New Sesquiterpenes from the Flowers of *Chrysanthemum indicum* L.

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Two new guaianolides (= guaianolactones), chrysanthguaianolactone A and B (**1** and **2**, resp.), and one new eudesmane sesquiterpene, chrysanthemdiol A (**6**), together with seven known sesquiterpenes were isolated from the flowers of *Chrysanthemum indicum* L. Their structures were elucidated on the basis of spectroscopic evidence.

Introduction. – *Chrysanthemum indicum* L. is a kind of traditional Chinese medicine with effects on clearing heat and detoxification and is widespread in China. The flowers of *Chrysanthemum indicum* L. have been used for the treatment of vertigo, hypertension, and virosis [1]. Previous phytochemical investigations led to the isolation of sesquiterpenes and flavonoids from the flowers of *C. indicum* L. [2–6].

Our pharmacological experiments revealed that the EtOH extracts of *C. indicum* indicated promising anti-HBV activities. Phytochemical investigation of the flowers of *C. indicum* led to the isolation of two new guaianolide (= guaianolactone) sesquiterpenes, named chrysanthguaianolactone A[1]) (**1**) and chrysanthguaianolactone B[1]) (**2**), and one new eudesmane sesquiterpene, named chrysanthemdiol A[1]) (**6**), together with seven known sesquiterpenes. By comparison of their spectroscopic data with those reported, the known compounds were elucidated as (3α,6α,8α)-8-tigloyl-3,4-epoxyguai-1(10)-eno-12,6-lactone (**3**) [7], apressin (**4**) [8], athanadregeolid (**5**) [9], cumambrin A [10][11], (3β,6β)-eudesm-4(14)-ene-3,5,6,11-tetrol [12], (+)-eudesm-4(14)-ene-11,13-diol [13][14], and cryptomeridiol [15]. Herein, we report the isolation and structural elucidation of the new compounds.

Results and Discussion. – Compound **1** was obtained as colorless crystals. The HR-ESI-MS showed an accurate $[M+Na]^+$ ion peak at *m*/*z* 385.1627, in accordance with an empirical molecular formula $C_{20}H_{26}O_6$ with seven degrees of unsaturation, which was supported by the ^{1}H- and ^{13}C-NMR, and DEPT data. The IR spectrum of **1** showed the presence of OH groups (3563 cm^{-1}), C=O groups (1752 and 1718 cm^{-1}), and C=C bonds (1646 cm^{-1}). In the ^{1}H-NMR spectrum of **1**, the signals of two olefinic H-atoms (δ(H) 6.16–6.22 (*m*) and 5.28 (br. *s*)), three O-bearing CH groups (δ(H) 5.48 (br. *d*, *J* = 10.4 Hz), 3.95 (*dd*, *J* = 9.2 and 9.2 Hz), and 3.54 (br. *s*)), and five Me groups (δ(H) 2.02 (*d*, *J* = 7.2 Hz), 1.92 (*s*), 1.90 (*s*), 1.69 (*s*), and 1.29 (*d*, *J* = 6.8 Hz)) were observed (*Table 1*). The ^{13}C-NMR spectrum of **1** showed the signals of a total of 20 C-atoms,

