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ESSENTIAL OIL COMPOSITION OF THE CORIANDER (*Coriandrum sativum* L.) HERB DEPENDING ON THE DEVELOPMENT STAGE

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Abstract

The herbal material of *Coriandrum sativum* is the fruit. Fresh herb is also used as an aromatic spice. The aim of the present study was to evaluate the content and chemical composition of coriander herb obtained at different plant growth stages. Coriander plants were grown in a glasshouse, the herb was harvested at the initial stage of flowering and from regrowing shoots. Essential oil extraction from the herb was performed by the hydrodistillation method, whereas the assessment of the chemical composition – using GC-MS method. The examined material contained 0.17–0.29 ml × $100g^{-1}$ of essential oil, depending on the stage of plant development when the harvest was done. 61 (generative phase) and 65 (vegetative phase) compounds were found in the examined coriander oil. The essential oil from the coriander herb contained the highest amount of aliphatic aldehydes, among which was decanal, *E*-2-dodecanol and *E*-2-decenol had the highest percentages. The contents of most aliphatic aldehydes decreased with each subsequent harvest of the herb. In addition to the above-mentioned aliphatic aldehydes, the presence of linalool, phytol, and oleic acid was found in the essential oil extracted from the coriander herb.

Key words: Apiaceae, cilantro, volatiles, GC-MS, ontogeny, C_{10}–C_{16} aldehydes

INTRODUCTION

Coriander (*Coriandrum sativum* L.) – an annual of the Apiaceae family, is one of valuable medicinal, seasoning and oliferous plants. This species comes from the Mediterranean region and it is grown all over the world. The coriander fruit (*Coriandri fructus*) and the essential oil isolated from it are used for medicinal purposes (Duarte et al. 2012; Mahendra and Bisht, 2011; Sriti et al. 2011; Chung et al. 2012). The coriander fruit shows relaxant activity in the alimentary tract; coriander raw material and oil are also used as an aromatising agent in the pharmaceutical, food, cosmetics and perfume industries. The fresh coriander herb, containing essential oil (Potter and Fagerson, 1990; Telci and Hisil, 2008), fatty acids (Neffati and Marzouk, 2008), flavonoids (Oganesyan et al. 2007), carotenoids (Raju et al. 2007) as well as coumarin compounds (Taniguchi et al. 1996) is also an aromatic seasoning. Coriander leaves, known as "asotu" in eastern Anatolya and as "cilantro" in the United States, are eaten fresh. The aroma of the coriander fruit and herb is completely different. While aliphatic aldehydes (mainly C_{10}–C_{16} aldehydes), with their unpleasant odour, are the main components of the volatile oil from the fresh herb (Potter and Fagerson, 1990), linalool and other oxidized monoterpenes as well as monoterpene hydrocarbons predominate in the oil distilled from the fruit (Bhuiyan et al. 2009). The varying composition of coriander volatile oil is the reason why the herb from young plants is used to prepare curry, soups and sauces, whereas the fruit is mainly used as a seasoning for pickles, cold meats, confectionery products, and seasoning mixtures.

The coriander essential oil content and chemical composition are significantly affected by various factors. The accumulation and chemical composition of essential oil in plants are determined by different factors: environmental (Rakic and Johnson, 2002; Sriti et al. 2011), genetic (Zheljazkov et al. 2008; Ebrahimi et al. 2010), ontogenetic (Msaada et al. 2007; Mohammadi and Saharkhiz, 2011) as well as cultivation (Zheljazkov et al.

2008). In the case of coriander, the ontogenetic variability is especially important. The chemical composition of coriander essential oil undergoes significant changes during ontogenesis, which affects the aroma of the plant. The biggest change in the composition of coriander volatile oil during ontogenesis concerns the production of monoterpene linalool alcohol from aliphatic aldehydes (Bhuiyan et al. 2009). Large differences in the composition of coriander essential oil also relate to cultivated forms and cultivated (Telci et al. 2006a; b; Zheljazkov et al. 2008). The essential oil extracted from coriander fruit has been identified quite well as far as its chemical composition is concerned. However, the chemical composition of herbal volatile oil has not been determined sufficiently. Potter and Fagerson (1990) determined 41 compounds in the volatile oil distilled from coriander leaves, among which 82% were aldehydes and 16.65% – alcohol. Similarly, Telci and Hisil (2008) report aliphatic aldehydes as the main components of coriander herb essential oil. Bhuiyan et al. (2009) identified 44 compounds in the coriander herb oil, mainly from the group of aliphatic acids.

