Chemical Systematics of the Verbenaceae

CHAPTER 3

CHEMICAL SYSTEMATICS OF THE VERBENACEAE

3:1 Introduction

The Verbenaceae, a large family of 75 genera and 2500 species (Jafri and Ghafoor, 1974) is chiefly tropical and subtropical in distribution, although a few species extend into temperate regions of the new world.

Economically the family is important for teak (*Tectona grandis*), which is highly prized for its wood. A number of genera contain important ornamentals, notable among them are, *Clerodendrum*, *Duranta*, *Holmskioldia*, *Lantana*, *Petrea*, *Verbena* and *Vitex*.

In India the family is widely distributed upto an altitude of 4,000 feet (Clarke, 1885). However, most of its members are confined to central India. *Clerodendrum*, *Verbena* etc. are introduced from temperate regions of the world. *Avicennia* is confined to coastal regions of India.

Most of the Verbenaceae members are predominantly scandent or straggling shrubs, rarely herbs (*Verbena*), armed shrubs with axillary thorns (*Duranta*) or deciduous trees (*Tectona*) or Halophytes as in case of *Avicennia*. The leaves are simple, opposite deccusate, exstipulate; except in Vitex which show compound leaves. The presence of a pair of glands at the base of lamina is a characteristic feature of Citharexylum. Inflorescence is variable, usually determinate and one or other modification of dichosial cyme. Racemes with flowers arranged in dense or lax spike or corymbs are seen in Verbena, whereas the inflorescence is paniculate in Gmelina philippensis and Tectona grandis.

Flowers are typically bisexual, hypogynous and pentamerous. The calyx shows considerable variation in number of lobes or teeth. It is usually cupular having 5 lobes or teeth and persistent with the fruit. In Phyla the calyx is generally 2-4-dented, whereas 5 lobes and 5 scales in the throat are seen in Petrea. Calyx is membranous in Lantana. In Tectona the calyx is campanulate and 5-7-lobed. In other members like Vitex, Clerodendrum, and Holmskioldia the calyx is perfectly 5-toothed and slightly enlarged in fruit. The gamopetalous corolla is cylindrical or infundibuliform with as many lobes as the calyx. The lobes are mostly unequal but at times 5 equal or subequal lobes are present in genera like Gmelina, Holmskioldia and Premna. Hypocrateriform corolla with 5-7 subequal lobes is present in Tectona.

In the family, the stamens are invariably 4 and didynamous, inserted on the corolla tube and alternating with corolla lobes. The fifth stamen is generally represented by a staminode. Tectona is the only exceptional taxon showing 5 fertile stamens. The anthers are dithecous and mostly dehisce longitudinally. In Stachytarpheta ,of the 4 stamens only the 2 anterior ones are fertile and possess divergent anther lobes. Anther lobes sagittate in Duranta. Normally the stamens are included but exceptionally exerted in Tectona, Clerodendrum and Holmskioldia.

The gynoecium is usually bicarpellary; some times it is 4-carpelled as in Duranta. Ovary is superior, 4-lobed, 2-4-loculed with one ovule in each apparent cell on axile placenta. Ovules usually solitary, mostly erect, anatropous with micropyle facing downward. The capitate stigma is usually bifid but with unequal 4 lobes as in Duranta. A curved and acutely bifid stigma occurs in Clerodendrum. Fruit is generally a drupe with as many pyrenes as ovules in ovary or nutlets (Verbena). The fruit is a 2-4-valved capsule in Avicennia. The seeds are with a straight embryo and exalbuminous. However, fleshy endosperm is present in Avicennia, Stilbe, Chloanthes.

3:2 <u>Taxonomy</u>

The family Verbenaceae was included under subclass Gamopetalae, series Bicarpellatae and order Lamiales by Bentham and Hooker (1876). Engler and Prantl (1897) grouped it close to Lamiaceae under suborder Verbenineae of the order Tubiflorae. This scheme was later followed by Engler and Diels (1936). In the latest edition of Engler's syllabus der pflanzenfamilien (ed. Melchior, 1964), the earlier classification is maintained. Order Tubiflorae encompasses Verbenaceae under "Unterreiche" Verbenineae.

Hallier (1912) recognised 4 primary sub divisions for the various families of the Dicotyledons. The IVth sub division 'ochnigenes' comprised, amongst other families, Verbenaceae under the order Tubiflorae. Bessey (1915) on the basis of corolla zygomorphy and gynoecium character put Verbenaceae and Lamiaceae in a distinct order, the Lamiales. The order Lamiales according to him consists of Verbenaceae, Myoporaceae, Phrymaceae and Lamiaceae (Labiatae). Rendle (1925) and Wettstein (1935) followed the concept of Engler and Prantl (1897) regarding the placement of the Verbenaceae.

Hutchinson (1926) treated Verbenaceae and its allied family Phrymaceae under Lamiales, but latter (1948) segregated them to form order Verbenales, a group that represented the climax among the woody dicots. In Verbenaceae, if one looks for a primitive type, the choice falls on the genus Tectona, a hard wooded timber tree in which 5 stamens have been retained. In all other genera, the 5th stamen is absent or rarely represented by a staminode. Therefore, Hutchinson considered this group as unrelated to Labiatae and derived it from rubiaceous stock. Thus Verbenaceae was regarded as parallel to and not as a close relative of Lamiales. Hence, it topped the linear sequence of Lignosae while the Lamiaceae topped the Herbaceae. In the latest edition of Families of Flowering Plants (1973) he recognised as many as 5 families in order Verbenales namely Ehretiaceae, Verbenaceae, Stilbaceae, Dichrostyladaceae (Chloanthaceae) and Phrymataceae.

Skottsberg (1940) grouped Verbenaceae under Tubiflorae, while Gundersen (1950) erected a new order Boraginales wherein the Verbenaceae was grouped along with families such as Boraginaceae, Myoporaceae, Labiatae and Callitrichaceae. Lawrence (1951) adopted the system proposed by Engler and Diels (1936) and treated Verbenaceae under Tubiflorae. Benson (1957) treated this family under separate order Lamiales. This was also supported by Soo (1961). The systems proposed by Cronquist (1968) and Takhtajan (1969,1980) are more or less alike in broad treatment and placed Verbenaceae under Lamiales in sub class - Asteridae of the Dicotyledons (Magnoliatae). The families included under Lamiales are Labiatae, Boraginaceae, Phrymaceae, and Callitrichaceae. Cronquist (1968) visualises a close relationship of the Verbenaceae to the Labiatae. To quote him "Indeed it is difficult to know where to draw the line between them, and a case might be made for treating the present Verbenaceae and Labiatae as the more primitive and more advanced segments of a single family".

The family Verbenaceae is classified into 8 tribes by Bentham and Hooker (1876) on the basis of inflorescence character, number of locules in the ovary and nature of ovule. The various tribes recognised are Phrymeae, Stilbeae, Chloantheae, Verbeneae, Viticeae, Caryopterideae, Symphoremeae and Avicenniae (Table 11).