[1]) Trivial atom numbering; for systematic names, see *Exper. Part.*

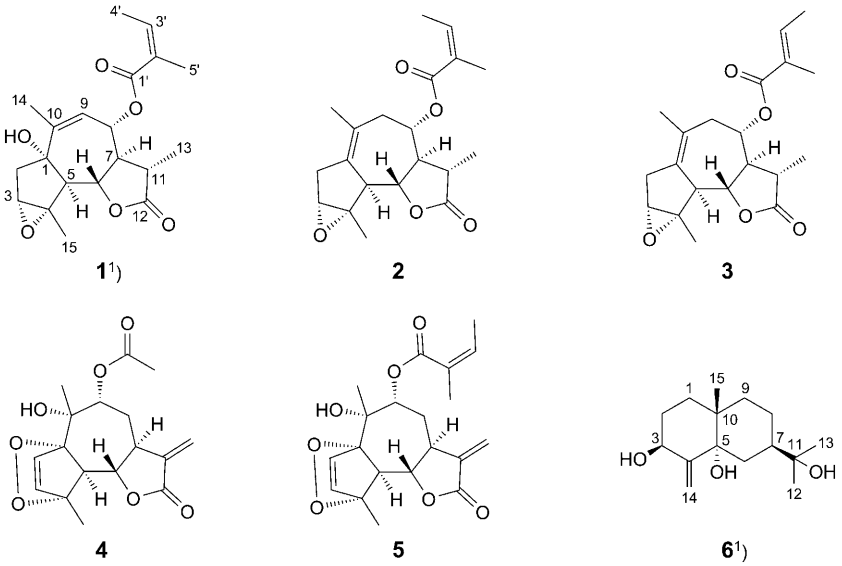


Fig. 1. *Compounds* **1**–**6**, *isolated from* Chrysanthemum indicum *L.*

including two C=O groups (δ(C) 177.3 and 166.9), four olefinic C-atoms (δ(C) 140.6, 138.9, 126.9, and 123.5), five O-bearing C-atoms (δ(C) 80.7, 76.3, 72.4, 67.4, and 63.1), five Me groups (δ(C) 15.7, 16.0, 19.6, 20.5, and 24.7), one CH_2 group (δ(C) 42.1), and three CH groups (δ(C) 59.8, 53.0, and 40.1) (*Table 2*). Analysis of the ^{1}H- and ^{13}C-NMR data as well as of the 2D-NMR spectra (^{1}H,^{1}H-COSY, HSQC, and HMBC) revealed the presence of an angeloyl (=(2*Z*)-2-methylbut-2-enoyl) moiety (δ(H) 2.02 (*d*, *J* = 7.2 Hz), 1.90 (br. *s*), and 6.16–6.22 (*m*); δ(C) 166.9, 140.6, 126.9, 16.0, and 20.5) in **1**. Based on the above evidence and the fact that some sesquiterpenoids were isolated from this genus, compound **1** was suggested to be a sesquitepernoid with a guaiane-type skeleton. In the HMBC spectrum (*Fig. 2*), the long range correlation between H–C(8) (δ(H) 5.48 (br. *d*, *J* = 10.4 Hz)) and C(1′) (δ(C) 166.9) suggested that the angeloyl moiety was linked at C(8) (δ(C) 72.4). The C=C bond between C(9) and C(10) was assigned by the ^{1}H,^{1}H-COSY cross-peak between H–C(8) (δ(H) 5.48 (br. *d*, *J* = 10.4 Hz)) and H–C(9) (δ(H) 5.28 (br. *s*)), and it was also confirmed by the following HMBCs: Me(14) (δ(H) 1.92 (*s*))/C(10 (δ(C) 138.9), Me(14) (δ(H) 1.92 (*s*))/C(9) (δ(C) 123.5), H–C(2) (δ(H) 1.86–1.88 (*m*))/C(10) (δ(C) 138.9). Seven degrees of unsaturation were attributed to two C=O groups, two C=C bonds, and three rings, the remaining degree of unsaturation indicating that **1** had one more ring. The presence of an O-bearing CH group at δ(C) 63.1 and a quaternary C-atom at δ(C) 67.4 suggested the occurrence of an oxirane ring in **1**. The position of the oxirane ring between C(3) (δ(C) 63.1) and C(4) (δ(C) 67.4) was established by the HMBCs between Me(15) (δ(H) 1.69 (*s*)) and C(3) (δ(C) 63.1) and C(4) (δ(C) 67.4). The OH group at C(1) (δ(C) 80.7) was assigned by the ^{1}H,^{1}H-COSY cross-peak between CH_2(2) (δ(H) 2.46–2.50 and 1.86–1.88 (2*m*)) and H–C(3) (3.54 (br. *s*)), and further confirmed by the HMBC cross-peak between Me(14) (δ(H) 1.92 (*s*)) and C(1) (δ(C) 80.7). All the H- and C-

Table 1. *^{1}H-NMR* ($CDCl_3$, 400 MHz) *Data of Compounds* **1**–**6**. δ in ppm, J in Hz.

