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Effects of Crop Season and Maturity Stage on the Yield and Composition of Essential Oil of Coriander (*Coriandrum sativum* L.) Fruit

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ABSTRACT

The aim of this study was to determine the yield and chemical composition of the essential oil (EO) extracted from fruits of coriander (*Coriandrum sativum* L. cv. 'Menzel Temime' with high essential oil yield: 0.35%, w/w) as affected by two successive crop seasons and different stages of maturity using gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS). The maximal oil yields (0.17 and 0.32%) reached at the final stage of maturity in the season 2003 and 2004, respectively. Oil yields were significantly ($P < 0.001$) affected by the crop season, stage of maturity and their interaction. EO composition varied significantly ($P < 0.05$) with the stages of maturity. The compound linalool was the main compound in the season 2003 (79.86 ± 8.16) as well as in 2004 (80.04 ± 9.12). The strong effect of crop season, maturity stage and their interaction was found on 36 EO compounds. Several compounds (α-terpinene, α-terpineol, terpinene-4-ol, carvone and *p*-cymen-8-ol) showed a different response to crop season, stage of maturity and their interaction.

Keywords: essential oil composition, linalool, seasonal and maturational effects, Umbelliferae

INTRODUCTION

Coriander (*Coriandrum sativum* L.) is also known as *kuzbara* or *cilantro* in Arabic and Spanish, respectively, belongs to the family Apiaceae (synonymous with Umbelliferae). The essential oil (EO) of coriander is obtained by steam distillation of the dried fully ripe fruits (seeds). The EO has a characteristic odour of linalool compound. It has a mild, sweet, warm, aromatic flavour. In the food industry, coriander EO is used as a flavouring agent and adjuvant. The fruits contain an EO (up to 1.0%), with a monoterpenoid, linalool, being the main EO component. Coriander is a popular spice and finely ground fruit is a major ingredient of curry powder (Wangensteen *et al.* 2006).

Coriander fruits are mainly used as a drug for indigestion, worms in the stomach, rheumatism and pain in the joints (Wichtl 1994). Coriander fruits have a pleasant flavour owing to the particular composition of the EO. The fruits are used in the preparation of fish and meat, and also for baking. The extracted EO is used in the flavouring of a number of food products and in soap manufacturing. It is principally used as a flavouring agent in the liquor, cocoa and chocolate industries (Diederichsen 1996). It is also employed in medicine as a carminative or flavouring agent. It has the advantage of being more stable and of retaining its agreeable odour longer than any other EO (Diederichsen 1996).

Climatic conditions, geographical position of growing site, cultivation technology, as well as the growth stage of plants at the time of harvest and the extraction technique applied influence both qualitative composition and content of the individual components of the EOs (Bauer *et al*. 1992; Soliman *et al*. 1994; Voitkevich 1999).

Like all secondary metabolites, the EOs are known to have several important ecological functions such as protection against predators (microorganisms, fungi, insects, herbivores) and UV radiation (Khan *et al*. 1997). Apart from this, they also have secondary functions such as attracting natural enemies of herbivores (Turlings *et al*. 1990), attracting pollinators (de Moraes *et al*. 1998), or inhibiting germination and growth (Kessler *et al*. 2001).

Recent studies on the compositional analysis of *C. sativum* L. fruits have revealed changes in EO content during maturation (Msaada *et al*. 2006; Msaada 2007; Msaada *et al*. 2007a, 2009a), changes in EO composition of different plant parts (Msaada *et al*. 2003; Msaada 2007; Msaada *et al*. 2007b), changes in fatty acid composition during fruit maturation (Msaada 2007; Msaada *et al*. 2009b, 2010), effects of stage of maturity and growing region on fatty acid composition (Msaada *et al*. 2009c) and effects of growing region and stages of maturity on EO yields and composition (Msaada *et al*. 2009d) regarding this medicinal and aromatic plant.

Coriander EO composition (relative percentages of main compounds) cultivated in different regions of the world is presented in **Table 1**. There is significant variability in the EO composition of coriander fruit suggesting a significant regional effect. The main compound was linalool in all studied oils and its percentage varied significantly among the studied regions. For example, the percentage linalool was 41.4 and 82.9% in India (Gupta *et al.* 1977) and Argentina (Gil *et al.* 2002), respectively. In addition, camphor was 9.9% in coriander fruit EO cultivated in Italy (Pino *et al.* 1993) but absent in India (Gupta *et al.* 1977), China (Zhu 1993) and Pakistan (Karim *et al.* 1979) EO. However, variations in camphor concentrations within one country such as India (0-9.6%), Italy (3.3-9.9%) and USA (1-5.5%), were observed. This variation could be explained by the plant's sensibility to temperature, photoperiod, agronomic and ecologic factors during fruit maturation and

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Table 1 Yield of main EO composition (%, w/w) of coriander fruit from different geographic origins.

Origin	Linalool	α-Pinene	α-Terpinene	Geraniol	Geranyl acetate	Limonene	*p*-Cymene	Camphor	References
Tunisia	87.54	0.02	0.01	tr	0.83	0.02	tr	0.17	Msaada *et al*. 2007a
Albania	57.40	3.10	nd	4.60	2.60	3.60	4.10	4.80	Pino *et al*. 1993
China	81.92	8.72	nd	0.33	4.02	nd	0.67	nd	Zhu 1993
India	41.40	5.30	nd	0.70	1.10	1.90	nd	nd	Gupta *et al.* 1977
India	49.20	5.40	0.05	2.60	0.60	2.90	14.70	9.60	Pino *et al.* 1993
Italy	67.50	1.80	0.05	2.30	1.70	1.40	2.60	8.90	Chialva *et al.* 1982
Italy	72.00	2.80	nd	1.60	3.70	1.50	2.80	3.30	Bourrel *et al.* 1995
Italy	56.00	3.10	nd	3.40	2.40	3.10	13.00	9.90	Pino *et al.* 1993
Italy	64.50	5.10	tr	0.40	0.40	3.60	nd	6.40	Cantore *et al.* 2004
Pakistan	69.00	0.90	nd	6.30	5.60	3.10	1.60	nd	Karim *et al.* 1979
USA	63.80	3.30	0.10	1.80	1.00	2.40	4.90	5.50	Anitescu *et al.* 1997
USA	57.65	10.15	nd	1.95	0.95	3.35	7.00	1.00	Redshaw *et al.* 1971
Finland	66.00	0.01	0.02	1.30	3.80	3.10	2.80	6.70	Taskinen and Nykänen 1975
Finland	70.40	0.60	nd	2.80	7.20	3.80	0.60	4.70	Hirvi *et al.* 1986
Finland	71.90	0.60	nd	4.00	7.50	2.60	0.70	4.30	Hirvi *et al.* 1986
Japan	69.29	5.85	0.09	1.00	2.04	7.28	2.56	2.46	Chou 1974
Argentina	81.00	1.70	3.60	2.60	1.40	1.00	1.00	4.00	Bandoni *et al.* 1998
Argentina	82.90	2.10	nd	1.90	1.10	0.70	nd	3.00	Gil *et al.* 2002
Russia	69.00	5.20	tr	1.60	2.70	2.81	1.60	5.20	Buronfosse and Sellier 1997
Russia	58.80	4.90	nd	2.50	4.20	3.80	3.90	6.60	Pino *et al.* 1993

water availability and soil quality (Hornok 1986).

Unlike the reports published regarding the EO of the fruits of other species, the reports regarding the changes in the fruit EO of *C. sativum* as influenced by cultivation season, developmental stage and the interaction between the two are still very scarce. In the present study, we studied for the first time the effects of cultivation season, stage of maturity and that of interaction of cultivation season and stage of maturity on the EO composition isolated from the Tunisian coriander fruit. The results indicate the potential economic utility of *C. sativum* L. as a source of raw material for useful industrial and food-EO components.