Briquet (1897) based the infrafamilial or tribal classification on cohesion of ovary and number of ovules in each locule. The family is sub-divided into 8 subfamilies and 15 tribes. This treatment is followed in the latest edition of Engler's syllabus (ed. Melchior, 1964). Hutchinson (1969) elevated Bentham and Hooker's tribes Stilbeae, Chloantheae and Phrymeae to separate familial rank. Thus the family Verbenaceae as now circumscribed

Table-11. VERBENACEAE, taxa screened and their arrangement in various schemes of classification

BENTHAM and HOOKER (1876)		BRIQUET (1897)			HUTCHINSON (1926), (1969)		MOLDENKE (1959)	
TRIBE I	PHRYMEAE				FAMILY	PHRYMACEAE	FAMILY	PHRYMACEAE
II	STILBEAE	Div.B	SUBFAMILY	STILBOIDEAE		STILBACEAE		STILBACEAE
III	CHLOANTHEAE	Div.A	SUBFAMILY	CHLOANTHOIDEAE		CHLOANTHACEAE	SUBFAMILY	CHLOANTHOIDEAE
IV	VERBENEAE		SUBFAMILY	VERBENOIDEAE	TRIBE	VERBENEAE	SUBFAMILY	VERBENOIDEAE
	Lantana, Phyla, Stachytarpheta		TRIBE	LANTANEAE				
	Verbena		TRIBE	VERBENEAE				
	Petrea		TRIBE	PETRACEAE				
	Citharexylum, Duranta		TRIBE	CITHAREXYLEAE				
V	VITICEAE							
	Tectona		SUBFAMILY VITICOIDEAE	TRIBE TECTONEAE				
	Premna, Gmelina, Vitex		SUBFAMILY VITICOIDEAE	TRIBE VITICEAE	TRIBE	VITICEAE	SUBFAMILY	VITICOIDEAE
	Clerodendrum, Holmskioldia		SUBFAMILY VITICOIDEAE	TRIBE CLERODENDREAE				
VI	CARYOPTERIDEAE	Div.B	SUBFAMILY	CARYOPTERIDOIDEAE	TRIBE	CARYOPTERIDEAE	SUBFAMILY	CARYOPTERIDOIDEAE
VII	SYMPHOREMEAE	Div.C	SUBFAMILY	SYMPHOREMATOIDEAE	TRIBE	SYMPHOREMEAE	FAMILY	SYMPHOREMACEAE
VIII	AVICENNIEAE	Div.C	SUBFAMILY	AVICENNIOIDEAE	TRIBE	AVICENNIEAE	FAMILY	AVICENNIACEAE
	Avicennia	Div.B	SUBFAMILY	NYCTANTHOIDEAE				

includes only tribes Verbeneae, Viticeae, Caryopterideae, Symphoremeae and Avicenniae.

Many controvercies exist regarding the position of the genus Avicennia. Endlicher (1838) in his classification of Verbenaceae, appended Avicennia as an unnumbered tribe Avicenniae. Latter in 1843, he proposed separate monogeneric family Avicenniaceae. Schauer (1847), Bentham and Hooker (1876) however, maintained Avicennia as constituting a separate and last tribe of the Verbenaceae.

Briquet (1897) and Melchior (1964) gave the genus Avicennia, the status of a sub-family Avicennioideae Briq. Moldenke and Moldenke (1946) and Moldenke (1959) accepted Avicenniaceae of Endlicher as a family distinct from Verbenaceae under the Lamiales, on the basis of wood anatomy, articulate branches, free central placentation and pendent orthotropous ovule. Moldenke (1959) also recognized Phrymaceae, Stilbaceae and Symphoremaceae as separate families.

3:3 Earlier chemical work

The known chemical data (Table 12) of various Verbenaceae taxa include flavonoids, lignans, quinones, alkaloids and steroidal saponins. The phenolic chemistry is

Table: 12.

EARLIER CHEMICAL WORK

Plant name	Part	Compound reported	Author
FLAVONOIDS			
Callicarpa macrophylla	Lvs.	5,4'-di-OH,3,7,3'-tri-Ome flavone	Chaudhary, et al. (1978).
Citharexylum subserratum		7,3'-diOme-5,6,4'-tri-OH flavone	Subramanian, et al. (1976).
Clerodendrum indicum	Lvs.	Hispidulin, Scutellarein	Subramanian. S.S. and A.G.R.Nair (1973).
C. infortunatum	Lvs.	Glucuronoides of Hispidulin, Scutellarein but no free aglycones.	Subramanian, S.S. and A.G.R.Nair (1973).
	Fls.	Apigenin, Acacetin	Sinha et al. (1981).
C. inerme	Lvs.	7-o glucuronide of apigenin and scutellarein	Subramanian, S.S.. A.G.R.Nair and T.N.C.Vedantham (1973).
C. nerifolium	Lvs.	Scutellarein-4'-arabinoside, scutellarein Acacetin, Hispidulin.	Subramanian, S.S. and A.G.R.Nair (1972).
C. phlomidis	Lvs.	Scutellarein and Pectolinoringenin	Subramanian, S.S. and A.G.R.Nair (1972).
C. serratum	Lvs.	Acacetin-7-glucuso-glucuronide	Okigawa, et al. (1971).
		6-OH Luteolin, Caffeic and Ferulic acid.	Nair, A.G.R., T.N.C. Vedantham and Kannabiran, B. (1979).
C. trichotomum	Lvs.	Acacetin-7-β-glucoso (1-2)β-glucuronide	Okigawa, et al. (1971).
Duranta repens	Lvs.	Malvidin	Forsynth, W.G.C. and N.W.Simmonds (1954).
		Scutellarein, Pectolinoringenin	Subramanian, S.S. and A.G.R.Nair (1972).
Gmelina arborea	Lvs.	Luteolin	Rao, et al. (1967).
		Apigenin, Luteolin, Quercetin..	Rao, D.V. and E.V Rao (1970).
		Quercetagetin, Apigenin, Luteolin, Kaempferol, Kaemp-3-rutinoside, Api-3-rutinoside, Api-7-glucuronide, Luteo-7-glucuronide.	Nair, A.G.R. and S.S.Subramanian (1975).
G. asiatica	Lvs/ Fls/ Frs.	Luteolin	
		Apigenin, Luteolin, Quercetin.	
		Quercetagetin, Apigenin, Luteolin, Kaempferol, Kaemp-3-rutinoside, Api-3-rutinoside, Api-7-glucuronide, Luteo-7-glucuronide, Carotenoids.	

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Table :-12. Contd.

