CHEMICAL COMPOSITION OF THREE *XYLOPIA* LEAF ESSENTIAL OILS FROM PASOH FOREST RESERVE, NEGERI SEMBILAN, MALAYSIA

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SITI HUMEIRAH AG, NOR AZAH MA, MAILINA J, MUHAJIR H & PUAD A. 2010. Chemical composition of three *Xylopia* leaf essential oils from Pasoh Forest Reserve, Negeri Sembilan, Malaysia. Hydro distillation of the leaves of *Xylopia malayana, X. fusca* and *X. elliptica* collected from Pasoh Forest Reserve, Negeri Sembilan, Malaysia yielded 1.39, 1.25 and 0.56% (v/w) of pale yellow and yellowish oils respectively. The chemical compositions of the oils were analysed by gas chromatography (GC) and gas chromatography mass spectrometry (GCMS). A total of 29 (94.4% of the total oil), 22 (78.8%) and 34 (80.7%) compounds were identified from essential oils of the leaves of *X. malayana*, *X. fusca* and *X. elliptica* respectively. Analyses of the oils revealed that the main components from *X. malayana* leaf oil were β-pinene (42.0%), α-pinene (15.2%), elemol (11.6%) and bicyclogermacrene (5.2%) whilst the principal components of the *X. fusca* leaf oil were germacrene D (17.0%), bicyclogermacrene (12.0%), β-elemene (11.5%) and β-pinene (10.1%). Major compounds of *X. elliptica* leaf oil were bicyclogermacrene (11.5%), sabinene (10.6%), α-pinene (9.0%), elemol (8.1%) and β-pinene (5.4%).

Keywords: *Xylopia malayana*, *X. fusca*, *X. elliptica*, Annonaceae

SITI HUMEIRAH AG, NOR AZAH MA, MAILINA J, MUHAJIR H & PUAD A. 2010. Komposisi kimia minyak pati daun tiga spesies *Xylopia* dari Hutan Simpan Pasoh, Negeri Sembilan, Malaysia. Penyulingan daun *Xylopia malayana*, *X. fusca* dan *X. elliptica* yang dikutip dari Hutan Simpan Pasoh, Negeri Sembilan, Malaysia, masing-masing telah menghasilkan 1.39%, 1.25% and 0.56% minyak pati yang berwarna kuning pudar dan kekuningan. Kandungan komponen kimia minyak pati setiap spesies telah dianalisis menggunakan kromatografi gas dan gabungan kromatografi gas/spektrometri jisim. Sebanyak 29, 22 dan 34 komponen yang masing-masing merupakan 94.4%, 78.8% dan 80.7% daripada keseluruhan minyak pati *X. malayana, X. fusca* dan *X. elliptica* telah dikenal pasti. Minyak pati daun *X. malayana* didapati mengandungi β-pinena (42.0%), α-pinena (15.2%), elemol (11.6%) dan bisiklogermakrena (5.2%) sebagai komponen utama manakala minyak pati bagi daun *X. fusca* pula terdiri daripada germakrena D (17.0%), bisiklogermakrena (12.0%), β-elemena (11.5%) dan β-pinena (10.1%) sebagai komponen utamanya. Bagi *X. elliptica*, komponen utama minyak pati daun ialah bisiklogermakrena (11.5%), sabinena (10.6%), α-pinena (9.0%), elemol (8.1%) dan β-pinena (5.4%).

INTRODUCTION

The genus *Xylopia* (Annonaceae) comprises about 170 species and they are widely distributed throughout South-East Asia, Australia and Melanesia (Hyland & Whiffin 1993). In Malaysia, about eight species with three varieties of *Xylopia* have been recognised (Kochummen 1972). The plants are usually shrubs or small trees in nature. Many species have been noted for their fragrant flowers and also used as spices and flavouring agents in foods (Moraes & Roque 1988). Many *Xylopia* species have been known to possess volatile components, diterpenes and alkaloid (Leboeuf *et al.* 1982).

Xylopia malayana is a small tree up to 20 m high and occurs in the northern part of East Malaysia. The leaves are elliptic-oblong and glabrous whilst the flowers are small and aromatic. Locally known as 'banit kijang', the leaves of *X. malayana* are traditionally used for treatment after childbirth (Kamarudin 1988). The wood is also used for making rafters in native houses. *Xylopia elliptica* or locally known as 'lilan'

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is a medium size tree which could grow up to 8–10 m tall and commonly found throughout Peninsular Malaysia (Kochummen 1972). The leaves are membranous and blade elliptic. The flowers are axillary and the fruit, oblong. *Xylopia fusca* is widely distributed in South-East Asia. It is a medium to large tree, reaching 125 m in height and commonly found in peat swamp forest. Locally known as 'jangkang paya', the wood of *X. fusca* is used in making pineapple box while the bark is used for walls of huts. To the best of our knowledge, there are no reports on the chemical composition of the essential oils from *X. malayana*, *X. fusca* and *X. elliptica* and this study was aimed at making this information available.

