

## Chemical composition of the leaf essential oils of *Murraya koenigii* (L.) Spreng and *Murraya paniculata* (L.) Jack

**Jasim Uddin Chowdhury, Md. Nazrul Islam Bhuiyan and Mohammed Yusuf**

*BCSIR Laboratories, Chittagong Cantonment, Chittagong 4220, Bangladesh.*

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### Correspondence:

MNIB  
e-mail: nazrul119@yahoo.com

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### Abstract

The chemical composition of the leaf oils of *Murraya koenigii* (L.) Spreng and *M. paniculata* (L.) Jack from Bangladesh was studied by gas chromatography mass spectroscopy (GC-MS). *M. koenigii* oil contained 39 compounds of which the major is 3-carene (54.2%) followed by caryophyllene (9.5%). Oil of *M. paniculata* contained 58 compounds of which the major are caryophyllene oxide (16.6%),  $\beta$ -caryophyllene (11.8%), spathulenol (10.2%),  $\beta$ -elemene (8.9%), germacrene D (6.9%) and cyclooctene, 4-methylene-6-(1-propenylidene) (6.4%). The compositions of both oils varied qualitatively and quantitatively.

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### Introduction

A systematic study on the medicinal and aromatic plants is being carried out in Bangladesh. Among these plants one finds *Murraya koenigii* (L.) Spreng and *M. paniculata* (L.) Jack of the Rutaceae family. *Murraya koenigii* (L.) Spreng is a small strong smelling perennial shrub or small tree commonly found in forests as undergrowth, cultivated in India for its aromatic leaves and for ornament. Leaves are used as a condiment in the preparation of curry powder, pickle, chutney, sausages and seasonings (Anonymous, 1962; Hiremath et al, 1998). The flavor and fragrance of leaves retain even after drying (Omankutty Amma et al. 1984). Leaves relieve nausea, indigestion, vomiting; eaten as a cure for diarrhea and dysentery (Anon. 1962; Ghani, 2003). 1996). Essential oil composition of

leaves has been studied by various workers. The major constituents responsible for the aroma and flavour have been reported as pinene, sabinene, caryophyllene, cadinol and cadinene (Anonymous, 1962; Nigam and Purohit, 1961; Prakash and Natarajan, 1974; Macleod and Pieries, 1982; Hiremath et al, 1998). Raina et al. (2002) reported four genetically divers chemotypes (1)  $\beta$ -pinene (70%),  $\beta$ -caryophyllene (6.5%) and  $\alpha$ -pinene (5.4%), (2)  $\alpha$ -pinene (65.7%),  $\beta$ -pinene (13.4%) and  $\beta$ -phellandrene (7.4%), (3)  $\beta$ -caryophyllene (53.9%), aromadendrene (10.7%) and  $\alpha$ -selinene (6.3%), (4)  $\beta$ -phellandrene (30.2%),  $\beta$ -caryophyllene (24.2%),  $\alpha$ -pinene (15%), (E)- $\beta$ -Ocimene (5%) and aromadendrene (4.5%) as major constituents from various parts of India. Walde et al. (2005) reported

$\alpha$ -pinene (52%) and cis- $\beta$ -ocimene (34%) as major constituents in Hyderabad plants.

*M. paniculata* (L) Jack. syn. *Chalcas paniculata* L. *Chalcas exotica* (L.) Millsp. is a very variable evergreen shrubby plant with small flowers and small oblong fruits and hard wood. It is commonly found in gardens as an ornamental plant in the country. Leaves are stimulant and astringent and are used in the treatment of diarrhea, dysentery and diseases of teeth and gum; useful against rheumatism, coughs and hysteria (Anonymous, 1962; Chopra et al., 1956; Ghani, 2003). The essential oils showed significant anti-inflammatory and analgesic activities (Dash et al., 2004). Sawangiaroen et al. (2006) reported, it showed anti-amoebic activities. The leaves and other tissues have both stimulant and astringent properties and are used to treat diarrhea, dysentery, cuts, joint pain, body aches (Parrotta, 2001), venereal disease (Kinoshita and Firman, 1996), and as an abortive (Xiao and Wang, 1991). In addition to essential oils, tissues of orange jasmine contain the indole alkaloid yuehchukene (Xiao and Wang, 1991) and at least eight highly oxygenated flavones (Kinoshita and Firman, 1996), leaves yield oil, which contains sesquiterpenes (1-cadinene), a sesquiterpene alcohol and methyl anthranilate (Anonymous, 1962; Chopra et al., 1956). Nureni et al. (2004) reported, the principal constituents of the leaf oil were  $\beta$ -cyclocitral (22.9%), methylsalicylate (22.4%), trans-nerolidol (11.7%),  $\alpha$ -cubebene (7.9%), (-)-cubenol (6.8%),  $\beta$ -cubebene (5.8%) and isogermacrene (5.7%). The most prominent compounds were  $\beta$ -caryophyllene (24.1%), with lesser amounts of germacrene D (11.9%) and bicyclogermacrene (11.8%). Some reports of the chemical composition of the leaf oil of *M. paniculata* of Asian or Australian origin have appeared (Garg and Nigam, 1970; Qiang et al., 1988; Brophy et al., 1994). In our study the abundance of 3-carene was the highest. It has various medicinal uses (Jeong et al., 2007; Lastbom et al. 1998). On the other hand, *M. paniculata* contained abundance of caryophyllene oxide which has antifungal activities (Yang et al. 1999). So the present study deals with the investigation of the chemical components in oils obtained from leaf of *M. koenigii* and *M. paniculata* grown in Bangladesh.

