

TITLE: THE CHEMOTAXONOMY OF THE "GERANIALES"

THE CHEMOTAXONOMY OF THE "GERANIALES"

by

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## INTRODUCTION

### A. History of taxonomy

Botany is an old science, one branch of which, taxonomy, antedates recorded history. In fact, the early Chinese, Egyptian, and Assyrian cultures were based, to a degree, on cultivated plants (Porter, 1959). The early developers of systematic botany included Theophrastus (370 - 287 B.C.), Pliny the Elder (23 - 79 A.D.), and Dioscorides (1st century, A.D.). Theophrastus, the "father of botany," classified plants according to the growth habit. Thus he had four groups: herbs, undershrubs, shrubs, and trees.

After their deaths, a lapse of more than 14 centuries followed in which there was <sup>little</sup> recorded botanical history. The next surge of interest came with the herbalists. These herbalists were many, and such names as Brunfels, Fuchs, Bock, Lobelius, Gerard, and Cordus, stand out. They wrote herbals, books which contained figures and descriptions from actual plant specimens. Gradually terminology appeared, and groupings resulted.

Andre Caesalpino belonged to a transition period, for the Renaissance with its ensuing changes was descending upon civilization. Botany broke away from tradition, as did the other arts and sciences, and developed a new system of classification, terminology of descriptions, and a system of nomenclature. This was the period of artificial classification, when plants were grouped according to their habit or number of a certain organ.

Eventually, there was a gradual break away from this artificial system to a more natural one, in which authors used what knowledge was known about the plants to devise a system. Among the men who broke away from the old tradition were John Ray (1627 - 1705), Joseph Pitton de Tournefort (1656 - 1708), Pierre Magnol (1638 - 1715), and Rudolf Camerarius (1665 - 1721).

The best known botanist of this period is Carolus Linnaeus (1707 - 1778), the father of modern botany. He is best known for his binary system, his close study of the plant world, and his definition of genera and species. His Species Plantarum (1753) marks the starting point of the binomial system of nomenclature. He initiated the use of a sexual system in classification, thus recognizing 24 classes based on the number of stamens, or some obvious character of it; this system was set forth in Systema Naturae (1735). In spite of its artificiality, this method was adopted because of the ease of classifying plants, and it was responsible for the period of relative stagnation which resulted in botany.

Systematic botany was revived in France by Bernard de Jussieu and his nephew Antoine Laurent. The birth of a natural system came with the memoir on Ranunculaceae, with the realization that characters should be weighed, not counted.

Other major works included those of A. P. de Candolle, Robert Brown, A. Brongniart, and Hofmeister.

Later systematists were G. Bentham and J. Hooker, van Tieghem, S. Endlicher, A. Eichler, and Adolf Engler. Others included C. Bessey, H. Hallier, R. von Wettstein, and J. Hutchinson.

Although there are many classification systems, no two are exactly-alike. Thus chaos in systematic botany still prevails. For example, Engler, Endlicher, Eichler, and Wettstein believed that the Gnetales or conifer-like line gave rise to woody dicotyledons, with unisexual, petal-bearing, wind-pollinated flowers of a few parts. Catkins were considered primitive. Bentham and Hooker, Bessey, Hallier, Arber and Parkin, and Hutchinson, on the other hand, believed in the "Ranalian theory," in which the ~~Corniferales~~ ~~Bennettitales~~ ~~Gnetales~~ gave rise to the Ranales, the primitive flowers being bisexual, many-parted,

and spirally-arranged. Simple flowers evolved from multi-parted ones.

This general chaos in taxonomy is further exemplified by comparing the classification of the Tubiflorae of Engler and Diels (1936) and Hutchinson (1959). According to Engler and Diels' scheme, the Tubiflorae consists of 22 families. Hutchinson, however, splits several of these families, and thus creates 28 families out of the original 22. To further complicate matters, he distributes these 28 families as several end-lines of evolution. Furthermore, he places the orders Verbenales and Bignoniales on the "woody" side, and Solanales, Personales, Polemoniales, the Boraginales, and the Lamiales, on the "herbaceous" side.

The goal of taxonomists is to devise a phylogenetic system in which organisms are placed together according to an evolutionary scheme. Thus organisms regarded as the most primitive are segregated from the more advanced. Since all plant parts do not evolve at the same rate, an organism would retain some primitive traits along with the advanced. Of course problems will be encountered, such as convergent, divergent, and parallel evolution. Often small mutations can cause a multitude of changes.

Research is being done to solve this problem of phylogenetic relationships between taxa. Paleobotany, embryology, cytology, and genetics are just a few of the fields. As Cronquist (1957) stated: "Every taxonomic character is potentially important, and no character has inherent, fixed importance; each character is only important as it proves to be in any particular instance in defining a group which has been perceived on the basis of all the available evidence. Experience shows us that some characters are much more stable and thus more likely to be important than others, and that there are many essentially unidirectional evolutionary trends...."

Morphology is the outward expression of genes. Since genes or chromo-

somes are biochemical in nature, the study of the chemistry of plants is just another method of investigation. But by no means should this line of research be emphasized to the exclusion of the others. This thought is echoed by the words of McNair (1935) who stated that "plants can be classified chemically in accordance with the substances made by them. Such a chemical classification may be compared with and used as a supplement to morphological classification and may be of some importance in the development of the true natural system of angiosperm phylogeny."

There is a great impetus in this field of research, but the development of the use of biochemistry in taxonomy has been a gradual one.

#### B. Development of chemotaxonomy

Chemotaxonomy is essentially the investigation of chemical compounds or groups of biosynthetically-related compounds, in a series of related, or supposedly-related plants (Erdtman, 1963).

Nehemiah Grew was perhaps the first to state that plants have things in common in "An idea of a phytological history propounded" (1673). James (1699) Petiver/wrote "Some attempts made to prove that Herbs of the same MAKE or CLASS for the generality, have the like Virtue and Tendency to work the same Effects."

de Candolle believed Rudolph Jacob Camerarius to be the first to clearly to express the connection between forms of plants and their properties (Gibbs, 1963). Hegnauer (1958), in turn, noted that de Candolle paid much attention to the chemical properties of plants as correlated to their morphological characters. For example, "all Convolvulaceae were laxative" and all Pinus species produced terpenes.

Another pioneer in this field, Helen C. de S. Abbott proposed in "Certain Chemical Constituents of Plants considered in Relation to Their Morphology and

Evolution" that plant chemistry be utilized in establishing phylogenetic relationships. She ~~exsaid~~ said: "There has been comparatively little study of chemical principles of plants from a purely botanical view. It promises to become a new field of research." This was in papers written from 1886 to 1887.

As early as 1891, Greshoff suggested the use of chemistry in taxonomy. He (1909) suggested that a "chemical description" be part of a formal description of a new genus. He also found the alkaloid laurotetanine to be common in the Lauraceae. At Kew, he looked for tannins, alkaloids, cyanogenetic substances, and saponins, in plants.

van Romburgh (1899) studied the occurrence of HCN, methyl salicylate, and acetone in plants, while Treub (1907) studied the HCN role. Greshoff (1909) emphasized the presence of HCN in plants, and vividly described it in Platanus:

"Indeed, in the ordinary plane-tree of the London streets (*P. acerifolia*), there is so much hydrocyanic acid present that the amount from every London plane-leaf would be enough to kill a London sparrow."

A school was set up at the turn of the century, when Nuttall published his significant paper on the use of essentially serological methods in establishing relationships. It gained momentum in the 1920's. Mez was a dominant figure in this area. He set up a group at Königsberg, Germany.. The work resulted in "Serodiagnostische Stammbaum", a phylogenetic tree derived almost in entirety by comparative serological methods (Mez and Ziegenspeck, 1926).

This is the basis of the serological methods: when antigens or foreign bodies are injected into the host, they elicit the formation of antibodies which agglutinate or otherwise affect the foreign object. Animals such as rabbits and sheep are used. In the precipitin reaction, aliquots of antigen are mixed in varying dilutions with antibody preparations (anti-serum), thus

producing a measurable precipitate corresponding to the "strength" of the reaction.

The validity of serology depends on its reliability. Seeds are used as the source of antigen. They are ground and extracted with petroleum-ether to remove lipids. The protein concentration is important.

Chester (1937) should be consulted for further information on serology.

Many constituents are looked for in plants today. The ten-plus methods used by the author and others are useful for preliminary surveys of the biochemistry of plants.

One method widely-used is that of paper chromatography. There is a vast amount of literature on this. The pioneers of this field were Day and Tswett. Others provided the impetus. Today this method is extensively used. Alston and Turner (1963) use it for amino acid separation, while the author used it for the separation and subsequent identification of phenolic compounds of leaves. Shaw (1961) used it to identify sedoheptulose and d-glucitol.

Gas chromatography is also being expanded in its use.

The outlook for chemotaxonomy appears bright. More and more compounds are being looked for, as more refined techniques are being employed, and as interest is being aroused.

## REVIEW OF LITERATURE

### A. The order "Geraniales"

There have been many different classifications of the "Geraniales". The number of families which have been included in it have varied from as little as four to more than twenty. Thus one can see that there is much disagreement as to which families this order should contain. In addition, there is the problem of raising genera to the familial rank and families to the ordinal rank.

According to the 11th "Syllabus" of Engler and Diels (1936), the "Geraniales" is classified as follows:

Suborder Geraniineae: Oxalidaceae

Geraniaceae

Tropaeolaceae

Linaceae (including Humiriaceae)

Erythroxylaceae

Zygophyllaceae

Cneoraceae

Rutaceae

Simarubaceae

Burseraceae

Meliaceae

Akaniaceae

Suborder Malpighiineae: Malpighiaceae

Trigoniaceae

Vochysiaceae

Suborder Polygalineae: Tremandraceae

Polygalaceae

Suborder Dichapetalineae: Dichapetalaceae

Suborder Tricoccae: Euphorbiaceae

Daphniphyllaceae

Suborder Callitrichineae: Callitrichaceae

The 12th "Syllabus" (1964) splits the "Geraniales" into two principle orders, the "Geraniales" sensu stricto and the Rutales. Other families are distributed in different orders. The "Geraniales" s.s. is divided as follows:

Suborder Limnanthineae: Limnanthaceae (2/8)

Suborder Geraniineae: Oxalidaceae (8/950)

Geraniaceae (11/780)

Tropaeolaceae (1/80)

Zygophyllaceae (30/250)

Linaceae (including Humiriaceae) (4/200)

Erythroxylaceae (including Nectaropetalaceae)  
(4/200)

Suborder Euphorbiineae: Euphorbiaceae (290/7500)

Daphniphyllaceae (1/35)

The Rutales, on the other hand, includes the following:

Suborder Rutineae: Rutaceae (150/1600)

Cneoraceae (2/3)

Simarubaceae (24/100)

Picrodendraceae (from Simarubaceae) (1/3)

Burseraceae (20/600)

Meliaceae (50/1400)

Akaniaceae (1/1)



Suborder Malpighiineae: Malpighiaceae (63/800)

Trigoniaceae (4/35)

Vochysiaceae (6/200)

Suborder Polygalineae: Tremandraceae (3/30)

Polygalaceae (13/800)

Dichapetalaceae (Chailletiaceae) (4/850) has been placed between Penaceae and Thymelaeaceae in Thymelaeales, while Callitrichaceae (1/44) has been placed in the suborder Verbenineae of order 8 of the Sympetales, the Tubiflorae, between Verbenaceae and Labiatae.

The numbers in parenthesis stand for the number of genera over the number of species. Thus, just in these 21 families, there are over 686 genera and 15,760 species.

Hutchinson (1959) included Geraniaceae, Limnanthaceae, Oxalidaceae, and Tropaeolaceae in the "Geraniales". This order was placed on the "herbaceous" side. In addition, he added Balsaminaceae to this order. He considered these plants as advanced from the Caryophyllales or direct from the Ranales. He considered that there was a "considerable gap" between these groups, and Limnanthaceae was the link between them.

Hutchinson placed the other families of the "Geraniales" s.g. of the 12th "Syllabus" on the woody side. He placed Humiriaceae, Linaceae, Erythroxylaceae, and Zygophyllaceae in the Malpighiales. He placed these families in the order with Malpighiaceae, Irvingiaceae (from Simarubaceae), Huaceae, Ledocarpaceae (including Ledocarpon, Wendtia, and Rhynchotheca of the Geraniaceae), Ctenolophonaceae (from Linaceae), Balanitaceae (from Zygophyllaceae), and Lepidobotryaceae (from Oxalidaceae).

Euphorbiaceae was raised to the ordinal rank, with roots derived from several stocks: Bixales, Tiliales, Malvales, Celastrales, and perhaps

Sapindales. He considered it to be closely allied with Tiliaceae and especially Sterculiaceae. He calls this family a "rubbish heap" of apetalous flowers which never arose from the Geraniales.

Hutchinson placed Daphniphyllaceae in the Hamamelidales, near Buxaceae and Bruniaceae.

Both Euphorbiaceae and Daphniphyllaceae are on the Lignosae side.

Hutchinson took Viviania from the Geraniaceae and raised it to the familial rank and placed it in the Pittosporales. Averrhoa of the Oxalidaceae was likewise raised to the familial rank and placed in the Rutales.

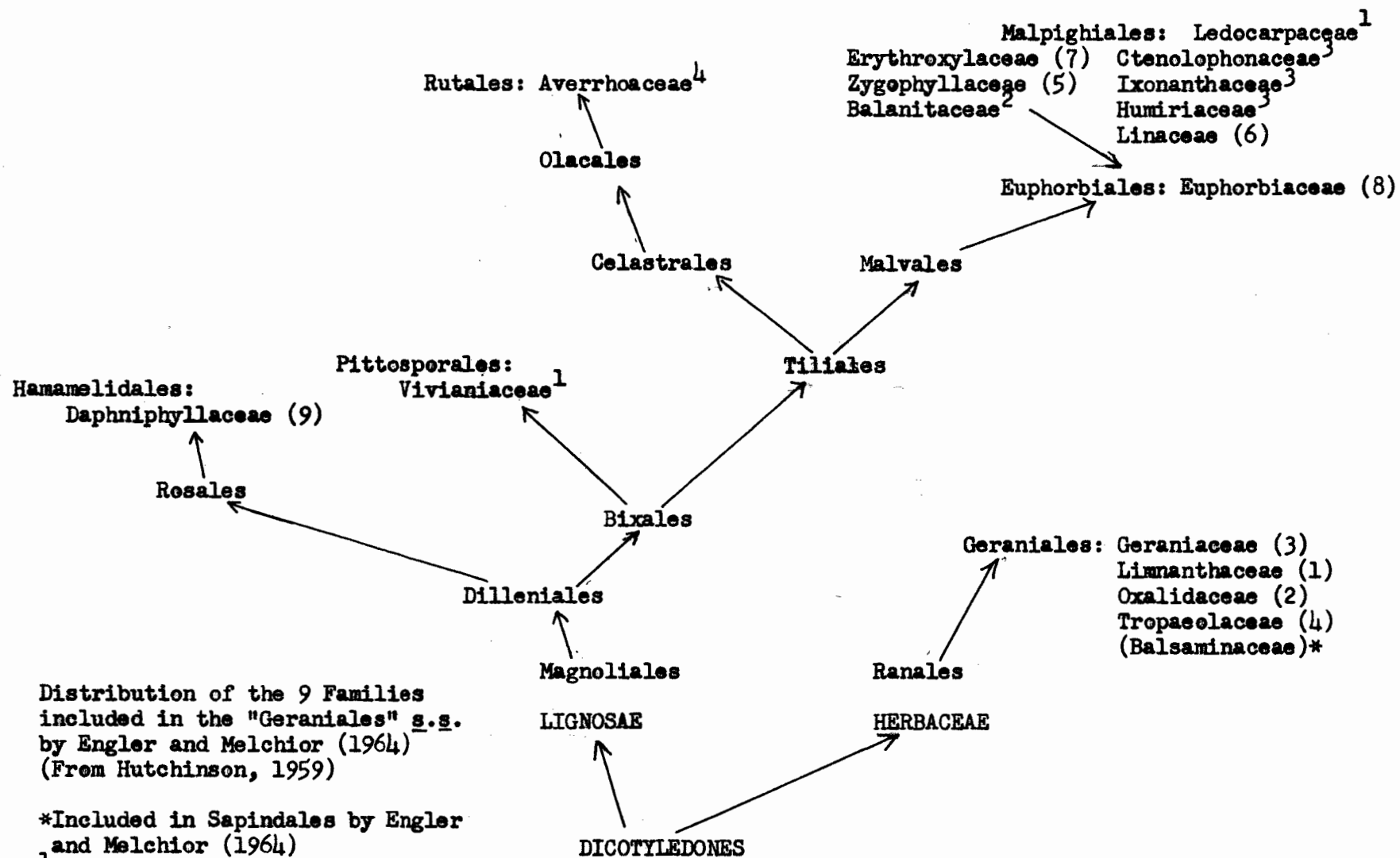
The other families of the 11th "Syllabus" (1936) were placed in the Polygalales, Rutales, Meliales, Sapindales, and Rosales, on the "woody" side, and Lythrales, on the "herbaceous" side. These orders will be discussed further in the next section.

The diagram on the following page gives the distributional pattern of the families of the "Geraniales" s.s. as devised by Hutchinson.

Another systematist, Gundersen (1950), divided the dicotyledons into different groups or complexes which were subdivided into orders. He formed a Geranium group, placed between the Malva group and the Dianthiflorae. The orders, in sequence, of the Geranium group consist of the Rutales, Juglandales, Sapindales, Celastrales, and Geraniales. Included in the Geraniales are Zygophyllaceae, Oxalidaceae, Geraniaceae, Tropaeolaceae, Limnanthaceae, Balsaminaceae, and Linaceae. Erythroxylaceae was made into a subfamily Erythroxylloideae of the Linaceae.

Euphorbiaceae (including Daphniphyllum) was raised to the ordinal rank and placed in the Malva complex.

Pulle (1950) devised this line of evolution: Reeks Rosales → Pandales → Malvales → Tricoccae → Geraniales → (Malpighiales) → (Polygalae) → Rutales → Sapindales → (Balsaminales) → Rhamnales



Distribution of the 9 Families included in the "Geraniales" s.s. by Engler and Melchior (1964) (From Hutchinson, 1959)

\*Included in Sapindales by Engler and Melchior (1964)

<sup>1</sup>Included in Geraniaceae

<sup>2</sup>Included in Zygophyllaceae

<sup>3</sup>Included in Linaceae

<sup>4</sup>Included in Oxalidaceae

"HYPOTHETICAL PROANGIOSPERMS"

The Geraniales consisted of Erythroxylaceae, Humiriaceae, ~~Linaceae~~, Dichapetalaceae, Oxalidaceae, Geraniaceae, ~~Limnanthaceae~~, Tropaeolaceae, Zygophyllaceae and Daphniphyllaceae.

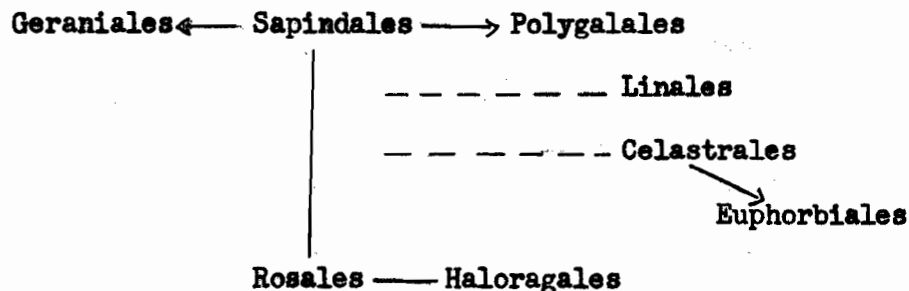
Again Euphorbiaceae was raised to the ordinal rank. The order was thus called Tricoccae.

The Geraniales is order 9 of Rendle's (1952) classification. He considered it to be allied to the Malvales, and placed in it seven families: Geraniaceae, Oxalidaceae, Balsaminaceae, Tropaeolaceae, Linaceae, Zygophyllaceae, and Malpighiaceae.

Euphorbiaceae (including the ~~tribe Daphniphyllae~~) was placed in the Tricoccae.

Cronquist (1957) placed four families in the Geraniales: Oxalidaceae, Geraniaceae, Limnanthaceae, and Tropaeolaceae. He raised Linaceae to the ordinal rank, and placed Humiriaceae, Erythroxylaceae, and Linaceae, in it. The Euphorbiales consisted of Euphorbiaceae and Daphniphyllaceae, in addition to Buxaceae, Aextoxicaceae, Pandaceae, and Didieraceae. Zygophyllaceae was placed in the Sapindales.

This was his arrangement of the relationships between these orders:



He believed that the Geraniales was an offshoot of the Sapindales, with a tendency towards simple leaves. In addition, the Euphorbiales is probably related to and derived from the Celastrales. The possibility of an unnatural polyphyletic Euphorbiaceae, he believed, was yet to be demonstrated.

Chadefaud and Emberger (1960) divided their orders into phyla.. One phylum or lineage consists of two orders, the Geraniales (Gruinales) and Malvales. These two orders, according to the authors, have striking resemblances to each other, thus:

"Il est donc impossible de séparer Gruinales et Malvales; leur souche est, selon toute vraisemblance, commune, et, remontant plus haut dans le passé, ces 2 ordres ont la même ascendances que les autres phylums du groupe IV, notamment ceux des Terebinthales - Umbelliflores, etc. et des Tricoques."

Their Geraniales consists of the following families: Zygophyllaceae, Geraniaceae, Tropaeolaceae, Oxalidaceae, Linaceae, Lepidobotryaceae, Humiriaceae, Erythroxylaceae, Malpighiaceae, and Limnanthaceae. Geraniaceae, Tropaeolaceae, and Oxalidaceae form a complex, with Limnanthaceae a little more evolved. Linaceae, Lepidobotryaceae, Humiriaceae, and Erythroxylaceae form another complex, with Malpighiaceae standing apart. Zygophyllaceae is a little more isolated from the rest of the families.

Chadefaud and Emberger placed Euphorbiaceae and Daphniphyllaceae in the Tricoques. They stated that there apparently is an affinity between this order and the Geraniales-Malvales complex.

Bessey (1915) placed the Geraniales near the Malvales and placed 22 families in it: Geraniaceae, Oxalidaceae, Tropaeolaceae, Balsaminaceae, Limnanthaceae, Linaceae, Humiriaceae, Erythroxylaceae, Zygophyllaceae, Cneoraceae, Rutaceae, Simarubaceae, Burseraceae, Meliaceae, Malpighiaceae, Trigoniaceae, Vochysiaceae, Polygalaceae, Tremandraceae, Dichapetalaceae, Euphorbiaceae, and Callitrichaceae.

van Tieghem and Constantin (1918) made the Geraniales alliance 8 of the Ranunculineae, with Geraniaceae as the transition towards the Malvales. The order included 31 families: Peganaceae, Nitrariaceae, Strasburgeriaceae, Elatinaceae, Cunoniaceae, Anacardiaceae, Burseraceae, Sapindaceae, Aesculaceae,

Melanthaceae, Aceraceae, Francoaceae, Stachyuraceae, Koeberliniaceae, Buxaceae, Meliaceae, Corynocarpaceae, Malpighiaceae, Polygalaceae, Caryophyllaceae, Portulacaceae, Rutaceae, Coriariaceae, Crassulaceae, Simarubaceae, Irvingiaceae, Leguminosae, Connaraceae, Rosaceae, and Crossosomaceae.

Skottsberg (1940, 1955) classified the Gruinales as follows:

Suborder Geraniineae: Oxalidaceae

Linaceae

Erythroxylaceae

Geraniaceae

Tropaeolaceae

Zygophyllaceae

Suborder Malpighiineae: Malpighiaceae

Trigoniaceae

Vochysiaceae

Tremandraceae

Benson (1957) has had an order Geraniales. It belongs to the "Thalamiflorae", and includes Limnanthaceae, Linaceae, Oxalidaceae, Tropaeolaceae, Balsaminaceae, Elatinaceae, Geraniaceae, Erythroxylaceae, Zygophyllaceae, and Malpighiaceae.

Perhaps one of the earliest orders formed with Geraniaceae as the type family was the Gruinales of Martius (1835). Gruinales was cohort 28, and included Balsamineae A. Rich., Oxalideae DC., Geraniaceae Juss., and Lineae DC.

Another systematist, Wettstein (1935), included the following families in the Gruinales: Linaceae, Humiriaceae, Oxalidaceae, Geraniaceae, Limnanthaceae, Tropaeolaceae, Erythroxylaceae, Malpighiaceae, Zygophyllaceae, and Cneoraceae.

Boivin (1956) made the Geraniales order 5 (XXX) of the Disciflorae, and placed in it Linaceae, Humiriaceae, Lepidobotryaceae, Erythroxylaceae, Zygophyllaceae, Cneoraceae, Oxalidaceae, Geraniaceae, Tropaeolaceae, Balsaminaceae, and Limnanthaceae.

Hallier (1912) placed the following families in the Gruinales: Oxalidaceae, Geraniaceae, Balsaminaceae, and Zygophyllaceae. Limnantheae, Tropaeoleae, and Balsamineae are in Balsaminaceae. In a previous classification, the order also included Linaceae and Humiriaceae (Hallier, 1908).

#### B. Families of the "Geraniales"

1. Geraniaceae. Geranium (Tourn.) L. is the type genus of this family. The members of this family are mostly herbaceous, occurring chiefly in the temperate region. Some are succulent and shrubby. The genera are homogeneous in structure. According to Metcalf and Chalk (1950), they form a homogeneous group except for Sarcocaulon which has modified structures adapted to arid conditions.

Chaudefaud and Emberger (1960) have placed 12 genera and more than 800 species in this family, the type family of their Geraniales.

Heimsch (1942) pointed out the similarity of the wood with that of Oxalidaceae, particularly in the tendency towards the elimination of the rays, scanty paratracheal parenchyma, and the common occurrence of septate fibres.

Bentham and Hooker (1862 - 1883) included Impatiens and Limnanthes in this family. Limnanthaceae and Geraniaceae both have members with widely-spaced vascular bundles, but there is no trace of the characteristic mechanical ring in the pericycle of Limnanthaceae. Thus a separation would be warranted.

Bentham and Hooker also included members of the Oxalidaceae in this family. The well-developed sclerenchymous ring in the pericycle and the ring of separate collateral bundles in the stem and petiole unite the two families.

Also included in the Geraniaceae by these authors was Tropaeolum, the only similarity between it and the Geraniaceae being the arrangement of vascular bundles in the stem and petiole.

Hutchinson (1948) tried to show that perhaps the Geraniaceae was descended from the Caryophyllaceae; however, he felt that there was a greater possibility that it arose directly from Ranalian stock with intermediate connections having disappeared. The general habit and leaf character of the Geraniaceae are largely like the Ranunculaceae and not of the Caryophyllaceae. Hutchinson (1959) made it the type of the Geraniales in the Herbaceae. He argued that some feel the Geraniaceae should be allied with the Malvaceae, but he thought the fibrous stems supported the origin of the Malvaceae on the woody side. Chadeaud and Emberger (1960) were two systematists who felt that the Geraniales, therefore Geraniaceae, should be allied with the Malvales.

Hutchinson (1959) removed Balbisia (Ledocarpon), Wendtia, and Rhynchotheca from the Geraniaceae of Bentham and Hooker (1862-1883) and also Harms (1931) and created the family Ledocarpaceae, placed near the Huaceae and Erythroxylaceae in the Malpighiales. Furthermore Viviania Cav. was raised to the familial rank by him and placed in the Pittosporales on the woody side.

Many other authors have created an order Geraniales or Gruinales; in addition, they have placed the Geraniaceae near the following families: Oxalidaceae, Tropaeolaceae, Linaceae, Erythroxylaceae, and Zygophyllaceae. (Martius, 1835; Bessey, 1915; Skottsberg, 1940, 1955; Chadeaud and Emberger, 1960; and others) In the 12th "Syllabus" (Engler and Melchior, 1964), this family is placed in the suborder Geraniineae between Oxalidaceae and Tropaeolaceae.

