J. Mater. Environ. Sci. 7 (6) (2016) 1921-1924
ISSN : 2028-2508
CODEN: JMESCN

Essential oil composition of *Piper majusculum* Ridl. from Indonesia

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Received 20 Mar 2015, Revised 22 Aug 2015, Accepted 24 Aug 2015
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Abstract
The essential oil of *Piper majusculum* from Piperaceae family was obtained by hydrodistillation technique in a Dean-Stark apparatus. The GC and GC-MS analysis of the essential oil had identified twelve components accounting for 93.79% of the total oil. The major components were β-caryophyllene (17.27%), caryophyllene oxide (14.26%), α-selinene (14.21%) and *cis*-calamenene (9.62%). This is the first report on the volatile components of *P. majusculum*.

Keywords: Essential oil, chemical composition, *Piper majusculum*, Piperaceae

1. Introduction
Genus *Piper* is largely distributed in tropical and subtropical regions of the world [1]. Most of the species are fast-growing aromatic shrubs or vines with distinctive swollen nodes and drooping spikes, frequently found in the understory and secondary vegetation of tropical forest canopy of the Americas and Asia [2]. For years, the chemistry of the *Piper* species has been widely investigated, and phytochemical studies have resulted in the isolation of numerous biologically active compounds such as alkaloids, lignans, neolignans, terpenes, steroids, kawapyrones, piperolides, and flavanoids [3]. Members of the *Piper* genus are of commercial, economical and medicinal importance. *Piper* species are important medicinal plants used in Chinese medicine, in the Indian ayurvedic system and in folk medicine practices of Latin America and West Indies [4]. They are applied to treat asthma, bronchitis, fever, hemorrhoidal afflictions, gastrointestinal diseases, and rheumatism [5], and the preparations obtained from these plants have shown anti-inflammatory, insecticidal, anti-hypertensive, antidiabetics, immunomodulatory and antimutagenic effects [6].
P. majusculum Ridl., is known as '*sirih hutan*' (Malaysia and Indonesia) and collected from Bungus, Teluk Kabung, Padang, West Sumatra, Indonesia. Phytochemical studies have been done on the extracts of *P. majusculum* and successfully isolated 2-(5′-methoxy-3′,4′-methylenedioxyphenyl)-6-(4″-hydroxy-3″,5″-dimethoxyphenyl)-3,7-dioxabicyclo[3,3,0]octane, oleic acid, β-sitosterol, and mixture of three sterols [7]. Previous study on various extracts of *P. majusculum* also revealed strong inhibition on anti-inflammatory activity by TPA-induced mouse ear oedema model, ranging from 68.39-85.29%, comparable to indomethacin, 85.50%. In the TPA-induced mouse ear edema model, the extracts of *P. majusculum* act as an inhibitor of either protein kinase C or phospholipase A_2 [7]. On the other hand, lipoxygenase is involved in AA metabolism, generating various biologically active leukotrienes that play an important role in inflammation [8]. To the best of our knowledge, there is no report found on the chemical composition of the essential oil of *P. majusculum*.

2. Materials and methods
2.1. Plant materials and extraction method
A sample of *P. majusculum* was collected from Bungus, Teluk Kabung, Padang, Indonesia (**Figure 1**). This sample was identified by Mr. Rusdi Tamin and Nurainas and the sample specimens (EM-02/1205) were deposited at the Andalas Herbarium (ANDA), University of Andalas. Sample (200 g) was hydrodistilled for 6 h in a Dean Stark apparatus [9]. The resulting oils were collected, preserved in a sealed amber glass sample tube and stored at 4°C under refrigeration until analysis.

