A Benzofuranoid and Two Clerodane Diterpenoids from *Pulicaria wightiana*[1])

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Methyl 5-formyl-1-benzofuran-6-carboxylate (**1**) and the two clerodane diterpenes, methyl 6-oxocleroda-3,13-dien-15,16-olid-18-oate (**2**) and 2β-(2-methylbutanoyl)cleroda-3,13-dien-15,16-olid-18-oic acid (**3**), together with 15 known compounds, were isolated from the aerial parts of *Pulicaria wightiana*. The structures of the new compounds **1**, **2**, and **3** were established by spectroscopic (mainly 1D- and 2D-NMR) methods.

Introduction. – The genus *Pulicaria* (Compositae) comprises around 100 species [1]. Many of these species and their constituents are known for their medicinal properties. The root of *P. dysenteria* is used in the treatment of dysentery [2], and the paste of the plant is applied externally to wounds [3]. Sesquiterpenoids from *P. crispa* [4] and from the ethanolic extract of *P. orientalis* [5] were found to be cytotoxic. *P. salviifolia* exhibits hypolipidemic effect [6]. Antispasmodic activity of *P. glutinosa* [7] and antihistaminic effect of *P. dysenterica* [8] have also been reported. *P. wightiana* C. B. Clarke is an annual or perennial herb distributed in different parts of India. Chemical studies on this species are limited [9][10], and so a detailed investigation on a recent fresh collection of the plant was undertaken. In earlier studies on aerial parts of this plant from Tirumala Hill (India) [10], we reported clerodane diterpenes with antibacterial activity. The results encouraged us to investigate the same species from different places. In this recent study we have investigated the aerial parts of *P. wightiana* from Thalakona forest (India), and we report herein the isolation, and structure elucidation of one new benzofuranoid, **1**, and two new clerodane diterpenes, **2** and **3**. Fifteen known compounds (*Fig. 1*) *viz.*, stigmasterol (**4**) [11], β-sitosterol (**5**) [11], methyl 15,16-epoxy-6α-hydroxycleroda-3,13(16),14-trien-18-oate (**6**) [10a], phyllemblin (**7**) [12], isopimara-8(14),15-diene-7-keto-2α-ol (**8**) [9], methyl 6α-hydroxycleroda-3,13-dien-15,16-olid-18-oate (**9**) [10a], methyl 6,15-dihydroxycleroda-3,13-dien-15,16-olid-18-oate (**10**) [10a], methyl 6α,7α-dihydroxycleroda-3,13-dien-15,16-olid-18-oate (**11**) [10a], 6α-hydroxycleroda-3,13-dien-15(16),4(7)-diolide (**12**) [10a], oxyanin B (**13**) [13], methyl 6α-hydroxycleroda-3,13-dien-18-oate (**14**) [11b], fraxetin (**15**) [14], gallic acid (**16**) [12], daucosterol (**17**) [15], and bergenin (**18**) [16] were also isolated.