Plant name	Part	Compound reported	Author
Lippia nodiflora	Lvs.	Nodiflorin, A and B	Joshi, B.C. and D.S. Bakuni (1959).
	L s.	Nodiflorin, A and B	Joshi, B.C. (1966).
		Nodifloretin-A (5,6,7,4'-tetra-OH,3' Ome flavone)	Barua, et al. (1969)
		Lippiflorin A,B; 6-OH-Luteolin 6-Ome luteolin (nepatin)	Nair, et al. (1973).
Stachytarpheta indica	Lvs.	Scutellarein	Subramanian, S.S., A.G.R.Nair and T.N.C. Vedantham (1974).
		Choline, chlorogenic acid, 6-OH luteolin, apigenin, glucuronide.	Duret, S.H., Jacquenin and R.R. Paris (1976).
Vitex sp. (V. agnus-castus, V. trifolia, V. negundo	Lvs. or Wood	Casticin (3',5'-di-OH,3,4'-6,7,tetra-Ome flavone), Luteolin 7-glucoside, Orientin and homoorientin.	Hansel, et al. (1965).
V. negundo	Lvs.	5-OH,3,6,7,3',4'-penta-Ome-flavone.	Banerjee, et al. (1969).
		p-OH benzoic acid, 3,4 di-OH benzoic acid	Joshi, V., J.R. Merchant and U. Nadkarni (1974).
	St. bark	Vanillic, p-OH benzoic acid, Luteolin.	Kodanda Rao, E.V.Rao and D.V.Rao(1977)
		Leucoanthocyanin	Subramanian, P.M. and G.S.Misra (1978)
		3,6,7,3',4'-penta Ome 5-o glucopyranosyl rhamnoside, vitexin, 4'-Ome myricetin-3-o galactopyranoside.	Misra, G.S. and P.M. Subramanian (198[illegible]).
V. lucens	Ht.wood	Vitexin	Perkin, A.G. (1900). Rao, C.B. and V.Venkateswarulu (1961).
V. peduncularis		Vitexin	Rao, C.B. and V.Venkateswarulu (1962).
V. altissima / V. leucoxylon		Vitexin	Rao, P.S. (1965).

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Table : 12. Contd.

Plant name	Part	Compound reported	Author
LIGNANS			
Gmelina arborea	Ht. wood	Gmelinol, n-octanosonol.	Joshi, K.C. and L.B. Singh (1970).
		Arboreol	Govindachari, T.R., P.C.Parthasarthy and H.K.Desai (1972).
		Gmelinol	Desai *et al.* (1973).
		Arboreol, isoarboreol, Gmelenone.	Anjaneyulu, *et al.* (1975).
		6' bromoisoarboreol, Gummadiol.	Anjaneyulu, *et al.* (1977).
Quinones			
Avicennia tomentosa		Lapachol	Bournot, K. (1914).
Tectona grandis	Root	Lapachol, Dehydrotectol, Tectoquinone (Pet. ether ext.)	Joshi, K.C., P. Singh and R.T. Pardasani (1977).
		β-lapachone, Dehydro-α-lapachone and Tectol (ethanol ext.)	
	Lvs.	1,4,5,8-tetra-Hydroxy-2-iscpentadienyl anthraquinone	Agarwal, S.C., R.G.Sarangarajan and T.R. Sheshadri (1965).
		Quinone A_6, A_7 (o-quinoid structure belonging to naphthalene series).	Sandermann, W. and M.H. Simatupang (1965).
	Stem tissue culture	2,5,di OH-1-Ome-3-methyl anthraquinone	Dhruwa, *et al.* (1973).
ALKALOIDS			
Clerodendrum inerme	Lvs.	A bitter principle, alkaloidal in nature.	Abdul Alim (1971).
C. serratum	Root Bark	Alkaloidal matter absent	Banerjee, *et al.* (1969).
Duranta repens	Fruit	Pyridine alkaloid	Yousef, *et al.* (1973).
Gmelina arborea	Lvs.	Alkaloids present	Bhattacharjee, A.K. & A.K.Das (1969).
Verbena officinalis	Aerial part	Alkaloids present	Ismailov, N.M. (1967).
Vitex negundo	Lvs.	Nishindine alkaloid	Basu and Singh (1944). Joshi, K.C., J.R.Merchant & U.Nadkarny (1974)

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Table 12. Contd.

Plant name	Part	Compound reported	Author
STEROIDAL SAPONINS			
Avicennia officinalis	Lvs/ Stem	β-Sitosterol, lupeol, lupenone	Subramanian, S.S. and T.N.C.Vedantham (1974).
Callicarpa arborea	Lvs.	β-Sitosterol	Sen, M. and U. Sarkar (1978).
Clerodendron infortunatum	Lvs.	Clerodin	Banerjee, H.N. (1937).
			Chaudhary, D.N. and P.C. Dutt (1951).
	Lvs/Root	β-Sitosterol	Sen, A.B. and S.P. Singh (1964).
	Root	β-Sitosterol	Barua, A.K., P. Sanyal and P.Chakrabarti (1967).
	Lvs.	Clerodin	Subramanian, S.S. and A.G.R. Nair (1973).
Clerodendrum paniculatum	Root	β-amyrin, B-Sitosterol	Joshi, K.C., P.Singh and Alka Mehra (1979).
C. phlomidis	Stem	β-Sitosterol	Gupta, R.K., Suresh Chandra and V. Mahadevan (1967).
C. serratum	Root Bark	γ-Sitosterol	Banerjee, *et al.* (1969).
Duranta repens	Fruit	β-Sitosterol	Kapil, R.S. (1960).
Gmelina arborea	Lvs.	β-Sitosterol	Rao, D.V. and ..V. Rao (1970).
	Ht.wood	β-Sitosterol	Joshi, K.C., L. Prakash and L.S.Singh (1971).
			Desai, *et al.* (1973).
Lantana camara		α-amyrin, β-Sitosterol	Ahmed, *et al.* (1972).
	Seeds	β-Sitosterol	Avadhoot, *et al.* (1978).
Stachytarpheta indica	Lvs.	Stigmasterol	Nair, A.G.R. and T.N.C.Vedantham (1974).
Tectona grandis	Bark	Saponin present	Bhattacharjee, A.K. and A.K.Das (1969).
Vitex negundo		β-sitosterol	Joshi, *et al.* (1974).
	Bark	β-Sitosterol	Kodanda Rao, *et al.* (1977).
V. trifolium	Lvs.	β-Sitosterol	Vedantham, T.N.C. and S.S.Subramanian (1976).

Contd...

Table : 12. Contd.

