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Chemical composition of essential oils of *Lantana camara* leaves and flowers from Cameroon and Madagascar

Martin Benoit Ngassoum,[1] Samuel Yonkeu,[1] Leopold Jirovetz,[2] Gerhard Buchbauer,[2]* Gerhard Schmaus[3] and Franz-Josef Hammerschmidt[3]

[1]University of Ngaoundere, BP 455, Ngaoundere, Cameroon
[2]Institute of Pharmaceutical Chemistry, University of Vienna, Althanstrasse 14, A-1090 Vienna, Austria
[3]Dragoco Gerberding & Co, D-37601 Holzminden, Germany

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ABSTRACT: The essential oils of leaves and flowers of *Lantana camara* (Verbenaceae) from Cameroon and Madagascar were analysed by GC–FID and GC–MS. The oils are characterized by a high percentage of sesquiterpenes. The major components in the oils from Cameroon are *ar*-curcumene (25%), β-caryophyllene (13%) and caryophyllene epoxide (7%), while the main components of the oil from Madagascar are davanone (15%) and β-caryophyllene (12%). The monoterpenes percentages are lower in the two essential oils and are represented by sabinene (1–9%), α-pinene (2–4%), 1,8-cineole (1–3%) and linalool (1–3%). A comparison with the composition of various essential oils of *L. camara* with different origin will also be given. Copyright © 1999 John Wiley & Sons, Ltd.

KEY WORDS: *Lantana camara*; Verbenaceae; essential oils; Cameroon; Madagascar; *ar*-curcumene; β-caryophyllene; davanone

Introduction

Lantana camara Linn. (Verbenaceae) is a straggling aromatic shrub, a native of tropical America, growing in Cameroon as an ornamental plant in the hedges of public and private gardens.[1] Infusions of the leaves provide a tea-like drink and are used as a bath against rheumatism and also as a treatment for coughs and colds.[2,3] It is also given to asthma patients to relieve dyspnoea and suffocation. The whole plant shows antimalarial activity, and a mixed infusion with *Ocimum* species is considered to show diaphoretic and antipyretic actions.[3,4]

Reviews on natural products from *L. camara* have been reported by Sharma:[5,6] the extracts from different parts of the plant are potential sources of triterpenoids, which are hepatotoxic, and of flavonoids, which exert antibacterial and antifungal activities. Avadhoot *et al.*[7,9] have tested the biological activities of essential oils from the seeds.

Essential oils from *L. camara* plant material from different origins have previously been investigated.[10–16] Recently Möllenbeck *et al.*[10] identified 19 compounds in *L. camara* leaf oil from Madagascar, with β-caryophyllene (19%) and δ-3-carene as the main components. Singh *et al.*[11] and Manavalan *et al.*[12] reported α-farnesene (29%), α-phellandrene (16%), longifolene (10%) and α-cedrene (8.6%) as main components of samples from India. For Salleh[13] and Ahmed *et al.*,[14] citral (16–22%), α-caryophyllene (6–13%), β-phellandrene (5–10%) and geraniol (4–11%) are the main components of the oil from flowers from Egypt. Mahmud *et al.*[15] analysed oil from flowers from Pakistan, and reported on the following main compounds: β-caryophyllene (19.8%), selinene (13.8%), 1,8-cineole (10.4%) and geranyl acetate (5.1%). Peyron *et al.*[16] identified 15 compounds in the oil of leaves and flowers from the Comores Islands. The oils show a high concentration of α-humulene (21.8%), β-caryophyllene (15%) and γ-terpinene (7.7%).

However, although the chemical constituents of the essential oils from plant material of *L. camara* from many countries have been reported, the essential oil from Cameroon has not been investigated until now. Also, no literature on headspace samples from plant parts could be found.

Therefore, the aim of this study was to analyse the volatiles of the oils responsible for the significant odour impression and to give information for a possible use in food or medicinal products. The investigation was

* Correspondence to: G. Buchbauer, Institute of Pharmaceutical Chemistry, University of Vienna, A-1090 Vienna, Austria. Tel: +43 1 31336 8610; fax: +43 1 31336 771.