MATERIALS AND METHODS

Plant culture

Coriander fruits were planted in the field on February 20 in two consecutive cultivation seasons (2003 and 2004) at Charfine (North-Eastern Tunisia; latitude 36° 44′ 29.12″ N; longitude 10° 40′ 51.26″ E, altitude 163 m). Charfine is characterized by annual rainfall of 700 mm and mean annual temperature of 16.8°C. The time required for complete maturity (harvest period) was extended from 5 days after flowering (DAF) to 31 DAF in 2003 and to 33 DAF in 2004. The colour and relative moisture content of the fruit were used as ripening criteria (**Table 2**). Only fully green fruits were harvested at the initial stages of maturity. Green-brown fruits were considered as indicators of the intermediate stages. Only brown fruits were selected for analysis during the final stages of maturity. At harvest, the fruits of the plants were collected by hand at different ripening stages during May and June in 2003 and 2004 crop seasons, respectively. Fruit analyses were undertaken after attaining a moisture content of 10% (**Table 1**). Moisture contents were determined using a hot air oven at 60°C for 2 weeks until constant weight.

EO isolation

Replicating three times, the fresh fruits (100 g) were subjected to hydrodistillation for 90 min in accordance with the method published by the European Pharmacopoeia (Council of Europe 1997). The distillate (100 mL) was extracted with 100 mL of 2-methylbutane (Analytical Reagent, LabScan Ltd., Dublin, Ireland) for 30 min (three times) and then the organic phase were separated and dried over anhydrous sodium sulphate (Sigma–Aldrich, Steinheim, Germany). The organic layer was then concentrated at 30°C using a Vigreux column (Labbox, Le Boulou, France) at atmospheric pressure and the resulting EO was subsequently analyzed.

Gas chromatography analysis (GC)

Analysis of volatile compounds of the EO by gas chromatography (GC) was carried out on a Hewlett-Packard 6890 gas chromatograph (Palo Alto, CA, USA) equipped with a flame ionization detector (FID) and an electronic pressure control (EPC) injector. A polar polyethylene glycol (PEG) HP Innowax and 5% diphenyl, 95% dimethylpolysiloxane a polar HP-5 capillary columns (30 m × 0.25 mm, 0.25 μm film thickness (Hewlett-Packard, CA, USA) were used in the GC. The flow of the carrier gas (N_2) was 1.6 mL/min. The split ratio was 60:1. The analysis was performed using the following temperature schedule: oven temperature kept isothermally at 35°C for 10 min and then the temperature was increased from 35 to 205°C at a rate of 3°C/min, and it was kept isothermally at 205°C for 10 min. Injector and detector temperatures were held at 250 and 300°C, respectively. The injected volume was 1 μL of neat (i.e. pure or crude) EO.

Gas chromatography-mass spectrometry (GC-MS) analysis

Analysis of the EO volatile compounds by GC-MS was performed using a gas chromatograph HP 5890 (II) interfaced with a HP 5972 mass spectrometer (Palo Alto, CA, USA) with electron impact ionization of 70 eV. A HP-5 MS capillary column (30 m × 0.25 mm, coated with 5% phenyl methyl silicone, 95% dimethylpolysiloxane, 0.25 μm film thickness) (Hewlett-Packard, CA, USA) was used in the analysis. The column temperature was programmed to rise from 50 to 240°C at a rate of 5°C/min. The carrier gas was helium with a flow rate of 1.2 mL/min and split ratio of 60: 1. Scan time and mass range were 1 s and 40-300 m/z, respectively.

Identification of volatile EO compounds

The tentative identification of the EO constituents was based on comparing their retention indices relative to (C_8-C_{22}) *n*-alkanes (Analytical Reagent: LabScan Ltd.) with those reported in the literature or with those of authentic compounds available in our laboratory. Further identification was made by matching their recorded mass spectra with those stored in the Wiley/NBS mass spectral library of the GC-MS data system and other published mass spectra (Adams 2001; Msaada *et al*. 2007a). Quantitative data were obtained from the electronic integration of the FID peak areas.

Statistical analysis

Data were expressed as mean ± S.D. The means of three determinations (replicates) were compared by using one-way analysis of

Table 2 Harvest date (days after flowering-DAF), fruit colour, stage of maturity, relative moisture content, EO yields and daily temperature of each harvest of coriander fruit cultivated at Charfine.

Crop season	Harvest date	DAF	Fruit colour, stage of maturity	Relative moisture content (%, w/w) ± SD	EO yield (%, w/w) ± SD	Mean temperature (°C) ± S.D
2003						
1	09 May 2003	5	Unripe, fully green	94.12(10.32)^{a}	0.014(0.00)^{h}	26.15(6.23)^{h}
2	13 May 2003	9	Unripe, fully green	84.12(9.25)^{b}	0.017(0.00)^{g}	26.46(6.45)^{g}
3	16 May 2003	12	Unripe, fully green	74.12(8.56)^{c}	0.021(0.00)^{f}	26.89(6.78)^{f}
4	20 May 2003	16	Unripe, green-brown	64.12(7.26)^{d}	0.023(0.00)^{e}	27.31(7.32)^{e}
5	23 May 2003	19	Half ripe, green-brown	54.12(6.59)^{e}	0.072(0.01)^{d}	27.84(7.59)^{d}
6	25 May 2003	21	Half ripe, green-brown	44.12(5.98)^{f}	0.076(0.01)^{c}	28.12(7.61)^{c}
7	29 May 2003	25	Half ripe, green-brown	34.12(4.68)^{g}	0.094(0.02)^{b}	30.26(7.80)^{b}
8	04 June 2003	31	Fully ripe, brown	24.12(2.98)^{h}	0.17(0.05)^{a}	32.30(8.21)^{a}
2004						
1	14 May 2004	5	Unripe, fully green	91.58(9.21)^{a}	0.085(0.01)^{h}	25.55(5.23)^{h}
2	18 May 2004	9	Unripe, fully green	81.58(7.78)^{b}	0.107(0.04)^{g}	25.88(5.62)^{g}
3	21 May 2004	12	Unripe, fully green	71.58(7.13)^{c}	0.154(0.06)^{f}	26.15(6.02)^{f}
4	24 May 2004	15	Unripe, green-brown	61.58(6.82)^{d}	0.189(0.08)^{e}	26.75(6.23)^{e}
5	28 May 2004	19	Half ripe, green-brown	51.58(5.81)^{e}	0.204(0.09)^{b}	27.01(6.75)^{d}
6	01 June 2004	23	Half ripe, green-brown	41.58(4.49)^{f}	0.260(0.09)^{c}	27.84(6.97)^{c}
7	07 June 2004	29	Half ripe, green-brown	31.58(3.36)^{g}	0.299(0.09)^{b}	28.43(7.16)^{b}
8	11 June 2004	33	Fully ripe, brown	21.58(3.01)^{h}	0.327(0.10)^{a}	29.33(8.33)^{a}

Values in the same row with different superscript (a-h) are significantly different at $P < 0.05$. Values in brackets are the respective standard deviations.

variance (ANOVA) followed by Duncan's multiple range test. The differences between individual means were deemed to be significant at $P < 0.05$. Correlation coefficients were calculated based on EO composition during fruit maturation in 2003 and 2004 seasons. All analyses were performed by the Statistica v 5.1 software (Statsoft 1998).

RESULTS AND DISCUSSION

EO yield

During the 2003 season, EO yield increased significantly ($P < 0.05$) from the first stage of maturity (0.014 ± 0.00%) to the last one (0.171 ± 0.05%) based on dry weight. At full maturity, the obtained yield was low in comparison with previous reports (Anitescu *et al*. 1997; Carrubba *et al*. 2002; Ravi *et al*. 2006; Telci *et al*. 2006; Msaada *et al*. 2007a; Msaada *et al*. 2009a; **Table 1**). EO yield in the 2004 season increased considerably from 0.085 ± 0.01 to 0.327 ± 0.10% at the first and final stages of maturity, respectively. The highest EO yield was recorded in the 2004 season at full fruit maturity (0.327 ± 0.10%). This yield was higher than the one investigated at Borj El Ifaâ region (0.30% w/w) (Msaada *et al*. 2009a) and lower than previously investigated samples (Anitescu *et al*. 1997; Carrubba *et al*. 2002; Ravi *et al*. 2006; Telci *et al*. 2006; Msaada *et al*. 2007a). Variation in the EO yield can be attributed to factors like cultivation conditions and, especially, the extent of use of fertilizers and irrigation (Ravi *et al*. 2006). EO yield during plant growth is particularly susceptible to environmental conditions such as light, nutrient availability, and day length (Circella *et al*. 1995; Skoula *et al*. 2000). Our data confirm the strong ($P < 0.001$) effects of season, stage of maturity and season × stage of maturity interaction on EO yield (**Table 3**). These results could be due to the inherent genetic variability in EO yield of coriander fruits by environmental factors, especially the stage of maturity and season of cultivation.