Plant Name	Part	Compound reported	Author
IRIDOIDS			
Citharexylum sp.		Lamiide. Durantoside	Milz, S. and H. Rimpler (1979).
Duranta plumerei	Lvs/St/	Lamiide, Durantoside-1,2,-3.	Rimpler, H. and T. Helmut (1974).
	Fruit	Lamiide, Durantoside	Rao, B.Ch., T.N.Rao and E.K.S. Vijaykumar (1978).
Lantana camara	Lvs/St.	Theveside (Iridoid glucoside)	Ford, C.W. and M.R. Bendall (1980).
Premna latifolia		7-deoxyloganic acid, geniposidic acid	Rao, B.Ch., E.K.S. Vijaykumar and K.V. Vijayalaxmi (1981).
Stachytarpheta indica	Aerial parts	Ipolamide	Tantisewie, B. and Ottosticher (1975).
S. mutabilis	"	Ipolamide	Garnier, J. (1979).
Verbena pulchella		Pulchelloside II.	Milz, S. and H. Rimpler (1978)
Verbena sp.		Verbenaline, Hastatoside, Griselinoside, Lamiide, Pulchelloside I & II	Milz, S. and H. Rimpler (1979).
V. sp.		Cornin, Hastatoside, Griselinoside, Lamiide, Pulchelloside I and II.	Damtoft, et al. (1979).
V. hastata / *V. officinalis*		Verbenalin, Hastatoside	Rimpler, H. and B. Schafer (1979).
Vitex megapotamica		Agnoside	Rimpler, H. (1972).

not much explored. There are random reports of flavonoids and other phenols from individual plants. These isolated reports are indicative of our meagre knowledge about the phenolic chemistry of this family. Not a single plant has been worked out thoroughly and systematically for phenolics. In the present work, 35 plants belonging to 14 genera have been systematically screened for the various phenolics. The leaf phenolics taken into consideration here include the flavones, flavonols, glycoflavones, leucoanthocyanins, quinones and phenolic acids. Therefore, in the present report an attempt is made to understand the interrelationships and phylogeny of Verbenaceae using reliable chemical markers. Data obtained from other disciplines of Botany are also properly weighed along with the chemical data for a proper appraisal of the present classifications of the family and to discern the evolutionary trends within this large taxon.

3:4 Materials and Methods

Most of the screening was done with fresh materials collected from gardens around Baroda. Supply of materials also came from the Botanical gardens at Waghai (Dangs), Aurangabad, Pune and Trivandrum. *Premna resinosa* was made available by Dr. K.S.S. Rao from the Kutch region. *Avicennia* was sent from Bulsar by Dr. P.G. More. The plant materials were properly dried, preserved and deposited in the Herbarium, Botany Department, The M.S. University of Baroda, Baroda (BARO).

The procedures followed in extraction, isolation, and identification of the various phenolics are the same as those described in Chapter 2.

3:5 Results

The distribution of flavonoid compounds (such as Flavones, Flavonols, Glycoflavones, Leucoanthocyanins), Phenolic acids, Alkaloids, Iridoids, Saponins, Tannins and Quinones from leaves of Verbenaceae members is presented in Tables 13, 14 and 15.

In the present work 35 members belonging to 3 tribes and 14 genera were studied using chemical markers like flavonoids and phenolic acids. 31 members contain one or the other type of flavonoids. 4 members are devoid of any flavonoid. A total of 30 o-glycosides and c-glycosides are detected, of which 19 are of flavones, 7 of flavonols and 4 c-glycosides.

Flavones are found singly in 19 members. Flavones and Flavonols occur together in 8 members. Flavones along with c-glycosides occur in 3 members. While Flavones/Flavonols/

Table .: 13. DISTRI.3UTION OF FLAVONOIDS IN THE VERBENACEAE.

Sr. No.	NAME OF THE PLANT	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30
	TRIBE VERBENEAE																														
1.	Citharexylum subserratum Sw.	+		+				+																			+				
2.	Duranta repen, Linn.	+	+		+								+																		
3.	Lantana camara var. acu'eata (L.) Mo denke	+	+															+													
4.	var. hybrida Moldenke		+		+										+																
5.	var. mista Bailey		+			+																									
6.	var. nivea Bailey	+	+			+																									
7.	L. salvifolia Jacq.							+		+					+								+	+							
8.	Petrea volubilis Jacq.																	+													
9.	Phyla nodiflora (Linn.) Greene														+		+	+													
10.	Stachytarpheta indi a Vahl				+	+												+		+		+									
11.	S. mutabilis Vahl		+							+											+		+								
12.	Verbena venosa Gill & Hook.			+																										+	
13.	V. bipinnatifida Nu.t.							+																+		+					
14.	V. tenuisecta Bric.	+	+																												
	TRIBE ~ ~ICEAE																														
15.	Clerodendrum aculeatum (Linn.) Griseb.																														
16.	C. fragrans Vent.		+																												
17.	C. indicum (Linn.) Kuntze							+						+														+			
18.	C. inerme (Linn.) Gaertn.		+		+																										
19.	C. infortunatum Gaertn.																														
20.	C. macrosiphon Hook. f.		+																												
21.	C. ultiflorum (Burm.f.) O. Ktze.				+													+													
22.	C. paniculatum Linn.		+																												
23.	Clerodendron siphonanthus R. Br.						+		+																						

Contd..

Table : 13. Contd..

Sr. No.	NAME OF THE PLANT	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30
24.	Clerodendrum splendens Don																	+	+												
25.	C. speciosum Hart.	+		+		+					+																				
26.	C. thomsoniae Balf.																														
27.	Gmelina arborea Roxb.	+	+									+												+							
28.	G. asiatica Linn.		+									+									+		+			+					
29.	G. philippensis Cha..		+																												
30.	Holmskioldia sangui[illegible]ea Retz.		+																												
31.	Premna resinosa (Hochst.) Schau.		+																												
32.	Tectona grandis Linn. f.				+							+	+														+				
33.	Vitex negundo L.				+								+			+															
34.	V. altissima Linn.																														
	TRIBE [illegible]NNIEAE																														
35.	Avicennia marina (Forsk.) Vierh. Syn. A. alba auct. non Blume												+								+	+							+		

Flavones -

1. Apigenin
2. 4'-Ome Apigenin
3. 7-Ome Apigenin
4. Scutellarein
5. 4'-Ome Scutellarein
6. 5-Ome "
7. 6-Ome "
8. 8-Ome "
9. 6,4'-di Ome Scutellarein
10. 7,4'-di Ome Scutellarein
11. Luteolin
12. 3'-Ome Luteolin
13. 4'-Ome "
14. 6-Ome "
15. 3',4'-di Ome Luteolin
16. 7,4'-di Ome Luteolin
17. 6-OH-Luteolin
18. 8-OH "
19. 6-OH,3',4'-diOme Luteolin

Flavonols

20. Kaempferol
21. 6-OH-4'-Ome Kaempferol
22. Quercetin
23. 3'-Ome Quercetin
24. 4'-Ome "
25. 3',4'-di Ome "
26. 6-OH,Quercetin (Quercetagetin)

Glycoflavones

27. Vitexin
28. 7-Ome vitexin
29. G.F.based on 7-Ome Apigenin
30. 6-Ome Orientin.

Glycoflavones together are present in only one member belonging to tribe Avicennieae.

14 members belonging to tribe Verbeneae were analysed for flavones/-ols. The results clearly indicate dominance of flavones. All the 14 members show presence of one or the other type of flavones. 8 members show exclusively flavone type of compounds. They are Duranta repens, Lantana camara var. aculeata, Phyla nodiflora and Verbena tenuisecta. The other 5 members possess both flavones and flavonols, as encountered in case of Citharexylum subserratum, Lantana salvifolia (= L.indica), Stachytarpheta indica, Stachytarpheta mutabilis and Verbena bipinnatifida. Verbena venosa is the only member from this tribe where flavones along with glycoflavones are detected.

