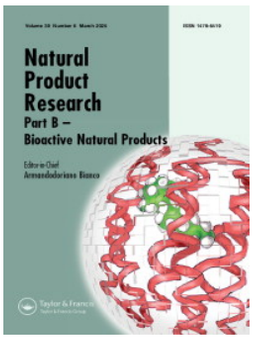

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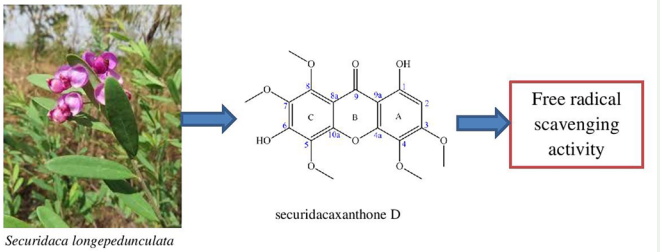
Securidacaxanthone D: a new xanthone with free radical scavenging activity from the roots of *Securidaca longepedunculata* (Polygalaceae)

Janvier Hinmara Tina[a], Cyrille Ngoufack Tagousop[b], Boubakary Abdouraman[a], Joël Abel Gbaweng[a], Roméo Feunaing Toko[a], Judith Djithe Djouakoua[a], Esther Lita Morsia[a], Céline Henoumont[c], Sophie Laurent[c] and Emmanuel Talla[a]

[a]Department of Chemistry, Faculty of Science, University of Ngaoundere, Ngaoundere, Cameroon; [b]Department of Basic Scientific Studies, University Institute of Technology, University of Ngaoundere, Ngaoundere, Cameroon; [c]Department of General Organic and Biomedical Chemistry, Faculty of Medicine and Pharmacy, University of Mons, Belgium, Mons, Belgium

ABSTRACT

A previously unpublished xanthone named securidacaxanthone D (**1**) and five known analogues: 1,7-dihydroxyxanthone (**2**), 7-hydroxy-1,2-dimethoxyxanthone (**3**), 1,5-dihydroxy-6,7-dimethoxyxanthone (**4**), 1,7-dihydroxy-3-methoxyxanthone (**5**), 1,7-dihydroxy-4-methxyxanthone (**6**) were isolated from the ethyl acetate soluble fraction obtained from the liquid-liquid partition of the crude hydro-ethanolic extract of the roots of *Securidaca longipedunculata*. Their structures were determined by analysis of 1D -(^{1}H and ^{13}C), 2D-(COSY, HSQC and HMBC) NMR data in conjunction with mass spectrometry (TOF-ESI-MS) and by comparison with reported data. Free radical scavenging activity were evaluated using 1,1-diphenyl-2-picrylhydrazyl (DPPH) as a free radical. Crude extract, ethyl acetate and *n*-butanol fractions as well as isolated compounds showed different degrees of free radical scavenging activity. Among the extract and fractions tested, the hydro-ethanolic extract was the most active with an IC_{50} value of 32 µg/mL. Among the compounds, compound **2** was the most active with an IC_{50} value of 282 µg/mL.



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CONTACT Cyrille Ngoufack Tagousop cyrilngoufack@yahoo.fr; Emmanuel Talla tallae2000@yahoo.fr
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1. Introduction

The genus *Securidaca* is a member of the Polygalaceae family and occurs throughout the tropical and some temperate environments. This genus comprises 80 species characterised by its keel with or without a small fringed crest, its eight stamens with filaments fused towards the base, and its 2- locular, winged fruit. Among the 80 recognised species only two recorded from Africa: *S. welwitschii and S. longipedunculata* (Johnson 1987). In Africa, the roots of *S. longipedunculata* are widely used in folk medicine in the treatment of a variety of ailments including coughs, fever, malaria, tuberculosis, skin diseases, stomach ache, rheumatism, pains, pneumonia and sexually transmitted diseases (Mongalo et al. 2015). Previous biological studies have demonstrated the antiplasmodial (Sekou et al. 2006), analgesic, anti-inflammatory (Ojewole 2008), anticonvulsivant (Adeyemi et al. 2010) and antioxidant (Muanda et al. 2010) properties of the roots of this plant. Previous phytochemical investigation on this plant led to the isolation of bioactive constituents such as: saponins (Stevenson et al. 2009; Mitaine-Offer et al. 2010), tannins, anthraquinones, alkaloids, terpenes, sterols, (Mongalo et al. 2015; Ogukwe et al. 2015; Obasi et al. 2020), methyl salicylate (Jayasekara et al. 2002) and xanthones (Lannang et al. 2006; Meli et al. 2007; Dibwe et al. 2013). Considering the previous results and in the course of our continuous search for secondary metabolites with potentially interesting bioactivity, we investigated the EtOAc soluble fraction from the crude hydro-ethanolic extract of the roots of *Securidaca longipedunculata*. We report here the isolation and structure elucidation of six xanthones, including one previously undescribed.

