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A Chemotaxonomic Study of the Rutales of Scholz

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ABSTRACT

On the basis of chemical evidence, the order <u>Rutales</u> seems to be a homogeneous one, if we consider it to contain the families <u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Burseraceae</u> and <u>Meliaceae</u>.

We know too little as yet of the chemistry of other families of 'our' order (the <u>Rutales</u> of Scholz, in Engler, Syll. 12, 1964) to say whether or not they constitute with the above four families a homogeneous group within the <u>Rutales</u>. The chemistry of the <u>Malpighiaceae</u>, however, is consistent with a position near the <u>Meliaceae</u>.

The chemical evidence also supports the creation of two more small families -- <u>Flindersiaceae</u> (<u>Flindersia</u> and <u>Chloroxylon</u>), and <u>Ptaeroxylaceae</u> (<u>Ptaeroxylon</u> and <u>Cedrelopsis</u>) -- which should be placed, perhaps, between <u>Rutaceae</u> and <u>Meliaceae</u>; and the separation of <u>Picramnia</u> (<u>Picramnioideae</u>) from the <u>Simaroubaceae</u>, <u>Mundtia</u> (if our material was genuine) from the <u>Polygalaceae</u>, and Fagara from <u>Zanthoxylum</u>.

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A CHEMOTAXONOMIC STUDY OF THE "RUTALES" OF SCHOLZ (IN ENGLER, SYLLABUS 12, 1964)

by

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INTRODUCTION

The term systematics can be used, as defined by Simpson (1961), as the scientific study of the kinds and diversity of organisms and of any and all relationships between them. Taxonomy is that part of systematics which deals with the study of classification, including its bases, principles, procedures and rules; while classification in a biological sense is the process of ordering plants into groups which are arranged hierarchically.

As stated by Davis and Heywood (1963):

"Certain disciplines of biology are, of course, so closely tied up with taxonomy that they cannot be practised without the use of a basic classification. These include Cytology, Genetics, Ecology, Phytosociology, Comparative Anatomy, Palynology, Palaeobotany and Plant Geography".

Taxonomy, therefore, is one of the very important biological sciences.

A. <u>History of Taxonomy</u>

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In order to survey the field of taxonomy at the present day, it is necessary to have some knowledge of the history of the subject and the development of ideas associated with it.

The early history of botany is largely a history of the uses of plants in medicine, and the root-gatherers and herbalists of the past began, many centuries ago, to group plants having similar "virtues" or medicinal properties, (Gibbs, 1963). Greene (1909), in his Landmarks of Botanical History, emphasized the major early descriptive developments in taxonomic botany, particularly as related to specific individuals and their contributions to systematics. Beginning with prehistoric time, he recognized as foremost (1) the descriptive contributions of Aristotle and Theophrastus, followed by a long period of lasting quiescence to the fifteenth century, (2) the significance of the observations of the herbalists Tragus, Brunfels, Bauhin, <u>et al</u>. of the sixteenth century (3) the first distinction of the monocotyledons and dicotyledons by John Ray in 1703, (4) recognition of sexual characters and their significance by Linnaeus and others in the mid-eighteenth century, and so on.

Lawrence (1951), in an excellent treatment of the history of classification, says:

"Many different classifications of plants have been proposed. They are recognizable as being or approaching one of three types: artificial, natural and phylogenetic. An artificial system classifies organisms for convenience, primarily as an aid to identification, and usually by means of one or a few characters. A natural system reflects the situation as it is believed to exist in nature and utilizes all information available at the time. A phylogenetic system classifies organisms according to their evolutionary sequence, it reflects genetic relationships, and it enables one to determine at a glance the ancestors or derivatives (when present) of any taxon. The present state of man's knowledge of nature is too scant to enable one to construct a phylogenetic classification, and the so-called phylogenetic systems represent approaches toward an objective and in reality are mixed and are formed by the combination of natural and phylogenetic evidence".

Theophrastus (370-287, B.C.), the "father of Botany", classified plants according to the growth habit. Thus he had four groups: herbs, undershrubs, shrubs, and trees. This was the period of artificial classification, when plants were grouped according to their habit or number of a certain organ.

The natural system came with John Ray (1628-1704) who for the first time recognized the importance of the embryo and the presence of one or two cotyledons. Several outstanding taxonomists during the 1800's classified plants by a "natural" system, they often made no serious or conscious attempt to place the major taxa together according to their evolutionary relationship. For example, such outstanding workers as Bentham and Hooker, in their classic <u>Genera</u> <u>Plantarum</u>, placed the gymnosperms between the Dicotyledons and Monocotyledons instead of placing the latter two together as most phyletic workers have done since that time. None-theless Bentham and Hooker's work remains to this day a useful system, mostly "natural", but not phylogenetic.

Nearly all systems of classification are based in part, at least, on arbitrary principles as to what constitutes primitiveness or, in turn, advancement. Several such principles have been advanced, some of a contradictory nature depending on the point of view of the systematist (Just, 1948; Constance, 1958). For example, Engler and Diels (1936) considered that the majority of plants with simple unisexual flowers are

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primitive, while Bessey, Hutchinson and others have considered these same floral types indicative of advancement, the condition having developed by reduction processes from complete, bisexual flowers.

Hutchinson (1959), in setting forth his view on the phylogeny of the angiosperms, adopts the principle that "the spiral arrangement of leaves on the stem and of the floral leaves precedes that of the opposite and whorled type". Cronquist (1955), on the contrary, in considering the phylogeny of the family <u>Compositae</u>, considered opposite leaves to be the primitive condition in that family, but this does not mean necessarily that he considers this to be the primitive condition for angiosperms generally. Similarly, Hutchinson's view that the herbaceous habit is primitive in the <u>Ranunculaceae</u> does not conflict with his supposition that woodiness is the primitive condition for the angiosperms generally.

Several systems of classification, particularly of the flowering plants and usually to the level of family, have been proposed in recent years. These include those of Cronquist

(1968), Benson (1957), Hutchinson (1959; 1969 for dicots), Takhtajan (1969) and others. However, only a few systems have gained wide acceptance or attention, the more important being those of Bentham & Hooker, Engler, Bessey, and Hutchinson.

No botanist, however, would claim that any one of the many present systems of classification that have been put

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forward, represents an ideal or final solution. There are still many problems at all taxonomic levels and, in particular, the interrelations between higher categories such as orders are very unclear. Many difficult taxonomic problems arise from phenomena such as parallel, convergent, and divergent evolution, which may cause botanists to assume closer or more distant genetic relations between plants or groups of plants than those actually existing (Erdtman, 1968). This is argued by Davis and Heywood (1963):

"Classification, many taxonomists claim, should be based on, or reflect, phylogeny. This aim we believe, is unrealistic in a group with an extremely inadequate fossil record, Indeed, the whole conception of phylogenetic classification is, we believe, a mistake except around the species level in favourable and well-studied groups; and even there phyletic relationship often conflicts with genetic relationship as expressed by phenotypic resemblances".

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) states

"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

Despite apparent disagreements over the purpose and interpretation of classifications, it is widely accepted that the

most useful assessment of the overall relationships of organisms is obtained by using the largest possible number of similarities and differences. In the original definition of phenetic classification (Cain and Harrison, 1960) it was made clear that it was based on all available observable characters (including genetic data), not just morphological ones. Thus phytochemical characters are included, along with cytological, anatomical, palynological and other attributes, in the best classifications.

Botanical classification rests largely on comparative studies of morphological and anatomical characters. Roads to phylogenetic systems were opened by Darwin, and a deeper understanding of the mechanism of heredity resulted from the fundamental experimental studies of Mendel and his successors. Their work made it abundantly clear that chemical as well as morphological characters of plants are determined by genetic factors by mechanisms the more precise natures of which are now gradually being unveiled.

Morphology is the outward expression of genes. Since genes or chromosomes 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 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".

It is evident that systematic investigations of the chemical characters of plants are likely to become of great supplementary value to classical plant taxonomy. Chemical characters of plants have the great advantage that they can be exactly defined.

B. Development of Chemotaxonomy

Chemical plant taxonomy or chemotaxonomy of plants, as defined by Hegnauer (1967), is a scientific investigation of the potentialities of chemical characters for the study of problems of plant taxonomy and phylogeny.

Chemical characteristics of plants have been noted and used by taxonomists for centuries. Gibbs (1965) wrote:

"Some of the earliest taxonomy was, in a sense, chemotaxonomic. The first groping towards some of our oldest families - oldest, that is, in establishment - was due to the recognition of 'virtues' in common."

De Candolle believed Rudolph Jacob Camerarius to be the first clearly to express the connection between forms of plants and their properties. In some of the writings of A.P. de Candolle, as Hegnauer (1958) has noted, considerable attention was given to the chemical properties of plants as correlated with their morphological characters. Examples from De Candolle cited by

Hegnauer were the observations that all <u>Cinchona</u> species combat fever, all <u>Pinus</u> species produce terpenes, all <u>Amentifere</u> have astringent bark, and all <u>Convolvulaceae</u> are laxative.