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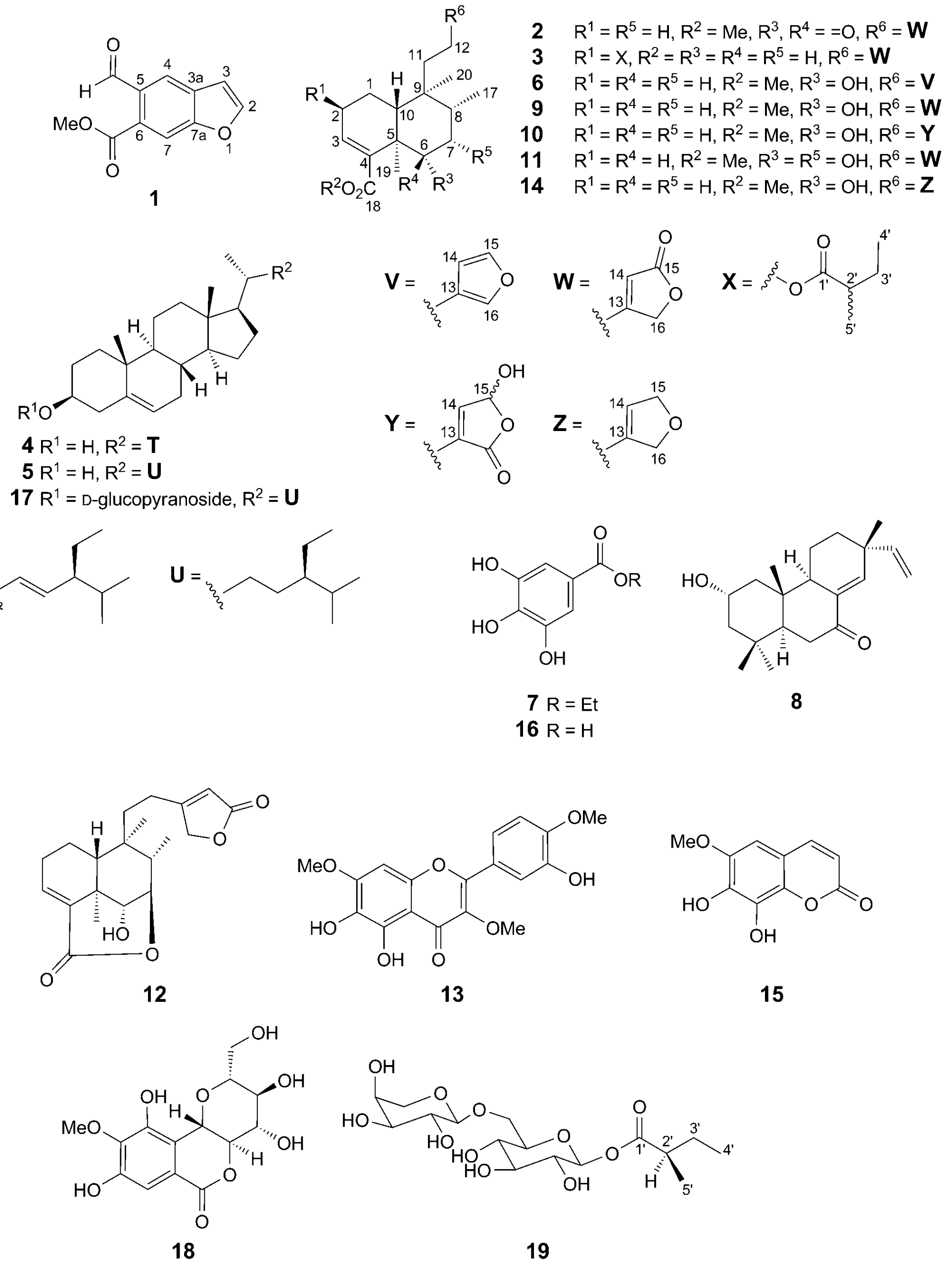
[1]) Part 65 in the series 'Studies on phytochemicals'.



2 $R^1 = R^5 = H$, $R^2 = Me$, $R^3, R^4 = {=}O$, $R^6 = $ **W**
3 $R^1 = X$, $R^2 = R^3 = R^4 = R^5 = H$, $R^6 = $ **W**
6 $R^1 = R^4 = R^5 = H$, $R^2 = Me$, $R^3 = OH$, $R^6 = $ **V**
9 $R^1 = R^4 = R^5 = H$, $R^2 = Me$, $R^3 = OH$, $R^6 = $ **W**
10 $R^1 = R^4 = R^5 = H$, $R^2 = Me$, $R^3 = OH$, $R^6 = $ **Y**
11 $R^1 = R^4 = H$, $R^2 = Me$, $R^3 = R^5 = OH$, $R^6 = $ **W**
14 $R^1 = R^4 = R^5 = H$, $R^2 = Me$, $R^3 = OH$, $R^6 = $ **Z**

4 $R^1 = H$, $R^2 = $ **T**
5 $R^1 = H$, $R^2 = $ **U**
17 $R^1 = $ D-glucopyranoside, $R^2 = $ **U**

7 R = Et
16 R = H

Fig. 1. *Structures of compounds* **1**–**19**

Results and Discussion. – Column chromatography (CC) of the EtOH and $CHCl_3$ (1:1) extract of the aerial parts of *P. wightiana* on silica gel afforded crude samples containing compounds **1** – **18**. Repeated CC of these mixtures on silica gel with AcOEt and hexane provided the pure compounds.

