

SUMMARY AND HIGHLIGHTS

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The Caryophyllales are unique in containing a number of features such as p-type plastids, betalains and pantoporate type of pollen. The absence of betacyanins in the Caryophyllaceae and Molluginaceae is often cited to recommend the expulsion of these taxa from this order. The families Didiereaceae and Cactaceae are included here after the discovery of betacyanins from them.

The thesis incorporates chemical studies on 98 plants belonging to the Amaranthaceae (23), Chenopodiaceae (13), Cactaceae (9), Nyctaginaceae (5), Phytolaccaceae (4), Portulacaceae (3), Basellaceae (1) Caryophyllaceae (24) and Polygonaceae (17). These members have been screened for various chemical markers such as leaf flavonoids, phenolic acids, proanthocyanidins, tannins, saponins, alkaloids, quinones and iridoids. The data have been used to assess the intrafamilial classifications, interrelationships among various families and the phylogenetic history of the different taxa at all levels of hierarchy. The position and status of certain controversial taxa are also evaluated. The highlights of the present investigation are summarised below:

× Flavonols formed the dominant phenolic pigments of the Caryophyllales. The Amaranthaceae, Chenopodiaceae, Cactaceae, Illecebraceae, Petiveriaceae, Nyctaginaceae and Polygonaceae

X are rich ⁿin these compounds. Flavones and glycoflavones
 y predominated ⁿthe Caryophyllaceae. All these groups of
 flavonoids were equally represented in the Basellaceae. In
 the Portulacaceae and in the Phytolaccaceae flavonoids were
 absent. Though proanthocyanidins and tannins were seen in
 both the Polygonaceae and Cactaceae, their distribution was
 very rare in the latter family. Alkaloids have a very
 restricted occurrence, obtained from the Phytolaccaceae,
 Cactaceae, and Chenopodiaceae. Quinones were located only
 in the Polygonaceae. Saponins and steroids were universally
 present. Iridoids were absent in all the families screened.

The presence of quinones as the dominant phenolic
 pigments and the occurrence of tannins and proanthocyanidins
 mark the Polygonaceae distinct from the other families screened.
 These characters justify the separation of this family from
 the Caryophyllales and placing them in a monotypic order the
 Polygonales. The presence of unilocular ovary and solitary
 ovules in basal placentation and more or less curved embryo
 validates treating the Polygonales in the subclass
 Caryophyllidae along with the Caryophyllales.

The remaining families of the Caryophyllales form a
 more or less homogenous order, characterized by the presence
 of p-type plastids, antoporate type of pollen and absence of
 quinones and tannins. But the distribution of various classes

of flavonoids demarcate two groups in the Caryophyllales.

The first group, containing the Amaranthaceae, Chenopodiaceae, Nyctaginaceae, Illecebraceae, Basellaceae, Petiveriaceae, ~~Phytolaccaceae~~ and Portulacaceae, ^{is} ~~are~~ characterized by the presence of flavonols as the main phenolic pigments. Apart from this, alkaloids are also detected from this group only. Except Illecebraceae all these families characteristically produce betacyanins. The second group, with the Caryophyllaceae and Molluginaceae ^{is} ~~were~~ characterized by the predominance of flavones and glycoflavones, and they produce anthocyanins instead of betacyanins. The Illecebraceae, differing from the former group in the absence of betacyanins and from the latter group in the absence of flavones/glycoflavones, are considered as an intermediate taxon. This family is placed in the 2nd group for the time being. These two groups correspond to two suborders, the Chenopodineae and Caryophyllineae proposed by Mabry et al. (1977).