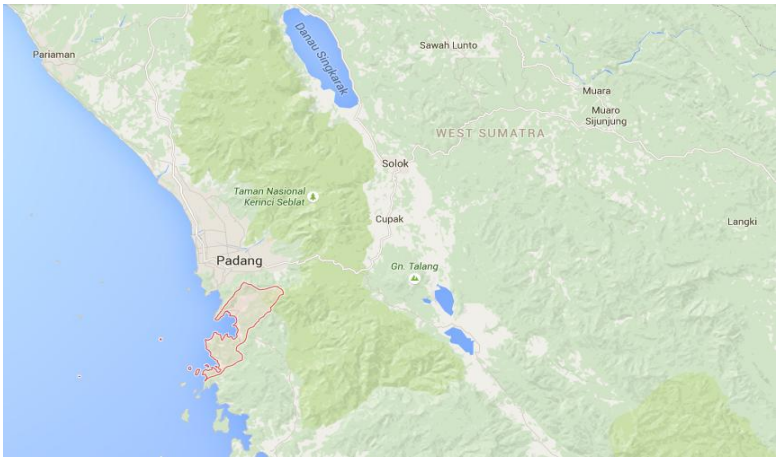

Figure 1: Location of Bungus, Teluk Kabung, Padang, Indonesia

2.2. Gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS)

GC analysis were performed on a Hewlett Packard 6890 series II A gas chromatograph equipped with an Ultra-1 column (25 m long, 0.33 µm thickness and 0.20 mm inner diameter). Helium was used as a carrier gas at a flow rate of 0.7 mL/min. Injector and detector temperature were set at 250 and 280°C, respectively. Oven temperature was kept at 50°C, then gradually raised to 280°C at 5°C/min and finally held isothermally for 15 min. Diluted samples (1/100 in diethyl ether, v/v) of 1.0 µL were injected manually (split ratio 50:1). The injection was repeated three times and the peak area percents were reported as means ± SD of triplicates. Calculation of peak area percentage was carried out by using the GC HP Chemstation software (Agilent Technologies). GC-MS chromatograms were recorded using a Hewlett Packard Model 5890A gas chromatography and a Hewlett Packard Model 5989A mass spectrometer. The GC was equipped with Ultra-1 column (25 m long, 0.33 µm thickness and 0.20 mm inner diameter). Helium was used as carrier gas at flow rate of 1 mL/min. Injector temperature was 250°C. Oven temperature was programmed from 50°C (5 min hold) to 250°C at 10°C/min and finally held isothermally for 15 min. For GC-MS detection, an electron ionization system, with ionization energy of 70 eV was used. A scan rate of 0.5 s (cycle time: 0.2 s) was applied, covering a mass range from 50–400 amu [9].

2.3. Identification of constituents

The constituents of the essential oil were identified by comparison of their mass spectra with reference spectra in the computer library (Wiley) and also by comparing their retention indices or data in the literatures [10]. The quantitative data were obtained electronically from FID area percentage without the use of correction factor.

3. Results and discussion

The yield of the essential oil obtained from the hydrodistillation of the whole parts (leaves and bark) of *P. majusculum* was 0.13 g, 0.026% (w/w). **Table 1** lists the chemical compositions identified with percentage composition along with their Kovats Index in two different columns, Ultra-1 (100% polymethylsiloxane) and HP-Wax. The GC and GC-MS (**Figure 2**) analysis of the essential oil successfully identified twelve components, together with four unidentified components, representing 93.79% of the total oil. The chemical structures of the essential oil components are shown in **Figure 3**. The essential oil were characterized by high amount of sesquiterpenoids which was dominated by β-caryophyllene (**3**) (17.27%), caryophyllene oxide (**11**) (14.26%), α-selinene (**6**) (14.21%) and *cis*-calamenene (**8**) (9.62%). The other major components that displayed more than 2% were spathulenol (**12**) (8.76%), α-bergamotene (**4**) (7.02%), α-copaene (**2**) (5.88%), α-(*Z*)-bisabolene (**7**) (4.95%), β-bisabolene (**9**) (4.13%), δ-cadinene (**10**) (2.80%), α-cubebene (**1**) (2.64%), and α-humulene (**5**) (2.25%). Meanwhile, monotepene were not detected in this oil.

The presence of β-caryophyllene in significant amounts in this essential oil is in agreement with the previous study from *P. obliquum* [11], *P. lancaefolium* [12], *P. guineense* [13], *P. tuberculatum* [14], *P. amapense* and *P. duckei* [15] indicating that the occurrence of β-caryophyllene as the major component may be a characteristic of *Piper* essential oils. β-caryophyllene is known for its anti-inflammatory and local anaesthetic activities [15]. Germacrene D and bicyclogermacrene which were abundant sesquiterpene markers for *Piper* species of *P. arboreum*, *P. cubotaonum* [16], *P. cernuum* [17], and *P. bisasperatum* [18].

Table 1: Chemical constituents identified from the essential oil of *P. majusculum*

Retention time	Compound	Kovats Index		Percentage (%)
		Ultra-1	HP-Wax	
14.41	α-Cubebene (**1**)	1353	1460	2.64
15.15	α-Copaene (**2**)	1369	1488	5.88
15.39	unidentified			tr
16.26	β-Caryophyllene (**3**)	1409	1617	17.27
16.47	α-Bergamotene (**4**)	1424		7.02
17.07	α-Humulene (**5**)	1461		2.25
17.92	α-Selinene (**6**)	1498	1716	14.21
18.06	α-(*Z*)-Bisabolene (**7**)		1720	4.95
18.13	unidentified			tr
18.45	*cis*-Calamenene (**8**)	1505	1857	9.62
18.53	unidentified			tr
19.84	β-Bisabolene (**9**)	1513		4.13
20.04	δ-Cadinene (**10**)	1531	1774	2.80
20.62	unidentified			tr
21.61	Caryophyllene oxide (**11**)		2001	14.26
21.91	Spathulenol (**12**)		2150	8.76
	Identified components			93.79

tr – trace amounts (< 0.1)

