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Comparison the Composition of Essential Oil Extracted of Coriandrum Sativum L by Hydro-distillation and Microwave-assisted Hydro-distillation and Evaluation Its Antimicrobial Activity

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Abstract
The essential oil has been extracted from Coriandrum sativum L seeds from Apiaceae collected in south west of Hama in Syria by hydro-distillation method and using the microwave-assisted hydro-distillation. The composition, the qualitative and quantitative analysis of the essential oil was achieved and characterized by means of GC-MS and comparing with the references in the literature, about 30 compounds of the essential oil were characterized in both methods. The components and the main oil yield obtained in the form of hydro-distillation (HD) were compared with those obtained by microwave-assisted hydro-distillation (MAHD). The major compounds of the oil extracted by the method of hydro-distillation are: Linalool (59.6%), alpha-pinene (10.1%) and p-Cymene (9.58%), while the major compounds of the oil extracted by microwave-assisted hydro-distillation are : Linalool (63.2%), Geraniol (5%) and Geranyl acetate (6.4%). The microwave extraction method showed a shorter time (30 min) than the time taken by the hydro-distillation extraction method (3 h) Innovative microwave-based extraction methods are faster, more energy efficient and environmentally friendly. The biological effects of essential oil were studied on some pathogenic bacteria including: *Staphylococcus aureus*, *Escherichia coli*.
Keywords: Apiaceae, Coriandrum sativum L, Essential oils, hydro-distillation, microwave, GC-Mass,, *Staphylococcus aureus*, *Escherichia coli*.

1. Introduction:

The Apiaceae (Umbelliferae) is one of the best known families of flowering plants, which comprise 300–450 genus and 3000–3700 species [1], They are aromatic plant and have a distinctive flavor with diverse volatile compounds from the fruits and leaves. *Coriandrum sativum* L, known as Coriander, is native to Iran, however it is widely distributed around the world. The seeds contain an essential oil (up to 1%) and the seeds are used in traditional medicine for indigestion, against worms, rheumatism and pain in the joints [2] It has also the nematicide activity [3], antibacterial activity [4] and larvicide activity [5].

Coriander seeds contain more than 80% of the fatty acid Petroselinic acid, the compound responsible for lowering the level of Arachidonic acid in the heart [6]. Coriander seeds are widely used in perfumery and cosmetics, and this oil is classified among the good oils but the ingredients responsible for aromatic properties are not yet fully known [7]. Despite intensive studies to determine the coriander oil components, research on the effect of extraction methods on the essential oil components of coriander plants is low. As a part of our study to characterize the chemical constituents of Syrian plants, we have investigated the essential oil of *Coriandrum sativum* L., In the present communication we wish to report the extraction and studying the chemical composition of the essential oil of *Coriandrum sativum* L and evaluate the effect of extraction methods on the chemical composition on the yield and composition of essential oil. And to determine the optimal method to extract the essential oil and evaluate its biological activity.

2. Taxonomic description of *Coriandrum sativum* L

The C. sativum L. (family Umbelliferae/Apiaceae) is an erect annual herb with pronounced taproot, and slender branching stems up to 20–70 cm in height. There are two varieties of C. sativum: vulgare and microcarpum; the former has larger fruits (3–5 mm diameter) with EO yields of 0.1%–0.35% (v/w) while the latter has smaller fruits (1.5–3 mm diameter) with EO yields of 0.8%–1.8% (v/w)[8] The leaves are lanceolate, green or dark green, glabrous on both surfaces and are variable in shape and lobed. The flowers are borne in small umbels, white or light pink, asymmetrical with the petals pointing away from the centre. The coriander seed is almost ovate globular dry schizocarp with two mericarps [9], and multiple longitudinal ridges on the surface possessing a sweet, slightly pungent, citrus like flavor with a hint of sage [10].

