

# Antioxidant Potential of *Eugenia caryophyllus*, *Satureja hortensis* and *Artemisia dracunculus* Essential Oils in Grape Seed Oil

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## Abstract:

**BACKGROUND:** Autoxidation is an irreversible reaction which occurs with the effect of oxygen in the air, and results in unpleasant taste and smell that are known as the signs of rancidity in oil.

**OBJECTIVES:** In this study, the antioxidant potential of clove, summer savory and tarragon essential oils (EOs) in grape seed oil was evaluated.

**METHODS:** Effects of EOs at different concentrations (0.3, 0.5, 1, and 1.5 %, v/v) on peroxide value (PV) and thiobarbituric acid reactive substances (TBARS) of grape seed oil at 60 °C were investigated.

**RESULTS:** Results showed 1.5% clove showed the lowest PV (52.13 meq/kg) at the end of the period. Among EO- treated samples, the highest PV was seen in samples treated with tarragon. There was no significant difference between the TBARS of samples containing 1% clove and 1.5% savory at day 10 of storage. TBARS of clove treated samples increased slightly toward the end of storage and similar trend was observed for savory-treated samples. TBARS values of tarragon treated samples at each storage time were higher than those for clove and savory EOs.

**CONCLUSIONS:** The antioxidant activity of EOs in grape seed oil was as follows: clove > summer savory > tarragon.

## Keywords:

Antioxidant activity, Clove, Grape seed oil, Summer savory, Tarragon

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## Introduction

The quality of edible oils and fats and also fatty foods is affected by oxidative deterioration during storage period depending on various factors such as light, heat, enzymes and sensitizers like trace metals in biological complexes. Autoxidation is an irreversible reaction which occurs with the effect of oxygen in the air, and results in unpleasant taste and smell that are known as the signs of rancidity in oil. Autoxidation goes on spontaneously when it begins, and its rate is directly related to the oil's unsaturation degree. The breakdown products formed after oxidation process such as peroxides, aldehydes and ketones shorten the shelf life of oils and turn products unacceptable for consumption (Zhang et al., 2018).

Antioxidants commonly used in food products today are butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT). In recent years the safety of synthetic food additives, including the possible toxicity of these chemicals used as antioxidants has received increasing attention. A common need is availability of natural extracts with a pleasant taste or smell combined with preservative effects, aimed to avoid lipid deterioration, oxidation and spoilage by microorganisms. To prevent the harmful effects of synthetic antioxidants, the use of antioxidants which are found in foods and various natural materials such as herbs is recommended (Embuscado, 2015). Cloves are the aromatic dried flower buds of a tree (*Eugenia caryophyllus*) of the family Myrtaceae (Chaieb et al., 2007). They exhibit anti-mutagenic, anti-inflammatory (Mektrirat et al., 2016), antioxidant (Embuscado, 2015) and anti-parasitic (Guldiken et al., 2018) properties. Also, it is report-

ed that eugenol which is one of the main components in clove essential oil (EO) is demonstrated as an effective antioxidant compound in several works (Ozcan and Arslan, 2011). Summer savory (*Satureja hortensis*) is cultivated in the West, Northwest and southern parts of Iran (Sefidkon et al., 2004). Summer savory might be used as antimicrobial agent, antispasmodic and stomach tonic. Researches on summer savory report that compounds which have antioxidant effects have phenol structure, such as thymol and p-cymene (Feyzioglu and Tor-nuk, 2016). Tarragon (*Artemisia dracunculus* L.) is a traditional medicinal plant and is used for the treatment of stomach pains, pyrexia, diabetes and parasitic or bacterial infections. The fresh and dried leaves are commonly used in salads, soups and barbecues. Based on the results of several studies, the EO of tarragon had biological properties as anti-oxidative and antibacterial activities (Sharafati Chaleshtori et al., 2013; Oswell et al., 2016).

The antioxidative effects of natural antioxidants, as herbs, on lipids were investigated in recent years as an extensive research area. It is presumed that herbal essential oils possess considerable antioxidative potential in food products. So, the purpose of this work was to investigate tarragon, summer savory and clove EOs' antioxidant effects on grape seed oil stored at 60 °C during a period of 30 days, in darkness.