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carried out on fresh and dried leaves and flowers from Madagascar and Cameroon.

Experimental

Plant Materials

Plant materials of *Lantana camara* were collected in the garden of the University of Ngaoundere (Adamaoua plateau of Cameroon) during the dry (March) and wet season (September). The species was identified and the voucher specimen was deposited at the National Herbarium of Yaounde (Reference No. 14711/SRFK).

Isolation of Volatiles Components

Fresh leaves and flowers were subjected to hydrodistillation for 4 h. The yield (v/w) of volatile oils was 0.06, 0.08 ml, respectively. The oils were dried over anhydrous sodium sulphate and stored at 4°C until analysed. The reference oil from Madagascar was obtained from Proimpex Agro Industries, Antananarivo.

The headspace of naturally dried leaves was produced by a dynamic method (trapping on charcoal-tubes from Dräger Co, Germany, which were integrated in a pumping-system of Brey Co, Germany) and eluted by 0.5 ml of dichloromethane.

Gas Chromatography

A GC-14A with FID and integrator C-R6A-Chromatopac (Shimadzu Co, Japan), respectively a GC-3700 with FID (Varian Co, Germany) and integrator C-R1B-Chromatopac (Shimadzu Co, Japan), were used for gas chromatographic analyses. Carrier gas, hydrogen; injector temperature 250°C; detector temperature 320°C; temperature programme, 40°C/5 min to 280°C/5 min with a heating rate of 6°C/min; columns, 60 m × 0.32 mm DB-WAX fused silica (film thickness, 0.50 μm; Restek Co, USA), 30 m × 0.32 mm bonded FSOT-RSL-200 fused silica column (film thickness, 0.25 μm; Biorad Co, Germany) and 30 m × 0.32 mm bonded Stabilwax fused silica column (film thickness, 0.50 μm; Restek Co, USA). Quantification has been performed by percentage peak area calculations and partial identification of single compounds by correlation of retention times with reference data.[17–21]

Gas Chromatography–Mass Spectrometry

Two systems were used for GC–MS analyses:

1. A GC-17A with QP5000 mass spectrometer (Shimadzu Co, Japan) and a data system Compaq-ProLinea (Compaq Co, USA; class 5k software), a GC-HP5890 with HP5970-MSD (Hewlett-Packard Co, USA) and data system Pentium-PC (Böhm Co, Austria; MSD-ChemStation software) and a GCQ (Finnigan-Spectronex, USA, Germany, Switzerland) with data system Gateway-2000-PS75-PC (Siemens-Nixdorf, Germany; GCQ software) were used. Carrier gas, helium; injector temperature, 250°C; interface heating, 300°C; ion source temperature, 200°C; EI-mode, 70 eV; mass range, 41–450 amu. Temperature programmes and columns: see section on GC. Mass spectra correlations with Wiley, NBS and NIST library spectra, respectively, and our own aromachemical libraries.
2. Hewlett-Packard HP5970 A GC–MS system. Conditions: 60 m × 0.25 mm i.d. DB-WAX column (df = 0.25 μm); temperature programme, 60–240°C at 4°C/min; injector temperature, 250°C; ion source temperature, 200°C; ionization energy, 70 eV; carrier gas, He, 1.2 ml/min; sample quantity, 2–3 μl of 10% solution in hexane; split, 1 : 30.

The identification of the constituents has been further proved by measurement of their Kováts indices (KI: calculated on the basis of the homologous series on *n*-alkanes; column: 60 m DB-WAX; see GC–MS 2 conditions, above).

Results and Discussion

The essential oils of the leaves and flowers of *L. camara* from Cameroon were obtained by steam-distillation and the headspace sample of each oil trapped by a dynamic method to obtain information on the volatiles effective in inhalation applications.

Three samples were olfactorically evaluated by professional perfumers and the results could be correlated to GC–MS data. The leaf essential oil exerts a green (hexenal-like), herbal and fatty odour, while the flower oil shows a fatty, floral irone- and ionone-like odour. Oils from natural dried leaves and flowers possess the following olfactory characteristics: floral in the direction of tagetes, herbal-like, dried camomile and tea-like in the direction of black tea (maté).

