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Oxidative Stability of Sunflower Oil Flavored by Essential Oil from *Coriandrum sativum* L. During Accelerated Storage

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Abstract

The aim of the work was to assess the oxidative stability of sunflower oil flavored by the essential oils extracted of *Coriandrum sativum* (coriander, CEO) during the accelerated storage of 24 days. In the study, CEO was extracted by Soxhlet apparatus, and its chemical composition was analyzed by gas chromatography with flame ionization detector (GC-FID) and gas chromatography and mass spectrometry (GC-MS), and then, the evaluation for antioxidant effect in vitro displayed that CEO possessed markedly antioxidant potential. Furthermore, after the addition of CEO, acid value (AV), peroxide value (PV), iodine value (IV), *p*-anisidine value (AnV), thiobarbituric acid reactive substances (TBARS), free fatty acid (FFA), total polar compounds (TPC), tocopherols (TOC) in sunflower oils during the accelerated storage were measured every 4 days, and the results herein exhibited the addition of CEO at 1200 ppm could not only increase the oxidative stability of the sunflower oils, but also exert synergistic effect with TBHQ. Meanwhile, the values of K_{232} and K_{268} and fatty acid composition were investigated. The sensory evaluation of the oil samples revealed that the addition of CEO at 1200 ppm could increase aroma flavor and consumers' acceptability, so that it could be developed as convenient condiment.

Keywords: Sunflower oil; *Coriandrum sativum*; Oxidative stability; Accelerated Storage

1. Introduction

Sunflower oil, as one of the four important edible oils after soybean oil, rapeseed oil and cottonseed oil all over the world, is widely involved in the preparation for daily food due to its high content of polyunsaturated fatty acids (PUFA, 85-95%, Noreen & Ashraf, 2010). Among them, linoleic acid (18:2 n-6), which takes up about 68-72% of the total fatty acid content and displays great hypocholesterolemic action, decreasing the cardiovascular risk (Upadhyay & Mishra, 2015a). Together with the fats, there are plenty of natural antioxidants in the oil, such as α-tocopherol, vitamins A, D and E, which accelerate its oxidative stability (Choi, Park, Kim, Hwang, Song, Choi, Lee, Paik, & Kim, 2013). Unfortunately, because of the high content of PUFA, the oil is more susceptible to oxidative deterioration which not only can produce rancid odours, but also can decrease the nutritional quality and safety, due to degradation products, giving rise to the harmful effects upon human health (Chen, Zhang, Zu., Yang, Lu, & Wang, 2014). In order to control the oxidative degradation of the oil, several methods were exploited, while the most popular method is to add antioxidants into it (Kiralan, Ulaş, Özaydin, Özdemir, Özkan, Bayrak, & Ramadan, 2017).

In terms of antioxidants, there are two kinds of antioxidants in uses, one kind is synthetic antioxidants, and the other kind is natural antioxidants. Among them, the synthetic antioxidants including butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) and tert-butyl hydroquinone (TBHQ) can display inhibitory effects against lipid oxidation of edible oils. However, the employment of these

synthetic antioxidants is critically regulated in terms of dosage and application because of their side effects and potential toxicity (Upadhyay, Sehwag, & Mishra, 2016). Nowadays, together with the most powerful synthetic antioxidant, TBHQ, both BHA and BHT have been restricted in Japan, Canada, USA and many European countries (Upadhyay & Mishra, 2015b). Consequently, searching for natural antioxidants to take the place of synthetic antioxidants has attracted more and more attentions in recent years. Meanwhile, plenty of extracts from spices and herbs has been demonstrated to hinder the oxidative degradation of edible oils (Embuscado, 2015; Blasia, Rocchetti, Montesano, Lucini, Chiodelli, Ghisoni, Baccolo, Simonetti, & Cossignani, 2018; Yanishlieva, Marinova, & Pokorný, 2006). Among them, the essential oils extracted from spices and herbs not only could reveal antioxidant activity to enhance the oxidative stability and thermal stability of edible oils, but also could ameliorate sensory analysis to increase the acceptability of customers (Chandran, Nayana, Roshini, & Nisha, 2017).

Coriandrum sativum L. (of Apiaceae or Umbelliferae family), also called coriander, cilantro and Chinese parsley, is a ubiquitous annual plant worldwide as medicinal plant, culinary spice and flavoring agent for its various medicinal/aromatic usages (Rudra, Shivhare, Basu, & Sarkar, 2008). In Chinese medicine, the whole plant of coriander has been documented in “Bencao Gangmu” and used for cold, measles airtight, and food stagnation (Zhang, Song, & Wang, 2004). In the past few years, many papers have published about its applications as spice or herb. Das et al. reported that the spice contained polyphenolic flavonoids including quercitin, kaemferol,

ramnetin and epigenin, and other polyphenolic compounds including caffeic and chlorogenic acids, and essential oil (Das, Raychaudhuri, & Chakraborty, 2012). Besides, the antioxidant activity of essential oil from coriander have been described repeatedly, indicating that the essential oil could be considered as a source of natural antioxidants and could be used as a potential substitute for synthetic antioxidants (Duarte, Luís, Oleastro, & Domingues, 2016). However, until now, although the plant extracts and seed oils of coriander have been applied to promote the oxidative stability of the edible oil for many times, there is no report about the application of the essential oil from the spice in the influence for the oxidative degradation for sunflower oils (Shyamala, Gupta, Lakshmi, & Prakash, 2005).

Therefore, the aim of this study was to evaluate the antioxidant and protective effects of the essential oil of coriander (CEO) on the oxidative stability of sunflower oil during accelerated storage, and the sensory analysis of the flavored sunflower oils was conducted as well.

