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A comparative study about antioxidant activity and phenolic composition of cumin (*Cuminum cyminum L.*) and coriander (*Coriandrum sativum L.*)

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Apiaceae (*Umbelliferae*) is a large family which involves approximately 455 genus and 3600 species. Coriander (*C. sativum L.)* and cumin (*C. cyminum L.*) are two members of *Apiaceae* family which commonly used for medicinal purpose due to their antioxidant activity. Since the antioxidants are compounds that prevent the oxidation by neutralizing free radicals, researches about their potential utilizations are of great interest for food science and technology. Due to the consumer preference and the worries about synthetic antioxidant compounds, the food industry shows tendency to use natural antioxidant compounds obtained from plant materials. FDA has recognized more than 150 plants as GRAS, without no limitations intake. Coriander, cumin, anise, fennel, thyme and oregano are some plants found in this list. The focus of this research is to contrast the composition of phenolic profile and antioxidant activity of ethanolic and methanolic extracts of these two medicinal herbs belonging to the *Apiaceae* family. For this purpose, coriander and cumin were analyzed for phenolic compounds and antioxidant assay. Antioxidant assay analyses were performed by applying cupric reducing antioxidant capacity (CUPRAC), 2,2'-azino-bis3-ethylbenzothiazoline-6-sulphonic acid (ABTS), ferric reducing antioxidant power (FRAP), 1,1-diphenyl-2-picrylhydrazyl methods (DPPH).

Keywords: Antioxidant activity, Coriander, Cumin, LC-QTOF, Phenolic compounds

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According to epidemiological and *in vitro* researches, foods that comprise of phytochemical components such as phenolics have potential defensive effects against many illness. As a result of this, there is an increasing demand for high dietary content of these phenolic components such as hydroxycinnamic acids and flavonoids by the reason of several biological characteristics; metal chelation activity, free radical scavenging, transition of enzymatic activity, inhibition of cellular proliferation[1]. Actually, these phytochemical components are important for adaptation of plants, especially in the stress conditions, to the environment[2]. Phenolic compounds are the secondary metabolites of plants, with more than 8000 phenolic structures (i.e., several hydroxyl groups on aromatic rings), have been recently determined. These secondary metabolites are generally classified as phenolic acids, flavonoids, terpenes, tannins, stilbenes and lignans[3,4]. Flavonoids like quercetin, naringenin, kaempferol, apigenin are the most common phenolic compounds in nutrition also recognized as primary antioxidants and superoxide anion scavengers.

Antioxidant activity is a parameter that can be used for characterizing plant materials. Free radical inhibitor, oxygen scavenger, peroxide decomposer and metal inactivator are some features of mechanisms of antioxidant activity[5]. Reactive oxygen species formed via oxidative processes are essential for various biochemical mechanisms in the human body, i.e., immune reactions, energy maintenance, detoxification, etc. The instability between antioxidant defense system and the generation of reactive oxygen species leads to 'oxidative stress' which may cause mutation, cell and tissue damage[3,6]. Oxidation, a non-microbial cause of food spoilage, damages to macro molecules such as proteins and lipids[7,8]. The food industry has a propensity to use natural antioxidant components derived from plant materials due to consumer preference and concerns regarding synthetic antioxidant compounds[7,9]. More than 150 plants have been approved as GRAS by the FDA,

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without any restrictions[10]. Several plants such as coriander, cumin, anise, fennel, thyme and oregano included in this list.

Apiaceae (Umbelliferae) is a large family comprising about 455 genus and 3600 species[11]. Coriander (*C. sativum L.),* cumin *(C. cyminum L.),* fennel *(Foeniculum vulgare L.)* and anise (*Pimpinella anisum L.*) are some of *Apiaceae* fruits which have been mainly used as flavoring agents in foods since the ancient times[12]. The *Apiaceae* family is commonly preferred for antioxidant activity worldwide[3]. Coriander (*C. sativum L.)* and cumin (*C. cyminum L.*) are two members of *Apiaceae* family utilized for treating various gastrointestinal problems[14].

