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EFFECTS OF SOME PLANT EXTRACTS ON THE OXIDATIVE STABILITY OF CANOLA OIL AND ITS PURIFIED TRIACYLGLYCEROLS

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ABSTRACT

In this study, the effects of rosemary (*Rosmarinus officinalis L.*), thyme (*Thymus vulgaris L.*), sage (*Salvia officinalis L.*) and bay (*Laurus nobilis*) extracts on the oxidative stability of canola oil and its purified triacylglycerols (TAGs) were studied. The effects of extraction solvents (methanol, ethanol and acetone) on the extraction yields, phenolic contents and antioxidant activities of these plant extracts were also determined. Methanol extracts had the highest total phenolics (78.4–177.4 mg gallic acid equivalent/g extracts), with the exception of thyme extract. In linoleic acid emulsion, antioxidant activities of all plant extracts were above 89.7% at the studied concentrations. The highest 2,2′-diphenyl-1-picrylhydrazyl radical scavenging activities were found for methanol extracts. There was no significant difference between induction periods of canola oils supplemented with ethanol, methanol or acetone extracts of rosemary, thyme and bay ($P > 0.05$) in differential scanning calorimeter. Protection factors of rosemary and thyme extracts were higher than those of the sage and bay extracts in canola oil and its purified TAGs in Rancimat test.

PRACTICAL APPLICATIONS

Rosemary, sage, thyme and bay extracts had high antioxidant and antiradical activities. They contain many bioactive components such as, carnosic acid, carnosol, methyl carnosate, rosmarinic acid, rosmanol, thymol, carvacrol and hydroxycinnamic acid. In addition to this, when the rosemary and thyme extracts were added into canola oil, they increased induction period of oil. These herbal extracts can be incorporated into oils or lipid-containing foods to prevent oxidation.

INTRODUCTION

Lipid oxidation is a major problem for lipids and lipid-containing foods. This process decreases the nutritional value of lipids and also makes the lipids hazardous to human health, by creating molecules such as free radicals. In order to increase the oxidative stability of lipids, the addition of antioxidants is required. Synthetic antioxidants, such as butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) and tert-butyl hydroquinone are widely used because they are very effective. However, recent literature has expressed concerns about the safety and health risks associated with the use of synthetic antioxidants. For this reason, the use of herbs and spices to inhibit the development of oxidative reactions in food systems has recently become popular (Suja *et al*. 2004; Iqbal *et al*. 2008; Gallego *et al*. 2013).

Extensive research studies have been conducted on the antioxidant activity and phenolic components of many species of the *Lamiaceae* family. From these studies, it has been determined that this family of species has a very strong antioxidant capacity (Lagouri *et al*. 2010). The most attention among these herbs and spices as sources of antioxidants has been concentrated on rosemary and sage.

These spices have a significant amount of diterpenoids and triterpenoids, phenolic acids and flavonoids. Carnosic acid, carnosol and rosmarinic acid are the main antioxidant compounds that are found in these spices (Erkan *et al.* 2008; Kontogianni *et al.* 2013).

Thyme and bay are also very common when trying to add a unique aroma and flavor to food. Antioxidant and antimicrobial activities and their phenolic compositions were found in several research reports (Elmastas *et al.* 2006; Lagouri and Nisteropoulou 2009; Gallego *et al.* 2013; Roby *et al.* 2013). Thymol, ferulic and gallic acids were found to be the most abundant phenolic compounds in thyme (Jabri-Karoui *et al.* 2012). The antioxidant activity of the plant extracts largely depends on the composition and concentration of the extracts as well as the test system that is used (Lagouri *et al.* 2010).

There have also been considerable amount of studies on the usage of plant extracts to decrease the rate of lipid oxidation. Abdalla and Roozen (1999) investigated the antioxidant activity of catnip, hyssop, lemon, balm, oregano, sage and thyme extracts in sunflower oil and its emulsion. Bandonienė *et al.* (2002) examined the antioxidant activity of acetone oleoresins and deodorized acetone extracts of sage, savory and borage extracts in rapeseed oil by Schaal oven test and Rancimat method. Babović *et al.* (2010) studied the effect of rosemary, sage, thyme and hyssop extracts on the oxidation of sunflower oil. In another study, sage and rosemary extracts were used to delay lipid oxidation when food was being deep-fried (CheMan and Jaswir 2000).

After purification of the vegetable oils, the antioxidative effects of herbal extracts were also investigated. Rapeseed oil triacylglycerols (TAGs) were used to evaluate the effect of rosemary extract on oxidative stability (Nogala-Kalucka *et al.* 2005). An oil-in-water emulsion was prepared with sunflower oil, which did not contain tocopherols and the antioxidant activity of rosemary, thyme and lavender extracts in this emulsion were reviewed (Gallego *et al.* 2013).

Some research studies have examined the efficiency of pure phenolics in bulk oil or oil-in-water emulsion. Frankel *et al.* (1996) studied the antioxidant activity of a commercial rosemary extract and its active constituents such as, carnosol, carnosic acid and rosmarinic acid. Erkan *et al.* (2009) determined the antioxidative effects of carnosic acid and sesamol on sunflower oil throughout a temperature-controlled microwave heating.

The purpose of the present study was to determine the effects of rosemary (*Rosmarinus officinalis L.*), thyme (*Thymus vulgaris L.*), sage (*Salvia officinalis L.*) and bay (*Laurus nobilis*) extracts on the oxidative stability of canola oil and its purified TAGs. In addition to that, the effects of extraction solvents on phenolic contents and the antioxidant activities were also discovered.

MATERIALS AND METHODS

Plant Materials and Canola Oil

Sun-dried thyme, sage, rosemary and bay were purchased from a spice seller. The leaves, stems and flowers of the plants were separated. The leaves were ground using a coffee mill to pass through a 1-mm sieve. The leaves were then stored in a dark at room temperature until they were used. Refined, bleached and deodorized canola oil was obtained from the local market in Turkey.

Reagents

All chemicals and reagents were of A.R. grade and were obtained from Merck (Darmstadt, Germany). 2,2′- diphenyl -1-picrylhydrazyl (DPPH) reagent, linoleic acid (purity 99%), gallic acid, potassium ferricyanide, trichloroacetic acid, BHT and BHA were purchased from Sigma-Aldrich (St. Louis, MO).

