JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

Composition, Quality Control, and Antimicrobial Activity of the Essential Oil of Long-Time Stored Dill (*Anethum graveolens* L.) Seeds from Bulgaria

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The essential oil of long-time stored seeds of dill (*Anethum graveolens* L.) from Bulgaria was analyzed by physicochemical methods, gas chromatography (GC), GC−mass spectrometry (MS) (achiral and chiral phases), and olfactometry, and its antimicrobial activity was tested by using different strains of microorganisms. More than 40 constituents of the essential dill oil, obtained from seeds stored for more than 35 years, could be identified as essential volatiles, responsible for the pleasant fresh (D-limonene) and spicy (D-carvone) odor of a high quality. As aroma impact compounds, D-carvone (50.1%) and D-limonene (44.1%) were found. Antimicrobial testings showed high activity of the essential *A. graveolens* oil against the mold *Aspergillus niger* and the yeasts *Saccharomyces cerevisiae* and *Candida albicans*.

KEYWORDS: ***Anethum graveolens*; stored seeds; aroma compounds; antimicrobial testing; quality control**

INTRODUCTION

The leaves and seeds of the herb dill (*Anethum graveolens* L. syn. *Peucedanum graveolens* Benth. et Hook, Apiaceae) have been well-known spices since antiquity, and they are mentioned in the Holy Bible as well as Sanskrit (*1*, *2*). The essential oils of different dill plant parts are also excessively used in many applications of the spice and food industry, especially for their pleasant and spicy aroma. The composition of an essential oil of *A. graveolens* is dependent on many factors, such as plant part, harvest time, extraction method, type of cultivar, geographic origin, storage conditions, etc. (*1*−*15*). In general, the monoterpenes D-carvone, D-limonene, and α-phellandrene are impact compounds of many essential dill oils and are responsible especially for the aroma and biological effects (*11*, *16*, *17*). The ratio of the D and/or L form of limonene and carvone can be analyzed by the use of chiral phase gas chromatography (GC) or GC-isotope ratio mass spectrometry (MS) (*5*, *11*, *18*−*20*), with chiral phase GC as the more common method.

Therefore, the aim of this investigation was to control for the first time the influence of unstandardized storage conditions for a longer period (more than 30 years) on the composition, aroma, and antimicrobial activity of the essential seed oil of *A. graveolens* from Bulgaria and to correlate this new data with that of an essential oil obtained from the same dill seeds 38 years ago.

MATERIALS AND METHODS

Plant Material. Ripe seeds of dill were harvested in the region of the town of Plovdiv in 1964. The seeds were stored for 38 years in double paper sacks (30 years in a refrigerator at 3−5 °C and 8 years at room temperature). The moisture of the original and stored seeds was estimated by azeotropic distillation to be 7.5 and 7.6%, respectively.

Essential Oil Isolation. Air-dried (whole) dill seeds were hydrodistilled in a laboratory glass apparatus from British Pharmacopoeia, modified in accordance to ref *21*. The water distillation was carried out in 1:10 proportions of raw material to water at the rate of 6% for 110 min. The yields and physicochemical properties of both oils (from original and stored seeds) are shown in **Table 1**.

Olfactoric Evaluation. The essential dill oil of stored seeds was olfactorally evaluated by professional perfumers, and the aroma of the samples was described as pleasant dill seed odor of high quality with characteristic fresh D-limonene-like top notes and later D-carvone (caraway) base notes.

Standards for Chiral GC Separations. L-Carvone (Sigma-Aldrich, Milwaukee, WI; W22490-1), d-carvone (W22492-8), L-limonene (W26330-3), and D-limonene (W50450-5) were used as the standards.

Instrumentation. GC analyses were performed with a Shimadzu GC-14A with flame ionization detection (FID) and Shimadzu Chromatopac C-R6A integrator and with a Varian GC-3700 with FID and Shimadzu Chromatopac C-R1B integrator (Shimadzu, Kyoto, Japan). Compounds were separated on 30 m × 0.25 mm (i.d.) fused silica columns coated either with a 0.25 μm film-bonded apolar FSOT-RSL-200 (Bio-Rad, Eke, Belgium) or with a 60 m × 0.25 mm (i.d.; 0.25

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10.1021/jf030004y CCC: $25.00
Published on Web 05/17/2003

Table 1. Yields and Physicochemical Properties of Original and Stored Essential Dill Oil

properties	essential dill oil from original seeds (*6*)	essential dill oil from stored seeds
yield (%, v/w of dry weight)	3.8	4.0
refractive index (20 °C)	nm[a]	1.4871
specific gravity (20 °C)	0.9144	0.9131
optical rotation	nm	+73.56
acid number	1.92	1.25
ester number	nm	6.35

[a] nm, not measured.

