Biphenylquinolizidine Alkaloids from *Lagerstroemia indica*

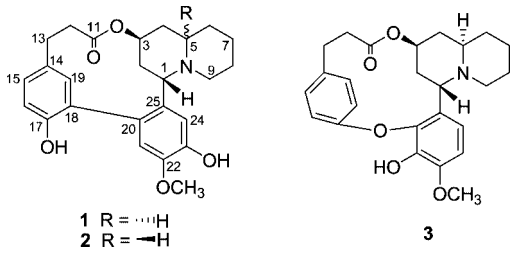
Hong Jin Kim,[†] Ik Soo Lee,[†] UiJoung Youn,[†] Quan Cheng Chen,[†] Tran Minh Ngoc,[†] Do Thi Ha,[†] Hongguang Liu,[§] Byung Sun Min,[‡] Jin Yong Lee,[§] Rack Seon Seong,[⊥] and KiHwan Bae*[,†]

College of Pharmacy, Chungnam National University, Daejeon 305-764, Korea, College of Pharmacy, Catholic University of Daegu, Gyeongsan 712-702, Korea, Department of Chemistry, Sungkyunkwan University, Suwon 440-746, Korea, and Korea Food and Drug Administration, Seoul 122-704, Korea

Received September 30, 2008

Two new biphenylquinolizidine alkaloids, 5-*epi*-dihydrolyfoline (**1**) and its stereoisomer, dihydrolyfoline (**2**), along with lagerine (**3**) were isolated from the aerial parts of *Lagerstroemia indica*. The structures of compounds **1**−**3** were elucidated by extensive spectroscopic techniques.

The plant *Lagerstroemia indica* L. (Lythraceae) is a decorative shrub common in China, Japan, and Korea.[1] It flowers early in the summer and produces seedpods that mature in the early fall. Preliminary studies showed that the alkaloids are concentrated in the seedpods with only trace amounts in the leaves and stems.[2] The isolation and structure determination of the phenylquinolizidine alkaloids that occur in plants of the Lythraceae family is due mainly to the work of Ferris and co-workers,[2−7] who investigated the species *Decodon verticillatus* (L.) Ell. (swamp loosestrife), and Schwarting and co-workers,[8−10] who examined *Heimia salicifolia* Link and Otto ('Hauchinal') and other *Heimia* species. After the report of phenylquinolizidine alkaloids from *L. indica* by Ferris and co-workers,[6] no other study has been carried out on the alkaloids of this plant. In our research on the alkaloids of this plant, two new biphenylquinolizidine alkaloids, 5-*epi*-dihydrolyfoline (**1**) and dihydrolyfoline (**2**), which exist as a diastereomeric pair differing only in the configuration at C-5, were isolated along with lagerine (**3**). We report herein the structures of the two new alkaloids, 5-*epi*-dihydrolyfoline (**1**) and dihydrolyfoline (**2**), as well as lagerine (**3**)[6,11,12] by presenting the complete ^{1}H and ^{13}C NMR assignments.



Aerial parts of *L. indica* were collected from Daejeon, Korea, and extracted with 70% ethanol. The extract was subjected to solvent−solvent extraction and repeated column chromatography on Si gel to obtain the alkaloids 5-*epi*-dihydrolyfoline (**1**), dihydrolyfoline (**2**), and lagerine (**3**). The structures of compounds **1**−**3** were determined using 1D and 2D NMR experiments in conjunction with the analysis of mass spectra and other spectroscopic data.