The different flavones identified in this tribe are mostly methylated or hydroxylated derivatives of Apigenin and Luteolin. Apigenin itself is detected in 6 members belonging to this tribe. They are Citharexylum subserratum, Duranta repens, Lantana camara var. aculeata, var. nivea, Phyla nodiflora and Verbena tenuisecta. The aglycone part of apigenin-glucuronide as reported earlier from Stachytarpheta indica (Duret et al., 1976) has been found to be absent. However, Stachytarpheta mutabilis shows presence of 4'-methoxy apigenin (Acacetin). Acacetin is also detected in

Duranta repens, *Lantana camara* var. *aculeata*, var. *hybrida*, var. *mista*, var. *nivea*, *Stachytarpheta mutabilis* and *Verbena tenuisecta*.

7-methoxy Apigenin, the other monomethyl derivative of apigenin is found in only two representatives of this tribe - *Citharexylum subserratum* and *Verbena venosa*.

Scutellarein (6-OH, apigenin) as reported formerly by Subramanian and Nair (1972) from *Duranta repens* and by Subramanian, *et al*. (1974) from *Stachytarpheta indica* is found to co-occur with 4'-Ome scutellarein in leaf extract. *Lantana camara* var. *hybrida* also shows presence of scutellarein. The other two varieties viz. var. *mista* and var. *nivea* show presence of 4'-Ome scutellarein. The occurrence of 7,3'-di-Ome, 5,6,4' tri-OH flavone (7,3'-di-Ome Scutellarein) as reported from *Citharexylum subserratum* Subramanian *et al*., (1976) could not be detected during the present flavonoid screening. However, it shows presence of another scutellarein derivative - identified as 6-Ome scutellarein (Hispidulin). 6-Ome scutellarein is also detected in taxa like *Lantana salvifolia* and is the only flavone in *Verbena bipinnatifida*.

From the dimethoxylated derivatives of scutellarein, 6,4'-di-Ome scutellarein (Pectolinaringenin) was detected in

Lantana salvifolia and Stachytarpheta mutabilis. But, found to be absent in Duranta repens contrary to an earlier report by Subramanian and Nair (1972).

3'-Ome Luteolin is found only in Duranta repens. The other monomethyl derivative of luteolin such as 6-Ome luteolin (Nepatin) and 6-OH luteolin reported by Nair et al. (1973) from Phyla nodiflora are also detected from the same plant. 6-Ome Luteolin is also found to be present in taxa like Lantana salvifolia and Lantana camara var. hybrida. In addition to these two reported compounds from Phyla nodiflora, 7,4'-di-Ome luteolin is also detected from the same plant.

The earlier report of occurrence of 6-OH luteolin from Stachytarpheta indica Duret, et al. (1976), is confirmed in the present chemical findings. 6-OH,3',4'-di-Ome Luteolin is also detected from the same plant.

The 6 flavonol compounds detected in this tribe include 6-OH quercetin (quercetagetin) from Citharexylum subserratum; Kaempferol from Stachytarpheta mutabilis, 6-OH,4'-Ome Kaempferol from S. indica, Quercetin from Lantana salvifolia and Stachytarpheta mutabilis, 3'-Ome quercetin from Lantana salvifolia and Verbena bipinnatifida and 4'-Ome quercetin only from Verbena bipinnatifida.

20 members of the second tribe Viticeae have been screened. 16 of them show presence of flavones/flavonols. Of these 16, 11 plants are with flavones only, 3 with both flavones and flavonols but no glycoflavone and 2 with flavone and glycoflavone. Thus in this tribe also, flavones are detected in majority of the members. The incidence of flavones, however, is comparatively more in tribe Verbeneae. Flavonols are of relatively rare occurrence.

The various taxa which possess only flavones are Clerodendrum fragrans, C. inerme, C. macrosiphon, C. multiflorum (= C. phlomidis), C. paniculatum, C. siphonanthus, C. splendens, C. speciosum, Gmelina philippensis, Premna resinosa and Vitex negundo. Gmelina arborea, G. asiatica and Tectona grandis are the only members of tribe which possess both flavones and flavonols. Clerodendrum aculeatum, C. infortunatum, C. thomsoniae and Vitex altissima are devoid of any flavonoid compounds.

In all 15 flavones have been identified from Viticeae members. Apigenin is encountered from two members - C. speciosum and Gmelina arborea. The presence of apigenin and luteolin as reported by Rao and Rao (1970) from G. arborea is confirmed. But, instead of quercetin its another methylated derivative (3'-Ome Quercetin) is detected along with acacetin. The other species G. asiatica shows presence of acacetin, Kaempferol, Quercetin and its dimethylether ($3',4'$-diOme quercetin) which is in agreement with the earlier report of Nair and Subramanian (1975) except with one additional flavone - the luteolin, identified during the present study. The third species, G. philippensis shows presence of acacetin only.

Out of the 12 Clerodendrum species screened, 9 contain flavones. The flavonols are completely absent. The 3 species totally lack flavonoid type of compounds. Among the flavones and their derivatives, acacetin has been identified from Clerodendrum fragrans, C. inerme, C. macrosiphon, C. paniculatum. 7-Ome Apigenin, $4'$-Ome scutellarein and $7,4'$-di-Ome scutellarein are detected from C. speciosum. The other Viticeae members with acacetin are Holmskioldia sanguinea and Premna resinosa.

Scutellarein, reported by Subramanian and Nair (1972) from Clerodendrum multiflorum and by Subramanian et al. (1973) from C. inerme is also found to be present, confirming the earlier reports. Scutellarein is also present in Tectona grandis and Vitex negundo.

5-Ome scutellarein is detected only from Clerodendrum siphonanthus. Hispidulin (6-Ome scutellarein) reported by Subramanian and Nair (1973) from C. indicum has also been noted.

Luteolin co-occurs with its 3'-Ome derivative in case of Tectona grandis. Whereas, 3'-Ome luteolin and 3',4'-di-Ome luteolin are detected from Vitex negundo. 6-OH Quercetin (Quercetagetin) is detected only from Tectona grandis. Vitex altissima is devoid of flavonoid compounds.

The genus Avicennia of tribe Avicennieae shows presence of a flavone, flavonols and a glycoflavone. The flavone and flavonols are 3'-Ome luteolin and kaempferol and its derivative respectively.

