

## Chemosystematics of *Gardenia*

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**Abstract.** The distribution of different chemical constituents in 6 species of *Gardenia* is numerically analysed. The quantified chemical data show that the taxa studied are closely related and suggest that they could be divided into 4 clusters. Based on the established criteria of simplicity and complexity in biosynthetic pathways, rarity and ubiquity in distribution of the chemical constituents and/or correlation studies *Gardenia longistyla*, *Gardenia resinifera* and *Gardenia thunbergia* have tentatively been found to be relatively advanced over the other 3.

**Keywords.** Numerical analysis; chemotaxonomy; *Gardenia*.

### 1. Introduction

There are a few published reports on the chemotaxonomy of Rubiaceae (Gibbs 1974; Kiran Mai *et al* 1984, 1985, 1986; Radhakrishnaiah *et al* 1987). The present account on distribution pattern of the phytochemical constituents in few Indian species of *Gardenia* is undertaken to establish the extent of kinship, re-evaluate their earlier infrageneric alignment and the relative advancement in the light of the quantified chemical data.

### 2. Materials and methods

The aerial parts of *Gardenia jasminoides* Ellis., *G. gummifera* Linn. f., *G. latifolia* Ait., *G. longistyla* Hook., *G. resinifera* Roth., and *G. thunbergia* Linn., of the present study were collected from Lalbagh Garden, Bangalore and Indian Botanic Garden, Howrah and screened for different phytochemical constituents (Amarsingham *et al* 1964; Weiffering 1966; Raffauf 1970; Harborne 1973; Gibbs 1974; Santaram 1983). The voucher specimens are deposited in the Herbarium, Department of Botany, Nizam College, Osmania University, Hyderabad. The unidentified chromatographic spots identical in spot colours and  $R_f$  values on different chromatograms were assigned a code and designated by the same.

### 3. Results and discussion

The distribution pattern of the chemical constituents encountered is presented in table 1. While there is uniform occurrence of syringyl radicals (components of lignin), there is restricted distribution of such constituents as aucubin compounds (monoterpenoid cyclopentanoid lactones), cardenolides (cardiac glycosides), dihydrochalcones, flavones, flavonones, proanthocyanidins (all flavonoids), ellagic acid

Table 1. Distribution pattern of the chemical constituents.

Chemical constituent	Name of the taxon*					
	1	2	3	4	5	6
Dihydrochalcones	+					+
Flavones		+	+	+		
Flavonols					+	
Proanthocyanidins	+	+	+			
Anthraquinones	+	+	+		+	+
Ellagic acid	+	+	+			
Tannins	+	+	+			
Aucubin	+			+		
Cardenolides			+			
Iridoids	+?		+			
Polyphenolase activity	+	+	+	+	+	+
Syringaldehyde	+	+	+	+	+	+
Saponins						+
Unusual amino acids						
a hRf-20		+	+	+		
b hRf-22	+				+	+
c hRf-28	+	+	+		+	+
d hRf-33				+		
e hRf-50	+					
f hRf-53	+	+		+		+
g hRf-77	+			+		+
Phenolic acids						
<i>p</i> -OH benzoic acid	+	+	+	+	+	+
Caffeic acid	+	+	+	+		+
<i>p</i> -Coumaric acid	+	+	+	+	+	
<i>o</i> -Coumaric acid	+					+
Ferulic acid	+	+		+		
Salicylic acid	+	+	+	+	+	+
Vanillic acid	+	+	+	+	+	+
Unidentified phenolic constituents						
A hRf 66/81		+	+		+	+
B hRf 16/7		+			+	+
C hRf 15/6		+	+	+	+	+
D hRf 19/10				+		
E hRf 16/15					+	+
F hRf 15/48		+	+		+	
G hRf 8/5				+		
H hRf 19/14						+
I hRf 12/10				+	+	+

1, *Gardenia jasminoides* Ellis; 2, *G. gummifera* Linn. f.; 3, *G. latifolia* Ait.; 4, *G. longistyla* Hook.; 5, *G. resinifera* Roth; 6, *G. thumbergia* Linn.