Compound **1** was obtained as a dark yellow powder and gave a positive Dragendorff's test. The molecular formula $C_{25}H_{29}NO_5$ was determined on the basis of the $[M]^+$ peak (*m*/*z* 423.2028, Δ −1.8 mmu) in the HREIMS spectrum and NMR data. The fragment ion observed at *m*/*z* 137 in the EIMS spectrum of **1** suggested the presence of a quinolizidine moiety in the structure. The ^{1}H NMR spectrum of **1** showed five aromatic resonances at δ_H 6.95 (dd, J = 8.0, 2.0 Hz), 6.86 (s), 6.82 (d, J = 8.0 Hz), 6.80 (s), and 6.69 (d, J = 2.0 Hz) and one *O*-methyl resonance (δ_H 3.77) along with aliphatic peaks. The 25 carbons present in the ^{13}C NMR spectrum (Table 1) comprised one *O*-methyl, eight methylene, eight methine, and eight quaternary carbons by using a DEPT experiment. Nine resonances [δ_C 19.0 (CH_2, C-8), 25.7 (CH_2, C-7), 26.2 (CH_2, C-6), 34.5 (CH_2, C-4), 38.8 (CH_2, C-2), 47.8 (CH, C-1), 50.1 (CH_2, C-9), 57.3 (CH, C-5), 71.1 (CH, C-3)] in the ^{13}C NMR spectrum seemed to be due to a *cis*-fused quinolizidine ring, as observed in decamine.[13] The downfield aliphatic carbons at δ_C 47.8, 50.1, and 57.3 indicated an adjacent nitrogen atom. The resonance at δ_C 71.1, which correlated with a proton at δ_H 4.89 in the HMQC spectrum, indicated oxygenation. The HMQC experiment revealed five aromatic methine carbons (δ_C 112.6, 114.0, 116.4, 129.0, and 132.0) of the two phenyl rings. HMBC correlations (Figure 1) observed for H-1 (δ_H 4.00) to C-9 (δ_C 50.1), C-5 (δ_C 57.3), C-3 (δ_C 71.1), C-24 (δ_C 114.0), C-20 (δ_C 127.5), C-25 (δ_C 132.8), and C-23 (δ_C 146.9) indicated that C-1 was the point of attachment of a phenyl ring. A proton singlet resonating at δ_H 6.80 (H-21) showed HMBC correlations with C-1 (δ_C 47.8), C-18 (δ_C 126.0), C-25 (δ_C 132.8), C-23 (δ_C 146.9), and C-22 (δ_C 146.4), and H-19 (δ_H 6.69) showed HMBC correlations with C-13 (δ_C 32.5), C-20 (δ_C 127.5), C-15 (δ_C 129.0), and C-17 (δ_C 152.3), suggesting that the two phenyl rings were linked between C-18 and C-20. H-12 and H-12′, appearing at δ_H 2.54 and 2.19, showed COSY correlations with H-13 (δ_H 3.00) and H-13′ (δ_H 2.71). Furthermore, H-12 and H-12′ showed HMBC correlations with the C-11 carbonyl carbon (δ_C 174.6) and C-14 (δ_C 131.6), and H-13 and H-13′ showed HMBC correlations with the C-11 carbonyl carbon (δ_C 174.6), C-15 (δ_C 129.0), C-14 (δ_C 131.6), and C-19 (δ_C 132.0), indicating that the carbonyl carbon and phenyl ring were linked by two methylene carbons. The HMBC correlation of the broad H-3 singlet (δ_H 4.89) with C-11 (δ_C 174.6) suggested formation of another ring by attachment of the lactone, and the fragment ion peak at *m*/*z* 379 $[M - CO_2]^+$ in the EIMS spectrum confirmed the presence of the lactone moiety. Additionally, the *O*-methyl proton (δ_H 3.77) showed HMBC correlation with C-22 (δ_C 146.4). The relative configuration of C-5 was confirmed using its ROESY correlation. H-1 and H-3, being axially and equatorially oriented on biogenetic considerations,[14,15] respectively, were used as the basis for correlations. H-1, at δ_H 4.00, showed ROESY cross-peaks with H-2eq, H-6ax, and H-8ax, while there was no detectable interaction between H-1 and H-5. Furthermore H-5 had ROESY cross-peaks with H-4eq, H-4ax, H-6eq, H-7ax, and H-9ax. These ROESY results clearly indicated the *cis*-quinolizidine structure that placed H-5 in an equatorial geometry in the six-membered ring containing carbons 1−4. The

* To whom correspondence should be addressed. Tel: +82-42-821-5925. Fax: +82-42-823-6566. E-mail: baekh@cnu.ac.kr.
[†] Chungnam National University.
[‡] Catholic University of Daegu.
[§] Sungkyunkwan University.
[⊥] Korea Food and Drug Administration.