Extraction Procedure

Ten grams of ground and sieved plants were extracted with 150 mL of methanol, ethanol and acetone in a shaking bath for 2 h at 25C. After the filtration, the residue was extracted again with 150 mL of the solvent to make sure the whole extraction of phenolic compounds. The combined extracts were evaporated under a vacuum at 40C to the required dryness using a rotary evaporator. The extraction process was repeated twice for every herb. After that, the extraction yields were calculated. The dried extracts were kept in a refrigerator until they were analyzed.

Total Phenolic Content

The total phenolic contents of the plant extracts were determined by Folin–Ciocalteu method (Iqbal *et al.* 2008). 0.01 g of dried plant extract was dissolved in 10 mL of methanol, ethanol and acetone. 0.2 mL of this extract was mixed with 0.8 mL of Folin–Ciocalteu reagent (freshly 1:3 diluted by volume) and 2 mL of sodium carbonate solution (7.5%). After the addition of 4 mL of distilled water, the tubes were mixed using a vortex mixer and were put in the dark for 2 h. Absorbance was then measured at 765 nm against the control (Shimadzu UV 1700 spectrophotometer, Kyoto, Japan). Gallic acid (Sigma-Aldrich) was dissolved in pure methanol at a concentration of 1 mg/mL and used as a stock solution. The calibration curve was obtained by using different concentrations (0.05–0.25 mg/mL) of gallic acid solutions. The total phenolic content was expressed as mg of gallic acid equivalent (GAE)/g of dry extract and mg of

GAE/g of dry plant. The analyses were done in triplicate and the results were given as average ± standard deviation.

Determination of Antioxidant Activities of Plant Extracts

Antioxidant Activity Using Conjugated Diene Method in Linoleic Acid Emulsion. Antioxidant activities of plant extracts were determined as described by Iqbal *et al.* (2008) and Mau *et al.* (2004). Linoleic acid (0.28 g) and Tween 20 (0.28 g) were mixed with the potassium phosphate buffer (50 mL, 0.05 M, pH 7.4) to obtain a linoleic acid emulsion (0.02 M). Then, 0.2 mL of plant extracts at the concentrations of 250, 500, 1,000 and 2,000 μg/mL were added to 2.5 mL of the linoleic acid emulsion, and 2.3 mL of the potassium phosphate buffer (0.2 M, pH 7.0). However, the solution of control was prepared with methanol, ethanol or acetone, which replaced the plant extract solution. The samples in brown bottles were incubated for 16 h in a dark incubator at 37C. The conjugated diene method was used to measure the degree of oxidation. Before and after the incubation, 0.1 mL of emulsion was mixed with 6 mL of 60% methanol in distilled water and the absorbance of this mixture was measured at 234 nm using a Shimadzu UV1700 spectrophotometer (Shimadzu). BHA and BHT at 200 μg/mL concentration were used for comparison purpose. The analyses were done in triplicate.

The antioxidant activity was calculated as follows:

$$\text{Antioxidant activity (\%)} = \left[\left(\Delta A_{234} \text{ of control} - \Delta A_{234} \text{ of sample}\right)/\Delta A_{234} \text{ of control}\right] \times 100.$$

where, ΔA_{234} was the absorbance increase of control or sample at 234 nm.

Radical Scavenging Activity Using DPPH Method. The radical scavenging activity of the plant extracts was determined using DPPH radical. 0.2 mL of various concentrations (250, 500, 1,000, 2,000 μg/mL) of plant extracts were added to 4 mL of daily-prepared DPPH solution (0.1 mM in methanol). Methanol, ethanol and acetone were used in the control instead of plant extract solution. After 20 min of waiting in the dark, the absorbance at 517 nm was measured by a Shimadzu UV 1700 spectrophotometer against methanol as blank (Nor *et al.* 2008). All analyses were carried out in triplicate. BHA and BHT at 200 μg/mL concentration were used for comparison purpose.

Reducing Power. In this assay, antioxidants in extracts reduce ferric chloride and ferricyanide complex to form ferrous complex. This produces a blue colored product with maximum absorbance at 700 nm. The increased absorbance of the reaction mixture indicated stronger reducing power (Bae *et al.* 2012). The reducing power of plant extract was determined according to Mau *et al.* (2004) with minor modifications. Various concentrations (250, 500, 1,000, 2,000 μg/mL) of plant extracts (0.2 mL) were added to 2.5 mL of 200 mM sodium phosphate buffer (pH 6.6) and 2.5 mL of 1% potassium ferricyanide mixtures. The mixtures were incubated at 50C for 20 min. After the incubation, 2.5 mL of 10% trichloroacetic acid (w/v) were added and centrifuged at 200 × g for 10 min. 5 mL of the upper layer was mixed with 5 mL of deionized water and 1 mL of 0.1% ferric chloride. After waiting 25 min, the absorbance was measured against a blank sample at 700 nm using a Shimadzu UV 1700 spectrophotometer BHA and BHT at 200 μg/mL concentration were used for comparison purpose.

Preparation of TAGs from Refined Canola Oil

Canola oil was purified from natural antioxidants as indicated by Karabulut *et al.* (2008) using activated carbon adsorption and alumina column chromatography treatments. The tocopherol analysis was conducted spectrophotometrically to determine the extent of purification of the TAGs (Wong *et al.* 1988). The purified TAGs, free from tocopherols, were stored at –18C under nitrogen until they were analyzed.

Addition of Plant Extracts into Refined Canola Oil and its TAGs. Each of the plant extracts was added into the refined canola oil (10 g) and its purified TAGs (7 g) at concentrations of 250, 500, 1,000 and 2,000 μg/g and then following this, they are waited at 40C for 10 min in an ultrasonic bath to dissolve the extracts in oil. Each of the samples was stored at –18C under nitrogen until they were used for further analysis.

Determination of the Induction Period Using Differential Scanning Calorimeter. Differential Scanning Calorimeter (DSC) analysis was conducted using a Shimadzu DSC 60. The equipment was calibrated with pure indium and the baseline was obtained with an empty open aluminum pan. The DSC chamber was heated from 50C to 150C at a rate of 10C/min, under nitrogen (50 mL/min) before the experiment. Then induction periods (IPs) of 1.5 ± 0.1 mg a sample in an open aluminum pan were determined to be 150C during isothermal heating of samples under a stream of purified oxygen at 50 mL/min. Another aluminum pan without any sample was used as a reference. Oxidation is an exothermic process in which the heat of reaction involved makes it viable to employ DSC for assessment of oxidative stability of oils. The time at which the onset of oxidation occurred was noted and this period was

taken as an indicator of the oxidative stability of the oil (Suja *et al.* 2004). IP of canola oils supplemented with different plant solvent (methanol, ethanol and acetone) extracts at 2,000 μg/g concentrations was found at these conditions. BHT and BHA were also used at 200 μg/g concentration for comparison. The analyses were performed in duplicate.

