lamiaceae | Leaves +Flowers |
| <i>Mentha spicata</i> L. | Mint | Lamiaceae | Leaves +Flowers |
| <i>Salvia fruticosa</i> L. | Sage | Lamiaceae | Leaves +Flowers |
| <i>Satureja hortensis</i> L. | Savory-sater | Lamiaceae | Leaves +Flowers |

3. Results and Discussion

The effects on some physico-chemical properties (free fatty acidity, peroxide number, viscosity) of olive oil of some spice essential oils and corresponding constituents during storage are given in Table 2a and 2b.

According to our results, it was observed an important difference of the free fatty acid values of olive oil containing several spice essential oils and some corresponding constituents ($p < 0.05$) (Table 2a and 2b). While the free fatty acidity of samples at the first week was found between 1.61% (sage) to 2.01% (fennel and ocimene), at the second week, the free fatty acid values of olive samples were determined between 1.81% (thymol) to 2.37% (laurel). These values ranged from 1.67% (myrtle-white) to 2.23% (thujene) at the third week of experiment (Table 2b). At the last week, the free fatty acid values of olive oil samples containing essential oil and corresponding constituents were found between 1.79% (basil and thymol) to 2.62% (myrtle-black). As a result, the free fatty acidity of olive oil was not effected from essential oil and corresponding constituents. But, the free fatty acid values of the most of essential oils and constituents were found partly lower compared to control group.

Table 2.a. Changes at the peroxide values, free fatty acid and viscosity values of treated olive oil

| Additives | 1th Week | | | 2th Week | | |
|----------------|---|------------------------------|-------------------|---|------------------------------|-------------------|
| | Peroxide value (meq O ₂ /Kg) | Free Fatty Acidity (oleic %) | Viscosity (m.Pas) | Peroxide value (meq O ₂ /Kg) | Free Fatty Acidity (oleic %) | Viscosity (m.Pas) |
| Bitter fennel | 12.95±1.17*d | 2.01±0.21a | 47.05±1.38ef | 19.64±0.78a | 1.86±0.11b | 59.25±1.67b |
| Myrtl (white) | 12.75±0.95d** | 1.68±0.29e | 39.90±0.98 | 15.80±0.76e | 2.17±0.16a | 58.25±1.43bc |
| BHA | 12.91±0.78d | 1.78±0.16d | 54.45±1.51b | 15.48±0.49e | 1.83±0.21b | 59.25±1.69b |
| Rosemary | 13.79±1.21c | 1.79±0.46d | 51.60±1.67c | 15.70±0.67e | 1.83±0.13b | 56.00±1.82cd |
| Carvacrol | 14.78±1.33b | 1.79±0.32d | 45.25±1.19e | 18.08±0.91b | 1.98±0.15ab | 53.35±1.71ef |
| Oregano | 13.17±0.87c | 1.82±0.21c | 42.60±1.39f | 18.53±0.84b | 1.91±0.17ab | 54.50±1.49e |
| Laurel | 12.88±1.06d | 1.78±0.18d | 49.10±1.71d | 17.09±0.82c | 2.37±10.28a | 58.90±1.56bc |
| Eucalyptol | 12.12±1.15d | 1.73±0.21d | 46.20±1.65ef | 16.45±0.38d | 2.19±0.18a | 58.15±1.67bc |
| Basil | 13.40±1.29c | 1.89±0.29c | 55.45±1.52a | 18.39±0.82b | 2.12±0.16a | 58.22±1.63bc |
| Thymol | 14.11±1.19b | 1.73±0.19d | 49.10±0.98d | 16.41±0.71d | 1.81±0.16b | 56.85±1.52cd |
| Myrtle (black) | 14.51±1.17b | 1.77±0.13d | 53.80±1.29bc | 15.93±0.81e | 2.10±0.34a | 58.95±1.59bc |
| Mint | 14.83±1.36b | 1.98±0.17ab | 53.50±1.78bc | 16.75±0.92d | 2.07±0.29a | 56.25±1.48cd |
| Ocimene | 12.22±1.52d | 2.01±0.28a | 42.65±1.12f | 16.42±1.01d | 1.82±0.17b | 59.00±1.62b |
| Sage | 13.33±1.28c | 1.61±0.15e | 43.50±1.21f | 16.23±1.09d | 2.09±0.38a | 55.85±1.39d |
| Savory sater | 15.10±1.34a | 1.84±0.17c | 51.10±1.42c | 16.33±0.96d | 2.14±0.32a | 57.50±1.53c |
| Thujene | 11.98±0.79e | 1.90±0.21ab | 53.90±1.36bc | 15.65±0.99e | 2.10±0.39a | 59.60±1.78b |
| Control | 13.40±1.72c | 1.67±0.11e | 54.35±1.87b | 15.46±0.78e | 2.27±1.39a | 62.00±1.78a |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different ($p < 0.05$)

