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Short Communication

The effects of drying on the chemical components of essential oils of *Calendula officinalis* L.

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Calendula officinalis **is a medicinal plant whose essential oils are used for various purposes. The oils were extracted by hydrodistillation from fresh leaves, dry leaves and fresh flowers of the herb yielding 0.06, 0.03 and 0.09%, respectively. The analysis of the oils by GC-MS revealed a total of 30, 21 and 24 compounds from the fresh leaves, dry leaves and the flowers in the same order. Sesquiterpenoids dominated the fresh leaves (59.5%) and flowers (26%), while the monoterpenes dominated the oil in the dry leaves (70.3%). T-muurolol (40.9%) predominated in the fresh leaf oil; α-thujene (19.2%) and δ-cadinene (11.8%) were also present in high quantities. Whereas, 1,8-cineole (29.4%), γ–terpenene (11.6%), δ-cadinene (9.0%), β–pinene (6.9%) and α-thujene (6.3%) were the major components in the dry leaf oil. In the fresh flower oil, α-thujene (15.9%), δ-cadinene (13.1%) and δ-cadinene (10.9%) were the major components. The significance of the effect of drying on essential oil composition of this plant is discussed.**

Key words: *Calendula officinalis*, essential oil, 1,8-cineole, T-muurolol.

INTRODUCTION

Calendula officinalis L is an aromatic herb that belongs to the Asteraceae family. While the biennial form grows wild in the Southern, Eastern and Central Europe (Van Wyk and Wink, 2004), the annual form is more widely cultivated. The composite flowers which are yellow and orange blossom in the spring-summer seasons (Gilman and Howe, 1999). The leaves and flowers of *C. officinalis* have a wide range of culinary usage in South Africa and because of their colour, aroma and flavour, they are used in food preparation to enhance taste and appearance (Marczal, 1987). They are also used in paint coating, cosmetic, nylon industries and for ornamental purposes (Muuse et al., 1992).

The essential oils of this herb are highly medicinal (Janke, 2004) with several therapeutic activities, such as anti-inflammatory, anti-tumorogenic (Jimenez-Medina et al., 2006) and cicatrizing (Hamburger et al*.,* 2003). In addition, the *in vitro* antimicrobial activities of its oils have been documented (Bassett et al., 1990; Sarrell et al., 2003). Extracts from the plant have been reported to show activity against HIV-1 replication (Kalvatchev et al., 1997). Genotoxic effect has also been reported for essential oils of *C. officinalis,* (Bakkali et al., 2005) and the plant have been known to exhibit antioxidant (Cetkovi et al., 2004) and wound healing properties (Lavagna et al., 2001).

Although there are several reports on the chemical composition of essential oils of this plant growing in different parts of the world (Miguel et al., 2004, Danielski et al., 2006) no such information is available on the wild *C. officinalis* growing in South Africa. This is the focus of an extensive study in our laboratory.

In this paper, we report the chemical composition of the essential oils of the vegetative and floral parts of *C. officinalis* growing in the Eastern Cape Province of South Africa. We also assess the effect of drying on the quality and quantity of oils from the leaves of the plant.

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MATERIALS AND METHODS

Plant collection and distillation of the essential oils

Fresh materials of *C. officinalis* were collected from the wild around the University of Fort Hare, Alice campus in the Eastern Cape Province of South Africa. (latitudes 30°00′–34°15′S and longitudes 22°45′–30°15′E) in September, 2005. A voucher specimen (OKOH/01) was deposited at the University herbarium.

The fresh plant materials were carefully separated into leaves and flowers. Some of the leaves were air dried at room temperature (18°C) for seven days. About 500, 200 and 250 g of the fresh leaves, dry leaves and fresh flowers respectively were hydro distilled separately for 3 h in an all-glass Clevenger apparatus in accordance with the British pharmacopoeia method (British pharmacopoeia, 1980).

GC-MS analyses and identification of components

The GC-MS analyses were carried out using Hewlett-Packard HP 5973 mass spectrometer interfaced with an HP-6890 gas chromatograph with an HP5 column. The following conditions were used: initial temperature 70°C, maximum temperature 325°C, equilibration time 3 min, ramp 4°C/min, final temperature 240°C; inlet: split less, initial temperature 220°C, pressure 8.27 psi, purge flow 30 ml/min, purge time 0.20 min, gas type helium; column: capillary, 30 m × 0.25 mm i.d., film thickness 0.25 µm, initial flow 0.7 ml/min, average velocity 32 cm/s; MS: EI method at 70 eV.

The components of the oils were identified by matching their mass spectra and retention indices with those of the Wiley 275 library (Wiley, New York) in the computer library and literature .The yield of each component was calculated per kg of the plant material, while the percentage composition was calculated from summation of the peak areas of the total oil composition. The whole experiment was replicated thrice.