2. Material and Methods

2.1 Material and Chemicals

5.0 kg of *Coriandrum sativum* L. plants (including stems, leaves and roots) was bought from New Century Shop Co., Ltd in Lankao County (Henan province, China). The plants were identified by Professor Wenchang Fan in Institute of Chinese Medicine Health Care, Guangdong Food and Drug Vocational College (Guangzhou, China), and its voucher specimens were deposited at the College of Food Science and

Technology, Henan University of Technology (Zhengzhou, China). 5.0 L sunflower oil produced by Henan Sunshine Oils and Fats Group was bought from Hongzhuan Shop of Dennis in Zhengzhou City (Zhengzhou, China). Ascorbic acid (Vit. C), potassium ferricyanide [$K_3Fe(CN)_6$] and trichloroacetic acid (TCA) were obtained from Sigma (Sigma Aldrich GmbH, Sternheim, Germany). 1,1-Diphenyl-2-picryl-hydrazyl (DPPH), nitroblue tetrazolium (NBT), nicotinamide adenine dinucleotide (NADH), and phenazine methosulfate (PMS) were obtained from Applichem (Darmstadt, Germany). Standards used for vitamin E (α-, β-, γ- and δ-tocopherol) were purchased from Merck (Darmstadt, Germany). Standards used for fatty acid (Luaric acid, myristic acid, palmitic acid, palmitoleic acid, stearic acid1, oleic acid, linoleic acid, linolenic acid, erucic acid, tetracosanoic acid) were purchased from (St. Louis, USA). Besides, the reagents and chemical materials used in this study were of analytical/HPLC grade and supplied by chemical suppliers within China.

2.2 Extraction of CEO

After roots removed, *C. sativum* plants were air-dried in the shade at room temperature until a constant weight, and crushed into powder by a grinder. The powder was placed in a flask and then hydrodistilled at 120˚C for 4 h, using a steam distillation apparatus. The resulting essential oil was dried over anhydrous Na_2SO_4 and kept in a dark flask at -4˚C by the refrigerator until the samples were analyzed by gas chromatography with flame ionization detector (GC-FID) and gas chromatography and mass spectrometry (GC-MS) or used in the storage study.

2.3 GC-FID and GC-MS of CEO

According to Msaada et al.' method, the GC-FID and GC-MS of CEO were carried out (Msaada, Taarit, Hosni, Hammami, & Marzouk, 2009). Briefly, for GC-FID, the analysis was performed by a polar Innowax column (30 m × 0.32 mm i.d., f.t. 0.25 mm, Agilent, USA) and an apolar HP-5 column (30 m × 0.32 mm i.d., f.t. 0.25 mm, Agilent, USA) on a Hewlett-Packard gas chromatograph (Agilent, USA) equipped with a FID. The temperature of oven was set as follows: 60-180˚C at 3˚C/min, 180-240˚C at 20˚C/min and held for 8 min. The temperatures of injector and detector were set at 280˚C and 290˚C, respectively. The carrier gas was He at the flow rate of 1 mL/min, and the split ratio was 1:50. For GC-MS, the analysis was performed using an Agilent 6890/5973 system operating at ionization energy of 70 eV, equipped with a HP-5 MS capillary column (30 m × 0.25 mm i.d., f.t. 0.25 mm, Agilent, USA). The carrier gas He at the continuous flow of 1 mL/min, and the split ratio was also 1:50. The temperature of oven was set as follows: 60-180˚C at 3˚C per min, 180-240˚C at 20˚C per min and held for 8 min. The temperatures of injector and detector were set as 280˚C and 290˚C, respectively. The carrier gas He at the flow rate of 1 mL/min, and the split ratio was 1:50. The chemical constituents of CEO were identified by comparison of their retention index (RI) with those reported in these literatures.

2.4 Evaluation of in vitro antioxidant activity of CEO

As described in our previous reports, the antioxidant activity *in vitro* of CEO was evaluated by scavenging effects on DPPH radical (DPPH$^{\bullet}$), hydroxyl radical (HO$^{\bullet}$) and superoxide anion ($O_2^{\bullet-}$), and measuring its reducing power (Wang, Zhao, Jiao, Yu, Yang, & Yang, 2012).

148 *2.5 Preparation of sunflower oil samples flavored by CEO*

149 For the preparation of CEO flavored sunflower oils, the essential oil of *C. sativum*
150 was added directly to refined sunflower oil at concentrations of 300, 600 and 1200
151 ppm (I: CEO-300 ppm, II: CEO-600 ppm and III: CEO-1200 ppm), respectively. The
152 synthetic antioxidant, TBHQ, was also applied at its legal limit of China (200 ppm) as
153 positive control (IV: TBHQ-200 ppm). In order to compare the synergistic or
154 antagonistic effects of them, the mixture of 100 ppm essential oil and 100 ppm TBHQ
155 (V: CEO-100 ppm + TBHQ-100 ppm) was employed. A normal control sample was
156 prepared as well with the same oil without any antioxidant (VI: CEO-0 ppm +
157 TBHQ-0 ppm). After preparation, all the samples (500 mL each) were stored
158 concurrently and respectively in dark brown bottles and stored in an oven at 65˚C.
159 According to Sadeghi et al.'s report, 24 h of storage in temperature of 65˚C is equal to
160 the storage of one month at room temperature (Sadeghi, Karami, & Etminan, 2017).
161 The examinations of the stored sunflower oil were conducted every 4 days for
162 successive 24 days except the analysis of fatty acid composition, which was
163 conducted every 8 days.

164 *2.6 Chemical analysis of sunflower oil samples*

165 *2.6.1 Measurement of thiobarbituric acid reactive substances (TBARS), free fatty*
166 *acids (FFA), total polar compounds (TPC), and tocopherols (TOC)*

167 TBARS were measured on the base of the National Standard (GB/T 5009.181-2003)
168 of China with a little modification (Chen et al., 2014). Briefly, 10 g of oil sample was
169 homogenised in 10 mL of TCA (7.5%)-EDTA (0.1%) solution. The oil sample was

incessantly shaken for 30 min and then immediately filtered. Together with 5 mL of TBA (2.88 g/L) solution, exactly 5.0 mL filtrate was added into a 25 mL colorimetrical tube and heated by a water bath (90˚C) for 45 min to develop pink colour. After cooled for 60 min, the tube was centrifuged at 3000 ***g*** for 10 min. Along with 5 mL of $CHCl_3$, the supernatant was added into another colorimetrical tube and shaken. This mixed solution should stand for at least 1 h, and its absorbance at 532 nm was measured by a spectrophotometer (UV-2550, Shimadzu, Japan). The TBARS was calculated from a standard curve of MDA prepared by acidification of TEP in the range from 0.02 to 0.3 μg/mL.

FFA was measured on the base of the National Standard (GB/T 5009.37-2003) of China with a little modification (Chen et al., 2014). Briefly, 2 g sunflower oil sample was dissolved in 50 mL mixed solution of neutral EtOEt-EtOH (2:1, v/v) to get a mixture, and the mixture was shaken by hand for 20 min. After cooled to ambient temperature, the mixture was titrated against KOH (0.05 M) using phenolphthalein solution (10 g/L) as an indicator. FFA was calculated according to the format: FFA (mg/kg) = ($V \times C \times 56.11$)/m, where V was the volume of KOH exhausted by oil sample (mL), C was the concentration of KOH (M), and m was the mass of oil sample (g).