The last complete classification of the Geraniaceae was done by Harms in 1931:



Subfamily Geranieae: Geranium L.

Erodium L'Hér.

Monsonia L. Mant.

Sarcocaulon (DC.) Sweet

Pelargonium L'Hér.

Subfamily Biebersteineae: Biebersteinia Steph. ex Fischer

Subfamily Wendtieae: Rhynchotheca Ruiz et Pav.

Wendtia Meyen

Balbisia Cav.

Subfamily Vivianieae: Viviania Cav.

Subfamily Dirachmeae: Dirachma Schweinf.

2. Oxalidaceae. Oxalis L. is the type of this family which most authors place in the Geraniales near Geraniaceae (Warming, 1895; Small, 1907; Gundersen, 1950; Pulle, 1950; Rendle, 1952; Cronquist, 1957; Engler and Melchior, 1964). The family differs from the Geraniaceae in the ten stamens united at the base and the five free styles (Rendle, 1952).

Oxalidaceae was placed in the Geraniaceae by Bentham and Hooker (1862-1883). Heimsch (1942), however, says this is a homogeneous group, and thus should be delimited as a family. In addition, on the basis of wood anatomy, Sarcotheca should be placed here rather than in Linaceae.

Hutchinson (1959) raised Averrhoa L. to the familial rank. Then the newly-created family Averrhoaceae Hutch. was placed by him in the Rutales on the woody side. The two species, A. bilimbi L. and A. carambola L., are thus in the Lignosae.

Lepidobotrys staudtii, placed by Engler and Melchior (1964) in this family, is sometimes placed in the Linaceae (Chadefaud and Emberger, 1960). Hutchinson (1959) removed this genus, and along with Sarcotheca and Dapania,

created a family Lepidobotryaceae which he placed in the Malpighiales.

Chadefaud and Emberger (1960) detached this genus Lepidobotrys from both the Linaceae and Oxalidaceae. They stated that the move was justified, for Lepidobotrys differs from both families by the jointed unifoliate leaves, the catkin-like inflorescences, the unisexual flowers, the tri-carpellate trilobulate ovary, and the three styles united except for the stigma. This family was thus placed in the Geraniales, intermediate between Linaceae, Erythroxylaceae, and Oxalidaceae.

3. Tropaeolaceae. Tropaeolum L. is the type genus for this family which was placed in the Geraniaceae by Bentham and Hooker (1862-1883). Most authors maintain such a family (Warming, 1895; Small, 1907; Gundersen, 1950; Pulle, 1950; Rendle, 1952; Cronquist, 1957; Hutchinson, 1959; Chadefaud and Emberger, 1960). While the stem and petiole anatomy is similar to that of the Geraniaceae, they differ in that Tropaeolum contains myrosin cells (Metcalf and Chalk, 1950).

This family has been placed near Limnanthaceae, Zygophyllaceae, Geraniaceae, or Balsaminaceae (Warming, 1895; Pulle, 1950; Cronquist, 1957). Buchenau (1902) stated that the "form of leaves of the ovary, also the embryo development" justify familial rank near the Geraniaceae, perhaps near Hippocastanaceae. Farenholtz (1931), on the other hand, stated that the family is highly-specialized and isolated; the stamens and carpels suggest Sapindaceae.

4. Zygophyllaceae. Zygophyllum L. and the other members of the Zygophyllaceae were placed in the Geraniales by Engler and Diels (1936) and Rendle (1952). This family was retained in the Geraniales in the 12th "Syllabus" (1964).

Chadefaud and Emberger (1960), Small (1907), and Pulle (1950), along with Gundersen (1950), are other systematists who include this family in the Gera-

niales.

Hutchinson (1959) placed this family in the Malpighiales near Balaniteaceae. Warming (1895) has this family in the Terebinthinae near Burseraceae and Simarubaceae, while Cronquist (1957) included this family in the Sapindales.

This is some disagreement between authors as to which genera should be included in this family. Some authors have removed Peganum L. from Zygophyllaceae. van Tieghem and Constantin (1918) set up a family Peganaceae that contained two genera, Malacocarpus and Peganum. Peganum is different from the other members of the family in its embryology, and in this respect, is said to be close to Helianthemum guttatum, Radiola linoides and Ruta graveolens (Chadefaud and Emberger, 1960).

Balanites Delile has also been raised to the familial rank (Hutchinson, 1959). Hutchinson places this family near Lepidobotryaceae and Zygophyllaceae in the Malpighiales. Bentham and Hooker (1862-1883) placed Balanites in the Simarubaceae. Scholz (1964), in the 12th "Syllabus", kept it in the Zygophyllaceae. It is, according to Hutchinson (1959), perhaps better to place it in neither family. Heimsch (1942) studied the wood of several members of the Zygophyllaceae and found the rays of this genus to differ from those of the others. Record (1921) added that the wood structure of this genus differs from Guaiacum, Porlieria, and Bulnesia, but if the other xylem characters are studied, this genus is nearer to the Zygophyllaceae than to the Simarubaceae.

van Tieghem and Constantin (1918) removed Nitraria from this family and raised the genus to the familial rank. Then they placed it, along with Polygalaceae, in the Geraniales; most authors include it in the Zygophyllaceae.

Heimsch (1942), on the basis of the tracheids in the xylem, believes that Zygophyllaceae should be placed with Polygalaceae, Trigoniaceae, Tremandraceae, Malpighiaceae, and Vochysiaceae, rather than with families which are

primarily composed of herbaceous families. Heimsch concludes that the woods of this family form a distinct and natural group that is highly-specialized.

5. Linaceae. Many authors have placed Linum (Tourn.) L. and the other members of the Linaceae in the Geraniales (Warming, 1895; Small, 1907; Engler and Diels, 1936; Gundersen, 1950; Pulle, 1950; Rendle, 1952; Chadeaud and Emberger, 1960; Engler and Melchior, 1964).

According to Metcalfe and Chalk (1950), the marked differences in the wood anatomy suggest that this group is unsound. The following discussion supports this statement.

Gundersen (1950) assigned Humiriaceae and Erythroxylaceae to the subfamily level and thus created the Humirioideae and Erythroxyloideae. Most authors have a family Erythroxylaceae; thus further discussion on its relationship to Linaceae will be found in that section. It is sufficient to say here that its wood anatomy differs considerably from the Linaceae. Humiriaceae will also be discussed under a separate title.

Many other genera of the Linaceae have been raised to the familial rank. Chadeaud and Emberger (1960) stated that this family has recently been divided into the Hugoniaceae, Nectaropetalaceae, Ctenolophonaceae, and Ixonanthaceae. Only the latter <sup>two</sup> will be discussed.

Ctenolophonaceae has been considered by several authors to be a member of the Olacales (Chadeaud and Emberger, 1960). Ridley (1922) placed the genus in the Olacaceae.

Winkler (1933) retained this genus in the Linaceae. Metcalfe and Chalk (1950), however, consider Ctenolophon Oliv. to be anatomically-primitive to the other members of the Linaceae.

Hallier (1921) moved this genus from Linaceae to Celastraceae, then to Ixonanthaceae. Exell and Mendonca (1951) placed this family between Linaceae

and Erythroxylaceae, while Hutchinson (1959) placed it in the Malpighiales between Erythroxylaceae and Malpighiaceae. Morphologically, anatomically, and palynologically, Ctenolophonaceae is related more to the Malpighiaceae than to any other family (Chadefaud and Emberger, 1960).

Heimsch (1942) concluded that the xylem structure of Linaceae and Ctenolophon is similar, but he notes that in this respect, it is closer to Humiriaceae. The 11th "Syllabus" (Engler and Diels, 1936) and the 12th "Syllabus" (Engler and Melchior, 1964) assign Ctenolophon to the subfamily Ctenolophonoideae of the Linaceae.

Hutchinson (1959), who split this family into many little families, raised Ixonanthes to the familial rank and placed it with Ochthocosmus (Phyllocosmus) in the Malpighiales. He also made Linaceae a member of the Malpighiales.

Cronquist (1957) raised Linaceae to the ordinal rank; in addition, he placed Humiriaceae and Erythroxylaceae, as families, in that order.

Hutchinson (1959), as stated earlier, placed Linaceae in the Malpighiales. Thus this family is located in the Lignosae. In 1948, he stated that Linaceae is a small but wide-spread and heterogenous group; he believed Linum to be descended from woody ancestors and should therefore be placed next to the Erythroxylaceae, following the Tiliales alliance of families. He had earlier considered it to be part of the Geraniales. The majority of Linum species, he added, are herbaceous, but some have woody woodstocks, and at least one species L. arboreum L. is a true shrub with a single woody and much-branched stem. Heimsch (1942) stated that there is no justification that non-woody members of the Linaceae were derived from woody members.

6. Humiriaceae. Bessey (1915) placed this family in the Geraniales. Pullé

(1950) also did this, as did Chadeaud and Emberger (1960). Warming (1895) placed this family in the Grinales near Limnathaceae. Cronquist (1957), on the other hand, placed it in the Linales.

As mentioned previously, several authors considered this group as a sub-family of the Linaceae (Engler and Diels, 1936; Engler and Melchior, 1964; Gundersen, 1950). Hutchinson (1959) placed this family in the Malpighiales.

Heimsch (1942), based on wood anatomy, stated that this is a homogenous family. On the basis of secondary xylem (tracheids), he added that it has the closest family affinity with Linaceae. Chadeaud and Emberger (1960) stated that this family is distinguished from Linaceae by the many stamens (ten to many) in several whorls and the intrastaminal disc which is cup-like or shell-like. In addition, they said that there may also be an affinity with Burseraceae, this based on the fruit structure. Metcalfe and Chalk (1950) noted that the idioblasts of Saccoglottis links the family with Theaceae.

7. Erythroxylaceae. Erythroxylum L. is the type genus of this family, the members of which are said to be closely allied to Linaceae. This family was placed in the Linaceae by Bentham and Hooker (1862-1883), Baillon (1878), and Hallier (1908, 1912, 1921). Gundersen (1950) also placed it in the sub-family Erythroxylaceae of Linaceae. Cronquist (1957), on the other hand, included this family in the Linales, between Humiriaceae and Linaceae.

Heimsch (1942) said that Erythroxylaceae and Linaceae differ greatly in their wood. Erythroxylaceae has members with scalariform vessel perforations. Wood types are more uniform in Erythroxylaceae. However, he concluded, its affinity with Linaceae is greater than with any other family.

Engler and Diels (1936) included this family in the Geraniales. It is retained there in the 12th "Syllabus" (Engler and Melchior, 1964). Pullé (1950) likewise placed this family in the Geraniales, as did Small (1907) and Chadeaud and Emberger (1960).

Hutchinson (1959) placed this family near Ledocarpaceae and Ctenolophonaceae in the Malpighiales. Warming (1895) has this family in Aesculinae, near Malpighiaceae and Vochysiaceae.

8. Limnanthaceae. This family contains two genera Limnanthes R. Br. and Floerkea Willd. Because of the ascending apotropous ovules and the binucleate pollen, they are often placed near the Sapindaceae; sometimes they are placed with the old "gamopétales" because of the uni-tegment and tenuinucellate ovule and the gynobasic style. (Chadefaud and Emberger, 1960)

Chadefaud and Emberger (1960) stated that many characters of the Geraniaceae are present in this family. Thus Bentham and Hooker (1862-1883) placed this family in the Geraniaceae because of the anatomical similarity between Floerkea and Geraniaceae. Most people place this family in the Geraniales (Warming, 1895; Small, 1907; Gundersen, 1950; Pulle, 1950; Hutchinson, 1959; Engler and Melchior, 1964). The family resembles the Geraniaceae, in one respect, in the presence of widely-spaced vascular bundles (Metcalfe and Chalk, 1950).

9. Euphorbiaceae. Euphorbia L. is the type genus of the Euphorbiaceae, a family which was first adequately delimited as a natural group of plants by A. L. de Jussieu in 1789 (Perry, 1943). Since then, there have been many contributions, phylogenetically, morphologically, and anatomically, to its classification.

This family consists mostly of tropical plants, each varied in habit. Even within a genus, there are varied habits. Euphorbia, for example, is a herbaceous plant in Britain; those of tropical Africa are cactus-like.

The members of this family have been placed in different orders by different authors. Several have raised this family to the ordinal rank (Small, 1933; Gundersen, 1950; Cronquist, 1957; Hutchinson, 1959).

It has also been placed in the Geraniales (Engler and Diels, 1936; Engler and Melchior, 1964). Still others have placed it in the Tricoccae or Tricoques (Warming, 1895; Pulle, 1950; Rendle, 1952; Chadeaud and Emberger, 1960).

Buxaceae and Daphniphyllaceae have been made subfamilies of this family. These families should be consulted for further discussion. Aextoxicon has also been included in this family. The family is discussed in another section.

Perry (1953) investigated a small per cent of genera of this family and concluded that they form a rather natural group although showing many lines of evolution. Cytologically, he added, certain tribes, or other taxonomical groups, appear to be natural in that one basic chromosome number is found in each, while in another group, nearly all the basic numbers present in the family are found, suggesting that the unit is merely descriptive and artificial and not phyletic. He found that within each of the genera studied there was considerable uniformity in the shape and size of the somatic chromosome, with the exception of the Euphorbia. Here the cytological complexity parallels the well-known morphological and taxonomical complexity.

Euphorbiaceae is a heterogenous family, more or less artificial (Chadeaud and Emberger, 1960). Chadeaud and Emberger agree with Perry that there is evidence of a polyphyletic origin. Again they cite the extreme diversity in morphology. Metcalfe and Chalk (1950) also reached the conclusion that this family is polyphyletic in origin. They found no anatomical homogeneity. They add that the petiolar structure supports an affinity with Malvaceae and Tiliaceae. There is also some support for its affinity with Buxaceae.

Janssonius (1906-1936) concluded from his studies of the woods of Acalypha, Antidesma, Bischofia, Bridelia, and Glochidion, that there is some relation-



ship to Burseraceae. Metcalfe and Chalk (1950) also found some resemblance between Euphorbiaceae and Apocynaceae.

On this matter of polyphyletic origin, Hutchinson (1959) adds that this family has some relationships with Rhamnales, Celastrales, Sterculiaceae, and Malvaceae. Willis (1960) remarked that this family is closely related to the Geraniales in the structure of the gynoecium, although separated a good deal from other families of this order by the amount of reduction in most of the flowers.

This order, the Euphorbiales, has often been placed near the Geraniales and Malvales (Gundersen, 1950; Pulle, 1950). But Hutchinson (1959) believes that it never arose from the Geraniales. Gundersen (1950), on the other hand, concluded that the Euphorbiaceae consists of members reduced in form, probably derived from the Geraniales and Malvales; in addition, the family is not very near to the Sapindales.

Euphorbia is an exceptional one in the family. Thus something will be said about it. It is a large and widely-spread genus with an advanced inflorescence composed of an involucre of connate bracts, between which are glands simulating small petals. Within the involucre are male and female flowers, the male flowers usually numerous and looking like stamens, the female flowers solitary and placed in the center, much resembling the ovary of the bisexual flower. (Hutchinson, 1959)

10. Daphniphyllaceae. This family has often been associated with Euphorbiaceae. In 1869, Daphniphyllum Blume was raised to the familial rank. In the 11th "Syllabus" (Engler and Diels, 1936) and 12th "Syllabus" (Engler and Melchior, 1964), this family is placed near the Euphorbiaceae. Rosenthal (1931) placed this genus near the Euphorbiaceae on account of the ovary, but separated on the basis of the small apical embryo and the ventral raphe. She was opposed to Hallier's view that Daphniphyllum should be near the Hamamelidaceae.

Gundersen (1950) included this genus in the Euphorbiaceae, while Chade-faud and Emberger (1960) placed this family in the Tricoques between Euphorbiaceae and Dichapetalaceae. Daphniphyllaceae is included in the Geraniales of Pulle's (1950), near Zygophyllaceae. Metcalf and Chalk (1950) placed this family after Buxaceae, stating that the wood is different from Euphorbiaceae's.

Hutchinson (1959) placed this family near the family Bruniaceae and Buxaceae in the Hamamelidales. Takhtajan (1954) also did this.

Whether or not it should be included in the Geraniales, Euphorbiales, or Hamamelidales, Janssonius (1929) stated that there was sufficient evidence from a study of the wood anatomy to form a new family. He added that it would have wide affinities with Theaceae (s.l.) and Hamamelidaceae.

11. Rutaceae. Ruta L. is the type of this family. A. L. de Jussieu, in 1789, delimited such a family. Rutaceae was included in the Geraniales in the 11th "Syllabus" (Engler and Diels, 1936), but it was made the type family of an order Rurales in the 12th "Syllabus" (Engler and Melchior, 1964). While Small (1907) included this family in the Geraniales, many authors (Gundersen, 1950; Pulle, 1950; Rendle, 1952; Hutchinson, 1959) include this family in the Rurales. It has also been made a member of the Sapindales (Cronquist, 1957), the Terebinthinae (Warming, 1895), and the Terebinthales sensu stricto (Chade-faud and Emberger, 1960).

Members of the Rutaceae are found in both temperate and warm regions.

The Rutaceae offers a diversity in morphological characteristics, such as the staminal corolla of Xanthoxylum, the polymorphic flower of Fagara, the pseudogynophore of Boenninghausia, the inflorescence of Diplolaena, the hypophyllic flowers of Erythrochiton, the calyx with very unequal sepals in Moniera, the stipulate stamens of Dictyoloma, and the androecia, nature of spines, and origin of <sup>the</sup> simple leaves of Citrus (Chade-faud and Emberger, 1960).

On the whole, the wood anatomy is very uniform and highly-specialized (Metcalf and Chalk, 1950). Heimsch (1942) considers the Rutaceae, Simarubaceae, Meliaceae, Sapindaceae, Burseraceae, and Anacardiaceae a natural group, with Rutaceae apart due to the lack of septate fibres, but nearest to Simarubaceae.

Flindersia R. Br., with 20 species, has sometimes been considered a member of the Meliaceae (Chadefaud and Emberger, 1960). Because of the homogenous rays and the non-septate fibres, Dadswell (1935) and Harrar (1937) believe it to be out of place in Meliaceae. The genus, however, is variable. Harrar suggests a separate family should be formed. Dadswell agrees. But Metcalfe and Chalk (1950) believe that it should remain with Rutaceae because of the secretory cavities and cells in the tissues other than the secondary xylem.

Kribs (1930) studied Ptaeroxylon and Chloroxylon and concluded that they should remain in Rutaceae. Chloroxylon has been placed in Meliaceae, while Aitonia and Ptaeroxylon have been placed in Meliaceae and Sapindaceae (Metcalf and Chalk, 1950). Heimsch (1942), on the basis of wood anatomy, concluded that Rhabdodendron does not belong in this family.

12. Cneoraceae. Cneoraceae, with Cneorum L. as the type, is a family of doubtful position. According to the 11th "Syllabus" (Engler and Diels, 1936), it is a part of the Geraniales. But in the 12th "Syllabus" (Engler and Melchior, 1964), it was moved to the newly-created Rutales.

Bessey (1915) made this family a member of the Geraniales. The nucleated albumen, the trinucleate pollen, and the ovary, are like the Geraniales (Chadefaud and Emberger, 1960). Engler (1931) stated that the carpels are somewhat like Zygophyllaceae, but distinguished by the single stamen whorl, the absence of stipules, and the presence of oil cells. This, he added, is

somewhat distinct from the other members of the Geraniales.

Gundersen (1950) placed this family near Coriariaceae and Simarubaceae in the order Rutales, while Bentham and Hooker (1862-1883) included it in the family Simarubaceae.

Chadefaud and Emberger (1960) raised this family to the ordinal rank, and made it the only family of the Cneorales. Hutchinson (1959) assigned this family to the Celastrales, near Koeberliniaceae and Cardiopteridaceae. Pulle (1950) and Cronquist (1957) both placed this taxon in the Sapindales.

13. Simarubaceae. Simaruba L. and the other members of this family are found in warmer regions of the world. They are usually trees or shrubs with bitter principles in the bark and wood (Rendle, 1952).

Engler and Diels (1936) placed this family in the Geraniales, as did Small (1907). In the 12th "Syllabus" (Engler and Melchior, 1964), it was moved to the Rutales, near Cneoraceae and Picrodendraceae.

Many authors have also assigned this family to the Rutales (Gundersen, 1950; Pulle, 1950; Rendle, 1952; Hutchinson, 1959). Cronquist included this family in the Sapindales, along with Akaniaceae, Burseraceae, Cneoraceae, Rutaceae, Meliaceae, and Zygophyllaceae.

This family has also been assigned to the Terebinthinae near Zygophyllaceae and Ochnaceae (Warming, 1895), and Terebinthales (Chadefaud and Emberger, 1960). These are essentially rutaceous orders. Chadefaud and Emberger (1960) stated that the simple leaves of this family have the same phylogenetic origin as the Rutaceae-Aurantioideae.

The family is divided into six subfamilies: Surianoideae, Simarouboideae, Kirkioideae, Irvingioideae, Picramnioideae, and Alvaradoideae, in the 12th Syllabus (Engler and Melchior, 1964). Metcalfe and Chalk (1950), in an earlier publication, stated that there are few anatomical characters common

to the group. Thus Jadin (1912) had earlier separated Irvingia from the family. In addition, he believed Suriana should be made into a family with affinities with Geraniaceae.

Metcalf and Chalk (1950) stated that three subfamilies, Kirkioideae, Irvingioideae, and Alvaradoideae, form distinct homogeneous groups. Surianoideae shows some divergence, while Simarouboideae is varied. Jadin (1912) removed Holacantha from Simarouboideae and created the family Holacanthaceae.

Webber (1936), however, said that the wood structure of Kirkioideae, Irvingioideae, Picramnoideae, and Alvaradoideae, show the group to be a natural one. In addition, the wood of Surianoideae is similar, but more heterogeneous. Simarouboideae is the more diversified group, especially in the xylem structure. Holacantha and Castela are widely-divergent from the other genera, and thus should be separated from Simaroubaceae. She also noted that the wood of Picrodendron, which Engler and Diels (1936) placed in the Simaroubaceae, also resembles the wood of Irvingioideae.

Hutchinson (1959) removed Irvingioideae Engl. from Simaroubaceae, and formed a family Irvingiaceae, which he placed in the Malpighiales, near Linaceae and Huaceae. van Tieghem and Constantin, as early as 1918, assigned a family Irvingiaceae to the Geraniales, near Simaroubaceae and Leguminosae. Small (1907) formed a family Surianaceae, which he placed near Rutaceae and Simaroubaceae in the Geraniales.

Thus, this family, obviously allied to Rutaceae and other members of that alliance, suffers from internal chaos.

14. Picrodendraceae. Picrodendron Planch. was raised to the familial rank in the 12th "Syllabus" (Engler and Melchior, 1964) and placed in the Rurales, near Simaroubaceae. It was formerly assigned to the Geraniales as part of

Simarubaceae. Metcalfe and Chalk (1950), in Anatomy of the Dicotyledons, stated that, anatomically, Picrodendron resembles Simarouboideae.

Record and Hess (1943) had a family Picrodendraceae. Hutchinson (1959) placed this family near Juglandaceae, in the Juglandales.

15. Burseraceae. Engler and Diels (1936) assigned this family to the Geraniales, but in the 12th "Syllabus" (Engler and Melchior, 1964), it was moved to the Rutales.

Other authors have also made this family part of the Rutales, amongst them, Gundersen (1950), Pulle (1950), Rendle (1952), and Hutchinson (1959). Chadeaud and Emberger (1960) assigned it to their equivalent of the Rutales, that is, the Terebinthales.

Small (1907) placed this family between Simarubaceae and Meliaceae in the Geraniales, while Cronquist (1957) assigned it to the Sapindales, along with Simarubaceae, Akaniaceae, Cneoraceae, Rutaceae, Meliaceae, and Zygophyllaceae.

Guillaumin (1909-1910) stated that Burseraceae has affinities with Rutaceae, Anacardiaceae, Simarubaceae, Cneoraceae, Meliaceae, Coriariaceae, Sapindaceae, and Hippocastanaceae, but the most marked affinities are with Anacardiaceae, Meliaceae, Rutaceae, and Simarubaceae. Because of the wood structure, Webber (1941) and Heimsch (1942) also conclude that Rutaceae, Simarubaceae, Meliaceae, Sapindaceae, Burseraceae, and Anacardiaceae, form a natural group. Heimsch (1942) stated that from a study of the wood anatomy of 1000 species in 37 families including Burseraceae, Meliaceae, Sapindaceae, Rutaceae, Simarubaceae, and Anacardiaceae, he concludes that these families are better classified by Wettstein, Hutchinson, and especially Hallier.

Thus Hutchinson (1959) has placed Burseraceae and several of those families in different but related orders. Hallier (1908) united Burseraceae,

Anacardiaceae, and other families in the Terebinthaceae. Jadin (1894) also united these two families on the basis of the longitudinal secretory canals found in the phloem.

As stated earlier, Chadeaud and Emberger (1960) placed this family in the Terebinthales, with affinities with Rutaceae and Simarubaceae, but distinguished by certain anatomical features, such as the secretory schizogenous cortical canals, and the radical or fundamental phloem which is sometimes more medullary or pithy.

16. Meliaceae. Small (1907), Bessey (1915), and Engler and Diels (1936) assigned this family to the Geraniales. In the 12th "Syllabus" (Engler and Melchior, 1964), it was transferred to the Rutales.

This family has been placed in the Rutales by several authors (Gundersen, 1950; Pulle, 1950; Rendle, 1952). Warming (1895) placed this family near Connaraceae and Rutaceae in the Terebinthinae, while Chadeaud and Emberger (1960) assigned it to the Terebinthales.

Hutchinson (1959) raised the family to the ordinal level (Meliales), and included Cedrela and Flindersia in the family. (See Rutaceae for more discussion on this.)

Cronquist (1957) assigned this family to the Sapindales. Heimsch (1942) stated that although Meliaceae, Rutaceae, Simarubaceae, Sapindaceae, Burseraceae, and Anacardiaceae, form a more or less natural group, the occurrence of septate fibres links Sapindaceae with Meliaceae rather than with Rutaceae.

There is some disagreement as to which genera should be included in this family. As stated in the discussion on Rutaceae, Chloroxylon, Flindersia, Ptaeroxylon, and Aitonia, are sometimes placed in this family. Kribs (1930) feels the first three genera should remain in Rutaceae, but argues that Swietenioideae is an anomalous subfamily and should thus be raised to the fami-

lial rank on the basis of its anatomy and morphology. Janssonium (1906-1936) stated that marked differences between the woods of Cedrela and Melia and those of the other genera suggest that these genera, Cedrela and Melia, should be placed in separate families.

17. Akaniaceae. Stapf (1912) first proposed that this family should be formed and placed in the Sapindales. This genus had previously been placed in the Sapindaceae. According to Metcalfe and Chalk (1950), Akania hillii Hook. was described imperfectly by J. D. Hooker and was thus assigned to Sapindaceae.

Akaniaceae has been placed in the Sapindales by many authors (Gundersen, 1950; Pulle, 1950; Cronquist, 1957; Hutchinson, 1959). Radlkofer (1890) and Solereder (1908) placed it in Staphyleaceae. Hutchinson (1959), in The Families of Flowering Plants, placed this family Staphyleaceae near Akaniaceae.

Chadefaud and Emberger (1960), although placing this family in the Terebinthales, consider the affinities of this taxon unclear. Anatomically, by the very large rays and the absence of uniseriate rays in the secondary wood, the family differs clearly from Sapindaceae.

Engler and Diels (1936) placed this family in the Geraniales, this mainly because of the ovule. In the 12th "Syllabus" (1964), it is assigned to the Rutales.