Petiver, in 1699, as Gibbs (1965) has noted, recognized the families <u>Umbelliferae</u>, <u>Labiatae</u>, and what we now call <u>Cruciferae</u>, largely on medicinal (chemical) characters.

Lindley, in 1830, as cited by Gibbs (1965), wrote of the <u>Amygdaleae</u> that they are:

"Distinguished from Rosaceae and Pomaceae by their fruit being a drupe, their bark yielding gum, and by the presence of hydrocyanic acid; from Leguminosae by the latter character, and ... from Chrysobalaneae by their hydrocyanic acid ..."

Abbott (1886) prophesied:

"There has been comparatively little study of the chemical principles of plants from a purely botanical view. It promises to become a new field of research".

Greshoff (1891) suggested the use of chemistry in taxonomy. He said that a "chemical description" should be part of a formal description of a new genus. At Kew, he looked at plants for tannins, alkaloids, cyanogenic substances, and saponins. In 1909, he emphasized the presence of HCN in plants, and vividly described it in Platanus:

"Indeed, in the ordinary plane-tree of 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".

Van Romburgh (1899) studied the occurrence of HCN, methyl salicylate, and acetone in plants, while Treub (1907) studied the role of HCN.

At present, botanists such as Gibbs and biochemists such as Florkin have long been aware that chemical data are potentially of great use to systematics as broadly defined, but it is only in the last ten years or so that biochemical systematics has come to occupy a major role. The two main reasons for this have been the development of rapid and efficient screening techniques such as chromatography and electrophoresis, and, as a result of this the rapid identification of a large number of organic compounds by these methods, and the realization that they have a wide systematic value and can contribute to the solution of many taxonomic problems.

Many constituents are looked for in plants today. Compounds found only in a single species (unique compounds) may only possess taxonomic value at the species level. Many natural products are ubiquitous and are, for that reason, of little of no taxonomic interest. Proteins and nucleic acids are truly ubiquitous but are, nevertheless, of potentially great taxonomic value. In spite of their high molecular weight, compounds such as cellulose, protein and nucleic acids can be assigned definite structures. The lignins are less regularly constructed, probably being mixtures of condensation products. Nevertheless, the lignins appear to possess great taxonomic value. Very recent developments suggest that biochemical data, especially at the "macromolecular" level, may assist in working out some of the evolutionary pathways by which groups have arisen.

Among the more popular small molecular weight compounds investigated in this fashion have been those of amino acids, alkaloids, terpenoids and flavonoids, these four chemical classes are widespread among plant groups but in each may be found certain subclasses which are restricted to closely related taxa.

Distributional surveys for secondary compounds will become much more meaningful after knowledge of their biosynthetic origins has accumulated. This point has been repeatedly emphasized by a number of workers (Swain, 1966), and in at least one recent text on phytochemistry (Mentzer and Fatainoff, 1964) the secondary compounds are arranged according to the metabolic pathways leading to their production; e.g., acetic acid (C_2)n derivatives; isoprenoid (C_5)n derivatives; shikimic acid derivatives, etc. Thus, as H*G.H. Erdtman (1963) writes:

"Chemotaxonomy is essentially the investigation of chemical compounds or groups of biosynthetically related compounds, in a series of related, or supposedlyrelated plants".

Therefore, for purposes of classification, knowledge about the genesis of plant constituents is just as important as knowledge of their structures. The general tendencies of evolution for metabolic patterns and for individual categories of constituents of angiosperms, are scarcely known at present. Alkaloids, for instance, have been detected in fungi, pteridophyta, gymnospermae and angiospermae, but it is virtually impossible to indicate evolutionary trends concerning their structure

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and distribution. Within taxa of lower rank, such as species in a genus, genera in a family, and even families in an order, such tendencies may emerge in the near future, but with regard to the whole plant kingdom such tendencies seem not to exist at all or are still far from being conceived clearly.

At the present time, there are many well developed methods available for both botanists and chemists who are interested in chemosystematic study.

The simple tests used by the present author and others are useful for preliminary surveys of the biochemistry of plants.

The techniques of paper-chromatography, although they require skill, are relatively simple and rapid, and they only need a small amount of leaf material. The author has used 2way paper chromatography for the separation and subsequent identification of simple phenolic compounds of leaves. Today this method, as well as thin-layer chromatography, is extensively used.

Gas chromatography, where the extracts are volatilized and run as a gas through a liquid column instead of on paper, requires more expensive apparatus but can be largely automated, the results being presented in the form of a graph in which the peaks represent the abundance of molecules with different features. It is employed for the screening of oils as in mints, or terpenes as in the pines.

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With the techniques of gel electrophoresis, Boulter and his associates at Liverpool worked on protein bands obtained from the albumin fraction of seeds of various genera of the <u>Leguminosae</u> and they have shown that the technique may be of great value in supporting taxonomic arrangements or suggesting where revision ought to be considered.

Mass spectrometry is increasingly replacing the classical elementary analyses and determinations of molecular weights. The combination of gas chromatography and mass spectrometry makes it possible to investigate products available only in trace amounts and to identify their components by comparing their mass spectra with those of known substances. The fragmentation reactions of organic compounds are being intensely studied and, no doubt, in the future it will become possible to elucidate the structure of many substances by mass spectrometry alone.

One of the most important recently developed techniques is that of nuclear magnetic resonance spectroscopy (NMR). It has opened new avenues of approach to a whole world of problems involving small molecules, including the phenolic and other secondary metabolic substances. The principles and methods involved make its use applicable to the small amounts of substances available in many types of biological experiments.

The UV, IR, and NMR spectra of many substances give valuable information about their structure, configurations, and

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conformations. Many important but difficult problems concerning the absolute configuration of natural products are now easily solved by examination of their ORD curves.

Structural investigation and studies on the distribution of natural products in the plant kingdom open roads to fields such as taxonomy and, perhaps, plant evolution.

C. Purpose of This Research

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The purpose of this research was to gather supplemental (that is, <u>chemical</u>) evidence to reveal (1) Whether the "Rutales" of Scholz (in Engler 12th Syllabus, 1964) is a natural group. (2) The affinities and/or absence of affinities between the families of Scholz's <u>Rutales</u>. (3) Gaps in our knowledge of these families, which might suggest further work to be done on this group.

REVIEW OF LITERATURE

Α. The Order "Rutales"

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The position of many taxa of Scholz's "Rutales" in the natural system of plants is still highly uncertain. This applies to all levels of taxonomic categories, e.g. species in a genus, genera in a family (e.g. <u>Ptaeroxylon</u> in Ptaeroxylonaceae?), families in an order (e.g. Polygalaceae in Polygalales?; Malpighiaceae in Malpighiales?).

Scholz in the 12th "Syllabus" of Engler and Melchior (1964) classifies the "Rutales" as follows:

Suborder Rutineae:

Rutaceae (204/1600) Cneoraceae (2/3)Simaroubaceae (26/100) Picrodendraceae (1/3)Burseraceae (20/600) Meliaceae (50/1400)Akaniaceae (1/1) Suborder Malpighineae: Malpighiaceae (65/800) Trigoniaceae (4/35) Vochysiaceae (6/200) Suborder Polygalineae: Tremandraceae (3/30)

Polygalaceae (13/800)

The above 12 families have been placed in several different arrangements and groups under various names over the years. The positions attributed to them in some systems of angiosperms are given in Table I. This table does not show all the other

families which some of the systematists include, but rather gives the more common ones and the names of the orders under which they have been grouped. I choose a few examples to illustrate this point:

Hutchinson (1969) believes that not all the "Rutales" of the 12th Syllabus are representatives of one line of development (woody). Accordingly, he splits them into no less than eight orders: <u>Rutaceae</u>, <u>Simaroubaceae</u>, and <u>Burseraceae</u> form for him the order <u>Rutales</u> which derives from the <u>Celastrales</u>. His <u>Polygalales</u> includes <u>Polygalaceae</u>, <u>Trigoniaceae</u>, <u>Vochysiaceae</u> (and <u>Krameriaceae</u>). <u>Malpighiaceae</u> and <u>Irvingiaceae</u> (from <u>Simaroubaceae</u>) with some other families form his order <u>Malpighiales</u>. The remaining five families <u>Meliaceae</u>, <u>Akaniaceae</u>, <u>Tremandraceae</u>, <u>Cneoraceae</u> and <u>Picrodendraceae</u>, Hutchinson considers to be members of the <u>Meliales</u>, <u>Sapindales</u>, <u>Pittosporales</u>, <u>Celastrales</u> and <u>Juglandales</u> respectively!

An order "Rutales" has been established by other systematists, too --Lindley (1853), Gunderson (1950), Rendle (1938, 1952), Takhtajan (1959, 1969), Pulle (1952), Benson (1957), and Boivin (1956).

