Food Chemistry 77 (2002) 75–80

Food Chemistry

www.elsevier.com/locate/foodchem

Cumin herb as a new source of essential oils and its response to foliar spray with some micro-elements

Salma A. El-Sawi[a,*], M.A. Mohamed[b]

[a]Pharmacognosy and Chemistry of Medicinal Plants Department, National Research Centre, Dokki (12311), Cairo, Egypt
[b]Cultivation and Production of Medicinal and Aromatic Plants Department, National Research Centre, Dokki (12311), Cairo, Egypt

Received 22 May 2001; received in revised form 21 August 2001; accepted 21 August 2001

Abstract

The effects of 50 mg/l levels of micronutrients (Zn and Mn), as single and combined treatments, on the growth, oil yield and oil constituents of cumin plants, were studied. Application of micronutrients had significant positive effects, in most cases, on growth measurements and chemical composition of cumin plants. A combined treatment of the two micronutrients gave the highest values in this respect. In the herb and seed oils, 21 constituents were identified, representing 90.2 and 95.6% of the total amounts, respectively. Eleven components were similar in both herb and seed oils. Cumin aldehyde was found as the main component at concentrations of 53.6% for seed oil and 40.5% for herb oil. Among the new identified components in the seed oil were perilla aldehyde, α-cis bergamotene, acoradiene and benzoic acid 4-(1-methylethyl). These components were found in the herb oil, as well. The oils of herb and seeds of cumin contained considerable amounts of oxygenated monoterpenes. Both oils were characterized by small amounts of monoterpenoid and sesquiterpene hydrocarbons. Qualitative and quantitative data indicated that oil production from cumin herb is a possibility.

Keywords: Cumin (*Cuminum cyminum*); Micronutrients; Zn; Mn; Growth; Herb; Seeds; Essential oil; GC–MS

1. Introduction

Cumin (*Cuminum cyminum* L) is an annual plant of the Umbelliferae family. This plant, which is one of the important spices in the world, is native to Egypt. Cumin is used as a condiment and as an ingredient in many food industries. Cumin seed oil is obtained by steam distillation and is used in perfumery. Shetty, Singhal, and Kulkarni (1994) reported that cumin seed oil had antimicrobial properties.

Composition of the cumin seed oil was extensively investigated many years ago. In 1981, Takahashi, Muraki and Yoshida reported that cumin oil contained mint sulfide as a trace constituent. Twelve years later, Anon (1993) and Shaath and Azzo (1993) reported that the main constituents of Egyptian cumin seed oil were cumin aldehyde, β-pinene, γ-terpinene, ρ-mentha-1,3-dien-7-al, ρ-mentha-1,4-dien-7-al and *p*-cymene. Composition of the cumin seed oil of Turkish origin was investigated by Baser, Kurkcuoglu, and Ozek (1992) and Borges and Pino (1993) who found that Turkish cumin seed oil was characterized by high amount of cumin aldehyde, *p*-mentha-1,3-dien-7-al, *p*-mentha-1,4-dien-7-al, γ-terpinene, *p*-cymene, β-pinene and perilla aldehyde.

Conditions of the arid soil of Egypt, with high pH, low organic matter content, irregular irrigation and compaction, reduce the availability of micronutrients (Zn, Mn, and Fe) to crop plants (El-Fouly, 1983). Application of micronutrients, as supplements to macroelements, has been reported to have significant effects on herb yield and oil contents of Ocimum, marjoram, mint and geranium (Sharma, Singh, and Tripathi, 1980; Wahab and Hornok, 1983). El-Sherbeny and Abou Zeid (1986) found that growth, volatile oil and carbohydrate contents of fennel were increased by foliar application of micronutrient fertilizers containing Cu, Zn, B and Mn. Also, Tarraf, El-Sayed, and Ibrahim (1994) found that application of micronutrients on to *Rosmarinus officinalis* had significant positive effects on growth, fresh and dry yields and oil production.

Until now, there have been no reports on composition of the herb oil of *C. cyminum*, which may serve as a

* Corresponding author.

