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# 学位論文全文に代わる要約
# Extended Summary in Lieu of Dissertation

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学位論文題目：
Title of Dissertation

Allelopathy of *Cerbera manghas*, *Filicium decipiens* and *Anredera cordifolia* from Indonesia
(インドネシア産*Cerbera manghas*、*Filicium decipiens*、*Anredera cordifolia*のアレロパシー)

学位論文要約：

Dissertation Summary

The term of allelopathy is derived from the Greek' root words *allelon*, 'of each other', and *pathos*, 'to suffer', hence it means, the injurious effect of one upon another. As a sub-discipline of chemical ecology, the study of allelopathy is concerned with the influence of biochemicals produced by one organism on the growth, survival, development, reproduction and distribution of the other organism in an ecosystem. In this case, the biochemicals, or known as allelochemicals may have beneficial or detrimental effects on the target organisms (Cheng & Cheng, 2015). The term of allelopathy to refer to any process, which involving secondary metabolites produced by plants, microorganisms, viruses and fungi that influence the growth and development of other biological systems, in addition, the donor and receiver of allelopathic should include animals (Rice, 1984). Plants are biochemical factories. Biochemicals with allelopathic potential may existent in all part of plant tissues, including leaves, stems, roots, rhizomes, flowers, fruits, and seeds. Allelochemicals may be released from plant tissues in a variety of ways, including volatilization, root exudation, leaching, and decomposition of the plant residues (Putnam, 1985).

As noted by Blum (2014), both organic and inorganic compounds are released from plants to the soil by active and passive processes (Figure 1). Active processes include secretion by glands and hairs, secretion and exudation of high molecular compounds by cells particularly root cells, and the active transport of organic compounds across cell membranes. Passive release includes exudation of low molecular weight compounds by cells, sloughing and lysis of cells, particularly root cells, abscission of vegetative and reproductive tissues and organs, leaching of living, dying, or dead tissues such as leaves, stems, roots, flowers, and fruits by condensates or rain events, and loss of volatile compounds by living and dead vegetative and reproductive tissues. As described by Rizvi, Haque, Singh, & Rizvi, (1992), the mode of action of allelochemicals can be divided into indirect and direct action. Indirect action may include effects through alteration of soil property, its nutritional status and altered population and/or activity of harmful/beneficial organisms like microorganisms, insects, nematodes, etc. Whereas, the direct mode of action, which includes effects of allelochemicals on various aspects of plant growth and metabolism. The following are some important processes known to be influenced by allelochemicals, including (1) cytology and ultrastructure, (2) phytohormones and their balance, (3) membrane and its permeability, (4) germination of pollens/spores, (5) mineral uptake, (6) stomatal movement, pigment synthesis and photosynthesis, (7) respiration, (8) protein synthesis, (9) leghaemoglobin synthesis and nitrogen fixation, (10) specific enzyme activity, (11) conducting tissue, (12) water relations of plants, and (13) genetic material.

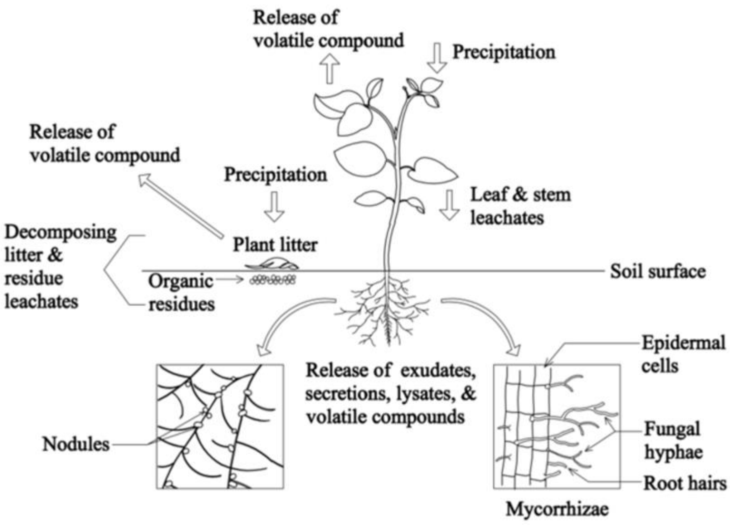


Figure 1. Pathways of organic and inorganic compounds are released from plants (Blum, 2014)

