



Flavoring Compounds of Essential Oil Isolated from Agriculture Waste of Coriander (*Coriandrum sativum*) Plant

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Abstract

Our program to study the optimization of agricultural residues after harvest. Dry waste of coriander (*Coriandrum sativum*) plant was collected for recycling in order to obtain the volatile oils. The purpose is, to benefit from the waste receiving more of coriander essential oil, in addition, reducing the environmental pollution resulting from the methods used to disposal of agricultural waste. The experiment was carried out at Asuit region -Egypt during two successive seasons 2015-2016 to study the yield and chemical constituents of the essential oils produced from the green herb, the dry seeds and the waste of coriander (*C. sativum*) plant. The chemical constituents of coriander oils were analyzed by means of GC/MS. The results of the analysis of coriander essential oil showed that, thirty five constituents representing 92.49% of the oil herb were identified, against twenty nine (84.6%) and seventeen (91.84%) for seed and waste oil, respectively. The main constituent was linalool in green herb and dry seeds oils, while trans-anethole was found as the major oil constituents of waste oil followed by linalool compound. The high percentage of linalool was found in the seeds oil (59.6%) followed by the waste (29.29%) and green herb oil (18.34%) respectively. Clear variation in the oil constituents of the three oils due to kind of plant materials as well as the growth stages was observed. So some compounds were found in all samples of the three oils with only differences in the concentrations of them. These compounds were α -pinene, camphene, β -pinene, p-cymene and limonene. Also some compounds were found in some oils and absent in the others. Cis-beta-ocimene, terpinene gamma, camphor, α -terpineol, β -caryophyllene and decanal were found in the green herb and seeds oils, while estragole and trans anethole were found only in green herb and waste oil. The composition of the waste oil was close to that of the green herb and dry seed oils, which means that waste oil resembles the herb and seed oil to a great extent.

1. Introduction

Through our program to study the optimization of the residues after harvesting plants of Umbelliferae family. the wastes of Coriander (*Coriandrum sativum*) plants after harvest were collected where it were recycled in order to obtain volatile oils. The purpose is to benefit from the waste receiving more of Coriander essential oil, in addition to reducing the environmental pollution resulting from the methods used to dispose the agricultural wastes Thousands of years ago essential oils have been used in different cultures for medical and health purposes. Uses of essential oil range from, personal beauty care, household cleaning products, aromatherapy and natural medicine treatments [1-4]. The benefits of essential oils come from their antimicrobial, anti-inflammatory and antioxidant properties. These curative oils are fast growing in popularity because they work as natural medicine not including any side effects. In addition, it is an important economic resource for the national economy of Egypt.

Coriander (*C. sativum* L.) an annual of the Umbelliferae family, is one of important medicinal, flavor plants. This plant comes from the Mediterranean region and it is cultivated all over the worlds [5-7]. The hydro-distillation of coriander aerial parts gave volatile oils at vegetative, full flowering, green immature fruit and dry

fruit (mature) with a yield of 0.14%, 0.23%, 0.37% and 0.31% (w/w), respectively [8]. The chemical class characters of coriander essential oils from different fruit samples have been represented by Sriti *et al.* [7]: Seeds of *C. sativum* yielded 0.8%, yellow oil, with a pleasing aroma, containing oxygenated monoterpenes (80.47%), monoterpene hydrocarbon (6.45%) [9]. Essential oils can differ among different parts of the same plant, such as, essential oils isolated from the *C. sativum* seed has different content from the essential oil of *C. sativum* flower as well as cilantro (immature leaves) [10]. The coriander essential oil from fully ripe and dried seeds is a colorless or pale yellow liquid with a characteristic odor and soft, sweet, warm and aromatic flavor, and linalool is its major compounds [11]. While aliphatic aldehydes (mainly C10–C16 aldehydes), are the main constituents of the volatile oil from the fresh herb [12], Many investigators reported that, the essential oil constituents of the coriander fruit and herb are completely different [8, 10,13,14].

2. Experimental details

2.1 Plant materials

The experiment was conducted at Asuit region –Egypt (380 km south of Cairo) during two successive seasons 2014-2015 to study the yield and chemical constituents of the essential oils produced from the green plants, the dry seeds and the waste of coriander plant. The seeds were sown in the 15th of November in the two seasons. All agricultural practices of cultivation were done. Samples of green herb were collected during the formation of the green coriander seeds in March of each season. The dry seeds samples of coriander (*C. sativum*) plant were taken in May (mature seeds stage) and the dry waste was taken after harvest.