Coriander, as most species synthesizing essential oil, has high light requirements, and at the stage of generative development – also high temperature requirements. Light and temperature are the factors affecting oil plant growth and essential oil synthesis, whereas the effect of the above-mentioned factors is stronger in plants whose oil reservoirs are on the surface of leaves and flowers (exogenous) rather than in fruits, seeds, or roots (endogenous). The aim of the present study was to determine and compare the composition of herbal essential oil of coriander grown in a glasshouse, obtained at two stages of plant development: vegetative and generative.

MATERIALS AND METHODS

Coriander (*Coriandrum sativum* L.) plants of the cultivar 'Jantar' were grown in a periodically heated glasshouse of the Experimental Farm of the University of Life Sciences in Lublin, situated in the north-southern direction, in the period from the first decade of March to the first decade of June 2008. During the heating period, lasting until the end of April, the temperature in the glasshouse was in the range from 18°C to 25°C (day) and 10°C – 16°C (night). In the remaining period, the temperature depended on the weather, but it was not allowed to exceed 28°C by airing and shading the site. The plants were grown in industrial pots with the dimensions of 9 × 9 × 10 cm. The substrate was peat used for growing vegetables and herbal plants (pH 5.5–6.5). Sowing was performed on the 28th of March; coriander seeds were evenly distributed over the whole pot surface. The herb was harvested was conducted at two stages of plant development: the generative phase (beginning of flowering: 11th May) and the vegetative phase (from regrowing shoots: 6th of June).

The essential oil was extracted from air-dried powdered material (40 g) in a glass Clevenger-type distillation apparatus by using a method following Polish Pharmacopoeia VI guidelines (2002) and subjecting the material to hydrodistillation for three hours. The extracted essential oil was stored in a dark glass container at a temperature of -10°C, until the time of chromatographic separation. Qualitative and quantitative analysis of the coriander essential oil was performed using a ITMS Varian 4000 GC-MS/MS (Varian, USA) GC-MS instrument, equipped with a CP-8410 auto-injector and a 30 m × 0.25 mm i.d. VF-5ms column (Varian, USA), film thickness 0.25 µm; carrier gas, helium at a rate of 0.5 ml × min^{-1}; injector and detector temperature, 250°C; split 1:100 × 1 µl of the solution was injected (10 µl of the sample in 1000 µl of hexane). A temperature of 50°C was applied for 1 min., then it was incremented to 250°C at a rate of 4°C. min^{-1}; 250°C was applied for 10 min. A VF- 5ms column was used (an equivalent of DB-5). Helium was the carrier gas, with a constant flow of 0.5 ml/min. Injector: 250°C; split 1:100. 1 µl of the solution was injected (10 µl of the sample in 1000 µl of hexane). A Varian 4000 MS/MS detector was used, recorded range: 40–1000 m/z, scan rate 0.8 sec/scan. The retention indices were determined based on the alkane series C_{10}–C_{40}. The qualitative analysis was carried out on the basis of MS spectra, which were compared with the spectra of the NIST library (Mass Spectral Library, 2008) and with data available in the literature. The identity of the compounds was confirmed by their retention indices, taken from the literature (Adams, 2004) and the author's own data.

RESULTS

The essential oil concentration in the coriander cultivar 'Jantar' at the beginning of flowering was 0.29 ml × $100g^{-1}$ (Table 1). In the examined oil, 61 compounds were detected, including 46 identified ones. Mass spectra were determined for the remaining compounds (Fig. 1). It was found that the predominant compounds in the investigated oil were aliphatic aldehydes: *E*-2-dodecanol (17.8%), decanal (15.3%), and *E*-2-decenol (11.9%). Moreover, the following compounds were found to have a significant percentage: dodecanal (4.7%), 1-decanol (4.2%), phytol (2.8%), undecanal (2.2%), tetradecanal (2.2%), *E*-2-undecenal (2.0%), oleic acid (2.0%), *E*-2-tridecenol (1.7%), cubenol (1.5%), and nonane (1.4%). The linalool content was 0.3%. The identified compounds constituted 99.8% of all the components of the examined oil. The

