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The rhizome essential oil of *Curcuma aeruginosa* Roxb. (Zingiberaceae) from Vietnam

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ABSTRACT

The volatile compounds identified from the rhizome oil of *Curcuma aeruginosa* Roxb (Zingiberaceae) collected from Cham Chu Natural Reserve, Tuyen Quang Province, Vietnam, are reported. The plant sample was air-dried for two weeks under laboratory shade. Essential oils were obtained by hydrodisitllation in a Clevenger-type apparatus. The yield of the oil was 0.18% (v/w) regarding the dry weight of the plant material. The constituents of the oil were analyzed by gas chromatography (GC-FID) and gas chromatography-mass spectrometry (GC/MS). The main constituents of the oil were found to be β-pinene (21.9%), neocurdione (16.1%) and curcumol (15.2%).

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1. Introduction

The role of medicinal plants is remarkable in different disciplines of food, agricultural and pharmaceutical sciences. Apart from being food for man, they act as sources of drugs and natural products for useful purposes. Medicinal plants and herbs have been used to cure and ameliorate several disorders from time immemorial (Ganesan and Xu, 2017; Rais and Ali, 2017). In fact, diverse plant materials have been major sources of natural products and nutraceuticals (Mohammadhosseini et al., 2017; Pavuranj et al., 2017; Venditti et al., 2018). Essential oils are valuable mixtures of natural products that have found important applications for industrial and pharmaceutical purposes (Aidi et al., 2017; Camilo et al., 2017; Mohammadhosseini, 2017; Erenler et al., 2017).

Curcuma L., a valuable genus in the family Zingiberaceae, includes approximately 110 species. The plants of this genus are native to Southeast Asia. *Curcuma aeruginosa* Roxb. is one of the most valuable oriental herbs. The plant is distinguished by red corolla lobes and ferruginous or greenish-blue rhizome. In the young plants, the rhizomes of *C. zedoaria* are easily confused with those of *C. aeruginosa* Roxb. and *C. mangga* because both have almost similar yellow color. However, a cross-section of the rhizomes of the mature plants of *C. aeruginosa* Roxb. is slightly dark purplish, whilst *C. mangga* has brighter yellow color (Malek et al., 2004). However, a cross-section of the rhizomes of *C. zedoaria* is slightly red in color (Nair, 2013).

The combination of 5% (*n*-hexane) extract of *C. aeruginosa* Roxb. and 5% minoxidil (anti-hair loss cream) slowed hair loss and increased hair growth, thereby acting as 5α-reductase inhibitor (Pumthong et al., 2012). Extracts of *C. aeruginosa* Roxb. possess strong antibacterial (Safitri et al., 2017), antioxidant (Nurcholis et al., 2015; Safitri et al., 2017), toxicity (Nurcholis et al., 2015) and anti-inflammatory (Wantana et al., 2006; Siska and Churiyah, 2015) activities. There were reports on the antinociceptive (Wantana et al., 2006), antipyretic (Wantana et al., 2006) and cytotoxic (Eka et al., 2014; Rilianawati et al., 2015) potentials of *C. aeruginosa* Roxb. extracts. Some compounds isolated from *C. aeroginosa*

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such as aeruginone and curcumenone exhibited cytoxic activity against some human cancer cell lines (Atun et al., 2016), while germacrone displayed anti-inflammatory (Chowdhury et al., 2015) and antiandrogenic (Safitri et al., 2017) properties. It has also been shown that sesquiterpene-enriched extract of *C. aeruginosa* Roxb. retards axillary hair growth (Srivilai et al., 2018).

