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Botany, uses, chemistry and bioactivities of mangrove plants II: *Ceriops tagal*

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Introduction

The genus *Ceriops* consists of *C. tagal*, *C. decandra*, *C. zippeliana* and *C. australis* (Giesen *et al*., 2007; Sheue *et al*., 2009). Of the four species, *C. tagal* has the widest geographical range, stretching from East Africa, Madagascar, South and Southeast Asia through to Papua New Guinea, northern Australia and the West Pacific. Ecologically, *C. tagal* occurs at the landward side of littoral mangroves, sometimes co-existing with *C. decandra*. The latter was considered to be synonymous to *C. zippeliana* until morphological and molecular evidence showed that they are distinct species (Sheue *et al*., 2009). The chemistry and pharmacology of *C. tagal* have been reviewed under the family Rhizophoraceae (Nebula *et al*., 2013) and under the genus *Ceriops* (Wang *et al*., 2012).

Botany and Uses

Ceriops tagal (Perr.) C.B. Robinson (family Rhizophoraceae) is a small tree with short buttresses and knee-like breathing roots (Giesen *et al*., 2007; Baba *et al*., 2013). The bark is pale greyish-brown, smooth in young trees and fissured in old trees (Figure 1). Leaves are simple, shiny, opposite, ovate, and dark green in shade and bright greenish-yellow in full sun. The leaf apex is rounded or notched. Inflorescences are axillary cymes with 5–10 flowers. Flowers have a deeply sunken calyx with five green lobes and five white petals. Fruits are ovoid and dark brown when ripe. Hypocotyls are pendulous, warty and pointed with a yellow collar (Figure 2). The fruit takes four weeks to mature and the collar appears 10 days before detachment of the hypocotyl (Solomon Raju & Karyamsetty, 2008).

In the Can Gio Mangroves, Ho Chi Minh City, Viet Nam, embankments of abandoned salt pans are breached to facilitate tidal flow before planting with *C. tagal* (Figure 3). The bark of *C. tagal* has been used for the treatment of infected wounds in Thailand, and obstetric and hemorrhagic conditions in the Philippines (Bamroongrugsa, 1999). The species is also used to treat sores, haemorrhages and malignant ulcers and malaria in China (Zhang *et al*., 2005a; Wang *et al*., 2012). In the Philippines, the reddish-brown ground bark of *C. tagal* (Figure 4) is added to toddy (an alcoholic drink from the inflorescence sap of coconut) as a preservative to delay the fermentation process by controlling spoilage microbes (Baba *et al*., 2013). The oyster-white toddy turns reddish-brown, and has a slight bitter and astringent taste. The demand for *C. tagal* bark in the Philippines has led to the harvesting and smuggling in Sabah by illegal immigrants (Figure 5). In Indonesia, tannin from *C. tagal* bark is used as dye for batik designs (Bandaranayake, 1998) and in Madagascar, the wood is used for boiling shrimp and fish (Rasolofo, 1997).

Figure 1 Tree of *Ceriops tagal*

Figure 2 Propagules of *Ceriops tagal*

Figure 3 An abandoned salt pan planted with *Ceriops tagal*

Figure 4 Ground bark of *Ceriops tagal*

Figure 5 Smuggled bark of *Ceriops tagal*

Chemistry

Phytochemical analysis of *C. tagal* showed that the major constituents are diterpenoids of tagalsins in leaves, stems, twigs and roots (Zhang *et al*., 2005a, 2005b; Chen *et al*., 2008, 2011; Hu *et al*., 2010; Ouyang *et al*., 2010), and tagalenes in leaves and twigs (Yang *et al*., 2015). Triterpenoids of lupeol, betulin, betulinic acid and cereotagalols have been isolated from aerial parts, roots, fruits and hypocotyls of *C. tagal* (Pakhathirathien *et al*., 2005; Wang *et al*., 2010; Chacha, 2011). Phytochemical constituents of the bark have not been reported except the isolation of a diterpene named *ent*-16β-17α-dihydroxykaurane (Chantrapromma *et al*., 2006).

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Bioactivities

Antibacterial activity: Isolated from roots of *C. tagal*, 16-hydroxyisopimar-8(14)-en-15-one had antibacterial activities with MIC values of 0.5 mg/mL against *Streptococcus pyrogenes*, 0.25 mg/mL against *Salmonella pooni*, and 0.1 mg/mL against *Bacillus cereus*, *Staphylococcus aureus* and *Micrococcus kristinae* (Chacha *et al*., 2008).

