SUPPLEMENTARY MATERIAL

Pleosporalone A, the first azaphilone characterized with aromatic A-ring from a marine-derived Pleosporales sp. fungus

Fei Cao, Jian-Kun Yang, Yun-Feng Liu, Hua-Jie Zhu, and Chang-Yun Wang

Key Laboratory of Pharmaceutical Quality Control of Hebei Province, College of Pharmaceutical Sciences, Hebei University, Baoding 071002, People’s Republic of China; Key Laboratory of Marine Drugs, the Ministry of Education of China, School of Medicine and Pharmacy, Ocean University of China, Qingdao 266003, People’s Republic of China

Abstract
A fungal strain, Pleosporales sp. CF09-1, was isolated from marine sediment collected from the Bohai Sea. A novel azaphilone derivative, named pleosporalone A (1), along with two known analogues, cohaerins A and B (2 and 3) were obtained and identified from the culture extract of Pleosporales sp. CF09-1. Their planar structures were elucidated by detailed analysis of spectroscopic data and by comparison with related known compounds. Pleosporalone A (1) represent the first azaphilone derivative characterized with A-ring aromatization. Compound 1 showed strong antifungal activity against three plant pathogenic fungi Botrytis cinerea, Rhizopus oryzae and Phytophthora capsici with the MIC values of 0.39, 0.78 and 0.78 μM, respectively.

Keywords: marine sediment; Pleosporales sp.; azaphilone derivative; antifungal activity

Correspondence
Prof. Dr. Hua-Jie Zhu
College of Pharmaceutical Sciences, Hebei University 180 Wusidong Road Baoding 071002 Hebei P. R. China
jackzhu2002@sia.com
*Corresponding author. Email: jackzhu2002@sia.com (H. J. Zhu)
List of Supplementary Material

Table S1. $^1$H NMR and $^{13}$C NMR data of 1 and Thunberginol B

Figure S1. $^1$H NMR (500 MHz, CDCl$_3$) spectrum of 1

Figure S2. $^{13}$C NMR (125 MHz, CDCl$_3$) spectrum of 1

Figure S3. HMQC (CDCl$_3$) spectrum of 1

Figure S4. COSY (CDCl$_3$) spectrum of 1

Figure S5. HMBC (CDCl$_3$) spectrum of 1

Figure S6. COSY and key HMBC correlations of 1

Figure S7. ESIMS spectrum of 1

Figure S8. HRESIMS spectrum of 1
Table S1. $^1$H NMR and $^{13}$C NMR data of I and Thunberginol B

| No. | I (CD$_3$OD, 500 MHz) | Thunberginol B (DMSO-$d_6$, 270 MHz)* |
|-----|----------------------|--------------------------------------|
| 1   | -                    | 168.8                                |
| 3   | -                    | 151.5                                |
| 4   | 6.50 (1H, s)         | 110.3                                |
| 4a  | -                    | 138.1                                |
| 5   | 6.43 (1H, s)         | 103.1                                |
| 6   | -                    | 165.2                                |
| 7   | -                    | 112.3                                |
| 8   | -                    | 162.0                                |
| 8a  | -                    | 99.7                                 |
| 9   | 2.12 (3H, s)         | 8.1                                  |
| 10  | -                    | 121.7                                |
| 11  | -                    | 140.1                                |
| 12  | 6.77 (1H, d, $J = 7.5$ Hz) | 122.3                                |
| 13  | 7.14 (1H, dd, $J = 8.0, 7.5$ Hz) | 131.6                                |
| 14  | 6.73 (1H, d, $J = 8.0$ Hz) | 114.3                                |
| 15  | -                    | 157.1                                |
| 16  | 2.26 (3H, s)         | 20.1                                 |

* Yoshikawa M, Harada E, Naitoh Y, Inoue K, Matsuda H, Shimoda H, Yamahara J, Murakami N. 1994. Development of biactive functions in hydrangea dulcis folium. III. On the antiallergic and antimicrobial principles of hydrangea dulcis folium. (1). Thunberginols A, B, and F. Chem Pharm Bull. 42:2225-2230.
Figure S1. $^1$H NMR (500 MHz, CDCl$_3$) spectrum of 1

Figure S2. $^{13}$C NMR (125 MHz, CDCl$_3$) spectrum of 1
Figure S3. HMQC (CDCl₃) spectrum of 1

Figure S4. COSY (CDCl₃) spectrum of 1
Figure S5. HMBC (CDCl₃) spectrum of 1

Figure S6. COSY (---) and key HMBC (→) correlations of 1.
Figure S7. ESIMS spectrum of 1

Figure S8. HRESIMS spectrum of 1