Natural Product Research

Formerly Natural Product Letters

ISSN: 1478-6419 (Print) 1478-6427 (Online) Journal homepage: https://www.tandfonline.com/loi/gnpl20

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To cite this article: T. Soumya, P. R. Jayasree, M. Deepak & P. R. Manish Kumar (2019): Chemical composition, antioxidant and antiproliferative activities of essential oil from rhizome and leaves of *Curcuma mutabilis* Škorničk., M. Sabu & Prasanthk., endemic to Western Ghats of India, Natural Product Research, DOI: 10.1080/14786419.2018.1533826

To link to this article: https://doi.org/10.1080/14786419.2018.1533826

NATURAL PRODUCT RESEARCH
https://doi.org/10.1080/14786419.2018.1533826

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SHORT COMMUNICATION

Chemical composition, antioxidant and antiproliferative activities of essential oil from rhizome and leaves of *Curcuma mutabilis* Škorničk., M. Sabu & Prasanthk., endemic to Western Ghats of India

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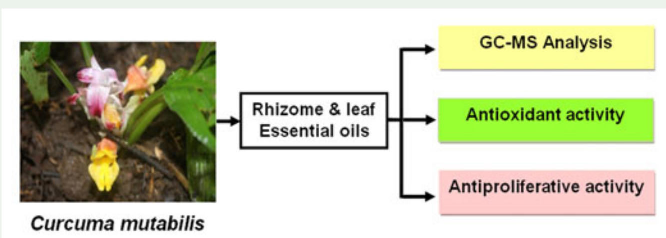
ABSTRACT

Hydro-distilled essential oils, from fresh rhizomes and leaves of *Curcuma mutabilis* Škorničk., M.Sabu & Prasanthk., characterized by GC–MS revealed the presence of thirty three and twenty three compounds therein respectively. Whilst estrone methyl ether (3-Methoxyestra-1,3,5(10)-trien-17-one) was the major component in rhizome oil (47.35%), sesquiterpene hydrocarbons predominated as the major group (63.92%) in leaf oil with a higher preponderance of β-caryophyllene (25.48%), β-farnesene (19.47%) and α-humulene (11.01%). Weak antioxidant activities observed in these oils determined by DPPH and ABTS methods were apparently influenced both by the oil composition and the assay conditions. Rhizome oil showed higher antiproliferative activity than leaf oil against leukemic K562 (IC_{50}-6.8μg/mL) and colorectal HCT116 (IC_{50}-8.5μg/mL) cancer cell lines. This first report reveals composition and biological activities of essential oils from *C. mutabilis*.



ARTICLE HISTORY
Received 2 June 2018
Accepted 5 October 2018

KEYWORDS
Curcuma mutabilis; essential oil; GC-MS analysis; estrone methyl ether; antiproliferative activity

1. Introduction

The genus *Curcuma* (Zingiberaceae) is a bioresource enriched with molecules credited with a plethora of bioactivities including anti-inflammatory, hypocholesterolemic, antidiabetic antimicrobial, antiviral, antioxidant and anticancerous among others (Chattopadhyay

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Supplemental data for this article can be accessed at https://doi.org/10.1080/14786419.2018.1533826

et al. 2004; Sasikumar 2005; Kumar et al. 2013). Apart from the major curcuminoid compounds, other bioactive phytochemicals including terpenoids have also been identified in essential oils (Afzal et al. 2013). Lesser known turmerones from *Curcuma longa* rhizome oil reportedly possess potent antioxidant, cytotoxic and apoptogenic effects on cancer cell lines (Ji et al. 2004; Funk et al. 2010). Oil from selected *Curcuma* species termed 'Zedoary' containing curzerene, eucalyptol, curcumol, pyridine, germacrone, β-elemene and curdione is used in Chinese medicine for treatment of various ailments including malignant tumors. This oil also reportedly inhibits gastric cancer cell line proliferation through cell cycle arrest and apoptosis induction (Pharmacopeia Commission of PRC 2010; Shi et al. 2013). *C. zedoaria* oil containing epicurzerenone and curzerene used as folk medicine reportedly possess antioxidant and anti-angiogenesis activities (Mau et al. 2003; Chen et al. 2011).

Curcuma species from different geographical areas have been analyzed for essential oil composition and bioactivities (Jantan et al. 2012; Devi et al. 2012; Angel et al. 2014; Jena et al. 2017). Major terpenoids such as camphor, 1,8-cineole, isoborneol and curdione have been reported from the essential oil of *C. haritha*, an endemic species from South India (Raj et al. 2008). However, a large number including those endemic to Kerala await analysis. In this first report on unexplored and endemic *C. mutabilis* Škorničk., M.Sabu & Prasanthk. from Western Ghats of Kerala, we have evaluated the chemical composition and bioactivities of rhizome and leaf essential oils. *C. mutabilis* is a 10–60 cm sized herb with light brown ovoid branchless faintly aromatic rhizome and shoot with 2–7 leaves (Skornickova et al. 2004).

