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Evaluation of essential oil isolated from dry coriander seeds and recycling of the plant waste under different storage conditions

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Abstract

Background: This study focused on the comparison of the essential oil percentage and constituents obtained from the dry seeds and the waste of coriander plant in order to reach the best ways to store the essential oil.

Results: The chemical composition of coriander oil was analyzed by gas chromatography–mass spectrometry (GC–MS). The analysis of coriander essential oil showed that linalool was the main constituent of dry seed oil under all conditions. It recorded 59.6, 59.28 and 47.69% of the treatments of the oil at zero time, stored in cool conditions and stored at room temperature, respectively. Concerning oil constituents of coriander waste (the remained herb after collecting seeds), the results showed that *trans*-anethole was the major oil constituent, followed by linalool compound. The quality of the stored oil in the refrigerator after harvest was better than the stored oil from seeds or waste under room temperature.

Conclusion: The changes have been observed in the chemical composition of coriander oil extracted from seed and waste subjected to different storage conditions. The waste of coriander can be considered a new source of essential oil.

Keywords: *Coriandrum sativum*, Coriander seeds, Coriander waste, Storage, Essential oils, Gas chromatography

Background

For thousands of years, essential oils have been used in different cultures for medical and health purposes. Uses of essential oil ranging from personal beauty care, household cleaning products, to aromatherapy treatments and natural medicine (Ryman 1984; Reeds 2000; Salma et al. 2018). Essential oil benefits come from their antimicrobial, antioxidant properties and anti-inflammatory. These therapeutic oils are growing rapidly in popularity because it works natural medicine and does not include any side effects. Coriander seeds are a good source of secondary plant metabolites such as polyphenols, especially phenolic acids and flavonoids (Zeković et al. 2014). It has industrial applications in pharmaceutical applications and tobacco where it is used to counteract unpleasant odors. Coriander seeds contain Δ6 palmitate desaturase enzyme which enables coriander seeds oil to accumulate over 85% of the industrially useful fatty acid and petroselinic acid (Jaworski and Cahoon 2003). Also, *Coriandrum sativum* is considered an important economic resource for the Egyptian national economy.

Coriander (*C. sativum* L.), an annual of the Umbelliferae family, is one of the important spice plants. It comes from the Mediterranean region and is grown all over the world (Mahendra and Bisht 2011; Sriti et al. 2011; Duarte et al. 2012). Seeds of *C. sativum* recorded 0.8%, yellow oil, with a pleasurable aroma, which contains oxygenated monoterpenes (80.47%) and monoterpene hydrocarbon (6.45%) (Pande et al. 2010). The coriander essential oil is colorless with a characteristic odor and soft, sweet, warm and aromatic flavor, and linalool is its major constituent (Msaada et al. 2007).

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The effect of storage conditions on the quality of the essential oils during storage has been investigated by a number of investigators. It has been reported that the values of essential oil decreased during the storage time in *Majorana hortensis* (Arabhosseini et al. 2007). Temperature, light, humidity and air are the most significant factors that affecting the volatile oils of the spices and herbs (Njoroge et al. 1996). There are a few reports about the effect of storage on quality of coriander seeds essential oil, for these reasons, storage is a significant factor to identify the most suitable way to get a high-quality product. This study focused on the comparison of the best way to store *C. sativum* essential oil, whether to store it in the form of essential oils or keep it inside the dry seeds until extracted by hydro-distillation. Also, to increase the utilization of post-harvest agricultural waste and reduce the environmental pollution caused by the burning of plant waste in the field, the main objective of the present investigation was to study the effect of different storage conditions on the essential oil extracted from the waste of coriander plant and compared with coriander seed oil under the same conditions.

Methods

Plant materials and isolation of essential oil

The experiment was conducted in Assiut region—Egypt (380 km south of Cairo) during the two successive agricultural seasons in the period of 2017–2019. Identification of *C. sativum* species was achieved by the staff of the Medicinal and Aromatic Plants Research Department at the National Research Centre. Voucher specimens are deposited in the herbarium of NRC, Cairo, Egypt.

