

A New Phenylalkanoid of *Alpinia Galangal*Tang Yu Kuo¹, Hsuan En Chen², Hung Chun Yeh³, Chung Yi Chen^{3*} and Su Ling Liu^{4*}

¹Kaohsiung Municipal Kaohsiung Senior High School, Sanmin Dist., 807302, Kaohsiung City, Taiwan.

²Kaohsiung Municipal Hsin Chuang Senior High School, Zuoying Dist., 81368, Kaohsiung City, Taiwan.

³School of Medical and Health Sciences, Fooyin University, 83102, Kaohsiung City, Taiwan.

⁴Experimental Forest, College of Bio-Resources and Agriculture, National Taiwan University, Nantou County, Taiwan.

***Correspondence:**

Chung Yi Chen, School of Medical and Health Sciences, Fooyin University, 83102, Kaohsiung City, Taiwan.
Su Ling Liu, Experimental Forest, College of Bio-Resources and Agriculture, National Taiwan University, Nantou County, Taiwan.

Received: 07 Jun 2022; Accepted: 15 Jul 2022; Published: 19 Jul 2022

Citation: Tang Yu Kuo, Hsuan En Chen, Hung Chun Yeh, et al. A New Phenylalkanoid of *Alpinia Galangal*. Chem Pharm Res. 2022; 4(3): 1-3.

ABSTRACT

A new phenylalkanoid, undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate (**1**) was isolated from the rhizomes of *Alpinia galangal* (Zingiberaceae). The structure of the new phenylalkanoid was elucidated by chemical and physical evidence.

Keywords

Alpinia galangal, Zingiberaceae, Rhizome, Phenylalkanoid.

Introduction

Traditional system of medicinal consists of large number of plants with various medicinal and pharmacological importances and hence represents a priceless tank of new bioactive molecules. *Alpinia galangal* (Zingiberaceae) is one amongst these, found all over the world [1]. It is used in medication, culinary and cosmetics for centuries. It is widely used in dietary intake as well as in the traditional system of medicine, viz. Ayurveda, Unani, Chinese and Thai folk medicine. It has a pungent, hot and spicy taste with an aromatic ginger like odour [2]. Different parts of this plant are traditionally claimed to be used for the treatment of ailments including anti-fungal, anti-tumor, anti-helminthic, anti-diuretic, anti-ulcerative, disease of heart, rheumatic pains, chest pain, dyspepsia, fever, diabetes, burning of liver and kidney disease to list of few [1]. In the course of screening for biologically and chemically novel agents from Formosan Zingiberaceous plants [3-31], *A. galangal* was chosen for further phytochemical investigation. In continuation of some studies of chemotaxonomy and biologically active metabolites from this plant, a methanol extraction of the rhizomes of *A. galangal* afforded 20 compounds, including 2 phenylalkanoids, 3 sesquiterpenes, 2 steroids, 8 flavonoids and 5

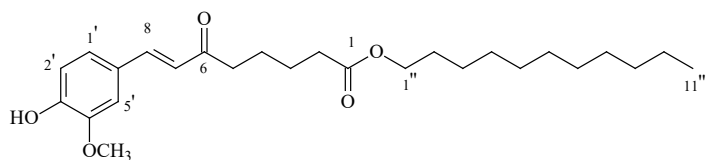
benzenoids [32]. In this paper, we report the isolation and structural elucidation of the new phenylalkanoid (undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate).

Undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate (**1**), a yellow oil, was deduced as C₂₆H₄₀O₅ by HRMS-ESI (*m/z* 455.2771 [M + Na]⁺; calc. 455.2773). Two IR bands at ν_{\max} 3400 and 1650 cm⁻¹ two signal appearing at δ 167.4 and 178.3 in the ¹³C NMR spectrum suggested that hydroxyl group and two carbonyl groups might be present. The ¹H NMR spectrum of **1** showed three aromatic protons at δ 6.91 (1H, d, *J* = 8.4 Hz, H-2'), 7.06 (1H, d, *J* = 2.0 Hz, H-5') and 7.08 (1H, dd, *J* = 8.4, 2.0 Hz, H-1'), four methylene protons at 4.18 (2H, t, *J* = 6.8 Hz, H-5), 2.34 (2H, t, *J* = 7.6 Hz, H-2), 1.69 (2H, m, H-4), 1.63 (2H, m, H-3), two methine protons at δ 6.29 (1H, d, *J* = 16.0 Hz, H-7) and 7.61 (1H, d, *J* = 16.0 Hz, H-8) and one methyl protons at δ 3.93 (3H, s, OCH₃), indicating that **1** was probably a disubstituted phenylalkanoid. The resonance of the ester moiety included those of the ester carbonyl (δ 178.3), one terminal methyl group, and those for the remaining methylenes of the aliphatic chain (Table 1). The carbons of **1** were assigned, from ¹³C-NMR and DEPT experiments, one methyl at δ 14.0 (C-11''), one methoxy at δ 55.9 (OCH₃), 14 methylenes at δ 64.6 (C-5), 33.8 (C-2), 29.3 (C-4), 24.7 (C-3), 26.0 (C-1'') and 28.8~29.7 (C-2''~C-10''), five methines at δ 109.3 (C-5'), 114.7 (C-