## Materials and Methods

**Plant material:** Fresh leaves of *M. koenigii* and *M. paniculata* were collected from the plants grown in the campus of BCSIR Laboratory, Chittagong during June 2007. Two-voucher specimen (Y-325) was deposited in the herbarium of BCSIR Laboratory, Chittagong.

**Extraction of essential oil:** Leaves were harvested and air-dried for about one week. The oils were obtained by hydrodistillation for 4 hour in a Clevenger-type apparatus. The oil yields (calculated per weight of dried material) were 0.5% for *M. paniculata* and 1.0% for *M. koenigii*. The oil samples were stored at 0°C in air-tight containers after drying them over anhydrous sodium sulfate for gas chromatography mass spectroscopy (GC-MS) analyses.

**GC-MS analysis:** The essential oil from leaves of *M. koenigii* and *M. paniculata* were analyzed by GC-MS electron impact ionization (EI) method on GC-17A gas chromatograph (Shimadzu) coupled to a GC-MS QP 5050A mass spectrometer (Shimadzu); fused silica capillary column (30 m x 2.5 mm; 0.25  $\mu$ m film thickness), coated with DB-1 (J&W); column temperature 100°C (2 min) to 250°C at the rate of 3°C/min; carrier gas, helium at constant pressure of 90Kpa. Acquisition parameters full scan; scan range 40-350 amu.

**Identification of the compounds:** Compound identification was done by comparing the NIST library data of the peaks with those reported in literature, mass spectra of the peaks with literature data. Percentage composition was computed from GC peak areas on BP-I column without applying correction factors.

## Results and Discussion

The essential oils from the leaves of *M. koenigii* and *M. paniculata* were analyzed by GC-MS presented in Table I. *M. koenigii* oil contains 39 compounds of which the major is 3-carene (54.22%) followed by caryophyllene (9.49%). Other notable compounds in the *M. koenigii* oil are  $\alpha$ -thujene (1.47%), allyl(methoxy)dimethylsilane (2.58%),  $\beta$ -myrcene (3.2%),  $\alpha$ -terpinene (2.39%),

$\gamma$ -terpinene (2.7%), cis-sabinenehydrate (1.46%), 4-terpineol (2.8%),  $\beta$ -elemene (1.92%),  $\alpha$ -caryophyllene (2.81%),  $\gamma$ -elemene (1.96%), caryophyllene oxide (1.02%) and 3-phenylbutyrophe-none

(1.15%). Oil of *M. paniculata* contains 58 compounds of which the major are caryophyllene oxide (16.63%),  $\beta$ -caryophyllene (11.81%), spathu-

Table I: Constituents of leaf essential oil from *M. koenigii* and *M. paniculata*

Compounds from <i>M. koenigii</i>		%	Compounds from <i>M. paniculata</i>		%
1.	$\alpha$ -Thujene	1.47		Sabinene	0.12
2.	3-Carene	54.22		3-Hexen-1-ol, formate	0.09
3.	Camphene	0.09		Limonene	0.34
4.	Allyl(methoxy)dimethylsilane	2.58		Linalool	0.13
5.	$\beta$ -Myrcene	3.20		Cyclohexene,3,4-diethenyl-3-methyl	0.58
6.	$\alpha$ -Phellandrene	0.07		Cyclohexene,5,6-diethenyl-3-methyl	3.30
7.	$\alpha$ -Terpinene	2.39		Azulene	0.08
8.	m-Cymene	0.30		Ocimene	0.06
9.	Limonene	0.84		cis-3-Hexenyl valerate	0.20
10.	$\beta$ -Phellandrene	0.49		2-Cycohexen-1-one, 2-methyl-5-(1-methylethenyl)	0.09
11.	Eucalyptol	0.11		1H-Imidazole -4-methanol,5-methyl	0.06
12.	(E)-Ocimene	0.23		$\delta$ -Elemene	3.57
13.	$\gamma$ -Terpinene	2.70		$\alpha$ -Cubebene	2.96
14.	cis-Sabinenehydrate	1.46		Germacrene D	6.95
15.	Linalool	0.19		3,9-Dodecadiene	0.09
16.	4-Terpineol	2.80		$\beta$ -Caryophyllene	11.81
17.	Naphthalene	0.09		Caryophyllene oxide	16.63
18.	Terpinyl acetate	0.14		Cyclooctene, 4-methylene-6-(1-propenylidene)	6.37
19.	cis-Piperitol	0.13		Retinal	1.00
20.	Isobornyl acetate	0.07		$\alpha$ -Caryophyllene	3.13
21.	$\delta$ -Elemene	0.04		$\beta$ -Humulene	0.17
22.	Neryl propionate	0.04		Copaene	2.33
23.	1-Chloroheptacosane	0.06		$\gamma$ -Elemene	0.25
24.	$\beta$ -Elemene	1.92		Cubenol	2.36
25.	Caryophyllene	9.49		$\alpha$ -Bulnesene	0.12
26.	$\alpha$ -Caryophyllene	2.81		Calamenene	0.27
27.	Eudesma-4(14),11-diene	0.18		3-Tetradecyonic acid	0.24
28.	$\gamma$ -Elemene	1.96		Lanceol, cis	0.30
29.	$\delta$ -Cadinene	0.10		$\beta$ -Vatirenine	0.14
30.	Nerolidyl acetate	0.15		$\beta$ -Elemene	8.94
31.	Caryophyllene oxide	1.02		Nerolidyl acetate	1.20
32.	12-Oxabicyclo[9.1.0]dodeca-3,7-diene,	0.07		Alloaromadendrene oxide	0.30