2.2 Plant extraction

The essential oils of the dry plant materials were extracted by hydro-distillation for 3 hr. (Clevenger, 1928)[15]. The essential oils were dehydrated over anhydrous sodium sulfate and subjected to GC/MS analysis.

2.3 Gas chromatography

GC analysis was performed using a Shimadzu GC- 9A gas chromatograph equipped with a DB5 fused silica column (30 m x 0.25 mm i.d., film thickness 0.25 µm). Oven temperature was held at 40°C for 5 min and then programmed until 250°C at a rate of 4°C/min. Injector and detector (FID) temperature were 260°C; helium was used as a carrier gas with a linear velocity of 32 cm/s.

2.4. Gas chromatography- mass spectrometry

GC-MS analyses were carried out on a Varian 3400 system equipped with a DB-5 fused silica column (30 m x 0.25 mm i.d.); Oven temperature was 40 to 240°C at a rate of 4°C/min, transfer line temperature 260°C, injector temperature 250°C, carrier gas helium with a linear velocity of 31.5 cm/s, split ratio 1/60, flow rate 1.1 ml/ min, Ionization energy 70 eV; scan time 1 s ; mass range 40-350 amu.

2-5. Qualitative and quantitative analysis of essential oil

Identifications were made by library searches (Adams, 1995) [16] combining MS and retention data of authentic compounds by comparison of their GC retention indices (RI) with those of the literature or with those of standards available in our laboratories. The retention indices were determined in relation to a homologous series of n-alkanes (C8–C22) under the same operating conditions. Further identification was made by comparison of their mass spectra with those stored in NIST 98 and Wiley5 Libraries or with mass spectra from literature. Component relative concentrations were calculated based on GC peak areas without using correction factors.

3. Results and discussion

3.1 Oil percentage

Table 1 showed that the dry waste contained the highest percentage of oil (0.89%) followed by the dry seeds (0.31%) and green herb (0.27%) respectively.

Table 1. Oil percentage of coriander (*Coriandrum sativum*) herb, seed and waste oil

Stage	%
Herb	0.27
Seeds	0.31
Waste	0.89

3.2 Coriander (*C. sativum*) herb oil

Data based on GC analysis were listed in Table 2 recorded that, thirty five compounds, represent 92.49% of the Egyptian coriander herb essential oil were identified.

Table 2. Oil constituents of coriander (*C. sativum*) herb essential oils

Peak No	Compound	KI	%
<i>(MH) monoterpene hydrocarbons</i>			
1	α -pinene;	939	3.23
2	Camphene	953	0.51
3	β -Pinene	980	0.56
4	Myrcene	991	0.45
5	α -Phellandrene	1005	1.47
6	Para cymene	1026	4.08
7	Limonene	1031	3.3
8	Cis-beta-ocimene	1040	0.31
9	Terpinene gamma	1061	2.21
	Total		16.12
<i>(OM) oxygenated monoterpenes</i>			
1	Cis-linalool oxide	1074	0.36
2	α -Terpinolene	1088	1.06
3	Linalool	1098	18.34
4	α -Thujone	1102	2.29
5	Camphor	1143	2.7
6	Terpinen-4-ol	1177	0.43
7	α -Terpineol	1189	1.22
8	Estragole	1195	7.71
9	Verbenone	1204	2.75
10	Geraniol	1255	1.9
11	Trans-anethole	1283	3.33
12	Carvacrol	1298	4.29
	Total		46.38
<i>(SH) sesquiterpenes hydrocarbons</i>			
1	Longifolene	1402	1.74
2	Beta-caryophyllene	1418	0.34
	Total		2.08
<i>(OS) oxygenated sesquiterpenes</i>			
1	Elemol	1549	1.19
<i>(VC) various compounds</i>			
1	Decane	999	1.19
2	1-octanol	1070	0.46
3	Ethyl heptanoate	1095	1.1
4	(E)-2-nonenal	1155	5.3
5	Undecenal	1156	0.45
6	1-nonanol	1171	0.53
7	1-decanol	1204	1.43
8	Decanal	1204	2.85
9	(E)-2-decenal	1261	5.1
10	Geranyl acetate	1383	3.47
11	Dodecanal	1407	0.89
12	Terpenyl acetate	1352	1.13
13	Eugenyl acetate	1524	1.12
14	Trans-nerolidol	1564	0.53
15	Tetra decanal.	1611	0.52
16	Phytol	1949	0.65
	Total		26.72