## Materials and Methods

**Grape seed oil:** The red grape seed oil with no antioxidants added was obtained from a local oil extraction shop in Babolsar (Mazandaran, Iran).

**Plant material and extraction of the**

**EOs:** Savory (*S. hortensis*) plant and clove (*E. caryophyllus*) buds were purchased from a local market in Tehran (Iran). Tarragon (*A. dracunculus*) plants were collected from Isfahan in October and November 2016. The samples were transported in polypropylene bags, and were dried to constant weight in room temperature for analyses. The taxonomic identification of plants materials was confirmed by the pharmacology department of Mazandaran University of Medical Sciences. Plant materials were identified according to the voucher herbarium specimens of plants in the repository of universities.

The air-dried clove buds and aerial parts of summer savory and tarragon plants were each subjected to water distillation for 3 h using a British-type Clevenger apparatus. The EOs obtained were dried over anhydrous sodium sulfate and stored in darkness at 4 °C in airtight glass vials closed under nitrogen gas.

**Gas chromatography/Mass spectroscopy:** EOs were analyzed by gas chromatography (GC) (Thermo Quest® 2000, UK). The chromatograph was equipped with a DB5 capillary column (Agilent Technologies, USA) (30 m × 0.25 mm ID × 0.25 µm film thickness) and the data were acquired under the following conditions: initial temperature 50 °C; rate of increase of temperature 2.5 °C, final temperature 265 °C and injector temperature 250 °C. An injection volume of 0.5 ml was employed using the autosampler (autosampler 7693 – 100 positions, Agilent Technologies, USA). The carrier gas was helium and the split ratio was 120. The column head pressure was 24.9 kPa. An Agilent 6890 Flame Ionization Detector (Agilent Technologies, USA), operated at 200 Hz, was used. EOs were also analyzed

by gas chromatography mass spectroscopy (GC/MS) (Thermo Quest Finnigan®, UK) using the same capillary column and analytical conditions indicated above. The MS was run in the electron ionization mode using an ionization energy of 70 eV. Components were identified based on the comparison of their relative retention times and mass spectra with those of standards (Guan, Li, Yan, Tang, and Quan, 2007). N-alkanes (C8-C20) were used as reference points in the calculation of relative retention indices (RRI) and data reported on reference books as well as standard libraries (Wiley 275.L and Wiley7n.L) (Adams, 2001).

**DPPH radical-scavenging activity:** The free-radical scavenging potential of the EOs was measured by 2,2-diphenyl-2-picrylhydrazyl (DPPH) according to the method explained by Hara et al. (2018). One milliliter of each EO at known concentration was added to 0.25 ml of a DPPH methanolic solution. The mixture was shaken vigorously and left at room temperature for 30 min in darkness. The absorbance of the resulting solution was then measured at 515 nm and corresponded to the ability of the EO to reduce the stable radical DPPH to the yellow colored diphenylpicrylhydrazine. Absorption of a blank sample containing the same amount of methanol and DPPH solution was considered as negative control.

**Treatments:** Four different concentrations of each EO (0.5, 1.0 and and 1.5 %, v/v) were added to grape seed oil and the samples were stored for 30 days at 60 °C, in darkness. Experiments were conducted at 5-day intervals.

**Determination of peroxide value (PV):** To evaluate the PV, 5.0 ± 0.05 g grape seed oil was put, along with 30 ml mixture (2:3, v/v) of chloroform and acetic acid into a

**Table 1.** Effect of type and concentration of EOs and storage time on the PV (meq/kg oil) of grape seed oil. The values marked with the same letters are not significantly different ( $P>0.05$ ).