Peroxide values of oil at the first week of experiment ranged from 11.98 to 15.10 meq O₂/kg. The effects of fennel, myrtl (white), BHA, oregano, laurel, eucalyptol, ocimene, sage and thujene were found partly higher compared to control group without food additive. The weakest effect was established in savory oil. Thujene exhibited the highest antioxidant effect, followed by eucalyptol, ocimene, myrtle-white, BHA and fennel. At the second week, it was observed an increase at the peroxide values. According to control group, both essential oils and their some constituents particularly stimulated peroxide values. The peroxide values of samples were determined between 15.46 to 19.64 meq O₂/kg. The highest peroxide value was recorded on olive oil with fennel essential oil. While this oil inhibited oxidation, it stimulated at the rate of about 49% at the second week. In addition, the effects of carvacrol, oregano and basil were weak on the stability of olive oil in 60°C ($p < 0.05$). The peroxide values of myrtle-white, BHA, myrtle-black and thujene were found partly similar compared to control group. At the 3rd week of experiment, the peroxide values of samples increased when compared with results of the first and second week. The highest peroxide value (27.5 meq O₂/kg) was established in olive oil containing carvacrol, followed by laurel (23.88 meq O₂/kg), myrtle-black (23.76 meq O₂/kg) and sage (21.57 meq O₂/kg) compared to control group (19.42 meq O₂/kg) and BHA (19.32 meq O₂/kg). The highest increase of the peroxide values of samples (except for myrtl-white) was observed at the 4rd

week of experiment. At the 4th week, peroxide values were determined between 18.70 to 39.60 meq O₂/kg. According to control group, the peroxide values of myrtle-white, BHA, eucalyptol and basil were found particularly low. In addition, peroxide value of fennel oil was found similar compared to control. Generally, the effects of both spices and corresponding constituents on oxidation of olive oil kept at 60 °C were found positive compared to control and BHA. At the same time, the most of constituents had more effects on the stability of olive oil compared to essential oils. Such as rosemary and sage oils are known antioxidants and find many applications in food preparations. Naturally occurring compounds in rosemary extracts have been reported to exhibit antioxidant properties greater than BHA and equal BHT (Wu et al., 1982; Ho et al., 1983, Özcan 1999). These differences may be probably due to phenolic compound structure of spice essential oils. In addition, it was observed statistically significant differences among peroxide value and viscosity values of olive oil treated with several spice and constituents compared to control group during storage periods ($p < 0.05$). It was observed a linear correlation between phenolic content and antioxidant activity in studies made by several researchers (Velioglu et al., 1998; Gheldof and Engeseth., 2002; Oktay et al., 2003).