18. Malpighiaceae. Malpighiaceae is a well-distributed tropical family. It was placed in the Geraniales by Engler and Diels (1936), but Engler and Melchior (1964) assigned it to the Rutales, in the subfamily Malpighiineae, near Trigoniaceae and Vochysiaceae.

Small (1907) placed this family in the Geraniales, near Zygophyllaceae and Rutaceae. Bessey (1915), Benson (1957), and Chadefaud and Emberger (1960) also placed this family in the Geraniales.



Heimsch (1942) described the wood of Malpighiaceae and stated that the more highly specialized wood dissociates this family from Linaceae, Humiriaceae, and Erythroxylaceae, families often placed in the Geraniales.

Many authors have raised this family to the ordinal rank (Pulle, 1950; Hutchinson, 1959). It has also been assigned to the Polygalales near Trigonaceae (Cronquist, 1957), Aesculinae near Aceraceae and Erythroxylaceae (Warming, 1895), and the Sapindales (Gundersen, 1950). Gundersen noted that the twisted seed of the Sapindaceae and other characters make Malpighiaceae distinct from that family.

van Tieghem and Constantin (1918) described the Malpighiaceae as "...une famille très homogène."

19. Trigonaceae. The systematic position of Trigon Aubl. and the other members of this family is still uncertain. Bentham and Hooker (1862-1883) assigned the genera to Vochysiaceae. Engler and Diels (1936) formed a family and placed it in the Geraniales. In the 12th "Syllabus", this family is included in the order Rutales.

Hutchinson (1959) made this a family of the Polygalales and placed it near Krameriaceae and Vochysiaceae, while Gundersen (1950) assigned Trigon to Tremandraceae. Pulle (1950) assigned this family to the Malpighiales near Vochysiaceae. Like Hutchinson, Cronquist (1957) placed the family near Vochysiaceae in the Polygalales. Warming (1895) assigned this family to Aesculinae, again near Vochysiaceae and Tremandraceae.

Thus, as one can see, the many authors assign this family to a position near Vochysiaceae. Metcalfe and Chalk (1950) described this family as being conspicuously different from Vochysiaceae in the absence of intraxylary phloem and in the presence of bordered pits in the ground tissue elements of the xylem.

Any position thusfar discussed may be correct, for Heimsch (1942) states that the wood anatomy suggests a relationship with Polygalaceae, Tremandraceae, Zygophyllaceae, Malpighiaceae, and Vochysiaceae.

20. Vochysiaceae. Vochysia Poir. is the type of this taxon. Heimsch (1942) states that this family is related to Polygalaceae, Trigoniaceae, Tremandraceae, Zygophyllaceae, and Malpighiaceae, but differs from them in the more pronounced development of banded parenchyma and the occurrence of intercellular canals.

Engler and Diels (1936) assigned this family to the Geraniales. In Syllabus der Pflanzenfamilien (Engler and Melchior, 1964), it was assigned to the Rutales, and thus placed near Malpighiaceae and Trigoniaceae.

Pulle (1950) made this family a member of the Malpighiales; Warming (1895) placed this family near Erythroxylaceae and Trigoniaceae in the Aesculinae. Chadeaud and Emberger (1960) described this family as one near Anacardiaceae in the Terebinthales.

Many authors prefer placing this family nearer to Polygalaceae. Thus Small (1924) assigned it to the Polygalales near Polygalaceae. Cronquist (1957) did likewise, but placed it near Trigoniaceae and Balsaminaceae. Hutchinson (1959) assigned this family to a position near Trigoniaceae in the Polygalales.

Gundersen (1950) placed this family near Malpighiaceae and Tremandraceae in the Sapindales.

21. Tremandraceae. This is a family of doubtful position. Tremandra R. Br. and the other genera seem isolated. According to Chadeaud and Emberger (1960), the family is presently placed near Polygalaceae, Pittosporaceae, Byblidaceae, or Sterculiaceae.

Engler and Diels (1936) assigned this family to the Geraniales, but it was subsequently moved to the Rutales, subfamily Polygalineae (Engler and Melchior, 1964). Pulle (1950) and Cronquist (1957) placed this family in the Polygalales.

Pritzel (1930) believed this family to be related to Pittosporaceae, as are perhaps Vochysiaceae and Polygalaceae. Thus Hutchinson (1959) placed this family in the Pittosporales, near Vivianiaceae which many authors assign to the Geraniaceae.

Chadefaud and Emberger (1960) assigned this family near Corynocarpaceae and Balsaminaceae in the Terebinthales. Warming (1895) made this family part of the Aesculinae, and placed it between Trigoniaceae and Polygalaceae.

Metcalf and Chalk (1950), after investigating this family, described it as one in which anatomical structures are uniform and thus do not aid in establishing affinities. Thus the relationships of this somewhat isolated group remain unestablished.

22. Polygalaceae. Polygala (Tourn.) L. is the type of this family which Engler and Diels (1936) assigned to a position near Tremandraceae in the subfamily Polygalineae of the Geraniales. This family was subsequently moved to the Rutales (Engler and Melchior, 1964).

Chadefaud and Emberger (1960) made this family a member of the Terebinthales, while Warming (1895) placed this family near Tremandraceae in the Aesculinae.

Many authors have raised this family to the ordinal rank and have thus named the order Polygalales (Small, 1924; Pulle, 1950; Cronquist, 1957; Hutchinson, 1959).

Other authors link this family with the sapindaceous type; subsequently this family has been placed in the Sapindales (Gundersen, 1950; Rendle, 1952).

But Rendle calls the Polygalaceae a family of doubtful position, the members of which differ from other members of the Sapindales by the presence of pendulous ovules with ventral raphe, as in the Geraniales.

Heimsch (1942) investigated some members of this family and found that the wood anatomy showed affinities with that of Trigoniaceae, Tremandraceae, Malpighiaceae, and Vochysiaceae. Although more specialized in wood structure, he found the family to have some relationship to Linaceae, Humiriaceae, and Erythroxylaceae.

Chodat (1891-1893) describes this family as "a very natural family, not closely allied with any others." The herbs, shrubs, and small trees, he adds, have distinct pollen grains. He remarks that this is the "surest mark of distinction in the family." The grains are ellipsoidal with coarse pitting at the poles and longitudinal bands broken in the center by an equatorial ring.

There are some genera which have been doubtfully placed here. Chodat noted that Krameria is not a member of Polygalaceae, but the type of a family near Leguminosae-Caesalpiniaceae (Krameriaceae). Le Maout and Decaisne (1873) assigned Krameria to Polygalaceae.

Gagnepain (in Chadeaud and Emberger, 1960) raised Xanthophyllum Roxb. to the familial rank (Xanthophyllaceae fam. nov. 1908, Gagnepain). Wettstein (1935) and Cronquist (1957) also maintained a family. Engler and Melchior (1964) left it in Polygalaceae.

According to Jauch (1918), Xanthophyllum differs in its wood parenchyma from the other members of Polygalaceae, but should remain in the family because of the floral anatomy and pollen grain structure. He also suggests a link between this small family and Anacardiaceae, this based on the occurrence of lysigenous canals.

Moutabea Aubl. differs from the classic members of Polygalaceae in the anatomy and appearance of the flowers, but the androecia, disc, and pollen

structure are like those of Polygalaceae (Chadefaud and Emberger, 1960).

Diclidanthera Mart. has a doubtful position in this family. Hallier (1921) considers this genus to be the type of a separate family. O'Donell (1941), on the basis of wood anatomy and pollen grain morphology, has retained the genus in Polygalaceae.

23. Dichapetalaceae. Dichapetalum Thon. (Chaillietia DC.) and the other members of this family were included in the Geraniales by Engler and Diels (1936); Pulle (1950) also included it in this order, and placed it near Linaceae and Oxalidaceae.

Engler and Melchior (1964) placed this family in the Thymelaeales. Hutchinson (1959) assigned it to a position near Rosaceae and Calycanthaceae of the Rosales. Cronquist (1957) included this family in the Euphorbiales, near Euphorbiaceae and Aextoxicaceae. Small (1924), on the other hand, included this family in the Polygalales.

While the affinities of Dichapetalaceae remain unestablished, Heimsch (1942) links this family with Malpighiaceae, Vochysiaceae, Tremandraceae, Polygalaceae, and Trigoniaceae; he considers this preferable to the Euphorbiaceae or Rosaceae link of Wettstein (1935) and Hutchinson (1926).

24. Callitrichaceae. This is a family of very doubtful position. Willis (1960) says this is not unusual in water plants. In addition, he states that Callitrichaceae has been placed near Caryophyllaceae, Verbenaceae, and Boraginaceae, but closest to Euphorbiaceae.

Engler and Diels (1936) assigned this family to the Geraniales. Bessey (1915) also did this. Engler and Melchior (1964) later moved the family to the sympetalous Tubiflorae.

Rendle (1952) placed it in the Tricoccae with Euphorbiaceae and Buxaceae; Warming (1895) also has it here. The affinity with Euphorbiaceae is in the

number and position of the ovules, and in the reduction of the number of flowers in Euphorbia.

Gundersen (1950) placed this family near Labiatae in the Boraginales. Schürhoff (1926) said this family should be assigned to Warming's order Nuculiferae, nearest to Labiatae and Boraginaceae.

While Brown, de Candolle, Hegelmaier, and Bentham and Hooker assigned this genus to the Haloragaceae (Rendle, 1952), Cronquist (1957) places this family doubtfully in the Haloragales. Hutchinson (1959) made this family a member of the Lythrales, alongside the Haloragidaceae.

Chadefaud and Emberger (1960) placed this family in the annex of the Myrtales between Hippuridaceae and Dialypetalanthaceae.

Pulle (1950) raised this family to the ordinal rank (Callitrichales).

25. Balsaminaceae. Impatiens Riv. ex L. was placed in the Geraniaceae by Bentham and Hooker (1862-1883). Others have made it a family of the Geraniales (Warming, 1895; Bessey, 1915; Benson, 1957; Hutchinson, 1959). Impatiens differs from the members of the Geraniaceae in the presence of raphide sacs, the absence of a ring of mechanical tissue in the pericycle, and the absence of bundles in the petiole (Metcalf and Chalk, 1950). Hutchinson, in 1948, said that on morphological grounds, this genus "seems to be with the Geraniaceae." Impatiens represents a complex type of development in its own alliance, that is to say, there are no plants on a higher plane which are related to it.

The resemblance to the Tropaeolaceae of the Geraniales is striking, but important characteristics distinguish the two groups, such as the tenuinucellate ovules (Chadefaud and Emberger, 1960).

Engler and Melchior (1964) assigned this family to the Sapindales. Chadefaud and Emberger placed it in the Terebinthales.

Cronquist (1957) included this family in the Polygalales near Vochysiaceae and Stackhousiaceae, while Pulle (1950) raised this family to the ordinal rank (Balsaminales).

26. Buxaceae. Buxus L. and the other members of this family were previously placed in the Euphorbiaceae, from which it is separated because the raphe of the pendulous ovule is turned away and not towards the axis of the ovary (Steward, 1958). Bentham and Hooker (1862-1883) considered this family a tribe of the Euphorbiaceae.

Others have placed this family in the Euphorbiales (Gundersen, 1950; Cronquist, 1957). This family was first described as the Buxineae by Loiseleur (1819). In 1853, Plee separated this family from the Euphorbiaceae on the grounds that the former lacked latex and had parietal placentation. The dehiscence of the fruit, the apotropous ovule, the absence of laticifers, and the embryology, separate the two families (Chadefaud and Emberger, 1960).

Buxaceae has been placed in the Tricoccae with Euphorbiaceae and Callitrichaceae (Rendle, 1952); it has also been placed near Hippocastanaceae and Aextoxicaceae in the Terebinthales (Chadefaud and Emberger, 1960), near Icacinaceae in the Celastrales (Pulle, 1950), and in the Sapindales (Engler and Diels, 1936; Engler and Melchior, 1964).

Although certain features are uniform throughout the family, Buxus, Simmondsia, and Styloceras differ remarkably in wood anatomy, more so than usual among genera of the same family (Metcalf and Chalk, 1950).

27. Aextoxicaceae. Aextoxicon punctatum Ruiz. et Pavon of Chili is the only member of this family. This genus was previously placed in the Euphorbiaceae (Bentham and Hooker, 1862-1883). The xylem is like the structurally-primitive Euphorbiaceae (Heimsch, 1942).

Hutchinson (1959) stated that Euphorbiaceae can well dispense of this genus, for it possesses a combination of characters not found in the Euphorbiaceae: alternate and densely lepidate leaves and a racemose with unisexual flowers enclosed in the bud by a bracteole, well-developed petals. In addition, it has a ruminant endosperm. He believed the relationship of Aextoxicaceae to be closer to Aquifoliaceae and Celastraceae.

Gundersen (1950) placed this family near Melianthaceae and Didiereaceae of the Sapindales. Pulle (1950) also placed it in the Sapindales, but near Akaniaceae and Aceraceae.



## PURPOSE AND METHODOLOGY OF STUDY

Mc Nair (1935) has made the following statement: "Plants can be classified chemically in accordance with the substances made by them. Such a chemical classification may be compared with and used as a supplement to morphological classification and may be of some importance in the development of the true natural system of angiosperm phylogeny."

There are many classifications of the "Geraniales", most of them based on morphological evidence. Thus the purpose of this research was to gather supplemental evidence, that is, chemical evidence, to differentiate between the systems, and to perhaps support one or another system. This was to be accomplished through 1) work done in the laboratory by the author, 2) a survey of literature, and 3) information cards of Dr. R. D. Gibbs.

Many questions arose in the course of this study. Some answers will be attempted.

At the beginning of the study, one of the questions which arose was whether Hutchinson (1959) was justified in splitting the "Geraniales" of Engler and Diels (1936) into two lines of evolution, the Lignosae in which most of the families were placed, and the Herbaceae. Was Hutchinson justified when he raised many genera to the familial rank and created from the more than twenty families six orders? (See page 11.)

While this does remain as one of the problems to be discussed, the purpose of this study has become more complicated. When the work was initiated, the classification of the "Geraniales" according to the 11th "Syllabus" was followed. Since then, the 12th "Syllabus" (Engler and Melchior, 1964) has appeared.

There are many differences between the two classifications, the major one being the split of the "Geraniales" into two orders, namely the "Geraniales"

s.s. and the Rutales.

The question of whether the "Geraniales" s.s. can be justifiably split into the Geraniales and Malpighiales will be discussed. Are the differences great enough to justify the placing of the former in the Herbaceae, and the latter in the Lignosae?

Since the basic problem concerned with in this thesis is not that of the Rutales, the author will not attempt to answer whether the Rutales is a natural group. Rather she will try to discuss whether the split of the original 21 families is justified. Should the Rutales and "Geraniales" s.s. be united or left as is?

Other classification systems will also be discussed.

## CHEMOTAXONOMY

### A. Criteria considered in chemotaxonomic studies

One of the first problems the chemical systematist encounters is what products should be included in his analysis. Generally primary products are not included in his analysis. These fundamental pathways, such as those which are involved in energy transfer and in the synthesis of basic protoplasmic constituents, are considered to have been formed from the beginning of life. However, some products, such as d-glucitol and sedoheptulose, are considered, for <sup>some</sup> more highly-evolved plants are thought to accumulate these in large amounts.

Thus secondary products, such as alkaloids, terpenes, and various water-soluble pigments, and other metabolites, are analyzed. It is believed that in the process of product-formation, specific enzymes are necessary. Thus sometimes an unusual compound is restricted; at other times, its distribution suggests some phylogenetic relationship. For example, the distribution of isoquinolines in groups of related families is additional evidence for their relationship. But one must be cautious when chemical constituents are widely-distributed. The presence of nicotine in Solanaceae, some members of the Rutaceae, and also in Equisetum, is an example of the wide distribution of a product. It is hardly likely that the angiosperms are related to Equisetum! Thus it would be postulated that the pathways for the synthesis of nicotine probably evolved independently of each other.

Price (1963) states that the biogenetic pathway is phylogenetically more significant, not the structure itself. Thus it is the series of gene-controlled reactions which give rise to the product that is important. As noted previously, similar products (e.g., nicotine) can form via independently-evolved pathways.

A classic example of an independently-evolved pathway is that of  $\phi$ -cyanin formation. With the exception of Caryophyllaceae,  $\phi$ -cyanins are found in all Centrospermae. This suggests that compounds evolved once and successfully replaced or substituted anthocyanins with <sup>in</sup> certain evolutionary lines, and these taxa in which  $\phi$ -cyanins are present are phylogenetically related. (Alston et al, 1963)

The presence or absence of a product may be due to biochemical or genetic differences; a single gene difference may cause the inability of an organism to synthesize or accumulate a product in sizable amounts. (Price, 1963)

In addition, complexity should be evaluated before making comparisons. Alkaloids which occur infrequently or are present in trace amounts must be distinguished from those found in all members of a given group. Predictions about phylogeny can be made only if one restricts the groups of defined chemical substances to avoid interfering factors. Different populations of the same species may contain different combinations of products.

The parts of the plant investigated must be recorded. Alkaloids of leaves and stems may differ. Also one product may be found in one organ and may be absent from another. Steroidal sapogenins of leaves and seeds of Agave, for example, differ (Wall and Fenske, 1961).

Changes in the course of growth illustrate that there are chemical changes in the course of growth and development. In Theobroma cacao, the distribution of flavonoids and other phenols differ. During leaf development and maturation, there are present anthocyanins and flavonols with traces of phenolic acids. Then the concentration of flavonoids decreases, and the concentration of phenolic acids increases. (Griffiths, 1959)

Thus, certain criteria must be known. The chemistry, biogenesis, and distribution, of the product must be known. The range of production must be

considered in comparing. As stated previously, caution must be taken, as groups of compounds produced via analogous and homologous pathways of metabolism must be distinguished.

B. Methods used in this study (including a discussion on the chemistry involved)

In most cases, fresh leaf, stem, and root material was used in the investigation. The specimens were collected in Montreal from the McGill University Greenhouse and the Montreal Botanical Gardens.

Additional specimens were obtained from botanical gardens in Europe, Africa, Asia, Australia, and North America. These plants were flown to Montreal, packed in polyethylene bags to ensure freshness.

1. Hot water and cigarette tests. Fresh matured leaves are used for these tests. The results of both tests are similar, but the reaction in the cigarette test is more rapid.

In the cigarette test, a lighted cigarette is pressed gently on the underside of the leaf for three seconds. The results are classified as follows:

- I....an immediate reaction (formation of a ring)
- II....a slower reaction
- III...a very slight reaction
- IV...no reaction
- o.r.."oxalis-reaction"

In the hot water test, the leaf is dipped partway into hot water (85 - 90° C.) for five seconds. At the juncture, a dark line appears immediately in a strong reaction (I). After about one minute, if a line should appear, the result is classified as II. Any formation of a dark line after 30 minutes is classed as III, and if no reaction occurs within that time, it is recorded as a IV reaction. Leaves of Hedera are used as controls.

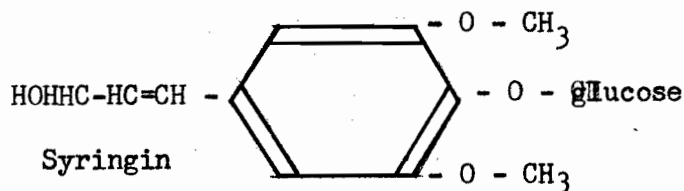
A yellow color in the dipped part is known as the "oxalis-reaction". It was first observed Oxalis. This particular result is perhaps due to

the highly acid cell sap; however, it has also been obtained in young leaves.

Dykyj-Sajfertova (1958) stated that positive results indicate the presence of polyphenols and their enzymes, the polyphenolases. This is supposedly indicative of an advanced character.

2. Syringin ( $1:1 \text{ H}_2\text{SO}_4/\text{H}_2\text{O}$ ) test and raphides. Freshly-cut cross-sections of stems, sometimes petioles, are mounted on a clean glass slide. On one section, a drop of 50 per cent sulphuric acid is added, and on the other, a drop of water. The latter is used as a control. The presence or absence of raphides and/or crystals is noted in this section too.

A blue color in the xylem, lignified fibres, and in other parts of the section, is recorded as a positive result. It is said to be due to syringin, the glucoside of 5-methoxy-coniferyl alcohol.



Other colors may appear in the xylem and fibres. These colors are correlated with negative reactions. A yellowish color often appears. A red color in the lignified tissue is closely-correlated with positive HCl/Methanol and leuco-anthocyanin reactions. Purpling, or darkening, especially in the cortex, is associated with the presence of aucubin or aucubin-like substances.

Raphides are calcium oxalate crystals, "slender, needle-shaped crystals arranged parallel to each other in tight bundles occurring in special raphide sacs" (Gibbs, 1963).

Perhaps the first published account of the use of raphides as a taxonomical tool was that of Robert Brown (in Gibbs, 1963). Brown remarked in a paper published around 1845 and concerning the parasitic Rafflesia: "That the whole of this covering belongs to the stock, is proved by its containing those raphides or acicular crystals which are so abundant in the root of Vitis

er Cissus, and which are altogether wanting in the parasite."

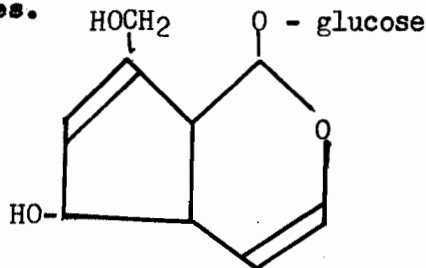
Gulliver (1866) noticed their restricted distribution, and remarked about their possible taxonomic importance.

3. Ehrlich test (A - B). Approximately 0.5 gm. of fresh plant material (usually leaf material) is placed in a test tube and covered with 50 per cent ethyl alcohol. The extract is then concentrated by evaporating it to a low level in a boiling water bath. Any frothing is recorded. The test tube is occasionally shaken.

Using a spotting pipette, three spots of the extract are placed on a 9 cm. filter paper. They are allowed to dry; the first spot is used as a control. On spot two is added a drop of acid alcohol (5 ml. conc. HCl: 200 ml. 95 per cent ethyl alcohol). To the third spot is added a drop of Ehrlich's reagent (1 gm. *p*-dimethylaminobenzaldehyde:5 ml. conc. HCl:200 ml. 95 per cent ethyl alcohol). These spots are allowed to dry. Any color change is recorded.

The filter papers are then placed in a preheated oven (100° C.) for one minute. The colors may become intensified. A blue reaction in the third spot is recorded as a positive reaction, and is caused by the presence of aucubin or aucubin-like substances.

Aucubin



A magenta color may appear in that third spot. This color change is correlated with a red color in the Syringin test, with a magenta color in the HCl/Methanol test, and with a "carmine" color in the leuco-anthocyanin test.

In what is called part B of this test, a drop of aqueous (10 per cent) ammonia is added to the first spot (the control). Sometimes a bright yellow

color appears. Although this has not been tested, this color may be indicative of flavonoids. ~~Other colors may appear.~~ A red or off-beat color is rare. More often a pale yellow color is retained.

4. HCl/Methanol test. Using a pencil sharpener, shavings of wood, usually sapwood, are obtained. These shavings are submerged in a few milliliters of HCl/Methanol solution (25 ml. HCl:1,000 ml. methanol) and left overnight.

The next day, the color of the shavings is recorded. A magenta reaction is recorded as positive, and a pale-yellow to no color is recorded as negative. Using Ridgeway (1912) as a guide, the specific colors are recorded. The positive reactions are also rated:

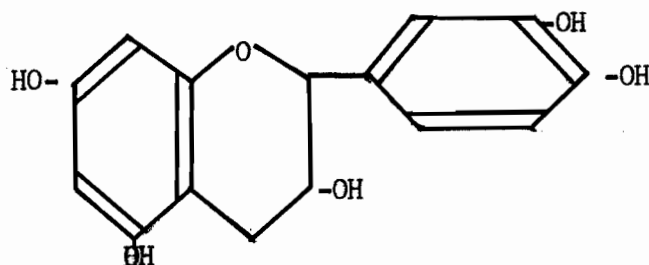
purple 1....very pale purple
3....magenta
4....darker than magenta

The magenta color corresponds to the magenta on Plate 26.

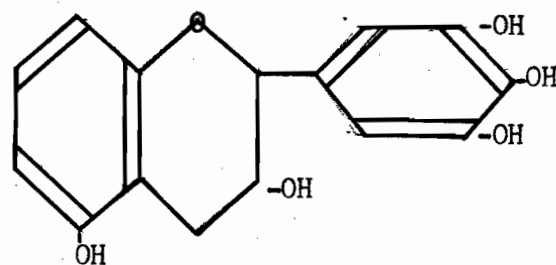
The color of the liquid in which the shavings were submerged is also recorded.

Isenberg and Buchanan (1945) described this test, and were among the first to note the taxonomical possibilities of this test.

Adler (1951), from his studies, concluded that catechol tannins may be responsible for the magenta color. These condensed tannins yield polyphenols on hydrolysis and may be condensation products of such compounds as catechin or gallic catechin. Therefore a positive reaction is due to the presence of flavanols.



Catechin



Gallic catechin



5. Leucoanthocyanin test (A). In this test, 0.5 to 1.0 gm. of freshly chopped leaf material is placed in a test tube. Five ml. of 2N HCl (186 ml. conc. HCl per liter of solution) is added to the leaf material. To facilitate matters, that is, to avoid having to measure 5 ml. solution each time it is needed, the test tubes are etched with a diamond-marking pencil at the 5 and 10 ml. points.

The test tube is then immersed in boiling water for 20 minutes, after which it is removed and cooled. Five ml. of amyl-alcohol is added to the 10 ml. mark on the test tube. The solution is vigorously-shaken.

in the amyl alcohol

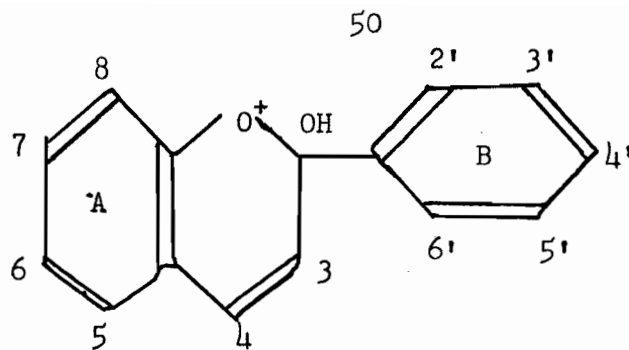
A red color is a positive reaction. Usually in a negative reaction, the upper layer of liquid is a yellowish-green color. The next day, the color of the upper layer is matched with the color charts in Ridgeway (1912). A positive reaction has a "carminé" color (Plate I).

In this test, leucoanthocyanins (leucoanthocyanidin glucosides), which are colorless and water-soluble, are hydrolyzed and oxidized to amyl-alcohol-soluble leucoanthocyanidins (the sugar-free phenolic substances) (Bate-Smith, 1954; Clark-Lewis, 1962).

Catechins, on heating in dilute HCl, may form red-brown polymers loosely-known as phlobaphenes or tannin-reds. The catechins under these conditions also form brown polymers. These phlobaphenes are amyl-alcohol-soluble.

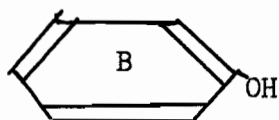
Glycosides of aucubins may interfere with reactions.

All naturally-occurring anthocyanins have 4', 3, 5, and 7, occupied by -OH or substituted groups. They usually occur as glycosides with sugars attached to positions 3 or 3 and 5. These sugars may be glucose, galactose, rhamnose, arabinose, disaccharides, or trisaccharides.