herb harvested on the second harvest date from regrowing coriander shoots contained 0.17 ml × 100g^{-1} of essential oil (Table 2). On the basis of GC-MS analysis, 65 compounds were determined, including 50 which were identified, in the essential oil extracted from the herb collected from regrowing shoots of coriander. Mass spectra were determined in the remaining unidentified compounds (Fig. 2). The determined compounds represented 99.8% of all the compounds in the coriander herb essential oil. Among the identified compounds, decanal (17.2%), *E*-2-dodecanol (16.5%), and *E*-2-decenol (14.2%) showed the highest percentages. Besides, the following compounds were found in higher amounts: nonane (3.4%), undecanal (2.1%), phytol (2.1%), tetradecanal (1.9%), and *E*-2-tridedecenal (1.7%). The concentration of linalool was 1.1%.

Table 1
Chemical composition of herb essential oil of *C. sativum* harvested in the generative stage

No	Compound	RT	RI	%	±SD	No	Compound	RT	RI	%	±SD
1	nonane	7.518	911	1.4	0.0	32	n.i.	27.707	1507	0.4	0.0
2	heptanal	7.701	916	tr.	-	33	tridecanal	28.187	1522	0.5	0.0
3	*α*-pinene	8.621	942	tr.	-	34	n.i.	29.061	1549	0.4	0.0
4	decane	10.677	999	0.1	0.0	35	*E*-2-tridecenal	29.477	1562	0.1	0.0
5	*n*-octanal	10.944	1007	0.2	0.0	36	*E*-nerolidol	29.646	1567	0.1	0.0
6	*p*-cymene	11.690	1027	tr.	-	37	germacrene D	29.751	1571	0.3	0.0
7	benzene acetaldehyde	12.577	1051	tr.	-	38	*E*-2-tridecenol	29.967	1578	1.7	0.0
8	*γ*-terpinene	12.845	1058	tr.	-	39	dodecanoic acid	30.112	1582	0.3	0.0
9	n.i.	13.326	1071	tr.	-	40	n.i.	30.771	1603	0.3	0.0
10	n.i.	13.959	1088	0.1	0.0	41	2-dodecenol	30.855	1606	0.6	0.0
11	undecane	14.203	1095	0.1	0.0	42	tetradecanal	31.198	1619	2.2	0.0
12	linalool	14.327	1098	0.3	0.0	43	cubenol	31.587	1634	1.5	0.0
13	nonanal	14.527	1104	0.7	0.0	44	n.i.	32.433	1666	0.6	0.0
14	cyclodecanol	14.944	1116	tr.	-	45	n.i.*	33.061	1689	11.8	0.0
15	*Z*-2-nonenal	16.577	1164	tr.	-	46	pentadecanal	34.037	1724	0.2	0.0
16	*n*-nonanol	16.925	1175	0.2	0.1	47	heptadecane	35.707	1783	1.6	0.0
17	4,*Z*-decenal	17.722	1198	0.2	0.0	48	1-dodecanal	37.116	1836	1.0	0.0
18	4,*E*-decenal	17.852	1202	0.5	0.0	49	dodecanal	37.306	1844	0.4	0.0
19	decanal	18.290	1215	15.3	0.1	50	n.i.	37.887	1866	0.4	0.0
20	*E*-2-decenal	19.653	1255	0.6	0.1	51	n.i.	38.191	1878	0.6	0.0
21	*E*-2-decenol	20.336	1275	11.9	0.0	52	*α*-humulene	38.348	1884	1.2	0.0
22	1-decanol	20.524	1281	4.2	0.1	53	trans-*β*-farnesene	38.724	1899	0.2	0.0
23	2-*n*-octylfuran	21.118	1298	0.3	0.0	54	1-dodecanol	38.980	1910	tr.	-
24	undecanal	21.689	1315	2.2	0.0	55	n.i.	40.264	1962	0.3	0.0
25	n.i.	23.089	1356	0.1	0.0	56	oleic acid	40.635	1978	2.0	0.1
26	*E*-2-undecenal	23.614	1372	2.0	0.0	57	phytol	43.799	2106	2.8	0.0
27	n.i.	24.672	1404	0.8	0.0	58	n.i.	44.886	2159	1.8	0.0
28	dodecanal	25.060	1417	4.7	0.0	59	n.i.	45.706	2199	0.5	0.0
29	*γ*-elemene	25.645	1437	0.3	0.0	60	n.i.	46.163	2218	0.4	0.0
30	*E*-2-dodecanal	26.375	1462	1.0	0.0	61	*n*-octadecanol	51.891	2487	0.6	0.1
31	*E*-2-dodecanol	27.067	1485	17.8	0.1						
Total/Identified compounds		>99.8%/>81.2%									
Essential oil content		0.29 ml × 100g^{-1}									