Within the suborder Chenopodineae, the Cactaceae, Aizoaceae, Nyctaginaceae, Portulacaceae, Petiveriaceae, and Cistaceae contained flavonols as the sole flavonoids and form a subgroup within the Chenopodineae. Flavones and glycoflavones were not located from this group and at least two families contained proanthocyanidins. The Amaranthaceae, Chenopodiaceae, Basellaceae and Phytolaccaceae

comprise a second group and appear to be very closely related because of the higher incidence of flavonols and in containing flavones and/or glycoflavones in a few members.

x Within the family Amaranthaceae the tribe Celoseae, being devoid of flavonoids, is found to be different from other taxa and thus deserved a separate status. This tribe is elevated to a subfamily Celosioideae in par with the Amaranthoideae and Gomphrenoideae. The subfamily Celosioideae, though considered primitive on morphological grounds, show advanced chemical characters like elimination of flavonoid system. Of the 3 subfamilies in the amaranthaceae, the subfamily Amaranthoideae is the most primitive because of predominance of flavonols. The subfamily Gomphrenoideae has a combination of advanced flavones and primitive flavonols and therefore occupies an intermediate position in the evolutionary sequence between the Celosioideae and Amaranthoideae.

Basella rubra differs from all other plants screened in the Chenopodiaceae in possessing all the three types of flavonoids (flavonols, flavones and glycoflavones). This uniqueness of Basella validates its separation and elevation to a unigeneric family, the Basellaceae. The chemical distinctiveness of Beta vulgaris, in containing glycoflavones which are not otherwise seen in the

Chenopodiaceae, is considered sufficient to assign a subfamily status, the Betoideae, to this genus.

Existence of two tribes in the Cactaceae is established by the chemical study. Of the two tribes, the opuntioae seem to be more primitive due to the higher percentage of incidence of quercetin and more variety of flavonols. The tribe Echinocactae is relatively advanced with a higher percentage of incidence of kaempferol, a less hydroxylated compound, and with fewer variety of flavonols. No chemical difference was observed between the leafy and leafless members of the Cactaceae.

Retiveria alliacea is quite distinct from the rest of the Phytolaccaceae screened in containing flavonols. It also has a rare distinction of possessing sulphides which are otherwise not observed in the Phytolaccaceae. This chemical uniqueness support the family status assigned to it.

The inter-relationships of Gisekia with all three families, the Phytolaccaceae, Aizoaceae and Molluginaceae, with which it is associated ^{at} one time or other, is evaluated. Gisekia shares some characters each with all the three families. But the unique combination of characters found in Gisekia makes this genus an odd member in all these three families. Therefore, the creation of a

unigeneric family, the Gisekiaceae is upheld.

The position of Sphenoclea, which is kept in the Phytolaccaceae by Airy shaw⁽¹⁹⁴⁸⁾ is evaluated and its placement in the Campanulaceae rather than the Phytolaccaceae is supported.

The Illecebraceae, with only flavonols, are identified as a separate group from the flavone/glycoflavone - rich Caryophyllaceae and thus the validity of the family Illecebraceae is justified. Within the Caryophyllaceae the chemical data bring forth the existence of two distinct groups, the Caryophylloideae and Alsinoideae.

The distribution pattern of the flavonoids and other chemical markers indicate the homogeneity of the family Polygonaceae. All the plants possess similar flavonoids and phenolic acids.

Based mainly on chemical characters, but supported by other evidences, a Rosalean or pro-rosalean origin of the Caryophyllales is proposed.

A cladistic analysis is carried out using chemical data and data from other disciplines and a Cladogram is prepared, based on Wagner's ground plan divergence method, to depict the Phylegenitic status of the families and the

dichotomies met within the group. The Amaranthaceae are found to be the most advanced family and Cactaceae the most primitive, while rest of the families are positioned in the intermediate levels in the evolutionary sequence.

In addition, the present work brings out data on the chemistry of many plants, which were not studied previously. A number of new sources of flavonoids (especially bio-flavonoids) tannins, saponins and alkaloids are identified.

