

Changes on essential oil composition of coriander (*Coriandrum sativum* L.) fruits during three stages of maturity

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Abstract

The essential oils composition of coriander (*Coriandrum sativum* L.) fruits obtained by hydrodistillation was studied at three stages of maturity by GC–FID and GC–MS. Essential oil yields showed marked increase during maturation process and forty one compounds were identified. Geranyl acetate (46.27%), linalool (10.96%), nerol (1.53%) and neral (1.42%) were the main compounds at the first stage of maturity (immature fruits). At the middle stage, linalool (76.33%), *cis*-dihydrocarvone (3.21%) and geranyl acetate (2.85%) were reported as the main constituents. Essential oils at the final stage of maturity (mature fruits) consist mainly on linalool (87.54%) and *cis*-dihydrocarvone (2.36%). Additionally, accumulation of monoterpene alcohols and ketones was observed during maturation process of coriander fruit.

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1. Introduction

Coriander (*Coriandrum sativum* L.) is a culinary and medicinal plant from the Umbelliferae family. This plant is of economic importance since it has been used as flavoring agent in food products, perfumes and cosmetics. As a medicinal plant, *C. sativum* L. has been credited with a long list of medicinal uses. Powdered seeds or dry extract, tea, tincture, decoction or infusion have been recommended for dyspeptic complaints, loss of appetite, convulsion, insomnia and anxiety (Emamghoreishi, Khasaki, & Aazam, 2005). Moreover, the essential oils and various extracts from coriander have been shown to possess anti-

bacterial (Burt, 2004; Cantore, Iacobellis, De Marco, Capasso, & Senatore, 2004; Kubo, Fujita, Kubo, Nihei, & Ogura, 2004), antioxidant (Wangenstein, Samuelsen, & Malterud, 2004), antidiabetic (Gallagher, Flatt, Duffy, & Abdel-Wahab, 2003), anticancerous and antimutagenic (Chithra & Leelamma, 2000) activities. Many phytochemical studies so far investigated the chemical composition of the essential oil from *C. sativum* L. seeds from different origins (Anitescu, Doneanu, & Radulescu, 1997; Bandoni, Mizrahi, & Juarez, 1998; Steinegger & Hansel, 1988). Evaluations of the oil composition extracted from leaves have also been reported (Eyres, Dufour, Hallifax, Sotheeswaran, & Marriott, 2005). Chemical essential oil composition of Italian coriander fruits was greatly influenced by the age and origins as mentioned by Carruba and la Tore (2002). Additionally, variations of the essential oil composition in many different fruits have been observed, depending on genetic and environmental factors as well as ontogeny and analytical methods (Eyres et al., 2005; Lawrence,

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2002). The effect of maturity stage on the essential oil composition was also reported (Sampaio & Nogueira, 2006; Vendramini & Trugo, 2000; Visai & Vanoli, 1997).

Although the substantial data of its chemical composition, there are no previous phytochemical reports that have been recorded for *C. sativum* L. from Tunisia.

In the present work, we investigated for the first time the chemical composition of the essential oil isolated from the Tunisian coriander fruits at different stages of maturity.

2. Materials and methods

2.1. Plant material

Coriander fruits were collected from cultivated plants in the region of Menzel Temime (Northeastern Tunisia) during May and June 2005. For collection at the initial stage of maturity, only full green fruits were harvested. For the middle stage, green-brown fruits were picked up. Only brown fruits were chosen as the final stage of maturity (mature).

2.2. Essential oil isolation

Fresh fruits (100 g, three times for each stage) were subjected to hydrodistillation for 90 min. The distillate was extracted with 2-methyl-butane (v/v) and dried over anhydrous sodium sulphate. The organic layer was then concentrated, at 30 °C using a Vigreux column and the resulting essential oil was subsequently analysed.

2.3. GC–FID and GC–MS analysis

2.3.1. GC–FID

Analytical gas chromatography was carried out on a Hewlett–Packard 6890 gas chromatograph equipped with a flame ionization detector (FID) and an electronic pressure control (EPC) injector. A polar HP Innowax (PEG) column and an apolar HP-5 column (30 m × 0.25 mm, 0.25 µm film thickness) were used. The flow of the carrier gas (N₂) was 1.6 ml/min. The split ratio was 60:1. The analysis was performed using the following temperature program: oven temps isotherm at 35 °C for 10 min, from 35 to 205 °C at the rate of 3 °C/min and isotherm at 205 °C during 10 min. Injector and detector temperature were held, respectively, at 250 and 300 °C. The injection volume was 1 µl.