C. cyminum L. is a traditional-aromatic plant, originated from Iran and Mediterranean district, has been used for so many years. It is also one of the most popular spice in Asia for both vegetarian and nonvegetarian diets[15,16,17]. The seeds, used for flavoring in pickles, soups, sausages, cheeses, also have numerous medicinal uses such as digestive system stimulant, painkiller in coughs[18,19]. One of the most important functional activities of cumin is its antioxidant activity described in some studies[15,20,21]. Rebey *et al.*[22] and Bettaieb *et al.*[20] described the phenolic profile for seeds, stems, leaves, flowers and roots of cumin.

C. sativum L. is an annual, aromatic-culinary herb and generally cultivated for seeds (fruits) and leaves. The medicinal use of coriander is due to some of its properties such as antibacterial, antifungal, antioxidant activities and digestive agent in the digestion process[23]. Numerous studies in the literature have described the antioxidant features of different coriander tissues such as seeds, shoots, leaves, roots, stems and also the whole plant which were extracted in different solvents such as acetone, methanol, etc.[24,25,26]. Phenolic compounds of coriander were determined for the whole plant, vegetative parts, leaves and stems, seeds[4,27,28,29].

The aim of this study is to contrast the phenolic composition and antioxidant activity of ethanolic and methanolic extracts of coriander (*C. sativum L.*) and cumin (*C. cyminum L.*) which are the two medicinal herbs of *Apiaceae* family. In accordance with this purpose, cumin and coriander were tested for phenolic compounds and antioxidant assay. Analysis of the antioxidant assay was carried out using CUPRAC (cupric reducing antioxidant capacity), ABTS (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid), FRAP (ferric reducing antioxidant power) and DPPH (1,1-diphenyl-2-picrylhydrazyl) methods.

Methodology

C. cyminum L. and *C. sativum L.* seeds were commercially provided by local farmers, cumin seeds were from Konya region and coriander seeds were from Isparta region. All standarts and reagents were purchased from Sigma-Aldrich Chemie GmbH, solvents were from Merck KgaA.

Extraction of samples

Plant materials were ground to 1 mm particle size with FRITSCH mill pulverisette 14 before extraction. Extraction was made according to procedure described by Roby *et al.*[30] with some modifications. One gram from each sample was weighed in 200 mL erlenmayer and then 50 mL of methanol:water (80:20 v/v) or ethanol:water (80:20 v/v) was put into the same erlenmayer. Extraction was carried through at ambient temperature for 24 h in shaking water bath (nüve ST30). At the end of the time, extracts were filtered out Whatman No 4, this step was repeated twice. After filtration, extracts were evaporated and then dried in vacuum dessicator at ambient temperature.

Analysis of phenolic compounds by LC-QTOF

For determining phenolic compounds an Agilent Tech 1260 Infinity LC coupled with quadrupole-time of flight 6550 and UHD accurate mass spectrometer configuration and Poroshell 120 EC-C18 HPLC column (2.7 µm, 46×100 mm) were used. The mobile phases were 5 mM ammonium acetate with deionized water and methanol. The gradient programme was started with 5% methanol, switched to 95% methanol in 25 min and stable with 95% methanol for more 5 min. The other instrument conditions were as follows; constant flow 0.5 mL/min, capillary 3500 V, skimmer 65 V, Q1 130 V, nebulizer gas pressure 45 psi, flow rate 15 L/min, temperature 225°C; sheath gas flow rate 12 L/min, temperature 350°C. Analysis was carried out in negative ionization mode (m/z range 100–1500) and definition was made in auto MS/MS mode. The phenolic components were determined by retention time and mass spectra of certified standards. Quantification was made with calibration curve and the results were expressed as µg per g of dried extract[31,32].

Determination of total phenolic content

The total phenolic content was determined using the Folin - Ciocalteu method, following the procedure of Bettaieb *et al.*[20] with some adjustments. 125 µL herbal extract was mixed with 500 µL of deionized water and 125 µL Folin - Ciocalteu reagent and left for 6 min to allow the reaction realized. After addition of 1.25 mL 7% sodium carbonate (Na_2CO_3), final volume was adjusted to 3 mL with deionized water. It was incubated at 25°C for 90 min, the absorbance was measured at 760 nm with three repetition for each extract. Total phenolic contents were quantified as mg of GAE/g of DW (mg gallic acid equivalents for per g dry weight of plant material). Shimadzu UV-1280 UV-VIS Spectrophotometer was used for all spectrophotometric measurements.