EO composition

The coriander fruits matured in 31 and 33 DAF during 2003 and 2004 seasons, respectively (**Tables 2, 4, 5**). **Tables 4** and **5** list the linear retention indices, percentage composition, and grouped compound of EOs of the coriander fruit collected in 2003 and 2004 seasons, respectively. EO compounds identified in *C. sativum* fruits are listed in **Tables 4** and **5** in order of their elution on the HP-5 column. In total, 41 compounds were identified.

1. 2003 season

Tables 3 and **4** show the EO composition as affected by seasonal changes. The chemical composition of EOs of coriander fruits at 8 stages of maturity are listed in **Table 4**. The EO analyses in triplicate revealed significant ($P < 0.05$) changes in chemical composition of EO. All 41 compounds were identified at all stages of maturity. The analyses of volatiles exhibited a clear difference, both in quality and in quantity, of major components, at each stage of maturity (**Table 4**). The first stage of maturity is represented mainly by monoterpene alcohols (39.70 ± 4.31%) dominated by linalool (26.99 ± 3.01%). Monoterpene esters (21.52 ± 4.65%) were the second main class of EO components containing geranyl acetate as the main compound (20.50 ± 3.12%), followed by sesquiterpenes (18.13 ± 2.12%), monoterpene hydrocarbons (8.75 ± 0.91%), monoterpene ketones (5.85 ± 0.43%), monoterpene ethers (2.30 ± 0.24%) and phenols (1.31 ± 0.14%). Our earlier study (Msaada *et al*. 2007a) demonstrated that unripe fruits of *C. sativum* cv. 'Menzel Temime' (high essential oil yield: 0.35%, w/w), collected from Menzel Temime (Northeastern of Tunisia) in 2005 had an EO composition as represented mainly by monoterpene esters (46.27%), monoterpene alcohols (14.66%) and monoterpene aldehydes (2.07%). These differences could be explained by a regional [(Tokat, located in the middle Black Sea region (36° 43' E; 40° 19' N, 650 m above sea level and Diyarbakir, located in the Southern Anatolian region (40° 14' E; 37°55' N; 660 m above sea level) (Telci *et al*. 2006) and eight regions of India (Ravi *et al*. 2006)) and/or seasonal [(samples of *Origanum vulgare* ssp. *Hirtum* collected from the same geographic area in the south of Croatia at different seasons of growth) (Jerković *et al*. 2001)] effect on EO composition. At the intermediate stages of maturity, EO compounds were affected differently by the stage of maturity, but linalool was the main constituent followed by geranyl acetate at the 9th, 12th, 16th, 19th, 21st and 25th DAF. Other compounds were detected at lower percentages. Linalool (79.86 ± 8.16%), α-humulene (3.30 ± 0.41%), geranyl acetate (3.06 ± 0.42%), α-terpineol (2.03 ± 0.22%) and geraniol (1.10 ± 0.12%) were the dominating compounds at full fruit maturity; the remaining compounds were present at levels less than 1%.

2. 2004 season

EO composition of coriander fruits at 8 stages of maturity are given in **Table 5**. The first stage of maturity (5 DAF) was marked by the prevalence of monoterpene alcohols (58.06 ± 7.85%) represented mainly by linalool (48.56 ±

Table 3 Effects of maturity stage [MS], season [S] and [MS] x [S] interaction on the yield and composition of EO of coriander fruit.

Variables	Factors	d.f	*F-value*	*P-value*	Sig.	Variables	Factors	d.f	*F-value*	*P-value*	Sig.
Heptanal	[MS]	11	112.1170	0.0000	***	*cis*–Hex-3-enyl	[MS]	11	107.6327	0.0000	***
	[S]	1	275.6250	0.0000	***	butyrate	[S]	1	12.4171	0.0013	***
	[MS] x [S]	3	92.2917	0.0000	***		[MS] x [S]	3	14.4551	0.0000	***
α-Thujene	[MS]	11	98.3167	0.0000	***	α-Terpineol	[MS]	11	1.554967	0.160571	NS
	[S]	1	25.7561	0.0000	***		[S]	1	1.238209	0.274106	NS
	[MS] x [S]	3	281.0111	0.0000	***		[MS] x [S]	3	1.660537	0.195092	NS
α-Pinene	[MS]	11	8.30417	0.0000	***	*cis*-	[MS]	11	96.79588	0.0000	***
	[S]	1	13.89291	0.0000	***	Dihydrocarvone	[S]	1	0.53850	0.468397	NS
	[MS] x [S]	3	14.88857	0.0000	***		[MS] x [S]	3	7.59108	0.00057	***
Sabinene	[MS]	11	1195.947	0.0000	***	Nerol	[MS]	11	401.942	0.0000	***
	[S]	1	1942.857	0.0000	***		[S]	1	18.149	0.000168	***
	[MS] x [S]	3	1251.137	0.0000	***		[MS] x [S]	3	1164.573	0.0000	***
β-Pinene	[MS]	11	316.6426	0.0000	***	β-Citronellol	[MS]	11	226.484	0.0000	***
	[S]	1	65.7094	0.0000	***		[S]	1	22485.413	0.0000	***
	[MS] x [S]	3	157.5613	0.0000	***		[MS] x [S]	3	605.265	0.0000	***
Δ^3-Carene	[MS]	11	417.3990	0.0000	***	Neral	[MS]	11	118.2884	0.0000	***
	[S]	1	23.1481	0.0000	***		[S]	1	65.0656	0.0000	***
	[MS] x [S]	3	46.2593	0.0000	***		[MS] x [S]	3	124.4754	0.0000	***
α-Terpinene	[MS]	11	62.22325	0.0000	***	Carvone	[MS]	11	82.15383	0.0000	***
	[S]	1	1.65746	0.207181	NS		[S]	1	0.3256	0.09563	NS
	[MS] x [S]	3	99.55801	0.0000	***		[MS] x [S]	3	62.03077	0.0000	***
p-Cymene	[MS]	11	111.8103	0.0000	***	Geraniol	[MS]	11	5519.86	0.0000	***
	[S]	1	189.3913	0.0000	***		[S]	1	55420.52	0.0000	***
	[MS] x [S]	3	413.8551	0.0000	***		[MS] x [S]	3	13993.79	0.0000	***
Limonene	[MS]	11	67.4606	0.0000	***	Geranial	[MS]	11	52.93182	0.0000	***
	[S]	1	26.8082	0.0000	***		[S]	1	61.06522	0.0000	***
	[MS] x [S]	3	197.0446	0.0000	***		[MS] x [S]	3	81.76087	0.0000	***
1,8-Cineole	[MS]	11	500.5721	0.0000	***	Anethole	[MS]	11	828.767	0.0000	***
	[S]	1	8.6835	0.0059	**		[S]	1	2701.125	0.0000	***
	[MS] x [S]	3	812.3291	0.0000	***		[MS] x [S]	3	2651.458	0.0000	***
(Z)-β-Ocimene	[MS]	11	237.804	0.0000	***	Thymol	[MS]	11	102.5745	0.0000	***
	[S]	1	1823.214	0.0000	***		[S]	1	192.2025	0.0000	***
	[MS] x [S]	3	673.164	0.0000	***		[MS] x [S]	3	93.7648	0.0000	***
γ-Terpinene	[MS]	11	35.96779	0.0000	***	Carvacrol	[MS]	11	88.59545	0.0000	***
	[S]	1	37.81102	0.0000	***		[S]	1	20.48000	0.0000	***
	[MS] x [S]	3	14.83990	0.0000	***		[MS] x [S]	3	70.13333	0.0000	***
cis-Linalool	[MS]	11	347.4886	0.0000	***	δ-Elemene	[MS]	11	1457.67	0.0000	***
oxide	[S]	1	240.2500	0.0000	***		[S]	1	11157.88	0.0000	***
	[MS] x [S]	3	792.4167	0.0000	***		[MS] x [S]	3	1570.88	0.0000	***
Terpinolene	[MS]	11	119.4650	0.0000	***	Eugenol	[MS]	11	94.5033	0.0000	***
	[S]	1	464.1421	0.0000	***		[S]	1	39.6826	0.0000	***
	[MS] x [S]	3	132.9425	0.0000	***		[MS] x [S]	3	101.8623	0.0000	***
trans-Linalool	[MS]	11	243.2831	0.0000	***	Neryle acetate	[MS]	11	293.7805	0.0000	***
oxide	[S]	1	182.3544	0.0000	***		[S]	1	316.1355	0.0000	***
	[MS] x [S]	3	323.5190	0.0000	***		[MS] x [S]	3	436.4861	0.0000	***
Linalool	[MS]	11	105527.3	0.0000	***	Geranyle acetate	[MS]	11	1216.080	0.0000	***
	[S]	1	164586.2	0.0000	***		[S]	1	2871.396	0.0000	***
	[MS] x [S]	3	19117.3	0.0000	***		[MS] x [S]	3	223.446	0.0000	***
Camphor	[MS]	11	389.6146	0.0000	***	β-Caryophyllene	[MS]	11	23442.39	0.0000	***
	[S]	1	205.0577	0.0000	***		[S]	1	33121.71	0.0000	***
	[MS] x [S]	3	199.2553	0.0000	***		[MS] x [S]	3	47919.05	0.0000	***
Borneol	[MS]	11	25.8051	0.0000	***	α-Humulene	[MS]	11	472.9114	0.0000	***
	[S]	1	88.4754	0.0000	***		[S]	1	698.6806	0.0000	***
	[MS] x [S]	3	121.9996	0.0000	***		[MS] x [S]	3	364.2505	0.0000	***
Menthol	[MS]	11	56.3369	0.0000	***	Germacrene-D	[MS]	11	689.3439	0.0000	***
	[S]	1	123.3128	0.0000	***		[S]	1	83.5652	0.0000	***
	[MS] x [S]	3	111.0421	0.0000	***		[MS] x [S]	3	377.8261	0.0000	***
Terpinene-4-ol	[MS]	11	38.8986	0.0000	***	Eugenyle acetate	[MS]	11	398.373	0.0000	***
	[S]	1	3.0000	0.0928	NS		[S]	1	1353.805	0.0000	***
	[MS] x [S]	3	108.8765	0.0000	***		[MS] x [S]	3	704.297	0.0000	***
p-Cymen-8-ol	[MS]	11	9.282229	0.0000	***	Oil yields	[MS]	11	236.671	0.0000	***
	[S]	1	1.102909	0.301495	NS		[S]	1	385.762	0.0000	***
	[MS] x [S]	3	1.134618	0.349850	NS		[MS] x [S]	3	429.382	0.0000	***