Determination of the IP Using Rancimat Apparatus

An automated Metrohm Rancimat model 743 (Herisau, Switzerland) was used for the determination of IPs of canola oil or its purified TAGs supplemented with different plant extracts at the concentrations of 250, 500, 1,000 and 2,000 μg/g according to AOCS method (1998) at 110C. In the experiment, 3 g of sample was used, the flow rate of dry air was 20 L/h and the temperature of the heating block was 110C. IP, the time elapsed from the beginning until the oil starts to become rancid, was measured by drawing tangents on both sides of induction curve, the intercept of which meets the time axis (Iqbal *et al.* 2008). All tests were performed in duplicate. BHT and BHA were used at 200 μg/g concentration for comparison. The oxidative stability was expressed as protection factor (PF) calculated as stated in the formula below (Bandonienė *et al.* 2000):

$$PF = \text{IP of oil withplant extracts} / \text{IP of control (canola oil or purified TAGs)}$$

Statistical Analysis

The statistical analysis was conducted with the SPSS 12.0 package software (SPSS Inc, Chicago, IL). The extraction process was repeated two times for every herb. Results were presented as means ± standard deviation of the two or three replicates of each experiment. The significance was evaluated by using the analysis of variance (ANOVA; one-way and two-way ANOVA). Significant differences among the means ($P < 0.05$) were determined by Tukey's test.

RESULTS AND DISCUSSION

Extraction Yields and Total Phenolics

Extraction yields and total phenolic contents of various plant extracts prepared by different solvents are shown in Table 1. Considering the extraction solvents, the highest extraction yield was found to be by methanol extraction of all the plants, and the lowest yields were found to be by acetone extraction. Among the methanol extracts of studied plants, bay had the highest (19.3%) extraction yields, followed by rosemary (15.6%), sage (10.4%) and thyme (9.1%).

Lagouri *et al.* (2010) reported that the extraction yields of methanol extracts of thyme and rosemary were 14.3% and 19.4%, which were higher than our findings. Pizzale *et al.* (2002) found that the extraction yields of *S. officinalis* and *S. fruticosa* were in the ranges of 18.7% to 27.8%.

The extraction yield varied in each plants and their solvent extracts. Turkmen *et al.* (2006) reported that extraction yield is dependent on the solvent and method of extraction. Roby *et al.* (2013) stated that the highest extraction yield was obtained by methanol extraction of thyme, sage and marjoram. They have also pointed out that the extraction yield of plants was decreasing in the following order: ethanol, diethyl ether and finally hexane.

TABLE 1. EXTRACTION YIELDS AND TOTAL PHENOLICS OF DIFFERENT PLANT EXTRACTS PREPARED BY ETHANOL, METHANOL OR ACETONE

Plants	Solvent extraction	Extraction yield (g/100 g plant)	Total phenolics (mg GAE/g dry extract)	Total phenolics (mg GAE/g plant)
Rosemary	Methanol	15.6	150.0 ±8.9[a]	23.9 ± 0.6[a]
	Ethanol	13.9	116.7 ±4.3[b]	16.2 ± 0.6[b]
	Acetone	10.5	97.3 ± 2.6[c]	10.2 ± 0.3[c]
Thyme	Methanol	9.1	177.4 ± 4.8[b]	16.0 ± 0.3[b]
	Ethanol	8.2	149.1 ± 4.5[c]	12.2 ± 0.4[c]
	Acetone	5.2	194.9 ± 3.2[a]	10.1 ± 0.2[a]
Sage	Methanol	10.4	158.8 ± 3.8[a]	16.4 ± 0.2[a]
	Ethanol	8.1	130.9 ± 7.2[b]	10.6 ± 0.6[b]
	Acetone	6.5	118.8 ± 1.6[c]	7.7 ± 0.1[c]
Bay	Methanol	19.3	78.4 ± 6.0[a]	15.2 ± 1.1[a]
	Ethanol	11.5	40.1 ± 1.3[c]	4.6 ± 0.1[c]
	Acetone	7.2	65.4 ± 5.7[b]	4.7 ± 0.4[b]

The values are expressed as mean ± SD of three independent experiments. The values with different letters are significantly ($P < 0.05$) different between the extraction solvents in the same plant extract.
GAE, gallic acid equivalents.

Phenolic compounds are common in secondary products of edible plants and medicinal plants (Lagouri *et al.* 2010). Phenolic compounds have high antioxidant activity. They are able to terminate free radicals and chelate metal ions that are capable of catalyzing formation of oxygen-reactive species that promote lipid peroxidation. Phenolic compounds restrict the oxidation process of lipids and free radicals by rapid donation of a hydrogen atom or electrons around the aromatic ring (Muchuweti *et al.* 2007).

The total phenolics of the plant extracts ranged from 40.1 mg of GAE/g dry extract to 194.9 mg GAE/g dry extract (Table 1). The lowest amounts (40.1–78.4 mg GAE/g dry extract) of phenolics were found in bay extracts. In a report, Koşar *et al.* (2005) stated that sage and thyme extracts had the most extractable phenolic components, whereas the bay extract had the least among aqueous methanol extracts compared with the other plants of *Lamiaceae* family.

The results showed that methanol extracts contained significantly higher total phenolics ($P < 0.05$) than those of the ethanol and acetone extracts, except for thyme. The solvent order in which phenolics were extracted from *D. hastata*, expressed in terms of total phenolic content, were methanol > water ≥ petroleum ether > acetone ≥ ethyl acetate (Erkan *et al.* 2011). As compared with less polar solvents (hexane and diethyl ether), higher polar solvents (methanol and ethanol) were more efficient in terms of the extraction of phenolic compounds from plant material (Roby *et al.* 2013).

According to the plant species, the phenolic contents of methanol extracts were in the following order: thyme > rosemary > sage > bay. Chen *et al.* (2007) determined total phenolic contents of water extract of rosemary as 185.0 mg GAE/g extract, which was higher than our findings. A study showed that aqueous ethanol extracts of rosemary and thyme contained 219 and 334 mg GAE/g dry extract, respectively (Gallego *et al.* 2013). Furthermore, Lagouri *et al.* (2010) reported that total phenolic contents of thyme and rosemary methanol extracts were 148.3 and 129.5 mg caffeic acid/g extract, respectively.