Determination of antioxidant Activity

ABTS Assay

2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid) (ABTS) was obtained after the reaction, carried out between 7 mM ABTS solution and 2.45 mM potassium persulfate ($K_2S_2O_8$) solution at ambient temperature in the dark conditions. After 16 h incubation, ABTS radical solution was diluted with methanol to get an absorbance reading of 0.675±0.025 at 734 nm. With this protocol fresh ABTS solution was prepared for each absorbance measurement. 2.9 mL of the ABTS solution was mixed with 0.1 mL seed extract, left to the incubation at ambient conditions for 30 min, in the dark conditions then the absorbance was obtained at 734 nm[33]. Results were calculated as mM trolox equivalents per kg dry weight of plant material.

DPPH Assay

DPPH (2,2-diphenyl-1-picrylhydrazyl) radical is a reagent in this spectrophotometric antixodant method. 4 mL 0,004% DPPH in methanol was mixed with 1 mL seed extract, incubated for 60 min at 25°C in the dark conditions, followed by measuring the absorbance at 517 nm against methanol. The formula is demonstrated below that used for calculating the inhibition of DPPH radical in percent (I%).

$$I\ \% = ((A_{blank} - A_{sample}) / A_{blank}) * 100)$$

where,

A_{blank} the absorbance of methanol

A_{sample} the absorbance of extract

The results were expressed as mg/mL extract concentration that provides 50% inhibition (IC_{50} %)[34].

CUPRAC Assay

1 mL neocuproine in alcohol (0.0075M), 1 mL NH_4Ac solution and 1 mL $CuCl_2$ solution (0.01 M) were mixed. After that step, (x) mL seed extract, (1.1-x) mL distilled water were added for adjusting final volume to 4.1 mL. Absorbance was measured at 450 nm against a reagent blank after 30 min incubation period. Results were expressed as mM trolox/kg dry weight of plant material through the calibration curve with trolox[35].

FRAP Assay

The extracts were analyzed according to the method described in Jimenez *et al.*[36] to determine if they reduce the (2,4,6-Tris(2-pyridyl)-s-triazine 2) TPTZ–Fe (III). The FRAP working solution was prepared in water at a ratio of 10:1:1 with 300 mM (pH 3.6) acetate buffer, 40 mM TPTZ and 20 mM $FeCl_3 \cdot 6H_2O$ respectively. FRAP solution and herbal extracts were mixed at 1:20 ratio and allowed reaction at 37°C in dark for 15 min. The absorbance was measured at 593 nm. The results are expressed in mM trolox/kg dry weight of plant material.

Statistical analysis

All analysis of each sample were carried out three times and obtained results were showed as means±SD. One-way-analysis of variance (ANOVA) ($p<0.05$) was applied and Pearson's correlation was used to determine the correlation coefficient of total phenolic content and antioxidant activity (Microsoft Excel 2013).

Results and discussion

Pesticide residues, toxic metal, mycotox in analyses were performed in all samples and none of these residues were detected. Methanolic and ethanolic extracts of cumin (*C. cyminum L.)* and coriander (*C. sativum L.*) were analyzed to determine their phenolic content, phenolic compounds and antioxidant activities. Antioxidant activities of extracts are shown in Table 1. As seen in Table 1, total phenolic content of cumin extracts are 7.0±0.2, 3.70±0.25 mg GAE/g DW while the values for coriander extracts are 4.2±0.3, 2.1±0.4 mg GAE/g DW, respectively. It can be said that total phenolic content of cumin, in the case of extraction with the same solvent, is two times more than content of coriander. These results indicate that higher antioxidant activities of cumin (*C. cyminum L.*) extracts compared to coriander (*C. sativum L.*) extracts may be in correlation with the phenolic content of plant. Compared with

Table 1 — Antioxidant activities of extracts

Sample Name	Total phenolic content (gallic acid equivalents mg/g dry weight)	DPPH (IC_{50}) mg/mL	ABTS (mM trolox/kg dry weight)	FRAP (mM trolox/kg dry weight)	CUPRAC (mM trolox/kg dry weight)
Cumin(methanolic extract)	7.0±0.2	1.48±0.03	55.3±1.5	65.0±3.4	57.1±2.5
Coriander(methanolic extract)	4.2±0.3	2.2±0.2	28.4±0.5	36.2±1.7	23.0±1.2
Cumin(ethanolicextract)	3.70±0.25	3.25±0.03	25.2±2.5	31.0±4.5	22.6±1.7
Coriander(ethanolic extract)	2.1±0.4	5.6±0.2	11.4±1.5	16.5±2.3	10.8±1.6

ethanolic extracts, methalonic extracts of cumin and coriander had higher amount of phenolic content, hence showed higher antioxidant activity.