2.3.2. GC–MS

GC–MS analysis was performed on a gas chromatograph HP 5890 (II) interfaced with a HP 5972 mass spectrometer with electron impact ionization (70 eV). A HP-5MS capillary column (30 m × 0.25 mm, 0.25 µm film thickness) was used. The column temperature was programmed to rise from 50 °C to 240 °C at a rate of 5 °C/min. The carrier gas was helium with a flow rate of 1.2 ml/min; split ratio was 60:1. Scan time and mass range were 1 s and 40–300 *m/z*, respectively.

2.4. Compounds identification

The identification of the oil constituents was based on a comparison of their retention indices relative to (C₈–C₂₂) *n*-alkanes with those of 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). Quantitative data were obtained from the electronic integration of the FID peak areas.

3. Results and discussion

The hydrodistillation of 100 g of *C. sativum* L. fruits gave essential oils at initial, middle and the final stage of maturity with a yield of 0.01%, 0.12% and 0.35% (w/w), based on dry weight, respectively. In this case, especially at complete maturity, obtained yields were low when compared to other previously investigated samples (Carruba & la Tore, 2002).

The composition of the essential oils at different stages of maturity is listed in Table 1. A total of 41 compounds representing 66.29%, 86.92% and 95.39% of the total detected constituents at initial, middle and the final stage of maturity respectively.

The first stage of maturity offered an essential oil which consists mainly on monoterpene esters (Table 2) represented exclusively by geranyl acetate (46.27%). monoterpene alcohols (14.66%) were the second main constituents of the oil and contained linalool as the main compound (10.96%). The remaining fractions as monoterpene aldehydes (2.07%), ethers (0.87%), hydrocarbons (0.24%) and monoterpene ketones (0.97%) as well as phenols (1.06%) and sesquiterpenes (0.15%) were weakly represented (Table 2).

At the middle and the final stage of maturity, the chemical composition of the fruits essential oil showed similar profiles and differed markedly from the first stage (immature fruits). Thus, over 84.92% of the compounds detected in the fruit at the middle stage were monoterpene alcohols (76.77%), ketones (3.43%), esters (2.85%) and ethers (1.87%) (Table 2). At this stage, the major constituents of the oil were linalool (76.33%), *cis*-dihydrocarvone (3.21%), geranyl acetate (2.85%) and anethole (1.41%). Other constituents were present in amount less than 1% (Table 1).

Essential oils of the mature fruit (final stage) were predominated by monoterpene alcohols (88.51%) and ketones (2.61%). Interestingly, the phenols advocated as a potent antioxidant were found with appreciable amount (2.31%). The mature fruit essential oil was composed mainly of linalool (87.54%) and *cis*-dihydrocarvone (2.36%).

The observed increase of the monoterpene alcohols fraction concomitant with marked increase of linalool may be linked, in quality and in quantity, with the maturity stages. On the other hand, the deep changes in the coriander fruit

Table 1
Essential oil composition (% w/w) of coriander fruit at three stage of maturity