Rosemary, thyme, sage and bay, with regard to their dry weight, had total phenolics within the following ranges: 10.2–23.9, 10.1–16.0, 7.7–16.4 and 4.6–15.2 mg GAE/g dry plant, respectively. Roby *et al.* (2013) stated that total phenolic content of thyme, sage and marjoram methanol extract were found to be 8.10, 5.95 and 5.20 mg GAE/g dry weight, respectively.

Among selected *Lamiaceae* species (basil, oregano, rosemary, sage, savory and thyme), Koşar *et al.* (2005) found that rosmarinic acid was the major component in all the extracts and was present in the range of 36.3–145.0 mg/g. Cuvelier *et al.* (1994) determined that the sage extract contained carnosol, carnosic acid, rosmanol, rosmadial, epirosmanol and methyl carnosate. Erkan *et al.* (2008) found that rosemary extract contained 6% carnosic acid and 8% of rosmarinic acid. 2-hidroxycinnamic acid and coumaric acid were determined in bay extracts (Muñiz-Márquez *et al.* 2013). Jabri-Karoui *et al.* (2012) reported that *T. capitalus* extract contained 14.25% gallic acid, 14.59% ferulic acid, 9.90% rosmarinic acid, 5.84% quercetin and 27.93% thymol.

Antioxidant Activity in Linoleic Acid Emulsion

Antioxidant activities of various plant extracts prepared by different solvents are given in Fig. 1. The antioxidant activities of plant extracts were close to each other and all of results were above 89.7% at the studied concentrations. Methanol extracts showed the lowest antioxidant activity values among the three solvent extracts in the concentration range tested, which can be attributed to the less effectiveness of polar antioxidants in linoleic acid emulsion (Gallego *et al.* 2013).

Rosemary ethanol extracts had the highest antioxidant activity followed by acetone and methanol extracts at 250 μg/mL. The strong activity of rosemary is related to compounds like carnosol, carnosic acid and rosmarinic acid (Erkan *et al.* 2008). Moreno *et al.* (2006) stated that ethanol and acetone extracts of rosemary had rosmarinic acid, carnosol and carnosic acid with a total yield for these polyphenols of 52.2% and 36.5% of extracts, respectively.

Erkan *et al.* (2008) determined that antioxidant activity was increased in the order of rosmarinic acid < carnosic acid < sesamol for the pure compounds in linoleic acid emulsion. This order was dependent on their hydrophobicity and solubility in linoleic acid emulsions.

Thyme extracts possessed antioxidant activity values ranging from 91.0% to 99.9% between 250 and 2,000 μg/mL. Thyme ethanol extracts showed the highest antioxidant activity values compared with other solvent extracts at all concentrations. The effect of concentration on the antioxidant activity of thyme ethanol extracts was found to be insignificant ($P > 0.05$). Antioxidant activity of thyme methanol extracts was 91.0%, while its acetone extracts had antioxidant activity of 95.9% at 250 μg/mL. The antioxidative effect of the thyme extract is associated with the high content of carvacrol and thymol (Yanishlieva *et al.* 2006).

Sage acetone extracts had the highest activity values at all the concentrations and bay acetone extracts showed the highest values except at 250 mg/mL.

Erkan *et al.* (2011) reported that the highest activity of petroleum ether extract of *D. hastata* in the thiobarbituric acid-reactive substances assay, which was attributed to the

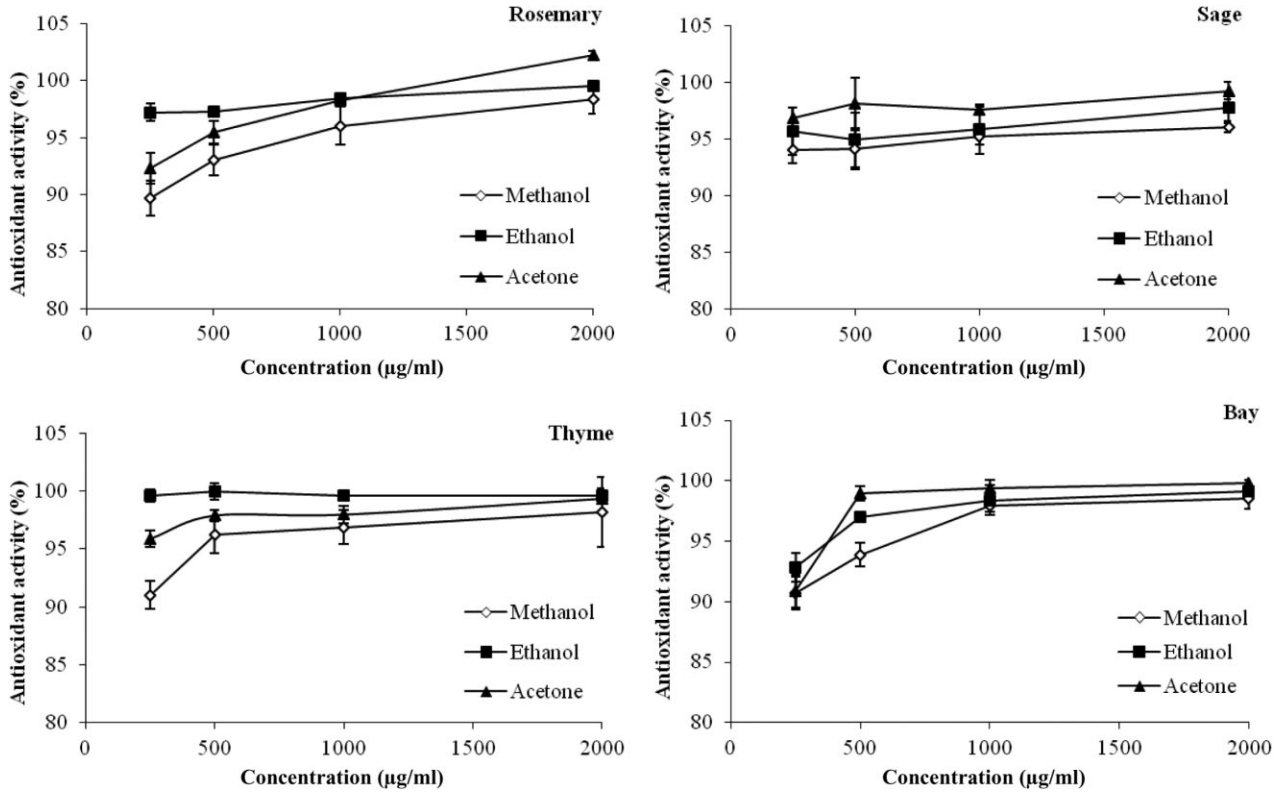

FIG. 1. ANTIOXIDANT ACTIVITY OF DIFFERENT PLANT EXTRACTS PREPARED BY METHANOL, ETHANOL AND ACETONE IN LINOLEIC ACID MODEL SYSTEM

higher solubility of the petroleum ether extract in the lipid phase.