Compound **1** was isolated as dark yellow needles (AcOEt), m.p. 182 – 183°. Its molecular formula was assigned as $C_{11}H_8O_4$ from elemental analysis, the *pseudo*-molecular ion peak at *m/z* 205 ($[M+H]^+$) in the ESI-MS spectrum and from the ^{1}H- and ^{13}C-NMR spectra (indicating the presence of 8 H-atoms and 11 C-atoms, resp.). The IR spectrum showed absorptions at ν_{max} 1723 and 1708 – 1530 (br.) cm^{-1}, indicating the presence of aldehyde and ester functions in the molecule. This information, together with the ^{1}H- and ^{13}C-NMR spectroscopic data (*Table 1*), suggested that the compound possesses a benzofuran type skeleton. The ^{1}H- and ^{13}C-NMR assignments were made with the aid of 2D-NMR (DQF-COSY, HSQC, HMBC, and NOESY) and APT experiments. The ^{1}H-NMR spectrum displayed a *doublet* at δ(H) 7.96 (H–C(2), $J = 3.1$) and a *doublet* of *doublet* at δ(H) 7.14 (H–C(3), $J = 3.1, 0.8$). The two H-atoms were strongly correlated with each other in DQF-COSY and NOESY. Furthermore, a *doublet* at δ(H) 9.18 (H–C(4), $J = 0.8$) and three *singlet*s at δ(H) 9.97 (CHO), 8.53 (H–C(7)), and 4.03 (COO*Me*) indicated that the compound was similar to methyl 5-formyl-1-benzofuran-6-carboxylate, recently isolated from *Aspergillus niger* [17]. The benzofuran skeleton and functionalities of both the molecules are similar. The ^{13}C-NMR spectrum of **1** presented signals for two CO groups (δ(C) 181.1 and 164.7), eight aromatic C-atoms including four quaternary (δ(C) 130.3, 125.1, 124.4, and 115.0) and four tertiary C-atoms (δ(C) 149.7, 148.2, 148.0, and 113.5) and of a Me ester (δ(C) 52.4, *q*). The HMBC experiment (*Fig. 2*) showed correlations between H–C(4) and C(6) (δ(C) 124.4), COO*Me* and *C*OOMe (δ(C) 164.7), and between H–C(7) and *C*OOMe, confirming the placement of the COOMe substituent at the 6-position. Correlations of CHO with C(6) and C(5) (δ(C) 125.1) and of H–C(4) with C(6) and C(5) were considered as strong evidence for the position of the formyl group at C(5). Thus, the structure of compound **1** was established as methyl 5-formyl-1-benzofuran-6-

Table 1. *^{1}H- and ^{13}C-NMR Spectral Data* (δ in ppm, *J* in Hz) *of* **1**[a])

	δ(H)	δ(C)
H–C(2)	7.96 (*d*, $J = 3.1$)	148.0
H–C(3)	7.14 (*dd*, $J = 3.1, 0.8$)	113.5
C(3a)	–	115.0
H–C(4)	9.18 (*d*, $J = 0.8$)	149.7
C(5)	–	125.1
C(6)	–	124.4
H–C(7)	8.53 (*s*)	148.2
C(7a)	–	130.3
CHO	9.97 (*s*)	181.1
*C*OOMe	–	164.7
COO*Me*	4.02 (*s*)	52.4

[a]) The values were assigned on the basis of 2D-NMR (DQF-COSY, NOESY, HSQC and HMBC) spectroscopy and DEPT experiments.

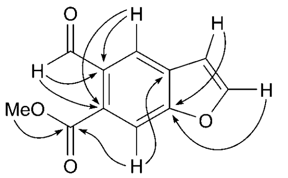


Fig. 2. *Selected HMBC* (H → C) *correlations of* **1**

carboxylate, which is a positional isomer of methyl 6-formyl-1-benzofuran-5-carboxylate [17] with only the exchange of the positions of the CHO and the COOMe groups.