Compound	RI ^a	RI ^b	Immature	Intermediate	Mature	Identification	
01	Heptanal	901	1194	tr	tr	tr	GC–MS
02	α -Thujene	931	1035	tr	tr	tr	GC–MS
03	α -Pinene	939	1032	0.01	tr	0.02	GC–MS, Co-GC
04	Sabinene	976	1132	tr	tr	0.03	GC–MS
05	β -Pinene	980	1118	tr	0.20	0.05	GC–MS, Co-GC
06	δ^3 -Carene	1011	1159	0.09	0.10	0.02	GC–MS
07	α -Terpinene	1018	1188	tr	tr	0.01	GC–MS, Co-GC
08	<i>p</i> -Cymene	1026	1280	tr	tr	tr	GC–MS, Co-GC
09	Limonene	1030	1203	0.04	tr	0.02	GC–MS, Co-GC
10	1,8-Cineole	1033	1213	0.23	0.14	0.20	GC–MS, Co-GC
11	(Z)- β -Ocimene	1040	1246	0.08	tr	tr	GC–MS, Co-GC
12	γ -Terpinene	1062	1266	tr	tr	tr	GC–MS
13	<i>cis</i> -Linalool oxide (furanoid)	1074	1478	0.32	0.32	0.27	GC–MS
14	Terpinolene	1088	1290	0.02	0.18	0.15	GC–MS, Co-GC
15	Linalool	1088	1553	10.96	76.33	87.54	GC–MS, Co-GC
16	<i>trans</i> -Linalool oxide (furanoid)	1088	1450	0.27	tr	tr	GC–MS
17	Camphor	1143	1532	0.86	0.13	0.17	GC–MS
18	Borneol	1165	1719	0.08	0.28	0.34	GC–MS
19	Menthol	1173	1628	0.14	0.16	0.05	GC–MS
20	Terpinene-4-ol	1178	1611	tr	tr	tr	GC–MS, Co-GC
21	<i>p</i> -Cymen-8-ol	1183	1864	1.36	tr	tr	GC–MS
22	<i>cis</i> -Hex-3-enyl butyrate	1188	1485	tr	tr	0.01	GC–MS
23	α -Terpineol	1189	1706	0.39	tr	0.05	GC–MS, Co-GC
24	<i>cis</i> -Dihydrocarvone	1193	1645	0.01	3.21	2.36	GC–MS
25	Nerol	1228	1797	1.53	tr	tr	GC–MS
26	β -Citronellol	1228	1772	0.11	tr	0.52	GC–MS, Co-GC
27	Neral	1240	1694	1.42	0.10	0.13	GC–MS
28	Carvone	1242	1751	0.10	0.09	0.08	GC–MS, Co-GC
29	Geraniol	1255	1857	tr	tr	tr	GC–MS, Co-GC
30	Geranial	1270	1742	0.65	tr	0.03	GC–MS
31	Anethole	1283	1828	0.05	1.41	0.01	GC–MS
32	Thymol	1290	2198	0.02	0.99	1.85	GC–MS, Co-GC
33	Carvacrol	1292	2239	1.04	0.11	0.46	GC–MS
34	δ -Elemene	1339	1479	tr	0.05	0.01	GC–MS
35	Eugenol	1356	2192	0.09	tr	0.01	GC–MS, Co-GC
36	Neryl acetate	1356	1733	tr	tr	tr	GC–MS
37	Geranyl acetate	1383	1765	46.27	2.85	0.83	GC–MS, Co-GC
38	β -Caryophyllene	1418	1612	0.02	0.07	0.03	GC–MS
39	α -Humulene	1454	1687	0.09	tr	0.02	GC–MS, Co-GC
40	Germacrene-D	1480	1726	0.04	0.19	0.05	GC–MS
41	Eugenyl acetate	1524	–	tr	tr	0.07	GC–MS
Total identified				66.29	86.91	95.39	

tr: trace (<0.01%).

^a Apolar HP-5 MS column.

^b Polar HP Innwax column.

Table 2
Volatile compounds classes percentages at three stage of maturity

Classes	Immature	Intermediate	Mature
Monoterpene hydrocarbons	0.24	0.48	0.3
Aromatic hydrocarbons	tr	tr	tr
Monoterpene alcohols	14.66	76.77	88.51
Phenols	1.06	1.1	2.31
Monoterpene esters	46.27	2.85	0.90
Monoterpene ketones	0.97	3.43	2.61
Monoterpene aldehydes	2.07	0.10	0.16
Monoterpene ethers	0.87	1.87	0.48
Sesquiterpenes	0.15	0.31	0.11
Non terpenic	tr	tr	0.01

tr: trace (<0.01%).

essential oils composition during stages of maturity can be used as a marker of the maturation process.

Since there are no previous citations about the evolution of the chemical composition of the coriander fruit essential oil during maturation, it is impossible to compare our results with earlier work. Nevertheless, such studies concerning other fruits were undertaken. Thus, acerola fruit (Vendramini & Trugo, 2000) and mangaba fruit (Sampaio & Nogueira, 2006) from Brazil were reported to be prevailed by the esters, alcohols, aldehydes and ketones which contribute markedly to the fruity note of the fruits as reported by (Mathesis, Buchanan, & Fellman, 1992). In contrast to our results, it is worth

mentioning that the esters fraction in the two previously mentioned studies was the less represented group compounds in the first stage of maturation for both acerola and mangaba fruits, while alcohols were the predominant fraction (Sampaio & Nogueira, 2006; Vendramini & Trugo, 2000).

Overall, the results reported on the chemical composition of the essential oil of the coriander fruits during different stages of maturity revealed great differences occurring during maturation process. It may be suggested that these differences were concomitant with modifications in secondary metabolism.

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