TPC was measured in the light of the procedure reported before (Sayyad & Farahmandfar, 2017). In brief, dried for 12 h at 160˚C, 63-100 μm of silica gel 60 (95 parts) was mixed with water (5 parts) and intensively shaked (1 min) and stay overnight, and then, 1 g of the silica gel was compressed and filled between two

192 cotton wool balls into a pipette tip (5 mL). 0.5 g oil sample was pipetted into a
193 volumetric flask (5 mL), and then, it was dissolved in 4 mL toluene and filled with
194 toluene. In the hood, 1mL solution was pipetted on top of the pipette tip. The solution
195 was soaked in and then the pipette tip was washed with 1mL eluent. When soaking in,
196 7 mL eluent was added. After 15 min of elution, the end of the tip was washed with
197 500 μL toluene. Finally, the solvent was eliminated and weighted, and TPC in
198 percentage (w/w) was calculated according to the format: TPC (w/w, %) = (W-W_1)/W
199 × 100, where W was the weight of oil sample in mg, and $W1$ was the weight of
200 non-polar compounds in mg.

201 TOC was measured in the light of the procedure reported before (Yang, Song, Sui,
202 Qi, Wang, Li, & Jiang, 2016). In brief, 0.3 g oil sample was dissolved in a 50 mL
203 brown volumetric flask, and diluted with MeOH, n-hexane and THF at a volumetric
204 ratio of 80:10:10. The mixture was vortexed and centrifuged, and an aliquot of the
205 supernatant was injected into a Waters 2695 Alliance HPLC system (Waters, USA)
206 equipped with a C18 column (250 mm × 4.6 mm × 5.0 μm, Waters, USA). A mixture
207 containing ACN and MeOH (55:45, v/v) was used as the mobile phase at a flow rate
208 of 1.2 mL/min. Quantitation of tocopherols was performed using a fluorescence
209 detector at an excitation wavelength of 290 nm with emission at 330 nm. The
210 calculations were conducted using the four tocopherol isomers as external standards,
211 and their concentrations were all calculated based on excitation peak area.

212 *2.6.2 Measurement of acid value (AV), peroxide value (PV), iodine value (IV) and*
213 *p-anisidine value (AnV)*

The measurement of acid value (GB/T 5530-2005), peroxide value (GB/T 5009.37-2003), iodine value (GB/T 5532-2008) and *p*-anisidine value (GB/T 24304-2009) were carried out according to the National Standard of China.

2.6.3 Determination of the absorbance at 232 (K_{232}) and 268 (K_{268}) nm

The determination of absorbance at 232 and 268 nm was obtained according to National Standard of China (GB/T 22500-2008). To be brief, after filtered by experimental gauze for three time, 0.1 g of sunflower clean oil was precisely weighted and steadily added to a 10 mL volumetric flask, and then, the volume of the flask was completed with HPLC-grade isooctane. The absorbance of the solution at 232 and 268 nm was tested to measure the conjugated dienes and trienes, the oxidation product formed during the storage.

2.6.4 Analysis of fatty acid methyl esters

Methyl esterification of fatty acids was carried out according to the National Standard (GB/T 17376-2008) of China with a little modification. Briefly, 350 mg of sunflower oil sample and 6 mL of 0.5% NaOH solution in MeOH was added in a 50 mL flask, and the mixture was heated under reflux and shaken for 7-10 minutes. Then, 7 mL of 20% BF_3 in MeOH was added and the condenser was installed with the reflux continued for 1 min, and 5 mL of n-heptane was added with the reflux continued 2 min. After that, the reflux and the flask were evacuated and an appropriate amount of saturated NaCl was added to the flask. After slightly shaken, saturated NaCl was added again. Finally, after anhydrous Na_2SO_4 was added to get rid of water, the supernatant was transferred to a vial for GC-FID analysis.

236 According to Wang et al.'s method (Wang, Wang, Wang, Yang, Wang & Wu, 2016),
237 the fatty acid composition of the sunflower oil was carried out on a Agilent 7890B gas
238 chromatograph (Agilent, USA) equipped with a FID detector, using a HP-88 column
239 (100 m × 0.25 mm i.d., f.t. 0.2 μm, Agilent, USA). The temperature was set as follow:
240 170-220˚C at 4˚C/min, 220-240˚C at 1˚C/min. The carrier gas He at the flow rate of 1
241 mL/min, H_2 at the flow rate of 30 mL/min and air at the flow rate of 400 mL/min, and
242 the split ratio was 1:50. The temperatures of injector and detector were set at 280˚C
243 and 290˚C, respectively.

244 *2.7 Sensory evaluation of sunflower oil samples*

245 Sunflower oil samples were estimated by 40 semi-trained panelists from consumer
246 representatives (20) of sunflower oil and undergraduate students (20) of Institute of
247 Chinese Medicine Health Care, Guangdong Food and Drug Vocational College,
248 Guangzhou, China. Before the sensory evaluation, all of panelists were trained how to
249 evaluate, and all sunflower oil samples were coded and presented in a randomized
250 arrangement. A 9-point hedonic scale was used to evaluation of aroma flavor and
251 overall acceptability that indicate 9 for extremely well and 1 for unacceptable.

252 *2.8 Statistical Analysis*

253 All experiments were conducted in duplicate. Unless otherwise indicated, the data
254 were reported as mean values of the measurements while presenting the standard
255 deviation values in tables and the standard deviation (SD, n=10) bars in figures. Mean
256 values among the treatment groups were compared by one way analysis of variance
257 (ANOVA) test. Probability values $p < 0.05$ and $p < 0.01$ were regarded as statistically

258 significant ($p < 0.05$) and highly significant ($p < 0.01$), respectively.