More than 105 compounds have been identified using GC–MS in the oils from Cameroon and reference essential oil of *L. camara* from Madagascar. Table 1 shows the chemical composition of two samples (60 m DB-WAX column used). In Table 2, we compared our data (30 m FSOT-RSL-200 column) with those from other authors,[10,11,13,16] but only compounds with a concentration higher than 1% have been quoted.

Table 1. Chemical composition of essential oils of *Lantana camara* from Cameroon and Madagascar

Compounds	RI (60 m DB-Wax)	Sample from Cameroon	Sample from Madagascar
Tricyclene	1013	–	tr
α-Pinene	1024	0.31	3.68
α-Fenchene	1062	–	tr
Camphene	1069	tr	1.99
β-Pinene	1113	0.56	2.59
Sabinene	**1123**	**0.15**	**9.02**
Verbene	1126	–	tr
3-δ-Carene	1152	–	2.26
Myrcene	1160	0.02	0.63
α-Phellandrene	1167	0.02	0.31
α-Terpinene	1190	tr	0.19
Limonene	1202	0.11	1.62
β-Phellandrene	1212	0.69	0.15
1,8-Cineole	1215	0.12	2.83
2-Pentylfuran	1227	–	tr
(*Z*)-β-Ocimene	1232	–	0.04
γ-Terpinene	1244	0.02	0.96
(*E*)-β-Ocimene	1249	0.04	1.11
3-Octanone	1254	–	tr
p-Cymene	1271	0.09	0.78
2-Methylbutyl butyrate	1281	–	0.02
Terpinolene	1285	tr	0.29
Tridecane	1300	tr	–
1-Octen-3-one	1310	tr	–
(*Z*)-3-Hexenyl acetate	1324	–	tr
5-Hepten-2-one, 6-methyl-	1335	0.01	–
Hexyl isobutyrate	1342	–	tr
(*Z*)-3-Hexenol	1379	tr	0.01
3-Octanol	1389	0.02	0.04
Nonanal	1393	tr	–
Tetradecane	1400	tr	–
Hexyl butyrate	1415	–	tr
4-Isopropenyl toluene	1437	–	0.01
1-Octen-3-ol	1444	0.28	0.14
Acetic acid	1460	tr	–
α-Cubebene	1464	0.11	0.16
trans-Sabinenhydrate	1464	–	tr
δ-Elemene	1476		0.04
Pentadecane	1500	tr	tr
Copaene	1502	0.16	0.44
Camphor	1526	–	0.54
Bourbonene	1531	tr	0.08
Linalool	1540	0.28	3.43
β-Cubebene	1547	0.21	1.01
cis-Sabinenhydrate	1549	–	0.01
Octanol	1552	tr	–
Linalyl acetate	1555	0.11	–
Sesquithujene	1560	1.06	0.01
α-Cedrene	1582	0.57	–
β-Ylangene	1586	–	0.19
β-Elemene	1598	1.26	0.89
Terpinen-4-ol	1605	0.27	tr
β-Caryophyllene	**1612**	**13.26**	**11.98**
Calarene	1617	0.49	–
Sesquisabinene*	1648	0.62	–
Aromadedrene	1659	0.34	0.24
(*E*)-β-Farnesene	1664	0.86	1.01
α-Humulene	1682	1.38	6.17
γ-Curcumene	1695	2.21	0.51
γ-Muurolene	1698	0.19	0.84
α-Terpineol	1698	–	tr
Borneol	1704	0.34	0.41
Germacrene-D	1721	0.91	1.35
Zingiberene	1724	2.75	0.02
β-Bisabolene	1731	0.67	2.98
α-Muurolene	1733	0.45	1.21
β-Selinene	1735	–	tr
α-Selinene	1737	–	0.84
β-Curcumene	1744	1.42	–