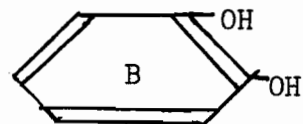


Leucoanthocyanin

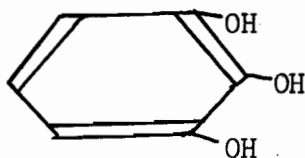
The aglycones of the anthocyanins include the following:



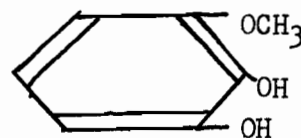
Pelargonidin



Cyanidin



Delphinidin



Malvidin

Bate-Smith (1954), who investigated many species, found anthocyanins to be more prevalent in woody plants. Thus it is believed that the ability to synthesize anthocyanins has been lost by many herbaceous plants. Therefore the absence of anthocyanins is considered a more highly-evolved characteristic.

6. HCN test A. About one gm. of fresh leaf material, preferably young leaves and tips, is ground with a speck of emulsin, a few drops of water, and 1 to 2 drops of chloroform. Emulsin is an enzyme which hydrolyzes the cyanogenetic glucosides and releases HCN.

The mixture is placed in a special glass test tube. This is stoppered with a glass stopper to which has been attached with wax an almost triangular piece of picric acid paper freshly dipped in 10 per cent  $\text{Na}_2\text{CO}_3$  and blotted.

A strongly positive reaction is one in which a red-brown color forms almost at once on the picric acid paper. The test tubes are left in a rack for at least a week, at which time the results are read.

Bohm (1803) first reported the presence of HCN or prussic acid in plants. He found it in Prunus amygdalus Stokes var. *amara*. But Lindley in 1830 probably made the earliest reference to the taxonomical use of the presence or absence of HCN in plants (Gibbs, 1963). He used it to separate the *Amygdaleae* (-laceae) from *Rosaceae* and *Pomaceae*, and also from *Leguminosae* and *Chrysobalaneae* (-lanaceae).

Cyanogenetic glycosides are rarely found in the entire plant; the leaves are more constantly found to contain them. However, seeds and racemes may also be cyanogenetic.

Dillemann (1958) wrote a rather comprehensive paper on cyanogenetic glycosides. Cyanogenetic substances are found in several families of the "Geraniales". Linamarin, found in Linum usitatissimum, Phaseolus lunatus (*Leguminosae*), Manihot usitatissimum (cassava), and Hevea brasiliensis, yields glucose, HCN, and acetone, upon hydrolysis. Hiptage madablota and Corynocarpus laevigata (*Corynocarpaceae*) contain hiptagin which yields glucose, tartronic acid, ammonia, and HCN, upon hydrolysis by an acid. Phyllanthin, in leaves of Phyllanthus gastroemii, yields glucose and  $\beta$ -hydroxy mandelonitrile. Zierin, found in Ziera laevigata, yields glucose,  $\beta$ -hydroxy benzaldehyde, and HCN.

These cyanogenetic substances in higher plants form a relatively small and somewhat heterogeneous group of glycosides of the cell sap. The test used by the author, however, merely shows the presence of prussic acid in the plant, not the chemical nature of the glycoside yielding it.

As noted, linamarin and the other cyanogenetic substances consist of one or more sugars, cyanhydric acid, and a third variable substance. The origin of the cyanogenetic substances is unknown, as is the role they play. (Dillemann, 1958)

The concentration varies with the environment. Trione (1960) found that the concentration of HCN increased with light and also diurnal variation in flax seedlings. Ermakov (1960) also found the concentration of linamarin was higher under controlled conditions of lower soil moisture, low temperature, after mechanical injury, and in young growing organs.

McNair (1941) stated that there are four times as many herbs as trees with cyanogenetic glycosides. Thus the presence of them must be characteristic of more highly-evolved plants.

7. Juglone tests A - C. This test is thus named because it was first described for Juglans. Only part A, however, is a test for juglone and its related compounds. Parts B and C are tests for ~~other~~ compounds and are conveniently included here.

In part A, a small amount of finely-chopped bark material (from the root or stem, but preferably the root) is placed in a test tube. It is then covered with chloroform and left overnight.

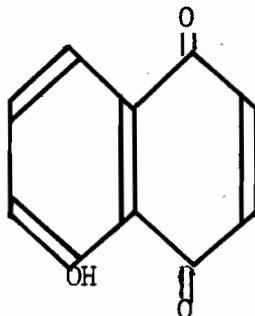
The next day, the liquid is filtered off, and this is evaporated to dryness in a boiling water bath. About 10 ml. of ether is added to the cooled test tube; this is shaken. An equal volume of 10 per cent  $\text{NH}_4\text{OH}$  is added to the test tube. The solution in the test tube is shaken.

A brilliant purple color in the ammonia layer (the lower layer) is a positive reaction and is due to the presence of juglone, a naphthoquinone, or to related compounds. An orange or wine color may be indicative of naphthoquinones also.

In test B, the color of the aqueous layer is recorded in a few days. A yellow color may be indicative of flavonoids.

Using a short wave ultraviolet lamp, any fluorescence in either layer is recorded. A brilliant blue fluorescence in the ammonia layer is said to be indicative of coumarins. This is part C of the Juglone test.

Juglone, the naphthoquinone, has the formula  $C_{10}H_6O_3$ , and has been known to be present in the walnut for over a century (Thomson, 1957).



Naphthoquinone

8. Tannin test A. A fresh solution of 2.5 per cent aqueous ferric ammonium citrate is prepared and placed in a petrie dish. Using a pair of small pliers, a half sheet of 11 cm. Whatman's No. 1 filter paper is dipped into the solution and then blotted. A piece of clean leafy material is laid on the paper, the paper is folded, and the entire thing is squeezed with the pair of pliers.

A positive reaction is one with a grey <sup>to purpley</sup> color on the filter paper. The colors may vary from purple, blue, brown, to green. Sometimes a green color may be due to the presence of chlorophyll which masks a reaction. A purple color, perhaps due to anthocyanins, may likewise mask a reaction.

A negative reaction is one in which no grey color is present, or if found, is present only on the edges of the filter paper.

Young leaves of a given plant contain more tannin than older leaves (Bates, 1953; Bates and Henson, 1955). In addition, the concentration of tannin varies within a single plant exposed to different environmental conditions. Ogden (1936) reported that the time of year, time of day, and the amount and character of light, affected tannin content. Thus Stitt and Clarke (1941) found tannin concentrations in <sup>lespedeza</sup>serica/leaves to increase until the 30th of June and gradually decrease until the 22nd of September. Stitt, Hyland, and McKee (1946) reported that tannin content varied significantly with soil type.

Stitt (1942) also found that hereditary factors also controlled tannin content. The number of pairs of genes involved with tannin inheritance of Lespedeza cuneata Don. is 20 - 25 (Bates, 1953).

Tannins are best regarded as phenolic substances. Non-hydrolyzable condensed tannins are flavonoid derivatives. These are complex polymers which may form insoluble products. These condensed tannins which yield no sugars on hydrolysis are considered condensation products of substances similar in nature to catechin or gallocatechin. (See page 48 for configurations of catechin and gallocatechin.)

Hydrolyzable tannins occur as glycosides, the aglycone often being a phenolic acid, e.g., gallic acid or ellagic acid.

9. Chromatography for phenolic constituents. Approximately 10 gram weight of pieces of fresh leafy material is covered with 80 per cent ethanol and heated on a steam bath for a half hour. The mixture is then blended in a Wareing blender and filtered through a Buchner funnel. The leaf-particles are re-extracted with alcohol on the steam bath, and the filtrate is added to the original filtrate. This process is repeated until the leaf material is colorless or almost so. The combined filtrates are evaporated to dryness under an air jet in a 250 ml. beaker.

The beaker in which the filtrate has been evaporated to dryness is half-filled with distilled water, and approximately 2 grams of celite is added. The beaker is then heated on the steam bath for 20 to 30 minutes.

The extract is divided into two parts, and to each part 5 ml. distilled water is added. One part is subjected to acid hydrolysis by adding one-fourth the volume present of conc. HCl and heating for one hour on the steam bath with a cold water bath resting on top. It is then cooled, and placed in a liquid/liquid extractor (Quickfit upwards displacement type) with ether and

heated either for six hours or overnight. The ether extract is then evaporated in a 250 ml. beaker under an air jet.

The other part of the water extract is subjected to alkaline hydrolysis. It is placed in a 250 ml. flask and 40 per cent NaOH equal to one-fourth the volume is added. Nitrogen gas is bubbled into the solution for about 30 seconds. The flask is then covered and left for 5 hours at room temperature, or 15 - 16 hours overnight in the refrigerator. The solution is then acidified with conc. HCl to approximately pH 2, cooled, and placed in a liquid/liquid extractor with ether for six hours or overnight. As with the acid hydrolysis, the ether extract is evaporated to dryness under an air-jet in a 250 ml. beaker.

Each residue is now taken up with a few drops of absolute ethanol, and the resulting solutions are chromatogrammed as follows: Two chromatograms of each solution (therefore four for each plant, two for the acid hydrolysis and two for the alkali hydrolysis) are prepared. The solutions are spotted at the origin of prepared Whatman's #1 filter paper (18 1/4" x 22 1/2"), and the papers are placed in well-saturated chambers for 5 hours, using benzene-acetic acid-water (6:7:3) as the solvent. The chromatograms are then removed and allowed to dry in a well-ventilated place. They are then turned 90° and run in 2 per cent formic acid for 3 - 3 1/2 hours. They are again removed from the saturated chambers and dried as before.

The resulting chromatograms are first read under long-wave ultraviolet light. Certain compounds fluoresce giving distinct colors. Reference compounds had been run by earlier workers, and a chart had been prepared giving the color and position of each known compound. The spots on the chromatograms are thus identified.

For further identification, one each of the acid and alkali chromatograms is sprayed with 3 per cent aqueous  $\text{FeCl}_3$ . Again the spots are identified by

their color and position on the chromatograms. The other set of chromatograms is sprayed with a diazotized sulfanilic acid mixture which is freshly made as follows: 4.5 gram weight of sulfanilic acid is added to 45 ml. of conc. HCl in a liter flask and 500 ml. of distilled water is added. This is then mixed with 5 per cent  $\text{NaNO}_2$  and 20 per cent NaOH in the proportions of 2:1:2. As before, the spots are identified by their color and position on the chromatograms.

Acid and alkali hydrolysates generally yield a large number of known phenolic acids. Many are unidentifiable. This suggests that phenolic acids in plants are in the bound form. Acids which do not move in the benzene solvent are undetected.

Acid-hydrolysable compounds occur, most likely, as glycosides, while alkaline-hydrolyzable compounds occur mostly as esters or alkali-sensitive glycosides. (Bohm and Towers, 1962)

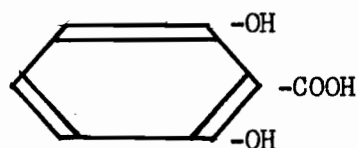
Over twenty phenolic compounds were identified in this work. They occur in the different classes of phenolic compounds. The literature available on this is vast. Therefore not much will be said about them here, and one is referred to Neish (1960), Whalley (1961), Seikel (1962), Robinson (1963), and others, for more detailed discussions.

Phenolic compounds present in tissues are characteristic of the species; this is based on the view that they are metabolically inert (Bate-Smith, 1958). Thus their taxonomic value is cited. Caution, however, is necessary, for phenolic acids may differ due to environmental conditions, age, physiological state of the material, and type of tissue used. Griffith (1959) studied the distribution of phenolic acids and found them to be more prevalent in the Lignosae of Hutchinson (1959).

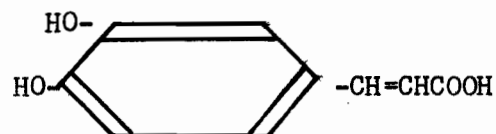
Phenolics may be taxonomically-important because of the many different types, many of them restricted. The most common phenolic substances are members of the cinnamic acid series ( $\text{C}_6\text{-C}_3$ ) and the benzoic acid series ( $\text{C}_6\text{-C}_1$ ). The



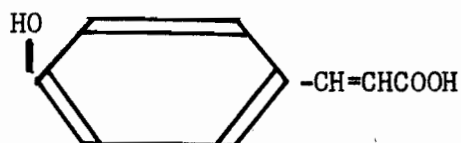
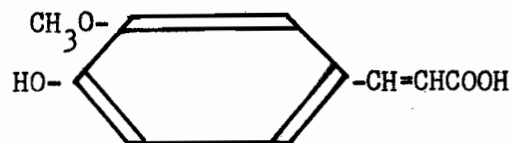




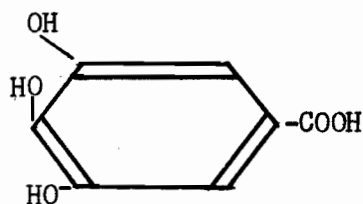
Gentisic Acid



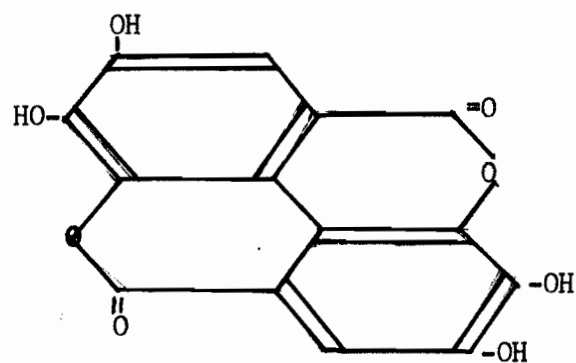
Caffeic Acid

p-coumaric Acid

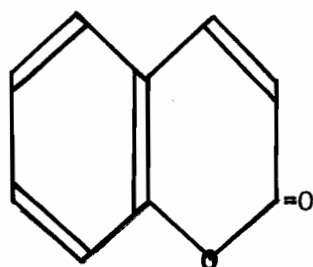
Ferulic Acid



Gallic Acid



Ellagic Acid



Coumarin  
[ o-hydroxycinnamic Acid  
(coumarins are lactones  
of this acid)]

Some of the More Common  
Phenolic Constituents

10. Saponin tests A - B. Test A. Saponins froth when boiled and shaken. Following Amarasingham and his co-workers (1964), finely-chopped fresh leaves are placed in a 15 x 1.5 cm. test tube marked at 5 ml. and 10 ml. volumes until the bottom is covered. Distilled water is added to the 5 ml. mark. The tube is heated until the liquid boils, after which it is boiled continuously for one minute. It is then cooled and vigorously shaken, and left standing for five minutes. Then the amount of foam is noted. Amarasingham et al. recorded as having saponins those species which gave 2 cm. depth or more of froth under these conditions. We record our results as follows:

1.5 or more cm. froth.....positive  
 1.0-1.5 cm. froth.....positive?  
 0.5-1.0 cm. froth.....negative?  
 0.0-0.5 cm. froth.....negative

The next day, any persistent froth is again recorded.

Paris (1963) reported saponins in 70 families; he added that the distribution is incompletely-known. Steroid saponins are less common, and apart from some families, e.g., Scrophulariaceae, they are found mostly in monocots. The triterpenes, on the other hand, are rare in monocots, but are present in Linaceae, Rutaceae, Sapindaceae, Polygalaceae, and other families. Paris concluded that this widespread character limits their use in taxonomy.

Test B. This is not, of course, a test for saponins but is conveniently included here. 10 per cent  $\text{NH}_4\text{OH}$  is added to the test tube to the 10 ml. mark. This is done the following day. The test tube is shaken <sup>the color recorded,</sup> and left to stand. Any deepening in color is recorded after one day and again after three days. At present, it is not known why aqueous ammonia causes darkening of the solution. The deepening of the color begins at the top, thus indicating that oxygen is necessary for the reaction. Therefore the test tubes are unstoppered and shaken slightly at various intervals. This test has been little used so far but it is evident that it has some taxonomic value (Gibbs, unpub'd).

C. Other chemical constituents which are known to occur in or which are thought to be absent from the taxa under consideration

1. Alkaloids. Alkaloids are not chemically-homogeneous. All contain nitrogen, frequently in a heterocyclic ring, and are thus basic in nature. Alkaloids often exist in plants in combination with various organic acids. Most are solids: a few are liquid. The molecular structure is complex, and there is often recognizable pharmaceutical activity. Classification is based on the ring system present.

There is a tendency for higher plants to contain more alkaloids than lower plants, but a few are known in clubmosses and horsetails, also fungi. None are thusfar known in bryophytes. (Robinson, 1963)

The alkaloids found in the same plant have generally the closest chemical relationship. Frequently they form a homologous series; often they are isomers, and sometimes stereoisomers. The same alkaloids are seldom found in different families, and often alkaloids are characteristic of a family. One kind may be confined to a single genus.

There are more than 1,000 alkaloids, most of them in vascular plants (Alston and Turner, 1963). As stated previously, they do not constitute a chemically-natural group, nor do they constitute a natural biological group, functionally, phylogenetically, or biosynthetically. McNair (1935) and Hegnauer (1963) reported that 15 - 20 per cent of all vascular plants contain alkaloids in one part or another.

Boas (1927) and Couch (1931) reported that alkaloids are found in 57 families of gymnosperms and angiosperms, the majority of which are found in warmer regions. This confirms what McNair (1935) stated, namely, that alkaloids are more frequent in tropical families than in temperate ones.

The taxonomic value of alkaloids is not restricted to distribution, but also to biosynthesis and enzyme and genetic mechanisms (Alston and Turner, 1963). The chemical, botanical, and pharmaceutical properties of alkaloids must be considered, as there is no sharp distinction between alkaloids and many nitrogen-containing compounds. On the other hand, the heterocyclic base, thiamine, which is widely-distributed, is not an alkaloid. (Hegnauer, 1963)

For taxonomic purposes, they can be defined as "more or less toxic substances which act primarily on the central nervous system. They have a basic character, contain heterocyclic nitrogen, and are synthesized in plants from amino acids or their intermediate derivatives" (Hegnauer, 1963).

Mothes (1964) did chemical studies on the Rhoeadales and used the restricted distributions of alkaloids in her discussion.

Alkaloids are found in some families of the "Geraniales", namely, Erythroxylaceae and Rutaceae. A summary of the alkaloidal distribution of this order is presented in Appendix V.

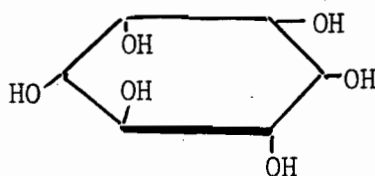
2. Aliphatic polyols and cyclitols. Plouvier (1963) examined the distribution of polyalcohols in order to determine the relationship between distribution and plant systematics. He noted that "as long ago as 1893, Monteverde examined 797 species in 199 genera of Scrophulariaceae with the aid of a simple test for the presence of mannitol and glucitol." Monteverde subsequently was able to divide the members of the family according to the presence or absence of these two substances.

Chromatography is the method used to isolate and identify these compounds.

Aliphatic polyols consist of straight chain carbons, while cyclitols are derivatives of cyclohexanes. These classes can be further subdivided according to the number of alcoholic hydroxylgroups present.

Aliphatic polyols have been found in Cryptogamae (algae, lichens, yeasts, molds, other fungi) and the higher plants. D-polygalitol (1,5-anhydrosorbitol) has been isolated from two families: Polygalaceae and Aceraceae,

A cyclitol, a hexol named L-inositol, has been found in two angiosperm families, Euphorbiaceae and Compositae. Its configuration has been worked out by Posternak (1936):



L - inositol

Another cyclitol is L-quercitol. It has only been found in Eucalyptus populnea (Plouvier, 1961), although sought for in families of the "Geraniales".

D-pinitol (3-O-methyl-D-inositol) has been found in six gymnosperm families and 13 angiosperm families. In the angiosperms, it has been isolated from Olacaceae, Aristolochiaceae, Loranthaceae, Nyctaginaceae, Phytolaccaceae, Aizoaceae, Caryophyllaceae, Magnoliaceae, Leguminosae, Cistaceae, and Apocynaceae. In addition, it has been found in two families of the "Geraniales", Zygophyllaceae and Euphorbiaceae.

Another cyclitol, L-quebrachitol, has been isolated from 11 angiosperm families: Ulmaceae, Moraceae, Proteaceae, Loranthaceae, Aceraceae, Hippocastanaceae, Sapindaceae, Elaeagnaceae, Apocynaceae, Compositae, and Euphorbiaceae.

3. Fatty acids of seeds. Extensive coverage of the distribution of fatty acids in seeds and the possible relationship of this to systematics has been done by Eckey (1954) and Hilditch (1956).

Shorland (1963) stated that the fatty acid composition differed in different parts of the same plant. Fatty acids are found in the lower

plants as well as in the angiosperms. Some of them are widespread, e.g., palmitic, oleic, linoleic, and to a lesser degree, linolenic acid. Others, however, may be confined to one family, e.g., the chaulmoogric fatty acid series in Flacourtiaceae, and the fluoroacetic acid series in Dichapetalaceae.

Hilditch (1956) stated that fatty (glyceride) components of seeds are specific and closely-related to the family in which parent plants have been grouped. It is therefore possible to classify according to the fatty acid content of seeds.

Chandra (1964) found the fatty acid composition to change in seeds. In Ricinus communis of the Euphorbiaceae, the concentration of ricinoleic, linoleic, and stearic acids increased with ripening, while the concentration of oleic and palmitic, after an initial increase up to 28 days, gradually decreased towards later stages of growth.

A summary of the fatty acids found in some members of the "Geraniales" can be found in Appendix IV.

4. Cardiac glycosides. Cardiac glycosides are related to the steroids, consisting of an additional lactone ring and a sugar (often a tetrasaccharide) attached to C - 3 of the cyclopentanophenanthrene skeleton. The aglycones are rarely found in the free state, and the sugar moiety varies in composition. The two subgroups differ according to the size of the lactone ring: a) cardenolides (5-membered ring) and b) bufanolides (6-membered ring).

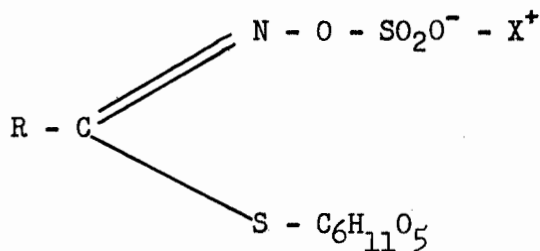
Since there is no mention in the literature of the occurrence of cardiac glycosides in the members of the "Geraniales", they are presumably absent from the order.

5. Sulphur-compounds - thioglucosides. Sulphur is one of the major elements which all plants need. Kjaer (1963) calls the isothiocyanate-producing

glucosides "a uniform but far from universally distributed group of plant constituents composed of thioglucosides, possessing the property of undergoing hydrolysis to glucose, sulphuric acid, and isothiocyanates induced by the enzyme myrosinase, which usually accompanies the thioglucosides in plant tissues." The classic examples of thioglucosides are sinigrin and sinalbin, both isolated from bark and mustard seeds over 100 years ago.

Guignard (1890, 1893) found the enzyme to be widely-distributed in members of the Cruciferae. It is also present in Capparidaceae and Resedaceae. Two families of the "Geraniales" contain the enzyme: Tropaeolaceae and Limnanthaceae.

The basic formula for all isothiocyanate-producing glucosides is as follows:



Thioglucosides have also been isolated from Euphorbiaceae, but Kjaer (1963) adds that it is not a typical constituent of the family and is sporadically-distributed.

6. Quinones. Quinone pigments are the largest class of natural coloring matters but make little contribution to natural coloring. They are found mainly in the bark or underground regions. They are easily oxidized and reduced, thus suggesting their probably importance in redox reactions.



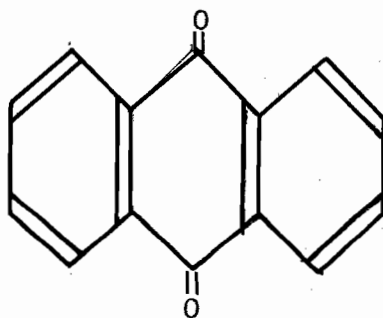
There are three major classes of quinones: benzoquinones, naphthoquinones, and anthraquinones. The first group is confined mainly to the fungi. However it is found in some angiosperms:

1. Adonis vernalis - Ranunculaceae
2. Myrsine, Embelia, Rapanea, Maesa - Myrsinaceae
3. Oxalis-purpurata var. jacquinii - Oxalidaceae
4. Perezia, Trixis - Compositae

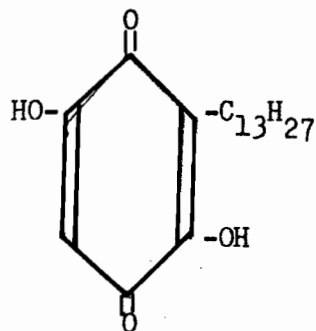
Most likely independent evolution was responsible for the presence of benzoquinones in diverse families.

Naphthoquinones are also rare. A discussion of these pigments is presented in the section on methods used in this study (under Juglone test).

Anthraquinones are widely-distributed, especially in Rubiaceae, Polygonaceae, and Rhamnaceae. Bate-Smith (1957) found these pigments to be confined to the Rutaceae in the "Geraniales".



Anthraquinone



Rapanone

7. Crystals. Crystals are usually "variously-shaped deposits of calcium oxalate", but gypsum may also occur. Some types are widespread, occurring as solitary prisms or clusters. However, the kinds secreted are relatively fixed in each species. If more than one kind occurs, its concentration varies with the time of the year and the nutrients available (Metcalfe and Chalk, 1950)

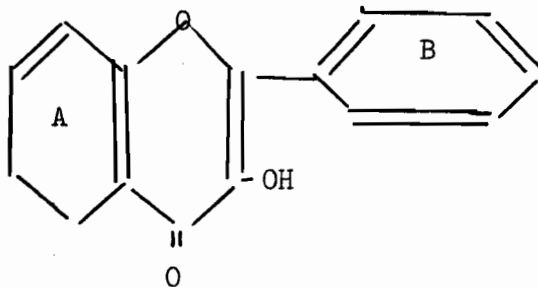
Raphides and crystal-sand are more restricted in distribution, therefore are taxonomically more important.

Calcium oxalate crystals are commonly found in vacuoles. At other times, they are found in specialized cells called crystal idioblasts. Still others are found in cell walls. They may completely fill the cell. (Esau, 1960)

8. Flavonols. There are three flavonols common to angiosperms: myricetin, quercetin, and kaempferol. Quercetin is one of the most common phenolic compounds of vascular plants, with kaempferol as a close second. These flavonols are commonly 3 - glycosides, the sugars being glucose, galactose, or rhamnose.

The works of Bate-Smith (1957, 1958, 1962) should be consulted for detailed information.

The configuration of flavonol is:



## RESULTS

The following tables represent the data collected through 1) work done in the laboratory, 2) a survey of literature, and 3) information cards of Dr. R. D. Gibbs. For the author's summary of her results, consult the Appendix.