The aim of this experiment was to study the percentage and chemical constituents of the essential oils produced from the dry seeds and waste of the coriander plant. Dried seeds and waste samples are divided into two groups. The essential oil of the first group was extracted by hydro-distillation using a Clevenger-type apparatus for 3 h and analysis after isolation directly (zero time treatment), the same oil placed in tightly closed dark vials, then stored in the refrigerator at 4–5 °C (cool conditions treatment) and analyzed after 1 year. And the second group (dry seeds and waste materials) packed in cartoon bags and stored in a dry place (at room-temperature treatment). After 1 year of storage, the coriander oil was extracted from the dried seeds and waste to identify its chemical components. The treatments were as follows:

T1 = Extracted and analyzed the essential oil from dry seeds at mature seed stage directly after harvest (at zero time).

T2 = Analyzed the essential oil, which extracted from dry seeds directly after harvest and stored in the refrigerator at 5 °C (cool conditions for 1 year).

T3 = Extracted and analyzed the essential oil from dry seeds which stored at room temperature for 1 year.

T4 = Extracted and analyzed the essential oil from dry waste immediately after harvest.

T5 = Analyzed the essential oil, which extracted from dry waste immediately after harvest and stored in the refrigerator at 5 °C (cool conditions for 1 year).

T6 = Extracted and analyzed the oil from dry waste which stored at room temperature for 1 year.

Plant extraction

The essential oils of the dry plant materials for all treatments were extracted by hydro-distillation for 3 h (Clevenger 1928). The essential oils were dehydrated over anhydrous sodium sulfate and subjected to GC–MS analysis under the same conditions. The laboratory investigations were conducted in the Medicinal and Aromatic Plants Researches Department, National Research Centre, Cairo, Egypt.

Gas chromatography (GC)

GC analysis was performed using a Shimadzu GC-9A gas chromatograph equipped with a DB-5 fused silica column (30 m × 0.25 mm i.d., film thickness 0.25 μm). The 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.

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 × 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.

Qualitative and quantitative analysis of essential oil

Identifications were made by library searches (Adams 1995), 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 the literature. Component relative concentrations were calculated based on GC peak areas without using correction factors.

Results

Oil percentage

In all cases, coriander wastes gave a high percentage of essential oil compared to those extracted from dry seeds. Dry coriander wastes contain a mixture of dried leaves, stems and broken seeds, which contain large areas of oily cells. In this respect, the percentage of essential oils of *C. sativum* plant recorded 0.31% and 1.09% for dry seeds and waste, respectively, at zero-time treatments (immediately after harvest) against 0.35% and 1.10% after storage at room temperature for 1 year, respectively (Fig. 1).

Comparison between the essential oil major constituents of seeds and waste of *C. sativum* plant

The essential oil components for both seeds and waste residues of *C. sativum* plant, which grew under the Egyptian conditions, were identified by GC–MS. Twenty-eight and sixteen components were identified in the essential oil extracted from coriander seeds and waste at zero time (immediately after harvest) accounting 84.78% and 94.94%, respectively. The data in Table 1 clearly show that the major components of the essential oil extracted from dry seeds and waste immediately after harvest were different. The major components of dry seeds essential oil were linalool (59.6%), hexanoic acid (4.92%), sabinene hydrate (4.36%), d-thujene (3.32%), caryophyllene (2.51%), α-pinene (2.28%) and camphene (1.3%) accounting 78.92% of total constituents, while the major components in essential oil of waste were *trans*-anethole (29.29%), linalool (20.06%), butanoic acid (14.17%), estragole (10.25%), longifolene (6.82%), carvacrol (5.1%) and germacrene-D (2.31%) accounting 88.00% of total constituents.