	1	**2**	**3**	**4**	**5**	**6**
CH_2(1)	–	–	–	–	–	1.09–1.12 (*m*), 1.80–1.81 (*m*)
CH_2(2) or H–C(2)	2.46–2.50 (*m*), 1.86–1.88 (*m*)	2.73 (*d*, *J* = 17.6), 2.30–2.51 (*m*)	2.73 (*d*, *J* = 17.6), 2.45–2.51 (*m*)	6.40 (*d*, *J* = 5.6)	6.46 (*d*, *J* = 5.6)	1.50–1.51 (*m*), 1.68–1.69 (*m*)
H–C(3)	3.54 (br. *s*)	3.41 (br. *s*)	3.40 (br. *s*)	6.37 (*d*, *J* = 5.6)	6.37 (*d*, *J* = 5.6)	4.59–4.63 (*m*)
H–C(5)	2.46 (*d*, *J* = 9.2)	3.02 (*d*, *J* = 10.4)	3.01 (*d*, *J* = 10.4)	2.67 (*d*, *J* = 10.4)	2.67 (*d*, *J* = 10.4)	2.74 (*t*, *J* = 10.0)
H–C(6) or CH_2(6)	3.95 (*dd*, *J* = 9.2, 9.2)	3.75 (*t*, *J* = 10.4)	3.74 (*t*, *J* = 10.4)	3.78 (*t*, *J* = 10.0)	3.80 (*t*, *J* = 10.0)	1.68–1.69 (*m*), 1.81–1.82 (*m*)
H–C(7)	2.46–2.50 (*m*)	2.26 (*dd*, *J* = 10.4, 10.8)	2.26 (*dd*, *J* = 10.4, 10.8)	3.43–3.49 (*m*)	3.47–3.50 (*m*)	1.88–1.91 (*m*)
H–C(8) or CH_2(8)	5.48 (br. *d*, *J* = 10.4)	4.77–4.80 (*m*)	4.76–4.81 (*m*)	2.30–2.38 (*m*), 1.94–2.02 (*m*)	2.41–2.42 (*m*), 1.96–1.95 (*m*)	1.49–1.50 (*m*), 1.87–1.88 (*m*)
H–C(9) or CH_2(9)	5.28 (br. *s*)	2.30–2.51 (*m*), 2.12–2.18 (*m*)	2.22–2.52 (*m*), 2.11–2.15 (*m*)	5.03 (*t*, *J* = 8.0)	5.14 (*t*, *J* = 8.0)	0.92 (*dd*, *J* = 17.6), 1.92–1.95 (*m*)
H–C(11)	2.54–2.56 (*m*)	2.45–2.56 (*m*)	2.45–2.52 (*m*)	–	–	–
Me(12)	–	–	–	–	–	1.14 (*s*)
Me(13) or CH_2(13)	1.29 (*d*, *J* = 6.8)	1.30 (*d*, *J* = 6.4)	1.31 (*d*, *J* = 6.4)	5.45 (*d*, *J* = 3.2), 6.19 (*d*, *J* = 3.2)	5.46 (*d*, *J* = 3.2), 6.19 (*d*, *J* = 3.2)	1.14 (*s*)
Me(14) or CH_2(14)	1.92 (*s*)	1.75 (*s*)	1.75 (*s*)	1.36 (*s*)	1.36 (*s*)	5.14 (br. *s*), 4.76 (br. *s*)
Me(15)	1.69 (*s*)	1.65 (*s*)	1.65 (*s*)	1.71 (*s*)	1.72 (*s*)	0.80 (*s*)
H–C(3′) or AcO	6.16–6.22 (*m*)	6.14–6.19 (*m*)	6.87–6.92 (*m*)	2.16 (*s*)	6.17–6.21 (*m*)	–
Me(4′)	2.02 (*d*, *J* = 7.2)	2.02 (*d*, *J* = 7.2)	1.84 (*d*, *J* = 7.2)	–	2.04 (*d*, *J* = 7.2)	–
Me(5′)	1.90 (br. *s*)	1.90 (br. *s*)	1.85 (br. *s*)	–	1.95 (br. *s*)	–