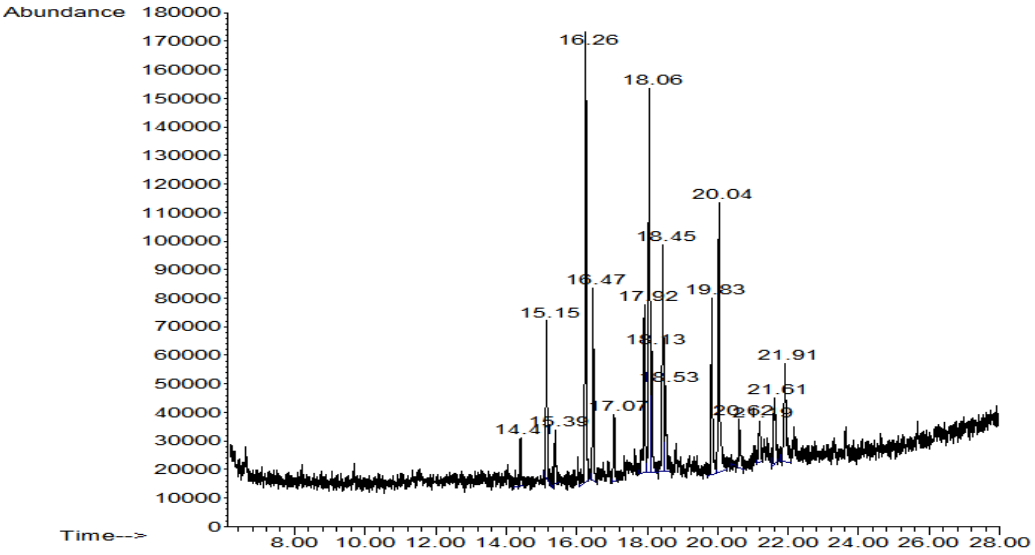

Figure 2: GC-MS chromatogram of the essential oil of *P. majusculum*

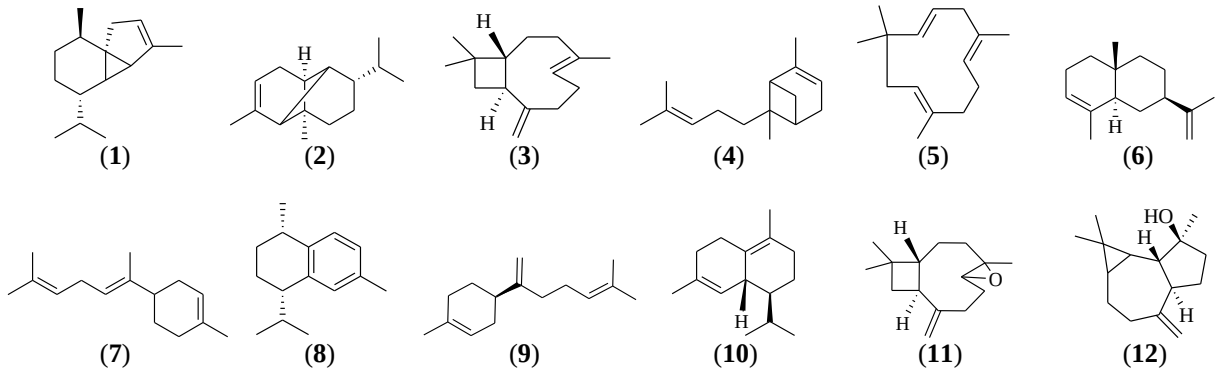

Figure 3: Chemical structures of the essential oil components of *P. majusculum*

However, these two constituents were not detected in the current essential oil. This could be due to the different environmental and genetic factors, chemotypes and nutritional status of the plants, which may influence the oil composition [19]. Our study can be considered as the first detailed report on the analysis of chemical composition of the essential oils of *P. majusculum* from Indonesia.

Conclusion

In conclusion, the high concentration of β-caryophyllene, α-selinene and caryophyllene oxide in the essential oil of *P. majusculum* contributed to the unique flavor of the plants. The presence of these components makes the plants useful in the medicines because they exhibit antibacterial, antifungal, anticancer, anti-inflammatory, antimalarial and are also used traditionally as flavoring agent and antimicrobials in food.

Acknowledgements-The authors are grateful to Research University Grant (GUP) under vote Q.J130000.2526.03H93 for financial support and Faculty of Science, Universiti Teknologi Malaysia for research facilities.

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