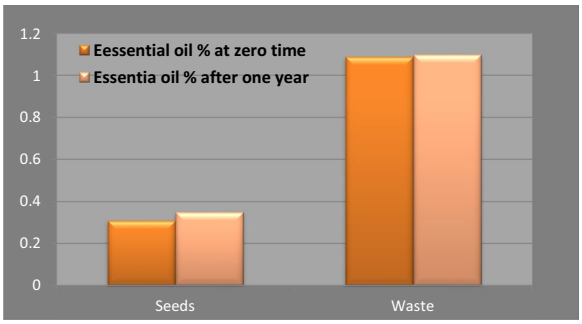

Fig. 1 Comparison of percentage of the essential oil of dry seeds and dry waste of *Coriandrum sativum* plant

Comparison between the chemical constituents of coriander seeds and waste essential oil stored under different conditions

The obtained data showed that the essential oil composition for each the seeds and waste of the coriander plant varied in their responses; some increased, while the others decreased during the different storage methods for 1 year.

Effect of different storage methods on constituents of seeds' essential oil

Concerning stored seeds' essential oil in cool conditions, T2 (analyzed the essential oil), which extracted from dry seeds directly after harvest and stored in the refrigerator at 5 °C (cool conditions for 1 year), and T3 (oil stored at room temperature for 1 year), the data showed that changes occurred in the essential oil constituents under different storage conditions as compared to zero-time treatment T1 (essential oil extracted from

Table 1 Comparison between the major constituents of essential oil for both seeds and waste of coriander plant

Seeds		Waste	
Compound	**Percentage**	**Compound**	**Percentage**
Linalool	59.6	*Trans*-anethole	29.29
Ethyl hexanoic acid <2->	4.92	Linalool	20.06
Sabinene hydrate trans	4.36	Butanoic acid, 2 methyl, 2-methoxy-4-(2-ropenyl) phenyl ester	14.17
α-Thujene	3.32	Estragole	10.25
Caryophyllene <beta>	2.51	Longifolene	6.82
α-Pinene	2.28	Carvacrol	5.1
Camphene	1.3	Germacrene-D	2.31
Total	78.29		88.00

seeds in mature stage directly after harvest). The main constituent linalool compound recorded 59.6% at zero-time treatment (T1) against 59.28% under cool conditions treatment (T2) and 47.69% with dry seeds stored at room-temperature treatment (T3) for 1 year. In other words, the linalool compound recorded a very slight decrease in cool conditions (T2), while the stored dry seeds at room temperature (T3) recorded high decrease proportion of this compound (linalool) compared to T1 (zero-time treatment). The other major compounds, ethyl hexanoic acid and sabinene hydrate, recorded 0.12% and 0.49% with cool conditions treatment (T2) and 0.56% and 0.0% at room-temperature treatment (T3) compared to zero-time treatment (T1) which recorded high percentage (4.92% and 4.36%), respectively (Table 2).

Oxygenated monoterpenes (OM) were the major ones in the essential oil extracted from coriander seeds under all storage treatments, followed by monoterpene hydrocarbons (MH) and then sesquiterpene hydrocarbons (SH). At zero time (T1), OM group recorded 66.72% against 64.65% and 53.73% for seeds oil stored in cool conditions (T2) and dry seeds stored at room temperature (T3), respectively. The same trend was observed with other groups: MH group decreased from 8.6% at zero time (T1) to 7.47% under cool conditions (T2) and 4.99% at room temperature (T3), while SH group decreased

Table 2 Percentage variation of the essential oil constituents of the seeds of coriander plant under different storage conditions for 1 year