RESULTS AND DISCUSSION

Pale yellow oils with yields of 0.06, 0.03 and 0.09% were obtained from the fresh leaves, dry leaves and fresh flowers of the plant, respectively. The oils gave a total of 30, 21 and 24 identified compounds, representing 91.7, 89.8 and 87.5% of the total oil composition from the fresh leaves, dry leaves and fresh flowers respectively (Table 1). Although the flowers had the greatest oil yield, the oil from the fresh leaves was richer in chemical constituents than the dry leaves and fresh flower. A total of 30 chemical constituents were identified from the fresh leaves oil, while 21 constituents were identified from the dry leaves. The result supports the observation by some workers who reported that there could be a 50-fold reduction in chemical composition when plant materials are dried (Fatemeh et al., 2006; Raghavan et al., 1997).

The fresh leaves oil was dominated by T-muurolol (40.9%), α-thujene (19.2%) and δ-cadinene (11.4%), while the dry leaves oil was found to be rich in 1, 8-cineole (29.4%), α-thujene (17.8%), β-pinene (6.9%), α-thujene (6.3%) and δ-cadinene (9.0%). On the other hand, the major components of the fresh flower oil were α-thujene

Table 1. Chemical composition of the essential oil from *Calendula officinalis* growing in the Eastern Cape Province of South Africa.

Compound	KI[b]	% Composition (oil)		
		Leaves		
		Fresh	Dry	Flowers
α- Thujene	908	19.2	17.8	26.9
α-Pinene	928	-	2.4	1.8
Sabinene	960	1.1	-	1.8
β-Pinene	969	0.6	6.9	-
Myrcene	971	-	-	1.1
Limonene	1020	0.8	-	-
1,8-Cineole	1022	-	29.4	1.7
Trans-β-ocinene	1033	0.2	-	-
γ-Terpenene	1049	0.4	-	0.7
δ-3-Carene	1050	-	0.3	-
Nonanal	1099	-	1.0	-
Terpenen -4-ol	1174	0.4	-	0.6
4-Methyl-3-cyclohexen-1-ol	1175	-	0.6	-
α-Terpeneol	1205	-	0.6	-
Bornyl acetate	1283	0.1	-	-
α-cubebene	1347	0.2	-	-
α-copaene	1376	0.3	0.2	0.2
α-Bourbonene	1385	0.3	0.2	-
β-cubebene	1389	0.4	0.2	0.5
α-Gurjunene	1409	0.6	-	0.6
Aromadendrene	1410	-	0.2	-
β-caryophyllene	1420	1.0	-	1.2
α-ylangene	1450	0.2	-	-
α-Humulene	1454	1.7	1.2	1.5
Epibicyclo sesquiphlandrene	1463	0.4	-	-
α-amorphene	1513	0.6	-	0.5
α-copaene	1376	-	-	2.7
Alloaromadendrene	1486	-	-	0.3
β-selinene	1486	0.5	-	-
Germacrene D	1481	1.1	0.6	2.8
α-Cubebene	1491	-	1.4	0.2
α-muurolene	1498	2.1	1.6	-
γ-Cadinene	1513	2.7	2.2	2.2
δ-Cadinene	1522	11.4	9.0	13.1
Cadina-1,4-diene	1531	0.5	-	0.4
α-cadinene	1537	0.6	0.4	0.4
Nerolidol	1559	-	-	0.9
Palustrol	1569	0.2	-	-
Calarene	1494	2.3	0.5	-
β-Endobourbonene	1575	0.6	-	0.5
Oplopenone	1609	0.3	-	-
T-muurolol	1659	40.9	13.1	24.9
Yield (%w/w)		0.06	0.03	0.09

[a]In order of elution.
[b]KI - Kovats retention indices on HP-5 (similar to DB 5).
[c]Correct isomer not identified.
Values are the mean of three replicates.

(26.9%), T-muurolol (24.9%) and δ-cardinene (13.1%). The inability of the GC-MS to detect the presence of 1,8-cineole in the fresh leaves and the quantity of the compound and its sudden appearance in the dry leaves (29.4%) is noteworthy. However changes in oil composition are known to be dependent on a number of factors including the species of plant (Mirjalili et al., 2007). The presence of 1,8-cineole in the dry leaf oil makes it superior to the fresh leaf oil due to the characteristic properties of the compound.

Generally, a lot of components were missing in the dried leaves oil as compared to the fresh leaves oil. The sesquiterpene hydrocarbons present in all the oils were α-humulene, germacrene D, δ-cadinene and δ-cadinene, while the monoterpene hydrocarbons were α-thujene, T-muurolol, the major component in the fresh leaves oil was also present in the other oils. T-muurolol is produced from the direct oxidation of α-muurolene. β-pinene present at 6.9% in the dry leaves occurred in minute amount in the fresh oil, and α-terpenene present at values of 6.9 and 11.6% respectively in the dry leaves oil, occurred in minute amounts in the fresh leaf oil. The changes in the regimes of volatile compounds during drying have been reported to depend on several factors such as drying method and change to species or family (Loughrin and Kasperbauer, 2003). The components of the essential oils that are lost in the dried leaves are those stored on or near the leaf surfaces (Moyler, 1994). However, Ibanez et al. (1999) observed no difference in the essential oil composition of fresh and dry rosemary plant. The results of this study have reinforced the fact that there are quantitative and qualitative differences in the essential oil components of fresh and dry plant materials.