Treatment	Storage period (day)						
	0	5	10	15	20	25	30
Control	2.20± 0.40 <sup>Fa</sup>	14.80± 0.72 <sup>Eb</sup>	30.53± 0.5 <sup>Da</sup>	51.20± 0.34 <sup>Ca</sup>	86.86± 0.5 <sup>Ba</sup>	157.6± 0.52 <sup>Aa</sup>	166.06± 0.5 <sup>Aa</sup>
Clove 0.3%	2.13± 0.35 <sup>Fab</sup>	12.66± 0.61 <sup>Ed</sup>	17.86± 0.50 <sup>Ee</sup>	51.20± 0.34 <sup>Ca</sup>	66.53± 0.50 <sup>Cf</sup>	102.13± 0.61 <sup>Be</sup>	127.06± 0.74 <sup>Af</sup>
Clove 0.5%	1.93 ± 0.41 <sup>Fab</sup>	9.00± 0.75 <sup>Eg</sup>	15.40± 0.40 <sup>Dg</sup>	25.00± 0.40 <sup>Ci</sup>	50.06± 0.50 <sup>Bk</sup>	89.00± 0.40 <sup>Ah</sup>	95.40± 0.40 <sup>Aj</sup>
Clove 1%	1.86± 0.11 <sup>Eab</sup>	7.20± 0.752 <sup>Fh</sup>	12.30± 0.41 <sup>Ei</sup>	22.40± 0.14 <sup>Dk</sup>	40.60± 0.72 <sup>Cm</sup>	68.53± 0.50 <sup>Bk</sup>	75.00± 0.40 <sup>Al</sup>
Clove 1.5%	1.60± 0.20 <sup>Fb</sup>	5.40± 0.52 <sup>Ei</sup>	9.40± 0.54 <sup>Dj</sup>	17.06± 0.50 <sup>Di</sup>	44.13± 0.61 <sup>Cl</sup>	62.40± 0.40 <sup>Al</sup>	52.13± 0.41 <sup>Bm</sup>
Summer savory 0.3%	2.09± 0.33 <sup>Fab</sup>	14.53± 0.41 <sup>Eb</sup>	25.80± 0.34 <sup>Dc</sup>	34.13± 0.41 <sup>Cd</sup>	75.40± 0.40 <sup>Bd</sup>	137.53 ± 0.46 <sup>Ac</sup>	134.66± 0.70 <sup>Ad</sup>
Summer savory 0.5%	1.93± 0.23 <sup>Hab</sup>	10.06± 0.50 <sup>Gf</sup>	22.65± 0.27 <sup>Fd</sup>	31.40± 0.40 <sup>Df</sup>	72.53± 0.23 <sup>Ce</sup>	113.00± 0.40 <sup>Bd</sup>	149.46± 0.23 <sup>Ac</sup>
Summer savory 1%	2.26± 0.41 <sup>Ga</sup>	9.33± 0.46 <sup>Ffg</sup>	15.66± 0.41 <sup>Ef</sup>	23.40± 0.22 <sup>Dj</sup>	60.46± 0.50 <sup>Ci</sup>	90.26± 0.23 <sup>Bg</sup>	114.40± 0.40 <sup>Ai</sup>
Summer savory 1.5%	1.90± 0.15 <sup>Fab</sup>	7.73± 0.30 <sup>Eh</sup>	13.40± 0.40 <sup>Dh</sup>	26.53± 0.50 <sup>Ch</sup>	55.00± 0.40 <sup>Bj</sup>	75.33± 0.50 <sup>Aj</sup>	81.00± 0.40 <sup>Ak</sup>
Tarragon 0.3%	2.00± 0.40 <sup>Fab</sup>	16.53± 0.38 <sup>Ea</sup>	28.73± 0.30 <sup>Db</sup>	44.53± 0.61 <sup>Cb</sup>	84.06± 0.30 <sup>Bb</sup>	139.693± 0.11 <sup>Ab</sup>	147.60± 0.40 <sup>Ab</sup>
Tarragon 0.5%	2.25± 0.10 <sup>Fa</sup>	13.60± 0.72 <sup>Ec</sup>	26.00± 0.34 <sup>Dc</sup>	37.00± 0.30 <sup>Cc</sup>	77.00± 0.40 <sup>Bc</sup>	137.13± 0.98 <sup>Ac</sup>	140.60± 0.60 <sup>Ac</sup>
Tarragon 1%	1.91± 0.25 <sup>Hab</sup>	11.46± 0.30 <sup>Fe</sup>	22.06± 0.30 <sup>Ed</sup>	32.90± 0.34 <sup>De</sup>	66.13± 0.23 <sup>Cg</sup>	99.86± 0.11 <sup>Bf</sup>	123.40± 0.40 <sup>Ag</sup>
Tarragon 1.5%	1.85± 0.11 <sup>Hab</sup>	9.46± 0.41 <sup>Gfg</sup>	16.40± 0.34 <sup>Ff</sup>	27.53± 0.64 <sup>Eg</sup>	61.33± 0.41 <sup>Ch</sup>	85.00± 0.40 <sup>Bi</sup>	117.20± 0.33 <sup>Ah</sup>
BHA 0.02%	1.60± 0.12 <sup>Gb</sup>	6.95± 0.16 <sup>Fj</sup>	15.53 ± 0.44 <sup>Ef</sup>	20.66± 0.37 <sup>Dm</sup>	28.73± 0.42 <sup>Cn</sup>	31.50± 0.65 <sup>Bm</sup>	39.47± 0.25 <sup>An</sup>