Compound no.	Compounds %	KI	Group	Treatment		
				T1	T2	T3
1	α-Pinene	939	MH	2.28	2.41	1.02
2	Camphene	953	MH	1.3	0.24	0.01
3	β-Pinene	980	MH	0.06	0.04	0.01
4	α-Thujene	995	MH	3.32	0.80	0.18
5	3-Carene	1011	MH	0.55	0.03	0.29
6	P-cymene	1026	MH	0.17	3.1	1.83
7	Limonene	1031	MH	0.11	0.42	0.8
8	(Z)-β-ocimene	1041	MH	0.66	0	0.42
9	γ-Terpinene	1061	MH	0.15	0.43	0.43
10	Sabinene hydrate trans	1097	OM	4.36	0.49	0
11	Tetra hydro linalool	1098	OM	0.26	0.68	0
12	Linalool	1098	OM	59.6	59.28	47.69
13	Camphor	1151	OM	0.25	3.50	1.02
14	Dihydro terpineol (trans-Alpha)	1161	OM	0.03	0.47	0
15	Borneol	1168	OM	0.34	0.05	0.55
16	α-Terpineol	1189	OM	0.04	0.06	0.25
17	Nerol	1228	OM	0.8	0.03	0.36
18	Citral A	1275	OM	0.27	0.05	1.67
19	Thymol	1290	OM	0.58	0.01	0.12
20	Carvacrol	1301	OM	0.19	0.03	2.07
21	Elemene <Delta->	1337	SH	0.05	0.02	0.22
22	Caryophyllene <beta>	1418	SH	2.51	0.05	0.45
23	Germacrene B	1556	SH	0.3	0.04	0.56
24	Dendrolasin	1574	OS	0.19	0.05	0.05
25	Caryophyllene oxide	1589	OS	0.08	0.06	0.1
26	Elemenone <cic-beta->	1591	OS	0.43	0.09	0.06
27	Ethyl hexanoic acid <2->	1129	VC	4.92	0.12	0.56
28	Dodecane	1199	VC	0.98	0.07	1.42
	Total			84.78	72.62	62.14

T1 = Extracted and analyzed the essential oil from dry seeds at mature seed stage directly after harvest (at zero time). T2 = Analyzed the essential oil, which extracted from dry seeds and stored directly after harvest in the refrigerator at 5 °C (cool conditions for 1 year).T3 = Extracted and analyzed the essential oil from dry seeds which stored at room temperature for 1 year

Table 3 Percentage of different chemical groups of coriander seeds' essential oil under different storage conditions for 1 year

Groups	Treatments		
	T1	T2	T3
Monoterpenes hydrocarbons (MH)	8.6	7.47	4.99
Oxygenated monoterpenes (OM)	66.72	64.65	53.73
Sesquiterpene hydrocarbons (SH)	2.86	0.11	1.23
Oxygenated sesquiterpenes (OS)	0.7	0.2	0.21
Various compounds (VC)	5.9	0.19	1.98
Total	84.78	72.62	62.14

T1 = Extracted and analyzed the essential oil from dry seeds at mature seed stage directly after harvest (at zero time). T2 = Analyzed the essential oil, which extracted from dry seeds and stored directly after harvest in the refrigerator at 5 °C (cool conditions for 1 year). T3 = Extracted and analyzed the essential oil from dry seeds which stored at room temperature for 1 year

dramatically from 2.86% for T1 to 0.11% and 1.23% for T2 and T3, respectively (Table 3).

Effect of different storage methods on essential oil constituents in waste of coriander plant

The results in Table 4 clear that essential oil, which stored in both cool conditions, T5 (essential oil of waste which stored in cool conditions for 1 year) and T6 (essential oil extracted from coriander waste stored at room temperature for 1 year), gave a similarity of constituents of coriander oil extracted at zero time, T4 (oil extracted from dry waste directly after harvest). But there is an obvious variation in the concentration of these compounds upon comparing the three treatments. Sixteen components have been identified that represent nearly 93.22% (oil stored in cool conditions), 92.6% (oil stored at room temperature) and 94.94% (oil extracted after harvest) for the three samples, respectively. *Trans*-anethole, linalool, butanoic acid, 2-methyl-, 2-methoxy-4-(2-ropenyl) phenyl ester, estragole and carvacrol were found as the main constituents of coriander waste oil in all treatments. Essential oil components of coriander waste oil found a variable response to storage treatments. Some increased, while other declined during storage up at 1 year. There is an obvious increase in the concentration of *trans*-anethole and butanoic acid due to the storage of coriander waste oil, T5 and T6 treatments. The first major constituents (*trans*-anethole) in this respect recorded 29.92% at zero time (T4), against 35.62% and 41.54% under cold and room-temperature conditions, T5 and T6 treatments, respectively. As for butanoic acid compound recorded 14.17%, 16.77 and 16.70% for the same treatments, respectively. The other major constituents of