***: $P < 0.001$, NS: Not significant; Sig. = significance

5.68%), followed by monoterpene esters (15.44 ± 3.26%) represented by geranyl acetate (14.24 ± 0.19%), monoterpene hydrocarbons (8.04 ± 1.02%) and sesquiterpenes (4.52 ± 0.05%). Other classes were detected in lower amounts. During maturity process, there were significant changes in volatile compounds, as shown in **Table 5**. In mature fruits, linalool (80.04 ± 9.12%), geranyl acetate (4.85 ± 0.56%), *p*-cymen-8-ol (1.82 ± 0.21%) and terpinolene (1.02 ± 0.12%) were the main compounds. Msaada *et al*. (2007a) reported that at full maturity, EOs of fruits collected from Menzel Temime in 2005 were predominated by monoterpene alcohols (88.51%), ketones (2.61%), phenols (2.31%), with the main compounds being linalool (87.54%) and *cis*-dihydrocarvone (2.36%). Linalool constitutes more than two-thirds

Table 4 Chemical composition (%, w/w) of the EO obtained from fruits of coriander in 2003.

Compound*	RI[a]	RI[b]	Days after flowering (DAF)								Identification
			5	9	12	16	19	21	25	31	
Heptanal	901	1194	0.24(0.03)^{cd}	0.30(0.02)^{bc}	0.33(0.03)^{ab}	0.19(0.02)^{de}	0.23(0.01)^{cd}	0.38(0.02)^{a}	0.15(0.01)^{c}	0.07(0.00)^{f}	GC-MS
α-Thujene	931	1035	0.47(0.05)^{c}	0.67(0.04)^{b}	0.76(0.07)^{ab}	0.70(0.08)^{ab}	0.50(0.05)^{c}	0.75(0.06)^{ab}	0.79(0.09)^{a}	0.80(0.07)^{a}	GC-MS
α-Pinene	939	1032	0.21(0.02)^{cde}	0.29(0.03)^{bcd}	0.22(0.02)^{cde}	0.33(0.03)^{b}	0.58(0.04)^{a}	0.30(0.02)^{bc}	0.20(0.02)^{de}	0.13(0.01)^{e}	GC-MS, Co-GC
Sabinene	976	1132	2.93(0.25)^{a}	2.04(0.13)^{c}	0.19(0.02)^{f}	2.67(0.30)^{b}	0.37(0.04)^{e}	0.40(0.04)^{e}	1.90(0.21)^{d}	0.27(0.02)^{f}	GC-MS
β-Pinene	980	1118	0.28(0.02)^{c}	0.14(0.01)^{cde}	0.70(0.04)^{b}	1.80(0.13)^{a}	0.75(0.06)^{b}	0.23(0.03)^{cd}	0.12(0.01)^{de}	0.05(0.00)^{e}	GC-MS, Co-GC
Δ^3–Carene	1011	1159	0.05(0.00)^{f}	0.03(0.00)^{f}	0.18(0.01)^{d}	0.14(0.01)^{e}	0.72(0.06)^{c}	0.88(0.08)^{b}	0.94(0.08)^{a}	0.04(0.01)^{f}	GC-MS
α-Terpinene	1018	1188	0.86(0.06)^{a}	0.40(0.03)^{b}	0.20(0.01)^{d}	0.27(0.02)^{c}	0.29(0.02)^{c}	0.34(0.03)^{bc}	0.08(0.01)^{d}	0.27(0.03)^{c}	GC-MS, Co-GC
p-Cymene	1026	1280	0.03(0.00)^{f}	0.09(0.01)^{e}	0.19(0.02)^{d}	0.53(0.02)^{b}	0.65(0.07)^{a}	0.49(0.05)^{b}	0.51(0.05)^{b}	0.24(0.03)^{c}	GC-MS, Co-GC
Limonene	1030	1203	3.21(0.35)^{a}	1.25(0.13)^{b}	0.06(0.00)^{h}	1.04(0.09)^{d}	0.78(0.08)^{f}	0.83(0.09)^{e}	1.16(0.26)^{c}	0.19(0.01)^{g}	GC-MS, Co-GC
1,8-Cineole	1033	1213	1.84(0.21)^{a}	0.31(0.03)^{b}	0.10(0.01)^{d}	0.23(0.02)^{c}	0.20(0.02)^{c}	0.22(0.02)^{c}	0.21(0.03)^{c}	0.24(0.02)^{c}	GC-MS, Co-GC
(Z)-β-Ocimene	1040	1246	0.45(0.05)^{d}	0.36(0.04)^{c}	0.13(0.01)^{g}	0.79(0.07)^{a}	0.65(0.07)^{b}	0.59(0.06)^{c}	0.34(0.02)^{e}	0.29(0.02)^{f}	GC-MS, Co-GC
γ-Terpinene	1062	1266	0.19(0.02)^{de}	0.69(0.07)^{a}	0.15(0.02)^{e}	0.28(0.03)^{cd}	0.37(0.04)^{c}	0.47(0.04)^{b}	0.20(0.02)^{de}	0.30(0.02)^{c}	GC-MS, Co-GC
cis-Linalool oxide	1074	1478	0.41(0.04)^{c}	0.95(0.10)^{a}	0.17(0.01)^{f}	0.25(0.02)^{d}	0.21(0.02)^{e}	0.59(0.06)^{b}	nd	0.21(0.02)^{e}	GC-MS
Terpinolene	1088	1290	0.10(0.01)^{g}	0.35(0.04)^{e}	0.02(0.00)^{h}	0.63(0.06)^{d}	0.75(0.07)^{c}	0.87(0.07)^{b}	2.19(0.25)^{a}	0.15(0.01)^{f}	GC-MS, Co-GC
trans-Linalool oxide	1088	1450	0.05(0.00)^{f}	0.43(0.04)^{d}	0.04(0.00)^{f}	0.54(0.04)^{b}	0.58(0.06)^{a}	0.47(0.03)^{c}	0.42(0.05)^{d}	0.28(0.02)^{e}	GC-MS
Linalool	1098	1553	26.99(3.01)^{h}	48.33(4.23)^{g}	51.88(6.52)^{f}	54.52(6.23)^{e}	56.14(6.22)^{d}	58.43(6.85)^{c}	60.30(8.45)^{b}	79.86(8.16)^{a}	GC-MS, Co-GC
Camphor	1143	1532	5.49(6.23)^{a}	0.67(0.07)^{bc}	0.05(0.00)^{d}	0.59(0.06)^{c}	0.86(0.07)^{bc}	0.92(0.08)^{bc}	1.00(0.12)^{b}	0.26(0.03)^{d}	GC-MS
Borneol	1165	1719	1.34(0.15)^{f}	1.57(0.15)^{e}	5.34(0.42)^{a}	2.75(0.30)^{b}	2.61(0.21)^{c}	0.76(0.05)^{g}	2.13(0.12)^{d}	0.49(0.05)^{h}	GC-MS
Menthol	1173	1628	2.21(0.21)^{a}	0.67(0.05)^{e}	1.09(0.15)^{c}	1.21(0.13)^{b}	1.07(0.12)^{c}	1.11(0.13)^{c}	0.90(0.07)^{d}	0.15(0.01)^{f}	GC-MS
Terpinene-4-ol	1178	1611	0.03(0.00)^{d}	0.45(0.05)^{c}	0.02(0.00)^{d}	1.02(0.12)^{a}	0.89(0.09)^{b}	0.47(0.05)^{c}	0.40(0.04)^{c}	0.48(0.06)^{c}	GC-MS, Co-GC
p-Cymen-8-ol	1183	1864	1.66(0.15)^{b}	1.35(0.10)^{c}	1.00(0.11)^{d}	1.24(0.13)^{cd}	2.31(0.25)^{a}	1.03(0.17)^{cd}	1.07(0.16)^{cd}	0.21(0.23)^{e}	GC-MS, Co-GC
cis–Hex-3-enyl butyrate	1188	1485	0.16(0.05)^{d}	0.64(0.06)^{bc}	0.68(0.08)^{ab}	0.33(0.02)^{cd}	0.86(0.07)^{ab}	1.00(0.15)^{a}	0.33(0.03)^{d}	0.03(0.00)^{d}	GC-MS