Family	H.W. Cig.	Syr.	Raph.	HCl/M.	L.A.	Ehr.	HCN	Jug./fl.	Tann.	Sap. A/B	Alk.	P.C. A/B/C	Al	Tart.	Card. glyc.	Cyc.
Geraniaceae	IV, or	-	-	(+)	(-)	-	-	-/+	(+)	-/+	+	(+)/(+)(-)	-	low to none	-?	-
Oxalidaceae	(or)	-	-	(+)	(+)	-	-	-/(+)	(-)	-/(-)		(+)(-)/-	-	-	-?	-
Tropaeolaceae	IV	-	-	-	-	-	-	-/+	+			(+)(+)(-)	-	tr	-?	-
Zygophyllaceae	IV	-	(-)	-	-	-	(-)	-/+	(-)	-/-	+	(+)(+)(-)	-		-?	+
Linaceae	I,II	-	-	-	-	-	(+)	-/+	(-)	-/-		(+)(+)(-)	-		-?	-
Erythroxylaceae		-	-	+	(+)	-	(-)	-/+	±		++	(+)(+)(-)	-		-?	
Limnaceae	IV	-	-	-	±	-	±	-/+	±			(+)(-)/-	-		-?	
Euphorbiaceae	(IV)	(-)	-	(-)	(-)	-	(-)	-/(+)	(+)	(-)/-	+	(+)(+)(-)(-)	-	tr	-?	(-)
Daphniphyllac.		-	-	+	?			-/								
Rutaceae	(IV)	(-)	(-)	(-)	(+)	-	(-)	-/+	(±)	(-)/+	+	(+)(+)(-)(-)			-?	
Cneoraceae	?	-	-	-	-	-	-	-/+	+			(+)(+)(-)			-?	
Simarubaceae	(IV)	-	-	(+)	(-)	-	-	-/+	+		+	(+)(-)(-)	-		-?	
Picrodendraceae	IV	-	-	+	-	-	-	-/+			+				-?	
Burseraceae	?	-	-	±	±	-	±	-/+	?			(±)(±)(-)	-		-?	
Meliaceae	(IV)	-	-	(+)	(±)	-	(-)	-/+	(+)	-/+		(+)(+)(-)	-		-?	
Akaniaceae	II	-	-	+	+	-	?	-/+	+		+	?			-?	
Malpighiaceae	(IV)	-	-	(+)	(+)	-	(-)	-/+	(±)	-/+	+	(±)(-)(-)	-		-?	
Trigoniaceae																
Vochysiaceae													+			
Tremandraceae	IV	-	-	(+)	-	-	(-)	-/+	?			(-)(+)(-)			-?	
Polygalaceae	(IV)	-	-	(-)	±	-	(-)	(-)/(±)	±	+/+	+	(+)(±)(-)(-)			-?	
Dichapetalaceae		-	-	-	+	-	-	-/-	+		+				-?	
Callitrichaceae					?	+						-/-/-			-?	
Balsaminaceae	IV	-	+	?	+	-	(-)	-/+	(+)	-/?		(+)(±)(-)			-?	
Buxaceae	IV	-	-	-	(-)	-	-	-/(+)	(±)	-/-	+	(+)(+)(-)			-?	

Key to symbols: (+) = majority are positive  
 (-) = majority are negative  
 (±) = few more negative/positive than positive/negative  
 ± = half positive, half negative  
 + = all positive  
 - = all negative  
 ? = questionable results

Comments: This is merely a summary chart. It is subjective in part. Thus for more accurate results, the tables on the following pages and in the Appendix should be consulted.

H.W. = hot water test  
 Cig. = cigarette test  
 Syr. = syringin test  
 Raph. = raphides  
 HCl/M. = HCl/Methanol test  
 L.A. = leucoanthocyanin test  
 Ehr. = Ehrlich test  
 Al = Al accumulators  
 Tar. = tartaric acid  
 Card. gly. = cardiac glycosides

HCN = HCN test A  
 Jug./fl. = Juglone test and fluorescence  
 Tann. = tannin test  
 Sap. A/B = saponin test A - B  
 Alk. = alkaloids  
 P.C. A/B/C = phenolic constituents groups A,B,C  
 Cyc. = cyclitols



Family	H.W. & Cig. tests			IV	or	?	Syringin test		?	Raphides		?	Ehrlich		HCN		?	HCl/Meth.		?	L.A.	+	?	Juglone		fl.		?	Tannin		?	Saponin			
	I	II	III				+	-		+	-		+	-	+	-		+	-					+	-	+	-		+	-		+	-	+	-
Geran.				3/8	1/7	1/3		3/6NR 1/2R 1/1PK		3/9				3/8ot 1/3M		3/15	1/1	1/3	1/2	1/1	1/1		3/14		3/9	3/9		1/4		3/10	1/1	1/1			
Oxalid.			2/2	1/2	1/12	1/1		1/2NR 2/3R	1/3R	3/5	1/3			1/3ot 3/4M 1/1PK		3/14		2/3		1/2	3/7		1/6	2/3	3/14	3/13	1/1	3/8	1/13	1/1		2/4	1/1	1/1	1/4
Trop.				1/2				1/1		1/1				1/2ot		1/1							1/2		1/1	1/1		1/1	1/1						
Zygophy.				3/5		3/3		4/5NR		1/1	5/6	-2/3		3/4ot 1/1PK	1/1	3/5			5/6				3/4		3/4	3/4		1/1	4/4		1/1sd	1/1		1/1	
Linac.	1/1	1/1				1/2		2/2NR	1/1R	2/3	1/1			1/1ot	1/2	3/5			2/3				2/8		2/3	2/3		1/3	1/1		2/3		2/3		
Eryth.								1/1NR 1/1R		1/2				1/1M	1/1	1/3			1/1	1/1			1/1	1/1	1/2	1/1	1/1	1/1	1/1						
Limnan.				1/1				1/1NR 1/1R		2/2				1/1ot 1/1M	1/1	1/1							1/1	1/1	1/1	1/1									
Euphor.	2/2	3/3	5/5	15/23	1/1	14/18	1/1	20/23NR 8/9R	2/3 2/2R	33/43	2/2			18/21ot 12/12M	16/22 5/5ot	23/41 6/8ot	10/10 2/2ot	13/16	22/37	5/5	15/18	22/31	8/8	26/33	25/28	2/3	1/1	22/26	3/3	8/9	1/1sd	2/3		2/3	
Daphn.								1/1NR		1/1				1/1PK				1/1					1/1		1/1										
Rutac.	4/5		1/1	22/26		13/15	1/1	28/39NR 5/5R	2/2NR 2/2R	3/3	33/50			16/18ot 16/20M	2/7ot 4/11	5/8ot 32/42	2/2	6/8	31/45	7/8	9/12	19/24	3/3	12/41 1/1	11/40 1/1		15/21 1/1 (inflorescence)	13/16	4/4		1/1	1/1			
Cneor. Simar.				1/1		1/1		1/1NR 3/3NR 1/1R		1/1 6/6				1/1ot 1/1ot 1/1M		1/1 3/3 1/1sd		1/1	1/1			1/1 3/3	1/1 1/1	1/1 5/5	5/5		1/1 1/1		1/1		1/1		1/1		
Picrod. Burs.				1/1				1/1R 2/2NR 1/1R		1/1 2/3				1/1M		1/1 1/1		1/1				1/1	1/1	1/1 3/4	1/1 3/4				1/1						
Meliac.			1/1	2/2		1/1		2/2NR 3/4R		5/6				2/2ot 3/4M	1/1	7/9	1/1	5/7	2/2		1/1 3/4	4/4		6/7	6/7		4/6	1/1 1/1 (bark)	1/1	1/1 (bark)	1/1 (bark)	1/1		1/1	
Akaniac. Malpigh.		1/1				2/2		1/1R 3/3NR 6/9R	1/1R	8/13				1/1M 5/6M	1/1	5/8	1/1 1/1	1/1 6/9	2/2	1/2	1/1 3/3		1/2	1/1 5/6	1/1 5/6		1/1 4/4	2/3	1/1		2/2	2/2			
Trig. Vochy. Treman.				1/3				1/2NR 1/2R	1/1R	2/5				1/1ot 1/2M	2/2	2/4	1/4	1/4	1/1			1/1		1/1	1/1				2/2						
Poly.				1/4		2/2		1/4NR 1/1PK	1/1	2/7	1/2			1/3ot 1/1B-M	1/1	3/5		1/1	1/3		1/1	1/1	1/1	1/1	1/1	1/1	1/1	1/1	1/1	1/1 (root)	1/2*	1/1		1/1	

Key to symbols: NR = no red; R = red; PK = pink; PK-B = pink-brown; ot (in Ehr.) = color other than magenta or blue;  
M = magenta; PU = purple; OM = orange-magenta; ot (in HCN) = part other than leaves and/or tips used  
in investigation; sd = seeds; B-M = brown-magenta

\*saponin found in plant;  
part investigated unknown



Family	H.W. & Cig. tests				or	?	Syringin test			Raphides		?	Ehrlich		HCN		?	HCl/Meth.		L.A.	-	?	Juglone		fl.		?	Tannin		?	Saponin					
	I	II	III	IV			+	-	?	+	-		+	-	+	-		+	-				+	-	+	-		+	-		A+	-	?	+	-	
Dichap.						1/1		1/1NR			1/1		1		1/3PKB						1/1				1/1		1/1			1/1						
Callit.													1/1		1/1M	1/1ot	1/1		1/1		1/1			1/2												
													1/1(part used unknown)																							
Balsam.				1/2		1/4		1/1R	1/1R?	2/9		1		1/2M	1/1	1/3	1/1		1/1	1/5					1/4	1/3		1/1					1/1	1/1?		
Buxac.				3/3		2/2		3/9NR	1/1		4/11				1/1PKY		4/11		4/9		2/2	3/10	1/1		4/8	4/5	1/1		2/3	2/4	1/1		1/1		1/1	
								1/1PK							2/5ot		1/1sd																			

Key to symbols: NR = no red; R = red; PK = pink; PKB = pink-brown; M = magenta; PKY = pink-yellow; ot (in Ehr.) = colors other than those symbolized obtained; ot (in HCN) = parts other than leaves or tips used; sd = seed



[illegible]



	gent.	fer.	caff.	ell.	sin.	p-c	gall.	umb.	van.	syr.	rutin	p-OH-B	p-cat.	scop.	aesc.	melil.	P/L	phlor.	H-G
Simarubaceae																			
Surianoideae			+1/1														-3/10	-3/10	-3/10
Simaruboideae	-1/1	-1/1		-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1		-1/1	-1/1	-1/1		-1/1	-1/1	-1/1	-1/1
	+2/3	+1/2	+1/4	+2/4			+1/1				+1/1	-1/1	+1/1	+1/1					
	-1/1	-2/2	-2/2		-2/4	-1/2	-1/1	-1/2	-1/2	-1/2	-1/1	-1/2	-1/1		-1/2	-1/2	-1/2	-1/2	-1/2
		?1/1		?1/1	?1/1									?1/1					
Kirkioideae	?1/1	-1/1	+1/1	+1/1	-1/1	-1/1	+1/1	-1/1	-1/1	?1/1	+1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1
Burseraceae	+1/1	+1/1	+2/2	+1/1		+1/2	+1/1										-1/1	-1/1	-1/1
	-1/1	-1/2	-1/1	-1/2	-2/3	-1/1		-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1
Meliaceae																			
Cedreloideae		+1/1	+2/2	+1/1		+2/3													
	-1/1	-1/2	-1/1		-1/2		-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1		-1/1	-1/1	-1/1	-1/1
					?1/1										?1/1				
Swietenoidae	+1/1	+2/2	+2/2	+2/2		+2/2	+1/1		+1/1		+1/1	+1/1							
					-2/2			-2/2	-1/1	-2/2	-1/1	-1/1	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2
	?1/1						?1/1												
Melioidae	+1/1	+4/4	+3/5	+3/4	+3/3	+2/2	+1/1			+1/1									
	-2/2		-1/1		-2/2	-3/3	-3/3	-3/4	-2/2	-2/2	-3/3	-3/4	-3/3	-3/4	-3/4	-3/4	-3/4	-3/4	-3/4
	?1/1	?2/2			?1/1	?1/1			?2/2	?1/1	?1/1		?1/1						
Malpighiaceae	+1/1	+1/2	+2/2	+1/2	+1/1	+2/2													
	-2/2	-2/2	-3/3	-1/1	-3/3	-3/3	-2/2	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3	-2/3
	?1/1	?1/1			?1/1		?1/1												
Trigoniaceae																			
Vochysiaceae																			
Tremandraceae				+2/2		+1/1	+2/2		+1/1										
	-2/2	-2/3	-2/3		-2/3	-1/1		-2/2	-1/1	-1/1	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2	-2/2
						?1/1				?1/1									
Polygalaceae	+1/1	+1/5	+1/2	+1/1	+1/4	+1/3			+1/1										
	-1/1	-1/1	-1/3	-1/1	-1/2	-1/3	-1/2	-1/2	-1/1	-1/2	-1/2	-1/2	-1/2	-1/2	-1/2	-1/2	-1/2	-1/2	-1/2
			?1/1																
Dichapetalaceae																			
Callitrichaceae	-1/1	-1/1	+1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1		-1/1	-1/1	-1/1		-1/1	-1/1	-1/1	-1/1
Balsaminaceae	+1/11	+1/7	+1/10	+1/2	+1/5	+1/12			+1/7	+1/1		+1/12	+1/3	+1/13					
	-1/5		-1/6	-1/3	-1/14	-1/2	-1/2	-1/2	-1/11	-1/17	-1/1	-1/5	-1/14	-1/4	-1/2	-1/2	-1/2	-1/2	-1/2
	?1/2	?1/1	?1/3								?1/1	?1/1	?1/1	?1/1					
Buxaceae																			
Buxee	+2/2	+3/6	+1/1	+3/4	+2/4	+3/4			+1/1			+1/2	+1/1						
	-1/2	-1/1	-3/5	-3/4	-2/3	-2/3	-3/5	-3/5	-3/4	-3/5	-2/3	-3/4	-2/3	-3/4	-2/2	-3/4	-4/5	-3/6	+1/1
	?2/2	?1/1	?1/1			?1/1							?1/1	?1/1	?1/1	?1/1	?1/1		-3/5
Stylocereae																			
Simmondsieae		+1/1	+1/1	+1/1	+1/1	+1/1			+1/1	+1/1									
	-1/1						-1/1	-1/1			-1/1	?1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1	-1/1



## DISCUSSION OF RESULTS

### A. Analysis of the "Geraniales" s.s.

individual

#### 1. The/families included in the order (according to the 12th "Syllabus")

a. Geraniaceae. Metcálfe and Chalk (1950) stated that except for Sarcocaulon, anatomically, the Geraniaceae forms a homogeneous group. Thus if their statement is correct, then chemically, this family should consist of species which react similarly to various chemical tests.

Unfortunately, it was impossible to obtain material of species other than those belonging to the following genera: Geranium, Erodium, and Pelargonium. These genera all belong to the subfamily Geranieae. Thus it will be impossible to discuss whether Hutchinson's (1959) splitting of the Geraniaceae into Ledocarpaceae, Vivianiaceae, and Geraniaceae s.s., is merited. The subsequent placing of these families into two lines of evolution, that is, Lignosae and Herbaceae, likewise cannot be discussed.

According to the results obtained, there is no evidence of the presence of polyphenolases in members of this family. Geranium and Erodium gave negative reactions, while many Pelargonium species gave an "oxalis-reaction", thus indicating high acid content in the cell sap and a close affinity with Oxalidaceae.

The nine species investigated in the Syringin test were negative; one Erodium species gave a pink color reaction, while two Pelargonium species gave a red color in the cross-sections of the stem.

As in the two previous tests, all results from the Ehrlich test were negative, thus indicating the absence of aucubin or aucubin-like substances in the leaves. Most specimens gave a pale yellow color, or hardly darkened at all upon heating. The magenta color reaction in Pelargonium correlated with the presence of a red color in the Syringin test.

The vast majority of this family are herbaceous; therefore it was impossi-

ble to do the HCl/Methanol test on the three genera. However Pelargonium did have some members sufficiently woody to be tested. Strips of wood were used, not shavings. As would be expected from the results obtained in the Syringin and Ehrlich tests, there were several species which gave a positive result. Thus, in reference to Pelargonium, the reactions were mixed.

Mostly negative results were obtained from the leucoanthocyanin test. Again Pelargonium gave mixed results. Although most species were negative in respect to this test, one species, P. peltatum, was doubtfully negative. There are many horticultural varieties of this species, and they seemed to differ in the Syringin-Ehrlich-HCl/Methanol-Leucoanthocyanin correlation. Often three correlated but the fourth differed. These experiments were repeated using the same horticultural variety, but the results obtained were the same.

Raphides are absent in this family. According to Metcalfe and Chalk (1950), solitary and cluster crystals which are widespread in the plant kingdom characterize this family. To a lesser degree, styloids and crystalline masses are present. Crystalline masses are also present to a small extent in Zygophyllaceae, a family often placed near Geraniaceae; but Euphorbiaceae, Rutaceae, Simarubaceae, and especially Malpighiaceae, contain them too. The solitary or clustered crystals are sometimes situated in idioblasts in Erodium. Styloids are present in Rhynchotheca, while sphaerocrystalline masses are found in Erodium and Monsonia. Short rows of cells containing cluster crystals characterize Wendtia.

As in all the families (except Polygalaceae), members of the Geraniaceae do not contain naphthoquinones. Fluorescence was observed in all species tested.

There were many species which were doubtfully positive in the tannin test. These gave a purple color on the filter paper; because of this masking effect, it was impossible to say with any degree of accuracy whether the results were positive. This color may have been due to anthocyanins. Milogradova and Mak-

hamadzhanov (1961) investigated two species of Geranium, G. collinum and G. rectum, and found the leaves and stems to contain tannins of the pyrogallol group. In addition, Baytop and Tarcan (1962) found the stalk of Pelargonium endlicherianum to contain tannin. Thus, in all probability, Geraniaceae is a tanniniferous family.

Just one species was tested in the saponin tests A and B; a positive result was obtained for part B. The leaves did not contain saponin. Baytop and Tarcan (1962) found the stalk of Pelargonium endlicherianum to contain no saponin.

Stafford (1961) investigated this family and found the leaves to be very low or non-accumulators of tartaric acid. Aluminum is not accumulated.

Although one Pelargonium gave a questionable reaction in the HCN test, the rest of the species tested of this genus and others were negative.

Group C phenolic constituents were limited in distribution. Group B compounds, especially gallic acid, were present, while members of group A phenolic compounds were widespread in this family.

The widely-distributed flavonols, kaempferol and quercetin, are present, while myricetin is absent (Bate-Smith, 1962).

Alkaloids have been recorded in Erodium, but there are no records of them in Pelargonium and Geranium (Gstirner, 1963; Gegil, 1964). Willaman and Schubert (1961), in their compilation of alkaloids found in plants, have records of unknown alkaloids being present in Geranium and Biebersteinia.

b. Oxalidaceae. Oxalidaceae has been associated with Geraniaceae by many authors, although van Tieghem and Constantin (1918) created an order Oxalidales and placed in it Oxalidaceae, Zygophyllaceae, Linaceae, Erythroxylaceae, Tropaeolaceae, Trigoniaceae, Tremandraceae, Cruciferae, and Frankeniaceae. If most authors are correct in their assumption, that is, that Oxalidaceae is closely-

allied to Geraniaceae, then members of this family should be chemically closely related to the Geraniaceae.

Only three genera were investigated: Oxalis, Averrhoa, and Biophytum.

Oxalis either gave a negative or "oxalis-reaction" in the hot water or cigarette tests, while species of the other two genera differed in giving "III" reactions. Thus Biophytum and Averrhoa differed from members of the Geraniaceae.

Closely-correlated with members of the Geraniaceae, Oxalis gave negative-non-red reactions in the Syringin test. Again Biophytum and Averrhoa differed, both giving negative reactions, but a red color in the xylem and/or fibres.

In the Ehrlich test, all results were neg. Brown colors, as well as a pink and a magenta one, were noted in Oxalis; many members of this genus have pink pigments in the petioles and parts of the leaves. Bate-Smith (1962) recorded the presence of delphinidin and cyanidin in the leaves of Oxalis and Averrhoa. Thus this may be the cause of the deviation of Oxalis from the Syringin-Ehrlich correlation. Averrhoa and Biophytum deviated from the norm in that both gave magenta reactions.

As in the Geraniaceae, most species are herbaceous. Thus it was impossible to employ the HCl/Methanol test to any extent. The Oxalis species tested had very little wood, so it was impossible to say with any degree of certainty whether the reactions were negative or positive. Averrhoa and Biophytum gave decidedly positive results.

Oxalis gave mixed reactions in the Leucoanthocyanin test. As mentioned previously, Oxalis species have anthocyanin-containing leaves. Thus this is probably responsible for the positive results. As expected, Averrhoa and Biophytum both gave positive results.

Thusfar Averrhoa and Biophytum have differed from the type of this family. Hutchinson (1959) removed Averrhoa from this family and raised it to the familial

rank. His move seems justified.

All species tested were negative to the HCN test, thus indicating the absence of cyanogenetic glycosides.

Raphides are likewise absent; however other calcium oxalate crystals have been noted by the author and others. Metcalfe and Chalk (1950) found solitary crystals to be present, and to a lesser degree, crystal sand.

No naphthoquinones were recorded; fluorescence was observed. Thus a consistency with the results obtained in the Geraniaceae.

The results in the tannin test were rather mixed. Oxalis species were predominantly negative, while the other two genera gave positive reactions.

Negative results were obtained from the Saponin A test. However, a negative reaction was obtained in Oxalis and a positive reaction was obtained in Averrhoa in part B.

Stafford (1959) examined the leaves of Oxalis, and found it (Oxalis) to be a very low or non-accumulator of tartaric acid. In this respect, it is similar to Geraniaceae. In addition, both families do not contain cyclitols (Plouvier, 1963).

No group C phenolic constituents were found; members of group B phenolic compounds were sparse, while group A compounds were found in the three genera. Gallic acid of group B must be a familial characteristic in Geraniaceae, for as noted in Oxalidaceae, and as will be noted in other geranialean families, this phenolic substance is absent.

Except for a trace of quercetin in Oxalis dispar, the three flavonol compounds are absent from this family.

c. Tropaeolaceae. Members of this family agreed with the Geraniaceae in the hot water and cigarette tests, syringin, leucoanthocyanin, and HCN tests. They also gave negative results in the Juglone test and positive fluorescence.

Bate-Smith (1962) recorded no anthocyanins in this family, and this agrees

with the results obtained.

Hutchinson (1959) placed this family in the Herbaceae. Therefore none could be tested for the HCl/Methanol test. But since negative results were obtained in the leucoanthocyanin test, it can be assumed that the results from the HCl/Methanol test would have been negative also.

Raphides are absent from this family. Calcium oxalate crystals other than raphides have been recorded in this family (Metcalfé and Chalk, 1950).

Metcalfé and Chalk also recorded the presence of myrosin cells in the sub-epidermal region as well as in the phloem of the stem. Myrosin cells have also been reported in the primary cortex and phloem of the root. Thus this characteristic separates this family from the Geraniaceae in which it was placed by Bentham and Hooker (1862-1883). Myrosin also characterizes Cruciferae, Resedaceae, and Capparidaceae. This is probably a case of parallel evolution.

Peter (1964) investigated the families Cruciferae and Tropaeolaceae, and found the families to contain mustard oil glucosides. Leucoanthocyanins, tannins, and polyphenolases were absent. In addition, it is significant that erucic and eicosenoic acids are the major fatty acids in Cruciferae and Tropaeolaceae. It is amazing that through parallel evolution, these two families should have a multitude of things in common.

Group C compounds are absent. Group B constituents are absent except for p-coumaric acid. Members of group A phenolic compounds are present.

Kaempferol, myricetin, and quercetin are absent.

d. Zygophyllaceae. Zygophyllaceae has been allied with Geraniaceae by many authors. Hutchinson (1959), however, removed this family from the "Geraniales" and placed this family in the Malpighiales in the Lignosae. Thus, if his

scheme is correct, Zygophyllaceae should differ greatly from Geraniaceae. Heimsch (1942) also stated that this family should be placed with less-herbaceous families.

This family agreed with Geraniaceae in the hot water and cigarette tests, the syringin, Ehrlich, leucoanthocyanin, and HCl/Methanol tests. Although a trace of HCN was observed in one species, the consensus was that all members are negative or show mere traces of cyanogenetic glycosides. The Juglone test was negative, and fluorescence was noted. Four out of five species contained no tannin in the leaves. Thus Hutchinson's placement of this family in the Malpighiales is challenged.

Raphides were not observed in the subfamilies other than Peganoideae. Metcalfe and Chalk (1950) recorded raphides in Peganum. This is one piece of evidence for raising this genus to the familial level as van Tieghem and Constantin (1918) did. They also included Malacocarpus in this family. It is now included in the genus Peganum.

Calcium oxalate crystals, other than raphides, have been recorded in Zygophyllaceae. Solitary and cluster crystals are dominant, but styloids, acicular crystals, and crystalline masses are found to a lesser extent. Crystals, however, are absent in Balanites, which Hutchinson (1959), for example, raised to the familial rank.

D-pinitol was recorded in Zygophyllum, but it is absent in Peganum. This is another argument for excluding Peganum from Zygophyllaceae. Of course one cannot exclude a genus from a family on the basis of one character, but on the basis of a multitude of characteristics.

Peganum is recorded as containing a host of alkaloids, most belonging to the indole group: harmine, harmalol, harmaline, and vasicine or peganine. Siddiqui et al. (1964) investigated P. harmala and found it to contain additional

alkaloids in the seeds: harmidine and pegaline. These alkaloids have not been reported in other members of the Zygophyllaceae. Therefore this is additional evidence for excluding this genus from the family.

Syringic and vanillic acids have been recorded in Zygophyllaceae, therefore differentiating this family from Geraniaceae. Group A compounds are present, and group B, but to a lesser degree.

No records of the chemistry of Nitraria were found. Thus the question of whether this genus should be raised to the familial rank remains unsolved.

e. Linaceae. This family is considered by some to be highly-unnatural. It has been divided into many families, for example, Hugoniaceae, Nectaropetalaceae, Ctenolophonaceae, and Ixonanthaceae. Unfortunately, except for Ixonanthes, none of the other genera were investigated for their chemical components.

Negative results were obtained for the syringin (no red was observed), Ehrlich, HCl/Methanol, raphide, leucoanthocyanin, and Juglone tests. These results agree with those obtained for Geraniaceae.

Cyanogenetic glycosides were present in Linum. Many people have recorded the presence of linamarin in L. usitatissimum. Butler (1965) found that plants previously reported to contain either linamarin or lotaustralin were found to contain both cyanoglucosides. This chemical characteristic separates Linum from the Geraniaceae. However, Dillemann (1953) reported that in Linaria, the presence of cyanogenetic glycosides is determined by one gene, this acting independently of those controlling morphological characters. Thus through independent evolution, this chemical characteristic could have arisen many times. Therefore unrelated families could possess cyanogenetic substances.

The majority gave negative reactions in the tannin test. Negative



results were also obtained from the saponin A - B test. Amarasingham (1964) also obtained negative results from the saponin test.

Bate-Smith (1962) has no records of myricetin, quercetin, and kaempferol in the family.

Group C phenolic compounds were absent, while groups A and B were found in various species.

f. Erythroxylaceae. Two species of Erythroxylum were tested.

This family has been placed in Linaceae by several authors (see Review of Literature). However, as will be seen, this family differs significantly from Linaceae. Thus its exclusion from that family is supported.

This family has also been placed in the Malpighiales by Hutchinson (1959). Thus it should possess characteristics chemically different from Geraniaceae.

The two species of Erythroxylum differed in their reactions to the various tests. For example, in the syringin test, both were negative, but a red color was observed in one species only. The Ehrlich and HCl/Methanol results corresponded with these findings. However, the species which gave a positive HCl/-Methanol reaction gave a negative leucoanthocyanin reaction. The first test is of course on wood, the second on leaves. Leucoanthocyanins, often associated with a positive HCl/Methanol reaction, must be absent from the leaves.

Cyanogenetic glycosides were absent, as well as naphthoquinones. Fluorescence was observed in one species.

Bate-Smith (1958) found this family to be tanniniferous. However tannin was found in trace amounts in one species only.

Rowson (1958) studied the alkaloids of this family, and found it to contain alkaloids of the tropane series. He said this group of alkaloids is highly specific, being thusfar isolated in Erythroxylum coca and E. truxillense. If for this reason only, this family should be separated from Linaceae.

Group C products were absent, while group A and B phenolics, with the exception of gallic acid, were present.

g. Limnanthaceae. Floerkia and Limnanthes were both tested by the author. Both gave negative syringin reactions; however, a red color was observed in Limnanthes. The presence and absence of the red color in the syringin test correlated with the data obtained in the Ehrlich, or leucoanthocyanin tests.

Raphides are absent.

