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Chemical Constituents from *Psychotria cadigensis* and their chemotaxonomic relevance

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Abstract
Phytochemical investigation on the leaves of *Psychotria cadigensis*, a Philippine endemic Rubiaceae species, led to the identification of three structurally-related *nor*-sesquiterpenoids (1-3). This represents the pioneering work on *P. cadigensis* and an addition to the list of *Psychotria* species which does not contain alkaloids. This is also the first report of three from the genus *Psychotria*.

Keywords: Psychotria, Rubiaceae, nor-sesquiterpenoid, chemotaxonomy.

1. Introduction
The genus *Psychotria* (Rubiaceae, Rubioideae, Psychotrieae) represents the largest member of the coffee family with 1800 species distributed worldwide [1]. This genus is also characterized by an intricate taxonomy and the relationships among the species are unclear, especially the endemic species found in Southeast Asia [2]. In the Philippines, there are about 112 species and almost all are endemic. In our interest on the phytochemical investigation on the Philippine endemic *Psychotria* species [3], we herein report the isolation of three norisoprenoids, vomifoliol (1), loliolide (2) and isololiolide (3) (Figure 1) from the leaves of *P. cadigensis*. Extensive literature search revealed that no studies have been conducted yet on *Psychotria cadigensis*.

2. Materials and methods
2.1 Plant Material
Fresh leaves of *Psychotria cadigensis* Merr. were collected at Mt. Malasimbo, PuertoGalera, Oriental Mindoro, Philippines in November 2012. It was collected and identified by one of the authors (MMU) and voucher specimens (USTH No. 011613, 011614, 011615, 011616, 011617) were deposited at the UST-Herbarium, Thomas Aquinas Research Complex, University of Santo Tomas, Philippines.

2.2 Extraction and Isolation
Air-dried, ground leaves of *P. cadigensis* (93 g) were percolated in technical grade MeOH (3.0 L) for three days and filtered. The combined filtrates were concentrated under reduced pressure to obtain the crude extract (20 g). The crude extract was suspended in distilled H_2O and partitioned with hexane (6.5 g), $CHCl_3$ (3.8 g) and *n*-BuOH (2.9 g), respectively. The $CHCl_3$ extract was initially separated by SiO_2 gravity column chromatography using neat $CHCl_3$, 2% MeOH/$CHCl_3$, 5% MeOH/$CHCl_3$, 10% MeOH/$CHCl_3$, 20% MeOH/$CHCl_3$, neat MeOH to afford 4 pooled fractions (Fr A-D). Fraction B (0.6 g) was chromatographed twice by flash CC using 70% EtOAc/hexane and 50% EtOAc/hexane to afford compound **1** (3.5 mg, colorless oil) and **2** (1.4 mg, colorless oil). Fraction C (0.4 g) was chromatographed by flash CC using 60% EtOAc/hexane (2x) and 40% EtOAc/hexane to afford **3** (1.1 mg, colorless oil).

3. Results & Discussion
This is the first report on the phytochemical work on *Psychotria cadigensis* which afforded the isolation and identification of structurally-related norisoprenoids (**1-3**). The isolated compounds were identified by 1D and 2D-NMR analyses and in comparison with the literature data as vomifoliol (**1**) [4], loliolide (**2**) [5] and isololiolide (**3**) [5]. The identification of **3** from the genus *Psychotria* is also reported for the first time. Compounds **1** and **2** have been previously isolated

from *P. yunnanesis* [6] while *P. gitingensis* earlier identified **1** as a metabolite [3]. Previous studies have also identified similar compounds such as megastigmanes and their glycosides from *P. correae* [7] and *P. stachyoides* [8].

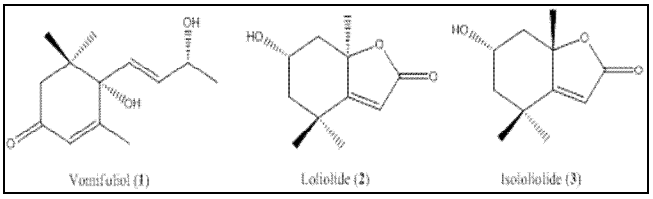

Fig 1: Isolated compounds from *Psychotria cadigensis*.

The *Psychotria* species are known to elaborate the presence of alkaloids [9, 10]. Thus, this work adds to those species which are devoid of the presence of alkaloids. This suggested that those non-alkaloid *Psychotria* plants, especially those distributed in the Asia Pacific region, may present an implication on their chemotaxonomic relationship to the other alkaloid-containing *Psychotria* species. Based on the proposition of Lu *et al*. [6], those species without alkaloids may exhibit a different biosynthetic route capable of forming another subgroup with the same genus. Further phytochemical investigation on other *Psychotria* species, especially those distributed from Asia, should be explored to facilitate the classification and molecular phylogeny of this abundant genus.

4. Conclusions

This is the pioneering work on the Philippine endemic *Psychotria cadigensis* leading to the identification of vomifoliol, loliolide and isololiolide. This study also adds to the number of *Psychotria* species which does not contain alkaloids. As such, some compounds can be used to classify infrageneric relationships within the *Psychotria* genus particularly in the assigning of their taxonomic sections.

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