10.1021/np800619g CCC: $40.75
Published on Web 03/04/2009

Table 1. NMR Data[a] of Compounds **1** and **2**

	1[b]				**2**[c]			
no.	^{1}H (δ)	m (*J*)	^{13}C (δ)	HMBC (H→C)	^{1}H (δ)	m (*J*)	^{13}C (δ)	HMBC (H→C)
1	4.00	d (11.0)	47.8, CH	3, 5, 9, 20, 23, 24, 25	2.99	d (11.0)	59.6,[g] CH	3, 5, 9, 20, 23, 24, 25
2	e[d] 2.33	br d (15.0)	38.8, CH_2		e 2.36	br d (15.0)	39.1, CH_2	
	a[e] 1.66	br t (13.5)			a 1.80	br t (13.5)		
3	4.89	br s	71.1, CH	1, 5, 11	4.97	br s	70.7, CH	1, 5, 11
4	e 1.57	br d (15.0)	34.5, CH_2		e 1.65[f]		37.4, CH_2	
	a 1.99	ddd (15.0, 6.5, 3.5)			a 1.65[f]			
5	2.99	dd (13.5, 2.5)	57.3, CH	1, 3	1.97	dd (14.5, 9.0)	59.7,[g] CH	1, 3
6	e 0.98	br d (13.5)	26.2, CH_2		e 1.47		33.5, CH_2	
	a 1.66	br t (13.5)			a 1.25			
7	e 1.44	br d (12.5)	25.7, CH_2		e 1.61		24.8, CH_2	
	a 1.14−1.11	m			a 1.14			
8	e 0.82−0.78[f]	m	19.0, CH_2		e 1.44[f]		26.2, CH_2	
	a 0.82−0.78[f]	m			a 1.44[f]			
9	e 2.81	br d (12.5)	50.1, CH_2		e 2.66	br d (11.0)	52.9, CH_2	
	a 2.29	br td (10.0, 4.0)			a 1.21			
11			174.6, qC				174.4, qC	
12	2.54	ddd (12.5, 5.5, 2.5)	38.8, CH_2	11, 14	2.61	ddd (12.5, 5.5, 2.5)	38.5, CH_2	11, 14
12′	2.19	td (13.0, 2.5)		11, 14	2.28	td (13.0, 2.5)		11, 14
13	3.00	td (13.5, 2.5)	32.5, CH_2	11, 14, 15, 19	3.14	td (13.5, 2.5)	32.7, CH_2	11, 14, 15, 19
13′	2.71	ddd (13.5, 5.5, 2.5)		11, 14, 15, 19	2.79	ddd (13.5, 5.5, 2.5)		11, 14, 15, 19
14			131.6, qC				132.5, qC	
15	6.95	dd (8.0, 2.0)	129.0, CH	13, 17, 19	7.08	dd (8.0, 2.0)	129.4, CH	13, 17, 19
16	6.82	d (8.0)	116.4, CH	14, 17, 18	6.96	d (8.0)	116.1, CH	14, 17, 18
17			152.3, qC				151.6, qC	
18			126.0, qC				125.9, qC	
19	6.69	d (2.0)	132.0, CH	13, 15, 17, 20	6.84	d (2.0)	131.6, CH	13, 15, 17, 20
20			127.5, qC				126.0, qC	
21	6.80	s	112.6, CH	1, 18, 22, 23, 25	6.83	s	111.0, CH	1, 18, 22, 23, 25
22			146.4, qC				145.6, qC	
23			146.9, qC				146.6, qC	
24	6.86	s	114.0, CH	1, 20, 22, 23	7.17	s	114.4, CH	1, 20, 22, 23
25			132.8, qC				136.6, qC	
−OCH_3	3.77	s	55.9	22	3.88	s	56.3	22

[a] ^{1}H (500 MHz) and ^{13}C NMR (75 MHz) (δ_H 7.24, δ_C 77.0). Carbon multiplicities were determined by DEPT 135° experiment. qC = quaternary, CH = methine, CH_2 = methylene, CH_3 = methyl carbons. Chemical shifts are expressed in ppm and *J* values in Hz. [b] Recorded in $CDCl_3$ + CD_3OD. [c] Recorded in $CDCl_3$. [d] e: equatorial proton. [e] a: axial proton. [f] Signal pattern unclear due to overlapping of two geminal protons. [g] Positions can be interchanged.