3. Experimental Procedure:

3.1. Plant Material

Seeds of C. sativum L., were collected and dried in April 2018, from South-Est of Hama, Syria. The plant was authenticated by the Atomic Agent in Syria. A voucher specimen of plant was deposited in the laboratory of chemistry of natural products, Department of chemistry, Faculty of sciences, AL Baath University, Homs, Syria.

3.2. Essential oils analysis

The analysis of the essential oil was performed with Shimadzu Bruker Ultra Shield 400MHz gas chromatograph with a capillary column DB5 (30m × 0.25 µm) With an internal character (0.25µm). Temperature program was as follows: 3 min at 40°C, increased to 100°C at a rate of 5°C min, then, increased to 120°C at a rate of 5°C min and held at that temperature for 1 min, increased to 180°C at a rate of 6°C min, increased to 200°C at a rate of 20°C min, increased to 220°C at a rate of 30°C min, then increased to 280°C at a rate of 40°C min and held at that temperature for 1 min. Injection temperature was 230°C. Injection volume was 1.0 µL. Helium was used as a carrier gas (1 mL/min). the identification of the constituents was performed by comparing the spectra obtained with database of Wiley Spectral Library Collection and NSIT library database. Quantitative data were obtained from the electronic integration of the FID peak areas

3.3. Extraction the essential oil:

3.3.1. hydro-distillation (HD):

The Coriander seeds (150 g) and 1,000 ml distilled water placed in a round bottom flask and connected to a Clevenger-type apparatus. Hydro-distillation was completed for 3h after boiling. Oil yield of the sample was calculated on a moisture free basis. The oil was dried over anhydrous sodium sulphate and kept at 4 °C in the sealed brown vial until the analysis by GC-MS.

3.3.2. Microwave-assisted hydro-distillation (MAHD):

The oil was obtained from (150g) of the seeds of Coriander by hydro-distillation for 30 min using a Clevenger type apparatus placed in a modified microwave oven (800 w). The oil was dried over anhydrous sodium sulphate and kept at 4°C in the sealed brown vial until the analysis by GC-MS.

4. Evaluation the biological activity:

The biological efficacy of the essential oil extracted from the seeds of C. sativum L., was studied in a against two bacteriostatic strains:Staphylococcus aureus, Escherichia coli. The lobster was highly effective on bacterium Effectiveness has been compared with gentamicin anti-inflammatory drug. Transfer (0.1) cm^3 from the diluted bacterial suspension to the center of nutritious Nutrient agar and spread on the surface of the center in a homogeneous manner and incubated for 30 minutes at a temperature of 37 °C for the purpose of sowing. In the meantime, the tablets were filled with oil extract and active ingredients. The discs were prepared from the filter paper with a perforation of the leaves and a diameter of 5 mm. These tablets were treated with different concentrations of the oil extract (100%, 50%, 25%). The steroid tablets containing the nutrient medium are then sterilized with sterile concentrates. At this time, the Gentamicin filter paper is coated with a concentration of 500 $\mu g/cm^3$. It is determined by the different concentrations of the oil extract and DMSO, all in one dish on the feeding medium and incubated at a temperature of (37) °C for a period of (16) hours.

5. Results and discussion:

The chemical composition of the essential oil obtained from C. sativum in two methods (Microwave-assisted hydro-distillation and hydro-distillation) are represented together with the retention indices in Table 1, the oils yields of the plant was determined as1.2 % and 0.7 % w/w in MAHD and HD respectively. The GC–MS analyses of both samples revealed the presence of a total of 32 components. Thirty compounds were identified in the hydro-distilled oil (Clevenger-type apparatus) which accounted for 95.18 % of the total oil composition. This oil was dominated by monoterpenoids such as, linalool, γ-terpinene, α-pinene and p-cymene. In the same way, thirty compounds were identified from the microwave extracted oil which accounted for 95.8 % of the total oil composition. The MAHD EO was dominated by monoterpenoids such as linalool and geranyl acetate.

