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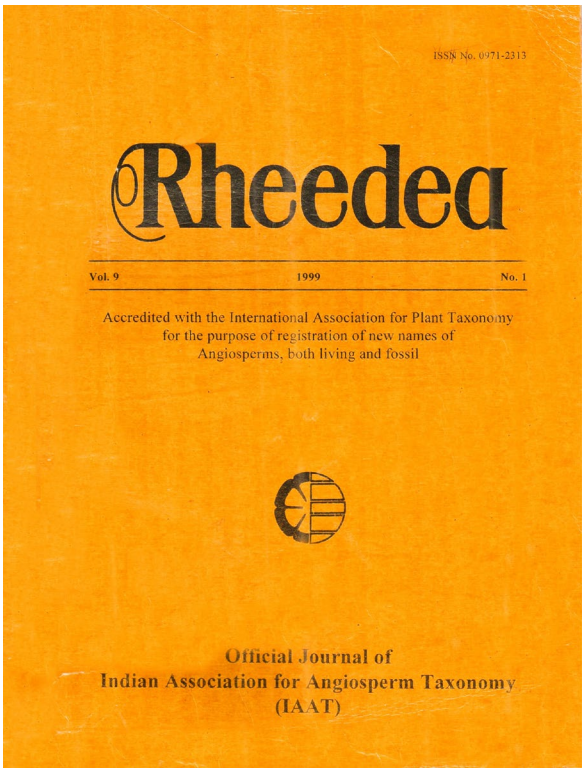
Padma Rao P.



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Chemotaxonomy of Some Indian *Clerodendrum* L.

P. Padma Rao
Drug Standardisation Unit-H
O.U. B-32, Street No. 4
Vikrampuri, Hyderabad-500 007, India.

Abstract

A comparison of chemical constituents in the leaves of eleven taxa of *Clerodendrum* L. is studied. In all 26 chemical compounds were detected and were statistically evaluated for their interrelationships. Present study shows that chemically 4 groups have evolved in *Clerodendrum*. The conclusions have been compared with the delimitations of the taxa proposed in earlier classifications. The groups presently arrived at are distinct and agree partially with Moldenke's subgeneric treatment.

INTRODUCTION

Earlier information on chemical studies in the genus *Clerodendrum* is extensive (Banerjee, 1936; Barton *et al.*, 1961; Bhakuni *et al.*, 1962; Sankarasubramanian and Nair, 1972; Abdul Alim, 1971; Sankarasubramanian, 1973; Nair *et al.*, 1979; Gibbs, 1974; Chaudhary & Roy, 1979; Hegnauer and Kooiman, 1978; Reddy *et al.*, 1988) but with meagre taxonomic interpretation. Hence, presently a qualitative comparison of chemical constituents is undertaken to assess the interrelationships in eleven taxa of *Clerodendrum*.

MATERIALS AND METHODS

Fresh leaves (100 grams) of each species was refluxed with 95% ethanol for 90 minutes. The extract was cooled and filtered. The extract was treated with animal charcoal powder to remove chlorophyll and then was concentrated. The extracts along with two identified compounds (Clerodin = CI-1, Clerodin hemiacetal = CI-2) was chromatographed on a TLC plate, using chloroform-methanol (8.5:1.5) as solvent system. TLC plates were coated with silica gel. Subsequent to drying the plates were activated in the oven at 110°C for 1 hour. The TLC plate was sprayed with concentrated H_2SO_4 and heated in hot-air oven at 95°C. In making the overall assessment of the constituents, Colour of spot (developed) and Rf values are taken into consideration (Table 1, Figs. 1, 2) and not their exact chemical nature. Following Ellison *et al.* (1962) paired affinity values (PAV) for all the species were calculated and represented in polygon graphs (Table 2, Fig. 3).

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RESULTS AND DISCUSSION

The TLC analysis of the leaf extracts in eleven species of *Clerodendrum* show in all, 26 different chemical compounds. However, compounds bearing spot numbers 14, 25 and 26 (Table 1, Fig. 2) appear common to all except in *C. serratum, C. inerme* and *C. calamitosum* respectively (Fig. 2). Spot Nos. 19 and 21 (Table 1, Fig. 2) occur in six species, while No. 18 and 22 only in *C. calamitosum* and *C. indicum* (Fig. 2 Table 1). Spot No. 12 is recorded in *C. philippinum* and *C. viscosum*, while spot 1-11, 13, 15-17, 20, 23 and 24 are unique to individual taxa (Table1).