Gunderson (1950) had a "Geranium group", placed between the "Malva group" and the "Dianthiflorae", with <u>Rutales</u>, <u>Juglandales</u>, <u>Sapindales</u>, <u>Celastrales</u> and <u>Geraniales</u>. Included in his <u>Rutales</u> are <u>Burseraceae</u>, <u>Cneoraceae</u>, <u>Simaroubaceae</u>, <u>Rutaceae</u>, and <u>Meliaceae</u> of 'our' Rutales. The <u>Akaniaceae</u>,

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TABLE I

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Numbers of Taxonomists Assigning the Families of Scholz's Rutales to the Rutales and to other Orders

		ORDERS									
	FAMILIES	Rutales	Geraniales	Terebinthales	Sapindales	Malpighiales	Polygalales	Juglandales	Meliales	Cneorales	
	Rutaceae	9	3	6	1	0	0	0	0	0	
	Cneoraceae	5	2	2	1	0	0	0	0	1	
H	Simaroubaceae	9	3	6	1	0	0	0	0	0	
NDER	Picrodendraceae	1	0	Ο.	0	0	0	2	0	0	
SUBORDER	Burseraceae	7	2	4	1	0	0	0	0	0	
S	Meliaceae	7	3	6	1	0	0	0	2	0	
	Akaniaceae	2	0	4	6	0	0	0	0	0	
2	Malpighiaceae	1	7	1	2	3	3	0	0	0	
0.	Trigoniaceae	1	1	3	1	1	7	0	0	0	
	Vochysiaceae	1	1	3	2	1	7	0	0	0	
	Tremandraceae	1	1	3	2	0	5	0	0	0	
s.0	Polygalaceae	1	1	3	3	. 0 .	10	. 0	0	. 0.	

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<u>Malpighiaceae</u>, <u>Vochysiaceae</u> (including <u>Trigonia</u>), <u>Tremandraceae</u> and <u>Polygalaceae</u> are in the <u>Sapindales</u>.

Rendle (1952) placed four of 'our' families in the <u>Rutales</u> --<u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Burseraceae</u> and <u>Meliaceae</u>. He included <u>Polygalaceae</u> doubtfully in the <u>Sapindales</u>, while <u>Malpighiaceae</u> was put in <u>Geraniales</u>. He also believed that <u>Geraniales</u>, <u>Rutales</u> and <u>Sapindales</u> are closely allied to each other.

Pulle (1952) had the following scheme:

	1	<u>Polygalales</u>
<u>Clusiales</u> ->	Rutales	Tremandr.
	(Zygophyll.)	Polygal.
	Cneor.	Malpighiales
	Rut.	Malpighi.
	Burser.	Trigoni.
	Simaroub.	Vochysi.
	Meli.	
	Akani.	

Takhtajan (1969) has a <u>Rutales</u> much like Pulle's with <u>Burser., Simaroub., Rut., Cneor., and Meli.</u> He puts <u>Malpighi</u>. in his <u>Geraniales</u>, and <u>Trigoni., Vochysi., Polygal.</u>, and <u>Tremandr</u>. in his <u>Polygalales</u>. The <u>Akani</u>. are in <u>Sapindales</u>, and the <u>Picrodendr</u>. in <u>Euphorbiales</u>.

Benson (1957) separated 'our' families into three different orders: <u>Rutales</u>, <u>Polygalales</u> and <u>Geraniales</u>. He regarded <u>Akaniaceae</u> and <u>Tremandraceae</u> to be of uncertain position, but he believed that they are related to the complex of the <u>Geraniales</u>, <u>Rutales</u> and <u>Sapindales</u>.

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Boivin (1956) placed the <u>Rutaceae</u> and <u>Simaroubaceae</u> in the <u>Rutales; Meliaceae</u> as the only family of <u>Meliales;</u> <u>Polygalaceae</u>, <u>Trigoniaceae</u> and <u>Vochysiaceae</u> as <u>Polygalales;</u> while <u>Malpighiaceae</u> was raised to ordinal rank again. <u>Akaniaceae</u>, <u>Cneoraceae</u> and <u>Tremandraceae</u> were put in <u>Sapindales</u>, <u>Celastrales</u> and <u>Pittosporales</u> respectively.

The early systematist Lindley (1853) included in the <u>Rutales</u> the <u>Rutaceae</u>, <u>Simaroubaceae</u> and <u>Meliaceae</u>; while <u>Malpighiaceae</u>, <u>Vochysiaceae</u>, <u>Tremandraceae</u> and <u>Polygalaceae</u> were in <u>Sapindales</u>.

On the other hand, Cronquist (1957 ; 1968) does not segregate <u>Rutales</u> from <u>Sapindales</u> as many taxonomists do. He places six of 'our' families -- <u>Akaniaceae</u>, <u>Burseraceae</u>, <u>Simaroubaceae</u>, <u>Cneoraceae</u>, <u>Rutaceae</u> and <u>Meliaceae</u> -- in <u>Sapindales</u>. He refers <u>Polygalaceae</u>, <u>Malpighiaceae</u>, <u>Trigoniaceae</u>, <u>Vochysiaceae</u>, <u>Tremandraceae</u>(and <u>Xanthophyllaceae</u> and <u>Krameriaceae</u>) to the <u>Polygalales</u>.

He says that <u>Picrodendron</u> seems best associated with the <u>Juglandaceae</u> in the <u>Juglandales</u>. Bessey (1951) included all of 'our' families except <u>Picrodendraceae</u> and <u>Akaniaceae</u> in his order <u>Geraniales</u>.

Soo (1953) and Wettstein (1935) included most of the families of the Syll. 12 <u>Rutales</u> in an order <u>Terebinthales</u>, but the family <u>Cneoraceae</u> was placed in <u>Geraniales</u> by Soo, while <u>Cneoraceae</u> and <u>Malpighiaceae</u> were referred by Wettstein to <u>Gruinales</u>.

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Skottsberg (1940) included in his order <u>Terebinthales</u> only <u>Rutaceae</u>, <u>Cneoraceae</u>, <u>Simaroubaceae</u>, <u>Meliaceae</u> and <u>Akaniaceae</u>. The <u>Malpighiaceae</u>, <u>Trigoniaceae</u>, <u>Vochysiaceae</u> and <u>Tremandraceae</u> form his <u>Gruinales</u>. <u>Polygalaceae</u> is the only family of his <u>Polygalales</u>.

Chaudefaud and Emberger's (1960) order <u>Terebinthales</u> is essentially the <u>Rutales</u>, but includes other families also. <u>Malpighiaceae</u> was placed by Chaudefaud and Emberger in <u>Geraniales</u>. They raised <u>Cneoraceae</u> to ordinal rank as <u>Cneorales</u>.

Hallier (1912) and Copeland (1957) placed the <u>Malpiqhiaceae</u>, <u>Trigoniaceae</u>, <u>Vochysiaceae</u>, <u>Tremandraceae</u> and <u>Polygalaceae</u> in the <u>Polygalales</u>. Hallier (1912) placed the other five families: <u>Rutaceae</u>, <u>Cneoraceae</u>, <u>Simaroubaceae</u>, <u>Burseraceae</u>, and <u>Meliaceae</u> in his <u>Terebinthinae</u>. Copeland (1957) referred <u>Rutaceae</u>, <u>Cneoraceae</u>, <u>Simaroubaceae</u> and <u>Meliaceae</u> to <u>Trihilatae</u>. While Warming (1895) included <u>Rutaceae</u>, <u>Simaroubaceae</u> and <u>Meliaceae</u> in <u>Terebinthinae</u>.

Finally, van Tieghem and Constantin (1918) referred <u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Meliaceae</u> and <u>Malpighiaceae</u> to <u>Geraniales</u>; <u>Trigoniaceae</u> and <u>Tremandraceae</u> to <u>Oxalidales</u>; and Vochysiaceae to Rhamnales.

These placings are summarized in Table I. We see that there are many authors placing <u>Rutaceae</u>, <u>Cneoraceae</u>, <u>Simaroubaceae</u>, <u>Burseraceae</u> and <u>Meliaceae</u> in <u>Rutales</u> or its equivalent order

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<u>Terebinthales</u>. On the other hand, <u>Malpighiaceae</u>, <u>Trigoniaceae</u>, <u>Vochysiaceae</u>, <u>Tremandraceae</u> and <u>Polygalaceae</u> are assigned to <u>Polygalales</u>; or the <u>Malpighiaceae</u>, <u>Trigoniaceae</u> and <u>Vochysiaceae</u> are grouped in <u>Malpighiales</u>; while <u>Picrodendraceae</u> and <u>Akaniaceae</u> are put in <u>Juglandales</u> and <u>Sapindales</u>, respectively.

B. Families of the "Rutales"

1. Rutaceae

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This family was first made by Jussieu in 1789. Small (1907), Bessey (1915), Van Tieghem and Constantin (1918) and Engler and Diels (11th "Syllabus", 1936) assigned this family to the <u>Geraniales</u>. Scholz (in the 12th "Syllabus", 1964) split the <u>Geraniales</u>, and the <u>Rutaceae</u> became the type family of an order Rutales.

