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Chemical Compositions and Tyrosinase Activity of the Essential Oils of *Alpinia aquatica*

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The essential oils obtained from the fresh rhizome, leaf, and pseudostem of *Alpinia aquatica* Roscoe. were hydrodistillated and analyzed using capillary gas chromatography (GC) and gas chromatography-mass spectroscopy (GC-MS). β-Pinene (11.7%), α-humulene (8.9%), aromadendrene (8.7%), and sabinene (7.7%) were the major components in the rhizome oil. The most abundant components of the leaf oil were germacrene D (21.3%), β-pinene (15.6%) and sabinene (12.1%), while α-humulene (19.8%), germacrene D (15.2%) and β-caryophyllene (8.7%) were the main constituents in the pseudostem oil. Antityrosinase assay revealed that all the three essential oils exhibited weak tyrosinase inhibition activities. The rhizome oil showed the highest inhibition activity with the value of 9.5% for the L-DOPA oxidation.

Keywords: *Alpinia aquatica*, Zingiberaceae, Essential oils, Tyrosinase inhibition.

Alpinia is one of the largest genera of the ginger (Zingiberaceae) family with 230 species found throughout tropical and subtropical Asian countries [1, 2]. Many studies have shown that *Alpinia* has many bioactivities including, antioxidant, anti-inflammatory, anticancer, immunostimulating, hepaprotective, antinociceptive [3], antimicrobial, antidermatophytic [4] and antiviral activities [5]. Hence, it has been used widely in traditional medicines due to the aforementioned pharmacological properties. For example, the rhizomes of *A. galanga* (L.) Willd. are used to cure stomach disorders, rheumatism, tuberculosis, diarrhea, ringworm, vomiting and skin diseases [6]. *A. conchigera* Griff. is used for treating bronchitis, jaundice, headache, vertigo, metritis [7], *A. zerumbet* (Pers.) B.L. Burtt. et Sin. is useful in treatment for anxiety disorder [8]. Besides that, *Alpinia* species are gaining attention for other purposes as well, such as, *A. caerulea* (R.Br.) Bentham is used as a flavouring agent in cooking due to its earthy resinous flavour [9], *A. zerumbet* "Variegata" is used for it's unique fragrance [10].

A. *aquatica* grows up to 10 feet high with a 3-foot spread in low to mid-elevation forests and forms clumps with stems from 1-3 meter high [11]. In further exploration on *Alpinia* species in Malaysia, we investigated the essential oil compositions of *A. aquatica* and their tyrosinase inhibition activity. Chemical compositions of the rhizomes oil of *A. aquatica* which was collected from Muar, Johor had previously been reported by Sirat *et al*. [12]. To the best of our knowledge, there is no report on the leaf and pseudostem oils of *A. aquatica*. Thus, the chemical compositions of the leaf and pseudostem oils of *A. aquatica* were analyzed and are reported here for the first time. Based on analysis of the GC chromatogram of the leaf oil led to the identification of 36 compounds amounting to 83.6% of the total oil composition. Among these compounds were monoterpene hydrocarbons (40.5%), monoterpenoids (1.9%), sesquiterpene hydrocarbons (39.5%) and sesquiterpenoids (2.2%). Germacrene D (21.3%) was the most abundant compound found in the leaf oil followed by β-pinene (15.6%) and sabinene (12.1%) which are monoterpene hydrocarbons. The occurrence of germacrene D as the major constituent in essential oils of *Alpinia* species has not been previously reported.

The pseudostem oil of *A. aquatica* comprised of 37 constituents, accounting for 80.4% of the analyzable oil components. The oil was rich in sesquiterpene hydrocarbons (60.8%), containing mainly α-humulene (19.8%), germacrene D (15.2%) and β-caryophyllene (8.7%) whereas low in monoterpenes hydrocarbons (11.2%) including β-pinene (3.7%). A total of 3.9% monoterpenoids, 4.3% sesquiterpenoids and 0.2% others were also identified in the oil. α-Humulene had previously been detected as the major constituent in the leaves oil of *A. allughas* Rosc. [13]. α-Humulene possessed several therapeutic efficacies such as anti-inflammatory and acts as a fumigant [14].

The analysis of the rhizome oil identified 49 constituents, representing 79.1% of the oil sample. This rhizome oil consisted of 34.7% monoterpene hydrocarbons, 5.8% monoterpenoids, 24.9% sesquiterpene hydrocarbons and 4.9% sesquiterpenoids. β-Pinene (11.7%) and sabinene (7.7%) were predominant in monoterpene hydrocarbons while, α-humulene (8.9%) and aromadendrene (8.7%) were found to be the highest sesquiterpene hydrocarbons in the oil. β-Pinene has been reported as the major compound in fruit oil of *A. pinnanensis* [15], leaf oils of *A. purpurata* (Vieill) K. Schum. [16], leaf and flower oils of *A. hainanensis* and *A. katsumadai*, respectively [17]. On the contrary, the chemical compositions of the rhizomes oil reported in the present investigation were entirely different from the previously isolated oil compositions of this species. Being the major constituents of previously isolated rhizomes oil, β-sesquiphellandrene (37.5%) and 1,8-cineol (21.2%), were completely absent in the rhizomes oil of the current investigation. In addition, β-pinene (0.9%) was detected in scarce amount in the preliminary study [12]. The variation in oil compositions of rhizomes in both studies could be attributed to sampling time, GC conditions and ecotypes or genotypes [18]. The results of this research revealed that the rhizomes yielded highest content of essential oils. Germacrene D (21.3%) and α-humulene (19.8%) were the highest content of sesquiterpenes detected in leaf and pseudostem oils, respectively, while β-pinene (11.7%) was the dominant monoterpene constituent present in the rhizome oils.