Table 1. Distribution of Chemical constituents on TLC with colour, Rf. and spot numbers in the*Clerodendrum* L. studied

Spot No.	Colour of the spot	Rf	Species No. I	II	III	IV	V	VI	VII	VIII	IX	X	XI
1.	Ash	0.05	-	-	-	-	-	-	-	-	+	-	-
2.	Brownish red	0.10	-	-	-	-	-	-	-	-	+	-	-
3.	Brown	0.11	-	-	-	-	-	-	-	-	-	-	+
4.	Green	0.16	-	-	-	-	+	-	-	-	-	-	-
5.	Yellow	0.20	-	-	-	-	-	+	-	-	-	-	-
6.	Yellow	0.22	-	-	-	-	-	-	-	-	-	-	+
7.	Reddish Green	0.24	-	-	-	-	-	-	-	-	-	+	-
8.	Brown	0.31	-	-	-	+	-	-	-	-	-	-	-
9.	Light Yellow	0.32	-	-	-	-	+	-	-	-	-	-	-
10.	Light brown	0.35	-	-	-	-	-	-	+	-	-	-	-
11.	Light Yellow	0.35	-	-	-	-	-	-	-	+	-	-	-
12.	Light brown	0.37	-	-	-	-	-	-	-	-	-	-	+
13.	Brownish yellow	0.47	-	-	+	-	-	-	-	-	-	-	-
14.	Violet	0.72	+	+	+	+	+	+	+	+	-	+	+
15.	Ash	0.73	-	-	-	-	-	-	-	-	+	-	-
16.	Yellow	0.76	+	-	-	-	-	-	-	-	-	-	-
17.	Bluish green	0.80	-	-	-	+	-	-	-	-	-	-	-
18.	Pinkish yellow	0.81	-	+	+	-	-	-	-	-	-	-	-
19.	Pink	0.82	-	-	-	-	+	-	+	+	+	+	+
20.	Light yellow	0.84	-	-	-	-	-	+	-	-	-	-	-
21.	Yellow of Light yellow	0.86	+	-	-	-	+	-	+	+	+	+	+
22.	Pinkish violet	0.88	-	+	+	-	-	-	-	-	-	-	-
23.	Pink	0.90	-	-	-	-	-	-	-	-	-	-	+
24.	Greenish yellow	0.91	-	-	-	+	-	-	-	-	-	-	-
25.	Yellow or Light yellow	0.94	+	+	+	-	+	+	+	+	+	+	+
26.	Pinkish violet	0.97	+	-	+	+	+	+	+	+	+	+	+

I = *C. aculeatum*; II = *C. calamitosum*; III = *C. indicum*; IV = *C. inerme*; V = *C. minahassae*; VI = *C. neriifolium*; VII = *C. philippinum*; VIII = *C. phlomidis*; IX = *C. serratum*; X = *C. splendens* and XI = *C. viscosum*. + = Present; - = Absent.

The present information on chemical analysis was statistically evaluated. Based on the paired affinity values (PAV) (Table 2) and cluster analysis (Fig. 4), the taxonomic position is disucssed hereunder.

From the Table 2 and Figs. 3 and 4, it is evident that 11 taxa presently studied fall into the following 4 groups. Group I includes *C. calamitosum* and *C. indicum* with a relationship of 92%. Group II consists of *C. aculeatum*, *C. minahassae*, *C. philippinum*, *C. phlomidis*, *C. serratum*, *C. splendens* and *C. viscosum* with a relationship of 56.5 to 92%. While Group III consists of *C. inerme* and Group IV of *C. neriifolium* with 32% and 50% relationship respectively (Fig. 4 and Table 2).

Table 2. Paired affinity values of *Clerodendrum* L. species studied based on distribution of their chemical constituents

Sl. No.	Name of the species	Species number 1	2	3	4	5	6	7	8	9	10	11
1.	*C. aculeatum*	100	44	55	40	67	60	73	73	50	67	57
2.	*C. calamitosum*		100	80	22	36	44	40	40	18	36	30
3.	*C. indicum*			100	36	46	55	50	50	31	46	40
4.	*C. inerme*				100	33	40	36	36	17	33	29
5.	*C. minahassae*					100	50	77	77	57	71	62
6.	*C. neriifolium*						100	55	55	36	50	43
7.	*C. philippinum*							100	83	62	92	67
8.	*C. philomidis*								100	62	77	67
9.	*C. serratum*									100	58	50
10.	*C. splendens*										100	63
11.	*C. viscosum*											100