Compound **2** was isolated as colorless crystals (AcOEt), with a m.p. of 175 – 176°, and $[\alpha]_D^{25} = -23.4$ ($c = 0.8$, $CHCl_3$). Its formula was deduced as $C_{21}H_{28}O_5$ from its elemental analysis, a *pseudo*-molecular ion peak at *m/z* 383 ($[M + Na]^+$) in the ESI-MS spectrum, and the ^1H- and ^{13}C-NMR spectra (*Table 2*; indicating 28 H-atoms and 21 C-atoms, resp.). The IR spectrum displayed absorption bands at ν_{max} 1778, 1746, and 1711 cm^{-1}, indicating the presence of keto, α,β-unsaturated ester, and lactone functions in the molecule. The ^1H- and ^{13}C-NMR spectral data (*Table 2*) suggested the compound to be a clerodane diterpenoid related to **9**, which was previously reported from the same species [10a]. The ^1H-NMR spectrum displayed signals for three Me groups (δ(H) 1.71 (*s*, Me(19)); 1.05 (*s*, Me(20)); 0.96 (*d*, $J = 6.7$, Me(17)), two olefinic H-atoms (δ(H) 6.63 (*dd*, $J = 4.0, 2.6$, H–C(3)); 5.84 (br. *t*, $J = 1.3$, H–C(14)), an oxygen-bearing CH_2 group (δ(H) 4.70 (br. *d*, $J = 1.3$, CH_2(16)) and a Me ester group (δ(H) 3.72 (*s*)) similar to that of **9**. The absence of H–C(6) in compound **2** and the presence of a signal at δ(C) 211.7 in the ^{13}C-NMR spectrum suggested that the compound contains a keto function at C(6) instead of a OH group present at this position in **9**. This was confirmed by correlations of Me(19), H–C(10) (δ(H) 1.69 – 1.53 (*m*)), and H–C(8) (δ(H) 1.97 (*m*)) with C(6) (δ(C) 211.7) in the HMBC experiment (*Fig. 3*). Furthermore, the ^{13}C-NMR spectrum displayed signals for 21 C-atoms (similar to **9**) including three CO groups (δ(C) 211.7, 169.5, and 168.5), four olefinic C-atoms (δ(C) 169.5, 137.3, 136.2, and 115.4), one oxygen-bearing CH_2 group (δ(C) 72.9), an ester Me group (δ(C) 51.4), three Me groups (δ(C) 20.8, 18.3, and 16.1), five secondary (δ(C) 42.4, 35.4, 26.3, 22.3, and 17.8), two tertiary (δ(C) 49.8 and 41.4), and two quaternary C-atoms (δ(C) 53.6 and 39.1). Thus it is clear that the structures of both the compounds **2** and **9** are similar, having only the difference that the former contains a keto group at C(6), while **9** contains a OH group. The HMBC experiment also showed correlations of Me(20), H–C(11) (δ(H) 1.85 – 1.73 (*m*)), and H–C(2) (δ(H) 2.31 (*m*)) with C(10) (δ(C) 49.8), CH_2(12) (δ(H) 2.23 – 2.01, *m*) with C(13) (δ(C) 169.5) and C(14) (δ(C) 115.5) and CH_2(16) with C(15) (δ(C) 169.5). In the NOESY experiment (*Fig. 3*), H–C(10) showed a correlation with H–C(8) but not with Me(20) and Me(19), indicating that H–C(10) and H–C(8) are β-configured. Me(17) showed correlations with Me(20) and Me(19), suggesting that these three Me groups are in α-configuration. From the above data and the co-occurrence of several related compounds with known configuration from the same plant, the structure of the new compound was established as methyl 6-oxocleroda-3,13-dien-15,16-olid-18-oate (**2**).

Table 2. *^{1}H-NMR and ^{13}C-NMR Spectral Data* (δ in ppm, J in Hz) *of* **2** *and* **3**[a])

