CONTRIBUTIONS TO THE CHEMICAL STUDY OF THE ESSENTIAL OIL ISOLATED FROM CORIANDER (OMAGIU CULTIVAR) FRUITS

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Abstract

We studied the nutritional space influence on the quality and quantity of the essential oil isolated from coriander fruits (Omagiu cultivar - O) cultivated in north-eastern Romania. The oil yield ranged between 1.4 and 1.8 mL/100 g dried fruits. Although the main components identified and quantified by gas-chromatography coupled mass spectrometry (GC-MS) were monoterpene alcohols (48.73-55.62%) and monoterpenes (28.40-36.49%), the quantity of linalool (40.59-51.20%) which should prevail in the volatile fraction was bellow 65.0%, the minimum limit required by European Pharmacopoeia, 6th ed. The most productive sample in volatile oil (11.37 kg/ha) was found to be O5, where the nutritional area was 50 cm x 15 cm, but in terms of quality, the most convenient results were obtained for O2, with a nutritional space of 25 cm x 15 cm.

Rezumat

În lucrarea de faţă am urmărit influenţa spaţiului de nutriţie asupra cantităţii şi calităţii uleiului volatil din fructele de coriandru soiul Omagiu (O), cultivat în nord-estul României. Conţinutul în ulei volatil, izolat prin hidrodistilare, s-a situat între 1,4 şi 1,8mL/100g produs vegetal uscat. Cu toate că grupele principale de componente identificate şi cuantificate semicantitativ prin GC-MS (gaz cromatografie cuplată cu spectometrie de masă) au fost monoterpenolii (48,73-55,62%), urmaţi de hidrocarburile monoterpenice (28,40-36,49%), linaloolul care ar trebui să predomine în fracţiunea volatilă se găseşte în cantităţi sub prevederi (40,59-51,20%, faţă de limita minimă de 65,0% cerută de Farmacopeea Europeană, ediţia a 6-a). Varianta cea mai productivă în ulei volatil (11,37kg/ha) s-a constatat a fi O5, la care spaţiul de nutriţie a fost de 50 cm x 15 cm, însă din punct de vedere calitativ, rezultatele cele mai convenabile s-au obţinut pentru varianta O2, la care acesta a fost de 25 cm x 15 cm.

Keywords: *Coriandrum sativum* L., essential oil, variability.

Introduction

Coriandrum sativum L. (*Apiaceae*) is a well known plant, originary from the Mediterranean region, intensively cultivated in Romania. Its fruits are used as a condiment and flavouring agent in the food industry, but also

in phyto- and aromatherapy for the treatment of gastrointestinal disorders such as dyspepsia, anorexia, flatulence or abdominal pains [1, 2, 3]. The coriander has stomachic and diuretic properties, while the volatile fraction is active in the topical treatment of abdominal aches, rheumatism and neuralgia; it is known to be very efficient as an antibacterial and antifungal agent [4, 5, 6].

Many parameters such as genetic and climatic factors, the maturity stage or agronomical practices can influence the yield and composition of the essential oil [7]. The plant spacing is an important factor that affects especially the productivity in organic cultures such as the ones included in our study.

In order to conclude on its pharmaceutical quality, we studied the effect of planting density on the yield and composition of the essential oil obtained from Omagiu coriander fruits.

Materials and Methods

Plant material

The coriander fruits were harvested during August 2009 from plants cultivated in the field of Agricultural Research and Development Centre, Secuieni – Neamț (Romania) (figure 1). The voucher specimens of samples were deposited at the Department of Pharmacognosy, Faculty of Pharmacy, University of Medicine and Pharmacy "Grigore T. Popa", Iași, Romania.

In order to investigate the influence of the nutritional space on the yield of coriander fruits crop and essential oil content, an experiment with different plant densities was developed. Therefore, the plants were cultivated as follows:

- distances between the plant rows: a=25 cm; b=50 cm;
- distances into the row: a= 0 cm; b=15 cm; c=20 cm.

The six experimental samples, encoded from O1 to O6 (table I), had a vegetation period of 111 days.

Figure 1

Coriandrum sativum L., Omagiu cultivar:

A- sprouting; B- rosette of basal leaves; C- flowering.