0308-8146/02/$ - see front matter
PII: S0308-8146(01)00326-0

valuable source of cumin oil in Egypt. The present investigation is the first attempt to investigate the oil production and oil constituents of the cumin herb, compared with the seeds, under the conditions prevailing in Egypt. Also, this work studies the effects of foliar application of Mn and Zn, singly and in combination, on plant growth and herb and seed yield, as well as essential oil production and oil constituents.

2. Material and methods

2.1. General

This work was carried out at the Experimental Farm of the Cultivation and Production of Medicinal and aromatic Plants Department of the National Research Centre at Giza during the two successive seasons, 1999 and 2000. Before planting, chemical and physical properties of the farm soil were determined. The soil was loamy, having a physical composition as follows: 33.0% clay, 23.5% silt, 42.62% sand and 0.88% organic matter. Soil chemical analysis was as follows: pH = 8.2; E. C. (m mohs/cm) = 0.64; cations (meq/l): Ca^{+2} = 1.43, Mg^{+2} = 1.5, Na^{+} = 4.46, K^{+} = 1.74; anions (meq/l) : CO_3^{2-} = zero, HCO_3^{-} = 3.6, Cl^{-} = 2.0, SO_4^{2-} = 5.6 (according to Jackson, 1973); elements (ppm): Fe = 2.0, Mn = 3.8, Zn = 1.8, Cu = 0.1 (according to Lindsay and Norvell, 1978).

Seeds were spread in plots (2×3.5 m) on 8 November at the range of 15 kg/feddan (feddan = 4200 m^2). The lay-out of the experiments was a complete randomized block design of three replicates for each treatment. All agricultural practices were performed in the same manner, as is usually done in the cumin production areas in Egypt.

Micronutrients in the form of zinc sulfate and manganese sulfate were applied as foliar sprays, prepared with twice distilled water in the following treatments:-

1. Control (sprayed with distilled water);
2. 50 ppm Zn;
3. 50 ppm Mn; and
4. 50 ppm Zn + 50 ppm/l Mn.

The plants were sprayed three times, the first was 45 days after germination; the second and the third were 15 and 30 days after the first spraying. Number of plants per plot, weight of plant, plant height, number of branches and mean yield of air-dried seeds and herb per plot and per feddan were determined.

The essential oil of each treatment was extracted by hydrodistillation, according to Guenther (1961), and dehydrated over anhydrous sodium sulfate.

The results of the two growing seasons were statistically analyzed using the F-test according to Snedecor and Cochran (1967).

2.2. Gas chromatography (GC)

A FID Hewlett-Packard 5890 was used using a fused silica capillary SE54 (30 m×0.25 mm id.) column. Temperature program was: 2 min at 60 °C, 60–100 °C

Table 1
Effect of foliar spray with micronutrients on cumin growth (mean of two seasons)

Treatment in (cm)	Number of plants in 1 m^2	Dry wt. of plants in 1 m^2	Mean dry wt. of one plant (g)	Plant height (cm)	Number of branches per plant
Control	75	693.9	9.25	22.0	6.0
Zn (50 mg/l)	80	762.6	9.53	22.3	7.2
Mn (50 mg/l)	80	763.3	9.54	22.5	7.2
Zn + Mn (50 + 50 mg/l)	85	837.4	9.85	22.9	8.6
LSD 5%	6.3	97.8	N.S.	N.S.	0.9

Table 2
Effect of foliar spray with micronutrients on cumin herb and seed oil yields (mean of two seasons)

Treatment	Herb yield			Seeds yield			Oil (%)		Oil yield (kg/feddan)	
	Unit area (7 m^2) (kg)	One plant (g)	Feddan (kg)	Unit area (7 m^2) (kg)	One plant (g)	Feddan (kg)	Herb	Seeds	Herb	Seeds
Control	3.786	7.20	2163.6	1.071	2.04	612.0	0.30	1.2	6.49	7.34
Zn (50 mg/l)	4.183	7.47	2390.4	1.155	2.06	660.0	0.29	1.3	6.93	8.58
Mn (50 mg/l)	4.174	7.45	2385.2	1.169	2.09	668.0	0.32	1.4	7.63	9.35
Zn + Mn (50 + 50 mg/l)	4.611	7.75	2634.8	1.253	2.11	716.0	0.33	1.6	8.7	11.46
LSD 5%			168.5			42.5			1.1	1.2

(2 °C/min) and 100–250 °C (5 °C/min), then isothermal for 20 min; carrier gas was helium at a flow rate of 1.0 ml/min.