Consistent with the Geraniaceae, <sup>no</sup> naphthoquinones were recorded. Fluorescence was observed. One genus showed a trace of HCN.

Tannin cells have been reported by Metcalfe and Chalk (1950), and these are found on the lower side of the leaves in Limnanthes. None have been found in Floerkia.

Maclay et al (1963) reported that Limnanthes douglasii is the richest source of glycerides with C<sub>20</sub> and C<sub>22</sub> straight chain acids. The chief fatty acid is eicosenoic.

Members of this family contained no group C phenolic constituents. Group B compounds were absent except for p-coumaric acid. Except for gentisic acid, group A compounds were present.

Although this group is a sound group anatomically, the two genera seemed to differ in their chemical reactions. Thus further investigations should be done on this family.

h. Euphorbiaceae. This is an extremely large family with diverse chemical characteristics. It is thought to be of polyphyletic origin.

There are two sections in this family, each with two subfamilies. Plant material was obtained from all subfamilies, and as would be expected if this family is of polyphyletic origin, the results were mixed.

The subfamily Phyllanthaceae gave mixed reactions for the hot water and cigarette tests. The other three taxa were more consistent. While the first subfamily showed some evidence of the presence of polyphenolases, the other subfamilies were negative. However, some doubtful "oxalis-reactions" were obtained in Crotonoideae.

Several genera in the first two subfamilies gave questionable results in the syringin test. These were doubtfully positive. Most, however, were negative and contained no red color.

As would be expected, the reactions from the Ehrlich tests were mixed. Many of the species which were tested gave a pink, magenta, or purple spot. This is inconsistent with the data obtained in the syringin test. If the vast majority gave no red color in the latter test, then they should likewise give no pink, magenta, or purple spot in the Ehrlich test. Again this must be due to the absence of leucoanthocyanins in the stems, and the presence of them in the leaves.

Mixed reactions were obtained for the HCl/Methanol test. However, more were negative. Since the same section of the plant was used here as for the syringin test, the results agree. Similar results were also obtained for the leucoanthocyanin test.

Phyllanthus paniculatus Oliv. is said to have raphides in the wood. However, raphides were not observed in any of our sections. Solitary and cluster crystals are present, but styloids and sphaero-crystals are found in a few species. (Metcalfe and Chalk, 1950).

As would be expected in a family of polyphyletic origin, mixed results were obtained for the HCN test.

The results for the Juglone test and for the fluorescence were consistent with those obtained from the Geraniaceae.

Tannin was present in nearly all the species tested. This is inconsistent with Bate-Smith's (1957) remark that this family can be considered non-tanninifer-

ous. Tanniniferous cells have been recorded (Metcalf and Chalk, 1950).

Amarasingham and his co-workers (1964) found saponin to be absent in 41 out of 46 species tested, but it was recorded in Glochidion, Cleistanthus, Drypetes, and Galearia.

Many of the members of this family are positive for one or another phenolic compound of the group C series. The three subfamilies investigated for phenolic compounds differed in the distribution of groups A and B phenolic constituents. Crotonoideae, for example, was positive for gallic acid, as Geraniaceae, but the other two subfamilies were generally negative.

The major fatty acids in the seeds are linoleic, oleic, and conjugated polyethenoic acids. These conjugated polyethenoic acids are found in various species:

Aleurites cordata -  $\alpha$ -elaeostearic  
Ricinus communis - ricinoleic  
Cephalocroton cordofanus - vernolic  
Mallotus philippinensis -  $\alpha$ -kamlolenic  
Sapium sebiferum - deca-trans - 2,cis 4 - dienoic  
Sebastiania ligustrina - dodeca - trans - dienoic

This would support the polyphyletic origin of the Euphorbiaceae.

In general, the polyphyletic origin of this family is supported.

i. Daphniphyllaceae. This family has been included in Euphorbiaceae. However that family is so chaotic that it is doubtful whether one can really say from the chemical tests employed whether the exclusion of this genus Daphniphyllum from Euphorbiaceae is justified.

Unfortunately, no fresh material was available during the author's tenure of research. Thus information on the chemistry of this family is incomplete. These results can be seen in the tables. Negative non-red reactions were obtained in the syringin test. No result/for the hot water and cigarette tests. A positive HCl/Methanol reaction and a questionable leucoanthocyanin result were obtained.

No raphides were observed. Cluster crystals have been reported in this family.

Bate-Smith (1957) investigated and thus considered this family tanniniferous.

No results were obtained for the group C phenolics. The data on groups A and B were incomplete.

## 2. The families as an order "Geraniales" s.s.

Generally speaking, except for Euphorbiaceae, the families seem to form a natural group. Except for Linaceae which gave a "I" or "II" reaction in the hot water and cigarette tests, most gave a "IV" or "o.r." reaction. All syringin tests were negative, and the presence or absence of a red color was mixed, although more were negative.

Many plants were herbaceous. Thus, in these, the HCl/Methanol tests were not performed. The reactions were mixed, especially in Euphorbiaceae. However, again, the majority of the species were negative in their response. The results were closely correlated with the leucoanthocyanin test.

All species were negative for the Ehrlich test, and if a magenta spot appeared, it was correlated with the other tests. A few members of the Euphorbiaceae, however, gave no red color in the syringin test and a negative result for the HCl/Methanol test, while giving positive results in the leucoanthocyanin test and magenta spots in the Ehrlich test.

HCN was absent in most families. If present, it is a familial character, and probably arose through independent evolution.

Naphthoquinones were absent from all the families. Some fluorescence was observed in all the families investigated. None of the fluorescences was bright enough to be indicative of coumarins.

Mixed results were obtained for the tannin test. Many were positive, especially in the Euphorbiaceae.

Saponins were generally absent, being present only in several members of the Euphorbiaceae. The saponin B test results were mixed, but many were negative. These tests must be carried out more extensively and intensively before anything more conclusive can be said.

Group C phenolic constituents were rarely found in the families. In Euphorbiaceae, they were more frequently found than in the other families. The other compounds of groups A and B were found in varying degrees. Some compounds, for example, gallic acid in Geraniaceae, were familial characteristics.

Except for raphides in Peganum of Zygophyllaceae, these calcium oxalate crystals were absent. Other calcium oxalate crystals, for example, the widely-distributed solitary and cluster crystals, were present. The other types varied in their distribution, and are therefore familial rather than ordinal characteristics.

Most of the families are alkaloid-containing. There are no records of alkaloids in Limnanthaceae, Oxalidaceae, Tropaeolaceae, and Linaceae. Thus in all probability, these families do not contain them.

Cyclitols are absent, except in Euphorbiaceae in which D-inositol has been recorded and Zygophyllaceae in which D-pinitol has been reported.

The fatty acid composition of the seeds vary, but Linaceae, Zygophyllaceae, Euphorbiaceae, and Daphniphyllaceae contain similar ones. Euphorbiaceae also contains some unusual acids.

As for aluminum accumulation, all the species were negative except Daphniphyllum species and several members of the Euphorbiaceae.

There is no evidence for Hutchinson's split of the "Geraniales" s.s. into the Geraniales and Malpighiales. In addition, there is no evidence for his placing the two orders in two lines of evolution, namely, the ~~Herbaceae~~ and

Lignosae. Additionally there is no evidence to support the other authors who have included just a few of these nine families in their Geraniales.

## B. Analysis of the Rutales

### 1. The families included in the Rutales

a. Rutaceae. This is obviously a very diverse and unnatural family. Thus the chemical reactions and other results obtained were mixed. There is so much confusion in this one family that a book in entirety could be devoted to this family. This will become more apparent as the chemical characteristics are discussed.

The presence of polyphenolases is indicated only in the first subfamily Xanthoxyleae; the majority of the members of this family are characterized by their absence. The first subfamily also had two questionable "o.r." reactions. This trend towards the absence of polyphenolases is consistent with the findings of the Geraniaceae.

One member belonging again to the subfamily Xanthoxyleae was recorded as being positive for the syringin test. While other members were doubtfully positive, the majority were negative. Most of the plants did not have any red color in the cross sections.

There was some inconsistency in the Ehrlich, HCl/Methanol, and leucoanthocyanin correlations. This was also observed by Dr. R. D. Gibbs. Many species gave a negative HCl/Methanol result, a magenta spot in the Ehrlich test, a positive leucoanthocyanin result, and a non-red reaction in the syringin test.

Thus it was that many plants gave a magenta color in the Ehrlich test, while as mentioned previously, the majority of the species contained no red color in the syringin test. Many of the leucoanthocyanin results were positive, while the results for the HCl/Methanol test were largely negative.

Raphides were absent in all the genera investigated excepted members of the

Xanthoxyleae. Raphides were limited in their distribution and were present in one subtribe: Cuspariinae. These genera are listed on the following page. Thus six out of sixteen genera in that subtribe are raphide-containing. They are also found in the same region of the world. This fact alone suggests that Rutaceae is an unnatural group.

Tannins were found in many of the species. This is consistent with Bate-Smith's findings (1957).

Naphthoquinones were absent, and all solutions fluoresced. The presence of coumarins was characterized by very brilliant fluorescences. Coumarins, found in this family, have not been reported in allied families of the Rutaceae, including Meliaceae, Burseraceae, Simarubaceae, Zygophyllaceae, and Cneoraceae (Price, 1963).

Anthraquinones have been recorded in the Rutaceae, but are absent from other members of Engler and Diels' (1936) "Geraniales".

Several genera of the Aurantioideae contain saponins. A positive result was obtained in the saponin B test. Thus in part A, there is some inconsistency.

The family is highly-diverse in its alkaloids. Price (1963) wrote a comprehensive paper on the distribution of alkaloids in this family. A summary of his findings can be found in the Appendix. As noted, furoquinolines are found in four of the subfamilies, and this perhaps link them together. Quinazolines are found in five other families including the Zygophyllaceae.

Bate-Smith (1958) reported that berberine alkaloids of the isoquinoline group are found only in the Ranales, Rhoeadales, and Rutaceae. Hegnauer (1958) believed the presence of protoberberine, aporphine, and chelidonine alkaloids in this family is indicative of a closer relationship to the Ranalean line. In 1963, Hegnauer added that this family is related to Polycarpicae via benzylisoquinoline alkaloids. He presented the simi-



## The Occurrence of Raphides in Rutaceae

I. Subfamily Rutoideae  
 Tribe Cusparieae  
 Subtribe Cuspariinae

genus	No. of species	Raphides	locality
<u>Spiranthera</u> A. St. Hil.	1	-?	Brazil
<u>Almeida</u> A. St. Hil.	4	-?	Brazil
<u>Euxylophora</u> Huber	1	-?	Brazil
<u>Adiscanthus</u> Kucke	1	-?	Amazon Valley
<u>Leptothyrsa</u> Hook. f.	1	-?	N. Brazil
<u>Ticorea</u> Aubl.	3	-?	French and Dutch Guiana, Amazon region
<u>Lubaria</u> Pittier	1	-?	Venezuela
<u>Rauia</u> Nees et Mart.	2	+ / styloids	S. Brazil
<u>Galipea</u> Aubl.	8	+ / styloids	Guiana, Brazil
<u>Raputia</u> Aubl.	9	+	tropical America
<u>Decagonocarpus</u> Engl.	1	-?	Amazon
<u>Erythrochiton</u> Nees et Mart.	5	+ / styloids	tropical America
<u>Cusparia</u> Humb.	25	crystal-sand	tropical America, Brazil
<u>Naudinia</u> Planch.	1	-?	Colombia
<u>Ravenia</u> Vell.	10	+ / styloids	Antilles, Brazil
<u>Monniera</u> L.	2	+ / styloids	equatorial America, tropical S. America

larities between Phellodendron amurense and Polycarpicae, and there is a close resemblance. This table is presented on the following page.

Quinazoline and harman alkaloids have been found in Peganum harmala which was placed by Hutchinson in Rutaceae; he then transferred it to Zygophyllaceae. Peganum species have several things in common with Rutaceae, including the alkaloids and the presence of raphides (Metcalf and Chalk, 1950; Price, 1963).

Hutchinson (1959) places Flindersia and Chloroxylon in Meliaceae. Dads-well (1938) suggests a separate family Flindersiaceae. Referring to alkaloids, one Chloroxylon species and 13 out of 14 Flindersia species (the exception is F. brayleana) contain furoquinolines. Furoquinolines are found only in Rutaceae. Secondly coumarins have been isolated from Chloroxylon and Flindersia, but none have been found in Meliaceae. Thus the subfamily Flindersioideae is chemically distinct from Meliaceae.

No aluminum accumulators were found.

In many ways, this family is homogeneous. But the degree of deviations is sufficient to make this an unnatural group. Thus further investigation is necessary. Perhaps one order should be created to contain the families which will eventually be created out of this one family. The sporadic appearances of group C phenolic constituents also support this idea.

b. Cneoraceae. This family is much like Geraniaceae in many respects. It gave a "TV-III" reaction for polyphenolases; most likely this is a negative reaction. In addition, it gave a negative-non-red syringin reaction that agreed with the negative leucoanthocyanin, Ehrlich, and HCl/Methanol tests. Raphides were absent, as well as naphthoquinones. Tannins were present, and saponins were absent.

Group C compounds were absent, and groups A and B compounds were present.

A Comparison between Polycarpicae and Phellodendron amurense

(from Hegnauer, 1963)

Substance	Polycarpicae	<u>Phellodendron amurense</u>
SiO <sub>2</sub> in membranes	widespread in woody members	+; also in other members of Rutaceae
Alkaloids: berberine palmatin jatrorrhizine magnoflorine	widespread	+
Essential oils	common in woody members (oil cells)	+; common in Rutaceae lysigenic cells
Ferulic acid	common	+ (lumecaerulic acid)
Amurensin	des-o-methylicariin from <u>Epimedium</u> has same aglycone	+
Isoprenoid bitter principles	columbine (C <sub>20</sub> ) in Menispermaceae	limonin (C <sub>26</sub> ) obacunone (C <sub>26</sub> )

c. Simarubaceae. As discussed in the Review of Literature, this family seems to be very diverse, and many authors have raised the subfamilies to the familial rank.

From the data obtained in the laboratory and from other sources, this seems to be a chemically-heterogeneous family. For example, Suriana of Surianoideae gave a "II" reaction in the hot water and cigarette tests; Picraena of Simaruboidae gave a "IV" reaction, while Kirkia of Kirkioideae gave a "TII" reaction. No members of Irvingioideae were tested. Thus, in this respect, only Picraena agrees with the results of Geraniaceae.

All species investigated were negative to the syringin test; however, members of subfamilies two and three did not show any red color in the cross-sections, while Suriana gave a red color. These results were consistent with those obtained for the Ehrlich, HCl/Methanol, and leucoanthocyanin tests.

No raphides were seen in the sections. The occurrence of solitary and cluster crystals have been reported. Styloids have been reported in one genus Alvaradoa. The size and distribution of the cluster crystals in Castela, Holacantha, and Picramnia are said to be of value in the identification of the genera. (Metcalfe and Chalk, 1950)

Cyanogenetic substances as well as naphthoquinones were absent. Fluorescence was noted.

The plants tested were positive in the tannin test. Amarasingham et al (1964) obtained a negative result for the saponin A test.

The fatty acid composition of the seeds is interesting. Shorland (1963) stated that "the members of the family Simaroubaceae thus show such wide variation in the composition of their seed fats as to prompt further inquiry into their botanical classification."

GENUS	CHIEF FATTY ACID(S)
<u>Ailanthus</u>	oleic, linoleic
<u>Picrasma</u>	petroselenic
<u>Picramnia</u>	tariric
<u>Irvingia</u>	myristic, lauric

These four genera are distributed among three subfamilies. Each contains a different fatty acid. This would be one source of evidence for splitting this family as Hutchinson (1959) did. The presence of myristic and lauric acids in Irvingia and other genera links this family with Vochysiaceae.

Results of the phenolic compound chromatography were mixed and comparable to Rutaceae's, but on a smaller scale. Group C compounds were present in subfamilies two and three, but absent in the first one. Group B substances were present again in subfamilies two and three, but absent in one. Only gallic acid of this group was present. Group A phenolics were present.

d. Picrodendraceae. The only genus in this family was formerly placed in the Simarubaceae. It gave a "IV" reaction to the hot water and cigarette tests. However it diverged from both Picraena and Geranium's results in that it gave a negative but red color in the syringin test. This was consistent with the data obtained from the Ehrlich and HCl/Methanol tests.

The absence of raphides, cyanogenetic glycosides, and naphthoquinones, and the presence of some fluorescence, are consistent with the results obtained for Geraniaceae.

e. Burseraceae. This family has been assigned to the Geraniales, Rutales, and Sapindales. It is also said to have affinities with Anacardiaceae, Meliaceae, Rutaceae, and Simarubaceae.

Very few members of this family were tested. The results for the hot water and cigarette tests were doubtful. All specimens were negative in the syringin test, and red was noted in one case only. The results were consistent with those obtained for the HCl/Methanol and leucoanthocyanin tests.

No raphides were seen; Metcalfe and Chalk (1950) recorded cluster and solitary crystals in this family.

Naphthoquinones were absent, and fluorescence was observed. Bursera has been recorded as cyanogenetic, while Commiphora has not. Tannins are doubtfully present. In addition, Amarasingham et al (1964) investigated four <sup>group of</sup> genera and six species for saponins, and found this/chemical substances to be absent.

Group C phenolic constituents were absent; group A compounds were present, while p-coumaric and gallic acids of group B were present.

The presence of stearic acid as the major fatty acid of the seed links this family with Meliaceae.

f. Meliaceae. There is general disagreement as to which genera should belong to this family, which families it should be allied with, and which subfamilies should be raised to the familial rank. Hutchinson (1959) further complicated things by raising this family to the ordinal rank.

Generally polyphenolases were absent from the species tested. In the syringin test, negative results were obtained. Four out of six species contained some red color in the xylem and/or fibres. These results were consistent with those obtained from the Ehrlich, HCl/Methanol, and leucoanthocyanin tests.

Raphides were absent.

Except for members of the Melioideae, all species investigated contained no HCN. As in the other families, the Juglone test results were negative. Fluorescence was observed.

All species tested except one were tanniniferous.

Amarasingham et al (1964) tested about 19 species and found the majority to contain no saponins. Chisocheton and Dysoxylum of the subfamily Melioideae were positive. The saponin test B was positive.

In general, group C compounds were absent; however Swietenioideae was similar to Rutaceae and Simarubaceae, in that it contained p-hydroxybenzoic acid, rutin, and vanillic acid. Gallic and sinapic acids of group B were absent. Group A compounds were present.

As mentioned in a previous section, the chief fatty acid of the seed is stearic, and this is a cross-link with Burseraceae, a family with which it has often been associated.

g. Akaniaceae. This family has been placed in the Geraniales, Rurales, and Sapindales. In contrast to Geraniaceae, it contains polyphenols and their enzymes, the polyphenolases. In addition, a negative result was obtained for the syringin test, but a red color was present. Correspondingly, it gave a magenta spot in the Ehrlich test, a positive HCl/Methanol reaction, and a positive leucoanthocyanin result.

This genus Akania contains no raphides. In addition, cluster crystals are present, and to a lesser extent, solitary crystals.

A doubtfully positive reaction was obtained in the HCN test. In addition, naphthoquinones or related compounds are absent. Fluorescence was observed.

Tannins were present in the leaves tested.

Group C phenolic constituents were absent from the leaves; only p-coumaric acid of group B was recorded. The results for group A were doubtful.

h. Malpighiaceae. As mentioned in the Review of Literature, many systematists have raised this family to the ordinal level. Hutchinson (1959) is among them.

He raised this family to the ordinal level and placed it along with many geranialean families in the Lignosae. Thus if it is allied with these families, it should contain many chemical characteristics in common with Geraniaceae.

The absence of polyphenolases was noted in the species investigated. All results were negative in the syringin test, but in contrast to Geraniaceae, the majority of the species contained some red color in the cross-sections. These results correlated with those obtained in the leucoanthocyanin, Ehrlich, and HCl/Methanol tests.

Raphides were absent from the species investigated. Solitary and cluster crystals, in addition to styloids, have been reported in this family by Metcalfe and Chalk (1950).

The majority of the plants contained no cyanogenetic substances. Heteropterys was the only positive genus.

Naphthoquinones were absent. As before, fluorescence was observed under the ultraviolet light. Many of the few plants tested were tanniniferous. While saponin test A was negative, part B was positive.

Group C compounds were absent, as well as many of group B. The results obtained for group A were mixed.

i. Trigoniaceae. No information was obtained for this family.

j. Vochysiaceae. Very little information was available on the chemistry of this family. While no raphides have been reported, solitary and cluster crystals have been recorded.

The members of this family are all aluminum accumulators.

The major fatty acids of the seeds are myristic and lauric acids. Many genera of Simarubaceae also contain these two acids.

k. Tremandraceae. The three genera of this family are native to Australia. The relationship of this family to others remains unestablished. Thus it has



been placed near many different families.

Polyphenolases were not recorded in Tetratheca. In addition, negative results were obtained for the syringin test. However, the two genera tested differed in the color reaction. There was no evidence of red in Platytheca; the reverse was true for Tetratheca. These results corresponded with those obtained for the related tests.

No raphides were seen in the cross-sections, although there have been reports on the occurrence of solitary and cluster crystals in this family. Cyanogenetic glycosides were present in several species, but did not predominate. Naphthoquinones were absent as in the other families thus far discussed. Fluorescence was observed.

Negative results were obtained for group C and most of group A phenolic compounds. However, most of group B compounds were present in the leaves.

1. Polygalaceae. This has been called a very natural family. However, many genera, for example, Krameria, Xanthophyllum, Moutabea, and Diclidanthera have been doubtfully placed in this family. Unfortunately, material of these genera were unavailable for investigation.

Polyphenolases were not recorded for most of the genera. However, Mundtia gave a "II-III" reaction. In addition, while one species was doubtfully positive, the other species were negative for the syringin test. There was no evidence of red in the cross-sections. Mundtia differed again, as there was a pink color in parts of the treated section. These results correlated with those of the related tests.

Polygala was negative in the Juglone test, and some fluorescence was observed. Mundtia again was the sole exception, for a positive Juglone test was obtained. The presence of any fluorescence was uncertain.

Polygala was non-tanniniferous, while Mundtia was doubtfully positive.

Amarasingham (1964) investigated two species of *Xanthophyllum* and found one of them to contain saponin. Saponins have been recorded in some species of Polygala. Part B was negative.

No raphides have been recorded, but calcium oxalate crystals are present. Mixed results were obtained for group B, while the majority of species contained group A substances.

Polygala contains D-polygalitol. This aliphatic polyol has only been recorded in one other family, Aceraceae. It has been isolated from Acer ginnala. (Plouvier, 1963)

Mundtia spinosa, on the basis of the chemical evidence, seems to be out of place in this family, but our material may possibly have been wrongly labelled Mundtia.

## 2. The relationship of the Rutales to the "Geraniales" s.s.

Rutaceae, on the basis of chemical evidence, is a very unnatural family. For example, many of the group C phenolic constituents were present in the various subfamilies. This is substantiated by the alkaloidal, quinone, and other chemical distributions. Thus further investigation must be done on this family. At present, no conclusion can be reached as to its affinity with the "Geraniales" s.s.

Simarubaceae is also a rather unnatural family. This is confirmed by the rather unusual fatty acid distribution in the seeds. Thus it is separated from the "Geraniales" s.s. Picrodendraceae in many ways is like the Geraniaceae. But the results from the HCl/Methanol and related tests are pieces of evidence against placing this family near Geraniaceae. More information should be obtained on the fatty acid composition of this family.

Meliaceae does not seem to be closely allied to Geraniaceae. The phenolic acid distribution is similar to those of Rutaceae and Simarubaceae. However,

more evidence is necessary.

Akaniaceae and Malpighiaceae, based largely on the HCl/Methanol correlation, do not seem to belong with Geraniaceae. In addition, Akania contains polyphenolases, enzymes lacking in most members of the "Geraniales" s.s.

Polygalaceae seems very much like Geraniaceae; however it differs in that it contains aliphatic polyols. Upon further investigation, more differences will undoubtedly turn up.

Tremandraceae is a family of doubtful position, and on the basis of chemical evidence, it remains as such. Few group A compounds were found in this family; group B compounds predominated.

Cneoraceae gave results very much like those of Geraniaceae. So, on the basis of chemical constituents, it should be placed in the "Geraniales" s.s. Burseraceae is also very much like Geraniaceae. However, very few specimens were examined. Thus further investigation is necessary before anything conclusive is said.

No information was available on Trigoniaceae and Vochysiaceae which would throw light on their systematic position.

Thus it seems that the order Rutales is heterogenous and should be more intensely investigated. Furthermore this order should not be reunited with the "Geraniales" s.s.

#### C. Analysis of other families included in the "Geraniales"

1. Dichapetalaceae. This is a family of doubtful position. Because of the limited material available, all of the tests could not be done. The syringin was negative, and there was no evidence of red in the cross-sections. The HCl/Methanol test was likewise negative, while the leucoanthocyanin test was positive. Thus a non-correlation.

This family is separated from the Geraniaceae in that it contains toxic

fatty acids which contain fluorine in their structure. Dear and Pattison (1963) found  $\omega$ -fluoroooleic acid to be identical with the toxic principle in Dichapetalum taxicarium. Also isolated was  $\omega$ -fluoro-elaidic acid.

On the basis of this, Dichapetalaceae is not allied with Geraniaceae.

2. Callitrichaceae. Engler and Diels (1936) placed this family in the "Geraniales", but it was subsequently moved to the Tubiflorae (Engler and Melchior, 1964). Unfortunately no material of this aquatic plant was available for investigation by me.

This was the only family in which the members gave a positive Ehrlich reaction (Gibbs, unpub'd), thus indicating the presence of aucubin or aucubin-like substances. This is in contrast to Geraniaceae, and in fact, all the families surveyed. Thus there is strong evidence against placing this family near Geraniaceae.

Gibbs (1962) did chemical work on the Tubiflorae and did find many members to be aucubin-containing, including those of the Verbenaceae, near which Callitrichaceae is placed in the 12th "Syllabus".

Since blackening occurred in the leucoanthocyanin test, it is postulated that the aucubin substances interfered with the reactions. Gibbs remarked that doubtful cases (in the Tubiflorae) "are mostly those in which darkening of the mixture occurs -- due to aucubin and aucubin-like substances -- or in which anthocyanin is already present in the untreated leaf."

Thus this family would be out of place in the "Geraniales" s.s.

3. Balsaminaceae. This family has been included in the Geraniales by many authors, among them, Hutchinson (1959). This family differed from Geraniaceae in that a red color was observed in the sections treated in the syringin test. The corresponding tests also correlated. In addition, numerous raphides were seen in the cross-sections.

In addition, parinaric acid was the major acid in the seeds of several

Impatiens. Furthermore, phenolic constituents of group C were found in many species. These included scopoletin and p-hydroxybenzoic acid.

These differences are evidence against placing this family near Geraniaceae.

4. Buxaceae. Members of the two subfamilies differed in their reactions to the various tests. Simmondsia gave a positive Leucoanthocyanin reaction, but a negative HCl/Methanol result. Thus perhaps anthocyanins were present in the leaves. The members of the other subfamily gave all negative results.

Group C compounds were generally absent, but Simmondsia contained vanillic and syringic acids. Gallic acid of group B was present, while group A compounds were present in all species.

Buxaceae contains many alkaloids, among them bebuxine, buxpiine, buxtauine, buxomegine, buxalphine, buxdeltine, buxetine, and cyclobuxine (Tomko et al, 1964). Kupchan et al (1964) reported the presence of a steroidal alkaloid in this family: buxetine-G. Other alkaloids include bebeerine and D-isochondrodendrine. Thus alkaloid-wise, this family is separated from Geraniaceae.