Bentham and Hooker (1865) split the genus into five subgenera and two of the present species viz., *C. aculeatum* and *C. inerme* have been kept under subgenus Volkameria. Clarke (1876) divided the genus into two sub-genera Euclerodendron and Siphonanthus, and *C. indicum* was kept in the latter while the rest five under the former. De Candolle (1825) divided the genus into two sections viz. Euclerodendron and Siphonanthus. The former was further subdivided into 4 sub-sections. He placed *C. indicum* in section Siphonanthus and the rest under various sub-sections of section Euclerodendron.

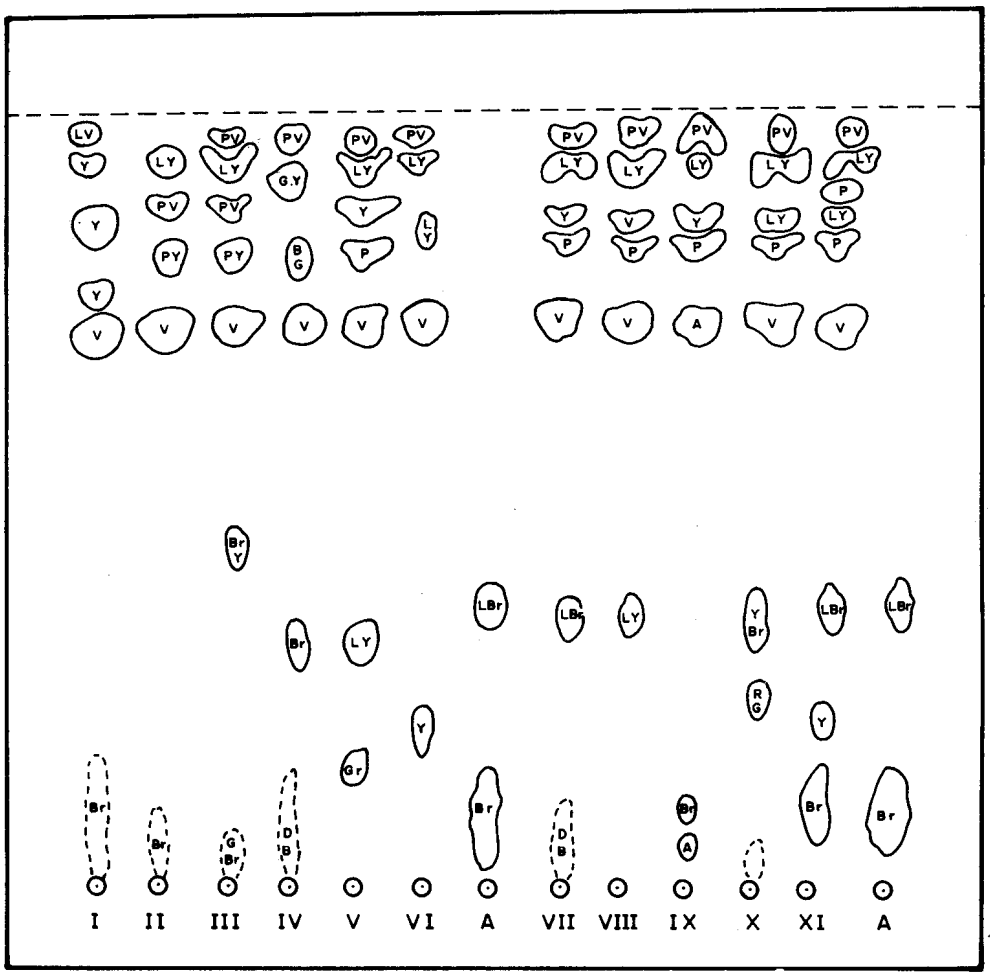

FIG. 1

Fig. 1. Thin layer chromatogram of the leaf extracts of eleven species of *Clerodendrum* spotted along with the mixture of Clerodin (CI-1) and its hemiacetal (CI-2) (A) depicting their colours (for details refer Table 1). I to XI, species number (see Table 1).

br = Brownish red; Br = Brown; Gr = Green; Y = Yellow; RG = Reddish green; LY = Light Yellow; L. Br = Light brown; Br. Y = Brownish yellow; V = Violet; A = Ash; BG = Bluish green; Py = Pinkish yellow; P = Pink; Pv = Pinkish violet; Gy = Greenish yellow; DB = Dark blue; G. Br = Brownish green.