2.3. Gas chromatography–mass spectrometry (GC–MS)

A Hewlett Packard 5989A GC-MS system, equipped with library software, Wiley 138 and NBS75, was used. Capillary GC conditions, as above were employed for the fused silica capillary column SE54. Injection volume was 1.0 ul at 1:50 split. Significant MS operating parameters: ionization voltage 70 eV, scan mass range 40–350 u.

2.4. Identification of components

Compounds were identified by matching of their mass spectra with those recorded in the MS library and further confirmed by injecting the authentic samples of different compounds with the volatile oil and by comparison of the mass spectra with those of reference compounds or with published data.

3. Results and discussion

3.1. Effect of micronutrients spray on the growth parameters, yield and essential oil percentages of cumin herb and seeds

Table 1 shows the effects of foliar spray with micronutrient on cumin growth. All treatments had positive effects on the number of plants in one square meter, number of branches per plant and plant dry weight. Combination of Zn and Mn gave the best results. These results are in agreement with those obtained by Razin and Shalaby (1989) on *Ocimum* spp., El-Sherbeny, Hussien, and Mandour (1987) on fenugreek and Tarraf et al. (1994) on rosemary plant.

Table 2 demonstrates that no marked changes were obtained in the oil percentage due to foliar spray with micronutrients. The increase in the yield of essential oils per feddan is due to the higher yield of herbage and seeds. The highest increment resulted from combined Zn and Mn treatments. Similar positive effects of micronutrients on oil yield were reported by several investigators such as El-Sherbeny and Abou Zeid (1986) and Bojenk and Koloniec (1968) on peppermint and Nair, Nair, Chendrasekharan and Chimamma (1979) on Lemongrass.

3.2. Oil constituents of cumin herb and seeds

Table 3 lists the identified compounds in the essential oil obtained by hydrodistillation of cumin herb and seeds, according to their chemical groups. In both herb and seed oils, 21 constituents were identified, representing (90.2 and 95.6%) of the total amounts, respectively. The oils of herb and seeds of cumin contained considerable amounts of oxygenated monoterpenes. Sesquiterpenes were found only in the herb oil at low concentrations together with octanal and estragole.

Table 3
Comparison between oil constituents of cumin herb and seeds

Compound	Herb oil area (%)	Seed oil area (%)
Monoterpene hydrocarbons		
α-Pinene	–	1.27
Sabinene	–	0.26
β-Pinene	2.31	6.26
Myrcene	–	0.72
α-Phellandrene	–	0.75
3-Carene	–	0.81
α-Terpinene	2.70	0.95
ρ-Cymene	3.51	1.54
γ-Terpinene	1.70	1.06
Oxygenated monoterpenes		
Terpinene-4-ol	1.89	–
α-terpineol	2.22	0.84
Cumin aldehyde	40.54	53.55
Perilla aldehyde	3.14	1.13
Thymol	2.38	1.27
Cumin alcohol	–	2.10
Sesquiterpene hydrocarbons		
α-*cis* bergamotene	2.46	1.21
β-Caryophyllene	–	3.14
cis β-Farnesene	–	1.72
Acoradiene	7.64	11.46
Cuparene	2.09	–
Oxygenated sesquiterpenes		
Caryophyllene oxide	3.42	–
Carotol	0.67	–
Daucol	0.77	–
Acids and esters		
Propyl tiglate	0.34	–
Hydrocinnamyl acetate		2.34
Benzoic acid 4-(1 methylethyl)-	5.36	1.09
ρ-Anisyl acetate		2.11
Menth-8-ene-3-ol, acetate	3.36	–
Hexadecanoic acid	0.23	–
Others		
Octanal	0.23	–
Estragole	3.27	–
Total		
No. of compounds	21	21
Monoterpene hydrocarbons	10.22	13.62
Oxygenated monoterpenes	50.17	58.89
Sesquiterpene hydrocarbons	12.19	17.53
Oxygenated sesquiterpenes	4.68	–
Acids and esters	9.29	5.54
Others	3.5	–

Table 4
Effect of foliar spray with micronutrients on the constituents of cumin herb oil