2'), 115.7 (C-7), 123.0 (C-1') and 144.6 (C-8), and five quaternary carbons at δ 127.0 (C-6'), 146.7 (C-3'), 147.9 (C-4'), 167.4 (C-6) and 178.3 (C-1). COSY correlations were observed between H-1 and H-2', between H-7 and H-8, between H-2, H-3, H-4 and H-5 and between H-1' to H-11'. The observation of the NOESY correlations from H-5' to methoxyl group suggested that methoxyl group was in the C-4' of this structure. Thus, the structure of this compound was determined to be undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate, which was further confirmed by HMBC experiments (Table 1).

Table 1: ^{13}C (100 MHz, CDCl_3) and ^1H HMR (400 MHz, CDCl_3) data of undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate (**1**).

C#	δ_{C}	δ_{H}	mult., J (Hz)	HMBC ($^1\text{H} \rightarrow ^{13}\text{C}$)
1	178.3	-	-	-
2	33.8	2.34	t, 7.2	C-1, C-3
3	24.7	1.63	m	C-1, C-2, C-4, C-5
4	29.3	1.69	m	C-2, C-3, C-5, C-6
5	64.6	4.18	t, 6.8	C-3, C-4, C-6, C-7
6	167.4	-	-	-
7	115.7	6.29	d, 16.0	C-6, C-8
8	144.6	7.61	d, 16.0	C-7, C-6'
1'	123.0	7.08	dd, 8.4, 2.0	C-2', C-6'
2'	114.7	6.91	d, 8.4	C-1', C-3'
3'	146.7	-	-	-
4'	147.9	-	-	-
5'	109.3	7.06	d, 2.0	C-4', C-6'
6'	127.0	-	-	-
4'-OCH ₃	55.9	3.93	s	C-3', C-4', C-5'
1''	26.0	1.39	m	C-1'', C-2'', C-3''
2''~10''	29.7~28.8	1.28	br s	C-1'', C-11''
11''	14.0	0.88	t, 6.5	C-9'', C-10''



Experimental

General. UV spectra were obtained in MeCN, IR spectra were measured on a Hitachi 260-30 spectrophotometer. ^1H NMR (400 MHz), ^{13}C NMR (100 MHz), HETCOR, HMBC, COSY and NOESY spectra were obtained on a Varian (Unity Plus) NMR spectrometer. Low-resolution ESI-MS spectra were obtained on an API 3000 (Applied Biosystems) and high-resolution ESI-MS spectra on a Bruker Daltonics APEX II 30e spectrometer. Silica gel 60 (Merck, 70–230 mesh, 230–400 mesh) was used for column chromatography. Precoated Silica gel plates (Merck, Kieselgel 60 F-254), 0.20 mm and 0.50 mm, were used for analytical TLC and preparative TLC, respectively, and visualized with 50% H_2SO_4 .

Plant Material

The rhizomes of *A. galangal* were collected from Chiayi County, Taiwan, in April 2015. Dr. Fu-Yuan Lu (Department of Forestry and Natural Resources College of Agriculture, National Chiayi

University) identified plant material. A voucher specimen was deposited at the Department of Medical Technology, School of Medical and Health Sciences, Fooyin University, Kaohsiung, Taiwan.

Extraction and Isolation

The rhizomes (3.24 kg) of *A. galangal* were extracted repeatedly with MeOH (10 L x 3) at room temperature for 24–48 hrs. The MeOH extract was dried and evaporated to leave a viscous residue (144.8 g). The residue was placed on a silica gel column (3.5 kg, 70–230 mesh) and eluted with CH_2Cl_2 gradually enriched with MeOH to afford 8 fractions. Part of fraction 3 (22.3 g) was subjected to silica gel chromatography (0.9 kg, 70–230 mesh) by eluting with *n*-hexane-acetone (90:1), enriched with acetone to furnish five fractions (3-1–3-5). Part of fraction 3-2 (6.8 g) was further purified on a silica gel column using *n*-hexane/acetone mixtures to obtain mixture of undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate (**1**) (16 mg).

Undecyl (*E*)-8-(4-hydroxy-3-methoxyphenyl)-6-oxooct-7-enoate (**1**)

Colorless oil. UV λ_{max} (MeCN, log ϵ) 211 (2.65), 262 (3.65), 282 (2.45) nm. IR (ν_{max} , cm^{-1}): 3400 (OH), 1650 (C=O). ESI-MS m/z 455 $[\text{M}+\text{Na}]^+$; HR-ESI-MS m/z 455.2771 $[\text{M}+\text{Na}]^+$ (calcd for $\text{C}_{26}\text{H}_{40}\text{O}_5\text{Na}$, 455.2773). ^1H and ^{13}C NMR data, see Table 1.

Acknowledgment

This investigation was supported by a grant from the Fooyin University.

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