The main components of the essential oil are (E)-2-Decenal > (5.1%), linalool (18.34%), (E)-2-nonenal (5.3%), and α -thujone (2.29%). Coriander herb oil classified into five groups which are MH (monoterpene hydrocarbons), OM (oxygenated monoterpenes), SH (sesquiterpenes hydrocarbons), OS (oxygenated sesquiterpenes) and VC (various compounds). Coriander oil is characterized by its higher content of OM compounds which recorded 46.38 % followed by VC (26.72%), MH (16.12 %), SH (2.08%) and (OS) group (1.19%) respectively. MH group amounted to 16.12 %. Para cymene (4.08%) was found as the major constituents of MH group. Significant amounts of α -pinene (3.23%), limonene (3.3%), terpinene gamma (2.21%), α -phellandrene (1.47%), β -pinene (0.56%), camphene (0.51%), myrcene (0.45%) and cis-beta-ocimene (0.31%) were detected in MH group (OM) group included, linalool (18.34%), estragole (7.71%), carvacrol (4.29%), trans-anethole (3.33%), verbenone (2.75%), camphor (2.7%), α -thujone (2.29%), Geraniol (1.9%), α -Terpineol (1.22%), α -terpinolene (1.06%), terpinen-4-ol (0.43%) and cis-linalool oxide (0.361%) (E)-2-nonenal (5.3%), as found as the main constituents of VC group followed E-2-decenal (5.1%), geranyl acetate (3.47%), decanol (1.43%), decane (1.19%), terpenyl acetate (1.13%), eugenyl acetate (1.12%), Ethyl heptanoate (1.1%), 1-nonanol (0.53%), trans-nerolidol (0.53%), tetra decanal (0.52%) -1 octanol (0.46%), undecenal (0.45%), decanal (2.85%), dodecanal (0.89%) and phytol (0.65%). Longifolene was found as the main constituent of (SH) group which recorded 1.74% while β -caryophyllene was found in a low concentration (0.34%) in the same group. (OS) group contained only elemol (1.19%) compound. These results were in agreement with those obtained by Renata Nurzyńska-Wierdak (2013) [17]

coriander seed essential oil:

The main constituents of coriander dry seed essential oil were linalool (59.6%), ethyl hexanoic acid <2> (4.92%), sabinene hydrate trans (4.36%) and α -thujone (3.32%). (OM) was found as the major group in the dry coriander seed oil. The total percentage of OM in this respect recorded (66.72%) against 8.6, 2.86, 0.7 and 5.9% for (MH), (SH), (OS) and (VC) group, respectively.

Similar results have been reported by many investigators [7, 8, 10, 17], which showed that the main compound in coriander seed essential oil was linalool. Among the other constituents of coriander seed oil identified in this work are, α -pinene (2.28%) and β -caryophyllene (2.51%). Some compounds that have concentration of less than 1% have been identified refer to table 3.

Coriander waste essential oil:

Seventeen constituents representing 91.84% of the Egyptian waste essential oil of coriander were identified. Anethole <trans-> was found as the main compound of coriander waste oil which recorded 29.29% followed by, linalool (20.06%), butanoic acid, 2-methyl-, 2-methoxy-4-(2-propenyl)phenyl ester (14.17%), estragole (10.25%) longifolene (6.82%), and carvacrol (5.1%)

Essential oil of coriander waste consists of five chemical groups. These groups were (MH), (OM), (SH) (OS) and (VC). Result in Table 4 revealed the presence of five components of (OM) group which recorded 68.42 % , against five, three, one and three compounds of (MH) (2.64%), (SH) (4.1%) (OS) (1.05) and (VC) (15.72%) group, respectively.

Comparing the oil constituents of the green herb, dry seeds and dry waste, it was found that, the main constituent was linalool in green herb and dry seeds oils, while trans-anethole was found as the major oil constituents of waste oil followed by linalool compound. The high percentage of linalool was found in the seeds oil followed by the waste and green herb oil respectively. At the same time, there is a clear variation in the oil constituents of the three oils due to kind of plant materials as well as the growth stage. So some compounds were found in all samples of the three oils extracted from green herb, dry seeds and waste of coriander plants, with only differences in the concentrations of them. These compounds were α -pinene. Camphene, β -pinene, p-cymene and limonene. Also it was observed that, some compounds were found in some oils and absent in the others. Cis-beta-ocimene, terpinene gamma, camphor, α -terpineol, β -caryophyllene and decanal were found in the green herb and seeds oils. While estragole and trans anethole were found only in the green herb and waste oil. Also some compounds have emerged only in one type of oil and did not appear in the other oil (Tables 3 and 4).