Essential oil isolation

The volatile fraction was isolated by hydrodistillation of powdered dried fruits in a Clevenger type apparatus (3 h). The essential oil samples were subsequently analyzed by gas chromatography coupled with mass spectroscopy GC-MS.

Analysis of essential oils

The essential oils have been characterized using gas chromatography coupled with mass spectroscopy GC-MS. GC-MS analyses were carried out on an Agilent type 7890A gas chromatograph, equipped with an Agilent 5975C mass spectrometer and a DB-5MS capillary column (30 m x 0.25 mm, 0.25 μm film thickness). The oven temperature was increased with a rate of 10°C/min from 35°C to 280°C (where kept isothermally for 7.5 min). The other characteristics of the analysis program were: inlet temperature: 250°C; split ratio: 50:1; carrier gas: helium (flow rate: 1 mL/min); injected volume: 0.2 μL of essential oil.

Volatile compounds identification

The compounds were identified by matching their recorded mass spectra with those stored in the Wiley mass spectral library.

Results and Discussion

The yield of the essential oil varied between 1.4 and 1.8 mL/100g dried fruits. The efficiency of the extraction and the oil productivity were determined and the results are listed in table I.

Table I

Fruits and essential oil yields for coriander (Omagiu cultivar)

Sample	Nutritional space* (cmxcm)	Fruits yield (kg/ha)	Essential oil (mL/100g dried fruits)	Essential oil yield (kg/ha)
O1	25 x 0	859.3	1.4	10.47
O2	25 x15	825.6	1.4	10.06
O3	25 x 20	808.4	1.6	11.25
O4	50 x 0	733.2	1.6	10.21
O5	50 x 15	726.1	1.8	11.37
O6	50 x 20	706.8	1.8	11.07

*Nutritional space=distance between plant rows x distance into the row

Our data showed that Omagiu coriander belongs to the sorts with a high productivity of fruits and essential oil. For the plants with a reduced nutritional space (O1-O3), the fruits yield was high (which is important for food industry), but the oil quantity was relatively low (1.4-1.6 mL/100g dried fruits). Consequently to the increase of the nutritional space (O4-O6),

the plants synthesized a higher quantity of essential oil, but they also had a lower fruits productivity. It remains to be seen if we can correlate these results with the pharmaceutical quality of the volatile fraction.

GC-MS analysis was carried out in order to evaluate the quality of the volatile fractions isolated from the samples of coriander fruits; the results of the identification and semiquantitative analysis of the compounds are presented in table II.

Table II

Chemical composition of the essential oils obtained from the Omagiu coriander fruits

Retention Time (min.)	Compound	Composition (%)					
		O1	O2	O3	O4	O5	O6
5.353	N-heptanal	-	0.02	0.01	0.01	0.02	0.02
5.860	α-thujene	0.15	0.14	0.23	0.31	0.35	0.33
6.011	**α-pinene**	**7.78**	**7.01**	**9.46**	**10.21**	**10.01**	**10.48**
6.281	camphene	1.20	1.17	1.44	2.00	2.01	2.05
6.626	sabinene	0.82	0.79	0.97	1.16	1.24	1.20
6.734	β-pinene	1.17	1.31	1.57	1.81	1.83	1.80
6.842	myrcene	1.59	1.78	1.97	2.19	2.30	2.20
7.014	decane	-	-	-	-	0.01	-
7.068	N-octanal	0.01	-	0.01	0.01	0.01	-
7.165	l-phellandrene	0.06	0.07	0.08	0.09	0.12	0.09
7.338	α-terpinene	0.16	0.20	0.19	0.23	0.27	0.22
7.468	**p-cymene**	**3.14**	**3.58**	**3.90**	**3.97**	**3.46**	**3.94**
7.543	**L-limonene**	**3.58**	**3.78**	**4.18**	**4.40**	**4.46**	**4.38**
7.770	trans-β-cymene	0.09	0.15	0.16	0.16	0.20	0.15
8.018	**γ-terpinene**	**11.58**	**10.72**	**12.02**	**12.07**	**11.14**	**11.91**
8.212	cis-sabinene hydrate	-	-	0.27	0.26	0.31	0.29
8.428	α-terpinolene	1.09	1.20	1.23	1.29	1.37	1.28
8.795	**L-linalool**	**51.20**	**51.04**	**46.19**	**43.78**	**40.59**	**42.66**
9.075	cis-ocimene	-	0.05	-	0.03	0.04	0.03
9.226	α-fenchene	-	-	0.06	-	0.04	0.02
9.248	Δ-3-carene	-	0.03	-	-	-	0.06
9.388	p-menth-2-en-1-ol	-	0.06	-	-	0.05	0.05
9.518	**camphor**	**6.57**	**6.97**	**5.88**	**5.64**	**6.11**	**5.52**
9.679	β-pinanone	0.12	-	-	-	-	-
9.701	pinocarvone	-	0.13	0.12	0.10	-	0.11
9.852	L-borneol	0.15	0.17	0.14	0.14	0.16	0.14
9.960	terpinene-4-ol	0.61	0.70	0.54	0.50	0.59	0.52
10.154	**α-terpineol**	**0.70**	**0.77**	**0.73**	**0.64**	**-**	**0.67**
10.251	N-decanal	0.07	0.07	0.08	0.07	0.10	0.07
10.359	(+)-verbenone	0.04	0.04	-	-	-	-
10.413	γ-isogeraniol	-	-	-	-	-	0.05
10.532	β-citronellol	0.20	0.23	0.24	0.26	0.35	0.27