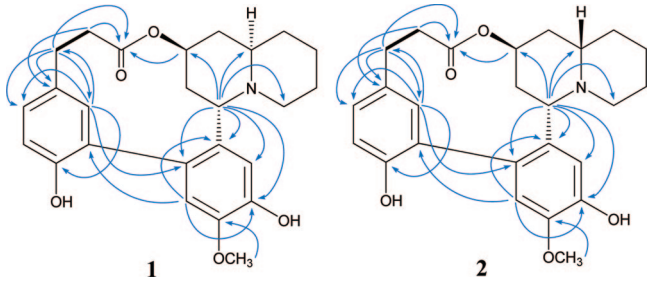
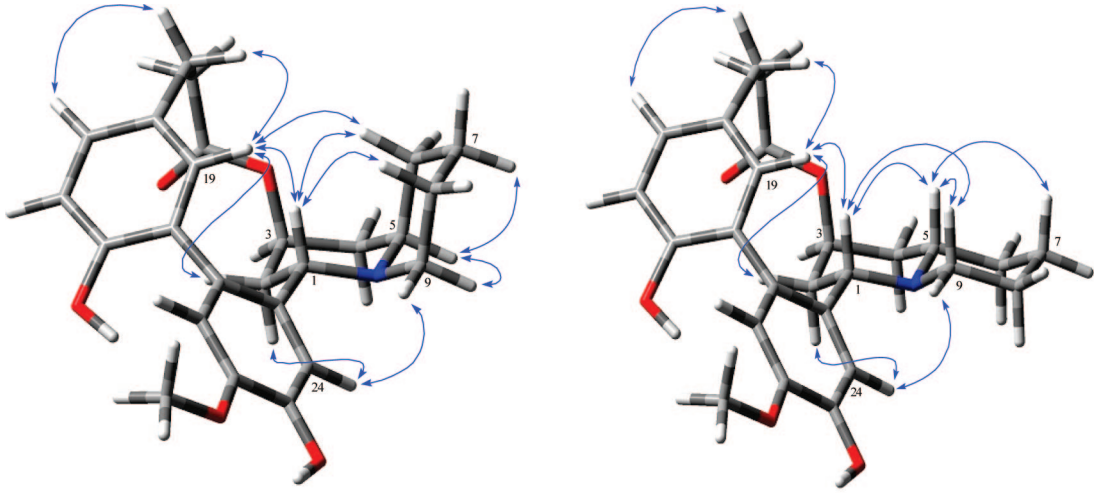


Figure 1. Selected HMBC (H→C) and ^{1}H−^{1}H COSY (−) correlations of **1** and **2**.

minimum energy conformation of **1** and key ROESY correlations are shown in Figure 2. Thus, compound **1** was elucidated as 5-*epi*-dihydrolyfoline (**1**).