Durling *et al.* (2007) determined that the key indicators of the antioxidant properties of sage extracts were connected to carnosic acid, carnosol and rosmarinic acid. The researchers claimed that the yield of lipophilic and hydrophilic antioxidants was highly dependent on solvent polarity. The mixture of acetone and water was a good solvent for polar antioxidants.

Sage and bay acetone extracts were followed by ethanol and methanol extracts. The antioxidant activity of bay ethanol extract was 92.8% at 250 µg/mL and it increased to 99.1% at 2,000 µg/mL.

Elmastas *et al.* (2006) reported that ethanol extracts of bay leaves at 20, 40 and 60 µg/mL showed 94.2%, 97.7% and 98.6% inhibition of lipid peroxidation in a linoleic acid emulsion, respectively.

On the whole, the antioxidant activities of the extracts were close to each other in all of the concentration levels. A low concentration of the extracts was enough to get the antioxidant activity above 90%.

At 200 µg/mL, BHA and BHT had 97.5% and 98.0% antioxidant activity values, respectively. Plant extracts had antioxidant activities that were comparable to synthetic antioxidants.

DPPH Radical Scavenging Activity

The DPPH radical scavenging assay has been widely used in several studies to evaluate free radical scavenging activities of plant extracts. The method is based on the reduction of alcoholic DPPH solutions at 517 nm in the presence of a hydrogen-donating antioxidant. Polyphenols in the extracts have been reported to be potent hydrogen donators to the DPPH radical (Turkmen *et al.* 2006).

DPPH radical scavenging activities of plant extracts ranged from 3.9% to 92.6% at the studied concentrations (Fig. 2). In contrast to the antioxidant activity values of extracts in linoleic acid emulsion, an increase in concentration led to an increase in radical scavenging activities of extracts. Generally, methanol extracts had higher scavenging activity values than ethanol and acetone extracts. High phenolic contents of methanol extracts compared with other solvent extracts confirmed this result.

Roby *et al.* (2013) determined that the extracts obtained using higher polarity solvents were more effective radical scavengers than those obtained using less polar solvents.

Among the plant methanol extracts, rosemary had the highest scavenging activity on the DPPH radicals. This could be attributed to its high phenolic content. It has been previously demonstrated that there are high correlation

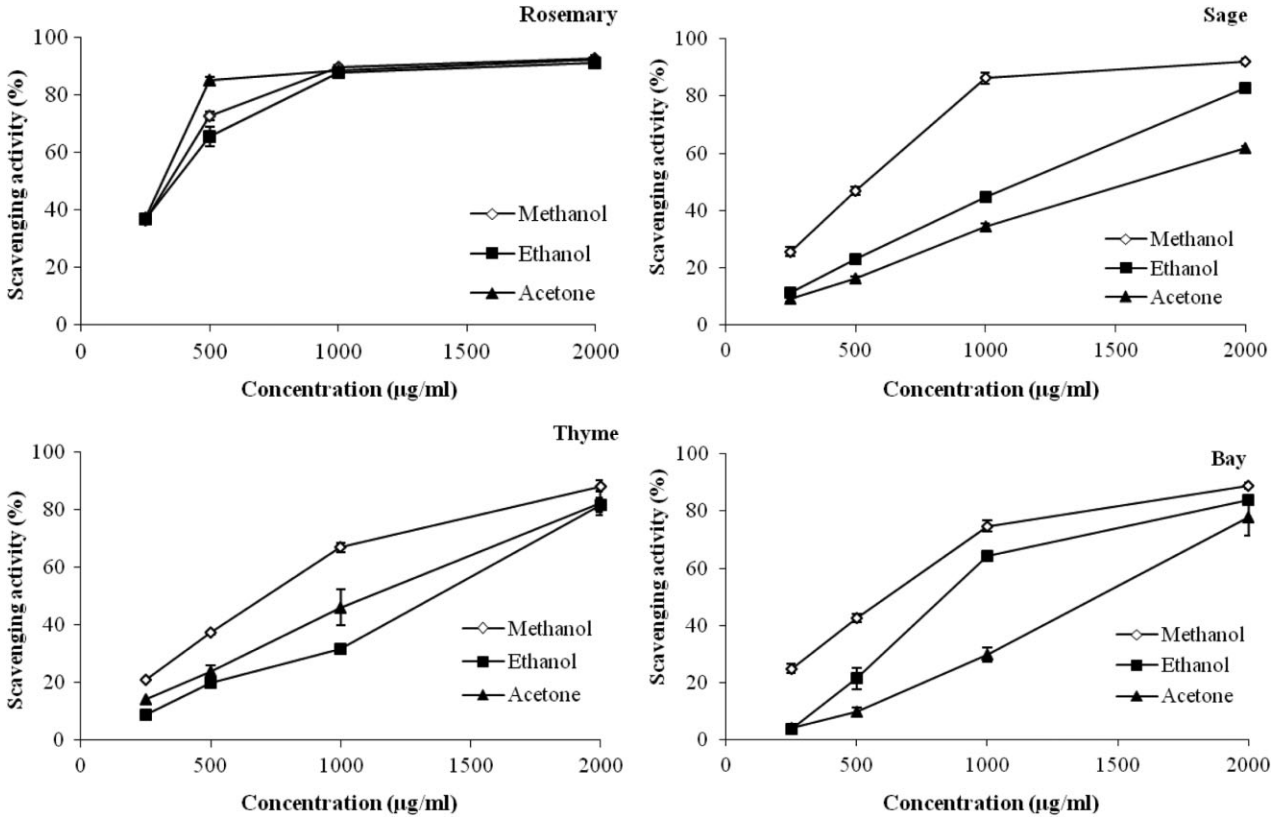

FIG. 2. RADICAL SCAVENGING ACTIVITY OF DIFFERENT PLANT EXTRACTS PREPARED BY METHANOL, ETHANOL AND ACETONE AT VARIOUS CONCENTRATIONS

coefficients between the total phenol content and DPPH radical scavenging activity of extracts (Rababah *et al*. 2011).