*see: Fig. 1

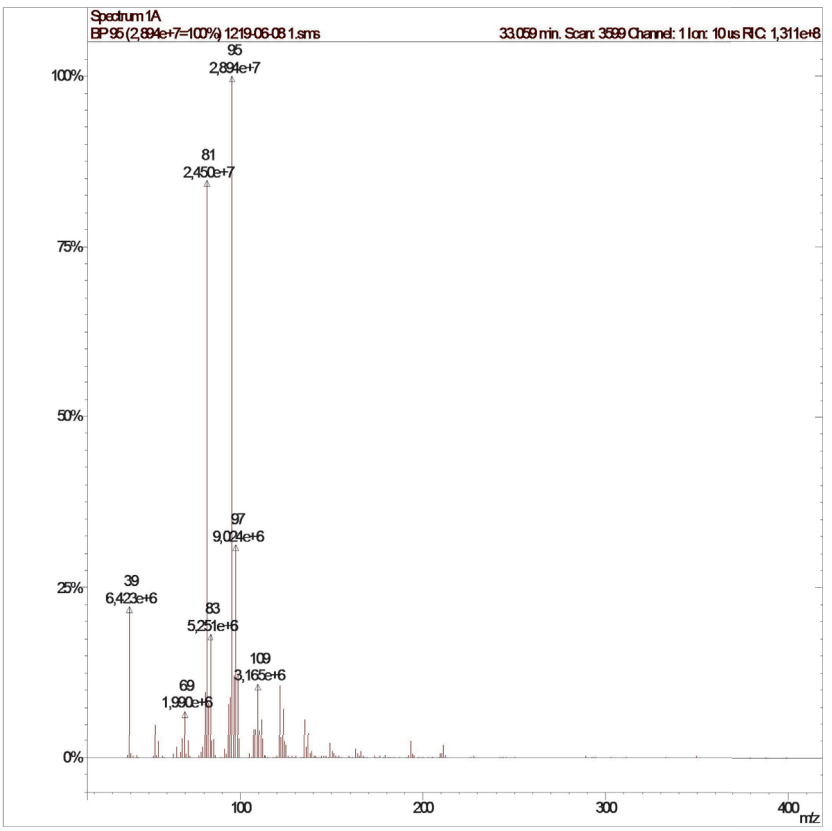

Fig. 1. Spectrum of unidentified compounds (RI 1689).