Compound	Control	Zn (50 mg/l)	Mn (50 mg/l)	(Zn + Mn) (50 + 50 mg/l)
β-pinene	2.31	0.51	0.44	0.31
Octanal	0.23	0.21	0.20	0.22
α-Terpinene	2.70	2.92	2.88	2.91
P-Cymene	3.51	2.83	4.34	3.77
Propyl tiglate	0.34	1.19	1.76	1.77
γ-Terpinene	1.70	1.88	1.20	1.81
Terpinene-4-ol	1.89	1.62	1.58	1.66
α-terpineol	2.22	1.15	3.52	3.40
Estragole	3.27	1.08	2.56	2.58
Cumin aldehyde	40.54	46.47	47.22	41.83
Perilla aldehyde	3.14	1.44	1.81	3.44
Menth-8-ene-3-ol, acetate	3.36	1.96	2.48	3.67
Thymol	2.38	1.89	3.76	3.58
a-*cis* pergamotene	2.46	2.97	2.75	2.72
Acoradiene	7.64	7.73	7.65	8.33
Cuparene	2.09	1.03	1.01	3.29
Benzoic acid 4-(1-methylethyl)-	5.36	3.60	4.70	4.74
Caryophyllene oxide	3.42	2.45	3.50	3.69
Carotol	0.67	0.34	0.32	0.31
Daucol	0.77	0.35	0.31	0.29
Hexadecanoic acid	0.23	0.21	0.19	0.21
Total	90.23	83.83	94.18	94.53

Table 5
Effect of foliar spray with micronutrients on the constituents of cumin seed oil

Compound	Control	Zn (50 mg/l)	Mn (50 mg/l)	Zn + Mn (50 + 50 mg/l)
α-Pinene	1.27	0.73	0.22	0.45
Sabinene	0.26	0.55	0.19	0.20
β-Pinene	6.26	2.11	1.11	0.43
Myrcene	0.72	0.18	0.18	0.22
α-Phellandrene	0.75	0.20	0.23	0.24
3 Carene	0.81	0.57	0.40	0.41
α-Terpinene	0.95	0.43	0.32	0.40
P-Cymene	1.54	1.06	0.80	1.05
γ-Terpinene	1.06	0.66	0.45	0.65
α-Terpineol	0.84	0.72	0.42	0.57
Cumin aldehyde	53.55	55.78	68.83	58.60
Perilla aldehyde	1.13	0.51	0.54	0.40
Tymol	1.27	0.33	0.61	1.09
Cumin alcohol	2.10	0.94	1.20	1.56
Hydrocinnamyl acetate	2.34	1.81	2.61	0.54
a-cis pergamotene	1.21	0.47	0.47	1.69
Benzoic acid 4-(1methylethyl)-	1.09	1.55	0.41	0.70
P-Anisyl acetate	2.11	0.89	1.85	0.61
β-Caryophyllene	3.14	0.71	2.36	2.45
Trans β-Farnesene	1.72	2.74	0.92	3.54
Acoradiene	11.46	11.87	10.88	13.63
Total	95.58	84.81	95.00	89.43

Cumin aldehyde was found as the main component in both oils. The seed oil had a higher concentration of cumin aldehyde than the herb oil. This compound was found as the main constituent of seed oil from Egypt and Turkey (Baser, Kurkcuoglu, & Ozek, 1992; Shaath & Azzo, 1993). Cumin seeds in Egypt also contained large amounts of acoradiene and β-pinene, while the three main components of the herb oil were cumin aldehyde, acoradiene and 4-(1-methylethyl) benzoic acid. Among the new components identified by GC/MS in the seed oil from Egypt, were perilla aldehyde, α-cis bergamotene, acoradiene and 4-(1-methylethyl) benzoic acid.

However, the composition of the volatile oil obtained from the herb markedly differed from that of the seeds.

Eleven components out of 21 were similar in both herb and seed oils, while some differences were observed between the relative amounts of β-pinene, α-terpinene, *p*-cymene, α-terpineol, perilla aldehyde, thymol, α-cis bergamotene, acoradiene and 4-(1-methylethyl) benzoic acid in the herb and seed oils.