The new sources of chemical compounds identified are present below:

1. NEW SOURCES OF BIOFLAVONOIDS

- | | |
|----------------|--|
| Luteolin | - <u>Polygonum plebejum</u> , <u>Rumex acetosella</u>
(Polygonaceae). |
| 7-OMe Luteolin | - <u>Arenaria foliosa</u> , <u>A.kashmirica</u>
(Caryophyllaceae) |
| Orientin | - <u>Dianthus jacquemontii</u> , <u>Polycarpea corymbosa</u> , <u>Silene conoidea</u> (Caryophyllaceae) |
| Quercetin | - <u>Herniaria hirsuta</u> , (Illecebraceae)
<u>Chenopodium murale</u> , <u>Agroglchin persicarioides</u> , <u>Suaeda fruticosa</u> ,
<u>S.nudiflora</u> (Chenopodiaceae), <u>Polygonum alpinum</u> , <u>P.amphibium</u> , <u>P.hydripiper</u> ,
<u>P.kashmirica</u> , <u>P.hetrophyllum</u> . <u>Antigonon leptopus</u> , <u>Persicaria glabra</u> . |

Opuntia Sp., Opuntia dillenii,

Mamillaria Sp. (Cactaceae)

7-OMe - Chenopodium hybridum (Chenopodiaceae)

2. FLAVONES

Apigenin - Sagina procumbens, (Caryophyllaceae)

4'-OMe Apigenin - Alternanthera sessilis (Amaranthaceae)
Sagina procumbens (Caryophyllaceae)

Luteolin - Alternanthera sessilis, (Amaranthaceae)
Polygonum plebejum, Rumex acetosella
(Polygonaceae)

3'-OMe - Alternanthera sessilis, (Amaranthaceae)

Luteolin Cerastium monosperma, Polycarpea
corymbosa (Caryophyllaceae)

7-OMe Luteolin - Arenaria foliosa, A. kashmirica, Sagina
procumbens, (Caryophyllaceae)
Polygonum amplexicaule (Polygonaceae)

4'-OMe Luteolin - Alternanthera sessilis (Amaranthaceae)

3',4'-DiOMe - Alternanthera sessilis, (Amaranthaceae)

Luteolin Arenaria festucoides, Polycarpea corymbosa
(Caryophyllaceae)

7,3'-4'-TriOMe - Oxygonum sinuatum (Polygonaceae)

Luteolin

3. FLAVONOLS

Kaempferol - Polygonum plebejum, P. heterophyllum

- (Polygonaceae), Cereus forbesii,
C.peruvianus, Cereus Sp., Mamillaria Sp.
Melocactus Sp., Opuntia Sp., Pereskia
grandifolia (Cactaceae), Agroglchin
persicarioides, Chenopodium murale,
Chenopodium botryodes, C.hybridum, Kochia
indica (Chenopodiaceae), Bougainvillea
spectabilis (Nyctaginaceae), Herniaria
hirsuta (Illecebraceae).
- 3-OMe kaempferol - Alternanthera pungens (Amaranthaceae)
- 4'-OMe
kaempferol - Polygonum kashmiricum (Polygonaceae),
Cereus peruvianus, Cereus Sp., Pereskia
grandifolia (Cactaceae), Agroglchin
persicarioides, Chenopodium botryodes,
Kochia indica (Chenopodiaceae),
Boerhavia procumbens (Nyctaginaceae),
Arenaria festucoides (Caryophyllaceae),
Herniaria hirsuta (Illecebraceae),
Allmania nodiflora (Amaranthaceae).
- 7,4'-DiOMe
Kaempferol - Nothosaerva brachiata (Amaranthaceae).
- Quercetin - Polygonum hydropiper, P.amphibium, P.
alpinum, P.kashmiricum, P.hetrophyllum,
Persicaria glabra, Rumex acetosella,
Antigonon leptopus, Coccoloba grandi-
flora, Muehlenbeckia platyclada

(Polygonaceae), Mamillaria Sp., Opuntia dillenii, Fereskia grandifolia, (Cactaceae), Agroglchin persicarioides, Chenopodium murale, Kochia indica, Salicornia brachiata, Suaeda fruticosa, S. nudiflora (Chenopodiaceae), Herniaria hirsuta (Illecebraceae).