In all cases the essential oils of coriander isolated from green herb, dry seeds or waste characterized by high amount of OM group. It ranged from 46.38 % in herb oil to 68.42% in waste oil, while it was 66.72 % in the seeds oil. The composition of the waste oil was close to that of the green herb and dry seed oils. This was also clear in the percentage of the main group (OM) which means that waste oil resemble the herb and seed oil to a great extent.

Table 3. Oil constituents of coriander (*C. sativum*) seeds essential oils

Peak No	Compounds	KI	%
<i>MH (monoterpene hydrocarbons)</i>			
1	α -pinene	939	2.28
2	Camphene	953	1.3
3	β -pinene	980	0.06
4	α -thujene	995	3.32
5	3-carene	1011	0.55
6	P-cymene	1026	0.17
7	Limonene	1031	0.11
8	(Z)- β -ocimene	1041	0.66
9	γ -terpinene	1061	0.15
	Total		8.6
<i>(OM) Oxygenated monoterpene</i>			
1	Sabinene hydrate trans	1097	4.36
2	Tetrahydrolinalool	1098	0.26
3	Linalool	1098	59.6
4	Camphor	1151	0.25
5	Dihydro terpineol (trans-Alpha)	1161	0.03
6	Borneol	1168	0.34
7	α -terpineol	1189	0.04
8	Nerol	1228	0.8
9	Citral A	1275	0.27
10	Thymol	1290	0.58
11	Carvacrol	1301	0.19
	Total		66.72
<i>(SH) sesquiterpenes hydrocarbons</i>			
1	Elemene <Delta->	1337	0.05
2	Caryophyllene < beta->	1418	2.51
3	Germacrene B	1556	0.3
	Total		2.86
<i>(OS) oxygenated sesquiterpenes</i>			
1	Dendrolasin	1574	0.19
2	Caryophyllene-oxide	1589	0.08
3	Elemenone <cic-beta->	1591	0.43
	Total		0.7
<i>(VC) various compounds</i>			
1	Ethyl hexanoic acid <2->	1129	4.92
2	Dodecane	1199	0.98
	Total		5.9

Table 4. Oil constituents of coriander (*C. sativum*) waste essential oils

Peak No	Compound waste	KI	%
<i>(MH)monoterpene hydrocarbons</i>			
1	α -pinene;	939	0.53
2	Camphene	953	0.31
3	β -Pinene	980	0.16
4	Para cymene	1026	0.86
5	Limonene	1031	0.78
	Total		2.64
<i>(OM)oxygenated monoterpenes</i>			
1	Linalool	1098	20.06
2	Estragole	1195	10.25

3	Trans-anethole	1283	29.29
4	Carvacrol	1298	5.1
5	Longifolene	1402	6.82
Total			68.42
<i>(SH) sesquiterpenes hydrocarbons</i>			
1	Germacrene-D	1480	2.31
2	Zingiberene	1495	1.05
3	β -Bisabolene	1509	0.65
Total			4.01
<i>(OS) oxygenated sesquiterpenes</i>			
1	Spathulenol	1576	1.05
<i>various compounds</i>			
1	Decanal	1204	0.7
2	Geranyl acetate	1383	0.85
3	Butanoic acid, 2-methyl-, 2-methoxy-4-(2-propenyl)phenyl ester	2149	14.17
Total			15.72
			91.84

Conclusion

In the field of utilization of agricultural waste and conservation of the environment, this research is interested in recycling agricultural waste of coriander produced after the harvest. Comparing the essential oils extracted from green herb and dry seeds with the oil extracted from the waste, encouraging results to take advantage of waste as a new source of oil. In most cases there was a great similarity between constituents in oil isolated from the remains of dry herb after harvest with coriander oil extracted either from dry seeds or from green herb at immature green seeds formation stage. The difference was only in the proportions of constituents with some additional new compounds. Linalool was found as the major constituents in most samples of coriander plant. Taking in consideration that the yield of coriander waste per unite cultivated area are greatly higher than that of dry seeds, it is obvious that the extraction of essential oil from wastes is of great economic benefit especially that the content of dry seed and dry waste oils are very close to each other. Therefore, we strongly recommend the recycling of the leaves of coriander aromatic plant by re-extracting the volatile oil from the plant waste after the harvest in order to reduce pollution and increase the economics of the crop.

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