2. Results and discussion

2.1. Structural elucidation of the isolated compounds

The purification by silica gel and Sephadex LH 20 column chromatography of the EtOAc soluble fraction obtained from liquid-liquid partition of the hydro-ethanolic crude extract of *Securidaca longipedunculata* led to the isolation of a new xanthone, securidacaxanthone D (**1**), together with five known analogues (**2–6**) (Figure 1). The known compounds were identified as: 1,7-dihydroxyxanthone **(2)** (Nagem and Oliveira 1997), 7-hydroxy-1,2-dimethoxyxanthone **(3)** (Lin et al. 2005), 1,5-dihydroxy-6,7-dimethoxyxanthone **(4)** (Cholpisut et al. 2016), 1,7-dihydroxy-3-methoxyxanthone **(5)** (Yukinobu et al. 1991), 1,7-dihydroxy-4-methoxyxanthone **(6)** (Marston et al. 1993). (See Supplementary Material, Tables S1 and S2).

Compound **1** was obtained as yellow amorphous powder. Mass measurement by TOF-ESI-MS revealed a sodium adduct peak at *m/z* 401.0 $[M+Na]^+$ and *m/z* 779.0 $[2M+Na]^+$ (see Supplementary Material, Figure S7). The UV spectrum displayed absorption bands at λ_{Max} 315 and 250 nm, which is typical of the xanthone chromophore (Kaennakam et al. 2015). The IR spectrum showed phenolic hydroxyl groups (3400–3200 cm^{-1}), a carbonyl group (1650 cm^{-1}) and aromatic carbons (1500 cm^{-1}). The molecular formula, $C_{18}H_{18}O_9$ (calcd. m/z 378.0) of compound **1** was established from its MS and NMR data, which accounted for 10 degrees of unsaturation. Its ^1H NMR spectrum (see Supplementary Material, Figure S1) exhibited the signal of five methoxy groups at δ_H: 3.96 (3H, *s*), 3.97 (3H, *s*), 4.03 (3H, *s*), 4.00 (3H, *s*), 4.14 (3H, *s*), an

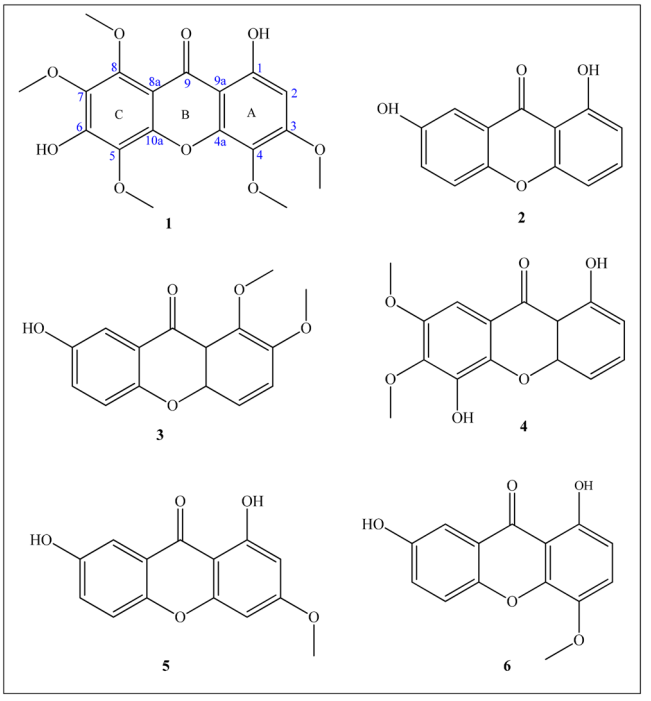

Figure 1. Structures of isolated compounds (1–6) from S. *longepedunculata*.