flask (Mehenbacher et al., 1997). To this mixture, 0.5 ml fresh saturated aqueous potassium iodide solution was added. The flask was shaken vigorously for about 1 min. Then, 30 ml distilled water was added and mixed thoroughly with the solution and titrated against 0.1 N and 0.01 N sodium thiosulphate solution, 0.5 ml soluble starch solution was used as an indicator. Also, a blank was prepared with no oil sample in it. PV was determined according to the following equation:

$$\text{PV (milliequivalents of peroxide/Kg oil sample)} = [(V_s - V_b) \times N \times 103] / w$$

Where  $V_s$  is the volume (ml) of sodium thiosulphate solution used for the sample,  $V_b$  that of the blank,  $N$  the normality of sodium thiosulphate solution and  $W$  the weight of the oil sample in grams.

#### Determination of thiobarbituric acid

**reactive substances (TBARS):** TBARS were analysed as a measurement of secondary lipid oxidation products, as described by Kristensen and Skibsted (1999). The thiobarbituric acid (TBA) reagent was prepared immediately before use by mixing equal volumes of freshly prepared 0.025 M TBA (brought into solution by neutralizing with NaOH) and 2 M  $H_3PO_4$ /2 M citric acid. Measurements were performed at 532 nm (red pigment) and 450 nm (yellow pigment) and the results are expressed as absorbance units in one gram of oil sample.

**Statistical analysis:** All determinations were performed in triplicate. The one-way ANOVA was performed to analyze the chemical parameters and significant differences were determined by using Tukey test. The analyses were performed in SPSS 20

**Table 2.** Effect of type and concentration of EOs and storage time on the TBARS (mg MDA/kg oil) of grape seed oil. The values marked with the same letters are not significantly different ( $P>0.05$ ).