α-Terpineol	1189	1706	0.25(0.02)^{g}	1.26(0.15)^{f}	2.56(0.27)^{b}	2.11(0.23)^{c}	1.43(0.11)^{e}	1.46(0.13)^{c}	2.84(0.31)^{a}	2.03(0.22)^{d}	GC-MS, Co-GC
cis-Dihydrocarvone	1193	1645	0.36(0.03)^{d}	0.12(0.01)^{e}	0.11(0.01)^{e}	0.20(0.01)^{e}	0.55(0.06)^{c}	1.40(0.12)^{b}	2.08(0.19)^{a}	0.13(0.01)^{e}	GC-MS
Nerol	1228	1797	0.15(0.01)^{e}	0.13(0.01)^{e}	0.63(0.05)^{c}	1.47(0.13)^{b}	1.55(0.16)^{a}	0.51(0.04)^{d}	0.50(0.04)^{d}	0.13(0.01)^{e}	GC-MS
β-Citronellol	1228	1772	0.87(0.07)^{d}	0.22(0.02)^{g}	0.72(0.06)^{e}	1.03(0.11)^{c}	0.15(0.02)^{g}	0.53(0.03)^{f}	1.66(0.14)^{a}	1.20(0.10)^{b}	GC-MS
Neral	1240	1694	0.71(0.06)^{c}	0.27(0.02)^{ef}	0.35(0.04)^{e}	0.86(0.06)^{b}	1.36(0.14)^{a}	0.50(0.06)^{d}	0.40(0.05)^{de}	0.19(0.02)^{f}	GC-MS
Carvone	1242	1751	tr	0.23(0.02)^{b}	0.29(0.03)^{a}	0.29(0.04)^{a}	0.26(0.02)^{ab}	0.10(0.01)^{c}	0.23(0.02)^{b}	0.02(0.00)^{d}	GC-MS, Co-GC
Geraniol	1255	1857	5.18(0.04)^{b}	11.26(1.23)^{a}	2.92(0.21)^{c}	2.77(0.25)^{d}	0.83(0.07)^{g}	0.79(0.06)^{g}	1.21(0.11)^{e}	1.10(0.12)^{f}	GC-MS, Co-GC
Geranial	1270	1742	nd	nd	0.24(0.02)^{e}	0.31(0.02)^{c}	0.33(0.03)^{bc}	0.28(0.03)^{d}	0.48(0.04)^{a}	0.35(0.04)^{b}	GC-MS
Anethole	1283	1828	0.47(0.05)^{c}	0.52(0.05)^{b}	0.28(0.02)^{d}	0.49(0.03)^{c}	3.04(0.37)^{a}	0.10(0.01)^{f}	nd	0.15(0.01)^{e}	GC-MS
Thymol	1290	2198	0.91(0.10)^{d}	1.17(0.15)^{b}	1.30(0.14)^{a}	1.01(0.11)^{c}	0.12(0.01)^{g}	0.43(0.04)^{f}	0.53(0.06)^{e}	0.12(0.01)^{g}	GC-MS, Co-GC
Carvacrol	1292	2239	0.40(0.03)^{bc}	0.67(0.04)^{a}	0.13(0.01)^{d}	0.62(0.05)^{a}	0.32(0.02)^{bc}	0.46(0.03)^{b}	0.30(0.02)^{c}	0.43(0.03)^{bc}	GC-MS
δ-Elemene	1339	1479	3.83(0.39)^{b}	3.40(0.36)^{c}	2.06(0.22)^{d}	1.85(0.16)^{e}	6.18(0.42)^{a}	1.66(0.14)^{f}	1.91(0.16)^{e}	0.05(0.00)^{g}	GC-MS
Eugenol	1356	2192	0.55(0.06)^{c}	0.53(0.06)^{c}	0.30(0.02)^{e}	0.77(0.06)^{a}	0.21(0.02)^{f}	0.65(0.05)^{b}	0.41(0.03)^{d}	0.64(0.05)^{b}	GC-MS, Co-GC
Neryle acetate	1356	1733	0.14(0.01)^{cd}	0.29(0.03)^{b}	0.20(0.01)^{bc}	0.54(0.06)^{a}	0.11(0.01)^{cd}	0.50(0.04)^{a}	0.57(0.06)^{a}	0.07(0.01)^{d}	GC-MS
Geranyle acetate	1383	1765	20.50(3.12)^{a}	12.00(2.03)^{d}	15.68(2.22)^{c}	3.44(0.42)^{g}	8.71(1.10)^{c}	17.01(1.64)^{b}	6.22(0.75)^{f}	3.06(0.42)^{g}	GC-MS, Co-GC
β-Caryophyllene	1418	1612	11.35(2.32)^{a}	2.66(0.32)^{d}	2.46(0.28)^{e}	3.31(0.25)^{b}	0.19(0.02)^{g}	0.08(0.01)^{h}	3.24(0.26)^{c}	0.25(0.02)^{f}	GC-MS
α-Humulene	1454	1687	2.41(0.27)^{b}	0.10(0.01)^{f}	1.48(0.16)^{c}	0.51(0.06)^{d}	0.18(0.02)^{ef}	0.34(0.03)^{de}	0.13(0.01)^{f}	3.30(0.41)^{a}	GC-MS
Germacrene-D	1480	1726	0.54(0.06)^{b}	0.11(0.01)^{de}	0.14(0.01)^{d}	0.24(0.03)^{c}	0.13(0.01)^{de}	0.21(0.01)^{c}	0.81(0.06)^{a}	0.09(0.01)^{e}	GC-MS
Eugenyle acetate	1524	nd	0.88(0.09)^{f}	tr	2.22(0.30)^{a}	1.36(0.20)^{c}	1.63(0.17)^{b}	1.21(0.14)^{d}	0.95(0.08)^{e}	tr	GC-MS
Grouped compounds											
Monoterpene hydrocarbons			8.75(0.91)^{a}	6.22(0.72)^{d}	2.61(0.31)^{g}	8.65(0.94)^{b}	5.76(0.60)^{c}	5.66(0.62)^{f}	7.92(0.66)^{c}	2.49(0.31)^{h}	
Aromatic hydrocarbons			0.03(0.00)^{g}	0.09(0.01)^{f}	0.19(0.02)^{e}	0.53(0.04)^{b}	0.65(0.05)^{a}	0.49(0.05)^{c}	0.51(0.05)^{bc}	0.24(0.02)^{d}	
Monoterpene alcohols			39.70(4.31)^{g}	66.29(7.45)^{f}	66.74(7.21)^{e}	69.38(6.91)^{d}	70.23(8.45)^{c}	65.84(7.64)^{g}	71.42(6.49)^{b}	86.44(9.73)^{a}	
Phenols			1.31(0.14)^{d}	1.84(0.17)^{a}	1.43(0.12)^{c}	1.63(0.14)^{b}	0.44(0.03)^{h}	0.89(0.06)^{e}	0.83(0.06)^{f}	0.55(0.05)^{g}	
Monoterpene esters			21.52(4.65)^{a}	12.29(2.13)^{d}	18.10(2.45)^{c}	5.34(0.84)^{g}	10.45(1.87)^{e}	18.72(2.13)^{b}	7.74(0.92)^{f}	3.13(0.42)^{h}	
Monoterpene ketones			5.85(0.43)^{a}	1.02(0.12)^{f}	0.45(0.05)^{g}	1.08(0.19)^{e}	1.67(0.17)^{d}	2.42(0.31)^{c}	3.31(0.46)^{b}	0.41(0.03)^{h}	
Monoterpene aldehydes			0.71(0.08)^{c}	0.27(0.02)^{h}	0.59(0.07)^{f}	1.17(0.14)^{b}	1.69(0.18)^{a}	0.78(0.09)^{d}	0.88(0.09)^{c}	0.54(0.06)^{g}	
Monoterpene ethers			2.30(0.24)^{a}	1.69(0.18)^{b}	0.31(0.02)^{g}	1.02(0.12)^{d}	0.99(0.11)^{d}	1.28(0.13)^{c}	0.63(0.07)^{f}	0.73(0.07)^{e}	
Sesquiterpenes			18.13(2.12)^{a}	6.27(0.71)^{c}	6.14(0.73)^{d}	5.91(0.62)^{f}	6.68(0.75)^{b}	2.29(0.37)^{h}	6.09(0.74)^{e}	3.69(0.41)^{g}	
Non terpenics			0.40(0.03)^{g}	0.94(0.08)^{d}	1.01(0.11)^{c}	0.52(0.06)^{e}	1.09(0.11)^{b}	1.38(0.12)^{a}	0.48(0.03)^{f}	0.10(0.01)^{h}	
Total identified (%)			98.7(10.23)^{d}	96.92(9.56)^{g}	97.57(9.63)^{f}	95.23(9.53)^{h}	99.65(9.23)^{c}	99.75(9.86)^{b}	99.81(10.58)^{a}	98.32(11.85)^{e}	