Retention Time (min.)	Compound	Composition (%)					
		O1	O2	O3	O4	O5	O6
10.748	neral	0.08	0.09	0.10	0.08	0.12	0.09
10.899	**geraniol**	**2.70**	**2.65**	**3.30**	**3.41**	**4.51**	**3.78**
11.039	piperitone	-	0.07	-	-	-	-
11.168	geranial	0.13	0.14	0.15	0.14	0.18	0.15
11.481	trans-anethole	0.41	0.50	0.43	0.32	0.47	0.26
11.611	carvacrol	0.04	0.04	0.05	0.04	0.06	0.04
11.730	dodecanal	0.03	-	-	0.05	-	-
11.740	undecanal	-	0.03	0.05	-	0.06	0.05
11.794	3-methoxy-acetophenone	-	-	-	-	0.02	-
11.891	methyl geranate	-	-	0.03	0.03	0.04	0.03
11.999	myrtenyl acetate	-	0.06	0.06	-	0.09	0.07
12.388	neryl propionate	-	-	0.05	-	-	-
12.431	geranyl butyrate	0.04	-	-	-	-	0.06
12.711	neryl acetate	-	0.05	-	-	4.85	-
12.722	lavandulyl acetate	3.38	3.27	3.21	3.64	-	-
12.744	**geranyl acetate**	**-**	**-**	**-**	**-**	**-**	**4.07**
12.852	myrtanyl acetate	-	-	0.06	-	-	-
12.927	β-bourbonene	0.07	-	-	-	-	-
13.175	pentylcyclopentane	0.03	-	-	-	-	-
13.197	dodecanal	-	0.02	0.04	-	0.06	-
13.305	junipene	-	-	0.01	0.01	0.01	0.01
13.434	β-caryophyllene	0.14	0.17	0.22	0.21	0.30	0.22
13.941	α-humulene	-	0.02	0.01	-	-	0.02
13.995	3-dodecen-1-al	0.07	-	0.15	0.15	0.18	0.20
14.017	2-dodecen-1-al	-	0.05	-	-	-	-
14.114	farnesol	-	0.01	-	-	-	-
14.470	bicyclogermacrene	0.03	0.03	0.04	0.04	0.06	0.06
14.546	tetradecanal	-	-	-	-	0.03	-
15.182	trans-nerolidol	-	-	-	0.01	0.01	-
15.603	spathulenol	0.02	-	-	-	0.01	-
16.002	tridecanal	-	-	-	-	0.03	0.04
16.121	α-amorphene	-	-	-	-	0.01	-
16.908	trans-2-tridecenal	-	-	-	-	0.13	-
17.405	hexatriacontane	-	-	-	-	0.02	-
	Others	0.75	0.62	0.37	0.54	1.61	0.34

RT= retention time

The chemical composition of the essential oils showed a great variability. The greatest number of compounds was identified for O5 sample (48) and the lowest number for O1 sample (37).