Compound **2** was obtained as a yellow powder and gave a positive Dragendorff's test. Its molecular formula was deduced to be $C_{25}H_{29}NO_5$ (*m*/*z* 423.2062, $[M]^+$, Δ +1.6 mmu) by HREIMS and NMR data. The EIMS showed an ion peak at *m*/*z* 137 corresponding to a quinolizidine ring in the structure, and fragment peaks were similar to those of **1**. Comparison of the ^{1}H and ^{13}C NMR data of **2** with those of **1** inferred that **2** was an analogue of **1** with a variation in the quinolizidine ring (Table 1). Nine resonances [δ_C 24.8 (CH_2, C-7), 26.2 (CH_2, C-8), 33.5 (CH_2, C-6), 37.4 (CH_2, C-4), 39.1 (CH_2, C-2), 52.9 (CH_2, C-9), 59.6 (CH, C-1), 59.7 (CH, C-5), 70.7 (CH, C-3)] in the ^{13}C NMR seemed to be due to a *trans*-fused quinolizidine ring, as observed in decinine.[13] The downfield proton H-1 at δ_H 2.99 showed the same HMBC correlations with C-9 (δ_C 52.9), C-5 (δ_C 59.7), C-3 (δ_C 70.7), C-24 (δ_C 114.4), C-20 (δ_C 126.0), C-25 (δ_C 136.6), and C-23 (δ_C 146.6) as those of **1**, indicating the attachment of an aromatic ring at C-1. Thus, the downfield aliphatic carbons at δ_C 52.9, 59.6, 59.7, and 70.7 were assigned to C-9, C-1, C-5, and C-3, respectively. H-1 (δ_H 2.99) was more upfield compared to **1**, with its *cis*-fused quinolizidine ring. A *trans*-fused quinolizidine ring can be readily distinguished by downfield shifts in ^{13}C NMR values of C-1 and C-5 in comparison with their counterpart. Hughes et al. reported that when H-5 is in a *cis*-fused quinolizidine system, C-1 and C-5 appear upfield at δ_C 47.2−47.5 and 56.6−57.0, respectively, whereas when H-5 is in a *trans*-fused quinolizidine system, C-1 and C-5 appear downfield at δ_C 59.5−59.6 and 59.7−59.9, respectively.[13] Moreover, H-5 (δ_H 1.97) also appeared significantly upfield compared to **1**. This proton showed a ROESY correlation with H-1 (δ_H 2.99) along with H-7ax (δ_H 1.14) and H-9ax (δ_H 1.21) (Figure 2), indicating a *trans*-fused quinolizidine ring system. Considering the configuration of C-1 and C-3,[14,15] the structure of compound **2** can be elucidated as dihydrolyfoline (**2**). The minimum energy conformation of **2** and key ROESY correlations are shown in Figure 2.

Although lagerine (**3**) had been isolated from this plant,[6] the structure suggested by Ferris et al. was shown to be in error by demonstrating that a substance synthesized unambiguously by Hanaoka et al. to reproduce the Ferris structure differed from natural lagerine in its chromatographic and spectroscopic properties.[11,12] Hence, here we report the reisolation of natural lagerine along with complete ^{1}H and ^{13}C assignments of **3** by using extensive 1D and 2D NMR spectroscopy (Table 2).



1 2

Figure 2. Minimum energy conformations and key ROESY (↔) correlations of **1** and **2**.

Table 2. NMR Data[a] of Compound **3**

no.	^{1}H (δ)	m (J)	^{13}C (δ)	HMBC (H→C)
1	3.37	d (12.5)	45.1, CH	2, 3, 5, 20, 24, 25
2	e 1.37	br d (13.0)	36.8, CH_2	
	a 1.57			
3	4.88	br s	68.2, CH	1, 5, 11
4	e 1.45	br d (15.5)	34.8, CH_2	
	a 1.99			
5	3.04		57.1, CH	1, 3
6	e 1.08	br t (15.0)	25.4, CH_2	
	a 1.75	dd (13.5, 4.0)		
7	e 1.86	br d (13.0)	26.1, CH_2	
	a 1.37	br d (13.0)		
8	e 1.08	br t (15.0)	19.6, CH_2	
	a 1.64	dt (13.5, 4.0)		
9	e 2.56		50.6, CH_2	
	a 2.42	br d (13.5)		
11			172.4, qC	
12	2.58	ddd (11.5, 4.5, 2.5)	39.7, CH_2	11, 14
12′	2.28	td (13.0, 5.5)		11, 14
13	3.04	ddd (13.0, 5.5, 3.0)	32.5, CH_2	11, 15, 19
13′	2.85	td (13.5, 5.0)		11, 15, 19
14			134.6, qC	
15	6.95	dd (8.0, 2.0)	130.9, CH	13, 17, 19
16	6.51	dd (8.0, 2.0)	116.2, CH	14, 17, 18
17			159.6, qC	
18	7.34	dd (8.0, 2.0)	118.5, CH	14, 16, 17
19	7.26	dd (8.0, 2.0)	129.9, CH	13, 15, 17, 18
20			143.6, qC	
21			148.0, qC	
22			146.8, qC	
23	6.74	d (8.0)	108.3, CH	22, 24, 25,
24	6.91	d (8.0)	119.9, CH	1, 20, 22, 23
25			138.4, qC	
-OCH_3	3.93		56.4	22, 23

[a] ^{1}H (500 MHz) and ^{13}C NMR (75 MHz) recorded in $CDCl_3$ (δ_H 7.24, δ_C 77.0). Carbon multiplicities were determined by DEPT 135° experiment. qC = quaternary, CH = methine, CH_2 = methylene, CH_3 = methyl carbons. Chemical shifts are expressed in ppm and J values in Hz.