Table continued over page

Table 1. Continued

Compounds	RI (60 m DB-Wax)	Sample from Cameroon	Sample from Madagascar
Bicyclogermacrene	1747	–	2.59
Geranyl acetate	1753	0.22	–
γ-Bisabolene	1762	tr	–
δ-Cadinene	1765	0.34	0.87
γ-Cadinene	1770	0.11	0.09
***ar*-Curcumene**	**1780**	**24.69**	**0.78**
Methyl salicylate	1783	–	tr
Cadina-1,4-diene	1793	–	0.11
Nerol	1794	0.11	–
α-Cadinene	1803	tr	0.03
β-Damascenone	1827	0.12	–
Geraniol	1840	0.46	–
Calamenene	1842	tr	0.21
epi-Cubebol	1895	0.17	0.82
Davanaether I**	1914	–	0.05
Davanaether II**	1921	–	0.03
β-Ionone	1947	tr	–
Cubebol	1947	0.39	1.63
Caryophyllene epoxide I	1993	1.51	0.25
Caryophyllene epoxide II	**2004**	**7.06**	**1.19**
Nerolidol	2031	0.25	2.28
Davanone	**2040**	**–**	**15.94**
Humulene epoxide	2058	0.68	1.94
Cubenol	2071	–	tr
Unknown	2073	–	1.94
epi-Cubenol	2078	–	0.41
Viridiflorol	2096	–	0.41
Zingiberenol	2113	1.81	–
Spathulenol	2129	1.48	0.68
β-Bisabolol	2151	0.19	–
Eugenol	2158	0.28	–
T-Cadinol	2174	0.99	0.49
T-Muurolol	2191	0.18	0.11
δ-Cadinol	2202	–	0.37
α-Bisabolol	2213	0.28	–
α-Cadinol	2233	–	0.34
Phytol	–	0.96	0.15
Xanthorrhizol	–	0.24	–

*See reference (21).
**Unknown stereochemistry.
tr = trace.

Samples from Cameroon

The main components in the oil sample from leaves and flowers (Table 1) are *ar*-curcumene (24.69%), β-caryophyllene (13.26%) and caryophyllene epoxide II (7.06%), zingiberene (2.75%), γ-curcumene (2.21%), zingiberenol (1.8%), caryophyllene epoxide I (1.5%) and spathulenol (1.5%). This sample contains many components (more than 60) with a concentration lower than 2%.

In comparison the oil from fresh leaves (see Table 2) is dominated by the sesquiterpenes, α-zingiberene (18.8%), *ar*-curcumene (15.6%), β-caryophyllene (11.3%) and nerolidol (11.3%). The monoterpenes are represented mainly by sabinene (7.0%), α-pinene (5.0%) and β-pinene (3.9%). Oxygenated monoterpenes are 1,8-cineole (1.7%) and linalool (1.3%).

The oil from the flowers contains also a high quantity of sesquiterpenes dominated by *ar*-curcumene (27.1%) and β-caryophyllene (8.1%). The oxygenated sesquiterpenes are nerolidol (13.3%), spathulenol (5.5%), caryophyllene oxide (2.5%) and T-cadinol (2.5%). The monoterpenes group are represented mainly by limonene (1.6%), β-myrcene (1.0%), 1,8-cineole (0.6%) and linalool (0.3%).

The headspace sample from the dried leaves contains also high quantities of sesquiterpenes, *ar*-curcumene (26.6%), α-zingiberene (11.4%) and β-caryophyllene (19.9%). The oxygenated sesquiterpenes are represented by nerolidol (2.5%) and caryophyllene epoxide II (2.8%). The monoterpene hydrocarbons are represented by sabinene (2.9%), α-pinene (2.5%) and β-pinene (2.5%) and limonene (2.6%), the oxygenated monoterpenes by 1,8-cineole (2.6%) and linalool (0.7%).