This family has been placed in the <u>Rutales</u> by many other authors (Lindley, 1853; Rendle, 1938, 1952; Gundersen, 1950; Pulle, 1952; Boivin, 1956; Benson, 1957; Hutchinson, 1969, and Takhtajan, 1969), while Cronquist (1968) included this family in the <u>Sapindales</u>. However, several authors (Soó, 1957; Emberger in Chadefaud and Emberger, 1960; Skottsberg, 1940; Wettstein, 1935) placed this family in the <u>Terebinthales</u>. It has also been made a member of <u>Trihilatae</u> (Copeland, 1957) and Terebinthinae (Hallier, 1912; Warming, 1895).

This family is divided into seven subfamilies: <u>Rutoideae</u>, <u>Dictyolomatoideae</u>, <u>Flindersioideae</u>, <u>Spathelioideae</u>, <u>Toddalioideae</u>, <u>Citroideae</u>, and <u>Rhabdodendroideae</u> by Scholz.

Metcalfe and Chalk (1950) stated that the wood anatomy is very uniform and highly-specialized in the whole group.

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The placing of the genera Chloroxylon and Flindersia of Scholz's subfamily Flindersioideae, has proved difficult for botanists (Kribs, 1930). According to Solereder (1908), these two genera have both Rutaceous characters, i.e. secretory cavities in the tissue of the leaf and cortex, and Meliaceous characters, i.e. the secretory cells in the cortical tissue of the axes. Bentham and Hooker (1862) have placed Chloroxylon and Flindersia with the Meliaceae, whereas placement in the Rutaceae was favoured by others (Engler and Prantl, 1897-1964; and Kribs, 1930. The latter view is supported by pollen morphology (Erdtman, 1952). Metcalfe and Chalk (1950) mention that Flindersia should remain with Rutaceae, while Chloroxylon should go to Meliaceae. Because of the homogeneous rays and the non-septate fibres, Harrar (1937) suggested that a separate family, the Flindersiaceae, should be formed. Dadswell (1935) agrees.

Bentham and Hooker took the genus <u>Zanthoxylum</u> to include the subgenus <u>Fagara</u>. The problem of whether or not to consider <u>Fagara L.</u> (with a biseriate, differentiated perianth) as distinct from <u>Zanthoxylum</u> L. sensu stricto (with a uniseriate, undifferentiated perianth) has recently been reviewed and discussed by Brizicky (1962) who concluded that ...

"the 'simple' perianth of Zanthoxylum is most likely a secondary condition, derived by reduction from that of the Fagara type by abortion of some or all the sepals"

and

"The occurrence of species of Zanthoxylum which appear in their perianth structure to be transitional to Fagara not only supports this view (of reduction), but also is ample reason to regard Fagara as a subgenus of Zanthoxylum".

Hartley (1966) agreed with these conclusion, except in the formal recognition of Fagara as a subgenus.

The only genus, <u>Rhabdodendron</u>, of the subfamily <u>Rhabdodendroideae</u> is also a problem. Heimsch (1942), on the basis of wood anatomy, concluded that <u>Rhabdodendron</u> does not belong in this family. It has been placed in <u>Rubiaceae</u> by Willis (1960), and in <u>Phytolaccaceae</u> by Record and Hess (1943). Dadswell (quoted by Price, 1963), however, considered it is possibly a mixed genus with species belonging to each of the three families. Recently, Prance (1968) states that <u>Rhabdodendron</u> is shown to differ from <u>Rutaceae</u> in nearly all important features of floral morphology, leaf and stem anatomy and pollen grain structure, and he writes:

"In these respects, it is closely related to Phytolaccaceae and other families of Centrospermae but sufficiently distinct from them to justify the description of a new family to accommodate it".

2. <u>Cneoraceae</u>

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<u>Cneorum</u> L. is the typical genus of this family which many authors have placed in the <u>Rutales</u> (Gundersen, 1950; Pulle, 1952; Benson, 1957; Engler and Melchior, 1964 and Takhtajan, 1969). On the other hand, other systematists have made this family a member of the <u>Geraniales</u> (Bessey, 1915; Engler and Diels, 1936; Soó, 1953). Engler (1931) stated that the carpels are somewhat like those of the <u>Zygophyllaceae</u>, but the single

stamen whorl, the absence of stipules and the presence of oilcells make the genus somewhat distinct from the other members of the <u>Geraniales</u>.

Cronquist (1968) placed this taxon in the <u>Sapindales</u>; Hutchinson (1959) and Boivin (1956) in the <u>Celastrales</u>. Wettstein (1935) had the <u>Cneoraceae</u> doubtfully at the end of the <u>Gruinales</u>. Bentham and Hooker (1862-1883) included it in the <u>Simaroubaceae</u>, while Hallier (1912) placed it near <u>Simaroubaceae</u> and Toddalineae in the order Terebinthinae.

Although Emberger (in Chadefaud and Emberger, 1960) stated that the nucleated albumen, the trinucleated pollen and the ovary are like those of the <u>Geraniales</u>, he made this family the only member of an order Cneorales.

Erdtman (1952) suggested that there is possibly some resemblance between the pollen grains of <u>Cneorum</u> and those of Rutaceae and related families.

3. Simaroubaceae

<u>Simarouba</u> and the other members of this family were included in the <u>Rutales</u> by Scholz, near <u>Cneoraceae</u> and <u>Picrodendraceae</u>.

Many other authors have also assigned this family to the <u>Rutales</u>, amongst them Lindley (1853), Gundersen (1950), Pulle (1952, Rendle (1952), Boivin (1956), Benson (1957), Hutchinson (1956) and Takhtajan (1969). Engler and Diels (1936), however, had this family in the Geraniales as did some other

authors (Small, 1907; Bessey, 1915). Cronquist (1968) included it in the <u>Sapindales</u>, along with <u>Akaniaceae</u>, <u>Burser</u>-<u>aceae</u>, <u>Cneoraceae</u>, <u>Rutaceae</u> and <u>Meliaceae</u>.

This family has also been assigned to their equivalent of the <u>Rutales</u>, that is, the <u>Terebinthales</u> by Wettstein (1935), Skottsberg (1940), Soo (1953) and Emberger (in Chadefaud and Emberger, 1960); the <u>Terebinthinae</u> by Warming (1895), and Hallier (1912); and the <u>Trihilatae</u> (Copeland, 1957).

Emberger stated that the simple leaves of this family have the same phylogenetic origin as those of the <u>Aurantioideae</u> of the <u>Rutaceae</u>.

Scholz divides this family into six subfamilies: <u>Surianoideae</u>, <u>Simarouboideae</u>, <u>Kirkioideae</u>, <u>Irvingioideae</u>, <u>Picramnioideae</u> and <u>Alvaradoideae</u>. Metcalfe and Chalk (1950) stated that there were few anatomical characters common to the group. Jadin (1901), as a result of his anatomical studies of stems, petioles and leaves, excluded the genera <u>Suriana</u> and <u>Holacantha</u> and assigned them respectively to the monotypic families <u>Surianaceae</u> and <u>Holacanthaceae</u>. Small (1907), too, formed a family <u>Surianaceae</u> which he placed near <u>Rutaceae</u> and <u>Simaroubaceae</u> in the <u>Geraniales</u>. Solereder (in Loesener and Solereder 1905) returned <u>Suriana</u> to the <u>Simaroubaceae</u>, recognized the genus <u>Rigiostachys</u> as belonging to it, recommended the re-establishment of the genus <u>Guilfoylia</u> (which had been combined in Cadellia), and included these four genera in the subfamily

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<u>Surianoideae.</u> Webber (1936) in her systematic anatomy of the woods of <u>Simaroubaceae</u>, supported Solereder's abolition of the monotypic family <u>Surianaceae</u>, his close grouping of <u>Suriana</u> and <u>Cadellia</u>, and his withdrawal of <u>Guilfoylia</u> from <u>Cadellia</u>. She also claimed that <u>Holacantha</u> and <u>Castela</u> differed markedly from the other genera, but that the differences do not support Jadin's erection of the monotypic family <u>Holacanthaceae</u>, since the woods of both <u>Castela</u> and <u>Holacantha</u> differ from those of other <u>Simarouboideae</u> in the same respects.

Hallier (1908) proposed that the <u>Irvingiaceae</u> and the genera <u>Picrodendron</u>, <u>Picramnia</u> and <u>Alvaradoa</u> be excluded from the <u>Simaroubaceae</u>. Whereas Webber (1936), from the standpoint of wood anatomy, wrote:

"the distinct type of wood structure characterizing the Kirkioideae, Irvingioideae, Picramnioideae and Alvaradoideae indicated that each of these subfamilies is a natural group. Whether these groups should be ranked as subfamilies of the Simaroubaceae, as distinct families, or as components of other families can be determined only after a consideration of all their characteristics. It is, however, of significance that the exclusion of the Irvingioideae, Picramioideae and Alvaradoideae from the Simaroubaceae has been proposed because of distinctive morphological characteristics other than wood structure".

In the meanwhile, she stated that the <u>Surianoideae</u> showed some diversity, and that the <u>Simarouboideae</u> exhibit rather wide variation, and suggested that the genus <u>Picrodendron</u> be assigned to a new monotypic family <u>Picrodendraceae</u>, since the woodstructure of <u>Picrodendron</u> bears a strong resemblance to that of the <u>Irvingioideae</u>.