Table 2. *^{13}C-NMR* ($CDCl_3$, 100 MHz) *Data of Compounds* **1**–**6**. δ in ppm.

No.	**1**	**2**	**3**	**4**	**4**[a])	**5**	**6**
C(1)	80.7 (*s*)	136.2 (*s*)	136.1 (*s*)	98.4 (*s*)	74.1 (*s*)	98.5 (*s*)	35.0 (*t*)
C(2)	42.1 (*t*)	33.3 (*t*)	33.3 (*t*)	133.3 (*d*)	133.3 (*d*)	133.5 (*d*)	33.0 (*t*)
C(3)	63.1 (*d*)	63.6 (*d*)	63.6 (*d*)	137.7 (*d*)	137.6 (*d*)	137.6 (*d*)	69.4 (*d*)
C(4)	67.4 (*s*)	67.0 (*s*)	67.0 (*s*)	93.6 (*s*)	78.5 (*s*)	93.6 (*s*)	153.9 (*s*)
C(5)	59.8 (*d*)	51.3 (*d*)	51.3 (*d*)	69.6 (*d*)	41.7 (*d*)	69.6 (*d*)	77.1 (*s*)
C(6)	76.3 (*d*)	77.6 (*d*)	77.6 (*d*)	78.4 (*d*)	72.1 (*d*)	78.4 (*d*)	32.2 (*d*)
C(7)	53.0 (*d*)	60.4 (*d*)	60.5 (*d*)	41.6 (*d*)	69.7 (*d*)	41.7 (*d*)	44.1 (*d*)
C(8)	72.4 (*d*)	70.8 (*d*)	71.2 (*d*)	30.2 (*t*)	30.4 (*t*)	30.3 (*t*)	17.1 (*t*)
C(9)	123.5 (*d*)	42.0 (*t*)	41.8 (*t*)	71.9 (*d*)	98.4 (*d*)	71.5 (*d*)	36.4 (*t*)
C(10)	138.9 (*s*)	128.9 (*s*)	128.9 (*s*)	73.9 (*s*)	93.7 (*s*)	74.1 (*s*)	37.5 (*s*)
C(11)	40.1 (*d*)	40.9 (*d*)	40.9 (*d*)	139.1 (*s*)	139.3 (*s*)	169.4 (*s*)	73.9 (*s*)
C(12)	177.3 (*s*)	177.2 (*s*)	177.3 (*s*)	169.4 (*s*)	169.2 (*s*)	139.2 (*s*)	28.7 (*q*)
C(13)	15.7 (*q*)	15.1 (*q*)	15.1 (*q*)	120.0 (*t*)	119.7 (*t*)	120.0 (*t*)	29.5 (*q*)
C(14)	24.7 (*q*)	22.2 (*q*)	22.2 (*q*)	21.6 (*q*)	21.6 (*q*)	21.9 (*q*)	21.4 (*q*)
C(15)	19.6 (*q*)	19.0 (*q*)	19.0 (*q*)	13.7 (*q*)	13.6 (*q*)	13.7 (*q*)	106.3 (*t*)
C(1′)	166.9 (*s*)	166.7 (*s*)	166.8 (*s*)	170.2 (*s*)	170.1 (*s*)	166.8 (*s*)	–
C(2′)	126.9 (*s*)	127.2 (*s*)	128.3 (*s*)	20.8 (*q*)	20.8 (*q*)	126.9 (*s*)	–
C(3′)	140.6 (*d*)	140.0 (*d*)	138.6 (*d*)	–	–	140.4 (*d*)	–
C(4′)	20.5 (*q*)	20.5 (*q*)	14.6 (*q*)	–	–	20.6 (*q*)	–
C(5′)	16.0 (*q*)	15.9 (*q*)	12.0 (*q*)	–	–	16.0 (*q*)	–

[a]) ^{13}C-NMR Data assignment reported by *Ognyanov* [8].