Treatment	Storage period (day)						
	0	5	10	15	20	25	30
Control	0.056±0.01 <sup>Fab</sup>	0.18± 0.11 <sup>Ea</sup>	0.22± 0.01 <sup>DEa</sup>	0.35±0.02 <sup>CDa</sup>	0.43± 0.00 <sup>Ca</sup>	0.57± 0.02 <sup>Ba</sup>	0.71± 0.02 <sup>Aa</sup>
Clove 0.3%	0.054±0.02 <sup>Fab</sup>	0.10±0.00 <sup>EFde</sup>	0.13±0.01 <sup>DEdef</sup>	0.17±0.01 <sup>Ddef</sup>	0.26±0.01 <sup>Cde</sup>	0.35± 0.00 <sup>Bd</sup>	0.46± 0.00 <sup>Ad</sup>
Clove 0.5%	0.052±0.02 <sup>Eab</sup>	0.09±0.01 <sup>DEef</sup>	0.12±0.00 <sup>Dfgh</sup>	0.15±0.07 <sup>Dfgh</sup>	0.23± 0.02 <sup>Cef</sup>	0.31±0.01 <sup>Be</sup>	0.41± 0.00 <sup>Ae</sup>
Clove 1%	0.047±0.02 <sup>Eb</sup>	0.07± 0.00 <sup>Efg</sup>	0.09± 0.02 <sup>Ehi</sup>	0.12± 0.02 <sup>Dh</sup>	0.18± 0.04 <sup>Ch</sup>	0.22± 0.01 <sup>Bf</sup>	0.30± 0.05 <sup>Af</sup>
Clove 1.5%	0.058±0.01 <sup>Ea</sup>	0.06± 0.00 <sup>Dh</sup>	0.07± 0.00 <sup>Di</sup>	0.10± 0.01 <sup>CDi</sup>	0.14±0.06 <sup>BCi</sup>	0.20± 0.01 <sup>Bg</sup>	0.24± 0.00 <sup>Ag</sup>
Summer savory 0.3%	0.053±0.01 <sup>Fab</sup>	0.11± 0.01 <sup>Ecd</sup>	0.15± 0.06 <sup>Ecd</sup>	0.20±0.05 <sup>Dcd</sup>	0.29± 0.00 <sup>Cc</sup>	0.40± 0.01 <sup>Bc</sup>	0.54± 0.01 <sup>Ae</sup>
Summer savory 0.5%	0.049±0.00 <sup>Fab</sup>	0.11± 0.00 <sup>Ecd</sup>	0.12±0.01 <sup>Eefg</sup>	0.19±0.00 <sup>Dcde</sup>	0.27± 0.04 <sup>Cd</sup>	0.36± 0.00 <sup>Bd</sup>	0.47± 0.01 <sup>Ad</sup>
Summer savory 1%	0.056±0.01 <sup>Fab</sup>	0.09±0.01 <sup>Efef</sup>	0.10±0.04 <sup>Egh</sup>	0.16±0.00 <sup>Dfgh</sup>	0.22± 0.00 <sup>Cfg</sup>	0.30± 0.02 <sup>Be</sup>	0.40± 0.01 <sup>Ae</sup>
Summer savory 1.5%	0.059± 0.00 <sup>Ea</sup>	0.07± 0.00 <sup>Def</sup>	0.09± 0.00 <sup>Dhi</sup>	0.14±0.01 <sup>CDE-gh</sup>	0.19±0.01 <sup>BCgf</sup>	0.27± 0.00 <sup>Bf</sup>	0.36± 0.01 <sup>Af</sup>
Tarragon 0.3%	0.054±0.01 <sup>Fab</sup>	0.13± 0.01 <sup>Eb</sup>	0.18± 0.02 <sup>Eb</sup>	0.25± 0.00 <sup>Db</sup>	0.36± 0.00 <sup>Cb</sup>	0.47± 0.01 <sup>Bb</sup>	0.61± 0.00 <sup>Ab</sup>
Tarragon 0.5%	0.051±0.01 <sup>Fab</sup>	0.12±0.05 <sup>Ebc</sup>	0.15±0.01 <sup>DEbc</sup>	0.20± 0.01 <sup>Dc</sup>	0.31± 0.01 <sup>Cc</sup>	0.40± 0.02 <sup>Bc</sup>	0.52± 0.01 <sup>Ac</sup>
Tarragon 1%	0.055±0.01 <sup>Fab</sup>	0.10± 0.00 <sup>Ed</sup>	0.13±0.00 <sup>Ede</sup>	0.18±0.05 <sup>Ddef</sup>	0.24± 0.05 <sup>Cef</sup>	0.35± 0.01 <sup>Bd</sup>	0.45± 0.01 <sup>Ad</sup>
Tarragon 1.5%	0.052±0.00 <sup>Fab</sup>	0.09± 0.02 <sup>Eef</sup>	0.12±0.05 <sup>Efgh</sup>	0.17±0.04 <sup>Defg</sup>	0.21±0.002 <sup>Cef</sup>	0.31± 0.05 <sup>Be</sup>	0.42± 0.02 <sup>Ae</sup>
BHA 0.02%	0.046±0.01 <sup>Eb</sup>	0.05± 0.00 <sup>Ei</sup>	0.05± 0.02 <sup>Ej</sup>	0.08± 0.01 <sup>Dj</sup>	0.10± 0.03 <sup>Cj</sup>	0.14± 0.01 <sup>Bh</sup>	0.17± 0.05 <sup>Ah</sup>

and MS Excel programs.