33.	1,4-Methanoazulen-9-ol, decahydro- 1,5,5,8a-tetramethyl	0.06	Spathulenol	10.21
34.	Cubenol	0.08	D-Verbenone	0.58
35.	2(1H)-Naphthalenone, 4a,5,6,7,8,8a- hexahydro-4a,8a-dimethyl-	0.05	Pyrimidine-2(1H) thione, 3,4-dihydro -6-methyl, 4- phenyl	0.47
36.	$\alpha$ -Cadinol	0.08	3-Carene	0.88
37.	Juniper camphor	0.45	12-Oxabicyclo(9,1,0) dodeca-3,7-diene,1,5,5,8 - tetramethyl	2.07
38.	3-Phenylbutyrophenone	1.15	Globulol	0.15
39.	Phytol	0.93	Eremophilene	1.54
40.			2(1H) Naphthalenone, 4a,5,6,7,8,8a-hexahydro, 4a,8a-dimethyl	0.30
41.			Tau -Muurolol	0.90
42.			Ledol	2.20
43.			Aromadendrene oxide	1.47
44.			$\alpha$ -Calaconene	0.29
45.			Longifolenealdehyde	0.61
46.			11-Hexadecyn-1-ol	0.21
47.			Cycloisolongifolene, 8-hydroxy-endo	0.28
48.			Longipinocarveol, trans	0.15
49.			Carveol	0.08
50.			1-Cyclohexene-1-ethanol, 2,6,6-trimethyl	0.12
51.			1-Methyl verbenol	0.21
52.			Cyclopropane, 1-bromo-2,2,3,3-tetramethyl-1-prop- 1-ynyl	0.18
53.			Corymbolone	0.13
54.			2(4a,8-Dimethyl-1, 3,3,4,4a,5,6,7-octahydro- naphthalene-2-yl) -prop-2-en-1-ol	0.11
55.			Ledene alcohol	0.65
56.			Aristolene oxide	0.34
57.			6-Isopropenyl-4, 8a-dimethyl 1,2,3,5,6,7,8,8a, octahydro naphthalen-2-ol	0.97
58.			Longifolene-[12]-epoxide	0.08

lenol (10.21%),  $\beta$ -elemene (8.94%), germacrene D (6.95%) and cyclooctene, 4-methylene-6-(1-propenylidene) (6.37%). Other major compounds in the *M. paniculata* oil are cyclohexene, 5,6-dietheyl-3-methyl (3.30%),  $\delta$ -elemene (3.57%),  $\alpha$ -cubebene

(2.96%), retinal (1%),  $\alpha$ -caryophyllene (3.13%), copaene (2.33%), cubenol (2.36%), nerolidyl acetate (1.2%), 12-oxabicyclo(9.1.0)dodeca-3,7-diene,1,5,5,8-tetramethyl (2.07%), eremophilene (1.54%), ledol (2.2%) and aromadendrene oxide (1.47%). The

presence of 3-carene as major compound in *M. koenigii* is not reported from elsewhere, completely differs from those reported by Raina et al. (2002) and Walde et al. (2005), where pinene, caryophyllenes and phellandrenes were the predominant compounds. This oil may be treated as a natural source of 3-carene, which may be studied for any bioactive properties for therapeutic uses. Also may serve as a useful compound of flavors and fragrances. The oil of *M. paniculata* also found different from the reports (Anonymous, 1962; Chopra et al., 1956). The study reveals that composition of two oils differs from the earlier reports and may, therefore be treated as different chemotypes. On the basis of above fact it may be concluded that *M. koenigii* and *M. paniculata*, growing widely in Bangladesh, may be utilized as a source for the isolation of natural 3-carene and caryophyllene oxide respectively.

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