In 1923, Hallier reviewed the early taxonomic history of the <u>Irvingiaceae</u> and included this group in his account of the <u>Linaceae</u>. Hutchinson (1959) formed a family <u>Irvingiaceae</u> which he placed in his <u>Malpighiales</u>, near <u>Linaceae</u> and <u>Huaceae</u>. Van Tieghem and Constantin, as early as 1918, assigned a family <u>Irvingiaceae</u> to the <u>Geraniales</u>, near <u>Simaroubaceae</u> and <u>Leguminosae</u>.

4. Picrodendraceae

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The affinities of Picrodendron are still obscure. It was placed in the Geraniales as part of Simaroubaceae by Engler and Diels (Syll. 11, 1936). In the 12th Syllabus, it was raised to familial rank and was assigned to the Rutales, near Simaroubaceae. Hallier (1923) suggested that Picrodendron be referred to the Bombacaceae rather than the Simaroubaceae. Engler (1931) considered that the carpel morphology of this genus was sufficiently distinctive to outweigh the anatomical evidence presented by Boas (1913) to show that Picrodendron should be included in the Irvingioideae; whereas, Webber (1936), according to the structure of wood, suggested that the genus Picrodendron should be assigned to a new monotypic family Picrodendraceae. Record and Hess (1943) had a family Picrodendraceae too. Hutchinson (1959) placed this family near Juglandaceae, in the Juglandales, and Cronquist (1968) stated that Picrodendron seems best associated with the Juglandaceae.

5. Burseraceae

Scholz, in splitting the <u>Geraniales</u> put <u>Burseraceae</u> in his <u>Rutales</u>.

This family has also been placed in the <u>Rutales</u> by many other authors (Rendle, 1938, 1952; Gundersen, 1950; Pulle, 1952; Benson, 1957; Hutchinson, 1959, and Takhtajan, 1969); and it has been assigned to their equivalent of the <u>Rutales</u>, the <u>Terebinthales</u>, by Wettstein (1935), Soó (1953) and Emberger (1960); and Terebinthinae (Hallier, 1912).

Small (1907) and Bessey (1915) placed this family between <u>Simaroubaceae</u> and <u>Meliaceae</u> in the <u>Geraniales</u>, while Cronquist (1968) assigned it to the <u>Sapindales</u>, along with <u>Akaniaceae</u>, <u>Burseraceae</u>, <u>Simaroubaceae</u>, <u>Cneoraceae</u>, <u>Rutaceae</u> and <u>Meliaceae</u>.

Guillaumin (1909-1910) stated that the most marked affinities of Burseraceae are with <u>Anacardiaceae</u>, <u>Meliaceae</u>, <u>Rutaceae</u> and <u>Simaroubaceae</u>. Webber (1941) and Heimsch (1942), on the basis of wood structure, also concluded that <u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Meliaceae</u>, <u>Sapindaceae</u>, <u>Burseraceae</u> and <u>Anacardiaceae</u> form a natural group. Heimsch (1942), in his study of the wood anatomy of 1000 species in 37 families including <u>Burseraceae</u>, <u>Meliaceae</u>, <u>Sapindaceae</u>, <u>Rutaceae</u>, <u>Simaroubaceae</u> and <u>Anacardiaceae</u>, <u>concluded that these families are better classified</u> by Wettstein, Hutchinson, and especially Hallier.

Emberger (in Chadefaud and Emberger, 1960) placed this family in the Terebinthales, with affinities with <u>Rutaceae</u> and

<u>Simaroubaceae</u>, but distinguished by certain anatomical feature, such as the secretory schizogenous cortical canals, and the radical or fundamental phloem which is sometimes more medullary or pithy.

Erdtman (1952) stated that the pollen grains in <u>Burseraceae</u> are more like those of certain genera of the <u>Simaroubaceae</u> than those of Rutaceae and Meliaceae.

6. Meliaceae

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Engler and Diels (1936) placed this family in the <u>Geraniales</u> as did Small (1907), Bessey (1915) and van Tieghem and Constantin (1918). In the 12th "Syllabus" it is in the <u>Rutales</u>, near <u>Burseraceae</u> and <u>Akaniaceae</u>.

Many other authors have also assigned this family to the <u>Rutales</u> (Lindley, 1853; Pulle, 1952; Gundersen, 1950; Rendle, 1938, 1952; Benson, 1957). Cronquist (1957; 1968) included it in the <u>Sapindales</u>, along with <u>Akaniaceae</u>, <u>Burseraceae</u>, <u>Simaroubaceae</u>, <u>Cneoraceae</u>, and <u>Rutaceae</u>. Takhtajan (1954) also assigned this family to the <u>Sapindales</u>, but later, in 1959 and 1969, he referred it to <u>Rutales</u> which he segregated from the <u>Sapindales</u>. Heimsch (1942) stated that <u>Meliaceae</u>, <u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Sapindaceae</u>, <u>Burseraceae</u> and <u>Anacardiaceae</u> form a more or less natural group.

Hutchinson (1969) and Boivin (1956) made this family the only member of the <u>Meliales</u>. It has also been assigned to the <u>Terebinthales</u> (Soo, 1953; Emberger in Chadefaud and Emberger, 1960; Skottsberg, 1940; Wettstein, 1935), and <u>Terebinthinae</u>

(Hallier, 1912; Warming, 1895). These are essentially rutaceous orders. Copeland (1957) placed this taxon in the order <u>Trihilatae</u>.

The genus <u>Ptaeroxylon</u> was placed in the <u>Sapindaceae</u> by Bentham and Hooker (1862); while Harvey and Sonder (1860) favoured a separate small subfamily, the <u>Ptaeroxyleae</u>, in the <u>Meliaceae</u>. Radlkofer placed this genus in the <u>Meliaceae</u> because of the presence of secretory cells in the leaves. Kribs (1930), on the other hand, having examined the wood of this monotypic genus, suggested that it more closely resembles the <u>Rutaceae</u>. Recently, it was placed with <u>Cedrelopsis</u> in a small family, the <u>Ptaeroxylaceae</u> (Leroy, 1959) and regarded as closely related to the <u>Sapindaceae</u>.

Kribs (1930) argued that <u>Swietenioideae</u> is the only subfamily of <u>Meliaceae</u> in which the genera form a distinct homogeneous group on the basis of its anatomy and morphology and shou'd thus be raised to familial rank as <u>Swieteniaceae</u>. Erdtman (1952), says that pollen morphology, does not support Harms's suggestion that the three subfamilies <u>Cedreloideae</u>, <u>Swietenioideae</u> and <u>Melioideae</u> should rank as separate families, and he also found pollen-grains more or less similar to those in <u>Meliaceae</u> to occur in <u>Rutaceae</u> (e.g. in <u>Aegle</u>, <u>Atalantia</u>, Citrus etc.).
7. Akaniaceae

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This is a tiny family with <u>Akania hillii</u> only. Stapf (1912) first proposed this family and placed it in the <u>Sapindales</u>, in which he was followed by many authors (Gundersen, 1950; Boivin, 1956, Hutchinson, 1959; Cronquist, 1968, and Takhtajan, 1969). Scholz places this family in Rutales, and this mainly because of the ovule.

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Cronquist (1968) wrote:

"The Akaniaceae, were unique in the Sapindales in their very wide wood rays, and they are somewhat unusual in lacking a floral disk and having endosperm. Their pollen is like that of some of the Sapindaceae, and they commonly have eight stamens and a pentamerous perianth, like the Sapindaceae. Although it has sometimes been included in the Sapindaceae, <u>Akania</u> seems amply to merit status as a separate family. On the other hand, there is nothing to indicate that it would be better placed in any other order".

In fact, in the presence of the very large rays and the absence of uniseriate rays in the secondary wood, this family differs clearly from Sapindaceae.

Benson (1957) says that <u>Akaniaceae</u> and <u>Tremandraceae</u> are of uncertain position, but he believes that they are related to the complex of the <u>Geraniales</u>, <u>Rutales</u> and <u>Sapindales</u>. Emberger (in Chadefaud and Emberger, 1960), although placing this family in the <u>Terebinthales</u>, considers the affinities of this taxon to be unclear.

8. Malpighiaceae

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This family was most often placed in <u>Geraniales</u> (Small, 1907; Bessey, 1915; van Tieghem and Constantin, 1918; Rendle, 1938, 1952; Benson, 1957; Emberger, in Chadefaud and Emberger, 1960; and Takhtajan, 1969); less often in <u>Malpighiales</u> (Pulle, 1952; Boivin, 1956; Hutchinson, 1959); or <u>Polygalales</u> (Hallier, 1912; Copeland, 1957 and Cronquist, 1968); while only Scholz (1964) treated it as a family of <u>Rutales</u>, in the suborder Malpighiineae, near Trigoniaceae and Vochysiaceae.

It has also been assigned to the <u>Sapindales</u> (Gundersen, 1950), but Gundersen noted that the twisted seed of the <u>Sapindaceae</u> and other characters make <u>Malpighiaceae</u> distinct from that family.