## Results

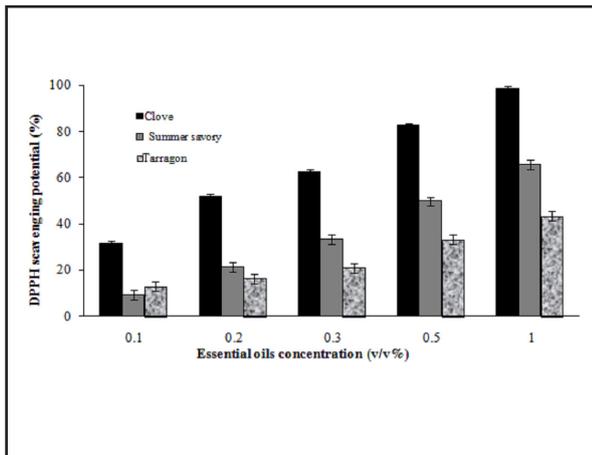
Thirty-two compounds were identified for summer savory EO; the main constituents were thymol (29.10%), carvacrol (26.60%) and  $\gamma$ -terpinen (24.76%). It is worthy to note that the majority of the compounds in savory EO were monoterpene hydrocarbons (more than 80%). Also, 23 and 31 compounds were detected for clove and tarragon EOs. The main compounds present in clove EO were eugenol (76.86%),  $\beta$ -caryophyllene (17.40%), and those in tarragon EO were *Z*-anethole (51.12%), (*Z*)- $\beta$ -ocimene (8.32%) and methyl eugenol (8.06%).

Antioxidant activities of the EOs of clove, summer savory and tarragon were determined by DPPH assay (Fig. 1). In DPPH method, the antioxidants react with the stable free radical 1,1-diphenyl-2-picrylhydrazyl (deep violet color) and convert

it to 1,1-diphenyl-2-picrylhydrazine along with discoloration. The degree of discoloration indicates the strength of free radical scavenging activities of the antioxidant and in the present study it has been found that clove EO at concentration of 1% was able to reduce the stable radical DPPH to 1,1-diphenyl-2-picrylhydrazine up to 98.5%, followed by summer savory and tarragon EOs with 62.8 and 43.4% inhibitory potential, respectively.

The effects of type and concentration of EOs and storage time on the PV of grape seed oil at 60 °C are illustrated in Table 1.

All of the PVs were found to be different among the oil samples. After 4 weeks, important increases in PV were determined but oil samples treated with 1.5% clove EO showed the lowest PV (52.13±0.41 meq/kg) at the end of the period ( $P<0.05$ ). Among EO-treated oil samples, the highest PV was seen in samples treated with tarragon EO



**Figure 1.** The effect of the essential oils on 1,1-diphenyl-2-picrylhydrazyl scavenging.

( $P < 0.05$ ).

Secondary lipid oxidation products were quantified using the TBARS method. The development of TBARS in the oil samples stored at 60 °C is shown in Table 2. There was no significant difference between the TBARS of samples containing 1% clove EO and 1.5% summer savory EO at day 10 of storage ( $P > 0.05$ ). TBARS of clove treated samples increased slightly toward the end of storage and a similar trend in TBARS was observed for samples treated with summer savory EO. TBARS values of tarragon treated samples at each storage time were higher than those for clove and summer savory EOs.