- 3-OMe Quercetin - Polygonum plebejum (Polygonaceae), Petiveria alliacea (Petiveriaceae), Aerva javanica, Gomphrena globosa (Amaranthaceae).
- 3'-OMe Quercetin - Persicaria glabra, Polygonum hydropiper, P. alpinum, P. kashmiricum, P. heterophyllum, Rumex hastatus (Polygonaceae), Melocactus Sp., Opuntia dillenii (Cactaceae), Beta vulgaris, Chenopodium album, Suaeda fruticosa, Salsola bryosma, Haloxylon recurvum (Chenopodiaceae), Digera muricata, Aerva javanica (Amaranthaceae), Bougainvillea spectabilis, B. glabra (Nyctaginaceae), Alternanthera pungens, Gomphrena globosa (Amaranthaceae).
- 4'-OMe Quercetin - Polygonum heterophyllum (Polygonaceae), Cereus forbesii (Cactaceae), Salicornia brachiata, Suaeda nudiflora (Chenopodiaceae), Digera muricata (Amaranthaceae).

- 7-OMe Quercetin - Chenopodium hybridum (Chenopodiaceae),
Polygonum amplexicaule (Polygonaceae).
- 3',4'-DiOMe Quercetin - Polygonum amphibium, P. alpinum, P. kashmiricum, Rumex hastatus (Polygonaceae),
Opuntia dillenii, Pereskia grandifolia (Cactaceae), Petiveria alliacea (Petiveriaceae), Amaranthus caudatus,
Digera muricata, Gomphrena decumbens, G. celosioides (Amaranthaceae),
- 7,3'-DiOMe Quercetin - Polycarpea corymbosa (Caryophyllaceae).
- 7,3'-4'-TriOMe Quercetin - Oxygonum sinuatum (Polygonaceae).

4. GLYCOFLAVONES

- 6-C-Glycosylated apigenin-(Isovitexin) - Muehlenbeckia platyclada (Polygonaceae),
Arenaria kashmirica (Caryophyllaceae),
- Vitexin - Cucubalus baccifer, Silene vulgaris,
Arenaria foliosa, A. kashmirica, Cerastium monosperma, Stellaria media, Vaccaria pyramidata (Caryophyllaceae)
- 4'-OMe Vitexin - Dianthus barbatus, Arenaria festucoides,
A. neelgeerrensis, Sagina procumbens,
Spergula arvensis, Lychnis apetala,
L. coronaria, Saponaria vaccaria,
Silene tenuis (Caryophyllaceae),
Basella rubra (Basellaceae).

- 7,4'-DiOMe vitexin - Lychnis coronaria, Stellaria media
(Caryophyllaceae).
7,4'-Isovitexin - Lychnis coronaria, Dianthus
accusmontii, Stellaria media,
(Caryophyllaceae).
Orientin - Rumex acetosella (Polygonaceae)

5. ALKALOIDS

Beta vulgaris, Chenopodium album, C. murale, C. botryodes,
C. hybridum, Suaeda fruticosa, S. nudiflora, Salsola bryosma,
Haloxylon recurvum (Chenopodiaceae), Boerhavia procumbens,
Bougainvillea spectabilis (Nyctaginaceae), Phytolacca
acınosa, P. latbena (Phytolaccaceae), Cereus forbesii,
C. peruvianus, Cereus Sp., Mamillaria Sp., Melocactus Sp.,
Opuntia dillenii, Opuntia Sp., Pereskia grandifolia
(Cactaceae).

6. PROANTHOCYANIDINS

Cereus peruvianus, Opuntia dillenii (Cactaceae), Polygonum
lepathifolium, P. amphibium, P. plebegum, Persicaria glabra,
Rumex patientia, Antigonon leptopus, Coccoloba grandiflora,
Muehlenbeckia platyclada (Polygonaceae)

7. TANNINS

Polygonum lepathifolium, P. amphibium, P. plebegum, Persicaria
glabra, Rumex patientia, Antigonon leptopus, Coccoloba
grandiflora, Muehlenbeckia platyclada (Polygonaceae).