The order of total phenolic content of extracts determined by Folin - Ciocalteu method is as follows: cumin (methanolic extract) > coriander (methanolic extract) >cumin (ethanolic extract) > coriander (ethanolic extract).

The order of antioxidant assay of extracts, independently of applied methods(ABTS, FRAP, CUPRAC or DPPH), were the same with order of total phenolic content results for the same extracts.

Total phenolic content of cumin extracts approximate to the findings of Queralt *et al.*[37] who reported 4.98 mg GAE/g DW for the same parameter. Shan *et al.*[38] found 2.3 mg GAE/g DW total phenolic content for 80% methanol extract while El-Ghorab *et al.*[16] determined 10.6 mg GAE/g DW for hexane extract of cumin, and also Hinneburg *et al*.[39] observed 37.4±0.32 mg GAE/g for aqueous extracts. Secondary metabolism of *C. cyminum L.*, a defence system for biochemical adaptation to environmental changes, may be effected by water deficit significantly[40]. Bettaieb *et al.*[41] reported that phenolic content of cumin increased under severe and moderate water deficit conditions as follows 15.3%, 43.7%, respectively. And also same authors determined that DPPH scavenging activity of cumin seed extracts increased significantly by 17.40 and 64.05% under moderate drought and severe drought, respectively. Ibrahim&Jaafar[42]explained this increasing trend of phenolic compounds, with the accumulation of resoluble carbonhydrates in a consequence of the reduced carry over of resoluble sugars. In a different research by Alinian *et al.*[43] effects of different irrigation regimes on phenolic content of *C. cyminum L.* were studied. They found that 150 and 200 mm regimes increased seed phenolics by 6% and 18%, respectively, while leaf phenolic components rised by 51% and 32%. Additionally, Salami *et al.*[44] and Rebey *et al.*[45] reported a progressive increase on phenolic content during ripening stages in fennel seed and aniseed populations, respectively.

In this research, total phenolic amount of coriander ethanolic extracts was found 2.1±0.4 mg GAE/g DW. Results of Wangensteen *et al.*[46] for the same parameter is similar with that finding. They observed 150 mg GAE /100 g DW for coriander material which was extracted by maceration in 80% ethanol over 24 h. Also Zekovic *et al.*[47] obtained total phenolic amount of microwave assisted(MAE) coriander extracts, in the range of 136.92-384.54 mg GAE/ 100 g DW. Total phenolic content yields obtained by using 50% ethanol 346.35–384.54 mg GAE/100 g DW, are in line with our findings for methanolic extracts of coriander. Gallo *et al.*[48] applied MAE (18 min, 200 W), using 50% ethanol for phenol extraction and determined 82.09 mg GAE/100 g DW. Concentration of ethanol solution may significantly modify the chemical composition of extracts. Lv *et al.*[49] reported a negative linear effect of ethanol concentration on total phenolic content, showing that increasing water amount is advantageous for phenolic extraction.

The similar results with our study were detected in a research carried out by Pavlic *et al.*[50] and the research showed that total phenolic content of coriander seed extracts is 365.44 mg GAE per 100 g DW. In the research total phenolic content was determined by ultrasound-assisted extraction (40min, 60^{o}C, 150 W) and 60% ethanol, which is similar with the results of methanolic coriander extracts. The similarity may be attributed to extraction time that applied in this study. Zhang *et al.*[51] observed phenolic compounds amount increased considerably with prolong extraction time. The positive linear effect of prolonged extraction time showed that there was no significant decrease in total phenolic content with that kind of extraction. In the same line with this pattern, Silva *et al.*[52]found that total flavonoids of chokeberry by-products, extracted by ultrasound, increased with increasing ethanol concentration (up to 50%) then started to decrease with much more increase in ethanol concentration.