Beyond the previously mentioned compounds, the phytochemicals of *C. aeruginosa* Roxb. include aeruginone and curcumenone (Atun et al., 2016), zedoarol, curcumenol, isocurcumeno (Sukari et al., 2007), germacrone, zederone, dehydrocurdione and zedoarondiol (Suphrom et al., 2012; Suphrom et al., 2014). Phytochemicals such as furanodiene, germacrone, zederone, dehydrocurdione and zedoarondiol from *C. aeruginosa* Roxb. displayed potential anti-androgenic activity (Jukkarin et al., 2011; Suphrom et al., 2014). The compounds 9-methyltetracyclo[7.3.1.0(2.7).1(7.11)] tetradecane, epicurzerenone and *cis*-1,3-dimethyl-2-methylene cyclohexane are responsible for the murine hepatocyte apoptosis and PARP-1 expression of *C. aeruginosa* Roxb. (Eka et al., 2014). Other sesquiterpenoid compounds such as curzerenone, curcumenone and curzeone were isolated from *C. aeruginosa* Roxb. (Che and Nur, 2014). Zedoalactone A, zedoalactone B, guaianolide and zedoarondiol were isolated from this plant, as well (Takano et al., 1995). Previous results showed that *C. aeruginosa* Roxb. rhizome which was extracted using two different techniques of solvent extractions contained various chemical classes of compounds including terpenoids, sterols, organic acids, fatty acids and sugars (Sanimah and Alizah, 2015).

The chemical compounds identified previously in the essential oils of *C. aeruginosa* Roxb. are given in Table 1.

The main compounds are mostly oxygenated sesquiterpenes (Fang et al., 1982; Zwaving and Bos, 1992; Dung et al., 1995; Dung et al., 1996; Sirat et al., 1998; Bin Jantan et al., 1999; Jirovetz et al., 2000; Jarikasem et al., 2005; Chantana et al., 2011; Che and Nur, 2014; Orawan et al., 2015; Nararat et al., 2017), oxygenated monoterpenes (Dung et al., 1995; Dung et al., 1996; Sirat et al., 1998; Bin Jantan et al., 1999; Jirovetz et al., 2000; Chantana et al., 2011; Che and Nur, 2014; Orawan et al., 2015; Nararat et al., 2017), aromatic compounds (George and Britto, 2015) and diterpenoids (Kamazeri et al., 2012). The essential oils of *C. aeruginosa* Roxb. are known to exhibit antityrosinase (Che and Nur, 2014), antimicrobial (Jirovetz et al., 2000; Orawan et al., 2015; Nararat et al., 2017; Wulan et al., 2017) and antioxidant (George and Britto, 2015) activities.

This paper reports the compounds identified in the rhizomes of *C. aeruginosa* Roxb. growing in Vietnam, in continuation of our research activity on volatiles from Vietnamese plants and herbs (Dai et al., 2018). The rhizome of *C. aeruginosa* Roxb. in Vietnam is used for various medicinal purposes such as antibacterial, anti-ulcer, anti-inflammatory features (Dai, personal information).

2. Experimental

2.1. Collection and authentication of plant

The rhizomes of *C. aeruginosa* Roxb. were harvested mid-day from Cham Chu Natural Reserve, Tuyen Quang Province, Vietnam (22.117ºN 105.250ºE), in August 2013. Botanical identification was achieved by Dr. Oanh P.T. A voucher specimen, PTO Z2, was deposited at the Herbarium Unit, VNU University of Science, Vietnam.

2.2. Preparation of plant sample

Prior to hydrodisitillation, the sliced rhizomes of *C. aeruginosa* Roxb. were air-dried under laboratory shade (21 ºC) for two weeks. In addition, sediments and other unwanted materials were separated from the samples by shaking and alternate hand picking. Afterwards, samples were pulverized to coarse powder using a locally made grinder.

Table 1
Major compounds identified in the previously analysed essential oil of *C. aeruginosa* Roxb..