	2		**3**	
	δ(H)	δ(C)	δ(H)	δ(C)
CH_2(1)	1.85 – 1.73 (*m*), 1.69 – 1.53 (*m*)	17.8	1.88 (*m*), 1.72 (br. *d*, $J = 14.1$)	24.7
CH_2(2) or H–C(2)	2.31 (*m*), 2.23 – 2.01 (*m*)	26.3	5.40 (*m*)	66.6
H–C(3)	6.63 (*dd*, $J = 4.0$, 2.6)	136.2	6.64 (*d*, $J = 3.9$)	132.8
C(4)	–	137.3	–	145.8
C(5)	–	53.6	–	38.4
C(6) or CH_2(6)	–	211.7	2.47 – 2.22 (*m*), 1.69 – 1.59 (*m*)	35.1
CH_2(7)	2.89 (*dd*, $J = 12.7$, 9.6), 2.23 – 2.01 (*m*)	42.4	1.58 – 1.42 (*m*)	26.9
H–C(8)	1.97 (*m*)	41.4	1.58 – 1.42 (*m*)	36.2
C(9)	–	39.1	–	37.8
H–C(10)	1.69 – 1.53 (*m*)	49.8	1.58 – 1.42 (*m*)	42.2
CH_2(11)	1.85 – 1.73 (*m*), 1.69 – 1.53 (*m*)	35.4	1.58 – 1.42 (*m*), 1.21 (*m*)	35.0
CH_2(12)	2.23 – 2.01 (*m*)	22.3	2.47 – 2.22 (*m*)	21.8
C(13)	–	169.5	–	170.6
H–C(14)	5.84 (br. *t*, $J = 1.3$)	115.4	5.83 (br. *t*, $J = 1.4$)	115.1
C(15)	–	169.5	–	171.5
CH_2(16)	4.70 (br. *d*, $J = 1.3$)	72.9	4.74 (*dd*, $J = 17.2$, 1.4), 4.68 (*dd*, $J = 17.2$, 1.4)	72.9
Me(17)	0.96 (*d*, $J = 6.7$)	16.1	0.84 (*d*, $J = 6.7$)	15.7
C(18)	–	168.5	–	171.5
Me(19)	1.71 (*s*)	20.8	1.24 (*s*)	18.8
Me(20)	1.05 (*s*)	18.3	0.81 (*s*)	17.9
COO*Me*	3.72 (*s*)	51.4	–	–
C(1′)	–	–	–	175.6
H–C(2′)	–	–	2.47 – 2.22 (*m*)	41.0
CH_2(3′)	–	–	1.69 – 1.59 (*m*), 1.58 – 1.42 (*m*)	26.8
Me(4′)	–	–	0.89 (*t*, $J = 4.7$)	11.5
Me(5′)	–	–	1.10 (*d*, $J = 7.0$)	16.4

[a]) The values were assigned on the basis of 2D-NMR (DQF-COSY, NOESY, HSQC, and HMBC) spectroscopy and DEPT experiments.

Compound **3** was obtained as a white solid with a m.p. of 123 – 124°, and $[\alpha]_D^{25} = -42.8$ ($c = 1.0$, $CHCl_3$). Its molecular formula was deduced as $C_{25}H_{36}O_6$ from elemental analysis, the $[M + Na]^+$ peak at *m/z* 455 in the ESI-MS spectrum, and ^{1}H- and ^{13}C-NMR spectral data (indicating 36 H-atoms, and 25 C-atoms, resp.). The IR spectrum showed absorptions at ν_{max} 2986, 1780, 1735 – 1715 (br.), and 1636 cm^{-1}, indicating the presence of carboxylic acid, ester, lactone, and olefinic functions in the molecule. The ^{1}H- and ^{13}C-NMR spectral data (*Table 2*) suggested that the molecule is related to 2β-hydroxyhardwickiic acid [18]. The ^{1}H-NMR spectrum showed signals for