259

260 **3. Results and discussion**

261 *3.1 CEO yield and its component contents*

262 In the study, 6.4 g essential oil was extracted from the stems and leaves of
263 coriander, with the extraction yield of 0.41%. As described in Table 1, during the
264 GC-FID/GC-MS analysis, the essential oil was found to contain 37 natural
265 compounds, representing 97.54% of the total essential oil. As seen, the major
266 components of coriander oil were linalool (w/w, 37.12%), geranyl acetate (35.72) and
267 menthol (5.07%), while the other components were quite a little. The results here
268 were in agreement with the literatures that the 37 compounds were identified from the
269 essential oils from the fruits of the plant (Msaada, Taarit, Hosni, Hammami, &
270 Marzouk, 2009). Compared with the literatures, 4 compounds, such as geraniol,
271 δ-elemene, neryl acetate and eugenyl acetate, were not found in this study. The
272 different results may attribute to the different origins of the plant materials, after all,
273 the growing environments of coriander in Tunisia and China were disparate. In
274 addition, in the study for the essential oil extracted from the leaves of coriander from
275 Yuxi County (Yunnan province, China), 41 natural compounds were identified as
276 well (Zhang, Chen, Yang, Zhang, & Yang, 2009). However, the major components of
277 coriander oil were lauraldehyde (14.69%), 9-tetradecenal (13.49%), decanal (13.04%)
278 and 2-dodecenal (9.46%). It means that the growing environments of coriander in
279 Lankao County and Yuxi County were also disparate.

3.2 Antioxidant effects of CEO in vitro

As described in Figure 1, the antioxidant potential of CEO *in vitro* can be appraised with the conventional $DPPH^{\bullet}$, $HO^{\bullet}$, $O_2^{\bullet-}$ systems and ferric-reducing antioxidant power assay. Interestingly, the results showed that the scavenging activity of CEO on $DPPH^{\bullet}$, $HO^{\bullet}$, $O_2^{\bullet-}$ was at a range from about 5% to75%, in a dose-dependent manner, at 10, 50, 100, 150 and 200 μg/mL, respectively. Meanwhile, the positive control, Vit.C, exhibited the scavenging activity of 98.82%, 72.54% and 96.87% against $DPPH^{\bullet}$, $HO^{\bullet}$, $O_2^{\bullet-}$ at 50 μg/mL, respectively. In the ferric-reducing antioxidant power assay, the phenomenon of dose-dependence was obvious as well, with about 0.1-0.5 at tested concentrations of 10-200 μg/mL. What's more, the effect of CEO was slight higher than that of Vit. C at the same concentration (200 μg/mL). In the past few years, although the antioxidant effects of coriander oil have been demonstrated, there were studies reported that coriander oil only exhibited quite low antiradical property. Undeniably, in Singh et al.'s and Duarte et al.' studies, the antioxidant activities of the essential oil, and its main compound, linalool, were demonstrated to be higher in comparison with several different extracts of this plant, or even make them potential alternatives to some synthetic antioxidants (Duarte et al., 2016; Singh et al., 2015). However, in Misharina's exploration, the value for antiradical efficiency of coriander oil was the lowest in the 10 spice oils/extracts (Misharina, 2016). In Wangensteen's investigation, the coriander oil was inactive in DPPH radical-scavenging assay (Wangensteen, Samuelsen, & Malterud, 2004). In China, plenty of researchers have studied the antioxidant effect of plant, but the antioxidant effect of it has attributed to

the flavonoid extract, and none of literatures about the antioxidant effect of coriander oil have been published until now (Yang, Zhu, & Li, 2015). Herein, the results were in consistent with that of Singh et al.'s and Duarte et al.' studies, and the antioxidant effect of coriander oil extracted from Chinese coriander was firstly reported.

3.3 Stability evaluation of CEO on sunflower oil during accelerated storage

The results of TBARS, FFA, TPC and TOC of the sunflower oil samples with and without antioxidants during the accelerated storage were depicted in Figure 2. TBARS, FFA and TPC values of all the samples were seen to markedly increase ($p < 0.01$), while TOC values of all the samples were found to markedly decrease ($p < 0.05$) during the whole storage period. After administrated by CEO at 1200 ppm, the increases of TBARS, FFA and TPC were prominently restricted to 0.20 μg/mL ($p < 0.01$), 0.34 mg/g ($p < 0.01$), 5.87 % ($p < 0.01$), respectively, and the decrease of TOC value was prominently limited to 61.8 mg/100 mL ($p < 0.01$). Meanwhile, the positive control, TBHQ, not only observably decreased the TBARS, FFA and TPC values ($p < 0.01$), but also observably increased the TOC value ($p < 0.01$), while that of CEO-100 ppm + TBHQ-100 ppm group displayed similar variation trend. Shyamala et al. reported that the EtOH extract from coriander leaves was able to exert a protective effect on peroxide formation during the storage of refined sunflower and groundnut oils heated to frying temperature for 4 weeks (Shyamala et al., 2005). Feng Li reported that several extracts of coriander seeds displayed antioxidant effects against the oxidative rancidity of lard, especially the 95% EtOH extract (Li, 2005). Furthermore, oleoresin and steam distilled extract from coriander were found to be

significantly effective in retarding the deterioration of ghee relative to control at the end of 21 days of storage, and the steam distilled extracts revealed higher antioxidant activity compared to oleoresin and BHA deep fat frying (Patel, Shende, Arora, & Singh, 2013). The study here confirmed the antioxidant effects of the plant on the base of these studies, and enriched the natural compounds of the plant which can be used as natural antioxidants during the storage of the edible oils and other foods.

The influences of CEO on AV, PV, IV and AnV values of the sunflower oil during thermal oxidation were depicted in Figure 3. In the study, the antioxidant effects of coriander oil were compared with the positive control, TBHQ. As seen, the addition of CEO at 1200 ppm not only markedly influenced the AV and PV values, but also markedly changed the IV and AnV values of the sunflower oil samples in the accelerated storage. Among them, the AV, PV and AnV values were observably decreased to 0.45 mg KOH/kg ($p < 0.01$), 60.1 meq O_2/kg ($p < 0.01$), and 189.4 ($p < 0.05$) after accelerated storage for 24 days. The IV values was observably increased to 37.8 g I_2/100 g oil ($p < 0.01$). Meanwhile, AV ($p < 0.01$), PV ($p < 0.01$) and AnV ($p < 0.05$) values of TBHQ group also significantly decreased, and IV value significantly increased ($p < 0.01$), while that of CEO-100 ppm + TBHQ-100 ppm group displayed similar variation tendency. Therefore, the CEO at 1200 ppm was able to be used as natural antioxidant in the storage for sunflower oil to some degree, and CEO and TBHQ displayed synergistic effects in their antioxidant effect during the storage for sunflower oil so that they can be used as a mixture. The results here were quite meaningful. In spite of the EtOH extract from coriander seed didn't show enough