aromatic proton at δ_H 6.40 (1H, *s*) and a chelated hydroxyl group at δ_H 13.20 (1H, *s*). Extensive analysis of its ^{13}C NMR spectrum (see Supplementary Material, Figure S2) in conjunction with the HSQC experiment (see Supplementary Material, Figure S5) and MS data indicated that compound **1** was a xanthone with five methoxy groups (Westerman et al. 1977). Comparison of the ^{13}C NMR data of compound **1** with that of 1,5-dihydroxy-3,4,6,7,8-pentamethoxyxanthone (securidacaxanthone B) (Meli et al. 2007; Dibwe et al. 2013) showed that, they were the same in the A and B rings. δ_C: 159.2 (C-1), 94.8 (C-2), 159.1 (C-3), 128.3 (C-4), 147.1 (C-4a), 148.2 (C-10a), 108.1 (C-8a), 103.0 (C-9a), 180.5 (C-9), 61.5 (3-OMe), 56.3 (4-OMe). The ^1H-^1H COSY spectrum (see Supplementary Material, Figure S4) confirmed the absence of coupled hydrogens in compound **1**. The position of two methoxy groups was confirmed by the correlation observed in the HMBC spectrum (see Supplementary Material, Figure S6) between the proton signals at δ_H: 3.96 (3H, *s*), 3.97 (3H, *s*) and carbons at δ_C: 128.2 (C-4), 159.1 (C-3). In addition, correlations observed in the same HMBC spectrum between the proton signal at δ_H 6.40 (1H, *s*) and the carbon signals at δ_C: 103.0 (C-9a), 128.2 (C-4), 159.1 (C-3), 159.2 (C-1), allowed us to confirm the position 2 of this proton. The main difference between the two compounds was observed in the C ring. Indeed, for 1,5-dihydroxy-3,4,6,7,8-pentamethoxyxanthone (securidacaxanthone B) (Meli et al. 2007; Dibwe et al. 2013) we have the following chemical shifts δ_C: 148.1(C-10a), 128.1(C-5), 137.7(C-6), 133.0 (C-7), 146.9 (C-8), 108.1 (C-8a) and for compound **1** we have δ_C: 148.2 (C-10a), 137.8 (C-5), 148.7 (C-6), 131.5 (C-7), 148.6 (C-8), 108.1 (C-8a) (see Supplementary Material, Table S1). The analysis of different chemical shifts showed the downfield of a signal δ_C: 128.1 (C-5) and 137.7 (C-6) for

securidacaxanthone B to δ_C: 138.0 (C-5) and 148.7 (C-6) for compound **1.** This observation suggests that compound **1** and securidacaxanthone B are a positional isomers. Considering the fact that the C ring does not have a proton, we exploited the reported data to determine the environment of the C ring. In fact, it was reported by Chaudhuri et al. 1978, that the chemical shift value for a methoxy carbon surrounded by two *ortho* substituents (OMe, O-aryl or CO-aryl) in the ^{13}C-NMR spectrum of a polymethoxyxanthone is shifted downfield to δ_C (60–62). The appearance of downfield methoxy signals at δ_C: 61.6, 61.7 and 62.1 indicated the presence of three methoxy groups flanked by two oxygenated substituents. In addition, according to Yukinobu et al. 1991, a methoxy group at position 1 or 8 of a polymethoxyxanthone, when surrounded by *ortho* and *meta* oxygenated substituents, is shifted downfield to 62–63 ppm. Therefore, in the ^{13}C-NMR spectrum of compound **1**, which has a chelated hydroxyl group at position 1, the methoxy group at δc 62.1 should be at position 8. The correlation observed in the HMBC spectrum (see Supplementary Material, Figure S6) between the proton at δ_H 4.00 and the carbon atom at δ_C 148.6 allowed us to identify the chemical shift value of carbon C-8. In addition, the correlation between the proton at δ_H: 4.03 (3H, *s*), 4.14 (3H, *s*) and the carbons at δ_H: 131.5 and 137.8, respectively, allowed us to locate the other methoxy group at position C-7 and C-5. The structure of compound **1** was thus established as: 1,6-dihydroxy-3,4,5,7,8-pentamethoxyxanthone and trivially named securidacaxanthone D.

2.2. DPPH radical scavenging capacity assay

The radical scavenging capacity of the hydro-ethanolic extract, ethyl acetate and the *n*-butanol fraction as well as isolated compounds and ascorbic acid used as reference was measured by the *in vitro* DPPH assay. The results are shown in Table S3 and Figure S10 (see Supplementary Material). The hydro-ethanolic extract (IC_{50}= 32 µg/mL) showed the highest radical scavenging activity, followed in decreasing order by the *n*-butanol fraction (IC_{50} = 80 µg/mL), ethyl acetate fraction (IC_{50}= 101 µg/mL), compound **2** (IC_{50} = 282 µg/mL), compound **1** (IC_{50} = 320 µg/mL), compounds **3 + 4** (IC_{50} = 479 µg/mL), compound **5** (IC_{50} = 637 µg/mL) and compound **6** (IC_{50}= 683 µg/mL).