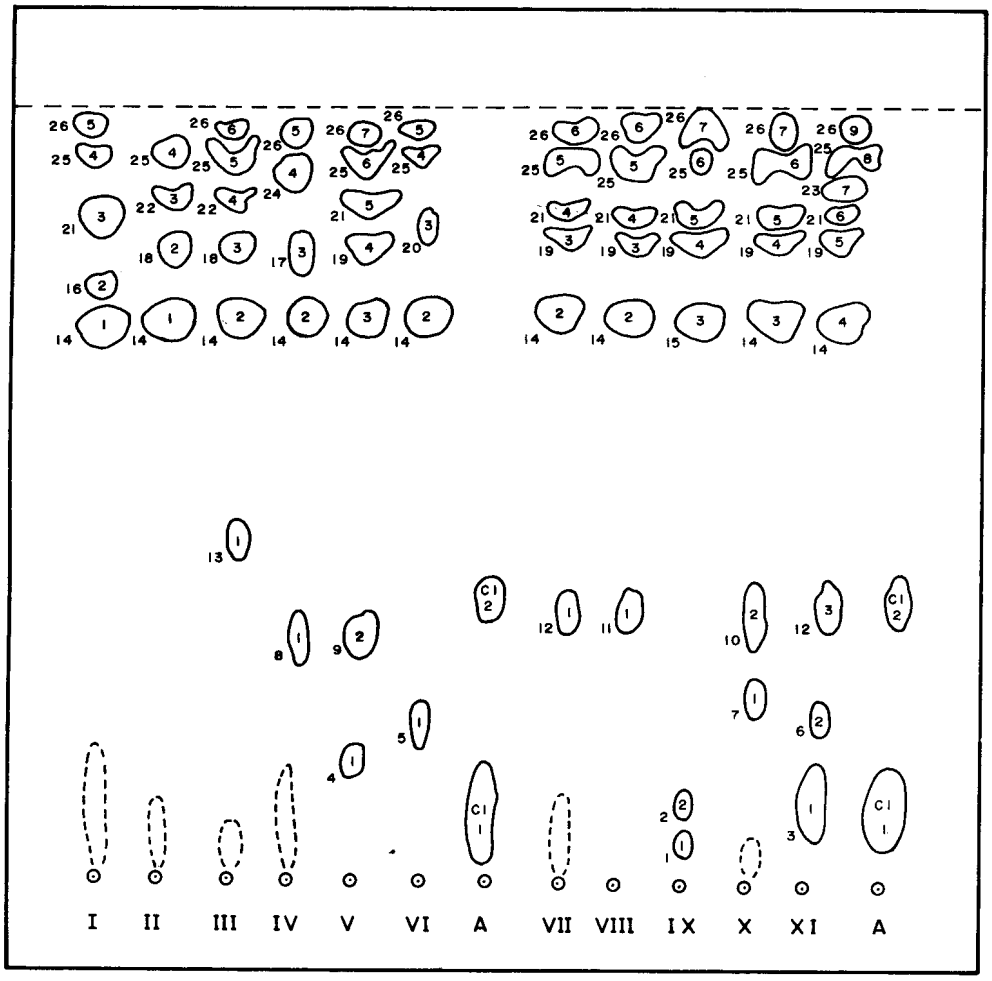

FIG. 2

Fig. 2. Thin layer chromatogram of the leaf extracts of *Clerodendrum* studied depicting the number of compounds (spots) and separated constituents in each species along with A, mixture of CI-1 (Clerodin) and CI-2 (Clerodin hemiacetal) I-XI (species number). The numbers outside the spot indicate compound number from the total pool. The numbers inside the spot represent spot numbers of the individual taxa from the origin to the solvent front.