tr: trace (<0.01%). nd: not detected. [a] Apolar HP-5 MS column. [b] Polar HP Innowax column. Volatile compounds percentages in the same line with different superscript (a–h) are significantly different at $P < 0.05$. Values in brackets are standard deviations.

of the volatile components of coriander fruit EO and is regarded as one of the flavour-impact compounds of the fruit EO (Mookherjee *et al*. 1989; Rogers *et al*. 1994; Cadwallader *et al*. 1999). The content of linalool increased regularly at all eight stages of maturity and its maximum content was obtained at the last stage of maturity in both crop seasons (**Tables 4, 5**). Linalool, an oxygenated monoterpene, was the main EO component in ripened fruits. It is well known that the linalool content in the EO of ripened coriander fruits is higher than that of premature fruits (Lawrence 1993). It has also been reported that climatic factors such as cloudy days and lower temperature and high amount of rainfall during maturation period may have adverse effect on the accumulation of linalool (Sangwan *et al*. 2001). In the two crop seasons, the level of linalool increased significantly during fruit maturation, this increase being concomitant with daily temperature, as indicated in **Table 2**. This could be due to the effect of temperature on linalool synthase enzyme involved in linalool biosynthesis and explains the gradual increase of monoterpene alcohols in both seasons (Pichersky *et al*. 1995; Dudareva *et al.* 1996; Martin *et al.* 2003). During the first stage of maturity, when green colour prevailed (**Table 2**), ester concentration was low. However, its content increased during maturation. The esters were probably formed due to reactions of alcohols and organic acids catalysed by alcohol acyl transferases. These reactions are influenced by temperature increase (**Table 2**). The alcohol acyl transferase is highly active during fruit maturation and appears to have an important role in ester biosynthesis (Olias *et al*. 1995). The changes, detected in

Table 5 Chemical composition (%, w/w) of the EO obtained from fruits of coriander in 2004.