In all 4 different glycoflavones (c-glycosides) have been detected. They are vitexin, 7-Ome vitexin, glycoflavone based on 7-Ome apigenin and 6-Ome orientin. The glycoflavones are almost absent from tribe Verbeneae except, in case of Verbena venusa where it is present in the form of glycoflavone based on 7-Ome apigenin. In the 2 members of Viticeae - Clerodendrum indicum and Holmskioldia sanguinea they are present as vitexin and 6-Ome orientin respectively. Vitexin has been reported uniformly in different parts of genus Vitex (Rao and Venkateswarulu, 1962). Its presence is reported in heartwood of V. altissima (Rao, 1965) and bark of V. negundo (Misra and Subramanian, 1980). However, the present study showed the absence of vitexin in both these species. The only member of Avicennieae screened here shows 7-Ome vitexin.

As in Bignoniaceae, the leucoanthocyanin test in most of the Verbenaceae members has been negative, except in Stachytarpheta mutabilis of Verbeneae and Clerodendrum splendens and Tectona grandis of Viticeae.

15 phenolic acids have been detected. p-Hydroxy benzoic, Gentisic, Syringic, p-Coumaric and Ferulic acids are common to all the three tribes. Tribe Verbeneae and Avicennieae show 100% incidence of p-hydroxy benzoic acid, while Viticeae show only 80%. Similarly Gentisic acid has 78.5, 55, and 100% incidence value in three respective tribes (Table 15). Vanillic acid is quite frequent in tribe Verbeneae and Viticeae, however, absent in Avicennieae. Syrigic acid is rather more common in tribe Verbeneae than in Viticeae. Genus Avicennia also shows presence of Syringic acid. p-Coumaric acid is widely distributed in tribe Viticeae and in genus Avicennia, whereas O-Coumaric acid is frequent in tribe Verbeneae.

Protocatechuic and Salicylic acids are confined to tribe Viticeae. Salicylic acid is detected in Clerodendrum fragrans, C. infortunatum, C. macrosiphon, C. paniculatum and Gmelina arborea. As against this only one Verbeneae member - Phyla nodiflora shows presence of this phenolic acid. Taxa like Citharexylum subserratum, Lantana salvifolia from tribe Verbeneae and Clerodendrum inerme, C. multiflorum, Table : 14. The Distribution of Phenolic Acids, Leucoanthocyanins, Alkaloids, Iridoids, Saponins, Tannins and Quinones in the Verbenaceae.

Sr. No.	NAME OF THE PLANT	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
	TRIBE VERBENEAE																							
1.	Citharexylum subserratum Sw.		+		+			+				+		+		+			+					
2.	Duranta repens Linn.		+	+				+	+		+	+											+	
3.	Lantana camara var. aculeata (Linn.) Moldenke		+	+				+	+	+									+	+			+	
4.	var. hybrida Moldenke		+	+				+				+		+					+	+		+		+
5.	var. mista Bail y		+	+					+		+			+	+	+		+	+				+	
6.	var. nivea Bail y		+	+				+	+		+	+		+					+	+		+		+
7.	Lantana salviiolia Jacq.		+	+	+			+				+		+				+		+				+
8.	Petrea volubilis Jacq.		+	+				+	+		+	+		+				+		+			+	+
9.	Phyla nodiflora (Linn.) Greene	+	+	+		+		+	+	+		+								+			+	
10.	Stachytarpheta indica Vahl		+	+					+			+		+				+	+	+			+	
11.	S. mutabilis Vahl		+	+				+	+		+	+					+	+	+	++			+	+
12.	Verbena venosa Gill & Hook.		+					+	+	+	+			+		+		+		+			+	
13.	V. bipinnatifida Nutt.		+	+				+		+	+	+		+		+		+		+		+	+	
14.	V. tenuisecta Briq		+					+		+	+	+		+		+				+				+
	TRIBE VITICEAE																							
15.	Clerodendrum aculeatum (Linn.) Griseb.		+					+				+												+
16.	C. fragrans Vent.	+	+					+						+		+				+			+	+
17.	C. [illegible]cum (Linn.) Kuntze		+					+										+		+		+		
18.	C. inerme (Linn.) Gaertn.		+	+	+			+	+		+	+		+				+		+				+
19.	C. infortunatum Gaertn.	+						+				+		+				+		+			+	
20.	C. macrosiphon Hook. f.	+	+					+				+		+									+	
21.	C. multiflorum (Burm. f.) O. Ktze.		+	+	+			+				+		+	+	+				+				+
22.	C. paniculatum Linn.	+						+		+	+	+		+		+				+				+

Contd.

Table : 14. Contd.

Sr. No.	NAME OF THE PLANT	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23
23.	Clerodendron siphonanthus R. Br.		+	+			+	+			+	+		+				+		+			+	
24.	Clerodendrum splendens Don		+					+				+		+		+	+	+		+			+	+
25.	C. speciosum Hort.		+	+				+				+						+		+			+	+
26.	C. thomsoriae Bal.							+				+						+					+	
27.	Gmelina arborea Roxb.	+	+	+				+	+		+	+	+	+				+					+	
28.	G. asiatica Linn.		+	+				+			+	+		+				+		+				+
29.	G. philippens s Cham.		+					+				+		+				+		+			+	
30.	Holmskioldia sanguinea Retz.		+	+	+			+				+		+		+		+		+			+	
31.	Premna resinosa (Hochst.) S hau.		+	+				+			+	+						+						
32.	Tectona grandis Linn. f.			+							+					+	+			+		+		+
33.	Vitex negundo L.		+	+	+			+	+		+			+				+	+	+			+	
34.	V. altissima Linn. f.		+	+							+	+		+				+		+				
	TRIBE AVICENNIEAE																							
35.	Avicennia ma r na (Forsk.) Vierh. Syn. A. alba auct. non Blume		+	+	+				+			+		+						+			+	

Phenolic Acids

1. Salycylic
2. p-OH-Benzoic Acid
3. Gentisic
4. Protocatechuic
5. 2-Hydroxy-5-methoxy benzoic Acid
6. 2-Hydroxy-6-methoxy " ".
7. Vanillic
8. Syringic
9. o-Coumaric
10. Melilotic
11. p-Coumeric
12. Phloretic
13. Ferulic
14. Sinapic
15. Chlorogenic
16. Leucoanthocyanins
17. Alkaloids
18. Iridoids
19. Saponins
20. Tannins

Quinones

21. Anthraquinones
22. Benzoquinones
23. Naphthaquinones

Table : 15. Percentage of incidence of phenolic acids in the tribe Verbeneae, Viticeae and Avicennieae.

PHENOLIC ACIDS	Verbenaceae (Tribes)		
	Verbeneae	Viticeae	Avicennieae
Salicylic	7.14	25	-
p-Hydroxybenzoic	100	80	100
Gentisic	78.57	55	100
Protocatechuic	14.28	20	100
2-Hydroxy 5-methoxy Benzoic	7.14	-	-
2-Hydroxy 6-methoxy Benzoic	-	5	-
Vanillic	85.7	90	-
Syringic	64.28	20	100
o-Coumaric	35.7	5	-
Melilotic	57.14	45	-
p-Coumaric	78.57	80	100
Phloretic	-	5	-
Ferulic	71.42	70	100
Sinapic	7.14	5	-
Chlorogenic	35.7	30	-

Holmskioldia sanguinea and *Vitex negundo* from tribe Viticeae show presence of protocatechuic acid and so also the genus *Avicennia*.