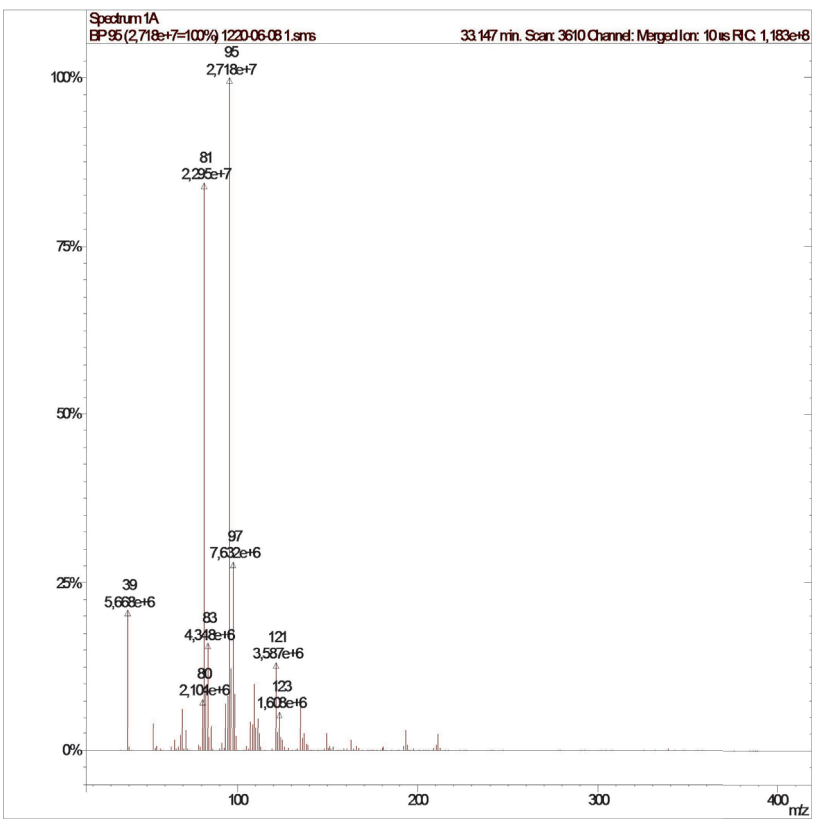

Fig. 2. Spectrum of unidentified compounds (RI 1692).

Table 2
Chemical composition of herb essential oil of *C. sativum* harvested in the vegetative stage

No	Compound	RT	RI	%	±SD	No	Compound	RT	RI	%	±SD
1	nonane	7.559	912	3.4	0.1	34	*E*-2-dodecanal	26.400	1463	1.0	0.1
2	heptanal	7.704	916	0.1	0.0	35	*E*-2- dodecanol	27.153	1488	16.5	0.3
3	*α*-pinene	8.633	942	tr	-	36	n.i.	27.736	1508	0.5	0.0
4	decane	10.687	1000	0.1	0.0	37	tridecanal	28.214	1523	0.6	0.1
5	*n*-octanal	10.950	1007	0.2	0.0	38	n.i.	29.081	1550	0.4	0.1
6	*p*-cymene	11.700	1027	tr	-	39	*E*-2-tridecenal	29.491	1563	0.2	0.0
7	limonene	11.834	1031	tr	-	40	*E*-nerolidol	29.666	1568	0.2	0.0
8	1,8-cineole	11.976	1035	tr	-	41	germacrene D	29.770	1571	0.2	0.0
9	benzene acetaldehyde	12.568	1051	tr	-	42	*E*-2-tridecenal	30.010	1579	1.7	0.1
10	*γ*-terpinene	12.857	1059	0.1	0.0	43	dodecanoid acid	30.132	1583	0.3	0.0
11	n.i.	13.333	1071	tr	-	44	n.i.	30.792	1604	0.4	0.0
12	n.i.	13.968	1089	0.1	0.0	45	2-dodecanol	30.873	1607	0.7	0.1
13	undecane	14.216	1095	0.1	0.0	46	tetradecanal	31.224	1620	1.9	0.2
14	linalool	14.354	1099	1.0	0.0	47	cubenol	31.607	1635	0.7	0.1
15	nonanal	14.544	1105	0.6	0.0	48	n.i.	32.456	1666	0.6	0.0
16	cyclodecanol	14.958	1117	tr	-	49	n.i.**	33.144	1692	11.7	0.3
17	camphor	16.221	1154	0.1	0.0	50	pentadecanal	34.058	1725	0.2	0.0
18	*Z*-2-nonenal	16.593	1165	tr	-	51	heptadecane	35.740	1784	1.6	0.0
19	*n*-nonanol	16.965	1176	0.2	0.0	52	1-dodecanal	37.129	1837	0.7	0.0
20	4,*Z*-decenal	17.743	1199	0.3	0.0	53	dodecanal	37.266	1842	0.1	0.1
21	4,*E*-decenal	17.877	1203	0.8	0.0	54	n.i.	37.896	1867	0.3	0.0
22	decanal	18.372	1217	17.2	0.7	55	n.i.	38.205	1879	0.5	0.0
23	*E*-2-decenal	19.683	1256	0.8	0.0	56	*α*-humulene	38.370	1885	1.0	0.0
24	geraniol	19.870	1262	tr	-	57	*trans*-*β*-farnesene	38.738	1900	0.2	0.0
25	*E*-2-decenol	20.423	1278	14.2	0.1	58	1-dodecanol	38.918	1907	0.1	0.0
26	1-decanol	20.581	1283	3.0	0.0	59	n.i.	40.278	1963	0.2	0.0
27	2-*n*-octylfuran	21.149	1299	0.3	0.0	60	oleic acid	40.636	1978	0.9	0.0
28	undecanal	21.731	1316	2.1	0.0	61	phytol	43.812	2106	2.1	0.1
29	n.i.	23.000	1357	0.2	0.0	62	n.i.	44.886	2159	1.1	0.1
30	*E*-2-undecenal	23.666	1373	2.2	0.0	63	n.i.	45.716	2199	0.3	0.0
31	n.i.	24.696	1404	1.0	0.0	64	n.i.	46.174	2218	0.3	0.0
32	dodecanal	25.101	1418	4.6	0.2	65	*n*-octadecanol	51.519	2471	0.2	0.2
33	*γ*-elemene	25.666	1438	0.1	0.0						
Total/ Identified compounds		>99.8%/>82,6%									
Essential oil content		0.17 ml × 100g^{-1}									