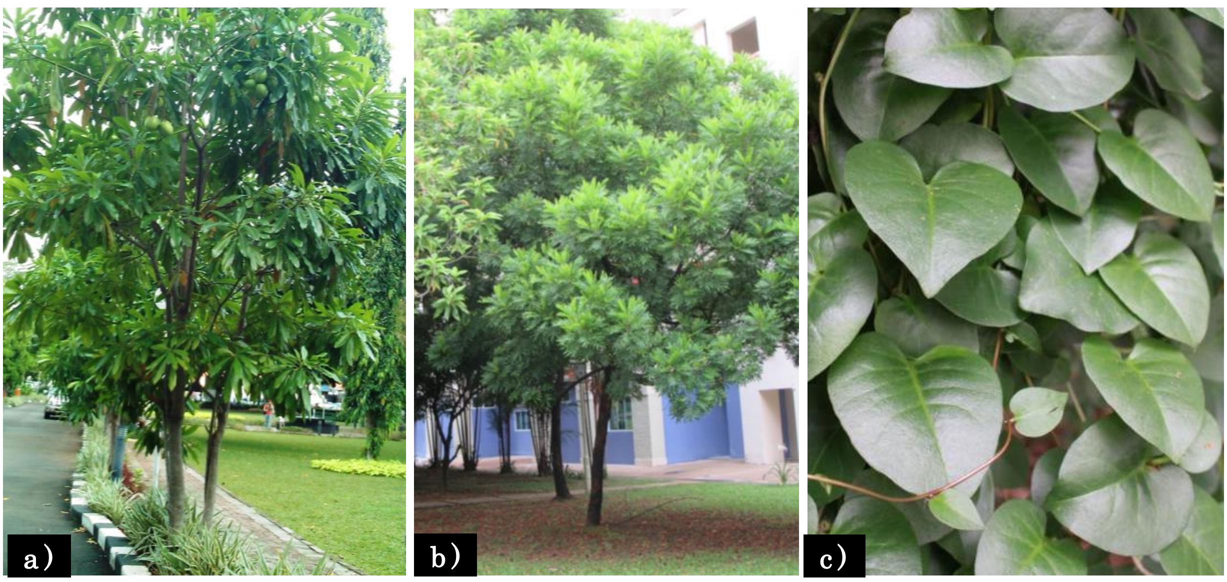


Figure 2. a) Sea mango (*Cerbera manghas*), b) ferntree (*Filicium decipiens*), and
c) Madeira-vine (*Anredera cordifolia*) from Indonesia

The allelochemicals with allelopathic effects are being explored intensively to reduce the dependency on synthetic pesticides in plant pest and diseases management. The present study examined allelopathic effects of sea mango (*Cerbera manghas*), ferntree (*Filicium decipiens*), and Madeira vine (*Anredera cordifolia*) from Indonesia (Figure 2). P-hydroxybenzaldehyde, benzamide, n-hexadecane acid monoglyceride, loliolide, β-sitosterol, cerberin, neriifolin, cerleaside A, and daucosterol were identified in sea mango (Zhang, Pei, Liu, Kang, & Zhang, 2010). Sitosterol *β*-D-glucoside, 3-O-*β*-D-glucopyranosyl kaempferol, 3-O-*β*-D- glucopyranosylquercetin and 3-O-*α*-L-rhamnopyranosyl(1 → 2)-*β*-D-glucopyranosylkaempferol were identified in ferntree (Jayasinghe, Balasooriya, Bandara, & Fujimoto, 2004). Saponins, alkaloids, polyphenols, flavonoid and mono polysaccharide including L-arabinose, D-galaktose, L-rhamnose and D-glucose were identified in Madeira vine (Astuti, Sakinah A.M, Andayani B.M, & Risch, 2011). The three plants are known as medicinal plants locally and the allelopathic potential of the plants are unidentified to date. The experiments used leaves as plants material because the leaf is one of main sources of allelopathic compounds (Khalid, Ahmad, & Shad, 2002; Rice, 1984). Whereas, fresh fruit of sea mango was used for the experiment of *R. argentiventer* repellent due to local experience of Indonesian farmers.

The results the experiments demonstrated that the aqueous methanol extracts of sea mango, ferntree and Madeira vine leaves had inhibitory effects on seedlings growth of test species, including monocot species (barnyard grass, foxtail fescue, Italian ryegrass, and timothy) and dicot species (alfalfa, garden cress, lettuce and rapeseed) under laboratory conditions. The obtained results indicated that the seedling elongation varied depending on test species and also the extract concentrations, whereas the sensitivities of shoots and roots were different among species, and the inhibition of seedling elongation increased along with the increase of the extract concentrations in all test species, in both case of shoots and roots. At some lower extract concentration, growth promotion was observed at root of lettuce and shoot of barnyard grass and foxtail fescue. The similar phenomenon was reported in case of the extract of *Cymbopogon nardus* (Suwitchayanon, 2013), *Leucas aspera* (Islam & Kato-noguchi, 2012) and *Aglaia odorata* (Kato-Noguchi et al., 2016). Dose response phenomenon characterized by a low dose stimulation and a high dose inhibition are known as hormetic responses. The phenomenon of phytotchemical phytotoxins and hormesis were also reviewed by Duke, whose generated a special issue dealing with dose-response aspects of allelochemicals (Duke, 2011). In Conclusion, these results suggested that sea mango, ferntree and Madeira vine may possess allelopathic substances. The crude extract of these plants leaves could be recommended to apply directly as natural herbicide or the residue also could be applied as mulch cover for weed control in farming practices, for practical purposes.