2-OH,5-Ome Benzoic and 2-OH,6-Ome Benzoic acids are identified from *Phyla nodiflora* and *Clerodendrum siphonanthus* respectively. Melilotic acid is fairly common in various members of tribe Verbeneae and Viticeae but absent in Avicennieae. The phenolic acid in *Gmelina arborea* gives colour reaction with spraying reagents. However, the colour fades soon afterwards. This indicates doubtful presence of phloretic acid in the plant. Sinapic acid is detected in *Lantana camara* var. *mista* and *Clerodendrum multiflorum*. Chlorogenic acid is common to Verbeneae (35%) and Viticeae (30%) but absent in Avicennieae.

Thus, from the distribution pattern of various phenolic acids in Verbenaceae, it is clear that a number of phenolic acids are common to all the 3 tribes. However, some of them are common to tribe Verbeneae and Viticeae but absent in tribe Avicennieae. The higher incidence of Gentisic, Vanillic, o-Coumaric, Melilotic in tribe Verbeneae, that of Salicylic, Protocatechuic in Viticeae, and absence of Salicylic, Vanillic, o-Coumaric, Melilotic and Chlorogenic in Avicennieae characterises the 3 tribes.

8 members of the Verbenaceae (7 of Verbeneae and 1 of Viticeae) have been found to possess iridoid compounds. Iridoids are absent in Avicennia. The presence of iridoids from Duranta repens (Rimpler and Helmut, 1974) could not be confirmed in the present study. The positive test for iridoids in case of Lantana camara, Stachytarpheta indica and S. mutabilis agrees with the earlier reports (Ford, et al., 1980; Tantisewie and Ottosticher, 1975 and Gurnier, 1979). The higher incidence of iridoids in the tribe Verbeneae as compared to Viticeae and their complete absence in the Avicennieae have taxonomic significance.

Saponins are present in 27 Verbenaceae taxa. Most of them show moderate amount of saponin (+). As in the Bignoniaceae, Tannins are absent in the Verbenaceae.

32 members show the presence of various Quinones. Tribe Verbeneae has benzoquinones and a higher incidence of anthraquinones. Tribe Viticeae has benzoquinones, naphthaquinones and a lower incidence of anthraquinones. Avicennieae, on the other hand has benzoquinones but no anthraquinones.

Around 62% of the plants gave positive test for alkaloids. However, when analysed chromatographically using BAW as solvent system, they did not show clearcut bands either in UV light or in iodine chamber, nor when sprayed with Dragendroff's reagent.

3:6 Discussion

Of the 8 tribes (Bentham and Hooker), only 3 tribes have been worked out in the present study. The chemical data points to the distinct nature of 3 tribes studied, due to their characteristic compounds. The tribe Verbeneae may be considered as chemically advanced due to occurrence of flavones in all the 14 members screened, absence of leuco-anthocyanins and glycoflavones accompanied by the presence of iridoid compounds. Tribe Viticeae also shows presence of flavones and flavonols. Although flavones are detected in majority of viticeae members, yet the percentage incidence of flavones is comparatively lesser. Loss of flavonoid compounds in some of the *Clerodendrum* species marks the advancement of the tribe. However, the tribe is, on the whole, slightly at a lower evolutionary level than the Verbeneae. The presence of a glycoflavone and absence of iridoids in *Avicennia*, the only member screened in the third tribe Avicennieae appears to be primitive.

The various representatives of these 3 tribes are arbitrarily grouped by Bentham and Hooker based on morphological similarities. The so-called groups are not designated by any taxonomic category. In the present discussion, more emphasis is laid on tribe Verbeneae and Viticeae. An attempt is being made to utilise chemical data to substantiate grouping of the various members based on morphological (flower and fruit particularly) criteria.

All the 14 Verbeneae members show presence of flavones. Of these 14 members, 5 also possess flavonol type of compounds.

The first group under Verbeneae includes Lantana, Phyla and Stachytarpheta. The genus Lantana as a whole, can be considered as an advanced member within the tribe Verbeneae due to higher incidence of flavones, iridoids and absence of glycoflavones and leucoanthocyanins.

Lantana camara and its 4 varieties studied in the present investigation show presence of acacetin in the leaf extract uniformly. However, the 4 varieties can be distinguished on other chemical grounds. Lantana camara var. aculeata and nivea, have apigenin as the flavone. But, the presence of 4'-Ome scutellarein along with anthra- and naphthaquinones in var. nivea and that of 6-OH luteolin and benzoquinones in var. aculeata clearly indicates that these two are distinct from each other not only morphologically but chemically as well. Similarly, var. hybrida with Scutellarein, 6-Ome-luteolin, anthra- and naphthaquinone and absence of alkaloids is quite different from var. mista, which shows presence of 4'-Ome scutellarein, alkaloids and benzoquinones.

The presence of iridoids in all the 4 varieties points to the distinct and advanced chemical feature of Lantana camara; thus characterising itself from the other species - L. salvifolia. The distribution pattern of flavonoids in the two species of Lantana also reveals chemical differences at species level. L. camara and its 4 varieties show apigenin and scutellarein type of flavones. Whereas, L. salvifolia is found to contain scutellarein and luteolin derivatives along with quercetin and its methyl derivatives. The presence of protocatechuic acid and absence of anthra- and benzoquinone, iridoids in L. salvifolia also substantiate the species difference.

Phyla nodiflora is characterised by presence of only flavones. Some of them it shares with Lantana.

Stachytarpheta possesses both flavones and flavonols. The two species of Stachytarpheta show different o-glycosides. S. indica contains 4 flavones and one flavonol. Whereas, S. mutabilis shows acacetin and scutellarein derivative as flavones and quercetin and kaempferol as flavonols. The absence of phenolic acids (Vanillic and Melilotic), leuco-anthocyanins and presence of only benzoquinone characterise

S. indica. On the other hand, S. mutabilis not only possesses all these compounds but has in addition naphthaquinones also.

Thus the arbitrary grouping of Lantana, Phyla and Stachytarpheta based on the ovary and fruit characters is justified on the basis of chemical findings. Phyla is chemically closer to Lantana than to Stachytarpheta.

Genus Verbena, which represents another group in Verbeneae shows flavones, flavonols and glycoflavones and absence of iridoids. The 3 species of Verbena screened, reflect chemical differences in their pattern of flavonoid distribution. The only common feature among these species is the presence of apigenin or its derivative as flavone. This supports their placement under one genus.

The presence of a single flavone and a glycoflavone in V. venosa reveals the presence of both advanced and primitive character in the species. The occurrence of 3'-methoxy quercetin and 3',4'-dimethoxy quercetin in addition to a single flavone - 6-methoxy scutellarein in V. bipinnatifida is suggestive of its primitive nature. V. tenuisecta appears to be an advanced species of the genus. The quinone distribution pattern also shows distinct identity of these species. V. venosa shows presence of a benzoquinone, V. bipinnatifida -

with anthra- and benzoquinones, whereas V. tenuisecta shows only naphthaquinone.