Until now there have been no reports on the composition of the herb oil of *C. cyminum*. Producing oil yield/feddan comparable to that of seed oil and being rich in cumin aldehyde, together with most of the major compounds found in cumin seed oil, cumin herb oil may serve as a valuable source of these compounds in Egypt.

3.3. Effects of microelements treatments on the oil constituents of cumin herb and seeds

Table 4 shows the differences in the oil constituents of cumin herb after the different microelements treatments. All treatments showed the same constituents. Application of the microelements spray increased the main constituents, such as cumin aldehyde, *p*-cymene, α-terpineol, thymol and acoradiene. On the other hand, spraying the cumin plant with microelements decreased other constituents, especially β-pinene. No marked differences between the relative percentages of the minor constituents of cumin herb oil, due to application of trace element treatments, were observed.

For seed oil, Table 5 indicates that application of the micronutrients spray gave the same trend as that in herb oil. Using the microelements spray increased the two major constituents, cumin aldehyde and acoradiene. A positive effect was also observed with propyl tiglate. Also, the application of micronutrient spray decreased the concentrations of some cumin seed oil compounds compared with the untreated plants. This effect was obvious with β-pinene.

Studies on the effects of nutrition on the constituents of the essential oil have been reported by several investigators. Among these were Maximoos (1985) on *Lathyrus odoratus* and Ibrahim (1989) on luisa plant. They found that the concentrations of different oil constituents were affected by the application of fertilizers. Essential oil biosynthesis is strongly influenced by several intrinsic (genotype, ontogeny) and extrinsic (environmental) factors (Lawrence, 1986). Among the latter, nutrients are important (Bars and Koster, 1981). Mn has important functions in plant metabolism, especially in chlorophyll synthesis, photosynthesis, nitrate reduction, amino acids and protein synthesis, activation of different enzymes and finally in photohormone regulation (Amberger, 1974). Zn is an essential micronutrient that acts either as a metal component of various enzymes or as a functional, structural or regulatory cofactor and is thus associated with saccharide metabolism, photosynthesis, and protein synthesis (Marschner, 1984). Carbon dioxide and glucose are precursors of monoterpene biosynthesis. Saccharides are also a source of energy and reducing power for terpenoid synthesis. CO_2 fixation, contents of primary metabolites and sucrose metabolism are closely linked with essential oil accumulation. As Zn is involved in photosynthesis and saccharide metabolism and as CO_2 and glucose are the most likely sources of carbon utilized in terpene biosynthesis, the role of Zn in influencing essential oil accumulation seems particularly important (Srivastava, Misra, & Sharma, 1997). The requirement of Zn for optimal growth and physiological processes (Misra, 1992), and critical Zn concentration for essential oil yield (Misra & Sharma, 1991) have been reported.

References

Amberger, A. (1974). Micronutrients dynamics in the soil and function in plant metabolism. *Proc. Egypt. Bot. Soc. Workshop* 1, (Cairo), 81–101.

Anon, A. (1993). Analytical methods of committee. Application of gas-liquid chromatography to analysis of essential oils Part XVI. Monography for five essential oils. *Analyst*, *118*, 1089–1098.

Bars, W., & Koster, J. (1981). Turnover and degradation of secondary (natural) products. In P. K. Stampf, & E. E. Conn (Eds.), *The biochemistry of plants (Vol. 7)* (pp. 35–84). New York: Academic Press.

Baser, K. H. C., Kurkcuoglu, M., & Ozek, T. (1992). Composition of the Turkish cumin seed oil. *J. Ess. Oil Res.*, *4*, 133–138.

Bojenk, E. S. O., & Koloniec, N. I. (1968). Effectiveness of micro-fertilizers in increasing the yield and quality of peppermint, onions and arvensis. *Tudy Vses, Naucissled Inst. Lekarstv. Rast*, *13*, 142.

Borges, P., & Pino, J. (1993). The isolation of volatile oil from cumin seeds by steam distillation. *Nahrung*, *2*, 123–126.

El-Fouly, M. M (1983). Micronutrients in arid and semiarid areas: level in soils and plants and needs for fertilizers with reference to Egypt. *Proc. 15th Coll. Int. Potash Inst.(Bern)*, p.163.