The results of experiment of the sea mango also suggested that sea mango fruit can be used as a natural repellent for *R. argentiventer*. Symptom of stress detected in the behaviour of *R. argentiventer*. Stress causes various physiological process disorders in mammals, including sleep–wake cycles, hormonal and nervous activity, body temperature, and exhibiting rhythmic changes over the course of 24h (Barker, Bobrovskaya, Howarth, & Whittaker, 2017; Hurtubise & Howland, 2016; Tahara, Aoyama, & Shibata, 2016). In these experiments, the comparison of metabolic parameters of *R. argentiventer* between with and without sea mango fruit treatments indicated that sea mango caused metabolic disorders. Sea mango fruit treatment decreased food and beverage intake by *R. argentiventer* and decreased its body weight significantly. Comparing the daily activities between with and without sea mango fruit treatments, *R. argentiventer* spent more time for locomotion and less time for foraging and resting. The results indicated that sea mango fruit has a potential as repellent for *R. argentiventer*. To date, sea mango fruit has no economic value yet in Indonesia. The fruits have not been utilised for any purposes. Farmers can find the fruits under their trees freely and easily. For practical purposes, farmers can put the sea mango fruit directly at any places where *R. argentiventer* are found.

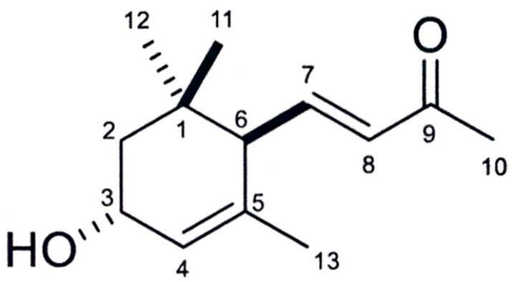


Figure 3. Structure of 3-Hydroxy-α-ionone

In this study, 3-hydroxy-α-ionone was isolated from Madeira vine leaf (Figure 3). The $C_{13}H_{21}0_2$ was determined as molecular formula of the allelopathic substance, which was characterized by HRESIMS at *m/z* 209.1530 $[M+H]^+$ (calcd for $C_{13}H_{21}0_2$, 209.1542, Δ = -1.2 mmu). The $^1$H NMR spectrum (400 MHz, $CDCl_3$ as internal standard) showed $\delta_H$ 6.53 (dd, *J*= 15.9, 10.3 Hz, I H, H-7), 6.10(d,*J*= 15.9, I H, H-8), 5.63 (br s, I H, H-4), 4.27 (br s, I H, H-3), 2.50 (d,*J*= 10.3, I H, H-6), 2.26 (s, 3 H, H-10), 1.84 (dd, J = 13.9, 6.1, I H, H-2), 1.62 (d, J = 0.7, 3 H, H-13), 1.40 (dd, *J*= 13.9, 6.7, I H, H-2'), 1.03 (s, 3 H, H-11), 0.89 (s, 3 H, H-12). The $^{13}$C NMR spectrum (100 MHz, $CDCl_3$ as internal standard) showed $\delta_c$ 198.3 (C-9), 147.3 (C-7), 135.6 (C-5), 133.8 (C-8), 125.9 (C-4), 65.6 (C-3), 54.4 (C-6), 44.0 (C-2), 34.0 (C-1), 29.4 (C-11), 27.3 (C-10), 24.8 (C-12), 22.8 (C-13). The specific rotation of the compound was $[\alpha]_D^{25} + 99$ (*c* 0.10, $CH_2Cl_2$). The active substance was determined as 3-hydroxy-α-ionone by referring the data compared with previous report (D'Abrosca et al., 2004).