## Discussion

Aromatic plants and medicinal herbs, due to their metabolites such as EOs and polyphenol compounds, are the source of natural antioxidants. These compounds inhibit radical formation by donating hydrogen to highly reactive radicals (Embuscado, 2015). Several studies have focused specifically on the strongly antioxidant activity of clove EO (Ozcan and Arslan, 2011; Shi et al., 2014). The main components of clove EO in this

work were eugenol and  $\beta$ -caryophyllene. It seems the high degree of its antiradical potential probably derived from the hydrogen donating activities exhibited by a wide range of its constituent compounds: eugenol [2-methoxy-4-(2-propenyl) phenol], eugenyl acetate, beta-caryophyllene, 2-heptanone, acetyl-eugenol, alpha-humulene, methyl salicylate, iso-eugenol, methyl-eugenol, phenyl propanoide, dehydrodieugenol, trans-coniferyl aldehyde, biflorin, kaempferol, rhamnocitrin, myricetin, gallic acid, ellagic acid and oleanolic acid (Khaleque et al., 2016).

The major constituents of summer savory EO are reported to be carvacrol, p-cymene,  $\alpha$ -thujene,  $\alpha$ -pinene,  $\beta$ -myrcene,  $\beta$ -terpinene, thymol, linalool, and  $\beta$ -caryophyllene (Hassanzadeh et al., 2016) as seen in the present work. The antioxidant activity of *S. hortensis* suggests that this plant can have the promising potential to be used as a natural compound in food industry to avoid food spoilage and oxidation while increasing the safety of the food products during the processing and also during the storages in various conditions (Hassanzadeh et al., 2016).

In a study conducted by Kordali et al. (2005), the predominant compounds of *A. dracunculus* EO were (z)-anethole (81.0%), z- $\beta$ -ocimene (6.5%), (E)- $\beta$ -ocimene (3.1%), limonene (3.1%) and methyl eugenol (1.8%). Also, it is showed by Ayoughi et al. (2011) that the main compounds in *A. dracunculus* EO were (Z)-anethole (51.72%), z- $\beta$ -ocimene (8.32%), methyleugenol (8.06%), limonene (4.94%) and linalool (4.41%) (2). In the present work, the major components of tarragon EO were Z-anethole (50.12%), (Z)- $\beta$ -ocimene (7.95%) and methyl eugenol (7.66%). In our results, the

amount of (Z)-anethole was high as in other studies and an almost similar amount of z- $\beta$ -ocimene was observed in comparison to the studies above. The different chemical compounds of the EOs might be related to harvest season, geographical situation, ground conditions and genetic parameters (Peter, 2004).

The EOs were capable of scavenging DPPH free radicals (Fig. 1) in a dose-dependent manner through hydrogen-donation converting it to the nonradical hydrazine form. The potential of scavenging DPPH radicals was determined as the clove > summer savory > tarragon. The DPPH radical scavenging activities were 98.5%, 65.8% and 43.4% for clove, summer savory and tarragon, respectively, which suggests that the components within clove EO are more efficient radical-scavenging components. Free radical scavenging potential of clove, summer savory and tarragon EOs has been showed in some literatures and their antiradical effect was mainly attributed to the presence of compounds such as thymol, carvacrol and g-terpinene, eugenol and ocimene (Momtaz and Abdollahi, 2010).

As shown in Tables 1 and 2, during the whole period of the assay, all the EOs showed antioxidant effect in varying degrees on oil samples. Antioxidant activities of clove, summer savory and tarragon EOs were weaker when compared to BHA added samples.

Antioxidant effect increased together with EO concentration. During the experiment, 1.5% level of clove EO showed a marked antioxidant activity, in comparison with BHA (Tables 1 and 2).

The peroxide content of edible oils is considered as one of their quality indices. Higher levels of PV in the edible oils also

results in the lower sensory scores (consumer acceptance, etc.) and also the shelf life of the product. It was shown that type of EO has influenced PVs significantly. All the three EOs used in the present study improved the oxidative stability of grape seed oil in comparison to the control but the different EOs resulted in different levels of PV. Totally, clove EO was more effective in reducing the oxidation rate in comparison to the other two EOs (summer savory and tarragon) and tarragon EO showed the weakest antioxidant activity. After 25 days of storage, a decrease in PV was observed in samples, decrease of PVs within the last five days of storage can be justified by an increase in the rate of hydroperoxide decomposition and forming secondary oxidation products during this period (Boselli et al., 2005). During the storage period, higher PV was observed in control than in EO-treated samples. The results indicate that clove followed by summer savory and tarragon EOs was effective in retarding the formation of hydroperoxide. Other authors have similarly found that phenolic compounds of clove demonstrate strong antioxidant properties (Shi et al., 2014).