The pharmaceutical quality of the volatile fractions isolated from the six samples of coriander fruits was compared with the requirements of

Coriandri aetheroleum monograph in European Pharmacopoeia, 6th edition [8]. It was found that the six essential oils presented significant variations, as shown in table III.

Table III

Pharmaceutical quality of Omagiu coriander fruits essential oil

Compound	European Pharmacopoeia 6th edition requirements	Composition (%)					
		O1	O2	O3	O4	O5	O6
α-pinene	**3.0-7.0 %**	7.78	7.01	9.46	10.21	10.01	10.48
limonene	**1.5-5.0 %**	3.58	3.78	4.18	4.40	4.46	4.38
γ-terpinene	**1.5-8.0 %**	11.58	10.72	12.02	12.07	11.14	11.91
p-cymene	**0.5-4.0 %**	3.14	3.58	3.90	3.97	3.46	3.94
camphor	**3.0-6.0 %**	6.57	6.97	5.88	5.64	6.11	5.52
L-linalool	**65.0-78.0 %**	51.20	51.04	46.19	43.78	40.59	42.66
α-terpineol	**0.1-1.5 %**	0.70	0.77	0.73	0.64	tr	0.67
geranyl acetate	**0.5-4.0 %**	tr	tr	tr	tr	tr	4.07
t-geraniol	**0.5-3.0 %**	2.70	2.65	3.30	3.41	4.51	3.78

tr – trace amounts (< 0.01 %)

The analysis revealed that our samples do not meet the requirements mentioned in the European Pharmacopoeia, 6th ed. It was found 40 to 51% linalool content in comparison to 65%, the minimum required value, and geranyl acetate was identified only in the O6 sample. Also, α-pinene, γ-terpinene, camphor and, in a lower extent, geraniol were in higher amounts in comparison to limits specified in *Coriandri aetheroleum* monograph.

The findings led to the conclusion that the essential oil isolated from the Omagiu cultivar coriander fruits (2009) does not meet the pharmaceutical qualities required by the European Pharmacopoeia, 6th ed.

The main classes of volatile constituents of each sample are shown in table IV.

Table IV

The chemical class characterization of *Coriandri aetheroleum* (Omagiu cultivar)

Volatile compounds classes (%)	Sample					
	O1	O2	O3	O4	O5	O6
Monoterpene hydrocarbons	29.27	28.40	33.83	36.21	35.69	36.49
Monoterpene alcohols	55.56	55.62	51.14	48.73	46.25	48.14
Monoterpene ketones	6.73	7.21	6.00	5.74	6.11	5.63
Monoterpene aldehydes	0.21	0.23	0.25	0.22	0.30	0.24
Monoterpene esters	3.42	3.38	3.41	3.67	4.98	4.23
Total Monoterpenes	**95.19**	**94.84**	**94.63**	**94.57**	**93.33**	**94.73**
Sesquiterpenes	0.26	0.23	0.28	0.27	0.40	0.31
Aliphatic compounds	0.21	0.19	0.34	0.29	0.65	0.38
Aromatic compounds	3.59	4.12	4.38	4.33	4.01	4.24

In all samples it was noticed a prevalence of monoterpene alcohols (48.73-55.62%), followed by monoterpene hydrocarbons (28.40-36.49%). Monoterpene ketones (5.63-7.21%), aromatic compounds (3.59-4.38%) and monoterpene esters (3.38-4.98%) were in lower amounts. Regarding the two major volatile compounds classes, monoterpene alcohols and monoterpene hydrocarbons, we note that their total amounts vary between 81.94% (O5) and 84.97 % (O3); these results justify the topical use of the essential oil as anti-inflammatory, analgesic and immunostimulating agent in the treatment of rheumatism, but also as an antibacterial and antiviral agent, although the content in linalool does not meet the pharmacopoeial requirements.

Conclusions

For high yields of coriander fruits and essential oils, the plants require an adequate nutritional space that should be provided by the agronomical practices. Regarding the oil yield, the best results were determined for O3 and O5 samples, but the volatile fraction with the most suitable chemical composition seems to be the one obtained from O2 sample with a nutritional space of 25 cm x 15 cm.

The O2 sample appears to be the best choice for the food industry concerning the fruits yield, but also for the aromatherapy due to its essential oil qualities, close to those required by the European Pharmacopoeia, 6th ed.

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