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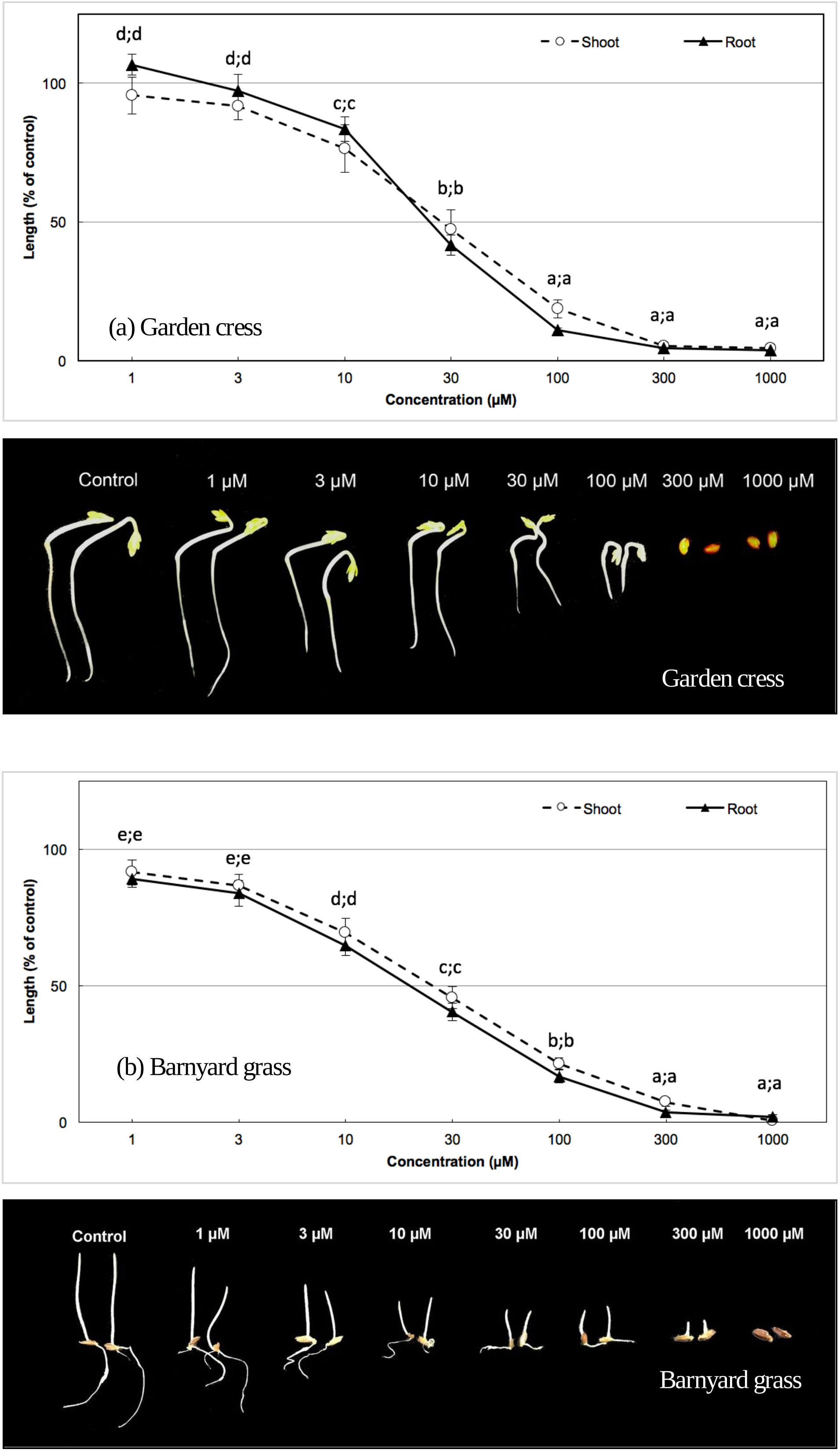


Figure 4. The 3-Hydroxy-α-ionone activity on seedling development of (a) garden cress and (b) barnyard grass. Values represent means ± SE from two replicate Petri dishes for each treatment (n = 20). Different letters indicated significantly different value of length at each concentration (Duncan, $P<0.05$)

The ionone and its derivatives involves in the terpenoids metabolism as essential intermediates (Lutz-Wahl et al., 1998). The substance have been isolated from various sources (Pabst, Barron, Sémon, & Schreier, 1992). In this research, the substance showed the ability to inhibit the seedling development of garden cress and barnyard grass (Figure 4). A similar substance, 3-hydroxy-β-ionone was isolated from moss *Rhynchostegium pallidifolium* (Kato-Noguchi & Seki, 2010). The substance was reported as a growth inhibitor in garden cress bioassay. Comparing 3-hydroxy-α-ionone and 3-hydroxy-β-ionone in garden cress bioassay, the $IC_{50}$ values were 35.60 µM; 38.03 µM (shoot; root) and 14.90 µM; 16.3 µM (shoot; root), respectively. This result suggested that 3-hydroxy-α-ionone may be less strong in activity than 3-hydroxy-β-ionone as allelopathic substance. Another ionone derivate, 3-hydroxy-5,6-epoxy-β-ionone was isolated from *Athyriurn yokoscense*. The substance was reported as inhibitor in lettuce seed germination (Kurokawa et al., 1998). In conclusion, the isolated compound, 3-hydroxy-α-ionone was one of the active substance that responsible for the allelopathic effect.

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