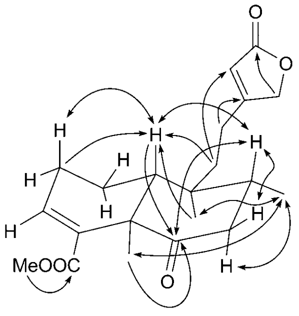


Fig. 3. *Selected HMBC* (→) *and NOESY* (↔) *correlations of* **2**

two olefinic H-atoms (δ(H) 6.64 (*d*, *J* = 3.9, H−C(3)); 5.83 (br. *t*, *J* = 1.4, H−C(14)), three Me groups (δ(H) 1.24 (*s*, Me(19)); 0.84 (*d*, *J* = 6.7, Me(17)); 0.81 (*s*, Me(20)), an oxygen-bearing CH_2 group (δ(H) 4.74 and 4.68 (*dd*, *J* = 17.2, 1.4, 1 H each, CH_2(16)) and an oxygen-bearing CH group (δ(H) 5.40 (*m*, H−C(2)). Thus, compound **3** is structurally related to 2*β*-hydroxyhardwickiic acid, but the former contains an *α*,*β*-unsaturated *γ*-lactone ring at C(12) (like compound **2**) instead of a furan ring present at this position in 2*β*-hydroxyhardwickiic acid. The ^{1}H-NMR spectrum of **8** also showed the signals of two additional Me groups (δ(H) 1.10 (*d*, *J* = 7.0), 0.89 (*t*, *J* = 4.7)). The correlations of H−C(2) (δ(H) 5.40 (*m*)) with H−C(3) (δ(H) 6.64 (*d*, *J* = 3.9)) and CH_2(1) (δ(H) 1.88 (*m*, 1 H), 1.72 (br. *d*, *J* = 14.1, 1 H)) in the DQF-COSY spectrum indicated the oxygen-attachment at C(2). The ^{13}C-NMR spectrum indicated the presence of 25 C-atoms which included three CO groups (δ(C) 175.6, 171.6 (2 C)), four olefinic C-atoms (δ(C) 170.6, 145.8, 132.8, and 115.1), two oxygen-bearing C-atoms (δ(C) 72.9 and 66.6), five Me groups (δ(C) 18.8, 17.9, 16.4, 15.7, and 11.5), together with two quarternary (δ(C) 38.4 and 37.8), three tertiary (δ(C) 42.2, 41.0, and 36.2) and six secondary C-atoms (δ(C) 35.1, 35.0, 26.9, 26.8, 24.7, and 21.8). The comparison of the ^{13}C-NMR values with those of 1*β*-vicianosyl-(2*S*)-methylbutyrate (**19**) [19] (*Table 3*) strongly indicated the presence of a 2-methylbutanoyl group in 2-position, which was confirmed by correlations of H−C(2) with C(1′) (δ(C) 175.6), C(10) (δ(C) 42.2), and C(4) (δ(C) 145.8), of CH_2(3′) (δ(H) 1.69−1.59 (*m*, 1 H), 1.58−1.42 (*m*, 1 H)) and Me(5′) (δ(H) 1.10) with C(1′) (δ(C) 175.6), and Me(4′) (δ(H) 0.89) with C(2′) (δ(C) 41.0) in the HMBC experiment (*Fig. 4*). It also showed correlations of H−C(14) with C(16) (δ(C) 72.9), CH_2(16) with C(14) (δ(C) 115.1) and C(15) (δ(C) 171.5), H−C(3) with C(7) (δ(C) 26.9), Me(19) with C(6) (δ(C) 35.1), and Me(19) and Me(20) with

Table 3. *Comparison of* ^{13}*C-NMR values* (δ in ppm) *of the 2-Methylbutanoyl Group of Compounds* **3** *and* **19**

	3	**19**
C(1′)	175.6	179.0
H−C(2′)	41.0	41.6
CH_2(3′)	26.8	27.0
Me(4′)	11.5	11.5
Me(5′)	16.4	16.4

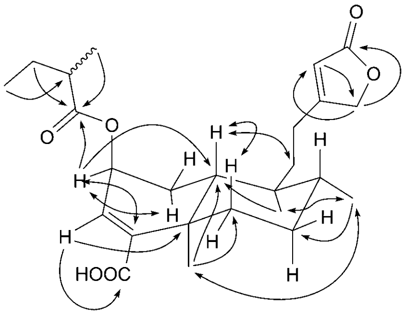


Fig. 4. *Selected HMBC* (H→C) *and NOESY* (↔) *correlations of* **3**

C(10). In the NOESY experiment (*Fig. 4*), H–C(10) was not found in correlation with Me(19) and H–C(2), but it was correlated to H–C(11) (δ(H) 1.21 (*m*)) and H–C(6) (δ(H) 2.47–2.22 (*m*)). Me(19) was correlated to Me(20) and Me(17). H–C(2) showed a correlation with only CH_2(1), but not with H–C(10). From the above observations the structure of the compound was deduced as 2β-(2-methylbutanoyl)cleroda-3,13-dien-15,16-olid-18-oic acid (**3**).

Along with the three new compounds **1**–**3**, the known compounds **4**–**14**, **16**, and **17** were also isolated, and they were identified by comparison of their physical (m.p., $[\alpha]_D$), and ^{1}H-NMR and MS data with those reported earlier. The occurrence of fraxetin (**15**) and bergenin (**18**) has been observed for the first time in the title species.

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Experimental Part

General. Column chromatography (CC) over silica gel (SiO_2; *BDH*, 100–200 mesh). TLC: silica gel *GF254*. The visualization of the spots in TLC plates was carried out either under UV light or by exposing the plates to I_2 vapor, or by spraying with 10% H_2SO_4 in MeOH and subsequently heating on a hot plate. M.p.: *Büchi 510* instrument; uncorrected. $[\alpha]_D$: *Jasco DIP-360* polarimeter. IR Spectra: *Perkin-Elmer RX 1* FT-IR spectrophotometer. ^{1}H- and ^{13}C-NMR Spectra: *Varian Gemini 200* MHz and *Unity 400* MHz spectrometers. ESI-MS: *Micromass Quattro*. LC-MS: *Micromass VG 7070 H* (70 eV).