Part	Origin	Main constituents	References
Rhizome	Malaysia	Epicurzerenone (19.5%), 1,8-cineole (15.9%), *trans*-β-farnesene (9.8%)	Che and Nur, 2014
''	''	Isocurcumenol (8.5%), curcumenol (9.9%), curcumanolides A,B (11.4%), dehydrocurdione (9.4%)	Zwaving and Bos, 1992
''	''	1,8-Cineole (23.2%) and curzerenone (28.4%)	bin Jantan et al., 1999
''	''	Curzerenone (24.6%), 1,8-cineole (11.0%), camphor (10.6%)	Sirat et al., 1998
Not stated[a]	''	Cycloisolongifolene, 8,9-dehydro formyl (35.3%) and dihydrocostunolide (22.5%)	Kamazeri et al., 2012
Leaf	Vietnam	Curzerene (16.2%), germacrone (13.6%), 1,8-cineole (13.5%)	Dung et al., 1995
''	''	Curdione (33.9%) and 1,8-cineole (26.3%)	Dung et al., 1996
Root	''	1,8-Cineole (13.4%), *cis*-β-elemenone (13.3%)	''
Small/ large rhizomes	''	*cis*-β-Elemenone (11.0%/14.5%), germacrone (3.8%/11.3%) and curdione (9.8%/8.4%)	''
Rhizome	India	1,8-Cineole (17.7%), curzerenone (10.5%), furanogermenone (7.8%), camphor (7.5%)	Jirovetz et al., 2000
''	''	Ethoxybenzene (33.4%)	George and Britto, 2015
Rhizome	Thailand	β-Pinene (7.7%), 1,8-cineole (9.6%) and curzerenone (41.6%)	Jarikasem et al., 2005
''	''	Camphor (16.85%),curzerenone (16.81%)	Chantana et al., 2011
''	''	Germacrone (23.5%), curzerenone (11.8%) and 1,8-cineole (10.9%)	Orawan et al., 2015
''	''	Camphor (29.4%) and germacrone (21.2%)	Nararat et al., 2017
Rhizome	China	Curzerenone[b]	Fang et al., 1982

[a]Part not stated; [b]Concentration not stated

2.3. Hydrodistillation of essential oil from rhizome of *C. aeruginosa* Roxb.

500 g of the pulverized rhizome of *C. aeruginosa* Roxb. was carefully introduced into a 5 L flask and distilled water was added until it covers the sample completely. Essential oils were obtained by separate hydrodistillation which was carried out in an all glass Clevenger-type distillation unit designed according to the established specification (Vietnamese Pharmacopoeia, 1997). The distillation time was 3 h and conducted at normal pressure (760 mmHg). The volatile oils distilled over water and were collected in the receiver arm of the apparatus into clean and previously weighed sample bottles. The oil sample was dried with anhydrous Na_2SO_4 and kept under refrigeration (4 ºC) until the moment of analyses (Dai et al., 2018).

2.4. Gas chromatography analysis of the oil

Gas chromatography (GC-FID) analysis was performed on an Agilent Technologies HP 6890 Plus Gas chromatograph equipped with an FID and fitted with an HP-5MS column (30 m x 0.25 mm, film thickness 0.25 µm, Agilent Technology). The analytical conditions were: carrier gas H_2 (1 mL/min), injector temperature (PTV) 250 ºC, detector temperature 260 ºC, column temperature programmed from 40 ºC (2 min hold) to 220 ºC (10 min hold) at 4 ºC/min. Samples were injected by splitting and the split ratio was 10:1. The volume injected was 1.0 µL. Inlet pressure was 6.1 kPa. Each analysis was performed in triplicate. The relative amounts of individual components were calculated based on the GC peak area (FID response) without using correction factors as previously described (Dai et al., 2018).

2.5. Gas chromatography-mass spectrometry analysis of the oil

An Agilent Technologies HP 6890N Plus Chromatograph fitted with a fused silica capillary HP-5 MS column (30 m x 0.25 mm, film thickness 0.25 µm) and interfaced with a mass spectrometer HP 5973 MSD was used for this experiment, under the same conditions as those used for gas chromatographic analysis (Dai et al., 2018). The conditions were the same as described above with He (1 mL/min) as carrier gas. The MS conditions were as follows: ionization voltage 70 eV; emission current 40 mA; acquisitions scan mass range of 35-350 m/z at a sampling rate of 1.0 scan/s.