Simmondsia californica (Link.) Schneider seems out of place in this family. The seeds have a high fatty acid content. An unusual liquid wax is present. Daugherty, Sineath, and Wastler (1958) investigated this plant of the California area, and found it was first mentioned by the Mexican historian Francisco J. Clavijero, who found that Indians used its fruits as food, and the oil as a medicine and hair-restorer. The liquid wax is a light yellow unsaturated liquid of unusual stability and is found in a relatively pure state. The chief fatty acid is eicosenoic.

## CONCLUSIONS

From the analysis of the data, the "Geraniales" s.s. forms a natural group if Euphorbiaceae, obviously of polyphyletic origin, is excluded. Furthermore, the data supports the separation of Averrhoa and Biophytum from Oxalidaceae, and Peganum from Zygophyllaceae.

There is no evidence for Hutchinson's (1959) splitting of the "Geraniales" s.s. into two major orders, the Geraniales and Malpighiales, and the placing of these orders in two evolutionary lines, namely, the Lignosae and the Herbaceae.

It is also concluded that the Rurales forms a heterogenous order. Therefore, more intense investigation should be done on Rutaceae, Simarubaceae, Meliaceae, and the other families. However, of all the families, Cneoraceae and Burseraceae seemed most to approach the Geraniaceae. There is no evidence to support the reunification of this order with the "Geraniales" s.s.

Dichapetalaceae, Callitrichaceae, Balsaminaceae, and Buxaceae, should not be placed in the Geraniales, but the latter two should be assigned to allied orders.

## SUMMARY

There is no systematic classification which is supported by all taxonomists. Therefore, there is disagreement as to which system is the correct one and therefore should be adopted. Most of these systems are based on morphological and anatomical evidence. Today, cytological, embryological, and genetical evidence is being increasingly used.

Since morphology and anatomy are expressions of the biochemistry of the plant, another method of investigation is to study the biochemistry of plants. If it can be assumed that related plants have similar products, ergo similar biosynthetic pathways, then plants can be grouped accordingly in an evolutionary scheme.

Thus, through work in the laboratory, a survey of literature, and information cards of Dr. R. D. Gibbs, data was collected and analyzed, and tentative conclusions reached on the classification of the "Geraniales".

These tentative conclusions include: 1) the "Geraniales" s.s. form a natural group if Euphorbiaceae, a family of polyphyletic origin, is excluded; 2) there is no evidence for Hutchinson's split of the "Geraniales" s.s.; 3) the Rutales form a heterogeneous group and more work should be done on it; 4) there is no evidence for the reunification of the "Geraniales" s.s. and the Rutales; 5) Dichapetalaceae, Callitrichaceae, Balsaminaceae, and Buxaceae, should not be placed near Geraniaceae.



## APPENDIX I

A list of plants (by genera) tested by the author, Gibbs and others

Key: NR = no red  
R = red  
Pu = purpleM = magenta  
tr = trace  
plt = plantsd = seeds  
oth. or ot = other parts

	Hot water & cig. tests				Syringin		Raphide	Ehrlich		HCN		HCl/Meth.		L.A.		Juglone		Fluores.		Tannin		Saponin			
																						A+		B+	
	I	II	III	IV	or	+	-	+	-	+	-	+	-	+	-	+	-	+	-	+	-	A+	-	B+	-
<b>GERANIACEAE</b>																									
I. Geranieae (5/646)																									
Geranium (Tourn.) L.			1?	5	2?		1NR	-1		3						5		1	1		4?		1		1
Erodium L'Hért.				2			2NR	-2		1						3		1	1		2?				
Pelargonium L'Hért.				1	7		4NR	-6		4	1?	7	3	2	1	6		7	7		4				
							2R				3M		1?								4?				
II. Biebersteinieae (1/5)																									
III. Wendtieae (3/10)																									
IV. Vivianieae (1/30)																									
V. Dirachmeae (1/1)																									
<b>OXALIDACEAE (8/950)</b>																									
Oxalis L.				2	12		3NR	-2		4		11	1?	1?	5	6		11	10	1	2	13	3		4
							3?NR	-3?		1M					2?						3tr, 3tr?		1?		
Biophytum DC.			1				1R	-1		1M		1	1		1		1	1			1				
Averrhoa L.			1	1?			2R	-2		2M		2	2		1		2	2			1		1	1	
															1?						1tr				
TROPAEOLACEAE (2/80)																									
Tropaeolum L.				2			1NR	-1		2		1				2		1	1		1	1			
											3(sd)														
<b>ZYGOPHYLLACEAE*</b>																									
I. Peganoideae (1/6)																									
Peganum L.				1?			1NR	+1		1		1	1			1		11			1				
II. Chitoniodeae (3/4)								-1																	
III. Tetradiclidoideae (1/1)																									
IV. Augeoideae (1/1)																									
V. Zygophylloideae (11/194)																									
Zygophyllum L.				3			2NR	-2		3		4	2		2		2	2			1	1	1		1
				III-IV-1				-1?																	
Guaiacum Plum. ex L.				1			1NR	-1		1	ltr.		1		1		1	11			1				
Tribulus Tourn. ex L.				1				-2?			1		1								1				
											1(oth.)										1				
Kallstroemia Scop.				1?			1NR	-1					1												
VI. Nitrarioideae (1/4) Nitrarias L.								-1																	
<b>LINACEAE</b>																									
I. Hugonieae (14/278)																									
Reinwardtia Dum.	1	1				1R?	1NR	-2		1		2			2					1?		1			1
								-1?																	
Linum Tourn. ex L. I-II-1							1NR	-1				2	1		6		1	11			3		2		2
II. Ctenolophoideae (1/4)																									
III. Ixonanthoideae (2/23)																									
Ixonanthes Jack.												2		2			2	2							
IV. Humirioideae (3/30)																									
<b>ERYTHROXYLACEAE (4/200)</b>																									
Erythroxylum P.Br.							1R	-2		1M	1	3	1		1	1?		2	1	11	ltr	1			
							1NR																		

\*the seventh subfamily of Zygophyllaceae is Balanitoideae (1/2)



	Hot water & cig. tests				Syringin	Raphides		Ehrlich		HCN		HCl/Meth.		I.A.		Juglone		fluores.		Tannin		Saponin			
	I	II	III	IV		or	+	-	+	-	+	-	+	-	+	-	+	-	+	-	A+	-	B+	-	
LIMNANTHACEAE (2/8)																									
Floerkia Willd.																									
Limnanthes R. Br.																									
EUPHORBIACEAE																									
A. Platylobeae																									
I. Phyllanthaceae (65/1731)																									
Antidesma Burm. ex.L.																									
Breynia Forst.																									
Putranjiva Wall.																									
Securinega Comm. ex Juss.																									
Phyllanthus L.																									
Andrachne L.																									
Drypetes Vahl.																									
Hemicyclia Wight et Arm.																									
Bischofia Blume																									
Bridelia Willd.																									
II. Crotonoideae (209/4331)																									
Croton L.																									
Aleurites Forst.																									
Hevea Aubl.																									
Claoxylon A. Juss.																									
Melanolepis Reichb. f. & Zoll.																									
Mallotus Lour.																									
Alchornea Sw.																									
Mercurialis (Tourn.) L.																									
Cleidion Blume																									
Macaranga Thou.																									
Acalypha L.																									
Tragia Plum. ex L.																									
Homonoia Lour.																									
Ricinus (Tourn.) L.																									
Dalechampia Plum. ex L.																									
Baloghia Endl.																									
Codiaeum Rumph. ex A. Juss.																									
Jatropha L.																									
Manihot Tourn. ex Adans.																									







	Hot water & cig. tests				Syringin	Raphides	Ehrlich	HHCN		HCl/Meth.		L.A.		Juglone		fluores.		Tannin		Saponin			
	I	II	III	IV	or	+	-	+	-	+	-	+	-	+	-	+	-	+	-	A+	-	B+	-
RUTACEAE																							
I. Xanthoxyleae (86/1169)																							
Xanthoxylum L.	2					2NR	-2							5	1	1		3	3		1		
Geijera Schott.	1					1NR	-1		1M	2		1?				1							
Evodia Forst.	1			1		2NR	-2		1	2		1		1		1		2	2		1		
Orixa Thunb.						1NR	-1			1				1		1		1	1		1		
Choisya H.B. et K.				1												1							
Ruta (Tourn.) L.				1		1NR	-1		1	1				1		1		1	1		2		
Cneoridium Hook. f.																1				1?			
Dictamnus L.				1		1NR	-1		1	1				1		1		1	1		1		
		III-IV-1																					
Boronia Sm.				1	2?	5NR	-6		5M	3	3			2	2	4		4	4		3		
				1?		1R			1		3(ot)			1?	1?					1tr			
																				1(inflorescence)			
Acrademia Kipp.				1		1?R	-1		1M	1				1									
Zieria Sm.				1		1NR	-1		1M	3	1?(ot)			1?	1								
		III-IV-1?								3(ot)													
Eriostemon Sm.	1					2NR	-2		1M	1,2(ot)				2	1			1	1		1		
Croton Smith				1		1NR	-1		1M	2(ot)				1									
Phebalium Vent.				1?					1M	1(ot)				1				1(leaves)	1				
Correa Andr.			1?			1?NR	-1		1M	2				1				1	1				
Nematolepis Turcz.			1			1NR	-1		1M	1				1	1			1	1		1		
Chorilaena Endl.				1		1NR	-1		1M	1				1	1			1	1		1?		
Diplolaena R. Br.			1?			1NR	-1		1	1				1		1		1	1			1	
Calodendron Thunb.				1		1R	-1		1	1				1		1		1	1			1	
Barosma Willd.				1		1NR	-1		1	1				1	1,1?			1			1		
Coleonema Bartl. et Wendl.				1		2NR	-2		2M	2				2		2		2	2		2		
Pilocarpus Vahl.				1		1NR	-1		1	1				1		1		1	1			1	
Galipea Aubl.							+1																
Raputia Aubl.							+1																
Erythrochiton Nees et Mart.			III?-IV-1			1			1M	1				1	1			1	1		1?		
II. Dictyolomatoideae (1/2)																							
III. Flindersioideae (2/20)																							
Flindersia R. Br.				1		1R	-1		1M	2				1	1			1	1		1		1
IV. Spathelioideae (1/10)																							
V. Toddalioideae (15/280)																							
Phellodendron Rupr.	3		II-III-1			1?NR	2NR	-4	2	1				2	1?	1		3	3		3		
						1R								2?									
Ptelea L.				1		1NR	-1							1		1		1	1		1, 1tr		
Casimiroa La Llave			II-III-1			1NR	-1		1	1				1		1		1	1		1?		
Toddalia Juss.						1NR	-1			1				1									
Acronychia Forst.			II-III-1			1NR				2				2									
Halfordia F. Muell.				1		1M			1M	1(ot)				1									
Skimmia Thunb.				1	1?	1NR	-1		1	1				2		2		1	1		1tr	2	
Amyris P. Br.														1									
Teclea Delile				1		1R	-1		1M	1				1	1?			1	1		1tr		
VI. Aurantioideae (28/193)																							
Glycosmis Correa				1		1NR	-1		1	1				1?		1		1	1		1		
Murraya Koen. ex L.			I-II-1	1		1R	-2		2	2				2		2		2	2		1		
						1NR														1?			
Triphasia Lour.				1		1NR	-1		1	1tr						1		1	1		1		



	Hot water & cig. tests				Syringin	Raphides	Ehrlich		HCN		HCl/Meth.		L.A.		Juglone		fluores.		Tannin		Saponin			
	I	II	III	IV			+	-	+	-	+	-	+	-	+	-	+	-	+	-	A+	-	B+	-
<u>Atalantia</u> Correa				1		1NR		-1		1		1		1		1		1		1				
<u>Poncirus</u> Rafin.						1NR		-1				1				1		1						
<u>Eremocitrus</u> Sw.										1														
<u>Citrus</u> L.				5		3NR		-5		1	4tr	1	1?	1	3	3	2		1	2				
											1(ot)													
											2tr(ot)													
											1tr?													
											1(ot)													
<u>Limonia</u> L.																								1?
<u>Aegle</u> Correa										1tr?		1												
<u>Feronia</u> Correa		II-III-1				1NR		-1		1M	1	1	1		1	1		1		1				
CNEORACEAE (2/3)																								
<u>Cneorum</u> L.			IV-III-1			1NR		-1		1	1	1		1		1				1				
SIMARUBACEAE																								
I. Surianoideae (4/6)																								
<u>Suriana</u> Plum. ex L.		1				1R		-1		1M	1	1				1		1						
II. Simaruboideae (22/109)																								
<u>Hannoa</u> Planch.						1NR		-1					1		1		1		1		1			
<u>Picrasma</u> Blume															1									
<u>Picraena</u> Lindl.				1		1NR		-1		1			1				1		1					
<u>Ailanthus</u> Desf.				1?		1NR		-1		1	1	1	1	1?			1		1					
III. Kirkioideae (1/4)																								
<u>Kirkia</u> Oliv.			1			1NR		-1			1			1		1		1		1?				
IV. Irvingioideae (3/13)																								
<u>Irvingia</u> Hook. f.											1(sd)													
V. Picramnioideae (1/40)																								
VI. Alvaradoideae (1/5)																								
PICRODENDRACEAE (1/3)																								
<u>Picrodendron</u> Planch.				1		1R		-1		1M	1	1				1		1						
BURSERACEAE (20/600)																								
<u>Bursera</u> Jacq.			1?										1	1	1		1		1					
<u>Commiphora</u> Jacq.		II-III-1				1R		-2			1	1			1		2		2		1?			
						1NR																		
<u>Canarium</u> (Rumph.) L.											1													
											1(ot)													
<u>Pachylobus</u> G. Don.						1NR		-1					1				1		1					
MELIACEAE																								
I. Cedreloideae (4/118)																								
<u>Cedrela</u> P. Br.													1			1								
<u>Toona</u> M. Roem.				1		1R		-1		1M	1	1		1		1		1		1tr				
<u>Ptaeroxylon</u> Eckl. et Zeyh.						1NR		-1		1	1	1		1		1		1		1		1	1(bark)	
II. Swietenioideae (8/51)																								
<u>Khaya</u> A. Juss.																					1			
<u>Swietenia</u> Jacq.			1			1R		-1		1M	1	1		1		1		1		2		1	1	







	Hot water & cig. tests					Syringin		Raphides	Ehrlich		HCN	HCl/Meth.		L.A.		Juglone		fluores.		Tannin		Saponin				
	I	II	III	IV	or	+	-		+	-	+	-	+	-	+	-	+	-	+	-	+	-	A+	-	B+	-
DICHAPETALACEAE																										
<u>Dichapetalum</u> Thou.		II-III-1						1NR	-1			1(ot)1	1	1		1		1		1						
CALLITRICHACEAE																										
<u>Callitriche</u> L.									1 1(plt)		1 1(plt)			2? (darkening)												
BALSAMINACEAE																										
<u>Hydrocera</u> Blume								+																		
<u>Impatiens</u> Riv. ex L.	1? III-IV-3			2			2R 1R?	+8		2M	ltr 3 ltr?		1?	5		4		3 1?		2 1		1		1?		
BUXACEAE																										
I. Buxae (3/25)																										
<u>Sarcococca</u> Lindl.				1			4NR	-4		3	4 1(sd)	4	1 2 1?		3		2 1		2tr							
<u>Pachysandra</u> Michx.				1			3NR	-3		3	2	2	3		3		1		1 1							
<u>Buxus</u> L.				1			1? 2NR	-3		3	4	2	5		1		1		3		1					
II. Stylocereae (2/4)																										
III. Simmondsieae (1/1)																										
<u>Simmondsia</u> Nutt.		III-IV-1						1NR	-1		1M	1	1	1		1		1		1?						



## APPENDIX II

Results of Miscellaneous Tests  
(done by author)

	H.W.				Cig.				Syr.	Rap.	Ehr.	HCN	HCl/M	L.A.	Jug.	fl.	T.T.	Sap.	A	B
	I	II	III	IV	or	I	II	III	or											
GERANIACEAE																				
I. Geranieae																				
<u>Geranium bicknelli</u>				+						-NR	-	-Y	-	-				++?		
<u>G. columbinum</u>				+								-Y	-	-						
<u>G. eriostem</u>														-				++?		
<u>G. molle</u>														-				++?		
<u>G. psilostemon</u>				+	?									-		-	+	++?	-	-
<u>G. sanguineum</u>				+	?									-Y	-	-				
<u>Erodium chamaedryoides roseum</u>																				
<u>    florenspluis (?)</u>										-PK	-						+			
<u>E. manescavii</u>					?III-IV					-NR	-	-Y	-	-				++?		
<u>E. pelargoniflorum</u>														-				++?		
<u>E. sp.</u>														-						
<u>Pelargonium burtoniae</u>				+				+		-NR	-	-BR	-	-?	-	-	+	+++?		
<u>P. graveolens</u>				+				+		-R	-	-M	-	+4	+++	-	+	+		
<u>P. hortorum</u>				+										-	-	-	+	+		
<u>P. inquinans</u>				+				+		-NR	-C	-YB	-	-	-	-	+	+		
<u>P. peltatum</u>				+				+		-NR	-C	-YB?	-	+4	-	-	+	+		
<u>P. peltatum</u> var. "Chester Frank"										-R	-C	-M?	-	+3	-	-	+	+++?		
<u>P. salmoneum</u>				+				+		-NR	-	-YB	-	-	-	-	+	++?		
II. Biebersteinieae																				
III. Wendtieae																				
IV. Vivianieae																				
V. Dirachmeae																				
OXALIDACEAE																				
<u>Oxalis bowiei</u>				+										-B	-	-	-	+	tr?	
<u>O. cernua</u>				+																
<u>O. chrysantha</u>				+										-YB	-	-	-			
<u>O. deppei</u>				+													+			
<u>O. dillenii</u>				+						-NR?	-C			-?		-	+			
<u>O. eriolepis</u>																		++		
<u>O. lasiandra</u>				+										-M	-	+	-	+	+	-
<u>O. lobata</u>				+						-NR	-?	-RPK	-		+			tr?	-	-
<u>O. ortgiesii</u>				+				+		-NR	-C					-	+	tr		
<u>O. peduncularis</u>				+				+		-NR	-C					-				
<u>O. piottae</u>				+				+		-NR	-	-B	-					tr?	-	-
<u>O. sp.</u>				+																
<u>Biophytum sensitivum</u>				+						-R	-	-M	-	+4	+	-	+	+++		
<u>Averrhoa bilimbi</u>				+	?					-R	-	-M	-	+1-2	+	-	+	tr		
<u>A. carambola</u>				+					III?-IV	-R	-	-M	-		+			++	-?	+
TROPAEOLACEAE																				
<u>Tropaeolum majus (?)</u>				+										-YB	-					
<u>T. sp.</u>				+						-NR	-C	-Y	-		-	-	+	+		
ZYGOPHYLLACEAE																				
I. Peganoideae																				
<u>Peganum harmala</u>				+	?					-NR	-	-O	-	-	-	-	+	-		
II. Chitonioideae																				
III. Tetradiclidioideae																				
IV. Augeoideae																				
V. Zygophylloideae																				
<u>Zygophyllum fabago</u>					III-IV			+		-NR	-	-PK	-	-	-	-	+	-	-	-
<u>Z. margsana</u>										-NR	-	-Y	-	-	-	-	+	+		
<u>Guaiacum officinale</u>				+									tr				+	-		
LINACEAE																				
I. Hugonieae																				
<u>Reinwardtia sp.</u>					I-II			+		-NR	-	-YB	-		-					
<u>Linum austriacum</u>														+++	-					
<u>L. grandiflorum</u> v. <u>rubrum</u>														++++	-	-	+			
<u>L. usitatissimum</u>					I-II															
II. Ctenolophonoideae																				
III. Ixonanthoideae																				
<u>Ixonanthes icosandra (?)</u>														-	-		+			
<u>I. reticulata (?)</u>														-	-		+			
IV. Humirioideae																				
ERYTHROXYLIACEAE																				
<u>Erythroxylum coca</u> var. <u>novo-granatense</u> ?										-NR	-				-	-		tr		
<u>E. novo-granatense</u>				+						-R	-	-M	-	+3-4?	-	-	+			



## APPENDIX II(cont'd)

	H.W.					Cig.					Syr.		Rap.	Ehr.	HCN	HCl/M	L.A.	Jug.	fl.	T.T.	Sap.	A	B
	I	II	III	IV	or	I	II	III	IV	or													
<b>LIMNANTHACEAE</b>																							
<u>Floerkia prosperpinacoides</u>											-NR	-		-Y	-								
<u>Limnanthes douglasii</u>				+							-R	-			tr		++						
<b>EUPHORBIACEAE</b>																							
<b>A. Platylobeae</b>																							
<u>Antidesma bunius</u>		II-III												-M			+	-	+		tr		
<u>Putranjiva roxburghii</u>		I-II												-YB			-	=	+		++?		
<u>Securinega suffructicosa</u>											-R	-				+4			+				
<u>Phyllanthus epiphyllanthus</u>				+										-M	-		-				+++		
<u>Andrachne colchiana</u>		III?-IV									-NR	-		-M		-?	+	-	+		+		
<u>Drypetes Gerrardii</u>		I-II									-NR	-		-M	-	-	++?	-	+		++		
<b>II. Crotonoideae</b>																							
<u>Aleurites moluccana</u>		IV-III				IV-III					-NR	-		-M	-	-	+	-	+		+++		
<u>Croton appendiculatus</u>		III-IV												-B	-	-?	-	-	+		-	-	
<u>Alchornea ilicifolia</u>				+							-R	-		-M	-		+	-	+		++		
<u>Cleidion Javanicus</u>			+								+?NR	-		-B	-	-	-	-	-		++		
<u>Macaranga grandifolia</u>											-R	-		-M	-	+3	+	-	+		-?		
<u>Acalypha godseffiana</u>				+							-NR	-C?		-B	-	-	-	-	-		-		
<u>A. hamiltoniana</u>		IV-III									+?NR	-		-	-	-	-	-	-		+		
<u>A. hispida</u>				+							+?NR	-C		-B	-	-	-	-	-?		+++		
<u>A. wilkensiana</u>																					+		
<u>A. wilkensiana v. musaica?</u>				+													-	-	-		+		
<u>Tragia involucratus</u>				+							-NR	-			-	-	-	-	+		++		
<u>Ricinus communis</u>				+					+		-NR	-C		-YB	-	-	-	-			+		
<u>Dalechampia roezliana</u>		III-IV							+		-NR	-		-YB	-	-	-	-			++		
<u>Jatropha curcas</u>				+							-NR	-		-M	-		+				+++		
<u>J. multifida</u>				+							-NR	-		-B	++++	-	-	-	+		-		
<u>Codiaeum sp.</u>											-NR	-		-B		-?	++?						
<u>C. sp.</u>			++?						+		-NR	-		-MB	-	-	+	-	+		tr?		
<u>Manihot palmata</u>				+							-NR	-		-BY	+++	-	-	-	+		tr		
<u>Baliospermum axillare</u>			+														++?	-	+		+		
<u>Omphalea trichotoma</u>		III-IV				III-IV					-R?	-C		-B	-	+3	++?	-	+		+		
<u>Homalanthus polypulneus</u>		III-IV						+			-NR	-		-Y	-	-	-	-	+		++?		
<u>Gymnanthes lucida</u>			+			III-IV								-M	tr		+	-	+		+		
<u>Hura crepitans</u>											-R?	-		-B	-	-	-	-	+		-?		
<u>Euphorbia balsamifera</u>				++?										-BY	-	-	-	-	+		++		
<u>E. characias</u>			+											-MP	-		+	-	+		-?	-	
<u>E. melliformis</u>																	-				+		
<u>E. monteiri</u>			+								-NR	-		-B	-	-	-	-			tr		
<u>Synandrium grantii</u>				+				+		+	-NR	-		-Y	-	-	-	-	+		tr?		
<u>Pedilanthus tithimalioides</u>			+						+		-NR	-		-B	-	-	-	-	++		+		
<u>Euphorbia pulcherrima</u>																							
<b>B. Stenolobeae</b>																							
<b>I. Porantheroideae</b>																							
<b>II. Ricinocarpoideae</b>																							
<u>Beyeria leschenaultii</u>			+								-NR	-		-M	-		+				+		

## DAPHNIPHYLLACEAE

## Key to symbols:

NR = no red

R = red

PK = pink

C = crystals

BR or B = brown

Y = yellow

M = magenta

YB = yellow-brown

RP = red-pink

O = orange

MP = magenta-pink

tr = trace

MA = maroon



APPENDIX II(cont'd)

	H.W.					Gig.					Syr.	Rap.	Ehr.	HCN	HCl/M	L.A.	Jug.	fl.	T.T.	Sap.	A	B
	I	II	III	IV	or	I	II	III	IV	or												
RUTACEAE																						
I. Xanthoxyleae																						
<u>Xanthoxylum simulans</u>											-NR	-			-					+		
<u>Evodia Danielli</u>											-NR	-			-					+		
<u>E. Henryi</u>	+										-NR	-	-Y	-	-					+		
<u>Orixa japonica</u>											-NR	-			-					+		
<u>Ruta graveolens</u>				+											-					++	tr	
<u>Cneoridium dumosum</u>																	+					
<u>Dictamnus albus</u> var. <u>turkestanicus</u>					III?-IV						-NR	-	-OB	-	-					++	+	
<u>Boronia denticulata</u>					+						-NR	-	-M	+	-?	+				++	+	
<u>B. lanagmusa?</u>											-NR	-	-M	-	-?	+				+	+	
<u>B. purdiana</u>											-R	-	-M	-	+3	+				++	tr	
<u>B. viminea</u>					+						-NR	-	-M	+	+3	+				++	tr	
<u>Eriostemon spicatum</u>											-NR	-	-M	-	-	+				+++	+++	
<u>Nematolepis pheballoides</u>			+								-NR	-	-M	-	-	+				++	++	
<u>Chorilaena quercitotia</u>				+							-NR	-	-M	-	-	+				+	+	
<u>Diplolaena angustifolia</u>			+								-NR	-	-M	-	-	+				+	+	
<u>Calodendrum capense</u>				+							-NR	-	-G	-	-	-				+++	-	
<u>Barosma scoparia</u>				+							-R	-	-Y	-	-?	-				+	-	
<u>Coleonema album</u>											-NR	-	-B	-	-	+	?				++	
<u>C. pulchrum</u>											-NR	-	-M	-	-	+				+	+	
<u>Pilocarpus pennatifolius</u>					+						-NR	-	-M	-	-	++				+	++	
<u>Erythrochiton brasiliensis</u>					III?-IV						-NR	-	-Y	-	-	-				+	-	
													-M	-	-	+				+	++?	
II. Dictyolomatoideae																						
III. Flindersioideae																						
<u>Flindersia australis</u>											-R	-	-M	-	+2-3	++				+		+
IV. Spathelioideae																						
<u>Ptelea trifoliata</u>											-NR	-C			-					+++		
V. Toddalieae																						
<u>Phellodendron amurense</u>	+										-R	-			-?					+		
<u>P. japonicum</u>											-NR	-			-					+		
<u>P. lavalleyi</u>					II-III						-NR	-	-Y	-	-	-				++	++	
<u>Casimiroa edulis</u>					II-III					II-III	-NR	-	-Y	-	-	-				+++	+	
<u>Skimmia foremanii</u>									+		-NR	-	-Y	-	-	-				+++	-	
<u>Teclea simplicifolia</u>					+						-R	-	-M	-	+4	+	?			+	tr	
VI. Aurantioideae																						
<u>Glycosmis pentaphylla</u>				+							-NR	-	-B	-	-?	-				+	-	
<u>Murraya exotica</u>				+					+		-R?	-	-Y	-	-	-				-	-	
<u>M. koenigii</u>					I-II								-YB	-	-	-				+	++	
<u>Triphasia trifolia</u>					IV-III						-NR	-	-B	-	-	-				+	-	
<u>Atalantia ceylanica</u>										+	-NR	-	-YB	-	-	-				-	-	
<u>Poncirus trifoliata</u>											-NR	-			-	-				+	-	
<u>Citrus limetta</u>				+					+		-NR	-			tr	-	-			+	+	
<u>C. limonia</u>				+							-NR	-	-Y	-	-	-				+	-	
<u>C. sp.</u>															-	-						
<u>C. sp. "otaheite orange"</u>															-	-						
<u>Feronia limonia</u>					+						-NR	-	-M	-	-	++				+	+	
CNEORACEAE																						
<u>Cneorum tricoccon</u>					IV-III-?								-YB							+	-?	-
SIMARUBACEAE																						
I. Surianoideae																						
II. Simaruboideae																						
<u>Hannoa klaineana</u>											-PK	-			-	-				+		
<u>Ailanthus altissima</u>					II-III								-BY	-	-	-				+		
III. Kirkioideae																						
<u>Kirkia acuminata</u>					+						-NR	-			-	-				+	+++?	
IV. Irvingioideae																						
V. Picramnioideae																						
BURSERACEAE																						
<u>Commiphora merkeri</u>					II-III						-MA	-			-	-				+	+++?	
<u>C. trothai</u>											-R	-			+4	-				+		
<u>Dacryodes (Pachylobus) klaineana</u>											-NR	-			-	-				+		
AKANIAACEAE																						
<u>Akania lucens</u>													-M								+++	
PICRODENDRACEAE																						
MELIACEAE																						
I. Cedreloideae																						
<u>Toona ciliata</u>				+					+		-R	-	-M	-	+3	+	-			++	tr	
<u>Ptaeroxylon obliquum</u>	+										-NR	-	-B	-	-	-	-			+	-	