Table 2.b. Changes at the peroxide values, free fatty acid and viscosity values of treated olive oil

| Additives | 3th Week | | | 4th Week | | |
|----------------|---|------------------------------|-------------------|---|------------------------------|-------------------|
| | Peroxide value (meq O ₂ /kg) | Free Fatty Acidity (oleic %) | Viscosity (m.Pas) | Peroxide value (meq O ₂ /kg) | Free Fatty Acidity (oleic %) | Viscosity (m.Pas) |
| Bitter fennel | 21.27±0.59b | 1.96±0.19b | 51.25±1.21d | 26.73±0.45e | 1.87±0.13 | 45.40±1.42bc |
| Myrtle (white) | 19.61±0.32d | 1.67±0.14d | 52.95±1.17c | 18.70±0.42h | 1.94±0.15 | 44.60±0.98bc |
| BHA | 19.32±0.59d | 1.83±0.13c | 52.45±1.27c | 23.89±0.51g | 1.88±0.21 | 43.10±1.21c |
| Rosemary | 21.26±0.89b | 1.79±0.29cd | 51.00±1.29d | 29.54±0.57cd | 1.95±0.23 | 50.80±1.45a |
| Carvacrol | 27.50±0.38a | 1.90±0.21b | 33.10±0.57h | 39.60±0.28a | 1.89±0.19 | 25.50±0.76g |
| Oregano | 21.80±0.49b | 1.95±0.27b | 45.40±0.98f | 33.90±0.32b | 2.17±0.27 | 34.00±1.28f |
| Laurel | 23.88±0.63b | 1.73±0.18cd | 47.20±0.82e | 28.99±0.2d | 1.93±0.18 | 42.70±1.53c |
| Eucalyptol | 20.00±0.87b | 2.13±0.19a | 50.75±1.12de | 25.22±0.19ef | 1.86±0.19 | 41.60±1.57cd |
| Basil | 18.22±0.58e | 1.83±0.13c | 53.22±1.37b | 24.41±0.22ef | 1.79±0.21 | 43.22±1.46c |
| Thymol | 20.87±0.87c | 1.80±0.23c | 43.15±1.12g | 29.13±0.43cd | 1.79±0.41 | 35.20±1.27f |
| Myrtle (black) | 23.76±0.78b | 1.89±0.37c | 47.95±1.21e | 32.20±0.18bc | 2.62±0.37 | 41.00±1.49d |
| Mint | 20.00±0.89c | 1.76±0.25cd | 50.10±0.97de | 27.85±0.27d | 2.00±0.19 | 47.20±1.63b |
| Ocimene | 19.90±0.67d | 1.69±0.21d | 54.70±1.11a | 28.57±0.16d | 1.87±0.49 | 46.00±1.34b |
| Sage | 21.57±0.78b | 1.69±0.22d | 51.95±1.38d | 30.70±0.45c | 1.97±0.41 | 46.60±1.52b |
| Savory sater | 21.27±0.94b | 1.90±0.28b | 42.05±1.27g | 29.00±0.52cd | 1.99±0.43 | 46.30±1.45b |
| Thujene | 19.80±0.76d | 2.23±0.38a | 47.55±1.39e | 30.71±0.46c | 2.07±0.39 | 41.00±1.59e |
| Control | 19.42±0.76d | 1.94±0.16b | 54.15±1.39a | 26.15±0.32e | 1.87±0.31 | 40.60±1.27ef |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different ($p < 0.05$)

Generally, the viscosity values of olive oil contained essential oil and their constituents were found low when compared with results of control group up to 3rd week of experiment. But, at the 4th week (Table 2b), the viscosity values of samples partly decreased together with control group. While the viscosity values of olive oil samples changed between 39.90 m.Pas (myrtle-white) to 55.45 m.Pas (basil), at the 3rd week, these values varied between 33.10 m.Pas (carvacrol) to 54.70 m.Pas (ocimene). But, at the 4th week of experiment, the viscosity values were measured between 25.50 m.Pas (carvacrol) to 50.80 m.Pas (rosemary) ($p < 0.05$). The viscosity values of carvacrol, oregano and thymol were found lower compared to control (40.60 m.Pas). Generally, the viscosity values of samples had probably changed depending on temperature, polymerization of oil and additives added into olive oil.