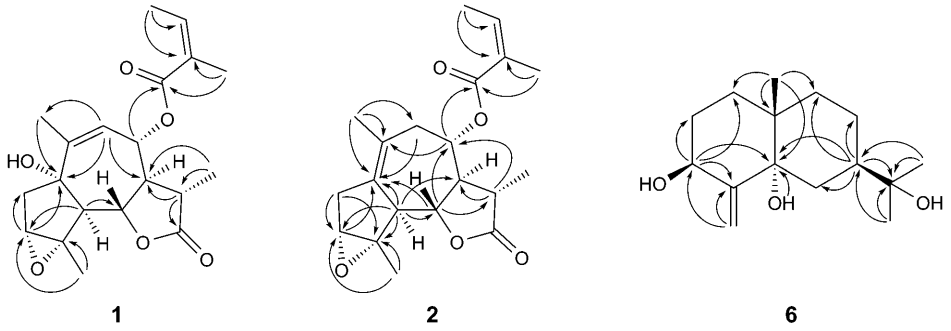
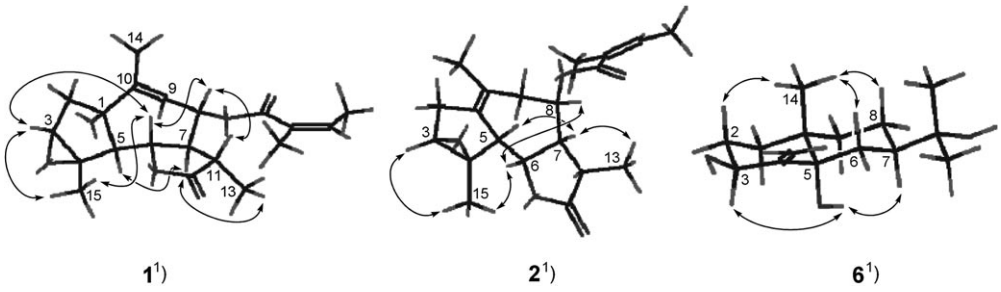


Fig. 2. *Selected HMBCs of compounds* **1**, **2**, *and* **6**

atoms were unambiguously assigned by the analysis of the ^{1}H- and ^{13}C-NMR, ^{1}H,^{1}H-COSY, HSQC, and HMBC data (*Tables 1* and *2*). The relative configuration of **1** was established by the NOESY correlations H–C(3)/Me(15)/H–C(6)/H–C(8)/H–C(11), suggesting that H–C(3), H–C(6), H–C(8), H–C(11), and Me(15) were β-oriented, and by the cross-peaks H–C(5)/H–C(7) and H–C(7)/Me(13), suggesting that H–C(5) and H–C(7) were α-oriented (*Fig. 3*). The downfield chemical shift of H–C(5) (δ(H) 2.46) supported the α-orientation of OH–C(1) [16]. Thus, the structure of **1** was elucidated as (3α,6α,8α)-angeloyl-3,4-epoxy-1-hydroxyguai-9-eno-12,6-lactone, and named chrysanthguaianolactone A[1]).