μm film) bonded polar Stabilwax (Restek, Bellefonte, U.S.A.). For chiral GC separations, a 25 m × 0.25 mm (i.d.; 0.25 μm film) FS-HYDRODEX-β-PM fused silica column (Macherey-Nagel Co., Düren, Germany) was used. The apolar achiral and the chiral columns were maintained at 40 °C for 2 min after injection and then programmed at 6 °C min^{-1} to 280 °C, which was maintained for 10 min (total analysis time 50 min), while the temperature program for the polar column was as follows: 40 °C for 2 min up to 250 °C for 10 min with a rate of 10 °C min^{-1} (total analysis time 38 min). Open split injection was conducted with split ratio of 1:20 and 1:50 for the nonpolar and polar columns, respectively; hydrogen was used as the carrier gas at 2.5 and 3.5 kPa, respectively. For all columns, the injector temperature was 230 °C and the detector temperature was 250 (carbowax) or 280 °C (RSL). After analysis on a RSL column, quantification was performed as % peak area using integration data. Some individual components could be identified by injection of pure compounds and comparison of their retention times (as Kovats indices, KI) with published (*11*, *22*−*26*) data.

GC-olfactometry analysis (sniffing technique) was performed with a Fractovap 2101 GC equipped with a splitting system, a model 230 LT-Programmer, a model 160 electrometer (Carlo Erba, Milano, Italy), and a Kompensograph-III recorder (Siemens, Munich, Germany). Compounds were separated on a 30 m × 0.32 mm (i.d.) fused silica column coated with a 0.25 μm film of nonpolar FSOT-RSL-200. The column was maintained at 40 °C for 5 min after injection and then programmed at 8 °C min^{-1} to 230 °C, which was maintained for 20 min. Compounds were injected in the splitless mode with hydrogen as the carrier gas (pressure, 1.8 kPa; column flow, 2 mL min^{-1}). The injector temperature was 250 °C, the detector (FID) temperature was 320 °C, and the sniffing capillary temperature was 250 °C. The column eluate sniffing split ratio was 1:50, FID:nose. Peak to odor impression correlations were performed by two professional perfumers and three fragrance chemists.

GC-MS was performed with a Shimadzu GC-17 gas chromatograph coupled with a Shimadzu QP5000 mass spectrometer (Compaq-Pro Linea data system, class5k software) and a GC-17A coupled with a QP5050 (Pentium-II data system, class 5k software). The columns (FSOT-RSL-200 and Stabilwax) and temperature programs used were for GC analysis. Split injection was performed with helium as the carrier gas. For the nonpolar column, the split ratio was 1:50, the column head pressure was 4.9 kPa, the flow rate was 0.5 mL min^{-1}, the linear velocity was 25.5, and the total flow was 25.6 mL min^{-1}; for the polar column, the split ratio was 1:126, the head pressure was 115.5 kPa, the flow rate was 1.0 mL min^{-1}, the linear velocity was 26.0, and the total flow was 131.1 mL min^{-1}. Injector, interface, and ion source temperatures were 230, 250, and 200 °C, respectively. The spectrometers were operated in electron-impact (EI) mode with 1.2 kV detection volts; the scan range was 41−400 amu; the scan interval was 0.50 s, and the scan speed was 1000 amu s^{-1}. Compounds were identified by use of NIST, Wiley, NBS, and our own mass spectra libraries as well as literature MS data (*24*, *27*−*30*).