Table 3.a. The effect of some essential oil and constituents on fatty acid composition of olive oil (%)

| Week | Additives | Fatty acid composition | | | | | |
|---------|----------------|------------------------|--------------|-------------|-------------|------------|-------|
| | | Palmitic | Oleic | Linoleic | Linolenic | Arachidic | Total |
| 1 | Bitter fennel | 7.80±0.19*c | 75.77±10.01a | 15.16±0.23c | 1.25±0.06 | -*** | 99.98 |
| | Myrtl (white) | 8.81±0.21b** | 74.57±0.11b | 13.51±0.34e | 1.29±0.04c | 0.66±0.03f | 98.84 |
| | BHA | 7.90±0.23c | 75.15±0.98a | 14.87±0.26d | 1.35±0.11a | 0.70±0.07b | 99.97 |
| | Rosemary | 9.10±0.32a | 75.65±0.91a | 13.93±0.37e | 1.30±0.07b | - | 99.98 |
| | Carvacrol | 8.50±0.38b | 75.22±0.89a | 13.74±0.21e | 1.22±0.21h | 0.69±0.06c | 99.37 |
| | Oregano | 7.56±0.29c | 75.67±0.71a | 14.83±0.58d | 1.25±0.17g | 0.67±0.03e | 99.98 |
| | Laurel | 9.00±0.27a | 74.99±0.68b | 12.85±0.41f | 1.28±0.14d | 0.68±0.05d | 98.80 |
| | Eucaliptol | 7.70±0.21c | 75.39±0.87a | 14.76±0.53d | 1.30±0.11b | 0.70±0.11b | 99.85 |
| | Basil | 6.82±0.42d | 75.80±0.99a | 15.35±0.25c | 1.20±0.09h | 0.70±0.13b | 99.87 |
| | Thymol | 6.87±0.51d | 75.03±0.78a | 14.43±0.34d | 1.22±0.09h | 0.18±0.03i | 97.73 |
| | Myrtle (black) | 9.70±0.32a | 75.34±0.77a | 12.97±0.19f | 1.26±0.09fg | 0.69±0.07c | 99.96 |
| | Mint | 6.70±0.28d | 75.40±0.71a | 16.37±0.23b | 1.28±0.06d | 0.22±0.03h | 99.97 |
| | Ocimene | 7.80±0.19c | 74.77±0.69b | 14.61±0.21d | 1.26±0.11fg | 0.67±0.07e | 99.11 |
| | Sage | 9.00±0.37a | 75.52±0.67a | 13.46±0.27e | 1.29±0.09c | 0.71±0.11a | 99.98 |
| | Savory sater | 8.80±0.28b | 74.97±0.56b | 14.05±0.31d | 1.27±0.17f | 0.28±0.03g | 99.37 |
| | Thujene | 5.79±0.37e | 74.09±0.96b | 17.09±0.23a | 1.28±0.19d | - | 98.25 |
| Control | 7.60±0.56c | 74.49±1.03b | 15.04±0.16c | 1.31±0.7ab | 0.64±0.09 | 99.08 | |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different (p <0.05);

***nonidentified

The fatty acid composition of olive oil containing several essential oils and some their constituents are presented in Tables 3a, 3b, 3c and 3d. During experiment, palmitic acid values of samples were determined between 5.70% (mint at 4th week) to 9.21% (myrtle-black) at the 3th week. While palmitic acid values of olive oils at the first week of experiment ranged from 5.79% (thujene) to 9.70% (myrtle-white), these values changed between 6.01% (thujene) to 9.20% (sage) at the second week. In addition, while palmitic acid values were found between 6.49% (basil) to 9.20% (sage) at the 3th week, these values were established between 5.70% (mint) to 9.0% (myrtle-black and savory).

Results exhibited partly differences according to control group. It was not observed an important difference in oleic acid contents of olive oil samples contained essential oils and their constituents. The oleic acid contents of samples were found between 71.78% (savory at the first week) to 77.15% (mint at the last week).