2. Results and discussion

Hydro-distillation of fresh rhizomes (500 g) of *C. mutabilis* yielded 0.32 mL (0.06% v/w) and fresh leaves (380g) yielded 0.58 mL (0.15% v/w) of colourless, faintly aromatic volatile oil. GC-MS analysis of rhizome oil revealed the presence of 33 components of the 82.38% identified whilst leaf oil contained 23 components of the 90.09% identified (Table S1).

Monoterpene hydrocarbons, oxygenated monoterpenes, sesquiterpene hydrocarbons and oxygenated sesquiterpenes in rhizome oil were 7.94%, 5.41%, 2.65% and 8.95% respectively in comparison to 0.77%, 0%, 63.92% and 7.58% found in leaf oil. Both rhizome and leaf oil respectively contained 1.37% and 12.22% of diterpenes. Other components including phenolics, carbonylic compounds and hydrocarbons comprised 20.75% and 9.81% of the rhizome and leaf oil respectively. In rhizome oil estrone methyl ether (3-Methoxyestra-1,3,5(10)-trien-17-one) (47.35%) which predominated as the major component was conspicuously absent in leaf oil; other constituents included camphene (4.02%), caryophyllene oxide (3.27%) and 6-octadecanoic acid (2.70%). Although the most commonly found essential oil components of *Curcuma* like α-pinene, camphene, β-pinene and camphor were detected in *C. mutabilis* rhizome, the volatile chemical profile was, nonetheless, found to be unique in comparison to that of *C. longa*, *C. aeruginosa, C. aromatica, C. caesia, C. malabarica, C. sylvatica and C. zedoaria* (Raina et al. 2005; Angel et al. 2014). β-caryophyllene (25.48%), β-farnesene (19.47%), phytol (11.69%), α-humulene (11.01%), caryophyllene oxide (5.41%) and α-fernesene (5.04%) were found to be present in *C. mutabilis* leaf oil and the volatile chemical profile was found similar to that of *C. longa* reported previously (Parveen et al. 2013). Varying amounts of β-pinene, β-panasinsene, β-caryophyllene, α-humulene and caryophyllene oxide were found to be present in both rhizome and leaf oil.

The results of antioxidant activity of *C. mutabilis* essential oils evaluated by DPPH/ABTS methods are given in Table S2. ABTS method was found to exhibit relatively better sensitivity as evidenced by values obtained, which were many folds higher, throughout the concentration range (10 μg – 1.0 mg) tested in a dose-dependent manner. DPPH method was apparently found to be refractory to doses below 500 μg/ml. Depending on the composition of essential oils, synergistic or antagonistic effects of the constituents therein cannot be ruled out. As pointed out by Amorati et al. (2013), the presence of a largely pro-oxidant hydrocarbon constituent fraction of essential oil, is capable of counteracting the antioxidant behavior of oxygenated fraction. This could perhaps be true in the case of leaf oil wherein the pro-oxidant sesquiterpene hydrocarbon amounted to 63.92%. As observed by us for *C. mutabilis*, low antioxidant activity has also been reported for *C. zedoaria* rhizome essential oil at a concentration of 1.0 mg/mL (Mau et al. 2003). Thus factors such as assay conditions as well as oil composition are likely to influence the observed antioxidant activities.

C. mutabilis rhizome oil treatment resulted in potent antiproliferative activity with IC_{50} values of 6.8 and 8.5 μg/mL determined for K562 and HCT116 cells respectively (Figure S1). Our results are in agreement with previous studies which showed that derivatives of the major constituent of rhizome oil, estrone methyl ether, possess antiproliferative activity (Maninder et al. 2003; Mueck and Seeger 2010). However, leaf oil treatment resulted in much lower inhibition of cell proliferation even at the highest dose tested (100 μg/mL) as evidenced by the observation of only 22 and 28% inhibition in K562 and HCT116 cells respectively. The mechanisms underlying antiproliferative activity of essential oils and their constituents, as revealed by more than a hundred studies exhaustively reviewed by Gautam et al. (2014), have been shown to target multiple, often overlapping, cellular regulatory pathways. These include those involved in apoptosis, cell cycle arrest, antimetastatic, antiangiogenic and increased generation of reactive oxygen species. Being complex mixture of several bioactive components, the scope of their synergistic action should also be taken into consideration (Bhalla et al. 2013; Gautam et al. 2014).

3. Conclusion

This study reveals for the first time comparative analyses of the chemical composition and biological activities of volatile oils from rhizome and leaf of the hitherto unexplored and endemic, *C. mutabilis* species. We show that differences in oil composition and assay conditions might influence the observed antioxidant activity. Our results also demonstrate that rhizome oil possessed significantly higher antiproliferative activity compared to leaf oil attributable to the presence of the major constituent estrone methyl ether. *C. mutabilis* should prove to be an important bioresource for isolation and evaluation of mode of action and therapeutic potential of the active constituents.

Acknowledgements

Financial assistance of Govt. of India DBT-Bio-CARe scheme to PRM, PRJ and ST (JRF) is acknowledged. Thanks are also due to Dr. Sabu, Botany Dept., Calicut University, Dr. Prabhukumar (plant authentication) and Dr. Suleiman (GC-MS analysis) from CMPR, Kottakal.

Disclosure statement

No potential conflict of interest was reported by the authors.

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