** see: Fig. 2

DISCUSSION

The concentration and composition of coriander essential oil shows genetic, ontogenetic, and environmental variability (Bhuiyan et al. 2009; Ebrahimi et al. 2010; Orav et al. 2011). However, this variability within coriander cultivars is mainly caused by chemotype variations and not by other factors (Bhuiyan et al. 2009). The essential oil content in the coriander fruit is very different: from 0.5 to 2.5% (Mahendra and Bisht, 2011), and it increases as the fruit ripens (Msaada et al. 2007). Coriander leaves contain less oil than the fruit (Bhuiyan et al. 2009) and the concentration of volatile substances is determined by, among others, cultivation factors (Neffati and Marzouk, 2008; Telci et al. 2006a; b). The leaves of coriander grown in Bangladesh accumulated 0.1% of oil and that value was determined in fresh plant material (Bhuiyan et al. 2009). Coriander harvested in Tunisia had 0.12% of oil in air-dry leaf weight (Neffati and Marzouk, 2008). In the present study, the amount of essential oil in the coriander herb was on average 0.23 ml × $100g^{-1}$ and it was higher in the generative phase (0.29 ml × $100g^{-1}$) than in the vegetative phase (0.17 ml × $100g^{-1}$). These values are comparable to those determined in the material from plants growing in warm and dry climatic conditions. This can be explained by advantageous light and temperature conditions in the glasshouse during the growth of coriander plants. Essential oil synthesis in coriander, which has high light and temperature requirements, is more intense under optimal climatic conditions (Telci et al. 2006a). Besides, the cultivar 'Jantar', whose plants are analysed in this paper, has a high oil concentration in the fruit and high oil yield (Zheljazkov et al. 2008).

The composition of the studied essential oil indicates the predominant share of aliphatic aldehydes, which is consistent with the literature data (Potter and Fagerson, 1990; Neffati and Marzouk, 2008; Padmakuniari, 2008; Telci and Hisil, 2008; Chung et al. 2012). Bhuiyan et al. (2009) reported that coriander leaf oil contained 44 compounds containing 2-decenoic acid, *E*-11-tetradecenoic acid and capric acid as the major constituents. It should be noted here that the demonstrated immunotoxic effect of coriander volatile oil against the larvae of *Aedes aegypti* L., the carrier of yellow fever, dengue, tularemia and meningitis, may result directly from the presence of aliphatic aldehydes – the main components of the oil distilled from leaves and stems (Chung et al. 2012). Thus, the share of above-mentioned components of coriander herb oil should be regarded as significant from the medical point of view. Changes in the contents of the above-mentioned aldehydes in the examined coriander oil depended on the plant developmental stage at which the herb was harvested. The volatile oil distilled from the coriander fruit, containing linalool, as the main component, demonstrates antifungal activity, whereas oleoresin, rich in oleic and linolenic acid, is indicated as an alternative source of natural antioxidants (Singh et al. 2006). As the present study revealed, the above-mentioned components also occur, though in much smaller amounts, in the oil extracted from the coriander herb. The material from the first harvest was characterized by a higher percentage of undecanal, *E*-2-dodecanol and tetradecanal, but a smaller percentage of decanal, *E*-2-decenol, linalool and oleic acid as well as phytol, compared to the herb harvested on the second harvest date. Among the above-mentioned compounds, linalool, oleic acid and phytol should be emphasized, as they are components with a significant biological value (Figs 3, 4, 5); their contents were different in the oil of the herb from the first and second harvest. The above relationships should be connected with the fact that coriander, as an annual plant, after the first herb harvest at the flowering stage, aims at forming fruits and seeds. Thus, the herb collected from regrowing shoots is not typical for the vegetative stage, which is proven, for instance, by an increased concentration of linalool – a compound that is mainly characteristic of the oil distilled from the fruit (Telci et al. 2006b; Bhuiyan et al. 2009) and depends on the phase at which the herb is harvested (Telci et al. 2006a). The plant development phase during the harvest of coriander fruits is a significant factor affecting raw material yield as well as the quality of essential oil because of the changes during the fruit ripening period (Telci et al 2006a; Msaada et al. 2007). As shown by the studies conducted by other authors (Potter, 1996; Telci and Hisil, 2008) as well as by the present research, the quality of coriander herb also depends on the plant developmental stage during the harvest period.