antioxidant activity in the expansion of oxidative stability of soybean oil in accelerated techniques including Rancimat and PDSC, the plant could improve the antioxidant effect of annatto during the storage of fish meatballs, evidencing a synergistic effect, probably through a regeneration mechanism of bixin by the phenolic compounds of coriander (Cordeiro, Medeiros, Silva, Silva, Soledade, Souza, Queiroz, & Souza, 2013; Sancho, de Lima, Costa, Mariutti, & Bragagnolo, 2011). Besides, the addition of the mixture of coriander H_2O extract and ascorbyl palmitate revealed a higher antioxidant effect than added them separately in sunflower oil under thermoxidation, demonstrating the synergistic effect under these conditions (Angelo & Jorge, 2008). The combination of natural antioxidant and/or synthetic antioxidant may be more effective in inhibiting the lipid oxidation than the usage of only an antioxidant, and the synergistic effect may lead to the joint action of sequestration of free radicals.

The influences of CEO on K_{232} and K_{268} values were described in Figure 4. After the addition of essential oil at 1200 ppm, K_{232} value was obviously ascended to 5.5 ($p < 0.01$) at the end of the accelerated storage, while K_{268} value was clearly ascended to 1.7 ($p < 0.05$) at the same time, indicating that the formations of conjugated dienes and trienes during the storage process were markedly inhibited. The results were also in agreement with that of AV, PV, IV, AnV, TBARS, FFA, TPC and TOC, verifying the antioxidant effect of the essential oil in the storage of sunflower oil. Consequently, CEO was able to be used as natural antioxidant during the storage of sunflower oil to substitute the synthetic antioxidant, TBHQ (Keramat, et al., 2017).

The influence of CEO on fatty acid composition was described in Table 2. Compared with the normal control group, the addition of CEO at 1200 ppm could markedly restrict the rapid decrease of oleic acid and linoleic acid at the 8th and 16th day of the storage ($p < 0.05$), and obviously inhibit the quick reduce of them at the end of the storage ($p < 0.01$). Furthermore, the addition of TBHQ at 200 ppm or TBHQ at 100 ppm and CEO at 100 ppm were also able to obviously inhibit the decline of oleic acid and linoleic acid at the similar level ($p < 0.05$ or $p < 0.01$). For other fatty acids, the essential oil couldn't significantly change their percentage in sunflower oil. The finding herein confirmed once again that the addition of CEO at 1200 ppm could be used as natural antioxidant to substitute TBHQ for the long term storage of sunflower oil. Although several essential oils have displayed antioxidant effects in sunflower oil, CEO was the first one that could influence the fatty acid composition of the oil (Hashemi, Niakousari, Saharkhiz, & Eskandari, 2011; Hashemi, Niakousari, Saharkhiz, & Eskandari, 2014).

3.4 Sensory evaluation of CEO on sunflower oil after accelerated storage

According to Table 3, the addition of CEO at 1200 ppm obviously ameliorated the aroma flavor and overall acceptability of the sunflower oil after the accelerated storage for 24 days at 65˚C, with 6.5, 6.5, respectively. Besides, the addition of TBHQ at 200 ppm or the combination of CEO at 100 ppm and TBHQ at 100 ppm also could improve the aroma flavor and consumers' acceptance of the sunflower oil, however, there is no significant differences between them and normal group. As known to all, the stems and leaves of the plant *C. sativum* are often used as spice for

cooking in China. In order to obtain convenient condiment in the preparation for daily food, plenty of flavored oils containing the spices have been developed. Interestingly, the study here firstly found that the coriander flavored sunflower oil possessed good oxidative stability, which could promote the rapid growth to a great extent in the developing for the flavored oil.

4. Conclusion

The results of the study revealed that CEO at 1200 ppm was able to inhibit the oxidative rancidity of sunflower oil during the accelerated storage, with its efficient was similar to that of TBHQ, which is considered as the most strongest and popular synthetic antioxidant. Meanwhile, the antioxidant effects of the essential oil *in vitro* were demonstrated as well. Above all, the sensory evaluation indicated that the aroma flavor and consumers' acceptance of the flavored oil markedly increased after the addition of the essential oil. Therefore, the CEO could be used as natural antioxidant to take place of TBHQ whose harmful effects on health have been frequently reported, and the flavored oil could be developed as cooking condiment in the market. Furthermore, the study point out that the addition of essential oils as a source of natural antioxidant can affect the condiment market as they may offer plenty of flavored oils with both good oxidative stability and delicious taste for consumers, after all, the study may be extendable to other essential oils from various other spices and commonly used edible oils. However, the study of the safety of the flavored oil has never been explored, so that there is still much work to be done before the final

412 launch of products.