MATERIALS AND METHODS

Plant material and preparation

Leaves of *X. malayana* (FRI 54729), *X. fusca* (FRI 54733) and *X. elliptica* (FRI 54736) were collected from Pasoh Forest Reserve, Negeri Sembilan. After identification, voucher specimens of the plant species were deposited at the Forest Research Institute Malaysia's (FRIM) herbarium. All plant materials (total weight: 63.90–511.21 g) were air dried for two days and were hydro distilled separately in a Clevenger-type apparatus for six hours. The oily layers obtained were separated and dried over anhydrous sodium sulphate. Oil yields were calculated based on dry weight (v/w) of the plant material.

Chemical analysis

The oils were analysed using gas chromatography (GC) and gas chromatography mass spectrometry (GCMS). The GC analysis was carried out on a Shimadzu GC-2010 gas chromatograph equipped with a flame ionisation detector (FID) using fused silica capillary column CBP-5 (25 m × 0.25 mm, 0.25 µm film thickness). Helium was used as the carrier gas and the injector and detector temperatures were set at 220 and 280 °C respectively. The oven temperature was programmed from 60 to 230 °C at 3 °C min^{-1} and finally held at 230 °C for 10 min. The volume of oil injected was 1.0 µl. Peak areas and retention times were measured by electronic integration.

The GCMS analyses were performed on Agilent GCMS 7890A/5975C Series MSD (70eV direct inlet) equipped with HP-5MS fused silica capillary column (30 m × 0.25 mm, 0.25 µm film thickness). The column and injector temperatures were the same as those for GC. The mass range was 50–550 Da in the full scan mode with a rate of 2.91 scans s^{-1}. The total scan time is 67.7 min. Chemical constituents were identified by comparison of their retention indices with literature values (Jennings & Shibamoto 1980, Adams 2001) and their mass spectral data with those from the Wiley7Nist0.5L, HPCH 2205.L and NIST05a.L mass spectral databases and, in some cases, by co-chromatography with authentic samples. Retention indices (RI) of the components were determined relative to the retention times of a series of *n*-alkanes. Relative proportions of the chemical compounds were expressed as percentages obtained by peak area normalisation and all relative response factors were taken as one.

RESULTS AND DISCUSSION

Leaf oils of *X. malayana, X. fusca* and *X. elliptica* obtained by water distillation yielded 1.39% (320.11 g), 1.25% (63.90 g) and 0.56% (511.21 g) (v/w) of oils respectively. Both *X. malayana* and *X. fusca* leaf oils were pale yellow in colour while the leaf oil from *X. elliptica* was yellowish in colour. The composition percentages of the leaf oils are listed in Table 1.

There were 29 (94.4% of the total oil), 22 (78.8%) and 34 (80.7%) compounds identified from the leaf essential oils of *X. malayana*, *X. fusca* and *X. elliptica* respectively. However, these three essential oils were different in terms of their major chemical composition. Main components of the essential oil from *X. malayana* were β-pinene (42.0%), α-pinene (15.2%), elemol (11.6%) and bicyclogermacrene (5.2%). Other components present in significant amounts were α-eudesmol (3.4%), guaiol (2.6%), β-phellandrene (1.9%), germacrene D (1.9%) and terpinen-4-ol (1.0%) in decreasing order of concentrations. Germacrene D (17.0%), bicyclogermacrene (12.0%), β-elemene (11.5%) and β-pinene (10.1%) were the main constituents of leaf oils from *X. fusca*. Other components with considerable amounts detected were spathulenol (6.3%), (*Z*)-β-ocimene (4.1%), α-cadinol (3.5%) and α-pinene (3.2%). Bicyclogermacrene was the most predominant constituent in *X. elliptica* which made up 11.5% of the total oil. This was followed by sabinene (10.6%), α-pinene (9.0%), elemol (8.1%) and β-pinene (5.4%).