Table 1. Chemical composition, retention time (RT)) and relative percent of coriander essential oils extracted by HD, MAHD

Peak number	Compound	RT		% Composition	
		HD	MAHD	HD	MAHD
1	α- pinene	5.27	5.27	10.1	4.4
2	p-Cymene	6.74	6.72	9.58	3.2
3	β- pinene	7.25	7.23	0.3	0.2
4	Myrcene	8.07	8.05	0.2	0.2
5	Limonene	8.35	8.32	0.3	0.2
6	ɣ-Terpinene	8.86	8.86	0.2	5
7	Terpinolene	9.09	9.07	0.1	0.1
8	Nonanal	9.3	9.29	0.1	0.1
9	trans-Linalool oxide	10.12	9.78	0.1	0.1
10	cis-Linalool oxide	10.42	10.42	0.3	0.1
11	Citronellal	10.65	10.65	0.1	0.1
12	Decanal	11.11	11.10	0.1	0.1
13	Camphor	11.39	11.35	0.6	0.7
14	Octanol	12.41	12.40	0.4	0.7
15	Terpinen-4-ol	12.62	12.60	0.4	0.1
16	Citronellyl acetate	12.76	12.72	0.1	-
17	Myrtenyl acetate	13.36	13.41	0.1	0.4
18	α-Terpineol	13.57	13.56	0.3	0.1
19	Borneol	13.69	13.69	0.1	0.1
20	Neryl acetate	14.27	14.10	0.1	-
21	Carvone	14.54	14.54	0.2	-
22	Linalool	15.93	15.93	59.6	63.2
23	(E)-2-Dodecenal	17.30	17.31	0.4	0.5
24	Caryophyllene oxide	19.02	19.02	-	0.2
25	Methyl eugenol	20.59	20.58	-	0.2
26	Geranyl acetate	22.38	22.39	4.2	6.4
27	Geraniol	24.24	24.22	3	5
28	Tetradecanoic acid	27.27	27.17	1.1	1.8
29	Pentadecanoic acid	29.17	29.10	0.2	0.2
30	Hexadecanoic acid	29.80	29.80	2.5	1
31	Hexamethyl-pyranoindane	30.80	30.02	0.3	1.3
32	n-Hexadecanoic acid	31.4	31.3	0.2	0.1
	Yield (w/w)%			0.7 %	1.2%
	Monoterpenes hydrocarbons			20.68	13.3
	Monoterpenes oxygenated			74.5	82.5
	Total			95.18	95.8