Table 4 Percentage variation of the essential oil constituents of coriander waste under different storage conditions for 1 year

Compound no.	Compound %	KI	Group	Treatments		
				T4	T5	T6
1	α-Pinene	939	MH	0.53	0.22	0.21
2	Camphene	953	MH	0.31	0.16	0.2
3	β-Pinene	980	MH	0.16	0.15	0.2
4	Para cymene	1026	MH	0.86	0.75	1.1
5	Limonene	1031	MH	0.78	0.89	1.88
6	Linalool	1098	OM	20.06	15.61	8.82
7	Estragole	1195	OM	10.25	9.04	8.1
8	Decanal	1204	VC	0.7	0.5	0.6
9	*Trans*-anethole	1283	OM	29.29	35.62	41.54
10	Carvacrol	1298	OM	5.1	3.26	2.7
11	Geranyl acetate	1383	VC	0.85	0.93	0.54
12	Longifolene	1402	OM	6.82	5.57	5.33
13	Germacrene-D	1480	SH	2.31	1.21	1.51
14	Bisabolene	1509	SH	0.65	0.59	0.47
15	Spathulenol	1576	OS	1.05	0.87	1.36
16	Butanoic acid, 2 methyl-,2-methoxy-4-(2-ropenyl) phenyl ester	2149	VC	14.17	16.77	16.7
	Total			94.94	93.22	92.6

T4 = Extracted and analyzed the essential oil from dry waste directly after harvest. T5 = Analyzed the essential oil which extracted from dry waste immediately after harvest and stored in the refrigerator at 5 °C (cool conditions for 1 year). T6 = Extracted and analyzed the oil from dry waste which stored at room temperature for 1 year

linalool, estragole and longifolene were decreased due to the storage treatments. They recorded the highest percentages at zero time (T4), lower percentages were observed by the storage of waste in cool conditions and the lowest percentages were got when stored at room temperature. The second major constituent (linalool) was 20.06% at zero-time treatment (T4) against 15.6% and 8.8% in cool conditions (T5) and at room-temperature treatment (T6), respectively. It is clear from the data in Table 4 that the percentages of *trans*-anethole and butanoic acid were higher under the cool storage than it in the room-temperature storage.

Oxygenated monoterpenes (OM) group was the major chemical group recorded in all treatments. It was observed that storage in cool conditions and at room-temperature treatments reduced the proportion of this group compared to zero-time treatment (T4). The ratio reached to 69.1% and 66.49% with cool (T5) and room-temperature (T6) treatments, respectively, while the same group was 71.52% at zero time. The same trend was observed with sesquiterpene hydrocarbons (SH). The percentage of monoterpenes hydrocarbons (MH) and oxygenated sesquiterpenes (OS) increased with stored essential oil in room-temperature treatments as compared to other treatments (Table 5).

The previous data in Tables 3 and 5 clear that oxygenated monoterpenes group was the major one (71.52% and 66.72%) for the seeds and waste essential oil, respectively. As for the percentage of groups monoterpenes hydrocarbons and sesquiterpene hydrocarbons were (8.6% and 2.86%) in seeds essential oil, respectively, while the same groups were 2.64% and 4.01% in waste essential oil, respectively.