2.6. Identification of the components of the oil

The identification of constituents of *C. aeruginosa* Roxb. was performed on the basis of retention indices (RI) determined with reference to a homologous series of *n*-alkanes (C_6-C_{40}), under identical experimental conditions. In some cases, co-injection with known compounds (Sigma-Aldrich, St. Louis, MO, USA) under the same GC conditions was employed. The mass spectral (MS) fragmentation patterns were checked with those of other known compounds and those in the literature (Dai et al., 2018).

3. Results and Discussion

3.1. Chemical composition of *C. aeruginosa* Roxb.

The yield of the essential oil was 0.18% (v/w) calculated on a dry weight basis, the oil sample was obtained as a light yellowish liquid. The volatile compounds were displayed in Table 2 along with their percentages and retention indices calculated on an HP-5MS column. Thirty-seven compounds representing 95.6% of the total volatile contents were identified in the oil sample of *C. aeruginosa* Roxb.. These involve monoterpene hydrocarbons (32.2%), oxygenated monoterpenes (6.8%), sesquiterpene hydrocarbons (20.4%), oxygenated sesquiterpenes (33.1%), and non-terpenes (2.1%). The main constituents of the oil were β-pinene (21.9%), neocurdione (16.1%) and curcumol (15.2%). There were significant amounts of β-elemene (6.6%), myrtenyl acetate (6.1%), β-caryophyllene (4.9%), α-pinene (3.4%) and α-selinene (3.3%).

3.2. Comparison of previous and present studies on essential oils of *C. aeruginosa* Roxb.

A comparison of the present data with previous ones indicated that the abundance of β-pinene in the studied rhizome essential oil of *C. aeruginosa* Roxb. from Vietnam was noteworthy. Literature data have shown that terpene hydrocarbons are less common in the essential oils of *C. aeruginosa* Roxb. when compared with oxygenated counterparts (Table 1). β-Pinene, an uncommon constituent of *C. aeruginosa* Roxb. essential oils, was only described in the rhizome oil of the plant from Thailand (Jarikasem et al., 2005). The contents of β-pinene in the Thailand sample of *C. aeruginosa* Roxb. essential oil were lower when compared with the present oil analysis from Vietnam. In addition, neocurdione, one of the major constituents of *C. aeruginosa* Roxb. in the present study was not previously found in *C. aeruginosa* Roxb. essential oils. A high content curcumol in the studied rhizome oil under investigation was also a significant point of difference with data previously reported from the essential oils of *C. aeruginosa* Roxb. (Table 1). This compound was not reported to be the major constituent of the oil of *C. aeruginosa* Roxb.. It was well-known that the qualitative and quantitative composition of an essential oil may be greatly influenced by several factors such as the climate, soil composition, the habitat, environmental changes, time collection and vegetative part. These factors may have been responsible for the variations

Table 2
Chemical constituents of essential oil of *C. aeruginosa* Roxb..