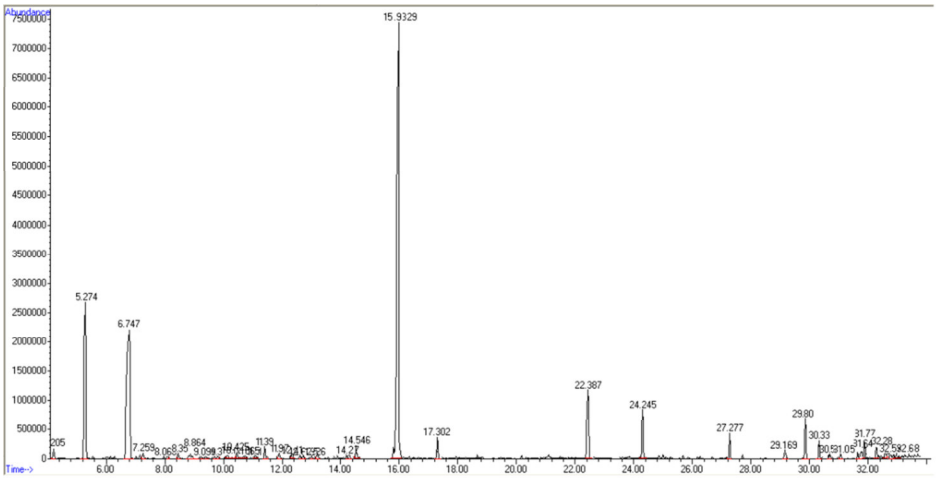

Figure -----: Chromatogram of oil obtained from coriander by HD method

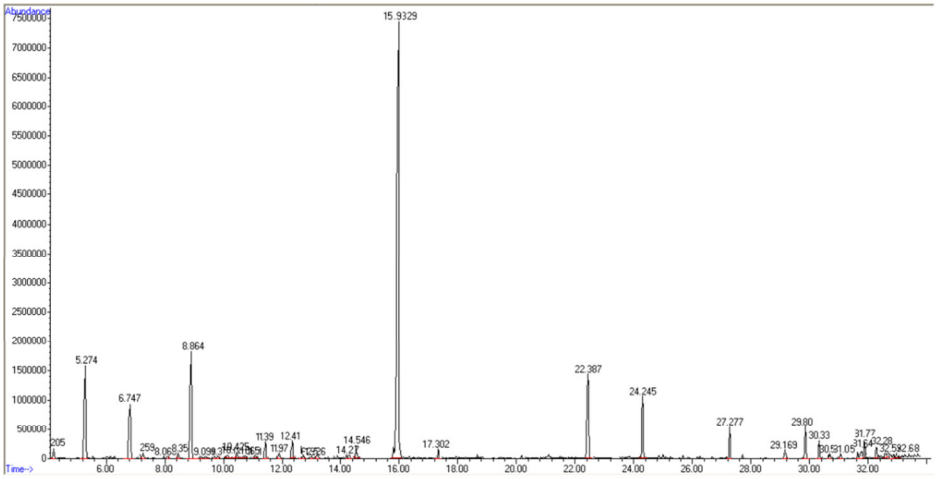

Figure -----: Chromatogram of oil obtained from coriander by MAHD method

Discussion

According to results in current study, the major compound in both EO was linalool which its amount in HD and MAHD was 59.6 % and 63.2%, respectively. Significant difference was observed in the linalool content of MAHD EO and HD EO. The total amount of monoterpene hydrocarbons in HD EO differ significantly with the amount in MAHD EO (20.68 % compare to 13.3 %). Moreover there were a difference between oxygenated monoterpenes content in MAHD EO and HD EO (82.5 % compare to 74.5 % respectively). The content of geranyl acetate in MAHD EO (6.4 %) increased compared to HD EO (4.2%), significantly. A critical observation of the oil compositions revealed that higher amounts of oxygenated monoterpenes are present in the essential oil isolated by MAHD (82.5 %) in comparison with the oil extracted by HD (74.5 %). MAHD method was important in terms of saving energy and extraction time (30 min compared to180 min in HD method) and the amount of oxygenated monoterpenes which play the great role in the essential oil properties increased, although the oil yield and total composition decrease by using this method.

The previous studies showed that linalool was the main compound on the EO Coriander collected in Serbia[11] , and Romania[12] , which agrees with our research that linalool was major component in the MAHD and HD EO . In the same way , Singhetal . investigated the EO chemical composition of Coriander from India and found that the major compounds were linalool (75.3 %), geranyl acetate (8.1%) and α-pinene (4.1 %)[13]. There is a report about the MAHD and HD EO of Coriander in which, their results were similar to our research. In MAHD essential oil the content of geranyl acetate increased while, monoterpene hydrocarbons content decreased. Furthermore oxygenated monoterpenes increased in MAHD EO [14].