Antioxidant activity, expressed as IC_{50} (mg/mL), is the concentration value of test solution required to obtain 50% radical scavenging capacity. IC_{50} for DPPH assay of methanolic coriander extracts was determined 2.2 mg/ mL as seen in Table 1. Martins *et al.*[53] found 1.93 mg/mL for the same parameter, similar to the present finding, however Wangensteen *et al.*[46] reported 510±12 µg/mL for ethanol extracts. In a different study, Zekovic *et al.*[47] determined IC_{50} for DPPH assay of coriander seed extracts as 0.0302-0.0665 mg/mL. The highest antioxidant activity, that means the lowest IC_{50} value (0.0302 mg/ mL), was determined at 400 W irradiation after 15 min extraction period within 70% ethanol. This could be explained by high microwave power which could be an alternative assistant technique for phenolic extraction by spending less time.

Methanolic extracts of cumin showed the highest free radical scavenging activity (1.48 mg/mL), directly related with its high content in phenolic compounds (7.0 mg GAE/g DW). Hinneburg *et al.*[39] determined approximately 2 mg/mL as IC_{50} value of aqueous extracts of cumin. The present findings for the same parameter are 1.48 mg/mL and 3.25 mg/mL. The different results are considered to be observed because of the different solvent (water, ethanol, methanol) application for the extraction. Besides, according to the different studies performed by Thippeswamy & Naidu[54], Einafshar *et al.*[34], Sultana *et al.*[55] the inhibition of DPPH radical in the methanolic extracts of cumin was observed as 0.52 mg/mL, 0.74 mg/mL and 15.48 µg/mL, respectively.

The antioxidant assay results observed against ABTS radical are shown in Table 1, as mM trolox/kg dry weight. Extracts obtained with 80% methanol showed higher antioxidant activity by comparison with 80% ethanol. Antioxidant activities of coriander seeds are as follows; 11.4, 28.4 mM trolox/kg dry weight. In a similar manner Przygodzka *et al.*[56] reported 14.1±2.9 µmol trolox/g dry matter for ethanol/water(1:1) coriander seed extracts. The same authors also observed 5.6±0.5 µmol trolox/g dry matter ABTS radical scavenging activity in 100% ethanol extracts of the seeds. Additionally, Gallo *et al.*[48] determined 2,671 mM trolox/100 g dry weight ABTS radical scavenging activity for microwave assisted extracts of cumin seeds, as in agreement with results of cumin ethanolic extracts. Also in a research by Queralt *et al.*[37], hydroalcoholic extracts of cumin seeds presented 3.26±0.29 mM trolox/g dry weight antioxidant activity through ABTS method. Compared with Table 1 it is seen that this value is ten fold higher than results of cumin extracts. This difference may be attributed to ultrasound assisted solid phase extraction technique applied by Queralt *et al.*[37]

The data obtained with FRAP method are represented in Table 1. According to the results, methanol extracts from cumin seeds showed highest antioxidant activity with 65.0±3.4 mM trolox/kg dry weight, while ethanol extracts from coriander seeds showed the lowest antioxidant activity with 16.5±2.3 mM trolox/kg dry weight. As in agreement with these results, Gallo *et al.*[48] reported 1.198 mM trolox/100 g for ultrasound assisted extracts of coriander seeds.

The CUPRAC (Cupric Reducing Antioxidant Capacity) method was developed by Reşat Apak and his research group. This method is based upon the redox reaction between neocuproine and CUPRAC reagent (Cu(II)-Nc), absorbance of the reaction product (Cu(I)-neocuproine (Nc) chelate) is obtained at 450 nm. With that CUPRAC method Apak *et al.*[57] studied antioxidant activities of several herbal infusions such as coriander seed, green tea, linden flower, common sage etc. They measured antioxidant activity of coriander seed infusion as 0.49 mM trolox/g herbal infusion, also they made this measurement with ABTS method as well and found 0.50 mM trolox/g herbal infusion, which is almost same with CUPRAC result. In the present study, due to the applied solvent, antioxidant assay of coriander extracts was measured as 10.8±1.6 mM trolox/kg dry weight and 23.0±1.2 mM trolox/kg dry weight. Dissimilarity between the measurements can be explained with different extraction techniques, infusion and maceration, applied in two researches.

