Influence of two extraction methods on essential oils of some Apiaceae family plants

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Background
Renewed interest in natural materials as food flavors and preservatives has led to the search for suitable essential oils. One factor that influences the essential oil content (%) is the extraction method used. *Carum carvi* (caraway), *Anethum graveolens* L. (anise), and *Pimpinella anisum* L. (dill) are well known plants from Apiaceae family widely spread in Egypt, where they have good climatic and soil conditions for high yield and good quality. Essential oil content is the main criteria for determining the quality of the fruits of these plant species. The aim of the study was to choose the best method for essential oil extraction.
Objective
The present research was conducted to evaluate the possible impacts of two types of distillation methods – hydrodistillation and hydro-steam distillation on essential oil content (%) and its main constituents of caraway, anise, and dill fruits.
Materials and methods
Seeds of the three species were subjected to two types of distillation methods – hydrodistillation and hydro-steam distillation. The essential oil content (%) of the three plants were determined and gas chromatography-mass spectrometry analyses was carried out to identify the chemical constituents of the oil samples and their percentage were calculated in order to clear the effect of the two extraction methods applied.
Results and conclusion
It was established that while hydrodistillation gave higher essential oil yields for caraway and dill seeds (3.14 and 2.36%, respectively), hydro-steam distillation gave the maximum mean values of essential oil content of anise seeds (0.76%). The maximum values of the main components such as carvone (54.45%), transanethole (98.97%), and carvone (57.71%) were obtained as a result of hydrodistillation method for caraway, dill, and anise seeds, respectively.

Keywords:
anise, caraway, dill, essential oil, hydro-steam distillation, hydrodistillation

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Introduction

Caraway, anise, and dill are well known plants from the Apiaceae family widely spread in Egypt, where they have good climatic and soil conditions for high yield and good quality. Essential oil content is the main criteria for determining the quality of these fruits. Caraway (*Carum carvi* L.) fruits contain from 1 to 6% of essential oil which gives its characteristic aroma [1]. Because of this, *Carvi fructus* is used as spice in ice creams, candies, baked goods, meat, cheese, pickles, condiments, soft drinks, and alcoholic beverages. Anise (*Pimpinella anisum* L.) fruits contain from 1.5 to 5.0% of essential oil which gives the sweet herbaceous odor and taste of this plant [2]. Because of its strong aroma, anise is often used in pharmaceutical industry for masking the bad taste of remedies, as well as for the preparation of many sweets (chocolate, cookies, and candies) and alcoholic beverages. The fruits of dill (*Anethum graveolens* L.) contain essential oil whose most important essential oil compounds are carvone and limonene. The dill essential oil has hypolipidemic activity and could be used as a cardioprotective agent [3].

Extraction methods of seed oils are an effective factor in the properties of oils. The traditional technologies pertaining to essential oil processing are of great significance and are still being used in many parts of the globe. Hydrodistillation, hydro-steam distillation, steam distillation, cohobating, maceration, and enfleurage are the most traditional and commonly used [4].

There are two main distillation methods for obtaining essential oil, namely (i) water distillation (or hydrodistillation) and (ii) hydro-steam distillation. In the manufacture of essential oils using the method of hydrodistillation, the botanical material is completely immersed in water and the whole is brought to boiling. This method protects the oils so extracted to a certain degree as the surrounding water acts as a barrier to prevent it from overheating. When the condensed material cools down, the water and essential oil are separated and the oil decanted, to be used as essential oil. The hydro-steam distillation method is basically a combination between the conventional hydrodistillation and that of steam distillation. When this second common method of distillation is used, the plant material is supported on a perforated grid or screen inserted some at distance above the bottom of the still. The lower part of the still is filled with water, to a level somewhat below this grid. The water may be heated by any of the methods previously mentioned. After being saturated in this way, wet steam of low pressure passes through the plant material. The typical features of this method are: first, that the steam is always fully saturated, wet, and never superheated; second, that the plant material is in contact with steam only, and not with boiling water [5].

The effects of different distillation methods on essential oil content and composition of aromatic plants has been previously reported [6] on *Thymus kotschyanus* and [7] on rose-scented geranium (*Pelargonium* spp.). It was established that the highest essential oil yields were obtained by the hydrodistillation method and the lowest by steam distillation.