1[1]) **2**[1]) **6**[1])

Fig. 3. *Selected NOESY correlations of compounds* **1**, **2**, *and* **6**

Compound **2** was obtained as colorless crystals. Its molecular formula was elucidated as $C_{20}H_{26}O_5$ by HR-ESI-MS, which gave a quasi-molecular-ion peak at *m*/*z* 369.1679. The ^{1}H- and ^{13}C-NMR spectra indicated the presence of an angeloyl moiety (δ(H) 2.02 (*d*, *J* = 7.2 Hz), 1.90 (*s*), and 6.14–6.19 (*m*); δ(C) 166.7, 140.0, 127.2, 15.9, and 20.5) in **2**. Comparison of its NMR data with those of **1** suggested that compound **2** possessed a skeleton similar to that of **1**. In compound **2**, the disappearance of the quaternary C-atom at δ(C) 80.7 and the presence of one more CH_2 group (δ(H) 2.12–2.18 and 2.30–2.51 (2*m*); δ(C) 42.0) suggested that the C=C bond in **2** was most likely between C(1) and C(10), which was confirmed by the HMBC correlations Me(14) (δ(H) 1.75 (*s*)/C(1) (δ(C) 136.2), C(10) (δ(C) 128.9), and C(9) (δ(C) 42.0). The relative configuration was also established by the NOESY correlations H–C(3) (δ(H) 3.41 (br. *s*))/Me(15) (δ(H) 1.65 (*s*))/H–C(6) (δ(H) 3.75 (*t*, *J* = 10.4 Hz))/H–C(8) (δ(H) 4.77–4.80 (*m*))/H–C(11) (δ(H) 2.45–2.56 (*m*)), suggesting that H–C(3), H–C(6), H–C(8), H–C(11), and Me(15) were β-oriented, and by the cross-peaks between H–C(5) (δ(H) 3.02 (*d*, *J* = 10.4 Hz)) and H–C(7) (δ(H) 2.26 (*dd*, *J* = 10.4 and 10.8 Hz)), suggesting that H–C(5) and H–C(7) were α-oriented (*Fig. 3*). The ^{1}H-NMR data of **2** were almost identical to those of the known isomer (3α,6α,8α)-3,4-epoxy-8-tigloylguai-1(10)-eno-12,6-lactone (**3**), except for the substituent at C(8) (δ(C) 42.0) (tigloyl = (2*E*)-2-methylbut-2-enoyl). In compound **2**, the Me groups of the substituent at C(8) appeared at more downfield chemical shifts (δ(C) 15.9 and 20.5) as compared to those of the Me groups of **3** (δ(C) 12.0 and 14.6; γ-*gauche* effect when the two Me groups are *cis*-oriented). Thus, the structure of compound **2** was unambiguously elucidated as (3α,6α,8α)-8-angeloyl-3,4-epoxyguai-1(10)-eno-12,6-lactone, and named chrysanthguaianolactone B[1]).

Compounds **3** and **5** were obtained as colorless crystals. These two compounds have been only reported by *Greger* and co-workers in 1986 [7], and *Bohlmann* and *Knoll* in 1979 [9], respectively, while the ^{13}C-NMR data of **3** and **5** have never been reported. We now unambiguously assigned all the H- and C-atoms of **3** and **5** by analysis of the 1D- and 2D-NMR spectra including ^{1}H,^{1}H-COSY, HSQC, HMBC, and NOESY data (*Tables 1* and *2*). *Bohlmann* and *Knoll* suggested that the endoperoxide moiety in **5** was β-oriented, while the NOESY correlations between Me(15), Me(14), and H–C(6) strongly suggested that the endoperoxide moiety in **5** was α-oriented, which well agreed with the result reported by *Ognyanov* and co-workers in 1981 [8]. In addition, *Ognyanov* and co-workers have also reported the isolation and structural elucidation of

a new sesquiterpene named apressin from *Achillea depressa*, whose ^{1}H- and ^{13}C-NMR data were completely identical to those of compound **4**. However, an unambiguous assignment of the NMR data of **4** revealed that the C-atom assignments reported in [8] were not completely accurate. Thus, we re-assigned all the H- and C-atoms of **4** on the basis of its NMR evidence including ^{1}H,^{1}H-COSY, HSQC, HMBC, and NOESY data (*Tables 1* and *2*).