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REFERENCES

Bakkali F, Averbeck S, Averbeck D, Zhiri A, Idaomar M (2005). Cytotoxicity and gene induction by some essential oils in the yeast *Saccharomyces cerevisiae*. Mutat. Res. 585: 1-13.

Bassett IB, Pannowitz DL, Barnetson RS (1990). A comparative study of tea-tree oil versus benzoylperoxide in the treatment of acne. Med. J. Austr. 153: 455-458.

British Pharmacopoeia (1980). H. M. S. Office. 2, London, pp. 109-110.

Cetkovi GS, Djilas SM, Canadanovi-Brunet JM, Tumbas VT (2004). Antioxidant properties of marigold extracts. Food. Res. Int., 37: 643-650.

Danielski L, Campos LMAS, Bresciani LFV, Hense H, Yunes RA, Ferreira SRS (2006). Marigold (*Calendula officinalis L.*) oleoresin: Solubility in SC-CO2 and composition profile. Chem. Eng. Proc. (in press).

Fatemeh S, Khadijeh A, Gholamreza BK (2006). Influence of drying and extraction methods on yield and chemical composition of the essential oil of *Satureja hortensis*. Food Chem. 99: 19-23.

Gilman EF, Howe T (1999). *Calendula officinalis*. Cooperative extension service. Institute of Food and Agricultural Sciences, University of Florida. Fact Sheet, FPS-87.

Hamburger M, Adler S, Baumann D, Forg A, Weinreich B (2003). Preparative purification of the major anti-inflammatory triterpenoid esters from marigold (*Calendula officinalis*). Fitoterapia, 74: 328-338.

Ibanez E, Oca A, de Murga G, Sebastian LS, Tabera J, Reglero G (1999). Supercritical fluid extraction and fractionation of different preprocessed rosemary plants. J. Agric. Food. Chem., 47: 1400-1404.

Janke R (2004). Farming a Few Acres of Herbs: Calendula, Kansas State University Agricultural Experiment Station and Cooperative Extension Service, MF-2610.

Jimenez-Medina E, Garcia-Lora A, Paco L, Algarra I, Collado A, Garrido F (2006). A new extract of the plant *Calendula officinalis* produces a dual *in vitro* effect: cytotoxic anti-tumor activity and lymphocyte activation. BMC Cancer, 6: 119.

Kalvatchev Z, Walder R, Garzaro D (1997). Anti-HIV activity of extracts from *Calendula officinalis* flowers. Biomed. Pharmacother. 51: 176-180.

Lavagna SM, Secci D, Chimenti P, Bonsignore L, Ottaviani A, Bizzarri B (2001). Efficacy of Hypericum and Calendula oils in the epithelial reconstruction of surgical wounds in childbirth with caesarean section. Il Farmaco, 56: 451-453.

Loughrin JH, Kasperbauer MJ (2003). Aroma Content of Fresh Basil (*Ocimum basilicum* L. Leaves as Affected by Light Reflected from Colored Mulches. J. Agric. Food. Chem., 51: 2272-2276.

Marczal G (1987). Data on the essential oil content and composition of *C. officinalis*. Herba Hung: 26(2-3): 179-89.

Miguel MG, Duarte F, Venancio F, Tavares R (2004). Composition of the essential oils from Portuguese *Thymus albicans* collected at different regions of *Ria Formosa* (Algarve). J. Essential Oil Res. pp. 308-311.

Mirjalili MH, Tabatabaei SMF, Hadian J, Ebrahimi SN, Sonboli A (2007). Phenological variation of the essential oil of Artemisia scoparia Waldst. et kit from Iran. J. Essential Oil Res. 19(44): 326-329.

Moyler DA (1994). Spices – recent advances in Charalambous (Ed.), Spices, herbs and edible fungi, Amsterdam: Elsevier., pp. 1-70.

Muuse BG, Cuperus FP, Derksen J (1992). Composition and physical properties of oils from new oilseed Crops. Indust. Crops Prod., 1: 57-65.

Raghavan B, Rao L, Singh M, Abraham K (1997). Effect of drying methods on the flavour quality of marjoram (*Origanum majorana* L.), Nahrung 41(3): 159-161.

Sarrell EM, Cohen HA, Kahan E (2003). Naturopathic treatment for ear pain in children. Pediatrics, 3(5): 574-579.

Van Wyk BE, Wink M (2004). Medicinal Plants of the World. Briza Publications, Pretoria.