As seen in Table 2, negative correlation -0,756 was observed between total phenolic content and DPPH results (IC_{50}), which means that IC_{50} decrease,

Table 2 — Correlation between total phenolic content, DPPH, ABTS, FRAP and CUPRAC results

	Total phenolic content	DPPH	ABTS	FRAP	CUPRAC
Total phenolic content		-0,756	0,975	0,969	0,995
DPPH	-0,756		-0,876	-0,882	-0,803
ABTS	0,975	-0,876		0,999	0,991
FRAP	0,969	-0,882	0,999		0,988
CUPRAC	0,995	-0,803	0,991	0,988	

antioxidant activity increases, with increasing total phenolic content. In a similar manner, Zekovic *et al.*[58] determined negative correlation among IC_{50} results and total phenolic amount of coriander seed extracts acquired by subcritical water extraction. Correlation between DPPH and ABTS, CUPRAC, FRAP was negative as well, due to the different mechanism and calculation procedures of methods. The correlation coefficient (R) between other antioxidant assay methods and total phenolic amount were 0.975, 0.969, 0.995, respectively, which shows that high amount of phenolic components means higher antioxidant activity.

The presence of positive correlation between antioxidant activity and total phenolic content of plant extracts has been reported in a wide range of studies[38,59]. In addition good positive correlation was determined between ABTS, CUPRAC and FRAP results as seen in Table 2 pointing out that the measures of antioxidant activity by three different assays were considerably correlated. Applied four antioxidant assay methods are based on electron transferred and mostly preferred for antioxidant activity measurement of natural plant extracts. This kind of reaction occurred between oxidant/radical and antioxidant. Radical takes an electron, which causes color change, from the antioxidant. Degree of color change depends on antioxidant concentration[60].

Quantitative analysis of extracts was carried out with LC-QTOF-MS and obtained results (as µg/g) are shown in Table 3. The compounds gallic acid, vanillic acid, caffeic acid, syringic acid, 4-coumaric acid,

Table 3 — Phenolic profiles of samples (µg/g)

Compound Name	Retention Time	Molecular ion [M-H]− (m/z)	MS / MS Fragments	Cumin		Coriander	
				Methanol	Ethanol	Methanol	Ethanol
Gallic acid	1.91	169.0136	125.0243	ND	ND	ND	ND
Vanillic acid	2.28	167.0344	167.0348 152.0113 123.0450	3.30±0.20	2.23±0.10	3.10±0.10	2.10±0.08
Caffeic acid	3.12	179.1495	59.0115 87.0059 161.0430	0.10±0.02	1.44±0.05	ND	13.75±5.25
Syringic acid	2.87	197.0449	179.0337 135.0445 123.0349	ND	0.15±0.06	0.26±0.04	ND
4-coumaric acid	3.72	163.0339	147.0438 119.0497 91.0544	2.34±0.15	2.86±0.25	2.78±0.25	2.26±0.15
Neochlorogenic acid	5.23	353.0872	191 179 135	137.29±11.51	8.47±0.30	33.00±6.56	9.48±0.35
Chlorogenic acid	5.33	353.0872	173.4700 135.0452 191.0587 179.0361	76.51±11.50	7.98±1.25	155.96±5.03	8.28±1.65
Ferulic acid	5.23	193.1761	193.0505 178.0264 134.0369	1.86±0.23	0.31±0.08	1.15±0.15	0.47±0.05
3-coumaric acid	5.17	163.0324	163.0401 119.0503 135.0452	0.34±0.06	0.01±0.003	0.18±0.04	0.15±0.002
2-cumaric acid	6.56	163.0333	119.0501 162.8392	1.33±0.05	0.15±0.01	0.45±0.13	0.22±0.02
Epigallocatehin	6.25	305.0666	125.0245 179.0353 305.0667	0.11±0.01	0.06±0.001	0.60±0.15	0.05±0.001

(*Contd.*)

Table 3 — Phenolic profiles of samples (µg/g) (*Contd*.)