413

414 **Conflict of interest**

415 The authors declare no conflict of interest.

416

417 **Acknowledgement**

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420

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527

528 **Figure captions**

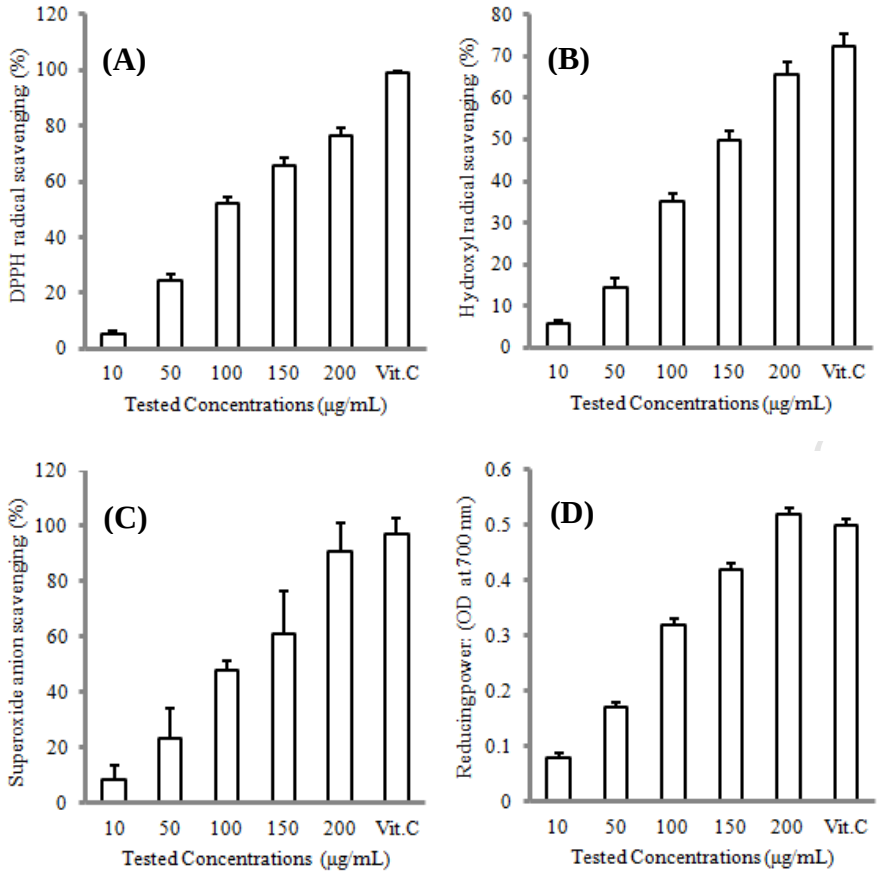
529 **Figure 1.** Antioxidant effects *in vitro* of CEO and positive Vit.C. Values are
530 expressed as means±SD (n=10). (A) $DPPH^{\bullet}$-scavenging activity of various
531 concentrations of ZSP and Vit.C (50 μg/mL). (B) $HO^{\bullet}$-scavenging effect of ZSP and
532 Vit.C (50 μg/mL). (C) $O_2^{\bullet-}$-scavenging capacity of ZSP and Vit.C (50 μg/mL). (D)
533 Reducing power of ZSP and Vit.C (200 μg/mL).

534 **Figure 2.** The influence of CEO on TBARS (A), FFA (B), TPC (C) and TOC (D) of
535 sunflower oil samples during the accelerated storage at 65˚C for 24 days. Values are
536 expressed as means ± SD (n=10).

537 **Figure 3.** The influence of CEO on AV (A), PV (B), IV (C) and AnV (D) of
538 sunflower oil samples during the accelerated storage at 65˚C for 24 days. Values are
539 expressed as means ± SD (n=10).

540 **Figure 4.** The influence of CEO on K_{232} (A) and K_{268} (B) of sunflower oil samples
541 during the accelerated storage at 65˚C for 24 days. Values are expressed as means ±
542 SD (n=10).

543 **Figure 1**



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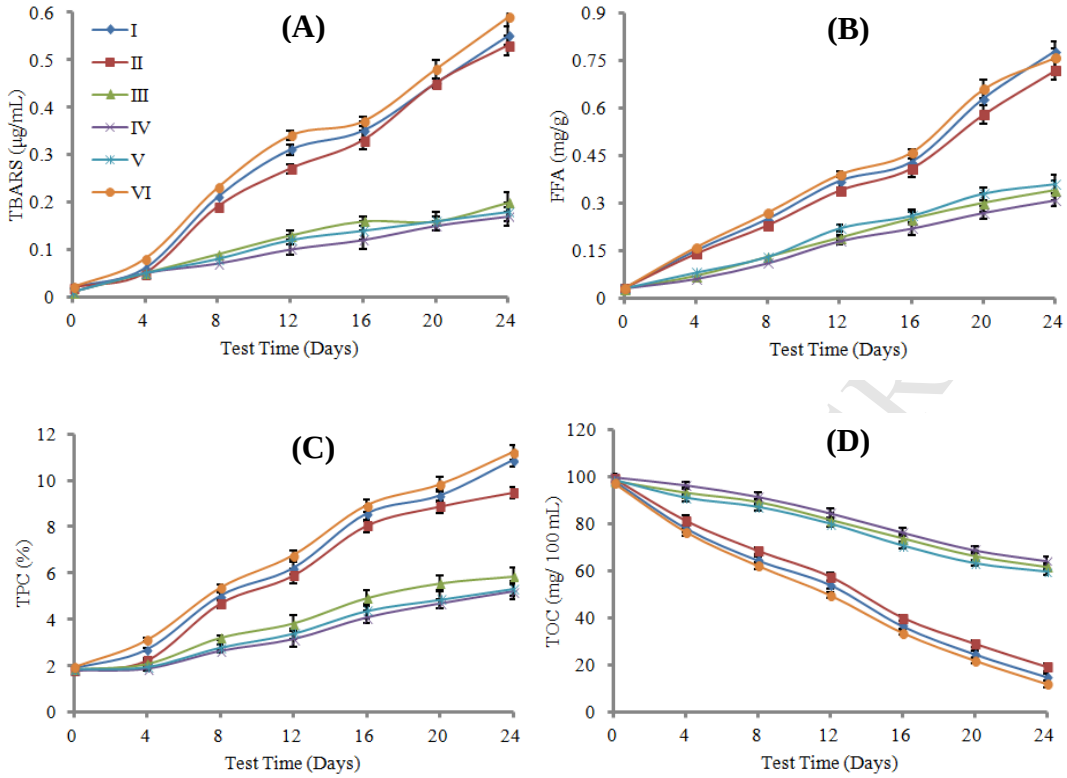
547 **Figure 2**