Antimicrobial Testings. The essential oil of stored *A. graveolens* seeds was prepared as a 20% solution of ethanol and dissolved in a 0.9% NaCl solution (ratio of 1:10). The investigations are carried out parallel with controls, using only ethanol in the NaCl solution in the ratio of 1:10. As test microorganisms, Gram (+) bacteria were used as follows: *Staphylococcus epidermidis* ATCC 12228, *Staphylococcus aureus* ATCC 6538 P, *Bacillus subtilis*, and *Bacillus pumilus*. The Gram (−) bacteria used were as follows: *Escherichia coli* ATCC 8739, *Pseudomonas aeruginosa*, and *Salmonella abony* NCTC 6017; the yeasts *Saccharomyces cerevisiae* ATCC 9763 *and Candida albicans* ATCC 10231 and the mold *Aspergillus niger* were also used. The test microorganisms were obtained from the National Bank of Industrial Microorganisms and Cell Cultures, Sofia, Bulgaria. The antimicrobial activity was studied by agar diffusion cup method (*31*) using cups (8 mm) and a quantity of 50 μL of the oil. After cultivation at 37 °C for 24 h (bacteria), at 27 °C for 24 h (yeasts), and for 72 h (mold), the resulting diameter of the zones was measured. All antimicrobial experiments were carried out 3-fold, and the results were statistically processed by level of confidence $\gamma = 0.95$.

Table 2. Composition of the Essential Oil (eo) of Stored Seeds of *Anethum graveolens* L. from Bulgaria

compd	KI[a]	eo[b]	odor[c]
hexanal[e,f]	785	0.1	fatty, green, grassy
cis-3-hexen-1-ol[e−g]	849	0.1	*fresh, green grasslike*
trans-2-hexen-1-ol[e,f]	853	tr[d]	green, leafy
hexanol[e−g]	859	0.1	herbal, mild woody, sweet
α-pinene[e−g]	953	0.1	*pinelike*
β-pinene[e−g]	981	tr	pinelike
sabinene[e−g]	983	tr	fresh, citrus note, spicy
myrcene[e−g]	988	0.2	*citrus note, spicy*
α-phellandrene[e−g]	1002	0.1	minty, herbaceous
p-cymene[e−g]	1016	0.3	*herbal, spicy*
β-phellandrene[e−g]	1023	0.2	herbal
D-limonene	1027	44.1	*fresh, citrus*
benzyl alcohol[e−g]	1032	tr	aromatic, floral
γ-terpinene[e−g]	1055	tr	herbal, citruslike
artemisia ketone[e,f]	1062	0.1	green herbal
artemisia alcohol[e,f]	1084	tr	herbal
linalool[e−g]	1095	0.1	*floral, lavender-like*
(*E*)-*p*-mentha-2,8-dien-1-ol[e,f]	1122	tr	fresh, minty
(*Z*)-limonene oxide[e,f]	1137	0.2	*fresh, citruslike*
(*E*)-limonene oxide[e,f]	1142	0.1	*fresh, citruslike*
estragole[e,f]	1180	0.2	sweet herbal
α-terpineol[e−g]	1187	0.1	floral
dihydrocarveol[e−g]	1190	tr	warm, spicy, woody
3,9-epoxy-p-menth-1-ene[e,f]	1192	tr	minty, spicy, dilllike
(*Z*)-dihydrocarvone[e−g]	1195	1.9	*warm, herbaceous*
(*E*)-dihydrocarvone[e−g]	1202	0.7	*warm, herbal*
citronellol[e−g]	1211	0.1	*roselike*
iso-dihydrocarveol[e−g]	1214	tr	warm, woody, spicy
(*E*)-carveol[e−g]	1217	0.4	*caraway-like*
cumin aldehyde[e−g]	1224	0.1	sharp, acid, woody, oily
(*Z*)-carveol[e−g]	1229	tr	caraway-like, spicy
D-carvone	1231	50.1	*herbal, caraway*
chavicol[e,f]	1249	0.2	medicinal, phenolic
geraniol[e−g]	1252	0.1	*citrus- and roselike*
cuminyl alcohol[e,f]	1284	0.8	*floral*
eugenol[e−g]	1358	0.1	*spicy, cloverlike*
geranyl acetate[e−g]	1378	0.1	citrus- and roselike
methyl eugenol[e−g]	1407	tr	spicy, clovelike
β-caryophyllene[e,f]	1418	tr	spicy, woody, terpenelike
dillapiol[e−g]	1602	tr	*warm, woody, spicy*

[a] KI using a nonpolar FSOT-RSL-200 column. [b] Concentrations calculated by % peak area of GC-FID analyses using a nonpolar FSOT-RSL-200 column. [c] Odor description using published data (*32*–*36*) and GC sniffing technique. [d] Trace compound (less than 0.1%); bold reference compounds (D and L form each) injected, and olfactoric evaluated by GC-olfactometry. [e] Compound tentatively identified by retention time (KI) correlations (GC-FID). [f] Compound tentatively identified by mass spectra correlations (GC-MS). [g] Compound partly and tentatively identified by olfactoric evaluations (published data and GC sniffing technique); italic odor description using GC sniffing technique (olfactoric evaluation by professional perfumers).