Experimental Section

General Experimental Procedures. FT-NMR spectra were recorded on a Bruker Avance 500 spectrometer (^{1}H NMR, 500 MHz) and a Bruker DRX-300 spectrometer (^{13}C NMR, 75 MHz) using $CDCl_3$ and CD_3OD as the solvent and TMS as an internal standard. Chemical shifts (δ) were expressed in ppm with reference to the TMS signals. 2D NMR (HMQC, HMBC, COSY, and ROESY) experiments were performed on a Bruker Avance 500 spectrometer. Mass spectra were obtained with a JEOL JMS-700 Mstation mass spectrometer. Semipreparative HPLC was conducted on TRILUTION LC with a UV/vis-151 detector, a 321 pump, a 402 syringe pump, and a GX-271 liquid handler (Gilson, Inc.), using a YMC-pack Pro C_{18} (250 × 20 mm, i.d.) column. Column chromatography was performed using Si gel (Kieselgel 60, 70−230 mesh and 230−400 mesh, Merck). TLC was performed on Merck precoated silica gel 60 F_{254}, and compounds were observed under 254 and 365 nm UV or visualized by spraying the dried plates with Dragendorff's reagent.

Plant Material. Aerial parts of *L. indica* were collected from Daejeon, Korea, in September 2006 and identified by one of the authors (K.H.B.). A voucher specimen (CNU 1517-1) was deposited at the herbarium in the College of Pharmacy, Chungnam National University.

Extraction and Isolation. The air-dried aerial parts of the plant (6.0 kg) were crushed and extracted three times with hot 70% EtOH. After evaporation of solvent under vacuum, the concentrate was suspended in H_2O, acidified to pH 3 with HCl, and extracted with EtOAc. The aqueous layer was then basified with NH_4OH to pH 9 and extracted with EtOAc. The EtOAc extract was dried with Na_2SO_4 and concentrated in vacuo to obtain the crude base (5 g). This was chromatographed on a Si gel column eluted with MeOH/$CHCl_3$/NH_4OH mixtures of increasing polarites, which afforded lagerine (**3**) (2.5 mg), dihydrolyfoline (**2**) (5.5 mg), and 5-*epi*-dihydrolyfoline (**1**) (7.0 mg). These compounds were purified by HPLC over a YMC-pack Pro C_{18} (250 × 20 mm, i.d.) column using MeCN/H_2O/NH_4OH (45:55:0.1) as eluent.

Computer Molecular Modeling. Molecular modeling and graphic display were performed on GaussView 3.09, and the resulting minimum energy conformations were gained by Gaussian 03 using the optimization method B3lYP/6-31G.

5-*epi*-Dihydrolyfoline (1): dark yellow powder; $[\alpha]^{20}_D$ −11.1 (*c* 0.70, $CHCl_3$); UV (MeOH) λ_{max} 298 nm; ^{1}H NMR (500 MHz, $CDCl_3$ + CD_3OD) and ^{13}C NMR (75 MHz, $CDCl_3$ + CD_3OD) see Table 1; EIMS *m/z* 423 $[M]^+$ (100), 407 $[M - O]^+$ (37), 379 $[M - CO_2]^+$ (19), 363 (40), 339 (100), 299 (23), 255 (77), 253 (34), 227 (69), 197 (12), 182 (17), 165 (15), 137 $[C_9H_{15}N]^+$ (27), 97 (12), 84 (60), 55 (29); HREIMS *m/z* 423.2028 $[M]^+$ (calcd for $C_{25}H_{29}NO_5$, 423.2046).