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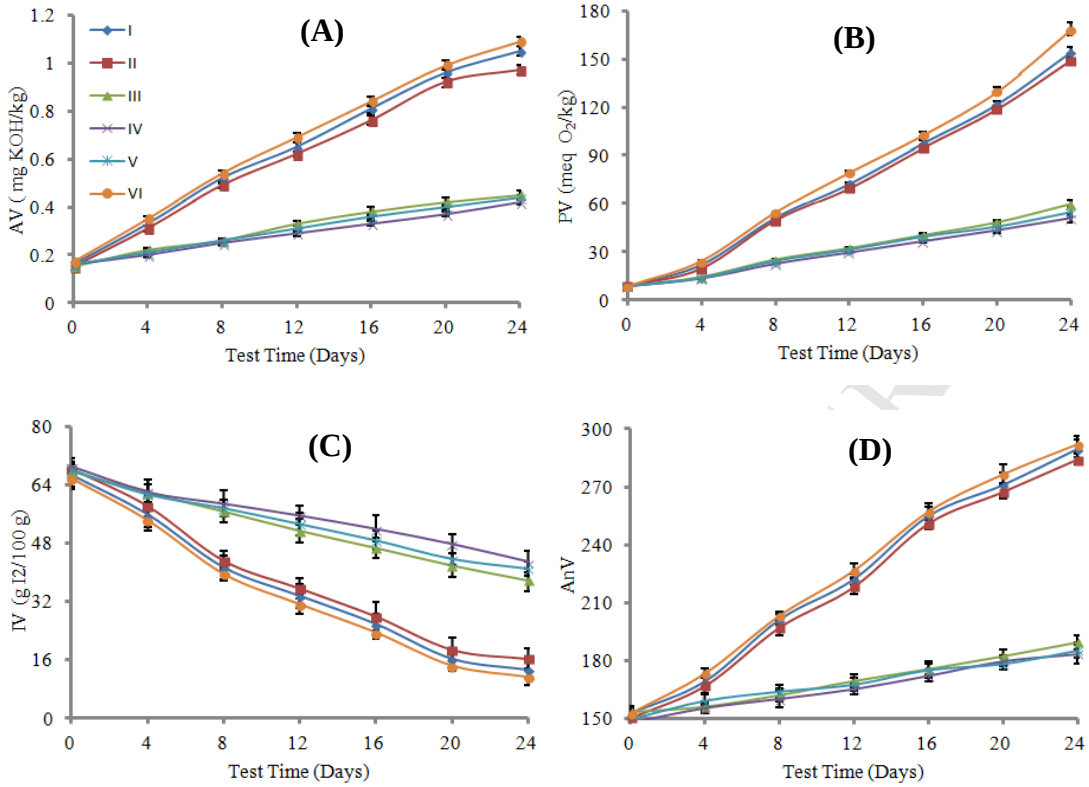
551 **Figure 3**



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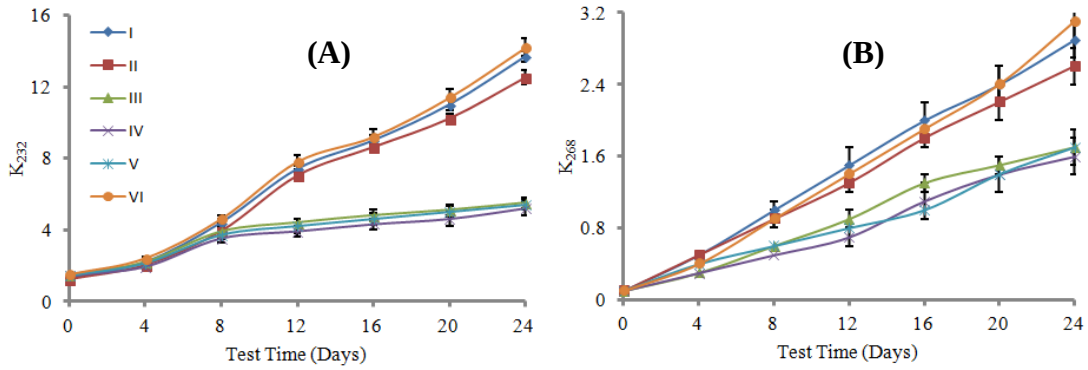
555 **Figure 4**



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558 **Table 1.** Essential oil composition (%) of coriander.

No.	Compound name*	RI[a]	RI[b]	Molecular formula	w/w (%)
1	Heptanal	901	1194	$C_7H_{14}O$	0.05
2	*α*-Thujene	931	1135	$C_{10}H_{16}$	0.11
3	*α*-Pinene	939	1032	$C_{10}H_{16}$	0.44
4	Sabinene	976	1132	$C_{10}H_{16}$	0.46
5	*β*-Pinene	980	1118	$C_{10}H_{16}$	0.36
6	δ^3-Carene	1011	1159	$C_{10}H_{16}$	0.17
7	*α*-Terpinene	1018	1188	$C_{10}H_{16}$	0.12
8	*p*-Cymene	1026	1280	$C_{10}H_{14}$	0.25
9	Limonene	1030	1203	$C_{10}H_{16}$	0.34
10	1,8-Cineole	1033	1213	$C_{10}H_{18}O$	0.45
11	(Z)-*β*-Ocimene	1040	1242	$C_{10}H_{16}$	0.97
12	*γ*-Terpinene	1062	1266	$C_{10}H_{16}$	1.73
13	*cis*-Linalool oxide	1074	1478	$C_{10}H_{18}O_2$	0.61
14	Terpinolene	1088	1290	$C_{10}H_{16}$	0.58
15	*trans*-Linalool oxide	1088	1450	$C_{10}H_{18}O_2$	0.31
16	Linalool	1098	1553	$C_{10}H_{18}O$	37.12
17	Camphor	1143	1532	$C_{10}H_{16}O$	0.64
18	Borneol	1165	1719	$C_{10}H_{18}O$	1.93
19	Menthol	1173	1628	$C_{10}H_{20}O$	5.07
20	Terpinen-4-ol	1178	1611	$C_{10}H_{18}O$	0.03
21	*p*-Cymen-8-ol	1183	1864	$C_{10}H_{14}O$	0.24
22	*cis*-Hex-3-enyl butyrate	1188	1485	$C_{10}H_{18}O_2$	1.22
23	*α*-Terpineol	1189	1706	$C_{10}H_{18}O$	0.09
24	*cis*-Dihydrocarvone	1193	1645	$C_{10}H_{16}O$	1.02
25	Nerol	1228	1797	$C_{10}H_{18}O$	0.13
26	Citronellol	1228	1772	$C_{10}H_{20}O$	0.02
27	Neral	1240	1694	$C_{10}H_{16}O$	0.15
28	Carvone	1242	1751	$C_{10}H_{14}O$	0.21
29	Geranial	1270	1742	$C_{10}H_{16}O$	1.51
30	Anethole	1283	1828	$C_{10}H_{12}O$	0.32
31	Thymol	1290	2198	$C_{10}H_{14}O$	0.06
32	Carvacrol	1292	nd	$C_{10}H_{14}O$	0.75
33	Eugenol	1356	2192	$C_{10}H_{12}O_2$	0.24
34	Geranyl acetate	1383	1765	$C_{12}H_{20}O_2$	35.72
35	*β*-Caryophyllene	1418	1612	$C_{15}H_{24}$	3.88
36	*α*-Humulene	1454	1687	$C_{15}H_{24}$	0.13
37	Germacrene-D	1480	1726	$C_{15}H_{24}$	0.11
	Total identified			97.54	

559 *Order of elution in HP-5 column.

560 [a]RI on apolar HP-5 column.

561 [b]RI on polar HP Innowax column.