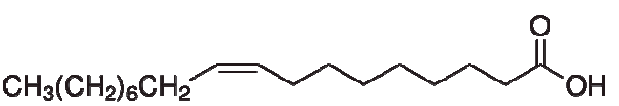

Fig. 3. Oleic acid – omega-9 fatty acid has been shown to slow the development of heart disease and to promote the production of antioxidants.

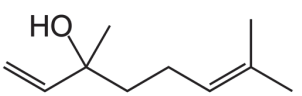

Fig. 4. Linalool – terpene alcohol, compound with anti-inflammatory, antimicrobial and sedative properties.

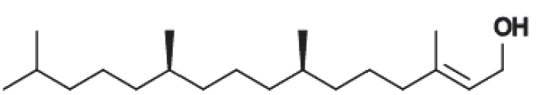

Fig. 5. Phytol – an acyclic diterpene alcohol used as a precursor for the manufacture of synthetic form of vitamins E and K_1.

To sum up, the obtained results indicate high variability of *Coriandrum sativum* L. herb as to the contents and chemical composition of its essential oil caused by the timing of harvest. Raw material harvested at different plant development stages has different compositions of volatile oil and this should be taken into account in producing oil for pharmaceutical purposes. The essential oil extracted from the coriander herb is marked by a significant percentage of aliphatic aldehydes, among which decanal, *E*-2-dodecanol and *E*-2-decenal predominate. These and the other aldehydes of coriander volatile oil should be considered to be important biologically active substances due to their possible toxic activity against tropical mosquitoes transmitting dangerous illnesses. The contents of most aliphatic aldehydes identified decreased with each subsequent herb harvest. The important components of coriander herb essential oil also include linalool, oleic acid and phytol – biologically active substances widely applied in the pharmaceutical industry. The percentage of the above-mentioned components varied with plant development: the herb collected at the initial stage of flowering had higher concentrations of oleic acid and phytol than the herb harvested from regrowing shoots. The latter had, however, higher concentrations of linalool, a compound characteristic of the oil distilled from the coriander fruit.

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Kompozycja olejku eterycznego ziela kolendry (*Coriandrum sativum* L.) w zależności od fazy rozwojowej rośliny

Streszczenie

Surowcem zielarskim *Coriandrum sativum* jest owoc, jako aromatyczną przyprawę wykorzystuje się również świeże ziele. Przedstawione badania dotyczyły oceny zawartości i składu chemicznego olejku kolendrowego, pozyskiwanego z ziela zbieranego w różnych fazach rozwoju rośliny. Kolendrę uprawiano w szklarni, zbiór ziela przeprowadzono na początku okresu kwitnienia oraz z odrastających pędów. Ekstrakcję olejku eterycznego z ziela przeprowadzono metodą hydrodestylacji, ocenę składu chemicznego metodą GC-MS. Badany surowiec zawierał 0,17–0,29 ml × $100g^{-1}$ olejku eterycznego, w zależności od fazy zbioru. Stwierdzono obecność 61 (faza generatywna) oraz 65 (faza wegetatywna) związków w badanym olejku kolendry. Olejek eteryczny z ziela kolendry zawierał w największej ilości aldehydy alifatyczne, z których największy udział miał dekanal, *E*-2-dodekanol i *E*-2-decenol. Zawartość większości oznaczonych aldehydów alifatycznych zmniejszała się wraz z kolejnym zbiorem ziela. Poza wymienionymi aldehydami alifatycznymi, w olejku eterycznym ekstrahowanym z ziela kolendry stwierdzono obecność linalolu, fitolu i kwasu oleinowego.