Table 5 Percentage of different chemical groups of coriander waste essential oil under different storage conditions for 1 year

Groups	Treatments		
	T4	T5	T6
Monoterpenes hydrocarbons (MH)	2.64	2.17	3.59
Oxygenated monoterpenes (OM)	71.52	69.1	66.49
Sesquiterpene hydrocarbons (SH)	4.01	2.88	3.32
Oxygenated sesquiterpenes (OS)	1.05	0.87	1.36
Various compounds (VC)	15.72	18.2	17.84
Total	94.94	93.22	92.6

T4 = Extracted and analyzed the essential oil from dry waste directly after harvest. T5 = Analyzed the essential oil which extracted from dry waste immediately after harvest and stored in the refrigerator at 5 °C (cool conditions for 1 year). T6 = Extracted and analyzed the oil from dry waste which stored at room temperature for 1 year

Discussion

The change in chemical contents for both seeds and waste is due to oxidative changes in the presence of temperature, light, air and oxygen during storage. Usually, the degree of change is the function of each temperature and time, and less stable compounds may also change due to chemical reactions with other components. Concerning the stored essential oil of seeds, the decrease in the main constituents at room temperature can be due to evaporation, oxidation and other unwanted changes in essential oil constituents during storage period (Mockute et al. 2005).

Changes in essential oil components during storage can occur through the re-arrangement, hydrogenation or the removal of hydrogen from the other components (Choi and Sawamura 2002; Orav et al. 2004; Ram et al. 2005). These results are consistent with the findings in the literature. Many researchers like Njoroge et al. 1996 on *Citrus junos* and Cesare et al. 2001 on basil oil have shown an increase in some oil compositions and decrease in others during storage, i.e., storage affected qualitatively and quantitatively of terpenoids that depended on conditions of storage (storage period and temperature). The variations in essential oil content and composition during storage under different conditions could be due to the effect of different conditions on an enzymes activity and metabolism (Burbott and Loomis 1969).

Some researchers noticed characteristic changes for some essential oils under different storage conditions. They stored the volatile oil of lavender, pine, rosemary and thyme for up to 72 weeks in the presence of atmospheric oxygen at 23 °C in the dark as well as at 23 °C and 38 °C under a cool white light, respectively. Remarkable degradation of monoterpenes was observed in rosemary oil, while α-terpinene was reduced to less than 10% within 3 weeks of storage at 38 °C under daylight, but did not change during the same period at room temperature in the dark, its amount of pine oil reduced to about 40 and 65%, respectively (Turek and Stintzing 2012).

For the essential oil extracted of waste, the increase in main components by cooling treatment might be due to that lower temperatures favor the solubility of oxygen in liquids, which may negatively affect essential oil stability (Bernhard and Marr 1960). Turek et al. (2012) emphasized this result, they announced that Rosemary oil pronounced stability at lower temperature and oxidation reactions could be prevented during the storage period when stored at refrigerator temperatures.

Similar results were registered on *Melissa officinalis* plant; it was recorded that the oil kept its primary quality when stored at low temperatures, particularly at −20 °C (Najafian 2014). Also, it was found that the essential oil content of damask rose plant was not affected by different cold storage treatments. However, changes in essential

oil compositions were observed during cold storage at 4 ± 0.5 °C and -20 °C up to 1 month as compared with control (Sharma and Kumar 2016).

Conclusion

The major constituent of the essential oil from dry seeds of *C. sativum* was linalool (59.6%), while the major constituents of dry waste essential oil were *trans*-anethole (29.29) and then linalool (20.06%). The constituents of coriander oil are greatly affected by the method of storage. It was observed that the largest change in essential oils was found in dry seeds and dry waste after 1 year of storage. Storage of coriander oil immediately after extraction in the refrigerator is less harmful than extracting from seeds or waste stored for 1 year at room temperature. However, this does not prevent the use of coriander oil from seeds or waste stored at room temperature if needed, but with a lower quality than those extracted immediately after harvest or if stored directly in cool conditions. Also, the waste of *C. sativum* can be used as a fertilizer or as a new source of essential oil, as well as protecting the environment from pollution resulting from the presence of coriander wastes.

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Authors' contributions
All authors participated in the idea and developed the research plan. MEI supervises the cultivation and collection of plant specimens from Assiut region—Egypt (380 km south of Cairo). HEW and HSA supervised laboratory experiments. All authors participated in the identification of coriander oil compounds and participate in the discussion and writing research. HSA is a corresponding author. All authors read and approved the final manuscript.

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