Compound **6** was obtained as colorless crystals. Its molecular formula $C_{15}H_{26}O_3$ was determined by HR-ESI-MS (*m/z* 277.1772 ($[M+Na]^+$)). The IR spectrum showed the existence of OH groups (3422 cm^{-1}) and a C=C bond (1649 cm^{-1}). The characteristic features of its ^{1}H- and ^{13}C-NMR spectra were very similar to those of (3*β*,6*β*)-eudesm-4(15)-ene-3,5,6,11-tetrol [12], suggesting that **6** was also an eudesmane-type sesquiterpene. The only difference between **6** and this tetrol was at C(6): for **6**, the C(6) was a CH_2 group without an OH group, which was confirmed by ^{1}H-NMR (δ(H) 1.81–1.82 (*m*, H_β–C(6)) and 1.68–1.69 (*m*, H_α–C(6)) and ^{13}C-NMR and DEPT (δ(C) 32.2). All the H- and C-atoms were unambiguously assigned by ^{1}H,^{1}H-COSY and HSQC, and confirmed by HMBC. The configuration of **6** was achieved by the analysis of its NOESY plot, the cross-peaks between Me(14) (δ(H) 0.80 (*s*)) and H_{ax}–C(2) (δ(H) 1.50–1.51 (*m*)), H_{ax}–C(6) (δ(H) 1.68–1.69 (*m*)), and H_{ax}–C(8) (δ(H) 1.49–1.50 (*m*)) suggesting a *trans*-ring junction, and the cross-peaks between OH–C(5) and H–C(3) (δ(H) 4.59–4.63 (*m*)) and H–C(7) (1.88–1.91 (*m*)) revealing the equatorial positions of the substituents at C(3) (δ(C) 69.4) and C(7) (δ(C) 44.1). Thus, the structure of compound **6** was elucidated as (3*β*)-eudesm-4(14)-ene-3,5,11-triol, and named chrysanthemdiol A[1]).

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Experimental Part

General. TLC: silica gel *GF$_{254}$* (SiO_2; 10–40 μm, *Qingdao Marine Chemical Factory*, Qingdao, P. R. China); detection at 254 nm or by heating after spraying with 1.0% anisaldehyde/H_2SO_4 in EtOH. Column chromatography (CC): SiO_2 (200–300 mesh), *Sephadex LH-20* (*17-0090-02*; *Amersham Bioscience*, Sweden) and *ODS* (prep. *C$_{18}$*, 12.5 nm, 55–105 μm; *Waters*). M.p.: *WC-1* micro-melting-point apparatus; uncorrected. Optical rotations: *Perkin-Elmer* model *341* and *Polar 3001*. IR Spectra: *PE-1710* FT-IR spetrometer; KBr pellets; $\tilde{\nu}$ in cm^{-1}. UV Spectra: *Shimadzu-UV-260* spectrometer; λ_{max} (log ε) in nm. NMR Spectra: *Bruker-DPX-400* NMR spectrometer; δ in ppm rel. to Me_4Si as internal standard, *J* in Hz. HR-ESI-MS: *Waters-Q-Tof* MS instrument; in *m/z*. X-Ray analysis: *Rigaku-R-AXIS* X-ray instrument.

Plant Material. The flowers of *Chrysanthemum indicum* L. were collected from Fangcheng County, Henan Province, P. R. China, in November 2008, and identified by Prof. *Ruo-Yong Liu* and *Cheng-Xue Pan* at the University of Zhengzhou. A voucher specimen (No. 2008012) was deposited with the Herbarium of the School of Pharmaceutical Science, University of Zhengzhou, P. R. China.