Rosemary extract was followed by sage, bay and thyme methanol extracts in descending order. Rosemary acetone extracts had higher DPPH radical scavenging activities (37.2% and 85.2%) than other solvent extracts at 250 and 500 µg/mL. At higher concentrations, all rosemary solvent extracts showed similar values ($P > 0.05$).

Scavenging activities of methanol, acetone and ethanol extracts of thyme sharply increased from 250 to 2,000 µg/mL with values of 20.9–88.1%, 14.2–82.2% and 8.7–81.5%, respectively.

Sage methanol extract had a radical scavenging activity of 92.0% at 2,000 µg/mL. Radical scavenging activity of bay solvent extracts were very low (3.9–25.0%) and increased with increasing concentrations ($P < 0.05$).

Scavenging activities were 81.9% and 59.5% at 200 µg/mL for BHA and BHT, respectively. Rosemary extracts at 1,000 and 2,000 µg/mL concentration had higher radical scavenging activities than these antioxidants. In addition, at 2,000 µg/mL, higher scavenging activities were found for methanolic extracts of sage and bay.

Muchuweti *et al*. (2007) found that radical scavenging activities of bay, rosemary and sage extracts were 91.1%, 88.5% and 88.3% at 5 mg/g concentrations. Koşar *et al*. (2005) determined that rosemary, sage and thyme extracts were significantly superior radical scavengers than basil, bay and oregano extracts. Hossain *et al*. (2011) reported the DPPH radical scavenging activity of rosemary, sage and thyme methanol (80% aqueous) extracts as 11.02, 6.39 and 4.34 g trolox/100 g dry weight of the spices, respectively.

Reducing Power

Reducing powers of plant extracts are given in Fig. 3. The reducing powers of ethanol, methanol, and acetone extracts were found to be significantly different ($P < 0.05$). An increase in concentration caused an increase in reducing power. Thyme and sage ethanol extracts and rosemary methanol extract had higher reducing power than the other plant extracts.

The highest reducing power was found in rosemary methanol extract at 2,000 µg/mL (0.614) followed by rosemary acetone (0.587) and finally bay acetone extract (0.469). BHA and BHT had 0.337 and 0.237 reducing powers at 200 µg/mL. Rosemary methanol extract had 0.330 at 1,000 µg/mL; this value was higher than that of BHA. Rosemary extracts also had higher reducing power than

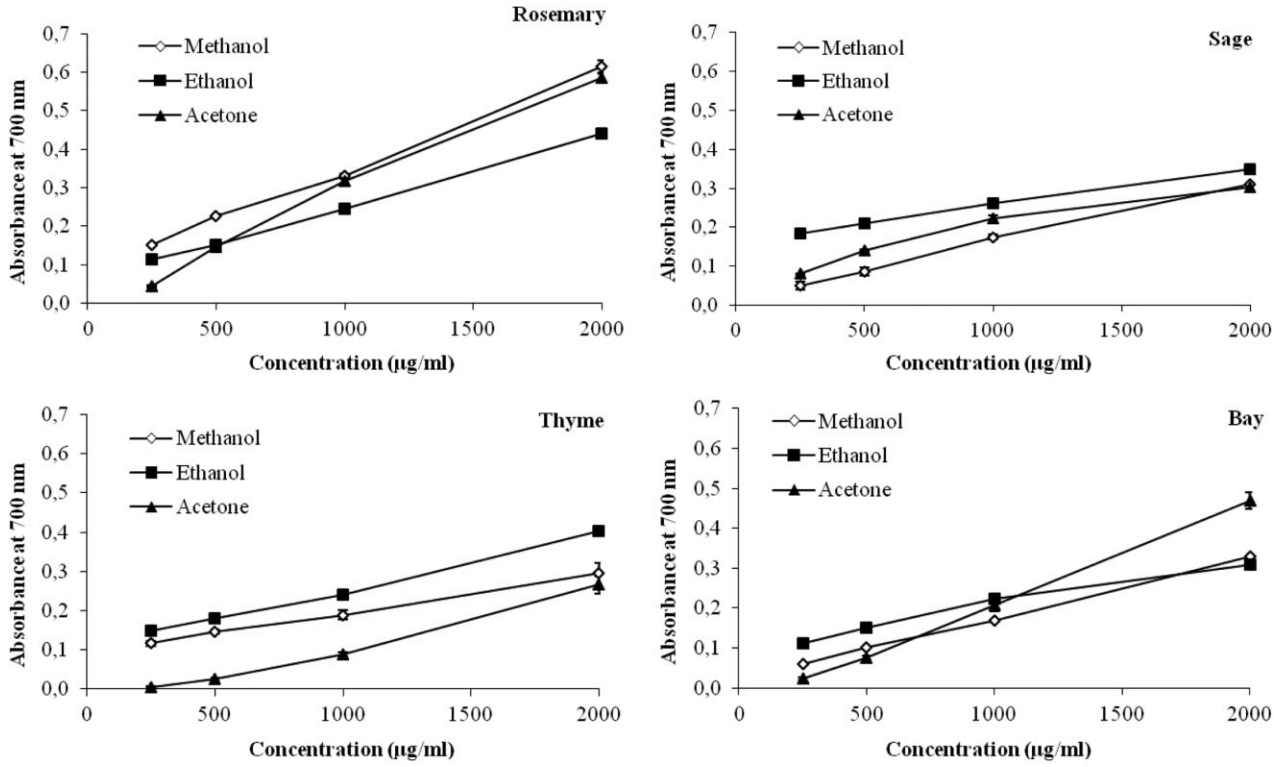

FIG. 3. REDUCING POWER OF DIFFERENT PLANT EXTRACTS PREPARED BY METHANOL, ETHANOL AND ACETONE AT VARIOUS CONCENTRATIONS

BHT at 1,000 and 2,000 µg/mL. Thyme and sage ethanol extracts showed 0.241 and 0.263 reducing powers at 1,000 µg/mL, which had higher values than BHT. Additionally, thyme, sage and bay solvent extracts had higher reducing powers at 2,000 µg/mL than synthetic antioxidants.

The phenolic compounds in methanolic extracts of spices managed to decrease potassium ferricyanide to a ferrous state (Muchuweti *et al.* 2007). They found that the reducing power of plant extracts was changing in the following order: sage > rosemary > mint > sweet basil > bay leaves at the concentration of 25 mg/g.