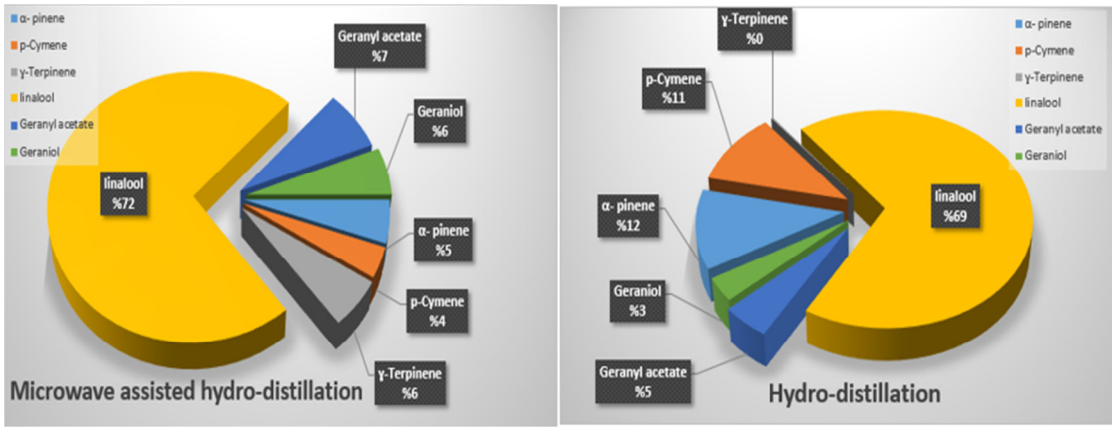

Figure 1: Comparison of the major components of the essential oil of coriander by HD and MAHD

The essential oil exhibit significant activity against the pathogenic bacteria including: *Staphylococcus aureus, Escherichia coli*.in comparing to the gentamicin. The damping area is measured by a

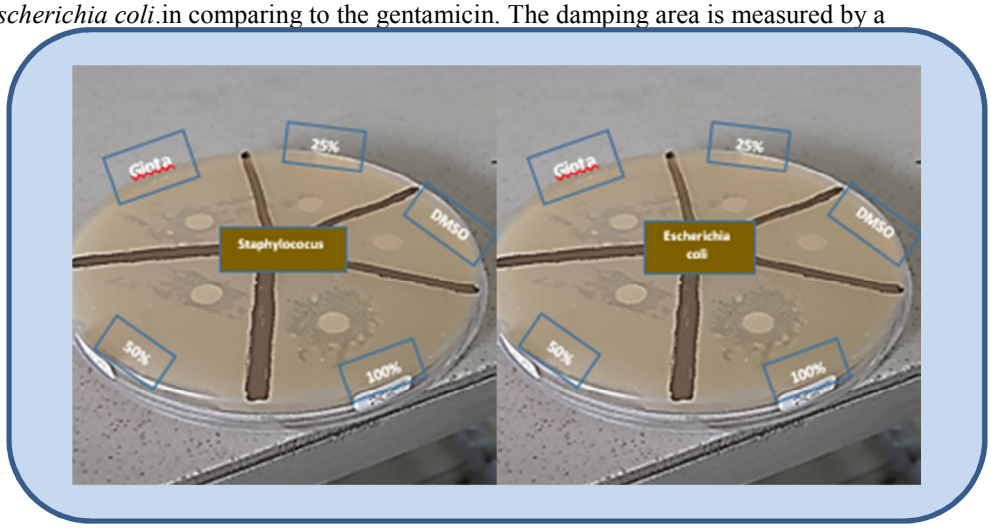

Figure 2: The zones of inhibition by essential oil of C. sativum L against to the growing of Staphylococcus aureus and Escherichia coli. in different concentration

Table 2. The activity of essential oil of C. sativum L, it is calculated by measuring the diameter of the inhibition zone (mm).

Org.	Essential oil Con.				
	100%	50%	25%	gentamicin	DMSO
E. coli	28.00 ±1.00	17.33 ±1.20	2.5 ±0.99	4.3	-
Staph. aureus	7.20 ±0.50	6.5 ±0.5	4.00 ±0.2	4.5	-

6. Conclusions

In MAHD method, time extraction is significantly shorter than HD method. The MAHD with lower energy consumption than HD is offered instead traditional methods. Therefore, considering the operation cost MAHD could be carried out using half of the expenses required by HD. The MAHD yielded less percentage of γ-terpinene, α-pinene and p-cymene compared to HD, therefore, MAHD is more efficient method for obtaining essential oil of Coriander.

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