The occurrence of a single flavone (6-hydroxy luteolin) in Petrea volubilis may be considered as an advanced feature leading to the elimination of entire flavonoid system.

The morphological grouping of Citharexylum and Duranta under a group in the Verbeneae is not supported on the basis of present study. Except for Apigenin, the distribution of other flavones is quite different in the two genera. The presence of quercetagetin, iridoids and complete absence of quinones in Citharexylum and absence of flavonols, iridoids and presence of quinones in Duranta indicate their chemical differences. Duranta shows a number of chemical similarities with Lantana rather than Citharexylum.

Tribe Viticeae has been divided into two broad categories, based on the corolla and fruit characters. The first group include members like Tectona with regular corolla and 1-4-seeded pyrene fruit.

Tectona grandis is considered as the most primitive taxon due to retention of 5 fertile stamens in the flowers. The present chemical data also point to its primitiveness due to presence of flavonols, leucoanthocyanins and absence of iridoids. Anatomically however, it is considered an advanced genus due to presence of ring-porous wood, paratracheal banded parenchyma (Metcalfe and Chalk, 1950; Chawdhary, 1964).

The second group in the Viticeae is characterised by irregular corolla and 4 didynamous stamens. As in tribe Verbeneae, this is divided into unnamed subgroups, mainly on fruit character.

Premna and Vitex are grouped along with Gmelina. All the 3 taxa show presence of flavones and absence of leucoanthocyanins and glycoflavones. Premna and Gmelina have acacetin in common. However, the presence of other flavonoid compounds indicate some amount of dissimilarity between these two genera. The presence of flavonols and absence of iridoids in Gmelina as against presence of iridoids in Vitex reflect the chemical difference. The pollen grains are also distinct in the two genera. Gmelina is exceptional in the tribe Viticeae having 3-zonocolporate type. While, Vitex and Premna have 3-zonocolpate type (Nair and Rehman, 1962).

The 3 species of Gmelina screened in the present study are also found to be chemically distinct. Acacetin is common to all of them but G. arborea, in addition to acacetin, shows presence of apigenin, luteolin and 3'-methoxy quercetin.

While G. asiatica has kaempferol and 3',4'-dimethoxy quercetin. The occurrence of acacetin in all the 3 species indicate the homogeneity of this taxon. G. arborea has advanced type of flavonoid compounds - mainly the flavones. The G. asiatica, due to presence of more flavonols than flavones indicates primitive nature. G. philippensis with single flavone may be considered as an advanced feature leading to the elimination of entire flavonoid system. The distinct nature of these 3 species is also evidenced on palynological grounds (Nair and Rehman, 1962).

The two species of Vitex screened, also appear to be distinctive chemically as well as palynologically. V. negundo possesses flavones, iridoids and prolate pollens. The other species - V. altissima is devoid of such compounds and exhibit subprolate pollen grains.

The other subgroup includes Clerodendrum and Holmskioldia due to presence of 4-pyrene drupe. The flavonoid distribution pattern also shows chemical similarity in the two taxa. However, the presence of glycoflavone in Holmskioldia points to its primitiveness within this subgroup. Palynologically also they have similar type of pollen.

The complete absence of leucoanthocyanins (except in Clerodendrum splendens), the presence of flavones and at times total absence of flavonoid system mark the advancement of Clerodendrum. The flavones when present, are mainly represented by apigenin and scutellarein derivatives except in Clerodendrum indicum which possesses 6-Ome scutellarein, 4'-Ome luteolin, Vitexin and anthraquinone. Occurrence of benzo- and naphthaquinone in most of the species of Clerodendrum are notable chemical characters. The genus, therefore, shows various levels of evolution represented by those with many flavones, those with a single flavone leading to a loss of flavonoid system in Clerodendrum aculeatum, C. infortunatum and C. thomsonae.

In the present study the monogeneric tribe Avicennieae is represented by Avicennia marina which possesses a single flavone, 2 flavonols and a glycoflavone. It also shows absence of leucoanthocyanins, alkaloids and iridoids. Although this taxon combines primitive and advanced characters, the primitive ones appear to have an edge.

Erdtman (1952) remarked that Avicennieae "probably should be regarded as a family of its own" on the basis of clear cut palynological differences. Such a contention has been supported by Nair and Rehman, 1962; Mukherjee and Chanda, 1973; Saxena, 1973, 1975, 1981; on the basis of the palynological studies of Indian members. Metcalfe and Chalk (1950) also considered the Avicennieae as anomalous in the

Verbenaceae and advocated its separation under a separate family on the basis of wood structures. Embryological studies of Junell (1934), Pal (1951), Rao (1952), Padmanabhan (1960), Maheshwari and Kapil (1966) have also suggested isolation of the Avicennieae from rest of the Verbenaceae.

Although there is an overwhelming evidence in support of separate family Avicenniaceae, the chemical characters presented here are not so different from the rest of the Verbenaceae to warrant a separate family status for the genus. Presently the author would be satisfied to retain Avicennieae within the confines of the family Verbenaceae.

It is highly probable that many of the character deviations that the group displays is a direct consequence of its adaptations to peculiar ecological niche. It is, therefore, felt improper to draw sweeping taxonomic conclusions on the basis of ecologically adaptive characters.

All the 3 tribes - Verbeneae, Viticeae and Avicennieae have distinctive chemical characters. However, the differences are of a minor nature and, therefore, raising these groups to subfamilial level is considered unwarranted.

A brief discussion on the taxonomic position of genus _Nyctanthes_ will not be out of place here. The genus _Nyctanthes_ has changed position at the hands of taxonomists. It has been traditionally grouped under Oleaceae. However, Airy Shaw (1952) on morphological and stant (1952) on anatomical evidences proposed its transfer as a new subfamily under the Verbenaceae. In the latest edition of Engler's syllabus (ed. Melchior, 1964), Nyctanthes is grouped under the Verbenaceae. Embryological investigations (Kapil and Vani, 1966), trichome studies (Inamdar, 1967) and anatomical studies on gynoecium (Kshetrapal and Tiagi, 1970) however, do not support the transfer of Nyctanthes from Oleaceae to Verbenaceae. Daniel and Sabnis (1981, in press) have proposed placement of Nyctanthes in the Oleaceae based on chemical characters and numerical analysis.

Chemically Nyctanthes is closer to subfamily Jasminoideae of Oleaceae. It has a high frequency of flavonol and absence of glycoflavones. The Verbenaceae on the other hand, is characterised by both flavonols and flavones (Daniel and Sabnis, 1979).

The presence of iridoids is considered an advanced character (Jensen et al., 1975). Out of the 35 plants screened under the present study, 7 of Verbeneae and 1 of Viticeae show presence of iridoids. This suggests an advanced nature of the tribe Verbeneae within the family Verbenaceae.

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