Compound*	RI[a]	RI[b]	Days after flowering (DAF)								Identification
			5	9	12	15	19	23	29	33	
Heptanal	901	1194	0.22(0.02)^{d}	0.84(0.06)^{a}	0.84(0.07)^{a}	0.73(0.06)^{b}	0.25(0.02)^{d}	0.12(0.01)^{c}	0.31(0.02)^{c}	0.23(0.02)^{d}	GC-MS
α-Thujene	931	1035	1.42(0.12)^{a}	0.17(0.14)^{de}	0.11(0.01)^{e}	0.50(0.04)^{b}	0.26(0.03)^{c}	0.24(0.02)^{cd}	0.17(0.02)^{de}	0.14(0.01)^{e}	GC-MS
α-Pinene	939	1032	0.33(0.03)^{b}	0.27(0.03)^{bc}	0.09(0.01)^{de}	0.11(0.01)^{d}	0.21(0.01)^{c}	0.28(0.02)^{bc}	0.40(0.03)^{a}	0.03(0.00)^{e}	GC-MS, Co-GC
Sabinene	976	1132	0.58(0.06)^{b}	0.49(0.05)^{c}	0.90(0.08)^{a}	0.18(0.02)^{d}	0.16(0.01)^{d}	0.12(0.01)^{d}	0.18(0.01)^{d}	0.11(0.01)^{d}	GC-MS
β-Pinene	980	1118	0.70(0.06)^{a}	0.19(0.02)^{bc}	0.24(0.02)^{bc}	0.29(0.03)^{b}	0.12(0.01)^{cd}	0.12(0.01)^{cd}	0.29(0.03)^{b}	0.05(0.00)^{d}	GC-MS, Co-GC
Δ^3-Carene	1011	1159	0.12(0.01)^{de}	0.27(0.02)^{c}	tr	0.40(0.03)^{b}	0.84(0.06)^{a}	0.26(0.02)^{c}	0.21(0.02)^{cd}	0.17(0.02)^{cd}	GC-MS
α-Terpinene	1018	1188	0.50(0.04)^{b}	0.21(0.01)^{c}	0.73(0.08)^{a}	0.21(0.02)^{c}	0.21(0.02)^{c}	0.24(0.02)^{c}	0.07(0.01)^{d}	0.07(0.00)^{d}	GC-MS, Co-GC
p-Cymene	1026	1280	0.88(0.07)^{a}	0.24(0.02)^{f}	0.12(0.01)^{g}	0.31(0.04)^{de}	0.38(0.04)^{c}	0.28(0.02)^{ef}	0.35(0.03)^{cd}	0.55(0.04)^{b}	GC-MS, Co-GC
Limonene	1030	1203	1.26(0.14)^{c}	1.34(0.15)^{b}	1.96(0.18)^{a}	0.64(0.07)^{e}	1.98(0.21)^{a}	1.11(0.15)^{d}	0.04(0.00)^{g}	0.09(0.01)^{f}	GC-MS, Co-GC
1,8-Cineole	1033	1213	0.85(0.09)^{b}	0.51(0.04)^{e}	1.06(0.11)^{a}	0.61(0.05)^{d}	0.16(0.01)^{g}	0.33(0.03)^{f}	0.52(0.06)^{e}	0.73(0.08)^{c}	GC-MS, Co-GC
(*Z*)-β-Ocimene	1040	1246	1.16(0.15)^{c}	1.48(0.16)^{b}	1.57(0.17)^{a}	0.63(0.06)^{d}	0.16(0.01)^{ef}	0.20(0.02)^{e}	0.04(0.00)^{g}	0.10(0.01)^{fg}	GC-MS, Co-GC
γ-Terpinene	1062	1266	0.20(0.02)^{c}	0.42(0.05)^{a}	0.16(0.01)^{cd}	0.32(0.04)^{b}	0.13(0.01)^{cd}	0.11(0.01)^{d}	0.12(0.01)^{d}	0.14(0.01)^{cd}	GC-MS, Co-GC
cis-Linalool oxide	1074	1478	0.34(0.03)^{e}	0.46(0.05)^{d}	0.99(0.01)^{a}	0.48(0.05)^{d}	0.57(0.06)^{c}	0.72(0.05)^{b}	0.06(0.01)^{g}	0.21(0.03)^{f}	GC-MS
Terpinolene	1088	1290	1.77(0.18)^{d}	2.81(0.32)^{b}	1.89(0.25)^{c}	1.63(0.19)^{e}	0.11(0.01)^{h}	0.55(0.06)^{g}	3.19(0.45)^{a}	1.02(0.12)^{f}	GC-MS, Co-GC
trans-linalool oxide	1088	1450	0.34(0.05)^{c}	0.16(0.02)^{c}	nd	nd	0.11(0.01)^{f}	0.46(0.05)^{b}	0.48(0.05)^{a}	0.19(0.02)^{d}	GC-MS
Linalool	1098	1553	48.56(5.68)^{h}	48.86(6.21)^{g}	68.37(7.65)^{f}	68.69(7.85)^{e}	70.29(6.94)^{d}	70.56(8.10)^{c}	73.01(8.41)^{b}	80.04(9.12)^{a}	GC-MS, Co-GC
Camphor	1143	1532	2.63(0.35)^{a}	0.33(0.04)^{d}	0.56(0.07)^{b}	0.48(0.05)^{c}	0.58(0.06)^{b}	0.27(0.03)^{e}	0.35(0.04)^{d}	0.11(0.01)^{f}	GC-MS
Borneol	1165	1719	0.48(0.05)^{e}	4.16(0.52)^{a}	0.15(0.01)^{f}	0.74(0.08)^{c}	0.60(0.05)^{d}	0.51(0.06)^{de}	0.90(0.05)^{b}	0.85(0.09)^{b}	GC-MS
Menthol	1173	1628	0.19(0.02)^{e}	0.25(0.02)^{d}	0.44(0.03)^{b}	0.09(0.01)^{f}	1.79(0.21)^{a}	0.29(0.03)^{c}	0.09(0.01)^{f}	0.25(0.02)^{d}	GC-MS
Terpinene-4-ol	1178	1611	0.30(0.04)^{b}	0.52(0.06)^{a}	0.49(0.06)^{a}	0.56(0.06)^{a}	0.08(0.01)^{c}	0.26(0.03)^{b}	0.24(0.02)^{b}	0.52(0.06)^{a}	GC-MS, Co-GC
p-Cymen-8-ol	1183	1864	2.33(0.26)^{b}	1.37(0.15)^{d}	0.74(0.08)^{e}	0.14(0.01)^{f}	3.68(0.43)^{a}	0.78(0.08)^{e}	1.40(0.18)^{d}	1.82(0.21)^{c}	GC-MS, Co-GC
cis–Hex-3-enyl butyrate	1188	1485	0.47(0.05)^{d}	tr	0.56(0.06)^{c}	0.38(0.04)^{e}	0.58(0.06)^{c}	2.88(0.34)^{a}	0.72(0.06)^{b}	0.26(0.03)^{f}	GC-MS
α-Terpineol	1189	1706	0.11(0.01)^{g}	1.24(0.16)^{d}	1.88(0.23)^{a}	1.83(0.22)^{b}	1.11(0.15)^{c}	1.44(0.14)^{c}	Tr	0.22(0.02)^{f}	GC-MS, Co-GC
cis-Dihydrocarvone	1193	1645	0.16(0.02)^{e}	nd	0.44(0.05)^{bc}	0.35(0.04)^{d}	0.41(0.03)^{c}	0.58(0.07)^{a}	0.45(0.05)^{b}	0.59(0.07)^{a}	GC-MS
Nerol	1228	1797	0.34(0.04)^{c}	1.37(0.21)^{a}	0.85(0.09)^{b}	0.11(0.01)^{f}	0.17(0.01)^{e}	0.15(0.01)^{e}	0.09(0.01)^{f}	0.22(0.01)^{d}	GC-MS
β-Citronellol	1228	1772	5.39(0.62)^{b}	6.81(0.66)^{a}	0.68(0.70)^{g}	0.75(0.70)^{f}	1.37(0.15)^{d}	2.13(0.31)^{c}	1.09(0.12)^{e}	0.63(0.07)^{h}	GC-MS
Neral	1240	1694	0.28(0.02)^{f}	0.69(0.07)^{b}	0.28(0.03)^{f}	0.48(0.05)^{d}	0.81(0.09)^{a}	0.38(0.05)^{e}	0.60(0.07)^{c}	0.63(0.07)^{bc}	GC-MS
Carvone	1242	1751	0.16(0.01)^{cd}	0.27(0.03)^{a}	0.23(0.02)^{b}	0.19(0.02)^{c}	0.12(0.01)^{e}	0.18(0.02)^{c}	Nd	0.13(0.01)^{de}	GC-MS, Co-GC
Geraniol	1255	1857	tr	0.72(0.06)^{cd}	0.67(0.07)^{d}	0.67(0.06)^{d}	0.76(0.07)^{bc}	0.90(0.11)^{a}	0.81(0.09)^{b}	0.95(0.08)^{a}	GC-MS, Co-GC
Geranial	1270	1742	0.57(0.04)^{a}	0.11(0.01)^{c}	0.12(0.01)^{c}	0.23(0.02)^{b}	0.30(0.03)^{b}	0.60(0.05)^{a}	0.08(0.01)^{c}	0.11(0.01)^{c}	GC-MS
Anethole	1283	1828	0.29(0.02)^{b}	0.40(0.03)^{b}	0.54(0.04)^{a}	0.62(0.05)^{a}	0.14(0.01)^{c}	nd	0.02(0.00)^{cd}	nd	GC-MS
Thymol	1290	2198	1.02(0.10)^{a}	tr	0.56(0.04)^{b}	tr	0.15(0.01)^{d}	0.24(0.02)^{c}	Tr	tr	GC-MS, Co-GC
Carvacrol	1292	2239	0.05(0.00)^{d}	0.55(0.04)^{a}	0.47(0.03)^{b}	0.45(0.03)^{b}	0.13(0.01)^{c}	tr	0.02(0.00)^{de}	0.05(0.00)^{d}	GC-MS
δ-Elemene	1339	1479	2.09(0.25)^{b}	2.52(0.28)^{a}	0.03(0.00)^{h}	1.25(0.12)^{c}	1.12(0.10)^{f}	1.84(0.23)^{c}	1.38(0.16)^{d}	0.97(0.08)^{g}	GC-MS
Eugenol	1356	2192	0.07(0.01)^{e}	0.13(0.01)^{d}	0.33(0.02)^{c}	0.65(0.07)^{a}	0.47(0.04)^{b}	0.65(0.05)^{a}	0.03(0.00)^{e}	0.04(0.00)^{e}	GC-MS, Co-GC
Neryle acetate	1356	1733	tr	1.57(0.12)^{a}	0.06(0.00)^{e}	0.25(0.02)^{c}	0.26(0.03)^{c}	0.97(0.07)^{b}	0.09(0.00)^{d}	0.10(0.01)^{d}	GC-MS
Geranyle acetate	1383	1765	14.24(0.19)^{a}	9.33(0.11)^{b}	4.89(0.51)^{e}	4.52(0.55)^{g}	3.84(0.41)^{h}	7.21(0.65)^{c}	5.81(0.60)^{d}	4.85(0.56)^{f}	GC-MS, Co-GC
β-Caryophyllene	1418	1612	2.31(0.31)^{c}	3.49(0.39)^{a}	0.11(0.01)^{f}	2.38(0.31)^{d}	2.41(0.21)^{c}	0.11(0.01)^{f}	3.22(0.29)^{b}	0.12(0.01)^{f}	GC-MS
α-Humulene	1454	1687	0.10(0.01)^{d}	0.27(0.03)^{b}	0.15(0.01)^{c}	0.16(0.02)^{c}	0.31(0.02)^{a}	tr	0.02(0.00)^{e}	tr	GC-MS
Germacrene-D	1480	1726	0.02(0.00)^{e}	tr	0.21(0.03)^{c}	1.02(0.10)^{a}	0.38(0.04)^{b}	0.23(0.02)^{c}	0.03(0.00)^{e}	0.10(0.01)^{d}	GC-MS
Eugenyle acetate	1524	nd	1.20(0.15)^{a}	nd	0.11(0.01)^{de}	0.12(0.01)^{d}	0.23(0.02)^{c}	0.13(0.01)^{d}	0.04(0.00)^{ef}	0.48(0.05)^{b}	GC-MS
Grouped compounds											
Monoterpene hydrocarbons			8.04(1.02)^{a}	7.65(0.98)^{b}	7.65(1.00)^{b}	4.91(0.64)^{c}	4.18(0.62)^{c}	3.23(0.41)^{f}	4.71(0.51)^{d}	1.92(0.23)^{g}	
Aromatic hydrocarbons			0.88(0.08)^{a}	0.24(0.03)^{f}	0.12(0.01)^{g}	0.31(0.02)^{de}	0.38(0.04)^{c}	0.28(0.03)^{ef}	0.35(0.04)^{cd}	0.55(0.04)^{b}	
Monoterpene alcohols			58.06(7.85)^{g}	65.83(8.23)^{f}	75.14(9.45)^{d}	74.85(9.79)^{c}	80.46(10.23)^{b}	77.67(7.56)^{c}	77.68(8.51)^{c}	85.54(11.23)^{a}	
Phenols			1.07(0.26)^{a}	0.55(0.06)^{b}	1.03(0.21)^{a}	0.45(0.05)^{c}	0.28(0.02)^{d}	0.24(0.02)^{d}	0.02(0.00)^{e}	0.05(0.00)^{e}	
Monoterpene esters			15.44(3.26)^{a}	10.90(1.25)^{b}	5.06(0.95)^{f}	4.89(0.84)^{f}	4.33(0.73)^{g}	8.31(0.99)^{c}	5.94(0.77)^{d}	5.43(0.64)^{e}	
Monoterpene ketones			2.95(0.42)^{a}	0.60(0.05)^{e}	1.23(0.15)^{b}	1.02(0.12)^{c}	1.11(0.11)^{bc}	1.03(0.10)^{c}	0.80(0.06)^{d}	0.83(0.07)^{d}	
Monoterpene aldehydes			0.85(0.07)^{bc}	0.80(0.06)^{cd}	0.40(0.03)^{e}	0.71(0.06)^{cd}	1.11(0.12)^{a}	0.98(0.08)^{ab}	0.68(0.05)^{d}	0.74(0.06)^{cd}	
Monoterpene ethers			1.53(0.13)^{b}	1.13(0.10)^{c}	2.05(0.22)^{a}	1.09(0.09)^{de}	0.84(0.06)^{f}	1.51(0.18)^{b}	1.06(0.10)^{e}	1.13(0.13)^{cd}	
Sesquiterpenes			4.52(0.05)^{d}	6.28(0.07)^{a}	0.50(0.04)^{h}	4.81(0.04)^{b}	4.22(0.03)^{c}	2.18(0.03)^{f}	4.65(0.03)^{c}	1.19(0.12)^{g}	
Non terpenics			0.69(0.08)^{f}	0.84(0.09)^{e}	1.40(0.16)^{b}	1.11(0.11)^{c}	0.83(0.07)^{e}	3.00(0.35)^{a}	1.03(0.12)^{d}	0.49(0.05)^{g}	
Total identified (%)			94.03(9.56)^{h}	94.82(9.12)^{e}	94.58(8.23)^{f}	94.15(9.87)^{g}	97.74(9.63)^{c}	98.43(9.48)^{a}	96.92(8.99)^{d}	97.87(9.27)^{b}	