Extraction and Isolation. Air-dried flowers (5 kg) were extracted with 95% EtOH (3 × 6 l) by a tissue-smashing extractor, and the combined extracts were concentrated (yield 1.3 kg). A portion of the residue (300 g) was suspended in H_2O (5 l) and then partitioned successively with petroleum ether, petroleum ether (60–90°)/AcOEt 1:1, and AcOEt (5 × 4 l). Each fraction was separately concentrated which afforded 10.0, 67.8, and 48.2 g of extract, resp. The petroleum ether/AcOEt 1:1 fraction (40 g) was subjected to CC (SiO_2, gradient of petroleum ether (60–90°)/acetone): *Fractions A–J. Fr. C* (5.25 g) was

subjected to CC (*ODS*, 60% MeOH) and continuously purified by CC (SiO_2): **1** (8 mg; with petroleum ether/AcOEt 9:2), **2** (40 mg; with petroleum ether/$CHCl_3$/AcOEt 7:1:2), **3** (10 mg; with petroleum ether/AcOEt 7:2), **4** (35 mg; with petroleum ether/AcOEt 2:1), **5** (11 mg; with petroleum ether/acetone 3:1), and cumambrin A (12 mg; with petroleum ether/AcOEt 5:2). *Fr. D* (2.52 g) was applied to CC (*ODS*, 40% MeOH) and repeated CC (SiO_2): **6** (10 mg; with petroleum ether/AcOEt/MeOH 10:2:1), (3*β*,6*β*)-eudesm-4(14)-ene-3,5,6,11-tetrol (35 mg, petroleum ether/AcOEt/MeOH 10:2:1), (+)-eudesm-4(14)-ene-11,13-diol (21 mg; with petroleum ether/acetone 3:2), and cryptomeridiol (45 mg; with petroleum ether/acetone 3:2).

Chrysanthguaianolactone A (= rel-*(3*R,*3a*S,*4*R,*6a*R,*7a*S,*8a*R,*8b*S,*8c*R*)-2,3,3a,4,6a,7,7a,8a,8b,8c-Decahydro-6a-hydroxy-3,6,8a-trimethyl-2-oxooxireno[2,3]azuleno[4,5-*b*]furan-4-yl (2*Z*)-2-Methylbut-2-enoate*; **1**): M.p. 164.0–165.0°. $[\alpha]_D^{25} = +17.3$ ($c = 0.052$, $CHCl_3$). IR: 3563, 1752, 1718, 1646. ^{1}H- and ^{13}C-NMR ($CDCl_3$, 400 and 100 MHz, resp.): *Tables 1* and *2*. HR-ESI-MS: 385.1627 ($[M+Na]^+$, $C_{20}H_{26}NaO_6^+$; calc. 385.1627).

Chrysanthguaianolactone B (= rel-*(3*R,*3a*S,*4*R,*7a*S,*8a*R,*8b*R,*8c*R*)-2,3,3a,4,5,7,7a,8a,8b,8c-Decahydro-3,6,8a-trimethyl-2-oxooxireno[2,3]azuleno[4,5-*b*]furan-4-yl (2*Z*)-2-Methylbut-2-enoate*; **2**): M.p. 164.0–165.0°. $[\alpha]_D^{25} = +17.3$ ($c = 0.052$, $CHCl_3$). IR: 3735, 1782, 1702, 1646. ^{1}H- and ^{13}C-NMR ($CDCl_3$, 400 and 100 MHz, resp.): *Tables 1* and *2*, resp. HR-ESI-MS: 365.1679 ($[M+Na]^+$, $C_{20}H_{26}NaO_5^+$; calc. 365.1680).

Chrysanthemdiol A (= rel-*(2*R,*4a*R,*7*S,*8a*R*)-Octahydro-7-(1-hydroxy-1-methylethyl)-4a-methyl-1-methylenenaphthalene-2,8a(1*H*)-diol*; **6**): Colorless crystals. M.p. 386–388°. $[\alpha]_D^{25} = +96$ ($c = 0.15$, acetone). IR: 3427, 3113, 1649. ^{1}H- and ^{13}C-NMR ($CDCl_3$, 400 and 100 MHz, resp.): *Tables 1* and *2*, resp. HR-ESI-MS: 277.1772 ($[M+Na]^+$, $C_{15}H_{26}NaO_3^+$; calc. 277.1780).

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