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A Novel Rearranged Type of Secoeudesmane Sesquiterpenoide from the Root of *Lindera strychnifolia* (SIEB. et ZUCC.) F. VILLARS

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A novel type of sesquiterpene lactone, called strychnilactone, has been isolated from the root of *Lindera strychnifolia*. Its structure was determined by 2D NMR techniques and X-ray analysis.

Key words *Lindera strychnifolia*, Lauraceae, sesquiterpene, strychnilactone, X-ray analysis

We have reported the isolation of a bisesquiterpene, bislindestenolide,[1] from the root of *Lindera strychnifolia* (SIEB. et ZUCC.) F. VILLARS, which has a strong fragrance and is used in Chinese folk medicine as a palliative and an antispasmodic. Takeda and co-workers reported the isolation of about 20 sesquiterpenes from this plant around 1970.[2] Our further investigation of the constituent from the root of *L. strychnifolia* has resulted in the isolation of another novel type of rearranged secoeudesmane sesquiterpene, called strychnilactone (**1**).

The ether-soluble part of the MeOH extract of the root of *L. strychnifolia* (2.9 kg) afforded strychnilactone (**1**) (17.1 mg) after chromatography on silica gel, medium-pressure liquid chromatography (MPLC) column, and purification by recrystallization.

Compound **1**, colorless rods, mp 181—182 °C, $[\alpha]_D^{20}$ $-267.6°$, was crystallized from $CHCl_3$–MeOH (98 : 2). Electron-impact mass spectrometry (EIMS) showed a molecular ion peak at *m/z* 262 [M^+] and the ^{13}C-NMR spectrum showed 17 carbon signals, confirming its molecular formula to be $C_{17}H_{24}O_6$. The ^{1}H-NMR spectrum of **1** showed the signals of CH_3-groups at δ_H ($CDCl_3$) 1.31, 1.66 (with a neighboring oxygen group), and 2.02 (vinyl methyl group), and an ethoxyl group at δ_H 1.26 (3H) and 4.22, 4.17 (each 1H). ^{1}H–^{1}H COSY confirmed the presence of oxygenated methylene group (δ_H 3.51, 3.48 [each 1H]) and cyclopropane ring (δ_H 1.25 [H-1], 1.58, 0.51 [H_2-2], 1.49 [H-3]). The ^{13}C-NMR spectrum of **1** indicated two signals of ester and/or lactone carbonyl carbons at δ_C 167.1 and 169.5, which was in good agreement with the IR spectrum (λ_{max} 1751 and 1641 cm^{-1}), indicating that one of the esters is conjugated.

Intensive analysis of the HSQC and HMBC spectra of **1** indicated the partial structure shown in Fig. 1. In the HMBC spectrum, no connectivity was shown between the H-9 methylene group and C-8 carbonyl carbon, indicating that a lactone ring was formed between C-4 and C-8 (δ-lactone). This was confirmed by the proton chemical shifts of the H-9 methylene group at δ_H 3.51 and 3.48. The NOE correlation analyzed by NOESY spectrum revealed the stereostructure shown in Fig. 2, except for the configuration of 6-OH. To elucidate the configuration at C-6 and confirm the entire sug-

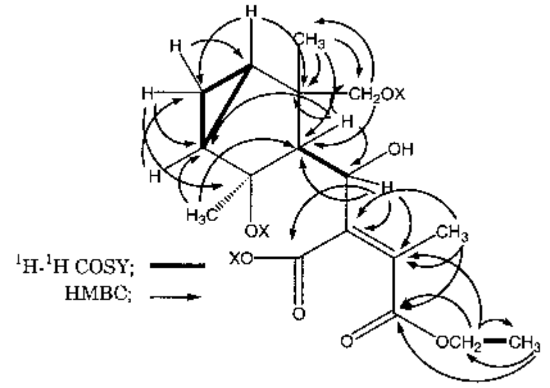

Fig. 1. Correlations of Compound **1**

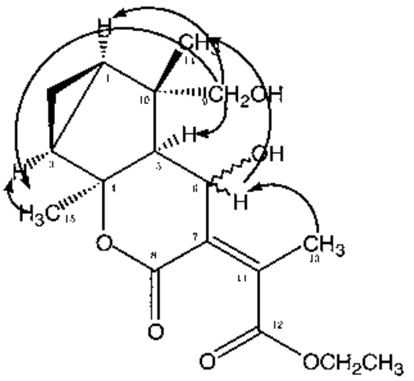

Fig. 2. NOE Correlations of Compound **1**

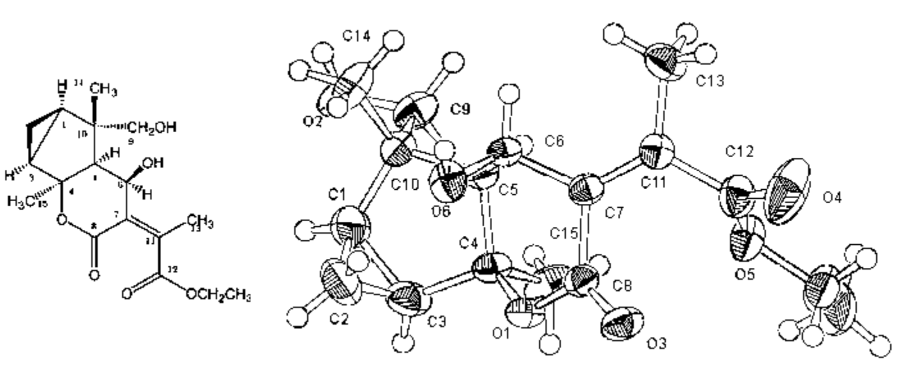

Fig. 3. Structure and ORTEP Drawing of Compound **1**

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gested structure, a single crystal X-ray analysis was performed. A single crystal belonging to a space group $P2_12_12_1$ ($Z=4$) was obtained by recrystallization from $CHCl_3$. Data collection was performed on a RIGAKU AFC 7S diffractometer, and the structure was solved by direct method (SIR92),[3] then refined by full-matrix least-squares based on 1925 observed reflections ($I>3.00\sigma[I]$). Hydrogen atoms were refined isotropically, and nonhydrogen atoms were treated anisotropically. The final refinement converged at $R=0.032$.[4] Thus the suggested whole structure (Fig. 3) including the configuration of 6β-OH was established, although the absolute structure remains to be clarified.

More than 10 secoeudesmane-type sesquiterpenes have been identified so far, but compound **1** is a unique and the first example of a secoeudesmanolide, cleaving between the C-8 and C-9 bond of the eudesmane skeleton.

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References and Notes

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4) The weighted agreement factor is $Rw=0.035$.