Erdtman (1952) found the pollen grains of <u>Malpighiaceae</u> to be slightly similar to those of <u>Tremandraceae</u> and <u>Trigoniaceae</u>, but more or less different from those of <u>Vochysiaceae</u>. van Tieghem and Constantin (1918) described the <u>Malpighiaceae</u> as ".... ... une famille trés homogéne".

9. Trigoniaceae

This family was included in the <u>Vochysiaceae</u> by Bentham and Hooker (1862-1883). Scholz (in Syll. 12, 1964) has it as a distinct family in the Rutales.

Several authors (Benson, 1957; Hutchinson, 1959; Cronquist, 1968, and Takhtajan, 1969, etc.) made this a family of the Polygalales and placed it near Vochysiaceae. Pulle (1952)

assigned this family to the <u>Malpighiales</u>, again near <u>Vochysiaceae</u>. Warming (1895) assigned this family to <u>Aesculinae</u>, again near <u>Vochysiaceae</u> (and <u>Tremandraceae</u>).

Thus, as one can see, many authors assign this family to a position near <u>Vochysiaceae</u>. Metcalfe and Chalk (1950) described it as being conspicuously different from <u>Vochysiaceae</u> in the absence of intraxylary phloem and in the presence of bordered pits in the ground tissue elements of the xylem. However, Heimsch (1942) stated that the wood anatomy suggests a relationship with <u>Polygalaceae</u>, <u>Tremandraceae</u>, <u>Malpighiaceae</u>, and <u>Vochysiaceae</u>; while Erdtman (1952) says that there is some similarity between the pollen-grains of <u>Lightia licanioides</u> (Trigoniaceae) and those of some genera of the Vochysiaceae.

10. Vochysiaceae

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Scholz assigned this family to the <u>Rutales</u>, and thus placed it near <u>Malpighiaceae</u> and <u>Trigoniaceae</u>.

Many authors (Small, 1907; Benson, 1957; Hutchinson, 1959; Cronquist, 1968 and Takhtajan, 1969) have preferred to put this family in <u>Polygalales</u>. Pulle (1952) made it a member of the Malpighiales.

Heimsch (1942) said that this family is related to <u>Polygalaceae</u>, <u>Trigoniaceae</u>, <u>Tremandraceae</u>, and <u>Malpighiaceae</u>, but differs from them in the more pronounced development of banded parenchyma and the occurrence of intercellular canals. Erdtman (1952) found that certain pollen types in <u>Malpighiaceae</u> and <u>Trigoniaceae</u> (<u>Lightia</u>) have at least some characters in common with the pollen of Vochysiaceae.

11. Tremandraceae

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Scholz assigns this family to <u>Rutales</u> and has it with <u>Polygalaceae</u> as the suborder <u>Polygalineae</u>. This family has been included in the <u>Geraniales</u> (Bessey, 1915) and in the <u>Sapindales</u> (Lindley, 1853; Gundersen, 1950), but many authors place it in <u>Polygalales</u> (Pulle, 1952; Cronquist, 1968; Takhtajan, 1969; etc.).

Warming (1895) made this family part of the <u>Aesculinae</u>, and placed it between <u>Trigoniaceae</u> and <u>Polygalaceae</u>.

Metcalfe 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. According to Erdtman (1952) the pollen grains of <u>Tremandra</u> are moreor-less similar to those of <u>Galphimia</u> (<u>Malpighiaceae</u>).

12. Polygalaceae

This family has been variously placed by different systematists. Scholz (Syll. 12, 1964) has it in the <u>Rutales</u> near <u>Tremandraceae</u>. Warming (1895) also placed this family near <u>Tremandraceae</u> but in the <u>Aesculinae</u>.

Many authors have it in <u>Polygalales</u> (Small, 1907; Pulle, 1952; Benson, 1957; Hutchinson, 1959; Cronquist, 1968 and Takhtajan, 1969 etc.). Others (Lindley, 1853; Gundersen, 1950; and Rendle, 1952) have placed this family in the <u>Sapindales</u>, but Rendle calls the <u>Polygalaceae</u> a family of doubtful position.

Heimsch (1942) investigated some members of this family and found that the wood anatomy shows affinities with that of <u>Trigoniaceae</u>, <u>Tremandraceae</u>, <u>Malpighiaceae</u>, and <u>Vochysiaceae</u>.

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.

Some genera have been doubtfully placed here.

<u>Diclidanthera</u> was made the type of a family <u>Diclidanthereae</u> (=<u>Diclidantheraceae</u>) by Agardh (1858). The family has been associated with <u>Ebenales</u>, but also with <u>Polygalales</u>. Metcalfe and Chałk (1950) have it after <u>Polygalaceae</u> and before <u>Vochysiaceae</u>. From pollen-grain morphology and wood anatomy, however, it was referred to <u>Polygalaceae</u> (0'Donell, 1964).

Chodat (1891-1893) noted that <u>Krameria</u> was not a member of <u>Polygalaceae</u>, but the type of a family (<u>Krameriaceae</u>) near <u>Leguminosae-Caesalpiniaceae</u>. Le Maout and Decaisne (1873), however, assigned <u>Krameria</u> to <u>Polygalaceae</u>.

<u>Xanthophyllum</u> Roxb. was raised to familial rank as <u>Xanthophyllaceae</u> (Gagnepain, in Chadefaud and Emberger, 1960). Scholz keeps it in <u>Polygalacea</u>e.

According to Jauch (1918), <u>Xanthophyllum</u> differs in its wood parenchyma from other members of the <u>Polygalaceae</u>, but should remain in the family because of its floral anatomy and pollen-grain structure.

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C. <u>Some Useful Chemistry in Plant Taxonomy</u>

There has been a fever of intellectual activity in botanical chemosystematics during the past 10 years. Chemical characters are ideally suited to be used, together with morphological criteria, in a numerical approach to taxonomy. Many books on chemotaxonomy have appeared since 1962 (Swain, 1963, 1966; Alston and Turner, 1963; Hegnauer, 1962, 1963a, 1964, 1966a, 1969; Harborne, 1967; Harborne and Swain, 1969), Reviews have been written for both books (Davis and Heywood, 1963; Alston, 1967; Harborne, 1968) and journals (Bate-Smith and Swain, 1965; Hegnauer, 1965) and many papers have appeared in journals such as Phytochemistry and Lloydia.

Many samples of plant material can be quickly and efficiently surveyed for a particular class of compound by one or other of the many chromatographic procedures available today. <u>Terpenoids</u> are usually separated and identified by gas-liquid chromatography and mass spectrometry; <u>phenolic</u> <u>compounds</u> by paper or thin-layer chromatography and absorption spectroscopy; <u>amino acids</u> by electrophoresis and ion-exchange chromatography, and so on.

The contributions of chemosystematics to plant taxonomy will be discussed under two general headings, micromolecules and macromolecules, depending on the relative molecular weights of the compounds under consideration.

1. Micromolecules

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Most of the early chemosystematic literature, and much of the present, has concerned itself with the mere notation of the distribution of relatively simple compounds from only one or, less often, several collections of a given taxon.

In this review, it is proposed to consider the various classes of micromolecular plant constituents, to outline their present contribution to taxonomy, and to estimate their future potential in this field.

a. Phenolic Constituents

Some phenols present in plant tissue are characteristic of the species, and thus they are of taxonomic value. Analysis of coumarins, flavonoids and other phenolic compounds in the study of taxonomic relationships, hybrids, ecological differentiation, etc. in various botanical taxa, has been receiving increased attention. The popularity of using this group of compounds as a criterion is partly due to the advent of thin-layer chromatography (Dedio, <u>et al</u>, 1969) which enable one to analyze rapidly a large number of samples.

i. Coumarins

In the plant kingdom, coumarins are widely but by no means universally distributed. That <u>coumarins</u> may be useful in chemotaxonomy will be clear from Price (in Swain, 1963) who says:

"Further support for the view that the Rutaceae is a distinct and homogeneous group is provided by its essential oils and coumarins ... within the Rutaceae coumarins are distributed throughout the four subfamilies Aurantioideae, Rutoideae, Toddalioideae

and Flindersioideae ... On the other hand, though it is negative evidence, there is not one report of the isolation of a coumarin from the Meliaceae, Burseraceae, Simaroubaceae, Zygophyllaceae, or Cneoraceae"

However, <u>Cedrelopsis</u>, <u>Ptaeroxylon</u> and <u>Ekebergia</u>, three genera of the <u>Meliaceae</u> (Scholz) have been reported to contai: coumarins (Eshiett <u>et al.</u>, 1968; Dean and Taylor, 1966; Bevan <u>et al.</u>, 1965). Especially, two botanically very close genera --<u>Cedrelopsis</u> and <u>Ptaeroxylon</u> -- both contain <u>Ptaeroxylin</u> which has an unusual seven-membered ring structure not encountered elsewhere. This would seem to be good evidence in support of Airy Shaw (in Willis, 1966), who places the two genera in a little family <u>Ptaeroxylaceae</u> somewhat intermediate between Meliaceae and Rutaceae.