Arabshahi-Delouee and Urooj (2007) discovered that the most effective reducing agent was methanol extracts of mulberry leaves, while the least effective reducing agent was found to be water extract containing the least amount of phenolics.

Bae *et al.* (2012) stated that the higher reducing power was in direct relation with the amount of total phenolics from different pepper extracts. They discovered that ethyl acetate and acetone extracts had the most reducing power in different pepper cultivars.

Rababah *et al.* (2011) reported that the reason for the differences in the antioxidant activity of the plant extracts could have been due to differences in the methods of estimation, the solvents used and/or the types of phenolic compounds in these extracts

Lagouri *et al.* (2010) claimed that the different polarity of the extracts could have different antioxidant constituents that revealed a variety of reactivity in the different *in vitro* models used in their study. It should also be considered that the level of antioxidants and the synergistic effect happening between them and the other plant constituents could influence the differences in the antioxidant ability of plant extracts.

IPs by DSC

IPs of plant extracts added to canola oils at 150C are given in Table 2. According to the DSC results, addition of plant extracts increased the induction period of canola oil. The highest induction period was found in oils containing rosemary solvent extracts (33.11–31.61 min) followed by sage, thyme and bay extract. Along with rosemary's and sage's antioxidant compounds, carnosic acid is thought to have the highest antioxidant activity. Carnosic acid is a lipophilic antioxidant that scavenges singlet oxygen, hydroxyl radicals and lipid peroxyl radicals, consequently inhibiting lipid peroxidation and causing disturbance of biological membranes (Babović *et al.* 2010).

Canola oil supplemented with rosemary methanol extract or sage acetone extract had higher induction periods than oil containing other solvent extracts. Oil with thyme solvent

TABLE 2. INDUCTION PERIOD (IP) OF CANOLA OILS SUPPLEMENTED WITH DIFFERENT PLANT SOLVENT EXTRACTS AT 2,000 μG/G CONCENTRATION DETERMINED BY DSC AT 150C

Plants	Solvent extraction	IP (min, DSC, 150C)	PF
Control		17.11 ± 0.35	
Rosemary	Methanol	33.11 ± 0.18[a]	1.94
	Ethanol	31.92 ± 0.29[a]	1.87
	Acetone	31.61 ± 0.59[a]	1.85
Thyme	Methanol	25.76 ± 0.39[a]	1.51
	Ethanol	24.73 ± 0.49[a]	1.45
	Acetone	25.85 ± 0.11[a]	1.51
Sage	Methanol	27.94 ± 0.14[b]	1.63
	Ethanol	26.04 ± 0.43[c]	1.52
	Acetone	30.08 ± 0.47[a]	1.76
Bay	Methanol	22.81 ± 0.11[a]	1.33
	Ethanol	22.91 ± 0.06[a]	1.34
	Acetone	22.82 ± 0.17[a]	1.33
BHA	–	21.24 ± 0.14	1.24
BHT	–	18.20 ± 0.29	1.06

The values are expressed as mean ± SD of two independent experiments. The values with different letters are significantly ($P < 0.05$) different between the extraction solvents in the same plant extract.
BHA, butylated hydroxyanisole; BHT, butylated hydroxytoluene, at 200 μg/g concentration; PF, protection factor.

extracts had induction periods between 24.73 and 25.85 min and their protection factors ranged from 1.45 to 1.51. Canola oils containing bay solvent extracts had the lowest induction periods and were the least effective among the studied extracts. Protection factors were 1.33 and 1.34.

According to the statistical analyses, no significant differences were found between the induction periods of canola oils supplemented with methanol, ethanol and acetone extracts of rosemary, thyme or bay ($P > 0.05$). Only the addition of sage extracts led to significant differences in the induction periods ($P < 0.05$). On the other hand, significant differences were discovered IP of canola oils with the same solvent extracts of rosemary, thyme, sage and bay extracts.

The induction periods of canola oil supplemented with BHA and BHT at 200 μg/g was 21.24 min and 18.20 min, lower than that of the plant extracts containing canola oils. These results indicate that even at higher temperatures, rosemary, thyme, sage and bay solvent extracts were capable of protecting canola oil.

IPs by Rancimat

IPs and PF of canola oil or TAGs-containing plant methanol extracts are given in Table 3. As seen in Table 3, induction periods found by Rancimat were higher than the ones by DSC. Moreover, in Rancimat, the induction periods of the oils were given in hours, whereas in the DSC experiment, they were given in minutes. With that being said, this could connect to the difference of the low quantity of samples, high surface–volume relationship between the oil and oxygen and usage of pure oxygen in DSC compared with Rancimat analysis. In fact, DSC would reach an induction period much quicker rather than utilizing Rancimat (Pardauil *et al.* 2011).

TABLE 3. INDUCTION PERIOD (IP) AND PROTECTION FACTOR (PF) OF CANOLA OIL OR ITS PURIFIED TRIACYLGLYCEROLS (TAGs) SUPPLEMENTED WITH PLANT METHANOL EXTRACTS DETERMINED BY RANCIMAT AT 110C

		Canola oil		Purified TAG	
Sample	Concentration (μg/g)	IP (h)	PF	IP (h)	PF
Control		9.39 ± 0.01		1.37 ± 0.06	
Rosemary	250	15.13 ± 0.14[d]	1.61	8.12 ± 0.05[d]	5.92
	500	17.54 ± 0.03[c]	1.87	12.12 ± 0.31[c]	8.85
	1,000	20.16 ± 0.05[b]	2.15	17.74 ± 0.40[b]	12.95
	2,000	22.82 ± 0.08[a]	2.43	22.22 ± 0.14[a]	16.22
Thyme	250	10.03 ± 0.04[d]	1.07	1.41 ± 0.01[d]	1.03
	500	10.53 ± 0.04[c]	1.12	2.77 ± 0.04[c]	2.02
	1,000	11.15 ± 0.07[b]	1.19	4.30 ± 0.33[b]	3.14
	2,000	11.94 ± 0.01[a]	1.27	6.52 ± 0.30[a]	4.76
Sage	250	9.64 ± 0.06[b]	1.03	0.54 ± 0.02[d]	0.39
	500	9.78 ± 0.09[b]	1.04	0.84 ± 0.01[c]	0.61
	1,000	9.89 ± 0.11[ab]	1.05	1.68 ± 0.04[b]	1.22
	2,000	10.24 ± 0.01[a]	1.09	2.64 ± 0.06[a]	1.92
Bay	250	10.32 ± 0.03[a]	1.10	0.52 ± 0.01[d]	0.38
	500	10.27 ± 0.18[a]	1.09	0.67 ± 0.03[c]	0.49
	1,000	10.20 ± 0.02[ab]	1.09	1.00 ± 0.00[b]	0.73
	2,000	9.70 ± 0.03[b]	1.03	1.71 ± 0.04[a]	1.24
BHT	200	9.34 ± 0.03	0.99	4.64 ± 0.50	3.39
BHA	200	9.74 ± 0.08	1.04	7.23 ± 0.10	5.27

The values are expressed as mean ± SD of two independent experiments. The values with different letters are significantly ($P < 0.05$) different in the same plant extract-added canola oils.