Table 3.b. The effect of some essential oil and constituents on fatty acid composition of olive oil (%)

| Week | Additives | Fatty acid composition | | | | | |
|---------|----------------|------------------------|-------------|-------------|-------------|-------------|-------|
| | | Palmitic | Oleic | Linoleic | Linolenic | Arachidic | Total |
| 2 | Bitter fennel | 7.60±0.31c | 74.21±0.73b | 14.32±0.37c | 1.30±0.31c | 0.78±0.16c | 98.21 |
| | Myrtl (white) | 7.81±0.29c | 73.89±0.71c | 14.52±0.39c | 1.18±0.13g | 0.86±0.07a | 98.26 |
| | BHA | 7.34±0.27c | 74.65±0.86b | 15.36±0.41b | 1.34±0.11b | 0.38±0.03j | 99.07 |
| | Rosemary | 8.81±0.25b | 72.82±0.95d | 13.47±0.38d | 1.23±0.08e | 0.68±0.06e | 97.01 |
| | Carvacrol | 7.59±0.37c | 74.62±0.83b | 14.55±0.56c | 1.20±0.07e | 0.63±0.04g | 98.59 |
| | Oregano | 7.21±0.41c | 74.25±0.78b | 15.26±0.47b | 1.17±0.06g | 0.78±0.11c | 97.97 |
| | Laurel | 8.90±0.42b | 75.42±0.72a | 14.00±0.43c | 1.27±0.05d | 0.36±0.02j | 99.95 |
| | Eucaliptol | 6.78±0.39d | 75.38±0.88a | 16.02±0.51a | 1.28±0.08cd | 0.52±0.06i | 99.98 |
| | Basil | 7.42±0.33c | 75.66±0.86a | 13.56±0.21d | 1.41±0.11a | 0.60±0.04h | 98.65 |
| | Control | 7.26±0.37c | 75.26±0.91a | 15.50±0.29b | 1.26±0.09d | 0.71±0.07d | 99.99 |
| | Myrtle (black) | 9.21±0.49a | 73.96±0.94c | 13.31±0.18d | 1.29±0.16cd | 0.49±0.03i | 98.26 |
| | Mint | 6.81±0.35d | 75.25±0.85a | 15.52±0.38b | 1.23±0.17e | 0.55±0.03i | 99.36 |
| | Ocimene | 7.90±0.41c | 75.78±0.93a | 14.06±0.32c | 1.26±0.31d | 0.65±0.06f | 99.61 |
| | Sage | 9.20±0.53a | 75.81±0.69a | 12.61±0.37e | 1.22±0.21e | 0.72±0.09d | 99.56 |
| | Savory sater | 9.18±0.57a | 74.93±0.78b | 13.05±0.17d | 1.19±0.05f | 0.85±0.09ab | 98.96 |
| | Thujene | 6.01±0.37d | 75.53±0.77a | 16.51±0.14a | 1.28±0.07cd | 0.65±0.03f | 99.98 |
| Control | 7.60±0.56c | 74.49±1.03b | 15.04±0.16c | 1.31±0.7ab | 0.64±0.09 | 99.08 | |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different (p <0.05);

***nonidentified

The oleic acid values of control group ranged from 74.49% to 75.36%. Linoleic acid contents of treated olive oils ranged from 10.75% (myrtle-black at the last week) to 17.09% (thujene at the first week). Linoleic acid values of olive oils with carvacrol, ocimene and thymol were found lower compared to control group during experiments.

Linolenic and arachidic acid contents of treated olive oils were identified under 1.5% level. Total amount of fatty acids changed between 96.86% to 99.99%. During storage, it was observed significant differences among palmitic, oleic and linoleic acid contents of olive oil compared to control group ($p < 0.05$). As a result, fatty acid compositions of olive oils had been partly affected from essential oil (0.1%) and some corresponding constituents (0.01%).