The aim of this study was to test the effect of two different distillation methods (hydrodistillation and hydro-steam distillation) on the essential oil content and composition of *C. carvi*, *P. anisum*, and *A. graveolens* plants.

Materials and methods

Plant material

Seeds of caraway, anise, and dill obtained from local medicinal plant grower were used in this study. Essential oil isolation: three replicates for the seeds of each of the plant species under study were performed. A total of 100 g per each replicate were subjected to hydrodistillation and hydro-steam distillation for 3 h using a Clevenger type apparatus [8]. The laboratory scale hydrodistillation apparatus, described in the European Pharmacopoeia [9], contains 500 ml volume round-bottom flask connected to a slightly modified Clevenger head [10] and a capillary collector tube with 1.0 ml (0.01 ml scaling). For hydro-steam distillation the plant materials are placed into a sieve or on a grate atop the distillation vat/pot/still. The water below is heated up causing steam distillation of the material. The steam and water are pushed through the plant material where the steam and oils are captured and then separated out to produce the essential oil.

Gas chromatography-mass spectrometry

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 40–240°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. The components of the oils were identified by comparison of their mass-spectra with those of a computer library or with authentic compounds and confirmed by comparison of their retention indices either with those of authentic compounds. Kovat's indices [11] were determined by coinjection of the sample with a solution containing a homologous series of n-hydrocarbons, in a temperature run identical to that described above.

Statistical analysis

All data were calculated with three replicates. Differences were analyzed with one way analysis of variance using completely randomized design and least significant difference. P values of less than 0.05 were considered to be significant according to Snedecor and Cochran [12].

Results and discussion

A qualitative and quantitative comparison of the essential oil constituents based on the different distillation methods applied to *C. carvi*, *P. anisum*, and *A. graveolens* plants oil is presented in Tables 1–4.

Essential oil content (%)

Data tabulated in Table 1 and illustrated in Fig. 1, clear that distillation methods had a significant effect on essential oil percentage of caraway (3.140 and 2.110%) and dill seeds (2.360 and 1.850%). The highest mean values for oil percentage of caraway and dill seeds were obtained by hydrodistillation and the lowest by hydro-steam distillation. On the other hand, the highest mean values of essential oil percentage for anise seeds was

Table 1 Effect of water distillation and water-steam distillation methods of essential oil percentage of caraway, anise, and dill seeds

Plants	Hydro distillation	Hydro-steam distillation	LSD at 5%
Caraway	3.14[a]	2.111[b]	0.059
Anise	0.673[a]	0.763[a]	NS
Dill	2.360[a]	1.850[b]	0.086

Same letter within the same row are not significant. LSD, least significant difference; NS, not significant.

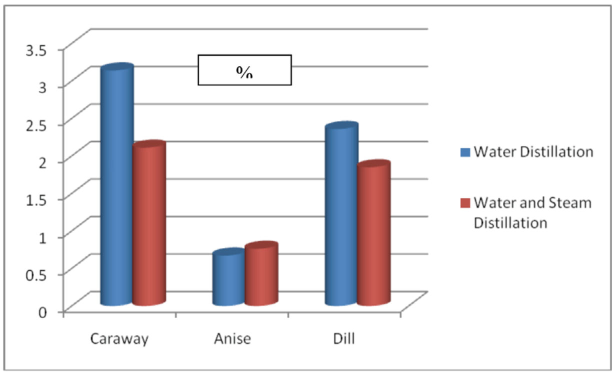
obtained by hydro-steam distillation. These results are in agreement with previous work obtained by the authors [5–7,13].

Essential oil components

Data tabulated in Table 2 show the effect of two distillations methods on essential oil components of *C. carvi* seeds. Fifteen constituents were found in *C. carvi* seeds essential oil extracted by both methods. The main constituents were carvone (30.66–54.45%) and limonene (22.47–38.80%). The highest relative percentages of carvone (54.45%) and limonene (38.80%) were obtained as a result of the hydrodistillation method. The main quality criterion for caraway essential oil is the carvone/limonene ratio [14], which is variable during ripening. In green, immature seeds the limonene concentration is higher [15]. Although it is not clear as yet which component(s) play the most important role in producing taste and fragrance, it can be expected that higher limonene-to-carvone ratio means better quality of caraway [16]. The overall quality of fruits is considered to correlate with the content of essential oil and its carvone/limonene (C/L) ratio: the higher the ratio, the better the quality [17].