tr: trace (<0.01%). nd: not detected. [a] Apolar HP-5 MS column. [b] Polar HP Innowax column. Volatile compounds percentages in the same line with different superscript (a–h) are significantly different at $P < 0.05$. Values in brackets are standard deviations.

the composition of EO of coriander fruit as affected by maturity stage, could occur as a result of complex chemical modifications of terpenes, including temperature-induced oxidative processes (Herath *et al.* 1979; Usai *et al.* 1992). In fact, in this study, the significant changes in the composition of EO of coriander fruit at different stages of maturity studied in this investigation can be used as an important marker of the fruit-maturation process. In addition, our earlier studies (Msaada 2007; Msaada *et al*. 2009b, 2009c) showed that at full ripeness, the chemical composition of coriander fruit exhibits high amount of petroselinic acid (≈80% of total fatty acid). In this study also, linalool was present near 80%. Thus, it appears that these metabolites can be used as systematic markers of coriander.

These results are in good agreement with the findings of Cristiana *et al*. (2006), who reported that the composition of EO obtained from *Ocimum gratissimum* leaves at different seasons of the year varied markedly. Kofidis *et al*. (2006) also reported seasonal variation in the chemical composition of EOs of *Mentha spicata*, grown in the wild conditions in Greece. The EO was reported to contain the maximum amounts of linalool in October (mid-autumn) and the minimum in June (summer). Celiktas *et al*. (2007) also reported variation in the chromatographic profile of *Rosmarinus officinalis* due to different seasons. In contrast, da Silva *et al*. (2003) reported that there were no considerable changes in the chromatographic profiles of EO of basil with regard to two different seasons of the year.

Effects of season [S], stage of maturity [SM] and [SM] × [S] on EO compounds

The combined analysis of variance (**Table 3**) showed that among the 41 identified compounds, only α-terpineol was insignificantly ($P < 0.05$) affected by the crop season, the stage of maturity, and the season x stage of maturity interaction. α-Terpinene, terpinene-4-ol and carvone were not affected by the season, while the *p*-cymen-8-ol was affected neither by the season nor by the season × stage of maturity interaction. The ramaining 36 compounds were strongly ($P < 0.001$) affected by the crop season, stage of maturity and the season × stage of maturity interaction. These significant effects could be due to the difference in the environmental factors between the two seasons like mean daily temperature (**Table 2**), photoperiod, quality of soil, cultural practices, weeds, plant diseases, crop insects and pests that could also affect EO composition. However, these factors were not investigated in this study. EO composition could be sensitive to the maturation process, year variation and weather conditions as well as to variation in the soil environment caused by cropping history. The observed differences in the EO composition between the two crop seasons in the present study could perhaps be explained by the variation in climatic conditions. Moreover, since the EO content in the fruit is related to the volatile compounds emitted by the crop (Mookherjee 1989), differences in the chemical signals emitted by the crop may produce different impacts on the arthropod community. It is important to remark that host chemical shifting is an important mechanism by which plants regulate insect fauna (Jones 1991). Many different insect species are pollinators or visitors of coriander (Diederichsen 1996). More stable chemical signals such as those given by the both crop season may transmit arthropod-specific information, whereas variable chemical signals may be related to generalist arthropod species (Rausher 1992). The relationship between the environmental conditions (wind, soil, temperature, moisture, etc.) and the emission of volatile compounds could be used as a method to improve agricultural practices such as flower pollination (Gil *et al.* 2002). In particular, EO emissions could then be targeted to attract communities of arthropods that are neutral or positive to crop production. In addition, coriander EO showed high activity against nematodes, especially *Bursaphelenchus xylophilus*, as indicated by Kim *et al*. (2008) and this could be due to the toxicity of linalool and related esters such as linalyl acetate (Bickers *et al.* 2003). The volatile oil of coriander also possesses high antifungal activity and can be used as a natural sprout suppressant (Singh *et al*. 2006).

CONCLUSIONS

In summary, the results showed that EO yields were maximally affected by the stage of maturity, the crop season and their interaction. The EO compounds were also significantly affected by the stage of maturity, the crop season and their interaction. Significant changes were found in the EO composition of coriander, as influenced by the stage of maturity and the crop season. It indicates the existence of complex chemical transformations of terpenes resulting in alterations in the organoleptic features of the oil and in the accumulation of substances that change the oil flavour and can be hazardous to human health. Our data suggest that the constantly growing use of natural EOs in the medicine, cosmetics and food industry, requires attention to be paid for consideration of the effects of maturity stage and crop season on the EO or on the oil based substances that relate with the composition and quality of the products.

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