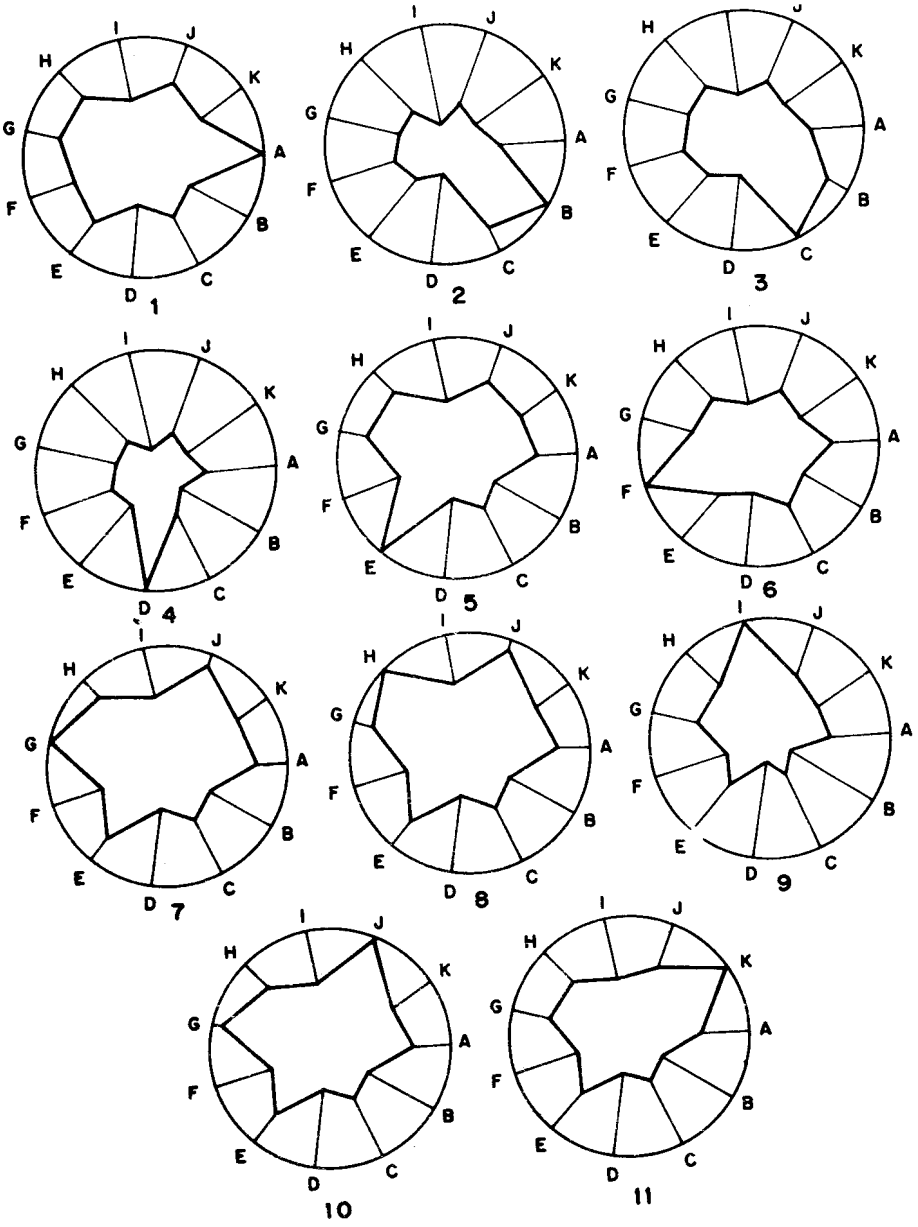

FIG. 3

Fig. 3. 1-11, Polygon patterns in the *Clerodendrum* species studied based on chemical characters.