Compound Name	Retention Time	Molecular ion [M-H]− (m/z)	MS / MS Fragments	Cumin		Coriander	
				Methanol	Ethanol	Methanol	Ethanol
Catechin	8.01	289.2602	289.0712 123.0456 109.0301	1.04±0.15	3.13±0.90	4.10±0.27	3.46±0.80
Epigallocatechin 3gallate	8.85	457.0779	459.0917 460.0949 289.0705	5.13±0.10	0.11±0.04	0.64±0.05	1.49±0.40
Epicatechin	9.63	289.0711	221.0815 123.0449 125.0240	0.10±0.03	0.75±0.10	0.03±0.01	1.75±0.30
Naringin	13.44	579.1713	119.0501 151.0032 271.0605	7.97±0.50	4.61±0.75	5.25±0.50	1.18±0.04
Trans-resveratrol	12.11	227.0715	185.0557 159.0834 143.0483	0.16±0.04	0.12±0.03	0.90±0.18	0.03±0.005
Myricetin	14.45	317.0297	151.0037 178.9988 137.0244	4.87±0.20	0.73±0.02	ND	0.19±0.03
Cis-resveratrol	15.95	227.0707	185.0557 159.0834 143.0483	0.31±0.06	3.52±0.80	7.55±0.50	0.27±0.03
Quercetin	16.29	301.0348	151.0034 121.0291 107.0140	1.14±0.11	0.56±0.01	17.89±0.15	4.83±1.20
Luteolin	16.81	285.0398	174.9697 199.0401	130.11±9.02	120.35±10.05	5.97±0.50	2.01±0.08
Kaempferol	17.01	285.0398	285.0329 286.0397	17.94±1.00	4.99±0.5	6.30±0.25	2.52±0.02
Apigenin	18.00	269.0449	151.0033 225.0544 119.0492	ND	ND	ND	ND

neochlorogenic acid, chlorogenic acid, ferulic acid, 3-coumaric acid, 2-coumaric acid, epigallocatechin, catechin, epigallocatechin-3-gallate, epicatechin, naringin, trans resveratrol, myricetin, cis resveratrol, quercetin, luteolin, kaempferol and apigenin were identified by comparison to the retention time (RT) and mass spectra of those 22 authentic phenolic standards.

Chlorogenic acid and neochlorogenic acid are the most abundant phenolic compounds, followed by luteolin and kaempferol. Gallic acid and apigenin were not determined in any sample. The main phenolic acids in all seed extracts are chlorogenic and neochlorogenic acid, which ranged from 7.98 µg/g to 155.96 µg/g. Levels of these two phenolic acids are almost same in all ethanolic extracts independently of plant species, while they differ from 33.0 µg/g to 155.96 µg/g in methanolic extracts. According to Fig. 1 which shows the three most abundant components found in extracts, concentration of neochlorogenic acid was the highest amount of all components in methanolic extract of cumin. On the other hand, chlorogenic acid was determined as the most abundant compund in methanolic coriander seed extract. Concentration of chlorogenic and neochlorogenic acid were 3,5-19 folds higher in methanol extracts in comparison with ethanol extracts; that indicates methanol is much more proper for these two acids extraction from cumin and coriander. On the contrary of this template, luteloin levels in methanolic and ethanolic extracts of cumin were almost same. Additionally, caffeic acid was not determined in methanol extracts of coriander while it was the most abundant phenolic in ethanolic coriander

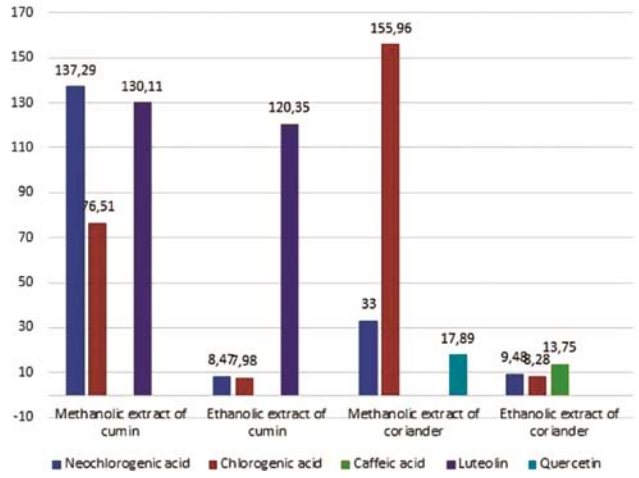

Fig. 1 — Three most abundant components found in extracts

extract. And also, the highest levels of quercetin and kaempferol were observed in methanol extracts of coriander and cumin, respectively. In the case of vanillic acid and coumaric acid derivative levels, they are much the same again. Coumaric acid is an essential component which can be present in all spices and plants, usually with p-hydroxybenzoic and chlorogenic acids[38,49,61]. The phenolic components of plants such as flavonoids, coumarins and phenolic acids may differ in varieties of the same species. From this point of view, there is an increasing interest to use phenolic compounds for taxonomic purposes or identifying the adulteration in food products[62].