Table 3.c. The effect of some essential oil and constituents on fatty acid composition of olive oil (%)

| Week | Additives | Fatty acid composition | | | | | Total |
|--------------|-------------------|------------------------|-------------|-------------|-------------|-------------|-------|
| | | Palmitic | Oleic | Linoleic | Linolenic | Arachidic | |
| 3 | Bitter fennel | 8.60±0.42b | 75.98±0.84a | 12.77±0.56d | 1.17±0.06f | 0.71±0.02d | 99.23 |
| | Myrtl (white) | 6.81±0.38d | 74.12±0.93b | 14.29±0.42b | 1.30±0.04c | 0.34±0.03j | 96.86 |
| | BHA | 7.80±0.39c | 75.45±0.87a | 14.20±0.37b | 1.24±0.06d | 0.67±0.06f | 99.36 |
| | Rosemary | 8.80±0.81b | 74.45±0.81b | 12.76±0.32d | 1.09±0.11g | 0.69±0.09e | 97.79 |
| | Carvacrol | 7.60±0.69c | 74.88±0.83b | 13.79±0.25c | 1.05±0.07g | 0.51±0.07i | 97.76 |
| | Oregano | 8.01±0.71b | 73.90±0.67c | 14.05±0.27b | 1.25±0.09d | 0.92±0.09a | 98.13 |
| | Laurel | 9.00±0.59a | 75.12±0.94a | 13.18±0.37c | 1.09±0.03g | 0.47±0.03ij | 98.86 |
| | Eucaliptol | 7.10±0.42c | 75.53±0.86a | 15.22±0.33a | 1.20±0.11d | 0.60±0.07i | 99.65 |
| | Basil | 6.49±0.47d | 74.90±0.82b | 15.71±0.41a | 1.60±0.16ab | 0.38±0.03j | 99.08 |
| | Thymol | 7.43±0.56c | 74.73±0.69b | 14.45±0.31b | 1.13±0.09f | 0.61±0.07i | 98.17 |
| | Myrtle (black) | 9.10±0.68a | 73.90±0.85c | 12.25±0.23d | 2.01±0.26a | 0.73±0.07c | 97.99 |
| | Mint | 6.60±0.71d | 75.37±0.89a | 15.56±0.21a | 1.22±0.18d | 0.34±0.03j | 99.09 |
| | Ocimene | 8.31±0.74b | 74.69±0.85b | 13.73±0.32c | 1.32±0.11d | 0.67±0.08f | 98.72 |
| | Sage | 9.20±0.87a | 75.49±0.99a | 12.89±0.38d | 1.19±0.09e | 0.64±0.03h | 99.33 |
| Savory sater | 9.13±0.76a | 71.78±0.87d | 12.25±0.36d | 1.65±0.21b | 0.81±0.09b | 94.81 | |
| Thujene | 6.71±0.59d | 75.07±0.82a | 15.14±0.27a | 1.22±0.18d | 0.67±0.05f | 98.56 | |
| Control | 7.20±0.53c | 75.36±0.78a | 14.99±0.36b | 1.20±0.17d | 0.65±0.06g | 99.40 | |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different ($p < 0.05$); ***nonidentified

The most effected acids were linoleic acid, followed oleic and linolenic acids. Chang et. al., (2013) have been determined the antioxidant activity of fennel seed extracts (FSE) was evaluated by synthetic antioxidant. During 28 days of storage, a compromise was accomplished based on the results assessed by peroxide value, at which the antioxidant activity of FSE was higher than BHA (75 ppm), BHT (75 ppm) and BHA to BHT ratio of 1:1 at the concentration of 150 ppm. Among them, concentration of 150 ppm showed the best antioxidant activity.

Another study was to investigate the effect of rosemary essential oil on the physico-chemical properties of extra-virgin olive oil. Free fatty acid and peroxide values of olive oils stored in different coloured bottles increased partly during storage. After 90 days of storage, free fatty acid values of samples changed between 0.78 and 0.89 mg KOH/g oil.

By the 90th day of storage peroxide values of samples had changed from 32.75 to 79.46 meq O₂/kg oil, whereas the peroxide value of the control group on the 90 th day was 94.55 meq O₂/kg. Linoleic acid (40.95-43.92%), oleic acid (33.04-34.99%) and palmitic acid (12.38-13.58%) were the major fatty acids of olive oils (Juhaimi et.al., 2015).