We determined the composition of the essential oil and studied the variation of the ratio of the two main components under two distillation methods. The ratio of carvone/limonene were higher (54.45/38.80) for hydrodistillation than for hydro-steam distillation which recorded 30.66/22.47. Recently, (+) carvone extracted from caraway seeds has been introduced as an effective sprouting inhibitor of potatoes [18]. The expanding commercial potential of (+) carvone has now generated interest in maximizing the yield of this substance from caraway seeds, a goal that requires an understanding of the process of carvone biosynthesis. More than 30 years ago[19], hypothesized that in dill fruits, which also contain (+) carvone and (+) limonene as the main components of its essential oil, limonene is an intermediate in the biosynthesis of carvone. During fruit development the content of carvone (as a percentage of fruit weight) increases at the expense of limonene, providing support for this hypothesis.

Figure 1



Effect of different distillation methods on essential oil percentage.

However, after measuring the changes in the absolute amounts of limonene and carvone in caraway fruits and performing in-vivo radiolabeling experiments[20,21], showed that limonene is no longer available as a precursor for carvone biosynthesis once it has been secreted into the essential oil ducts.

The pathway of (+) limonene and (+) carvone biosynthesis in caraway has been assumed [22] to be analogous to the biosynthesis of (−) limonene and (−) carvone in spearmint [23]. In this process, GPP, the ubiquitous precursor of the monoterpenes, is cyclized by a monoterpene syntheses to (+) limonene. The product is either stored in the essential oil ducts or oxidized to (+) trans-carveol by a Cyst P-450-dependent hydroxylase. Subsequently, a NAD^+ or $NADP^+$-utilizing dehydrogenase oxidizes (+)-trans-carveol to (+) carvone, which is then stored exclusively in the essential oil ducts. Concerning the effect of two distillation methods on the components of *A. graveolens* seeds essential oil, results indicate in Table 3, clear that 15 components were identified as a result of both methods. The main components of essential oil extracted by hydrodistillation were carvone (57.71%), p-cymene (26.62%), and camphene (8.49%) whereas the main components of essential oil obtained by hydro-steam distillation were carvone (23.36%), camphene (20.85%), and β-phellandrene (16.85%). The dill fruit oil contents and compositions varied with extraction method. Other authors studied the chemical composition of essential oil extracted by hydro-steam distillation and hydrodistillation from dill fruits cultivated in Thailand [24]. The main constituents of dill oils were dillapiole (19.98–48.9%), carvone (18.05–28.02%), and limonene (26.96–44.61%). Minor components, β-pinene (0–0.79%), β-myrcene (0.16–0.21%), decane (0.44–0.49%), 1,5,8-p-menthatriene (0.19–0.27%), undecane (0.34–0.38%), naphthalene (1.63–2.11%), cis-dihydrocarvone

Table 2 Essential oil components by gas chromatography-mass spectrometry analysis of *Carum carvi*) seeds

Essential oil compounds	Kovat's index	% compound	
		A	B
α-pinene	939	0.12	7.88
Myrcene	994	0.04	0.10
ρ-cymene	1026	0.13	3.18
Limonene	1030	38.80	22.47
1,8-Cineol	1031	3.79	4.52
γ-terpinene	1062	0.09	3.86
Cis-limonene oxide	1138	0.32	1.07
α-terpineol	1189	0.02	0.22
Cis-dihydrocarvone	1193	0.14	0.41
Trans-dihydrocarvone	1199	0.74	17.78
Carvone	1243	54.45	30.66
Carvenone	1252	0.12	0.31
Linalool acetate	1257	0.02	1.62
ΔCadinene	1524	0.02	0.30
Aciphyllene		0.02	5.60
Total		98.82	99.98
Monoterpene hydrocarbons		37.49	39.18
Sesquiterpene hydrocarbons		0.30	6.02
Oxygenated compounds		62.19	59.62

A, hydrodistillation; B, hydro-steam distillation.