Ellagic acid and its derivatives are in almost all families of <u>Myrtiflorae</u>, except <u>Hippuridaceae</u> (Hegnauer, 1964, 1966). In addition, members of <u>Rhizophoraceae</u>, <u>Combretaceae</u> and <u>Myrtaceae</u> are well-known sources of ellagitannins (Bate-Smith, 1962a). Ellagic acid is also found as an occasional constituent throughout the <u>Archichlamydeae</u>. It is of rare occurrence in the <u>Sympetalae</u>; plants containing it here (e.g. some <u>Ericaceae</u>) are those with close affinities with the <u>Archichlamydeae</u> (Bate-Smith, 1962b). Bate-Smith (1968) says it is absent from <u>Monocotyledons</u>. Its close association in occurrence with "woodiness" in plants indicate that it is more of phylogenetic rather than of practical interest to the systematist (Harborne, 1968).

Another aspect of coumarins in plants which shows promise of being of systematic interest is the study of their metabolism. Harborne and Corner (1961) and Harborne (1964) have noted differences in metabolic pathways of cinnamic acids and coumarins. <u>Datura</u>, for example, was the only genus among some dozen genera of the <u>Solanaceae</u> studied with the ability to convert caffeic acid into the coumarin scopolin. Again, while the more primitive angiosperms convert 6,7-dihydroxycoumarin (aesculetin) into the 6-glucoside, the more advanced plants (e.g. the <u>Compositae</u>, <u>Solanaceae</u>, etc.) change it into a mixture of 6- and 7-glucosides.

ii. Flavonoids

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There has recently been considerable interest in the range of plants in which flavonoids occur (Harborne, 1967). This has been due, in part, to their significance as taxonomic and evolutionary markers (Bate-Smith, 1963).

To date flavonoids have been isolated from or identified in a wide range of the lignin-containing Angiosperms, Gymnosperms and ferns (Harborne, 1967). Of the plants normally considered to be non-ligniferous, only the Mosses have so far been proven to contain flavonoids (chiefly flavone C-glycosides and anthocyanins) (Harborne, 1967; Markham and Porter, 1969a). However, several existing reports (Harborne, 1967; Reznik and Wierman, 1966) indicate that flavonoids might also occur in the Liverworts (<u>Hepaticae</u>) and this occurrence has recently been confirmed by Markham and Porter (1969b). The flavonoid data

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do not help us to establish the phylogenetic relationship between mosses and other plants although it is interesting, of course, to know that flavonoids occur among liverworts.

The flavonoid pigments of plants vary chemically according to class or according to number and position of extra skeletal substituents (e.g. hydroxyl, methoxyl, O-glycosyl, C-glycosyl, or isoprenoid residues). Lebreton (1964) set forth some rules by which one may presumably judge primitive versus advanced features of flavonoids. Saturation of the heterocyclic ring (as in catechins, leucoanthocyanins, flavanones, dihydrochalcones and flavononols) is regarded as a primitive character. Thus, anthocyanins, flavones and flavonols are more advanced than are the leucoanthocyanins. Hydroxylation of the B-ring is primitive; thus the evolved types are characterized by lack of B-ring hydroxyls or by methylation or glycosylation. Alston (1968), in discussing the C-glycosyl flavonoids, wrote:

"Indeed, if the implications of this fragmentary chemical data were accepted at face value, we might reconsider the question of which types of flavonoids are 'primitive' as opposed to 'advanced'. Although this question of chemical primitiveness among flavonoids has not apparently been considered comprehensively on biogenetic grounds, it seems to be generally regarded that anthocyanidins such as cyanidin, and O-glycosides of flavonoids are representatives of more primitive flavonoids. Primitive, as used here, signifies that such compounds may have appeared early among the flavonoids following the appearance of the characteristic C15 structural unit".

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These criteria may be valid if applied as broad generalizations, although it is doubtful that they have any significance at all when applied as specific criteria in individual situations.

To the taxonomist, variation in flavonoid class appears to be of most significance, since there is good evidence that some classes act as "replacement" characters. For instance, the <u>betacyanin/anthocyanin</u> criterion has been useful in supporting the inclusion of the <u>Cactaceae</u> and the <u>Didiereaceae</u> in the order <u>Centrospermae</u>, and for indicating that the affinities of the <u>Caryophyllaceae</u> (and <u>Molluginaceae</u>?), the only families to retain anthocyanin pigmentation, should be re-examined.

Flavones and flavonols do not often co-occur in the leaves of higher plants. Flavones instead tend to replace flavonols, a change which is correlated with evolutionary advancement (Bate-Smith, 1962b). Harborne (1967), for example, has shown that the occurrence of flavones versus flavonols is correlated with tribal division in the main subfamily of <u>Umbelliferae</u>, the <u>Apioideae</u>. The eight tribes can be divided into three groups according to whether the species have only flavonols, mainly flavonols or mainly flavones. Again, Crowden (1969), in a study of 52 per cent of the genera of <u>Umbelliferae</u> discovered that nearly all species have either flavonols <u>or</u> flavones, but not both. Furthermore, he also found that flavones were almost entirely in taxa generally considered to be more specialized or advanced (e.g. <u>Daucus</u>,

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<u>Torilis</u>), while flavonols predominated in the less advanced genera (e.g. <u>Hydrocotyle</u>). Thus, the replacement of flavonol by flavone appears to have an evolutionary significance with the family, as it probably has among the angiosperms generally. (Harborne, 1967; Bate-Smith, 1962b).

From the taxonomic viewpoint, a difficulty is the disconcerting habit of some types of flavonoids which had seemed to characterize a certain group appearing sporadically in quite unrelated taxa. Thus the value of <u>biflavonyls</u> for defining the Gymnosperms in chemical terms is lessened by the recent discovery of their presence in two widely different Angiosperms (<u>Casuarina</u> and <u>Viburnum</u>); in two lower plants (<u>Selaginella</u> and <u>Psilotum</u>); and their absence from the <u>Pinaceae</u> (one of the largest families of gymnosperms).

A similar situation exists in the case of <u>isoflavones</u>. It is well known that they occur quite characteristically in one subfamily of the <u>Leguminosae</u>, the <u>Faboideae</u>, and are rarely found elsewhere. But recently they have been found also in related (Rosaceae) and unrelated (Iridaceae) taxa.

The inheritance of flavonoids in the leaves of plants is usually straight-forward in that hybrids normally contain all or most of the constituents of the two parents. For example, in <u>Baptisia</u> the flavonoid patterns of hybrids were identical with those obtained by superimposing the pattern of one species on the other (Alston and Turner, 1962).

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iii. Other Simple Phenols

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There is a vast number of papers published on the phenolic acid contents of plants in which the presence or absence of certain hydroxycinnamic acids or even, sometimes, of a particular acid has been considered to be of taxonomic value. For instance, gentisic acid appears to be associated with woody habits and is perhaps associated with lignification or some associated process (Griffiths, 1959). In contrast, syringic acid was found in 35 per cent of the species investigated by Ibrahim (1961) and was more frequently found in Monocotyledons. More recently, Harborne (1968) chose the distribution of seven of the many systematically interesting simple phenols to illustrate the point that phenolic substances are useful at all levels or hierarchies of classification. However, since the assumption that phenolic acids are metabolically inert (Bate-Smith, 1958) is no longer valid, before any taxonomic use can be made, more studies are needed to determine to what extent genetical factors and environmental factors are responsible for the types and amounts of the phenolic acids accumulating in plants.

b. <u>Alkaloids</u>

Alkaloids are of very many different kinds and not a chemically natural group. Higher plants are the chief source of alkaloids, yet alkaloids are also known from club mosses (<u>Lycopodium spp.</u>), horsetails (<u>Equisetum spp.</u>), and fungi (Robinson, 1968). Although between two and three thousand

different alkaloids have been isolated from plants, some are taxonomically quite restricted in occurrence and others are found widely in unrelated plants. A statistical analysis of 3600 alkaloid plants by Willaman and Li (1963) showed caffeine occurring in the largest number of families (14), lycorine in the largest number of genera(30), and berberine in the largest number of species (89).

Henry, as early as 1949, reviews a number of instances where alkaloids were used to contribute to taxonomic problems, and Manske (1949) considered them in relation to the whole phylogenetic scheme of the angiosperms. Some authors are of the opinion that generalizations as to their phylogenetic significance may be of little value (Alston and Turner, 1963; Hegnauer, 1962, 1963b) or that "there does not appear to be any facile generalization to be made about alkaloid distribution" (Robinson, 1963). However, alkaloids are frequently treated as a related group of substances for both theoretical and practical purpose, and studies on their general patterns of distribution and occurrence have been attempted in the past (Manske, 1949; McNair, 1941, 1945; Willaman and Li, 1963). Their value in interrelating families is limited because of parallelism and convergence (Hegnauer, 1966b). Nevertheless, they have been shown to be of systemic interest in several families, particularly in the Amaryllidaceae (Hegnauer, 1963b), Liliaceae (Hegnauer, 1963b), Papaveraceae (Hegnauer, 1966b) and Rutaceae (Price, in Swain, 1963).