Table 3.d. The effect of some essential oil and constituents on fatty acid composition of olive oil (%)

| Week | Additives | Fatty acid composition | | | | | |
|---------|----------------|------------------------|-------------|-------------|-------------|-------------|-------|
| | | Palmitic | Oleic | Linoleic | Linolenic | Arachidic | Total |
| 4 | Bitter fennel | 8.43±0.81b | 76.69±0.82b | 12.83±0.18d | 1.13±0.06bc | 0.70±0.09d | 99.78 |
| | Myrtl (white) | 7.11±0.97c | 76.32±0.68b | 14.43±0.31b | 1.11±0.07c | 0.69±0.11de | 99.66 |
| | BHA | 7.20±0.85c | 75.16±0.69c | 14.23±0.21b | 1.14±0.07b | 0.68±0.16e | 98.41 |
| | Rosemary | 7.90±0.71c | 76.22±0.83b | 13.10±0.18c | 1.23±0.03a | 0.59±0.05g | 99.04 |
| | Carvacrol | 7.80±0.77c | 76.56±0.96b | 12.57±0.16d | 1.01±0.06d | 0.70±0.06d | 98.60 |
| | Oregano | 8.00±0.89b | 75.93±0.84c | 12.60±0.28d | 1.03±0.07d | 0.79±0.09ab | 98.35 |
| | Laurel | 7.90±0.81c | 76.60±0.74b | 13.51±0.26c | 1.09±0.03d | 0.69±0.03de | 99.79 |
| | Eucalyptol | 7.00±0.88c | 75.88±0.71c | 14.58±0.31b | 1.11±0.07c | 0.66±0.03f | 99.23 |
| | Basil | 6.65±0.54d | 76.08±0.77b | 15.36±0.39a | 0.92±0.03 | 0.92±0.11a | 99.93 |
| | Thymol | 7.43±0.74c | 75.23±0.83c | 13.07±0.31c | 1.23±0.09a | 0.68±0.06e | 97.64 |
| | Myrtle (black) | 9.00±0.71a | 76.74±0.92b | 10.75±0.41e | 1.12±0.08c | 0.71±0.9c | 98.30 |
| | Mint | 5.70±0.48e | 77.15±0.84a | 15.13±0.48a | 1.10±0.05c | 0.72±0.13b | 99.80 |
| | Ocimene | 8.20±0.98b | 76.98±0.77b | 12.78±0.36d | 1.13±0.09bc | 0.71±0.09c | 99.80 |
| | Sage | 8.00±0.76b | 75.98±0.85c | 13.23±0.31c | 1.09±0.07d | 0.69±0.06e | 98.99 |
| | Savory- sater | 9.00±0.99a | 76.11±0.79b | 12.15±0.28d | 1.10±0.06c | 0.72±0.07b | 99.08 |
| Thujene | 5.80±0.67e | 76.16±0.76b | 15.45±0.23a | 1.14±0.11bc | 0.70±0.11d | 99.25 | |
| Control | 6.90±0.51d | 74.96±0.81d | 14.06±0.37b | 1.13±0.09bc | 0.70±0.13d | 97.75 | |

*mean±standard deviation (n:3); **Values within each column followed by different letters are significantly different ($p<0.05$);

***nonidentified

Baiano et.al., (2010) showed that addition of some essential oils reduced lipid oxidation and showed an antioxidant effect when compared with the control group olive oil.

4. Conclusions and Recommendations

As a result, some spices and their some corresponding constituents have strong antioxidant effects (thujene, eucalyptol, ocimene, myrtle-white and BHA (butylated hydroxyanisole). These plant materials are expected to be a valuable food constituents for promoting good health in daily live. It can be concluded that essential oils extracted from these plants can supply a good opportunity as an antioxidant agent in food industry, if any sensory effects are acceptable. After these screening experiments, further works will be performed to describe the antioxidant in more details.

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