3. Experimental

3.1. General experiental procedure

The 1D (^{1}H and ^{13}C-NMR) and 2D (COSY, HSQC and HMBC) spectra were performed in deuterated solvents ($CDCl_3$ and CD_3OD) on Bruker spectrometer at 500/600 MHz for ^{1}H and 125/150 MHz for ^{13}C. All chemical shifts (δ) are given in ppm units with reference to tetramethylsilane (TMS) as internal standard and the coupling constants (*J*) are in Hz. Mass spectrometry (TOF-ES-MS) was conducted using QTOF-MS-LD+ equipment. Column chromatography (CC) was carried out using silica gel (Merck, particle size 230–400 mesh) as the adsorbent, while thin-layer chromatography (TLC) was performed on silica gel pre-coated aluminium sheets (Merck KGaA). The spots were visualised by spraying with 10% H_2SO_4 and heating at 100 °C for 2 min.

3.2. Plant collection and identification

The roots of *S. longepedunculata* were harvested in the northern region of Cameroon in August 2023. The plant material was identified by Dr. Bathelemy Tchiengue at the National Herbarium of Cameroon, Yaoundé, where a voucher specimen was deposited under the reference Number 8686/SRF/CAM.

3.2. Extraction and isolation

The air-dried plant material (1.3 kg) was powdered and extracted at room temperature with Hydro-Ethanolic mixture (30:70), (3 × 7 L, 72 h). The solvent was evaporated under reduced pressure, leaving crude extract (399.4 g). Part of this extract (385 g) was suspended in water (250 mL) and successively extracted with equal volumes (400 mL) of ethyl acetate (EtOAc) and *n*-BuOH yielding, respectively, 15.2 and 152.6 g of fractions after evaporation of the solvent under reduced pressure. Part of the EtOAc extract (12.2 g) was subjected to silica gel column chromatography using *n*-Hexane-EtOAc (100:0→0:100) followed by EtOAc-MeOH (100:0→90:10) gradient elution. A sub-fraction of 150 mL each were collected and subsequently combined on the basis of their TLC profiles, resulting in the formation of eight distinct sub-fractions labelled A, B, C, D, F, G and H. Sub-fraction B (100 mg) was purified on silica gel column chromatography eluted with *n*-hexane-EtOAc (8.5:1.5) to give compound **1** (18.7 mg) and compound **2** (11.6 mg). Sub-fraction C (118.6 mg) yielded to compounds **5** (13.1 mg) and **6** (26 mg) after purification on silica gel column chromatography with *n*-hexane-EtOAc (8:2) as eluent. Sub-fraction D (150 mg) was purified on Sephadex LH-20 column eluted with methanol to give the mixture of compounds **3** (16,2 mg) and **4** (42.4 mg).

3.3. New compound information

Securidacaxanthone D: yellow amorphous powder; TOF-ESI-MS (positive ion mode) *m/z* 401.0 $[M+Na]^+$ and *m/z* 779.0 $[2M+Na]^+$ (see Supplementary Material, Figure S7); UV ($CHCl_3$) λ_{max}: 315 and 250 nm, (see Supplementary Material, Figure S8); IR (KBr) υ_{max}: 3400–3200, 1620 and 1500 cm^{-1} (see Supplementary Material, Figure S9); ^1H and ^{13}C NMR data (see Supplementary Material, Figures S1 and S2).

3.4. DPPH radical scavenging capacity assay

2,2-diphenyl-1-picryl-hydrazyl (DPPH) is a stable radical with an odd electron. When reacted with antioxidants, DPPH radical could acquire an electron or a hydrogen from antioxidants. The *in vitro* radical-scavenging activity of extracts as well as isolated xanthones was performed according to Brand-Williams et al. 1995.

4. Conclusion

The results of the present study indicate that the six purified xanthones including a novel one: 1,6-dihydroxy-3,4,5,7,8-pentamethoxyxanthone (securidacaxanthone D) (**1**), the ethyl acetate and n-BuOH soluble fractions as well as the hydroethanolic extract

of the roots of *S. longepedunculata* showed different free radical scavenging activity against 2,2-diphenyl-1-picryl-hydrazyl (DPPH).

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Disclosure statement

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