Samples from Madagascar

The main components in the sample from Madagascar are davanone (15.94%), β-caryophyllene (11.98%),

Table 2. Comparative study of the chemical composition of essential oils of *Lantana camara* from different origins (Cameroon, Madagascar, Egypt, India and the Comores Islands)

Compounds	Leaves and flowers from Cameroon	Leaves from Cameroon	Flowers from Cameroon	Headspace of leaves from Cameroon	Leaves and flowers from Madagascar	Data from Madagascar[10]	Data from India[11]	Data from Egypt[13]	Data from Comores[16]
α-Pinene	0.3	5.0	0.60	2.5	3.7	3.5	2.0	4.0	1.4
Camphene	tr	0.3	0.05	–	2.0	1.3		2.0	0.1
β-Pinene	0.6	3.9	0.20	2.5	2.6	2.8	0.5	3.0	1.0
Sabinene	0.1	7.0	0.40	2.9	9.0	–	–	–	0.9
3-δ-Carene	–	–	0.05	0.1	2.3	10.4	tr	–	–
Myrcene	tr	0.2	1.00	0.2	0.6	–	–	0.5	0.9
α-Phellandrene	tr	tr	0.05	–	0.3	–	14.9	tr	1.6
Limonene	0.1	1.3	1.50	2.6	1.6	1.8	1.3	1.0	1.9
β-Phellandrene	0.7	–	–	–	0.1	–	–	8.0	–
1,8-Cineole	0.1	1.7	0.60	2.6	2.8	2.7		5.0	1.0
(*Z*)-β-Ocimene	–	1.3	0.05	–	tr	tr	3.1	–	–
γ-Terpinene	tr	0.3	0.05	–	1.0	–	–	2.0	7.7
(*E*)-β-Ocimene	tr	–	–	–	1.1	–	–	–	–
p-Cymene	0.1	–	0.20	–	0.8	0.7	0.7	0.4	3.5
Terpinolene	tr	–	–	–	0.3	–		1.5	
α-Copaene	–	0.5	0.60	1	0.4	1.4	1.1	–	2.5
Bourbonene	tr	–	–	–	0.1	–	–	–	–
Linalool	0.3	1.3	0.30	0.7	3.4	2.7		1.0	0.4
β-Cubebene	0.2	0.2	0.20	–	1.0	–	–	–	–
Sesquithujene	1.1	–	–	–	0.0	–	–	–	–
α-Cedrene	0.6	0.1	0.05	0.5	–	–	8.6	–	–
β-Elemene	1.3	1.0	1.20	2.6	0.9	–	–	–	–
β-Caryophyllene	13.3	11.3	8.10	19.9	12.0	18.8	7.1	1.0	15.0
Aromadedrene	0.3	0.5	–	–	0.2	3.5	–	–	–
(*E*)-β-Farnesene	0.9	0.6	0.90	0.5	1.0	–	2.4	–	–
α-Humulene	1.4	0.5	0.70	–	6.2	–	–	11.0	21.8
γ-Curcumene	2.2	–	–	–	0.5	–	–	–	–
Germacrene-D	0.9	1.0	0.40	0.3	1.3	–	–	–	–
Zingiberene	2.7	18.8	1.00	11.4	tr	–	–	–	–
β-Bisabolene	0.7	4.0	0.90	1.0	3.0	3.2	–	–	–
α-Muurolene	0.4	1.0	0.60	1.0	1.2	–	–	–	–
β-Curcumene	1.4	–	–	–	–	–	–	–	–
Bicyclogermacrene	–	–	–	–	2.6	–	–	–	–
δ-Cadinene	0.3	1.0	0.50	3.3	0.9	–	–	–	–
ar-Curcumene	24.7	15.5	27.10	26.6	0.8	–	–	–	–
Geraniol	0.5	–	0.05	–	–	–	0.3	11.0	–
Cubebol	0.4	–	–	–	1.5	–	–	–	–
Caryophyllene epoxide I (2)	1.5	–	0.20	0.5	0.2	1.8	–	–	–
Caryophyllene epoxide II (2)	7.1	0.4	2.50	2.8	1.2	–	–	–	–
Nerolidol	0.2	11.3	13.30	2.5	2.3	–	–	–	–
Davanone	–	–	–	–	15.9	–	–	–	–
Humulene epoxide	0.7	–	–	–	1.9	–	–	–	–
Zingiberenol	1.8	–	–	-	–	–	–	–	–
Spathulenol	1.5	0.3	5.50	0.1	0.7	–	–	–	–
T-Cadinol	1.0	0.4	2.50	0.2	0.5	–	–	–	–
T-Muurolol	0.2	0.4	1.40	0.2	0.1	–	–	–	–
α-Bisabolol	0.3	–	1.80	–	–	–	–	–	–
α-Farnesene	–	–	–	–	–	–	28.8	–	–
Longifolene	–	–	–	–	–	–	9.9	–	–
α-Bisabolene	–	–	–	–	–	–	1.3	–	–
β-Himachalene	–	–	–	–	–	–	1.8	–	–
Nonyl benzene	–	–	–	–	–	–	1.3	–	–
β-Cedrene	–	–	–	–	–	–	3.5	–	–
Dipentene	–	–	–	–	–	–	–	1.5	–
(+)-Citronellol	–	–	–	–	–	–	–	2.0	–
Citral	–	–	–	–	–	–	–	22.0	–