Queralt *et al.*[37]quantified catechin and epicatechin in cumin with levels 14.1 µg/g DW, 6.43 µg/g DW, respectively that are higher than present findings. On the other hand, in the same study levels of p-coumaric acid and chlorogenic acid were reported as 0.74 µg/g, 4.18 µg/g, respectively. As can be seen in Table 3 those levels are lower than the amounts of related compounds. Rebey *et al.*[63] used two different extraction methods, maceration and soxhlet, for determining their effects on phenolic compounds of cumin. They did not report gallic acid in maceration extracts that is in line with this study. Also they did not determine catechin in extracts of cumin which were obtained by soxhlet method. Sulaiman *et al.*[26] stated the main flavonoids of methanolic coriander seed extracts as apigenin, quercetin and kaempferol. As in line with that research quercetin and kaempferol were the most abundant phenolics in coriander methanolic extracts, while apigenin was not determined in any samples of this research. Additionally, Barros *et al.*[27] described that phenolic components of coriander fruits mostly consist of, i.e., chlorogenic acid, ferulic acid, p-coumaric and caffeic acids. The same authors reported that vegetative tissues and fruits of coriander showed different phenolic profiles. As major phenolic components, vegetative tissues comprised of hydroxycinnamic acid, quercetin and kaempferol derivatives, while seeds just contain hydroxycinnamic acid derivatives. In the literature there are a wide range of studies that describe quercetin derivatives as being the major components in vegetative parts of coriander[64,65,66]. Amount of caffeic acid is 13.75 µg/g DW in ethanol extracts of coriander seeds while kaempferol level is at the range of 2.52-17.94 µg/g DW in all extracts. Caffeic acid and kaempferol were determined by other authors[38]. The same authors observed main components of cumin phenolics as coumarin derivatives, caffeic acid, kaempferol, flavonoid deriatives and essential oils. Differences in phenolic compound levels can be explained by genotypic factors, growing conditions, and also the plant tissue which was analysed. In addition to this, type and amount of phenolic components may be different due to the factors such as different species, growing practices, geographical origins, post harvest practises and processing procedures, environmental effects, climatic differences, seasonal changes[67]. Lower temperatures in higher altitude can result in increasing the rate of biosynthesis of some kind of antioxidants[68]. Likewise, Sytar *et al.*[69] reported that the accumulation of phenolic acids, anthocyanins and flavonoids in lettuce species increased in direct sunlight in comparison to high temperature and low UV radiation conditions.

As seen in Table 1, extracts that include high amount of phenolic compunds showed high antioxidant activity. Cumin methanolic extract, that showed high antioxidant activity, also had high amount of phenolic constituents. In the similar manner, coriander extract showed the lowest antioxidant activity consequently the lowest level of total phenolic content. From this point of view, it can be said that phenolics present in medicinal herbs may be the most important constituents since they have influence over the antioxidant activity[70]. The good positive correlation among these two phytochemical parameters of medicinal herbs was previously revealed by Aliakbarlu *et al.*[71], as well.

In this study, total phenolic content and antioxidant activity of 2 *Apiaceae* species (*C. cyminum L.*, *C. sativum L.*), extracted with two different solvents,

were evaluated using 4 model systems. *C. cyminum L.* showed higher phenolic content and antioxidant activity in comparison with *C. sativum L*. Variations observed in total phenolic components and antioxidant activity of the same plant species, may be ascribed to used extraction solvent, varieties of herbs and cultivation conditions such as location, fertilizers and climate[72].

Conclusion

Despite the fact that they belong to same botanical family, some differences were observed in phenolic profile composition and antioxidant activities of these two plants. Production procedures, climatic changes such as average precipitation, harvesting time, altitude, storage conditions significantly influence the composition of phytochemicals in plants. For determining an optimum harvest time, ratio and antioxidant activity of these bioactive compounds are important. Also *in vivo* studies of the medicinal plants are needed to check the modes of action in living organism. Thus, their use as dietary supplement might be possible. In addition to these, medicinal plants are safe and precious sources with their potential antioxidant activities for food industry. In the study, cumin showed the highest antioxidant activity and total phenolic content that make it natural antioxidant additive material for utilizing in foods to replace synthetic ones, which have side effects such as carcinogenecity. Further studies are necessary to apply these natural antioxidants in various food systems.

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