Compounds[a]	RI[b]	RI[c]	Percent composition[d]
α-Pinene	939	932	3.4
α-Fenchene	948	941	0.4
Camphene	953	946	0.2
Verbenene	968	954	0.7
β-Pinene[e]	980	978	21.9
β-Myrcene	990	988	0.2
Limonene	1032	1030	0.8
(*E*)-β-Ocimene	1052	1044	0.1
α-Terpinolene	1090	1089	0.1
Linalool	1100	1100	0.4
p-Mentha-1,4(8)-diene[e]	1110	1093	5.4
Pinocarvone	1165	1165	0.1
α-Thujenal	1195	1189	0.2
Myrtenyl acetate[e]	1326	1328	6.1
α-Copaene	1378	1377	0.4
β-Elemene	1391	1389	6.6
β-Caryophyllene	1419	1417	4.9
γ-Elemene	1437	1434	0.3
α-Humulene	1454	1452	0.9
α-Amorphene	1485	1485	1.0
β-Selinene	1486	1486	1.4
δ-Selinene	1493	1493	1.0
(*E,E*)-α-Farnesene[e]	1506	1503	0.2
Pentadecene	1507	1500	0.1
α-Selinene	1512	1510	3.3
δ-Cadinene	1525	1522	0.2
trans-γ-Bisabolene[e]	1526	1531	0.2
Elemol	1550	1548	0.9
(*E*)-Nerolidol	1563	1561	0.8
Curcumol[e]	1623	1623	15.2
(*Z*)-3-Heptadecene[e]	1688	1688	0.1
Albicanol	1736	1740	0.1
Neocurdione[e]	1765	1761	16.1
n-Hexadecanoic acid	1964	1964	0.4
Octadecanoic acid	2188	2182	0.7
(*Z*)-9-Octadecenamide[e]	2375	2375	0.4
(*Z*)-13-Docosenamide[e]	2649	2625	0.5
Total			**95.6**
Monoterpene hydrocarbons			**33.2**
Oxygenated monoterpenes			**6.8**
Sesquiterpene hydrocarbons			**20.4**
Oxygenated sesquiterpenes			**33.1**
Non-terpenes			**2.1**

[a]Elution order on an HP-5MS column; [b]Retention indices on an HP-5MS column; [c]Literature retention indices; [d]Standard deviation (±SD) were insignificant and were excluded from the Table; [e]Co-injection with authentic compounds

between the previous and present data of essential oils of *C. aeruginosa* Roxb..

3.3. Proposed chemical combination of essential oils of *C. aeruginosa* Roxb.

The previous data on the essential oils of *C. aeruginosa* Roxb. revealed the high frequency of oxygen-containing monoterpenes and sesquiterpene compounds. The main identified monoterpene compounds were camphor and 1,8-cineole, while the sesquiterpene compounds mainly consisted of curdione, curzerenone, epicurzerenone, germacrone, *cis*-β-elemenone, curcumenol, curcumanolides A,B and dehydrocurdione. The combination of main abundant compounds obtained from previous studies on *C. aeruginosa* Roxb. oils were epicurzerenone/1,8-cineole (Che and Nur, 2014), curcumenol/curcumanolides A,B/dehydrocurdione (Zwaving and Bos, 1992), 1,8-cineole/curzerenone (Dung et al., 1996; Bin Jantan et al., 1999; Jirovetz et al., 2000; Jarikasem et al., 2005), curdione/1,8-cineole (Dung et al., 1996), 1,8-cineole/*cis*-β-elemenone (Dung et al., 1996), *cis*-β-elemenone/germacrone (Dung et al., 1996), camphor/curzerenone (Chantana et al., 2011), germacrone/curzerenone/1,8-cineole (Orawan et al., 2015), camphor/germacrone (Nararat et al., 2017). The two other uncommon constituents of the oils of *C. aeruginosa* Roxb. were cycloisolongifolene, 8,9-dehydro formyl/dihydrocostunolide (Kamazeri et al., 2012) and ethoxybenzene (George and Britto, 2015).

4. Concluding remarks

In the present work, chemical compositions of the essential oils from the rhizome of *C. aeruginosa* Roxb. have been reported for the first time. The main components characterized in the studied essential oil were β-pinene, neocurdione and curcumol. The present compositional pattern of the essential oil was found to be different from the other *C. aeruginosa* Roxb. plants grown in Vietnam or other parts of the world. The difference in chemical constituents of the essential oil of *C. aeruginosa* Roxb. between the previous and present studies could be attributed to climatic and environmental conditions as well as age and nature of the plant. The time of collection, plant parts, handling and processing methods may also contribute significantly to the differences in chemical profiling. However, more research works would be required to delineate a proper chemotaxonomy of essential oil of *C. aeruginosa* Roxb..

Conflict of interest

The authors declare that there is no conflict of interest.

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