562 **Table 2.** The change of CEO on fatty acid composition (%) of sunflower oil [a].

Groups	Days	C12:0[b]	C14:0	C16:0	C16:1	C18:0	C18:1	C18:2	C18:3	C22:1	C24:0
I	0	nd	0.31±0.04	15.19±0.31	0.58±0.05	4.61±0.13	19.18±0.36	58.45±2.22	0.48±0.06	nd	0.22±0.05
	8	nd	0.33±0.03	15.20±0.33	0.55±0.05	4.64±0.15	19.15±0.41	58.38±2.12	0.45±0.04	nd	0.31±0.03
	16	nd	0.35±0.03	15.22±0.32	0.53±0.04	4.66±0.16	19.15±0.39	58.31±2.31	0.41±0.04	nd	0.31±0.03
	24	nd	0.37±0.04	15.27±0.34	0.51±0.04	4.69±0.18	19.11±0.44	58.25±2.26	0.35±0.05	nd	0.33±0.04
II	0	nd	0.30±0.03	15.17±0.34	0.58±0.04	4.61±0.17	19.20±0.35	58.48±2.21	0.49±0.04	nd	0.21±0.05
	8	nd	0.30±0.03	15.20±0.32	0.56±0.06	4.63±0.16	19.19±0.42	58.39±2.25	0.44±0.04	nd	0.29±0.04
	16	nd	0.34±0.05	15.21±0.35	0.52±0.05	4.64±0.13	19.15±0.36	58.32±2.31	0.39±0.06	nd	0.30±0.04
	24	nd	0.38±0.04	15.28±0.36	0.51±0.05	4.68±0.15	19.12±0.38	58.23±2.16	0.34±0.03	nd	0.32±0.05
III	0	nd	0.31±0.05	15.18±0.37	0.57±0.05	4.61±0.16	19.18±0.41	58.48±2.32	0.48±0.04	nd	0.22±0.05
	8	nd	0.34±0.05	15.21±0.31	0.56±0.04	4.61±0.14	19.17±0.42	58.45±2.16[c]	0.45±0.04[c]	nd	0.31±0.04
	16	nd	0.35±0.02	15.21±0.35	0.52±0.04	4.64±0.16	19.12±0.35	58.44±2.22[c]	0.44±0.05[c]	nd	0.32±0.04
	24	nd	0.38±0.03	15.28±0.33	0.51±0.06	4.69±0.13	19.11±0.37	58.42±2.17[d]	0.43±0.05[d]	nd	0.32±0.05
IV	0	nd	0.31±0.05	15.20±0.38	0.59±0.05	4.61±0.12	19.22±0.39	58.49±2.35	0.49±0.04	nd	0.22±0.06
	8	nd	0.32±0.05	15.21±0.32	0.55±0.05	4.61±0.15	19.19±0.41	58.46±2.17[c]	0.46±0.04[c]	nd	0.29±0.04
	16	nd	0.36±0.03	15.22±0.36	0.53±0.05	4.64±0.16	19.15±0.40	58.45±2.41[c]	0.44±0.05[c]	nd	0.29±0.04
	24	nd	0.38±0.03	15.28±0.35	0.51±0.04	4.68±0.14	19.14±0.39	58.43±2.25[d]	0.42±0.03[d]	nd	0.30±0.04
V	0	nd	0.31±0.05	15.19±0.33	0.59±0.04	4.61±0.14	19.21±0.36	58.47±2.19	0.48±0.03	nd	0.21±0.05
	8	nd	0.32±0.05	15.20±0.37	0.54±0.04	4.61±0.15	19.18±0.34	58.45±2.35[c]	0.44±0.04[c]	nd	0.28±0.06
	16	nd	0.36±0.03	15.22±0.32	0.52±0.04	4.66±0.16	19.17±0.41	58.43±2.37[c]	0.43±0.04[c]	nd	0.29±0.06
	24	nd	0.38±0.04	15.27±0.39	0.51±0.05	4.69±0.14	19.13±0.42	58.41±2.25[d]	0.41±0.05[d]	nd	0.31±0.04
VI	0	nd	0.31±0.03	15.18±0.32	0.57±0.05	4.61±0.16	19.23±0.41	58.48± 2.29	0.49±0.06	nd	0.21±0.04
	8	nd	0.32±0.04	15.20±0.35	0.54±0.06	4.64±0.14	19.19±0.35	58.37±2.28	0.44±0.05	nd	0.28±0.05
	16	nd	0.36±0.04	15.21±0.33	0.52±0.04	4.67±0.16	19.17±0.38	58.30±2.32	0.40±0.05	nd	0.31±0.06
	24	nd	0.36±0.05	15.29±0.31	0.51±0.06	4.69±0.15	19.12±0.42	58.24±2.34	0.34±0.06	nd	0.32±0.06

563 [a]Values are expressed as means±SD (n=10).
564 [b]C12:0, luaric acid; C14:0, myristic acid; C16:0, palmitic acid; C16:1, palmitoleic acid; C18:0, stearic acid; C18:1, oleic acid; C18:2, linoleic
565 acid; C18:3, linolenic acid; C22:1, erucic acid; C24:0, tetracosanoic acid.
566 [c]As compared to normal control group at the same day: $p<0.05$.
567 [d]As compared to normal control group at the same day: $p<0.05$.

568 **Table 3**. The changes of CEO on aroma flavor and consumers' acceptability of
569 sunflower oil[a].

	Aroma flavor	Consumers' acceptability
I (CEO-300 ppm)	4.9±0.9	5.2±0.6
II (CEO-600 ppm)	5.0±0.8	5.4±0.8
III (CEO-1200 ppm)	6.5±0.5[b]	6.5±0.7[b]
IV (TBHQ-200 ppm)	6.0±1.0	5.9±0.9
V (CEO-100 ppm +TBHQ 100 ppm)	6.1±1.2	5.9±0.8
VI (Norman Control, No antioxidant)	4.9±0.6	5.4±0.7

570 [a]Values are expressed as means±SD (n = 40).
571 [b]As compared to normal control group: $p < 0.05$.

Highlights:

► The essential oil extracted from coriander was named CEO.

► CEO could exhibit significant antioxidant effect *in vitro*.

► CEO could increase the oxidative stability of sunflower oil during storage.

► CEO could improve the consumers' acceptance of sunflower oil.

► Sunflower oil added with CEO was able to be developed as a flavored oil.