Table 1 Chemical composition (%) of the essential oils of leaves from *Xylopia malayana, X. fusca* and *X. elliptica*

Compound	Retention index	*X. malayana* (%)	*X. fusca* (%)	*X. elliptica* (%)	Identification method
α-Thujene	927	0.4	-	1.4	RI, MS
α-Pinene	935	15.2	3.2	9.0	RI,MS,CO
Camphene	950	0.2	-	0.2	RI,MS
Sabinene	977	-	-	10.6	RI,MS
β-Pinene	980	42.0	10.1	5.4	RI,MS,CO
Myrcene	989	0.9	0.1	1.1	RI,MS,CO
α-Phellandrene	998	0.3	-	0.8	RI,MS
α-Terpinene	1013	0.3	-	2.3	RI,MS
ρ-Cymene	1023	0.1	-	0.2	RI,MS
β-Phellandrene	1029	1.9	0.2	2.5	RI,MS
1,8-Cineole	1037	-	-	0.2	RI,MS,CO
(*Z*)-β-Ocimene	1038	0.5	4.1	0.1	RI,MS
(*E*)-β-Ocimene	1048	-	0.2	-	RI,MS
γ-Terpinene	1059	0.6	-	4.0	RI,MS
Terpinolene	1085	0.2	-	1.0	RI,MS
Linalool	1099	0.7	-	0.3	RI,MS,CO
Cis-ρ-menth-2-en-1-ol	1021	-	-	0.2	RI,MS
Allo-ocimene	1127	-	0.3	-	RI,MS
Trans-ρ-menth-2-en-1-ol	1141	-	-	0.1	RI,MS
Terpinen-4-ol	1177	1.0	0.1	5.3	RI,MS,CO
α-Terpineol	1192	0.4	-	0.3	RI,MS
Cis-piperitol	1194	-	-	0.1	RI,MS
Bornyl acetate	1280	0.1	-	0.2	RI,MS,CO
δ-Elemene	1333	0.9	1.7	1.6	RI,MS
α-Terpinyl acetate	1344	0.2	-	0.7	RI,MS
α-Cubebene	1345	-	0.2	-	RI,MS
α-Copaene	1367	-	0.9	0.4	RI,MS
α-Ylangene	1372	-	0.1	-	RI,MS
β-Bourbonene	1381	-	0.8	-	RI,MS
β-Elemene	1386	0.4	11.5	0.7	RI,MS
β-Caryophyllene	1415	0.3	1.8	0.6	RI,MS
Germacrene D	1480	1.9	17.0	2.8	RI,MS
Bicyclogermacrene	1498	5.2	12.0	11.5	RI,MS
δ-Cadinene	1518	0.2	1.8	-	RI,MS
Germacrene B	1558	-	1.4	-	RI,MS
Elemol	1561	11.6	-	8.1	RI,MS
Spathulenol	1584	1.2	6.3	2.2	RI,MS
Viridiflorol	1590	-	1.4	1.3	RI,MS
Guaiol	1601	2.6	-	3.5	RI,MS
γ-Eudesmol	1637	1.1	-	0.9	RI,MS
α-Cadinol	1661	-	3.5	-	RI,MS
α-Eudesmol	1663	3.4	-	-	RI,MS
Bulnesol	1671	0.6	-	1.3	RI,MS

Percentage was calculated based on results obtained from column CBP-5; MS = mass fragmentation; CO = co-chromatography with authentic samples

Results of this study showed that all three leaf oils were made up of monoterpenoids and sesquiterpenoids. This is in agreement with literature reports on essential oils from other *Xylopia* species (Lemos *et al.* 1992, Fournier *et al.* 1994, Nor Azah *et al.* 1996a, b, Brophy & Goldsack 1998, Lago *et al.* 2005, Tavares *et al.* 2007, Costa *et al.* 2008). β-Pinene was identified as the major monoterpene component in the leaf oils from *X. caudata* (60%) and *X. ferruginea* (40.7%) (Nor Azah *et al.* 1996a, b). This compound was also reported in some *Guatteriopsis* species from the same family (Costa *et al.* 2008). Presence of bicyclogermacrene and germacrene D (both sesquiterpenoids) has also been reported in *X. maccraea* and *X. langsdorffiana* respectively (Brophy & Goldsack 1998, Tavares *et al.* 2007). Other chemical constituents, for example spathulenol, ρ-cymene and α-pinene detected in the leaf oils in this study have also been found in other *Xylopia* species. Spathulenol was found mainly in *X. aromatica* and *X. emarginata* leaf oils (Fournier *et al.* 1994, Lago *et al.* 2005) whilst ρ-cymene (16%) and α-pinene (6.1%) were detected from the leaf oils of *X. sericea* studied by Lemos *et al.* (1992). Chemical investigation of the three *Xylopia* species studied represented a valuable chemotaxonomic characterisation for the identification of the genus *Xylopia*. Considering the relatively high yield of monoterpenoids and possible bioactivity potential in the oils, *Xylopia* could be a new source of perfumery ingredient for personal care and cosmeceutical industries.

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