(hydrolysed product of ellagitannins, but can occur independently also), iridoids (a broad category of monoterpenes) and saponins (glycosides of terpenoidal or steroidal alcohols) in a few taxa. A uniform absence of alkaloids (both quinolizidine and indole types) aurones (a type of flavonoids in some yellow flowers), catechol-tannins (condensation products of catechins), flavonones (a minor

flavonoid), juglone (a 5-hydroxy naphthaquinone or hydroxyquinone), lignans (dimers of cinnamic acid or their derivatives), syringin (syringic acid) triterpenoids steroids and tannins (hydrolysable tannins) is observed in the present study. Except for the reported occurrence of triterpenoids in *G. latifolia* and *G. gummifera* (Reddy *et al* 1975, 1977) the rest of the results are in consonance with those of Gibbs (1974) on the taxa studied by him. From the distribution pattern of free amino acids it is evident that some of the usual free amino acids (not shown in the table 1) have been found to be absent in the hydrolysed extracts which are otherwise ubiquitous. This may be due to the metabolic state of the tissue or environmental conditions or both. Hence their presence or absence has been excluded from taxonomic purview. However, some uncommon (presumably non-protein) amino acids (table 1) with different  $R_f$  values and spot colours noticed are designated by code letters a-g and taken into consideration for establishing the relationship among the species.

Phenolic constituents like *p*-OH benzoic, *p*-coumaric, salicylic and vanillic acids are uniformly present in all the taxa, while the distribution of ferulic acid, *o*-coumaric acid and unknown compounds like A-I are restricted to a fewer taxa (table 1). Both known and unknown compounds are taken into consideration for numerical analysis.

The distribution of secondary metabolites, unusual amino acids and phenolic constituents, all of which are stable and show least environmental variation are quantified. The Jaccard coefficient of similarity ( $S_J = nJK/nJK + U$ ) where  $nJK$  stands for number of positive matches between taxon *J* and *K* and *U* for the mismatches, ranges from 0.23-0.8 (table 2), thus exhibiting close kinship among the species. The group affinity index (Ellison *et al* 1962) i.e., the affinity of one taxon with all others ranges from 363-427 (table 2) also indicates the same.

The dendrogram of cluster analysis (figure 1) based on weighted pair group method average (WPGMA) of chemical characters shows that they could be divided into 4 clusters. On morphological grounds the species of *Gardenia* are accommodated under 3 sections viz., Eugardenia, Ceriecus and Rothmania (Hooker 1882). *G. gummifera*, *G. latifolia* and *G. resinifera* are placed in *Eugardenia*. The placement of rest of the species of the present study has not been recorded earlier. The infrageneric placement of *G. gummifera* and *G. latifolia* in one section is justified chemically. However, the *G. resinifera* seems to be more closer to the other species, then the former two.

Earlier, Bate-Smith (1962) categorised the families of dicotyledons into 4 classes ab, a<sub>0</sub>b, ab<sub>0</sub>, a<sub>0</sub>b<sub>0</sub> depending on the occurrence or absence of selected phenolic

Table 2. Synthetic numerical indices.

Name of the taxon*	Jaccard coefficient similarity						Group affinity
	1	2	3	4	5	6	
1	1.0	0.5	0.46	0.40	0.35	0.48	370
2		1.0	0.80	0.40	0.52	0.39	427
3			1.0	0.27	0.23	0.27	416
4				1.0	0.30	0.28	369
5					1.0	0.56	401
6						1.0	363

\*Same as in table 1.

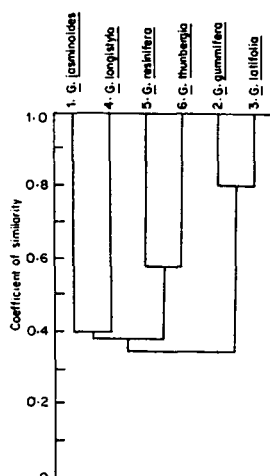


Figure 1. Dendrograms of cluster analysis based on the coefficient of similarity.

Table 3. Distribution of primitive and advanced characters.