TBARS value of all samples increased significantly with the advancement of storage period. Compared with treated samples, control samples showed higher formation of TBARS during storage and clove treated samples showed comparatively lower TBARS value. Similarly, Tajik et al. (2012) and Shi et al. (2014) who observed strong antioxidant activity from clove extract, reported that the antioxidant effect of this extract significantly restrained TBARS value. At a concentration of 1.5%, clove EO was more effective in reducing the TBARS level (0.28 mg MDA/kg oil at the end of stor-

age period) than did the others. At the same concentration, summer savory resulted in a TBARS of 0.36 mg MDA/kg oil but that of the tarragon EO was 0.41 mg MDA/kg oil at day 30. BHT showed the most antioxidant activity among antioxidants. The concentration of 1.5% (v/v) of all three EOs had the highest inhibitory effect on the formation of primary and secondary oxidation products in comparison to other concentrations.

Our results showed that herbal EOs investigated in this study possess considerable antioxidative activity in grape seed oil samples. The antioxidant activity of EOs was as follows: clove > summer savory > tarragon. The concentration of 1.5 % (v/v) of clove EO was the most effective in decreasing peroxide and secondary oxidation products formation rate in grape seed oil samples. However, more studies are needed in order to determine the cytotoxicity and biological activity of the herbal species evaluated in present work.

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### Conflicts of interest

The author declared no conflict of interest.

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## بررسی پتانسیل آنتی اکسیدانی اسانس های میخک، مرزه و ترخون در روغن هسته انگور

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### چکیده

زمینه مطالعه: اتواکسیداسیون یک واکنش غیرقابل برگشت است که تحت تأثیر اکسیژن در هوا رخ می دهد، منجر به ایجاد طعم و بوی ناخوشایند شده و به عنوان نشانگر فساد در روغن شناخته می شود.

هدف: در این تحقیق پتانسیل آنتی اکسیدانی اسانس های میخک، مرزه و ترخون در روغن هسته انگور مورد ارزیابی قرار گرفت. روش کار: تأثیر اسانس ها در غلظت های مختلف (۰/۳، ۰/۵، ۱ و ۱/۵ درصد) بر ارزش پراکسید (PV) و عدد اسید تیوباربتوریک (TBARS) روغن هسته انگور در دمای ۶۰ درجه سانتیگراد مورد بررسی قرار گرفت.

نتایج: نتایج نشان داد که نمونه های حاوی ۱/۵ درصد میخک دارای کمترین عدد پراکسید (۵۲/۱۳ میکروگرم در کیلوگرم) در انتهای دوره بودند. در نمونه های تیمار شده با اسانس، بیشترین PV در نمونه های تحت تیمار با ترخون مشاهده شد. در روز دهم نگهداری، اختلاف معنی داری بین TBARS نمونه هایی که حاوی ۱ درصد اسانس میخک و ۱/۵ درصد اسانس مرزه بودند، وجود نداشت. TBARS نمونه های تیمار شده با اسانس میخک تا انتهای دوره نگهداری به میزان اندک افزایش پیدا کرد و وضعیت مشابهی در نمونه های حاوی مرزه مشاهده شد. مقادیر TBARS در نمونه های حاوی اسانس ترخون در کل دوره ذخیره سازی بیشتر از نمونه های حاوی میخک و مرزه بود.

نتیجه گیری نهایی: فعالیت آنتی اکسیدانی اسانس ها در روغن دانه انگور به شرح زیر بود: میخک < مرزه < ترخون.

واژه های کلیدی:

ترخون، روغن هسته انگور، فعالیت آنتی اکسیدانی، مرزه، میخک