El-Sherbeny, S. E., & Abou Zeid, E. N. (1986). A preliminary study on the effect of foliar micro elements on growth and chemical constituents in *Foeniculum copillacum*. *Bull. NRC, Egypt*, *II*, 606.

El-Sherbeny, S. E., Hussein, M. S., & Mandour, M. S. (1987). A competitive study on the effect of some foliar fertilizers on Fenugreek plant. *Egypt J. Agron.*, *1–2*, 17.

Guenther, E. (1961). *The Essential Oils*, VIII. New York: Robert. E D. Von Nostrand comp., Inc.

Ibrahim, M. E. (1989). Physiological and chemical studies on luisa plants PhD thesis. Egypt: Faculty of Agric Science at Moshtohor, Zagzaig Univ.

Jackson, M. L. (1973). *Soil chemical analysis*. Engle-Wood Cliffs, USA: Prentice-Hall.

Lawrence, B. M. (1986). Essential oil production. A discussion of influencing factors. In T. H. Parliment, & R. Croteau (Eds.), *Biogeneration of aroma* (pp. 363–369). New York: American Chemical Society.

Lindsay, W. L., & Norvell, W. A. (1978). Development of DTPA soil test for zinc, iron, manganese and copper. *Soil Soc. Amer. J.*, *42*, 25–27.

Marschner, H. (1984). Function of mineral nutrients: micronutrients. In *Mineral nutrition of higher plants* (pp. 269–300). New York: Academic Press.

Maximoos, S. (1985). Physiological studied on *Lathyrus odoratus* plant. MSc thesis Fac. Agric. Zagazig Univ.

Misra, A. (1992). Effect of zinc stress in Japanese mint as related to growth, photosynthesis, chlorophyll content and secondary plant products—the monoterpenes. *Photosynthetica*, *26*, 225–2234.

Misra, A., & Sharma, S. (1991). Critical Zn concentration for essential oil yield and menthol concentration of Japanese mint. *Fertilizer Res.*, *29*, 261–265.

Nair, E. V. G., Nair, K., Chendrasekharan, K., & Chimamma, N. P. (1979). Field experimental with micronutrients on yield of grass and oil citral control of oil of East Indian Lemongrass Variety O. *D. 19 Indian Perfum*, *33*, 55.

Razin, A. M., & Shalaby, A. S. (1989). Response of *Ocimum basillicum* L. to the new fertilizer Agrispon. *Annals of Agric. Sc. Moshtohor*, *27*(2), 821–828.

Shaath, N. A., & Azzo, N. R. (1993). Essential oil of Egypt. In G. Charalambous (Ed.), *Food flavor ingredients and composition* (pp. 591–603). Amsterdam: Elsevier Sci. Pub.

Sharma, S. N., Singh, R., & Tripathi, S. (1980). Effect of NPK and micronutrients on herb, oil and menthol yield of Japanese mint. *Indian J. Agron*, *25*(2), 279–281.

Shetty, R. S., Singhal, R. S., & Kulkarni, P. R. (1994). *Antimicrobial properties of cumin. World J. microbiol. biotechnol.* Oxford, OX, UK. Published by Rapid Communications of Oxford Ltd. in association with UNESCO and in collaboration with the International Union of Microbiological Societies, c1990-. Mar (1994). 10(2), 232–233.

Snedecor, W. G., & Cochran, G. W. (1967). *Statistical methods* ((6th ed.)). The Iowa State Univ.

Srivastava, N. K., Misra, A., & Sharma, S. (1997). Effect of Zn deficiency on net photosynthetic rate, ^{14}C partitioning, and oil accumulation in leaves of peppermint. *Photosynthetica*, *33*(1), 71–79.

Takahashi, K., Muraki, S., & Yoshida, T. (1981). Synthesis and distribution of (−)-mint sulfide a novel sulfur-containing sesquiterpene. *Agro. Bio. Chem.*, *45*, 129–131.

Tarraf, S., El-Sayed, A. A., & Ibrahim, M. E. (1994). Effect of some micronutrients on Rosmarinus officinalis L. *Egypt. J. Physiol. Sci.*, *18*(1), 201–210.

Wahab, A. S. A., & Hornok, L. (1983). Effect of NPK fertilization on yield and oil content of *Ocimum basilicum. Kerteszeti Egypten Kozlemenye*, *45*(13), 66.