Plant Material. The aerial parts of *P. wightiana* were collected form Thalakona forest, Andhra Pradesh in August 2006 and identified by Prof. *C. Rajugopal*, Department of Botany, Osmania University, Hyderabad. A voucher specimen (No PW-AP-10) is preserved in our laboratory and another specimen (No. IICP 101008) in IICT herbarium.

Extraction and Isolation. The shade dried plant material (5 kg) was powdered and extracted three times with a 1:1 mixture of EtOH and $CHCl_3$ (4 l) for 72 h at r.t. The combined extracts were collected to afford a brownish mass (102.5 g). The residue (102 g) was subjected to CC over SiO_2 using hexane/AcOEt or AcOEt as eluent and gave eight fractions (*Fr. A–H*). The eluents and the compounds isolated after further chromatography are as follows: *Fr. A*, 5% AcOEt in hexane, compounds **4** (0.13 g), **5** (0.21 g), and **1** (0.012 g); *Fr. B*, 10% AcOEt in hexane, compounds **7** (0.10 g) and **2** (0.02 g); *Fr. C*, 20% AcOEt in hexane, compounds **8** (0.043 g), **3** (0.03 g), **6** (0.018 g), **9** (0.011 g), and **10** (0.009 g); *Fr. D*, 30% AcOEt in hexane, compound **11** (0.022 g); *Fr. E*, 40% AcOEt in hexane, compounds **12** (0.062 g) and **13** (0.091 g); *Fr. F*, 60% AcOEt in hexane, compounds **14** (0.017 g) and **15** (0.032 g); *Fr. G*, 80% AcOEt in hexane, compounds **16** (0.041 g) and **17** (0.17 g); *Fr. H*, AcOEt, compound **18** (0.034 g).

Methyl 5-formyl-1-benzofuran-6-carboxylate (**1**). Dark yellow needles. M.p. 182–183°. IR (KBr): 2922, 2852, 1723, 1708–1503 (br.), 1468, 1388, 1269, 1084, 757. ^{1}H- and ^{13}C-NMR: *Table 1*. ESI-MS: 205 ($[M+H]^+$). Anal. calc. for $C_{11}H_8O_4$: C 64.71, H 3.92; found: C 64.89, H 3.81.

Methyl 6-Oxoclerodа-3,13-dien-15,16-olid-18-oate (= *Methyl* (4*a*R,5S,6R,8*a*R)-*3,4,4a,5,6,7,8,8a-Octahydro-5,6,8a-trimethyl-8-oxo-5-[2-(5-oxo-2,5-dihydrofuran-3-yl)ethyl]naphthalene-1-carboxylate*; **2**). Colorless crystals. M.p. 175–176°. $[\alpha]_D^{25} = -23.4$ ($c = 0.8$, $CHCl_3$). IR (KBr): 2925, 2855, 1778, 1746, 1711, 1639, 1432, 1260, 1024, 770. ^{1}H- and ^{13}C-NMR: *Table 2*. ESI-MS: 383 ($[M + Na]^+$). Anal. calc. for $C_{21}H_{28}O_5$: C 70.00, H 7.78; found: C 69.89, H 7.89.

*2β-[(2*S*)-Methylbutanoyl]clerodа-3,13-dien-15,16-olid-18-oic Acid* (= *(3*S*,4a*R*,5*S*,6*R*,8a*R*)-3,4,4a,5,6,7,8,8a-Octahydro-5,6,8a-trimethyl-3-[(2-methylbutanoyl)oxy]-5-[2-(5-oxo-2,5-dihydrofuran-3-yl)ethyl]naphthalene-1-carboxylic Acid*; **3**). White solid. M.p. 123–124°. $[\alpha]_D^{25} = -42.8$ ($c = 1.0$, $CHCl_3$). IR (KBr): 2986, 2925, 2845, 1780, 1735–1715 (br.), 1636, 1458, 1219, 772. ^{1}H- and ^{13}C-NMR: *Table 2*. ESI-MS: 455 ($[M + Na]^+$). Anal. calc. for $C_{25}H_{36}O_6$: C 69.44, H 8.33; found: C 69.59, H 8.21.

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