## APPENDIX II (cont'd)

	H.W.					Cig.					Syr. Rap. Ehr. HCN HCl/M L.A. Jug. fl. T.T. Sap.									
	I	II	III	IV	or	I	II	III	IV	or									A	B
II. Swietenioideae																				
<u>Swietenia mahogani</u>			+			II-III					-R	-	-M	-		+	-	++	++	
III. Melioideae																				
<u>Carapa guianensis</u>	+										-R	-	-M	-	+4	+	-	+	+++	
<u>C. procera</u>	+										-NR	-	-M	-	-	+	-	+	+++	
<u>Cipadessa cinerascens</u>				+													-	++	+	
<u>Melia azedarach</u>	III-IV					III-IV					-NR	-	-Y	tr?		-	-	+	+	
MALPIGHIACEAE																				
I. Pyramidoterae																				
<u>Tristellateia australasiae</u>				+	?				+	?	-NR	-		-	-		-	+	tr	
<u>Gaudichaudia cynanchoides</u>	I-II										-NR	-	-M			-		-		
<u>Acridocarpus sp.</u>				+							-R	-	-M	-	+4	+	-	+	+	+++
<u>Thryallis glauca</u>											-R	-	-M	-	+4	+	-	+	+++?	
II. Planitorae																				
<u>Malpighia coccigera</u>				+							-R	-	-M	-	+2-3	+	?	+	-	+
<u>M. cubensis</u>																		+++		
<u>M. glabra</u>	III-IV										-R	-	-M	-	-?	+	?	+	-	
TRIGONIACEAE																				
VOCHYSIACEAE																				
TREMADRACEAE																				
<u>Platytheca verticillata</u>											-NR	-	-Y	-	-	-	-	+	+	?
<u>Tetratheca thymifolia</u>											-NR	-		-	-	-	-		+++?	
POLYGALACEAE																				
I. Polygaleae																				
<u>Polygala myrtifolia</u>	IV-III					IV-III					-NR	-	-B	tr	-	-	-	-	-	
<u>P. dalmaisiana</u>																				
<u>P. virgata</u>																		+		
<u>Mundtia spinosa</u>	II-III												-BM			+	+	tr?	+++	
II. Xanthophylleae																				
III. Moutabeae																				
DICHAPETALACEAE																				
<u>Dichapetalum cymosum</u>	II-III										-NR	-		-	-	+	-	-	++	
CALLITRICHACEAE																				
BALSAMINACEAE																				
<u>Impatiens oliveri</u>	III-IV												-M			+			+	?
<u>I. sultanii</u>	III-IV												-BM	tr?		++			+	
BUXACEAE																				
I. Buxaeae																				
<u>Sarcococca confusa</u>				+							-NR	-	-PKB				-	++	tr?	
<u>S. ruscifolia</u>				+					+				-PK				-	++	tr	
<u>Pachysandra procumbens</u>				+					+		-NR	-	-B				-	++	-	
<u>Buxus microphylla</u>													-B						-	-
II. Simmondsieae																				
<u>Simmondsia chinensis</u>	III-IV					III-IV							-M			+++	-	+	+++?	



APPENDIX III

Results of Chromatography  
of Phenolic Constituents (done  
by author)

	Group A				Group B			Group C											
APPENDIX III Results of Chromatography of Phenolic Constituents (done by author)	gent.	fer.	caff.	ell.	sin.	p-c	gall.	umb.	van.	syr.	p-OH-B	rutin	aesc.	p-cat.	scop.	melil.	P/L	phlor.	h-gent.
GERANIACEAE																			
<u>Geranium psilostemon</u>	-	tr?	tr	tr	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>G. robertianum</u>	+	+	+	+	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>Erodium pelargoniflorum</u>	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Pelargonium burtoniae</u>	+	+	tr	-	tr	-	++	-	-	-	-	-	-	-	-	-	-	-	-
<u>P. graveolens</u>	+	+	tr?	++	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>P. inquinans</u>	+	+	+	+	-	tr	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>P. peltatum</u>	+	tr	+	tr	-	-	+	-	-	-	-	+	-	-	-	-	-	-	-
<u>P. salmoneum</u>	+	+	+	+	-	+	+	-	-	-	+	-	-	-	-	-	-	-	-
OXALIDACEAE																			
<u>Oxalis bowiei</u>	+	+	+	tr	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Oxalis eriolepis</u>	+	-	-	++	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>O. europaea</u>	tr	+	+	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>O. lasiandra</u>	+	-	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>O. ortgiesii</u>	-	+	tr	tr	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Averrhoa bilimbi</u>	-	-	-	+	-	+	tr	-	-	-	-	-	-	-	-	-	-	-	-
<u>Biophytum sensitivum</u>	-	-	tr	+	-	-	tr?	-	-	-	-	-	-	-	-	-	-	-	-
TROPAEOLACEAE																			
<u>Tropaeolum majus</u>	-	-	++	+	-	++	-	-	-	-	-	-	-	+	-	-	-	-	-
ZYGOPHYLLACEAE																			
<u>Zygophyllum fabago</u>	-	+	tr	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Guaiacum officinale</u>	+	+	tr	tr	-	tr	+	-	+++	tr	-	-	-	-	-	-	-	-	-
LINACEAE																			
<u>Linum extraaxillare</u>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>L. usitatissimum</u>	-	+	-	tr	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
ERYTHROXYLACEAE																			
<u>Erythroxylum novo-granatense</u>	-	+	+	+	tr	++	-	-	-	-	-	-	-	-	-	-	-	-	-
LIMNANTHACEAE																			
<u>Limnanthes douglasii</u>	-	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
EUPHORBIACEAE																			
<u>Andrachne colchiana</u>	-	+	++	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Drypetes Gerrardii</u>	+	+	+	+	-	-	-	-	+	tr	-	-	-	-	-	-	-	-	-
<u>Croton appendiculatus</u>	+	+	tr?	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Cleidion Javanicus</u>	-	++	++	+	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>Acalypha godseffiana</u>	-	+	+	tr?	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Tragia involucrata</u>	-	tr	+	tr+	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>Baliospermum axillare</u>	-	+	+	+	-	tr	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Omphalea trichotoma</u>	-	+	+	++	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Euphorbia melliformis</u>	-	+	++	+	-	-	++	-	-	-	-	+	-	-	-	-	-	-	-
<u>Pedilanthus tithimalioides</u>	tr	+	+	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
DAPHNIPHYLLACEAE																			
RUTACEAE																			
<u>Evodia Danielli</u>	-	+	++	+	+	+	tr	-	-	-	+	-	-	-	-	-	-	-	-
<u>Evodia Henryi</u>	+	+	++	+	-	+	-	-?	-	-	-	-	-	-	+	-	-	-	-
<u>Orixa japonica</u>	+	+	++	-	-	+	-	-	-	+	-	-	-	-	-	-	-	-	-
<u>Ruta graveolens</u>	-	+	-	tr	+	+	-	-	-	-	-	-	-	+	-	-	-	-	-
<u>Dictamnus albus</u>	tr	+	tr	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Boronia denticulata</u>	tr?	+	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>B.? lanagmusa</u>	+	+	+	+++	+	+	-	-	+	-	+	-	-	-	-	-	-	-	-
<u>B. purdiana</u>	+	+	+	+++	+	++	-	-	+	-	+	-	-	-	-	-	-	-	-
<u>B. viminea</u>	+	+	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Eriostemon spicatum</u>	+	+	-	++	+	tr	-	-	tr	tr	-	-	-	-	-	-	-	-	-
<u>Nematolepis phebaloides</u>	-	+++	tr?	+++	-	+++	-	-	tr	-	-	+	-	-	-	+	-	-	-
<u>Diplolaena angustifolia</u>	-	+	tr?	+	+	-	tr	-	+	-	-	-	-	-	-	tr	-	-	-
<u>Calodendrum capense</u>	-	+	+	-	-	++	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Barosma scoparia</u>	-	+	+	++	+	-	-	-	-	-	-	-	+	-	-	-	-	-	-
<u>Coleonema album</u>	+	tr	tr?	+	+	-	-	-	-	-	-	-	-	-	+++	-	-	-	-
<u>C. pulchrum</u>	+	+	+	+++	+	+	tr?	-	tr	-	-	-	-	-	+	+	-	-	-
<u>Diosma ericoides</u>	+	+	tr	+	+	tr	-	-	-	-	-	-	-	-	+	-	-	-	-
<u>Pilocarpus pennatifolius</u>	-	+	+	+	+	+	-	-	tr	+	-	-	-	-	+	-	-	-	-
<u>Erythrochiton brasiliensis</u>	+	+++	++	+	+++	-	-	+	++	++	-	+	-	-	-	-	-	-	-
<u>Phellodendron japonicum</u>	tr	-	+	+	-	+	-	+	-	-	-	-	-	-	+	-	-	-	-
<u>P. lavalleyi</u>	-	tr?	+	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Ptelea trifoliata</u>	tr?	++	++	+	+	+	+	-	-	++	-	-	-	-	-	-	+	-	-
<u>Casimiroa edulis</u>	tr?	++	+	+	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-
<u>Skimmia reevesiana</u>	tr	+	tr	-	+	+	-	+++	-	+++	-	-	-	-	+	-	-	-	-
<u>Teclea simplicifolia</u>	+	+	+	-	++	+	-	-	-	+	-	-	-	-	-	-	-	-	-
<u>Glycosmis pentaphylla</u>	+	tr	-	+	tr	+	-	-	+	-	-	-	-	-	-	-	-	-	-
<u>Murraya exotica</u>	+	+++	+	+	+	+	-	-	+	-	-	-	-	-	-	+	+	-	-
<u>Triphasia trifolia</u>	-	+	tr	+	+	tr	-	-	-	-	-	-	-	-	+	-	-	-	-
<u>Atalantia ceylanica</u>	+	+	tr	+	-	+	-	+	+	-	-	-	-	-	-	-	-	-	-
<u>Citrus limetta</u>	+	+++	+	+	+	+++	-	-	+	-	-	-	-	-	+	-	-	-	-
<u>C. limonia</u>	-	++	+	+	tr	+++	-	-	+	-	-	-	-	-	-	-	-	-	-
<u>Limonia trifolia</u>	tr?	++	-	tr	++	+	-	+	-	-	-	-	-	-	-	-	-	-	-
<u>Feronia limonia</u>	+	+	+	+	+	+	+	-	+	-	-	-	-	-	-	-	-	-	-



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APPENDIX III  
(cont'd)

APPENDIX III (cont'd)	Group A				Group B			Group C											
	gent.	fer.	caff.	ell.	sin.	p-c	gall.	umb.	van.	syr.	p-OH-B	rutin	aesc.	p-cat.	scop.	melil.	P/L	phlor.	h-gent.
CNEORACEAE																			
SIMARUBACEAE																			
<u>Ailanthus altissima</u>	-	++	+++	+++	tr	+	+	-	-	-	-	+	-	-	-	-	-	-	-
<u>Kirkia acuminata</u>	+?	-	tr	+++	-	-	+++	-	-	tr?	-	+	-	-	-	-	-	-	-
PICRODENDRACEAE																			
BURSERACEAE																			
<u>Commiphora merkeri</u>	-	tr	++	++	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-
MELIACEAE																			
<u>Ptaeroxylon obliquum</u>	-	+++	+	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Khaya nyasica</u>	+?	+	+	++	-	+	+	-	-	-	-	+++	+	-	-	-	-	-	-
<u>Swietenia macrophylla</u>	+	tr	+	++	-	+	tr?	-	+	-	+	-	-	-	-	-	-	-	-
<u>Carapa guianensis</u>	+	tr?	+	++	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>C. procera</u>	-	tr	+++	+	tr	-	tr	-	tr?	tr?	-	+	-	+	-	-	-	-	-
<u>Cipadessa cinerascens</u>	+?	tr	+	tr	+	+	-	-	-	+	-	-	-	-	-	-	-	-	-
<u>Melia azedarach</u>	-	+	+	tr	+	tr?	-	-	-	-	-	-	-	-	-	-	-	-	-
AKANIACEAE																			
<u>Akania lucens</u>	+	+	+	+	-	++	-	-	tr?	-	+	-	-	-	-	-	-	-	-
MALPIGHIACEAE																			
<u>Gaudichaudia cynanchoides</u>	-	tr?	-	-	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Malpighia coccigera</u>	-	tr	-	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>M. cubensis</u>	tr?	tr	+	+	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-
TRIGONIACEAE																			
VOCHYSIACEAE																			
TREMANDRACEAE																			
<u>Platytheca verticillata</u>	-	tr?	+	+++	+	+	+++	-	+	tr?	-	-	-	-	-	-	-	-	-
<u>Tetratheca t(h)ymifolia</u>	-	-	-	+	-	+	+	-	-	-	-	-	-	-	-	-	-	-	-
POLYGALACEAE																			
<u>Polygala myrtifolia</u>	+	+	tr?	+	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-
DICHAPETALACEAE																			
CALLITRICHACEAE																			
BALSAMINACEAE																			
<u>Impatiens flaccida</u>	+	+	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>I. oliveri</u>	-	tr	tr	+	-	-	-	-	+	-	-	+	-	-	-	-	-	-	-
BUXACEAE																			
<u>Sarcococca confusa</u>	-	+	-	+	-	tr	-	-	-	-	-	-	-	-	-	-	+	-	-
<u>S. ruscifolia</u>	-	tr?	-	+	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Pachysandra procumbens</u>	+?	+	+	++	+	+	-	-	-	-	-	-	-	-	-	-	-	-	-
<u>Simmondsia chinensis</u>	-	+	+	+++	+	+	-	-	tr	tr	+	-	-	+	+	+	-	-	-

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Key to phenolic constituents:

gent. = gentisic  
fer. = ferulic  
caff. = caffeic  
ell. = ellagic  
sin. = sinapic  
p-c = para-coumaric  
gall. = gallic  
umb. = umbelliferone  
van. = vanillic  
syr. = syringic

p-OH-B = para-hydroxybenzoic  
rutin = rutin  
aesc. = aesculetin  
p-cat. = protocatechuic  
scop. = scopoletin  
melil. = melilotic  
P/L = paraphenyl-hydroxylactic  
phlor. = phloretic  
h-gent. = homogentisic

## APPENDIX IV

Fatty Acid Composition of Seeds  
(from Eckey, 1954; Hilditch,  
1956; and others)

Family	Major Fatty Acids	Comments
Geraniaceae		No information
Oxalidaceae		No information
Tropaeolaceae	erucic, eicosenoic	Very similar to Cruciferae
Zygophyllaceae	palmitic, oleic, linoleic	
Linaceae	linolenic	
Erythroxylaceae		No information
Limnanthaceae	eicosenoic	
Euphorbiaceae	linoleic, oleic, conj'd polyethenoic acids	Unusual conj'd poly- ethenoic acids
Daphniphyllaceae	palmitic, oleic, lino- leic	
Rutaceae	palmitic, oleic, lino- leic	
Cneoraceae		No information
Simarubaceae	<u>Picrasma</u> -petroselinic <u>Picramnia</u> -tariric <u>Irvingia</u> -myristic, lauric <u>Ailanthus</u> -oleic, lino- leic	Acids vary with genera
Burseraceae	stearic	
Meliaceae	stearic	
Akaniaceae		No information
Malpighiaceae		No information
Trigeniaceae		No information
Vochysiaceae	myristic, lauric	
Tremandraceae		No information
Polygalaceae		No information
Picrodendraceae		No information
Dichapetalaceae	monofluoroacetic	
Callitrichaceae		No information
Balsaminaceae	linoleic, oleic; in 2 <u>Impatiens</u> spp., parinaric	
Buxaceae	eicosenoic in <u>Simmondsia</u>	

## APPENDIX V

## Alkaloidal Distribution

(from Henry, 1949, and  
others)

family	alkaloids present	absent
Geraniaceae	x	
Oxalidaceae		x
Tropaeolaceae	no information*	
Zygophyllaceae	x	
Linaceae	no information*	
Erythroxylaceae	xxx	
Limnanthaceae	no information*	
Euphorbiaceae	xx	
Daphniphyllaceae	xx	
Rutaceae	xxx	
Cneoraceae	no information*	
Simarubaceae	xx	
Picrodendraceae	x?	
Burseraceae	x	
Akaniaceae	x	
Meliaceae	x	
Trigoniaceae	no information*	
Vochysiaceae	no information*	
Malpighiaceae	x	
Tremandraceae	no information*	
Polygalaceae	x	
Dichapetalaceae	x	
Callitrichaceae	no information*	
Balsaminaceae	no information*	
Buxaceae	xxx	

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\*since no information is available, it is more  
than likely that alkaloids are absent.

## APPENDIX VI

## The Distribution of Saponins in the "Geraniales"

(from Amarasingham et al, 1964)

genus	saponins	
	present	absent
<b>Linaceae</b>		
<u>Roucheria</u>		-1
<u>Ixonanthes</u>		-1
<b>Euphorbiaceae</b>		
<u>Baccaurea</u>		-4
<u>Aporosa</u>		-5
<u>Antidesma</u>		-2
<u>Glechidion</u>	+2	
<u>Phyllanthus</u>		-1
<u>Drypetes</u>	+1	
<u>Bischoffia</u>		-1
<u>Cleistanthus</u>	+1	-1
<u>Croton</u>		-4
<u>Mallotus</u>		-4
<u>Coecoceras</u>		-1
<u>Coelodepas</u>		-1
<u>Macaranga</u>		-2
<u>Acalypha</u>		-1
<u>Epiprinus</u>		-1
<u>Trigonostemon</u>		-2
<u>Galearia</u>	+1	
<u>Microdesmis</u>		-1
<u>Manihot</u>		-1
<u>Elateriospermum</u>		-1
<u>Gelonium</u>		-3
<u>Neoscortechinia</u>		-1
<u>Pimaleodendron</u>		-1
<u>Ptychopyxis</u>		-1
<u>Betryophora</u>		-1
<b>Rutaceae</b>		
<u>Xanthoxylum</u>		-2
<u>Glycosmis</u>	+1	
<u>Micromelum</u>		-1
<u>Luvunga</u>	+1	
<u>Atalantia</u>		-1
<b>Simarubaceae</b>		
<u>Eurycoma</u>		-1

## APPENDIX VI (cont'd)

genus	saponins	
	present	absent
Burseraceae		
<u>Triomma</u>		-1
<u>Canarium</u>		-2
<u>Dacryodes</u>		-2
<u>Santiria</u>		-1
Meliaceae		
<u>Walsura</u>		-1
<u>Aphanamixis</u>		-2
<u>Amoera?</u>		-1
<u>Aglaia</u>		-6
<u>Chisocheton</u>	+2	-2
<u>Dysoxylum</u>	+1	-4
Malpighiaceae		
<u>Hiptage</u>		-1
Polygalaceae		
<u>Xanthophyllum</u>	+1	-1
Dichapetalaceae		
<u>Dichapetalum</u>		-1



## APPENDIX VII

## Aluminum Accumulation in the "Geraniales"

family	Aluminum Accumulators	
	+	-
Geraniaceae		2/2
Oxalidaceae		1/1
Tropaeolaceae		
Zygophyllaceae		
Linaceae		
Erythroxylaceae		1/2
Euphorbiaceae	4/7	26/51
Daphniphyllaceae	1/1	
Rutaceae	1/1	17/58
Simarubaceae		3/3
Cneoraceae		
Burseraceae		1/2
Meliaceae		10/18
Akaniaceae		
Malpighiaceae		1/1
Trigoniaceae		
Vochysiaceae	6/?	
Tremandraceae		
Polygalaceae	2/?	5/15
Picrodendraceae		

(No information known on Dichapetalaceae, Callitrichaceae  
Balsaminaceae, and Buxaceae.)

## APPENDIX VIII

## The Distribution of Polyols in the "Geraniales"

(from Plouvier, 1963, and others)

genus	a. aliphatic	b. cyclitol	D-pinmitol	L-quebra- chitol
	D-polygalitol	L-inositol		
Geraniaceae				
<u>Geranium</u>			-4	
<u>Erodium</u>			-2	
<u>Pelargonium</u>			-2	
Oxalidaceae				
<u>Oxalis</u>			-2	
Tropaeolaceae				
<u>Tropaeolum</u>			-1	
Zygophyllaceae				
<u>Zygophyllum</u>			+1	
<u>Peganum</u>			-1	
Linaceae				
<u>Linum</u>			-2	
Euphorbiaceae				
<u>Securinega</u>			-1	-1
<u>Andrachne</u>			-1	-1
<u>Hevea</u>		+1		+1
<u>Mercurialis</u>			-1	-1
<u>Acalypha</u>			-1	+1
				-12
<u>Ricinus</u>			-1	-1
<u>Euphorbia</u>		+1	-6	-6
Polygalaceae				
<u>Polygala</u>	+4			

## APPENDIX IX

## Distribution of Leucoanthocyanins and Flavonols

(from Bate-Smith, 1957, 1962)

family	leucoanthocyanins		flavonols		
	delphinidin	cyanidin	myricetin	quercetin	kaempferol
Geraniaceae	-2/2/7	-2/7	-2/5 +1/2	++2/7	+2/7
Oxalidaceae	-2/5 +1/2	-1/3 +2/4	-2/7	-2/6 +1/1	-2/7
Tropaeolaceae	-1/2	+1/2	-1/2	-1/2	-1/2
Linaceae	-2/3	-2/3	-2/3	-2/3	-2/3
Erythroxylaceae	-1/1	+1/1	-1/1	++1/1	+1/1
Zygophyllaceae	-1/1	-1/1	-1/1	+1/1	+1/1
Limnanthaceae	+1/1	+1/1	+1/1	+1/1	+1/1
Eupherbiaceae	-7/10 +1/1	-6/9 +2/2	-7/11	-5/6 +3/5	-3/5 +4/6
Daphniphyllaceae	-1/1	-1/1	-1/1	+1/1	-1/1
Rutaceae	-10/12 +5/5	+9/10 +5/6	-8/9 +5/5	-4/4 +9/10	-4/4 +8/8
Cneoraceae	-1/1	-1/1	-1/1	+1/1	+1/1
Simarubaceae	-2/4	-2/3	+2/4	-1/1 +1/3	-1/1 +1/3
Burseraceae	-1/2	+1/2	-1/2	+1/2	+1/2
Akaniaceae	+1/1	+1/1	-1/1	+1/1	+1/1
Meliaceae	-3/4	-2/2 +1/2	-3/4	+3/4	-1/1 +2/3

## APPENDIX IX (Cont'd)

family	leucoanthocyanins		flavonols		
	delphinidin	cyanidin	myricetin	quercetin	kaempferol
Malpighiaceae	-4/4	-2/2 +2/2	-4/4	-3/3 +1/1	-2/2 +2/2
Tremandraceae	-2/2	-2/2	+2/2	+1/1	-2/2
Polygalaceae	-1/4	-1/4	-1/4	-1/1 +1/3	-1/2 +1/2
Callitrichaceae	-1/1	-1/1	-1/1	-1/1	-1/1
Balsaminaceae	-1/2 +1/1	-1/1 +1/3	-1/3	-1/1 +1/3	+1/4
Buxaceae	-2/2	-2/2	-2/2	+3/3	+3/3

## APPENDIX X

"Distribution of alkaloids in the Rutaceae" (from Price, 1963)

	acridines	furo- quinolines	quinolines	quinazolines	indolo- quinazolines	canthinones	imidazoles	benzyliso- quinolines	amines or amides
<b>Rutoideae</b>									
<u>Zanthoxylum</u>		X			X	X		X	X
<u>Fagara</u>		X						X	X
<u>Evedia</u>	X	X			X			X	
<u>Orixa</u>		X							
<u>Melicope</u>	X	X							
<u>Pentaceras</u>						X			
<u>Lunasia</u>		X	X						
<u>Medicosma</u>		X							
<u>Choisya</u>		X							
<u>Platydesma</u>		X	X						
<u>Boenninghausenia</u>		X							
<u>Ruta</u>		X	X						
<u>Haplophyllum</u>	X	X							
<u>Dictamnus</u>		X							
<u>Boronia</u>		X	X						
<u>Eriostemon</u>		X							
<u>Phebalium</u>		X							
<u>Geleznovia</u>		X							
<u>Pilocarpus</u>							X		
<u>Cusparia</u>		X	X						
<u>Galipea</u>			X						
<b>Dictyolomatoideae</b>									
<u>Dictyoloma</u>									X
<b>Flindersioideae</b>									
<u>Flindersia</u>		X							
<u>Chloroxylon</u>		X							
<b>Spathelioideae</b>									
<b>Toddalioideae</b>									
<u>Phellodendron</u>								X	X
<u>Balfourodendron</u>	X	X	X						
<u>Casimiroa</u>		X	X				X		X
<u>Vepris</u>		X							
<u>Toddalia</u>								X	
<u>Acronychia</u>	X	X							
<u>Hortia</u>		X			X				
<u>Skimmia</u>		X							
<u>Teclea</u>	X	X							
<b>Aurantioideae</b>									
<u>Glycosmis</u>	X	X		X					
<u>Aegle</u>		X		X					X
<u>Citrus</u>			X					X	X

Classified according to Engler and Harms, 1931

## APPENDIX XI

The occurrence of Ca Oxalate  
(from Metcalfe and  
Chalk, 1950)

	Solitary crystals	Cluster crystals	Styloids	Raphides	Acicular crystals	Crystal sand	Sphaerocrystals (incomplete list)	Crystalline masses
Geraniaceae	X	X	(X)					(X)
Oxalidaceae	X					(X)		
Zygophyllaceae	X	X	(X)	(X)	(X)			(X)
Linaceae	X	(X)						
Humiriaceae	X	X						
Erythroxylaceae	X							
Euphorbiaceae	X	X	(X)				(X)	
Daphniphyllaceae		X						
Rutaceae	XX	XX	(X)	(X)		(X)		
Cneeraceae	(X)							
Simarubaceae	X	X	(X)					
Burseraceae	X	X						
Meliaceae	X	X						
Akaniaceae	(X)	X						
Malpighiaceae	X	X	X					
Trigonaceae	X	X						
Vochysiaceae	X	X						
Tremandraceae	X	X						
Polygalaceae	X	X						
Balsaminaceae				XX				
Buxaceae	X	X				(X)		

Key to symbol: ( ) = limited in distribution

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