sabinene (9.02%) and α-humulene (6.17%). Many other dominating compounds in concentrations higher than 2% have been found in this sample. Monoterpene derivatives are represented by α-pinene (3.68%), linalool (3.43%), 1,8-cineole (2.83%), β-pinene (2.59%) and δ-3-carene (2.26%). Sesquiterpene derivatives are β-bisabolene (2.98%), bicyclogermacrene (2.59%), zingiberene (2.75%), nerolidol (2.28%), γ-curcumene (2.21%) and humulene epoxide (1.94%).

Davanone was formerly identified as a constituent of *Lantana* oil by Pieribattesti *et al.*[22] Davanaether I (0.05%) and davanaether II (0.05%) have been identified by us for the first time in an essential oil of *L. camara*. We have also identified other oxygenated sesquiterpenes which have not been reported until now, namely humulene epoxide (1.9%), epi-cubebol (0.8%), cubebol (1.6%), epi-cubenol (0.4%), T-muurolol (0.5%), viridiflorol (0.4%) and zingiberenol (0.4%).

Comparative studies with some essential oils of *L. camara* oil with various origins

We were able to compare also the samples from various origins, namely Cameroon, Madagascar, Egypt, India and the Comores Islands, as shown in Table 2. The data of the two samples from Madagascar show that β-caryophyllene (12% and 19%, respectively) is the main component in the sesquiterpene hydrocarbon group. Davanone has not been identified by Möllenbeck *et al.*;[10] the sample studied by these authors contains δ-3-carene (10.4%) as the main monoterpene, while our sample contains sabinene as the main component.

Essential oils from India[11] contain α-farnesene (28.8%) as the main sesquiterpene while the monoterpene group are dominated by α-phellandrene (14.9%). The essential oils of *L. camara* from the Comores Islands[16] contain α-humulene (21.6%) and β-caryophyllene (15%) as well as γ-terpinene (8%). Essential oils from Egypt[13] are dominated by α-humulene (11%) as the main sesquiterpene and by β-phellandrene (8%) as the main monoterpene. *Ar*-curcumene (16–27%) and β-caryophyllene (13–18%) are the main sesquiterpenes of the essential oils from Cameroon. Sabinene (3–7%) was found to be the dominating monoterpene.

From these observations we reach the following conclusions: the oils from *L. camara* materials are a rich source of sesquiterpene hydrocarbons. Proportions of sesquiterpene components differ, depending on the origin and on the part of the plant used, as dominating sesquiterpene components *ar*-curcumene, α-humulene, α-farnesene and β-caryophyllene could be found. Several oxygenated sesquiterpenes have been clearly identified in *L. camara* essential oils for the first time. The sesquiterpene composition of samples from Cameroon is drastically different from that from Madagascar. These data are possibly useful for differentiating *Lantana* oils from Madagascar and the African continent, but further samples must be analysed for unequivocal proof of the occurrence of different *Lantana* chemotypes.

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