(0.38–0.95%), trans-dihydrocarvone (4, 1.49–1.57%), and myristicin (12, 0.67–1.41%). The essential oil extracted using steam distillation contained higher content of limonene and carvone than oil extracted using hydrodistillation. Table 4 cleared that also, 15 components were found in *P. anisum* seeds were obtained as a result of both extraction methods. The main components of essential oil extracted by hydrodistillation was transanethole (98.97%), that decreased to 33.77% with hydro-steam distillation, whereas the main components obtained by hydro-steam distillation were carvone (33.99%), nerodiol (11.37%), p-cymene (6.86%), and elemicin (4.26%). These main components with hydro-steam distillation were in traces percentage with hydrodistillation. The presented results cleared that the main component of anise seed essential oil constituents is transanethole, this result is in harmony with the reports of other authors [25–30], who used GC-MS analysis for anise (*P. anisum* L.) seeds essential oil constituents and found that transanethole was the major component. Meanwhile Nhutnam *et al.* [31], cited that steam distillation decreased anethol, they added that steam distillation leads to the loss of many other components. The results of the present work, indicate that monoterpene and sesquiterpene hydrocarbons were increased with hydro-steam distillation, and oxygenated compounds were decreased with steam distillation. The decreasing of oxygenated compounds may repacks to changing of these compounds from the oxide form to reduced form with the high temperature of steam distillation.

Table 3 Essential oil components by gas chromatography-mass spectrometry analysis of (*Anethum graveolens*) seeds

Essential oil compounds	Kovat's index	% compounds	
		A	B
α-pinene	939	0.10	0.15
Camphene	954	8.49	20.85
β-pinene	979	0.08	2.86
α-terpinene	1018	0.12	2.48
ρ-cymene	1026	26.62	0.40
Limonene	1030	0.07	3.25
β-phellandrene	1042	0.07	16.85
Linalool	1098	6.23	1.48
Camphor	1143	0.10	10.71
Carvone	1243	57.71	23.36
Eugenol	1401	0.07	1.23
Myristicin	1520	0.33	5.36
Nerolidol	1546	0.07	0.20
Elemicin	1554	0.07	8.35
Apiol	1680	0.07	0.22
Total		99.80	99.82
Monoterpene hydrocarbons		8.74	30.24
Sesquiterpene hydrocarbons		0.07	16.85
Oxygenated compounds		90.99	52.73

A, hydro distillation; B, hydro-steam distillation.

Table 4 Essential oil components by gas chromatography-mass spectrometry analysis of (*Pimpinella anisum*) seeds

Essential oil compounds	Kovat's index	% compound	
		A	B
?-cymene	1026	0.04	6.86
Limonene	1031	0.03	0.10
Camphor	1143	0.03	0.37
Cis-dihydrocarvone	1193	0.06	0.17
Methyl chavicol	1195	0.10	0.93
Carvone	1243	0.06	33.99
Transanethole	1283	98.97	33.77
Cis-anethole	1284	0.04	0.99
β-elemene	1375	0.03	0.70
Eugenol	1401	0.48	2.48
Himachalene	1447	0.04	0.29
ϒ-himachalene	1499	0.04	0.44
Nerodiol	1546	0.03	11.37
Elemicin	1554	0.03	4.26
Apiol	1680	0.03	0.21
Total		99.94	96.93
Monoterpene hydrocarbons		0.07	6.96
Sesquiterpene hydrocarbons		0.11	1.43
Oxygenated compounds		98.92	88.61

A, hydrodistillation; B, hydro-steam distillation.

Conclusion

The method of extraction affected the percentage of essential oil quantity and quality of *C. carvi*, *A. graveolens*, and *P. anisum* essential oils, analyzed by GC-MS. Variations in the main components of the three seed essential oil constituents were obtained according to the used extraction method for essential oil. Thus, monoterpene and sesquiterpene hydrocarbons (the oxygenated compounds) were decreased with steam distillation. Water distillation gave significant increase in both of caraway and dill seeds essential oil percentages, compared with steam distillation. The results showed water distillation method was suitable for obtaining the highest amount of essential oil for caraway and dill seeds whereas water and steam distillation method was suitable for dill.

It can be concluded that methods of essential oil extraction play an important role for essential oil percentage and its constituents. So, other researchers can complete this idea using other extraction methods.

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Conflicts of interest
There are no conflicts of interest.

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