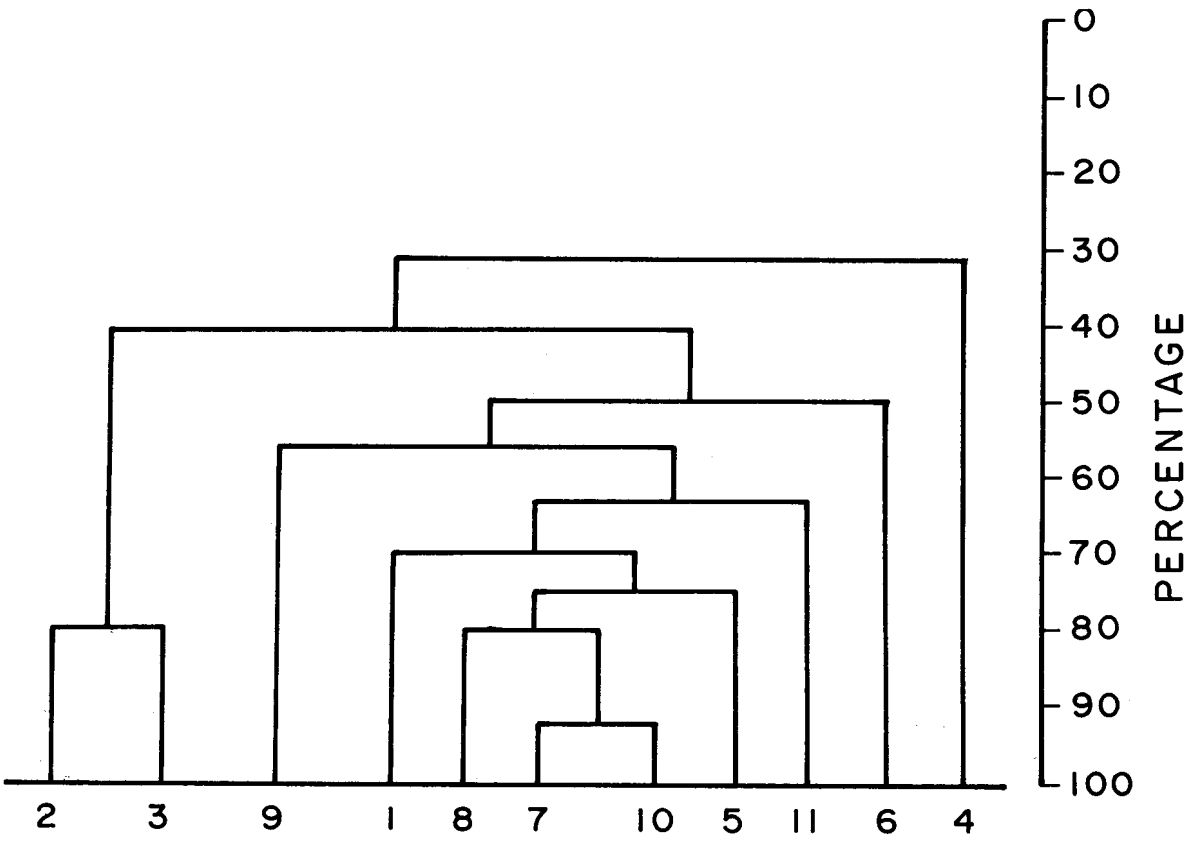

FIG. 5

Fig. 4. Dendrogram representing relationships in the *Clerodendrum* taxa based on chemical characters.

Junell (1934) on embryological grounds divided the genus into five sections. *C. indicum* was kept under his section Siphonanthus and *C. aculeatum* in section Volkameria.

Moldenke (1959) elaborated the genus into five sub-genera, which were further divided into sections and sub-sections. *C. serratum* was kept in section Stacheocymosa (sub-genus Cyclonema) and *C. aculeatum* under sub-genus Volkameria. The rest of the nine species presently studied were placed under various sections and sub-sections of the sub-genus Euclerodendron. For instance *C. inerme*, *C. neriifolium*, *C. phlomidis* and *C. calamitosum* under section Axilliflora; *C. indicum* and *C. minahassae* in section Siphonanthus; *C. phillippinum* and *C. viscosum* in

sub-section Paniculata (section Microcalyx) and *C. splendens* under sub-section Acuminata (section Oxycalyx).

Chemotaxonomically, correlation between morphological and chemical characters in splitting the genus with reference to the alignment of *C. indicum* under Siphonanthus and the rest under Euclerodendron (Clarke, 1876) except *C. viscosum* as a third cluster was reported (Reddy *et al.*, 1988). Chaudhary and Roy (1979) revealed biochemical affinities among some taxa of Verbenaceae which also included 6 species of *Clerodendrum*. However, they have not discussed interspecific relationships in the latter based on their chemical data.

In the light of earlier investigations and also various subgeneric treatments in the genus, the conclusions drawn in the present study based on TLC analysis is distinct and agrees partially in certain circles of affinities proposed by Moldenke (1959). For instance, the treatment of *C. phlomidis*, *C. minahassae*, *C. philippinum*, *C. viscosum* and *C. splendens* under sub-genus Euclerodendron of the latter is in agreement with their grouping in the present group II (Table 2 & 3 Fig. 4). However, a study based on chemical parameters of eleven species of such a large genus (564 spp. Cf, Moldenke, 1971) is only an attempt to understand the relationships amongst the Indian representatives of the genus *Clerodendrum*.

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