Chemical constituents	Name of the taxon*					
	1	2	3	4	5	6
<b>Primitive characters</b>						
Cardenolides			100			
Catechol-tannins				100		
Dihydrochalcones	100			100		
Ellagic acid	100	100	100			
Flavonols					100	
Flavonones						
Proanthocyanidins	100	100	100			
Syringin						
Tannins (hydrolysable)	100	100	100			
Triterpenoids/steroids						
Total score of primitive characters	1000	1000	1000	1000	1000	1000
Actual score of primitive characters	400	300	400	100	100	
Score of characters that are not primitive (= advanced)	600	700	600	900	900	1000
<b>Advanced characters</b>						
Alkaloids						
Aucubin	100			100		
Anthraquinones	100	100	100		100	100
Cyanogenic glycosides						
Flavones		100	100	100		
Indoles						
Juglone						
Lignans						
Saponins						100
Syringaldehyde	100	100	100	100	100	100
Total score of advanced characters	1000	1000	1000	1000	1000	1000
Actual score for advanced characters	300	300	300	300	200	300
Score of characters that are not advanced (= primitive)	700	700	700	700	800	700

\*Same as in table 1.

compounds of which the  $ab$  and  $a_0b_0$  groups represent the primitive and advanced situations respectively. The  $a_0b$  and  $ab_0$  groups represent the intermediate states. The families having the leucoanthocyanins (a) and flavonols and trihydroxy acids (b) are categorised under  $ab$ , those devoid of them as  $a_0b_0$  and those with one of them as  $a_0b$  and  $ab_0$ . In the present study, all the secondary chemical constituents (including phenolics) which are present in all climes (Fluck 1963) unaffected by the environmental fluctuations and get accumulated in the plant tissues in large amounts, have been taken into considerations for indicating the relative primitiveness/advancement on the basis of established criteria of simplicity or complexity in biosynthetic pathways and ubiquity or rarity of the compounds and constituents correlated with morphological features. Thus the presence of alkaloids (including indoles), anthraquinones, aucubin compounds, cyanogenic glycosides, flavones naphtho (=hydroxy) quinones (juglone), lignans, saponins and syringyl radicals among the angiosperms in general is recorded as advanced and their absence as primitive. Similarly the presence of cardenolides, catechol-tannins, dihydrochalcones, ellagic acid, flavonols, flavonones, proanthocyanidins, syringin and triterpenoids/steroids is regarded as primitive and their absence as advanced (Gibbs 1962; Sporne 1974; Smith 1976; Padhye *et al* 1981; Daniel and Sabnis 1982; Gershenson and Mabry 1983; Nageshwar 1986; Satyavati *et al* 1987; Anuradha *et al* 1987). The extent of primitiveness/advancement is calculated following Daniel and Sabnis (1982) and presented in tables 3–5, where the presence of each character is treated as 100 points. From the numerical analysis it is tentatively inferred that *G. longistyla*, *G. resinifera* and *G. thunbergia* are relatively advanced over the other 3 taxa studied.

Table 4. Total score of primitive and advanced characters.

Name of the taxon	Primitive score*	Total	Advanced score**	Total
<i>Gardenia jasminoides</i>	400 + 700	1100	300 + 600	900
<i>G. gummifera</i>	300 + 700	1000	300 + 700	1000
<i>G. latifolia</i>	400 + 700	1100	300 + 600	900
<i>G. longistyla</i>	100 + 700	800	300 + 900	1200
<i>G. resinifera</i>	100 + 800	900	200 + 900	1100
<i>G. thunbergiana</i>	0 + 700	700	300 + 1000	1300

\*Primitive score: Actual primitive score + score of characters that are not advanced.

\*\*Advanced score: Actual advanced score + score of characters that are not primitive.

Table 5. Net score.

Name of the taxon	Primitive score (-)	Advanced score (+)	Net score
<i>G. jasminoides</i>	1100	900	- 200
<i>G. gummifera</i>	1000	1000	0
<i>G. latifolia</i>	1100	900	- 200
<i>G. longistyla</i>	800	1200	+ 400
<i>G. resinifera</i>	900	1100	+ 200
<i>G. thunbergiana</i>	700	1300	+ 500

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