Dihydrolyfoline (2): yellow powder; $[\alpha]^{20}_D$ −8.0 (*c* 0.55, $CHCl_3$); UV (MeOH) λ_{max} 298 nm; ^{1}H NMR (500 MHz, $CDCl_3$) and ^{13}C NMR (75 MHz, $CDCl_3$) see Table 1; EIMS *m/z* 423 $[M]^+$ (100), 407 $[M - O]^+$ (41), 379 $[M - CO_2]^+$ (28), 363 (56), 339 (100), 299 (22), 255 (95), 253 (48), 227 (69), 197 (14), 182 (22), 165 (15), 137 $[C_9H_{15}N]^+$ (26), 97 (15), 84 (66), 55 (32); HREIMS *m/z* 423.2062 $[M]^+$ (calcd for $C_{25}H_{29}NO_5$, 423.2046).

Lagerine (3): yellow powder; $[\alpha]^{20}_D$ −13.6 (*c* 0.25, $CHCl_3$); UV (MeOH) λ_{max} 298 nm; ^{1}H NMR (500 MHz, $CDCl_3$) and ^{13}C NMR (75 MHz, $CDCl_3$) see Table 2; EIMS *m/z* 423 $[M]^+$ (100), 407 $[M - O]^+$ (7), 379 $[M - CO_2]^+$ (48), 365 (10), 339 (35), 299 (22), 255 (100), 253 (55), 227 (100), 182 (20), 176 (14), 137 $[C_9H_{15}N]^+$ (40), 122 (21), 84 (82), 77(26), 55 (48); HREIMS *m/z* 423.2039 $[M]^+$ (calcd for $C_{25}H_{29}NO_5$, 423.2046).

Acknowledgment. We are grateful to the Korean Basic Science Institute (KBSI) for measuring the NMR spectra. J.Y.L. acknowledges the KOSEF grant by MEST (No. R11-2007-012-03002-0) (2008).

References and Notes

(1) Lee, Y. N. *Flora of Korea*; Kyo-Hak Publishing Co., Ltd., 1996; pp 520−521.
(2) Ferris, J. P. *J. Org. Chem.* **1962**, *27*, 2985–2990.
(3) Ferris, J. P. *J. Org. Chem.* **1963**, *28*, 817–822.
(4) Ferris, J. P.; Boyce, C. B.; Briner, C. B. *J. Am. Chem. Soc.* **1971**, *93*, 2942–2953.
(5) Ferris, J. P.; Boyce, C. B.; Briner, C. B. *J. Am. Chem. Soc.* **1971**, *93*, 2953–2957.
(6) Ferris, J. P.; Boyce, C. B.; Briner, C. B. *J. Am. Chem. Soc.* **1971**, *93*, 2958–2962.
(7) Ferris, J. P.; Boyce, C. B.; Briner, C. B.; Weiss, U.; Qureshi, I. H.; Sharpless, N. E. *J. Am. Chem. Soc.* **1971**, *93*, 2963–2968.
(8) Appel, H.; Rother, A.; Schwarting, A. E. *Lloydia* **1965**, *28*, 84–89.
(9) Rother, A.; Schwarting, A. E. *Lloydia* **1975**, *38*, 477–488.
(10) Dobberstein, R. H.; Edwards, J. M.; Schwarting, A. E. *Phytochemistry* **1975**, *14*, 1769–1775.
(11) Hanaoka, M.; Kamei, M.; Arata, Y. *Chem. Pharm. Bull.* **1975**, *23*, 2191–2192.
(12) Hanaoka, M.; Hori, N.; Ogawa, N.; Arata, Y. *Chem. Pharm. Bull.* **1974**, *22*, 1684–1686.
(13) Hughes, D. W.; Spencer, L. D.; Wrobel, J. T. *Magn. Reson. Chem.* **2000**, *38*, 707–718.
(14) Koo, S. H.; Gupta, R. N.; Spencer, I. D.; Wrobel, J. T. *Chem. Commun.* **1970**, 396–397.
(15) Koo, S. H.; Comer, F.; Spencer, I. D. *Chem. Commun.* **1970**, 897–898.

NP800619G