Anti-fouling activity: Isolated from roots of *C. tagal*, diterpenoids of methoxy-ent-16-hydroxypimarenely-15-one and ent-8(14)-pimarene-15,16-diol exhibited significant anti-fouling activities against larvae of the barnacle (*Balanus albicostatus*) with EC_{50} values of 0.32 and 0.04 mg/cm^2, respectively (Chen *et al*., 2008). The anti-fouling activity of *C. tagal* warrants further research as it may address barnacle infestation, a problem for mangrove seedlings planted in highly saline environment, which requires close group planting (Figure 6) (Baba *et al*., 2009; Baba, 2011). Maxwell and Li (2006) have earlier reported that *Aegiceras corniculatum* seedlings with rough bark are more prone to barnacle infestation than *Kandelia candel* seedlings with smooth bark.

Figure 6 Stems of planted seedlings of *Rhizophora stylosa* infested with barnacles

Anti-feedant activity: Triterpenes of tagalsins Q, R & U obtained from stems and twigs of *C. tagal*, exhibited moderate anti-feedant activity against third-instar larvae of the coconut leaf beetle (*Brontispa longissima*) at concentration of 1 mg/mL (Hu *et al*., 2010).

Anti-hyperglycaemic activity: When tested on normal healthy sucrose-loaded and streptozotocin (STZ)-induced diabetic rats, the ethanol leaf extract of *C. tagal* significantly improved the glucose tolerance of sucrose-loaded rats by 33%, and resulted in 11% decline in hyperglycaemia in STZ-induced diabetic rats (Tiwari *et al*., 2008).

α-Glucosidase inhibitory activity: Out of six plant species screened for α-glucosidase inhibitory activity, the bark of *C. tagal* ranked second to that of *Rhizophora mucronata* (Lawag *et al*., 2012). IC_{50} values were 0.85 and 0.08 µg/mL, respectively.

Cytotoxic activity: Of 21 triterpenes isolated from fruits and hypocotyls of *C. tagal*, only 3*β*-*E*-feruloylbetulinic acid exhibited potent cytotoxic activity against KB, BC, and NIC-H187 human cancer cells, with IC_{50} values of 3.8, 3.0, and 1.8 µg/mL, respectively (Pakhathirathien *et al*., 2005). Triterpenoids of 3-epibetulinic acid and 3-*O*-acetyl-3-epibetulinic acid acetate isolated from *C. tagal* were found to inhibit H7402 human cancer cells with IC_{50} values of 14.4 and 10.0 mg/mL, and Hela cells with IC_{50} values of 11.8 and 11.3 mg/mL, respectively (He *et al*., 2007). Terpenoids from roots of *C. tagal* have been reported to induce apoptosis of cancer cells through activation of caspase-3 enzyme (Chacha, 2011). Tagalsin extracted from *C. tagal* inhibited H22 hepatoma cells in mice and the mechanism of action might be through the up-regulation of caspase-3 expression and down-regulation of bcl-2 expression (Song *et al*., 2010; Dong *et al*., 2011). Out of six new diterpenes along with 10 known diterpenes were isolated from leaves and twigs of *C. tagal*, tagalenes A & B, and tagalsins A, C, E & F were found to possess cytotoxic properties using the MTT assay (Yang *et al*., 2015). Tagalsin C was the most potent and broad-based with IC_{50} values of less than 10 µM against all human cancer cells of HCT-8, Bel7402, BGC823, A549 and A2780. Recently, it was reported that tagalsins isolated from *C. tagal* inhibited malignant hematologic cells (Neumann *et al*., 2015). The diterpenes induced a ROS-mediated damage of DNA, which led to apoptosis and blockage of cell cycle progression.

Conclusion

Tannin from the bark of *C. tagal* is being used as dye for batik in Indonesia and as preservative for fermentation of toddy in the Philippines. The wood is used for boiling shrimp and fish in Madagascar. Endowed with an array of diterpenoids and triterpenoids as chemical constituents in the various plant parts, *C. tagal* possesses properties of antibacterial, anti-fouling, anti-feedant, anti-hyperglycaemic, α-glucosidase inhibitory and cytotoxic bioactivities.

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