

学位論文要旨 Dissertation Abstract

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学位論文題目 : Assessment of Allelopathic Potential and Allelopathic
Title of Dissertation Substances in Myanmar Medicinal Plants for Weed Control
(雑草防除を目的としたミャンマー在来薬用植物のアレロパ
シーとその原因物質の探索)

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Plant allelopathy has been studied and investigated for a long time as an alternative weed control method to reduce the dependence on synthetic herbicides for sustainable weed management. The infestation of weeds reduces agricultural crop production and produce quality by competing with cultivated crops for available resources. Herbicides are a quick response to effectively suppress weed species, and many agricultural settings totally rely on them. However, heavy application of synthetic herbicides negatively affects human health and increases the number of herbicide-resistant weed biotypes. For these negative effects, searching for allelopathic plants and identifying allelopathic compounds with strong inhibitory activity are receiving the most attention. Among different plant groups, medicinal plants are described to have allelopathic potential and possess bioactive substances with pharmacological and allelopathic properties. However, many medicinal plants are still lacking sufficient evidence related to their allelopathic activity and potential allelopathic substances. Therefore, this research was conducted to assess the allelopathic potential and identify allelopathic substances from Myanmar medicinal plants.

Three medicinal plants used in this research are *Marsdenia tenacissima* (Roxb.) Moon, *Croton oblongifolius* Roxb., and *Aegle marmelos* (L.) Correa. Aqueous methanol extracts of *M. tenacissima*, *C. oblongifolius*, and *A. marmelos* significantly limited the seedling growth of cress, lettuce, alfalfa, barnyard grass, Italian ryegrass, and timothy in a concentration-dependent manner. The extract concentration of 300 mg DW (milligram dry weight) equivalent extract/mL totally inhibited the shoots and roots growth of examined plant species compared with the control, except for the shoots growth of barnyard grass. Concentrations of the leaf extracts of three medicinal plants needed for 50% plant (shoot and root) growth limitation (I_{50} values) ranged from 0.80–54.20, 3.22–65.75, and 1.61–36.14 mg DW equivalent extract/mL, respectively. The inhibitory effects of the

aqueous methanol extracts of three medicinal plants suggest that these plants may contain allelopathic potential and may involve allelopathic substances.

Therefore, the potent allelopathic compounds from the leaf extracts of *M. tenacissima*, *C. oblongifolius*, and *A. marmelos* were purified through bioassay-directed chromatographic steps. Then, molecular structures of isolated compounds were identified by spectral data analyses (HRESI-MS, $^1\text{H-NMR}$ and $^{13}\text{C-NMR}$). Three active substances were isolated from leaf extracts of *M. tenacissima*, and identified as steroidal glycoside-1 (a novel compound), 3-*O*-[6-deoxy-3-*O*-methyl- β -allopyranosyl(1 \rightarrow 4)- β -oleandropyranosyl]-5,6-dihydrogen-11 α -*O*-acetyl-12 β -*O*-tigloyl-17 β -marsdenin; steroidal glycoside-2, 3-*O*-[6-deoxy-3-*O*-methyl- β -allopyranosyl(1 \rightarrow 4)- β -oleandropyranosyl]-5,6-dihydrogen-11 α ,12 β -di-*O*-tigloyl-17 β -marsdenin; and steroidal glycoside-3 (a novel compound), 3-*O*-[β -cymaropyranosyl(1 \rightarrow 4)- β -cymaropyranosyl]-8-dehydroxy-11 β -*O*-acetyl-12 β -*O*-tigloyl-17 β -marsdenin. These compounds significantly suppressed the seedling growth of cress in a concentration-dependent way. The I_{50} values for shoots and root growth of cress were 0.46 and 0.03 mM for steroidal glycoside 1, 0.74 and 0.12 mM for steroidal glycoside 2, and 0.25 and 0.03 mM for steroidal glycoside 3, respectively. These results indicate that identified compounds may be responsible for the allelopathy of *M. tenacissima*.

Four active compounds; (3*R*,6*R*,7*E*)-3-hydroxy-4,7-megastigmadien-9-one, 2-hydroxy alpinolide (a novel compound), alpinolide and epialpinolide were isolated from *C. oblongifolius*. The I_{50} value for the shoot and root growth of cress were 0.24 and 0.15 mM for (3*R*,6*R*,7*E*)-3-hydroxy-4,7-megastigmadien-9-one, 0.11 and 0.04 mM for 2-hydroxy alpinolide, 0.12 and 0.07 mM for alpinolide and 0.16 and 0.09 mM for epialpinolide, respectively. The results suggest that these identified compounds might contribute to the allelopathy of *C. oblongifolius*.

Four active substances; umbelliferone, (*E*)-4-hydroxycinnamic acid, methyl ester, cinnamic acid, and methyl (*E*)-3'-hydroxyl-4'-methoxycinnamate, were identified from *A. marmelos*. The I_{50} values of shoots and roots growth of cress were 0.38 and 0.15 mM for umbelliferone, 0.18 and 0.08 mM for 2(*E*)-4-hydroxycinnamic acid methyl ester, 0.12 and 0.07 mM for cinnamic acid, and 0.26 and 0.18 mM for methyl (*E*)-3'-hydroxyl-4'-methoxycinnamate, respectively. The results indicate that these phytotoxic substances might be responsible for the allelopathic activity of *A. marmelos*.

In this study, the extract of three medicinal plants exhibited allelopathic potential against the seedling growth of the examined plant species, which indicate that these plants may possess allelopathic substances. Eleven inhibitory substances were isolated from the leaf extracts of *M. tenacissima*, *C. oblongifolius*, and *A. marmelos*, and these compounds affected the shoot and root growth of cress at different concentration interval. Therefore, the living and dead plant residues of these medicinal plants and their plant extracts could be used as a soil-additive resource for sustainable weed control, and their inhibitory substances could be utilized as a potential candidate for developing natural herbicides.