Canola oil had an IP at 110C of 9.39 h, whereas its TAGs had 1.37 h. During the purification process of the canola oil, antioxidants such as tocopherols and carotenoids were removed, which led to the decrease in IP. TAGs of edible oils are typically used to assess the anti- and/or pro-oxidant activities of their minor constituents; on top of that, it is also used to study the effectiveness of synthetic and natural antioxidants (Khan and Shahidi 2001).

Similar to DSC results, an addition of extracts increased the IPs of canola oil and TAGs. Generally, an increase in concentration of plant extracts led to a significant increase of IPs ($P < 0.05$). Rosemary was the most effective extract as it was also found to be the same result in the DSC results. It was followed by thyme, sage and bay methanol extracts. The induction period of canola oil containing 250 μg/g rosemary extract was 15.13 and 22.82 h with 2,000 μg/g rosemary extract; the protection factor increased from 1.61 at 250 μg/g to 2.43 at 2,000 μg/g. Gámez-Meza *et al.* (2009) reported that soybean oil containing rosemary extract at 0.5% (w/w) showed an IP higher than 48 h at 110C in Rancimat.

Similarly, when the concentration of rosemary extracts increased from 250 to 2,000 μg/g, induction period of TAGs increased to 22.22 h. This value was very close to the induction period of canola oil containing rosemary extract at the same concentration. Additionally, it had protection factor of 16.22. Rosemary methanol extract was more effective than BHA and BHT in TAGs at all the concentrations and showed higher IPs. Babović *et al.* (2010) found that rosemary extract had higher antioxidant activity than BHA in sunflower oil.

Nogala-Kalucka *et al.* (2005) reported that rapeseed oil TAGs containing 500 ppm rosemary had an IP of 3.9 h in Oxidograph and 7.61 h in Rancimat while the control had an IP of 0.43 and 0.84 h, respectively. They also claimed that rosemary extracts were more effective than synthetic antioxidants and tocopherol isomers in TAGs.

The addition of thyme extract at different concentrations increased the IP of canola oil, but this increase was lower than that of oils containing rosemary extract. Thyme extract increased the IP of canola oil to 11.94 h at 2,000 μg/g. Its protection factor was 1.27. When thyme extracts were added to TAGs at 250, 500, 1,000 and 2,000 μg/g, induction periods were 1.41, 2.77, 4.30 and 6.52 h. The protection factor ranged from 1.03 to 4.76. The thyme extract increased the oxidative stability of canola oil and its TAGs. It was more effective than BHA and BHT at 200 μg/g concentration.

IP of canola oil containing sage extract at the aforementioned concentration ranged from 9.64 to 10.24 h. The highest protection factor of sage extract in TAGs was found to be 1.92 at 2,000 μg/g concentration. However, when it was added to TAGs at 250 and 500 μg/g, IPs of these TAGs were found to be lower than the control. At low concentrations, the sage extract was not effective at increasing oxidative stability of TAGs, and it actually showed a pro-oxidant effect.

To the best of our knowledge, there is no published study about the effects of bay extract on vegetable oil oxidation in literature. Though the concentration of bay extract added to canola oil was changed from 250 to 2,000 μg/g, IP of the oil remained almost constant. At 2,000 μg/g, IP was found to be lower than that of the other concentrations, only slightly increasing the IP of canola oil. Bay extract was effective only at 2,000 μg/g concentration in TAGs. It increased IP of TAGs from 1.37 to 1.71 h.

IP values of canola oils supplemented with different concentrations of sage and bay extracts were close to that of the control. The addition of these extracts did not increase the oxidative stability of canola oil. One of the possible reasons for this could be the pro-oxidant effect of components in extracts with the tocopherol or other antioxidant compounds. Additionally, solubility of the phenolics in oil is also important. Solubility of hydrophilic phenolics in oil may be low.

At 200 μg/g, BHA was more effective in TAGs than canola oil. It increased the IP of TAGs to 7.23 h. Protection factor of BHA in TAGs was 5.27, which was higher than that of BHT amended TAGs (3.39).

Economou *et al.* (1991) reported that the oregano extract was found to be the most effective in stabilizing lard, followed by thyme, dittany, marjoram and lavender extracts in a decreasing order. They stated that the induction period of lard increased with antioxidant concentration. Bandonienė *et al.* (2000) reported that sage and sweet grass extracts were found to be effective in stabilizing rapeseed oil. Pizzale *et al.* (2002) found that IPs of lard containing sage extracts were 10 times higher than control.

CONCLUSIONS

In conclusion, this study showed that rosemary, sage, thyme and bay extracts had high antioxidant, antiradical activities and reducing powers between the concentrations of 1,000 and 2,000 μg/mL because of their phenolic compounds. Each different solvent extract of the plants had a different individual phenolic. It is necessary to review the phenolic profile of the extracts. Methanol and ethanol have similar polarities; furthermore, the polarity of acetone is lower than methanol and ethanol. Acetone extracts of plants contain more lipophilic antioxidants than ethanol and methanol extracts. Methanol led to the extraction of more phenolic compounds than any other solvent that was tested. In the DPPH assay, it was found that the methanol extracts had the highest radical scavenging activity.

Antioxidant effect of plant extracts was examined both in canola oil and its purified TAGs. According to the Rancimat and DSC results, rosemary extract was the most effective extract in stabilization of canola oil at 110C and 150C. TAGs containing rosemary and thyme extracts had longer induction periods than those of BHT or BHA-amended TAGs at 200 μg/g concentration. However, bay solvent extract was the least effective in canola oil. It had higher induction period than control at higher concentrations in TAGs. This is the first published study that indicates the effect of bay extracts on lipid oxidation. Among all of the plants that were studied, it was concluded that rosemary and thyme were most suited for the usage in canola oil against lipid oxidation.

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