RESULTS AND DISCUSSION

The essential oil of *A. graveolens* seeds, which were stored for 38 years in Bulgaria under unstandardized conditions, was

Table 3. Antimicrobial Activity of the Essential Dill Oil of Stored Seeds

test microorganisms	diameter (mm) of zone of inhibition	
	20%	1:10 control
S. epidermidis ATCC 12228	15.0 ± 0.2	13.0 ± 0.0
S. aureus ATCC 6538 P	25.0 ± 0.0	19.3 ± 0.1
B. pumilus	15.0 ± 0.0	11.0 ± 0.2
B. subtilis	17.0 ± 0.5	15.2 ± 0.1
E. coli ATCC 8739	13.3 ± 0.2	11.0 ± 0.5
P. aeruginosa	10.0 ± 0.0	
S. abony NTCC 6017	13.1 ± 0.1	10.0 ± 0.2
S. cerevisiae ATCC 9763	35.0 ± 0.0	28.0 ± 0.5
C. albicans ATCC 10231	28.3 ± 0.2	18.0 ± 0.2
A. niger	58.0 ± 0.5	38.5 ± 0.3

obtained by water distillation with a yield of 4.0%, and the physicochemical properties were measured (see **Table 1**). Olfactoric evaluations showed a high essential oil quality with a pleasant, characteristic dill odor dominated by a fresh (D-limonene) top note and a spicy (D-carvone) base note.

By means of GC-FID and GC-MS (achiral and chiral phases used), more than 40 volatiles could be identified as essential dill oil constituents of stored seeds (see **Table 2**) with D-carvone (50.1%) and D-limonene (44.1%) in high concentrations. In the essential oil of the original seeds, only carvone (71.0%) was found as the main compound. Beside these two monoterpenoids, other characteristic essential dill seed oil components, such as carveol, dihydrocarvones, α-phellandrene (main compound in essential dill leaf oils), and 3,9-epoxy-*p*-menth-1-ene (= dill ether), were identified in lower concentrations (trace up to 1.9%). Although D-limonene and D-carvone are responsible for the characteristic dill seed aroma, these minor concentrated volatiles have additional odor effects on the totally pleasant aroma impression of this essential *A. graveolens* oil sample, tested by GC sniffing analysis and correlation with aroma descriptions of single compounds published elsewhere (*32*−*36*).

In addition, antimicrobial testings with the essential dill oil of long-time stored seeds were done, using different strains of microorganisms (Gram (+) bacteria, Gram (−) bacteria, yeasts, and a mold) and results were obtained as follows: the antimicrobial activity of this essential oil is highest against the mold *A. niger*, high against the yeasts *S. cerevisiae* and *C. albicans* as well as the Gram (+) bacteria *S. aureus*, and somewhat weaker against the Gram (+) bacteria *B. subtilis*, *S. epidermidis*, and *B. pumilus*, as well as the Gram (−) bacteria *E. coli*, *S. abony*, and *P. aeruginosa* (see **Table 3**).

In conclusion, we can report that surprisingly the essential dill oil of long-time stored seeds (38 years under unstandardized conditions) possesses a high quality of aroma and composition. The monoterpenoids D-carvone and D-limonene (D form obtained from GC-FID and GC-MS analyses using chiral cyclodextrin phases) are the main compounds with concentrations of 50.1 and 44.1%, respectively. Antimicrobial testings showed high activities of this essential dill oil from Bulgaria especially against the mold *A. niger*, yeasts (*S. cerevisiae* and *C. albicans*), and some Gram (+) bacteria (*S. aureus* and *B. subtilis*). This result also certifies that the used, unstandardized storage method for dill seeds has no negative influence on the composition, aroma, and antimicrobial activity of the obtained essential oil, and therefore on its good quality, in a significant way. Applications of this essential oil of long-time stored *A. graveolens* seeds from Bulgaria are also possible in foods (e.g., flavoring of food products) and medicine (e.g., as disinfectant) without objection.

ACKNOWLEDGMENT

We acknowledge the olfactoric evaluations by Mr. V. Hausmann and Mr. W. Höppner, chief perfumers of Dragoco Co., Vienna, Austria.

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Received for review January 7, 2003. Revised manuscript received March 21, 2003. Accepted March 23, 2003.

JF030004Y