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In recent reviews by Alston and Turner (1963), Hegnauer (1963b), and Stermitz (1968), on the use of alkaloid content in biochemical systematics or chemotaxonomy, considerable emphasis was placed on interfamily and intergeneric relationships. Rather few alkaloid surveys have been carried out at the species level but Kuck <u>et al.</u> (1967) worked on the bark alkaloids of 7 Argentinian species of Fagara.

Li and Willaman (1968), in studying 7,740 alkaloid plants, suggested that presence or absence of alkaloids (as well as their relative abundance in the families and orders) may have both taxonomic and phylogenetic significance. Within the dicotyledons, there is a high incidence of alkaloid occurrence in the morphologically primitive Magnoliales-Ranales complex and related groups, a lower incidence in various phylogenetically intermediate groups, and a progression of high incidence in all of the phylogenetically advanced but unrelated groups. Notably, the generally wind-pollinated "Amentiferous" families, as well as the wholly aquatic families, are either alkaloid-free or show very low incidence. High incidence of alkaloid occurrence appears as a general character for some families or orders. In Ranunculaceae, Berberidaceae, Menispermaceae, Piperaceae, Cactaceae, Papaveraceae, and Gentianaceae, alkaloids occur in over 90% of the species tested. On the other hand, in Fagaceae, Betulaceae, Casuarinaceae, and Juglandaceae the alkaloid

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occurrence is almost zero. Thus, the presence or absence of alkaloids can be treated as a general family characteristic. At the ordinal level, high or low incidence may appear uniformly in all component families of a single order or sometimes pronouncedly in one of the component families.

McNair (1941) digressed from taxa and considered the size of alkaloids as it might be reflected in the habitat of the families containing them. For example, he deduced that the average molecular weight of alkaloids was greater in temperate than in tropical families; that the average number of nitrogen and carbon atoms was the same in the two groups, but that the number of oxygen atoms was greater in the temperate.

Alkaloids biogenesis and distribution even now are of considerable use, but as Hegnauer points out:

".... because of this structural diversity, the biosynthetic origin of an alkaloid should be known before reliance is placed on its use as a taxonomic marker"

and Gibbs (MSS) says:

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"When we do know enough of the biogenesis of alkaloids it will be possible to group them more naturally and to use the distribution of the groups as an important taxonomic character".

c. Terpenoids

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This name has been applied to a group of compounds distinguished by a singular chemical composition. Chemically, terpenoids all have carbon skeletons based on two or more isoprene (C_5) units. Thus there are monoterpenes (C_{10}), sesquiterpenes (C_{15}), diterpenes (C_{20}), triterpenes (C_{30}), tetraterpenes (C_{40}) and polyterpenes.

There is probably no living organism which does not contain at least one isoprenoid compound -- such as carotenoids, phytol or steroids -- belonging, respectively, to the tetra-, di- and mixed terpene groups. The occurrence of mono-, sesqui- and polyterpenes seems to be confined to the plant world, and there only to a limited number of species.

The distribution of terpenoids with respect to taxonomic classification can be found in Klein's <u>Handbuch der Pflanzen-analyse</u> (1932), in Guenther's <u>Essential</u> Oils (1948-1952) and in several recent reviews (Ponsinet <u>et al</u>., 1968; Harborne, 1968; Weissmann, 1966).

As early as in 1920, Baker and Smith published the chemotaxonomic investigations of the essential oils or terpenes of <u>Eucalyptus</u> species. In recent years, gas chromatographic separation of terpenes has proved valuable in taxonomic studies. Mirov (1948) studied the terpenes of <u>Pinus</u>, Von Rudloff (1966) the essential oils of <u>Picea</u>, and chromatographic analysis for chemotaxonomic purposes relating to the genus <u>Abies</u> were also

reported by Zavarin and Snajberk (1965). Other workers in the field have tried to find genetic relationships based upon the chemical composition of the essential oils (Haagen-Smit, 1953).

The contributions of these various terpenoid classes to taxonomy will be briefly considered.

i. Monoterpenes

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These are most abundant in the <u>Pinaceae</u>, <u>Labiatae</u>, <u>Umbelliferae</u>, <u>Rutaceae</u>, <u>Lauraceae</u>, <u>Myrtaceae</u>, and <u>Compositae</u>, but occur in at least 50 other families. The most extensively studied group at the present time are the members of the <u>Pinaceae</u>. (Mirov <u>et al</u>., 1966; Von Rudloff, 1966; Zavarin & Snajberk, 1965).

The exact nature of these variants is still in some doubt although some writers, e.g. Flück (1963), have suggested that the variation may be purely quantitative and not qualitative. Recently, Hellyer and coworkers (1969), in an examination of the oils from four "physiological forms" (chemical varieties) of <u>Eucalyptus dives</u>, established that the oils of <u>Eucalyptus dives</u> "type" all contain the same constituents, but in markedly varying proportions. These results tend to support Flück's hypothesis and indicates that monoterpene variation known in other <u>Eucalyptus</u> may be of the same type.

Harborne (1968) writes:

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"Perhaps the most striking correlation between terpene distribution and taxonomy has been found in a group quite unrelated to the Pines, in the angiosperm genus Hypericum (Guttiferae). In a survey of over 35 species, Mathis and Ourisson (51, 52a) found that the eight volatile constituents present varied both qualitatively and quantitatively. The result confirmed the existing classification at the sectional level. Species fell clearly into groups, those rich in limonene (12) and myrcene (13), and those poor in these substances (Table Interestingly enough, the distribution of mono-3). terpene alcohol, sesquiterpenes, and quinones also fitted in with these results".

Monoterpenes are also proving of value in taxonomic studies of plants of the family <u>Rutaceae</u>. Recent studies of the essential oils of the 12 genera in the <u>Aurantioideae</u> (Scora <u>et al.</u>, 1969) indicate that the essential oils conform to the botanical groupings, except for <u>Pleiospermium</u> in the <u>Citrinae</u> and <u>Murraya</u> in the Clauseneae.

ii. Sesquiterpenes

The sesquiterpenes are the class of terpenoids with 15 or fewer carbon atoms, the members of which originate from farnesyl pyrophosphate. The hydrocarbons $(C_{15}H_{24}-C_{15}H_{18})$, related <u>alochols</u>, <u>ketones</u> and a few other types have been commonly isolated from essential oils.

Bisset and Cowokers (1966), in analysing seventy-eight samples of resin which belong to 42 different species of the genus <u>Dipterocarpus</u> found that the <u>sesquiterpenes</u> are very variable, and defined six groups in the genus on the basis of the composition of the sesquiterpene fraction of their resin.

Sesquiterpene lactones may be useful for chemotaxonomic markers as will be clear from the following:

Novotný et al., (1966) say:

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"The occurrence of sesquiterpenoid lactones (compounds of the santonine, guaianolide, ambrosanolide, germacranolide and eremophilanolide type) may be taken for a new chemotaxonomic character"

Steelink and Spitzer (1966), in a paper on sesquiterpene lactones in chemotaxonomy, say:

"The guaianolides, a class of sesquiterpene lactones with the guaiane skeleton (I) appear to possess potential application in the chemotaxonomy of higher plants".

In fact, a number of workers (Steelink and Spitzer, 1966; Herz, 1968) have suggested that the distribution of sesquiterpene lactones, C₁₅ compounds which occur frequently in plants belonging to the <u>Compositae</u> tribes <u>Heliantheae</u>, <u>Ambrosieae</u>, <u>Anthem-</u> <u>ideae</u>, and <u>Helenieae</u>, might prove useful in understanding the evolutionary relationships among such genera as <u>Ambrosia</u>, <u>Parthenium</u>, Iva, and others.

The variation of chemical constitution within a plant species is commonly encountered. Perhaps the most striking examples of the diversity between morphological characterization and chemical constitution are found in the genus <u>Ambrosia</u> (fam. <u>Compositae</u>, trib <u>Ambrosiaceae</u>). The distribution of sesquiterpene lactones in <u>Ambrosia</u> species has been reviewed by Herz, 1968); its possible utility for clarifying evolutionary relationships has been discussed by Miller <u>et al.</u>, 1968); while Geissman and co-workers (1969), in examining and isolating sesquiterpene lactones of three distinct populations of <u>Ambrosia psilostachya</u> and four species of <u>Ambrosia</u> <u>acanthicarpa</u> (two of them being seedling and mature plants from a single location), found that different populations vary markedly in lactone content, and the seedling and mature forms show a wide qualitative difference.

Novotny <u>et al</u>., (1966), in a systematic study of the chemical components in representatives of the <u>Compositae</u>, revealed the occurrence of sesquiterpenoid substances of the less usual <u>eremophilane type</u> and found that these compounds can also be used as an important distinguishing character of the taxa of <u>Petasites</u>. They also noted that eremophilanolides have been found in two other genera -- <u>Ligularia</u> and <u>Euryops</u> -- of the <u>Senecioneae</u> and suggested that this shows the possibility of putting all three genera -- <u>Ligularia</u>, <u>Euryops</u>, and <u>Petasites</u> -into the one tribe <u>Senecioneae</u> which can be distinguished chemically from other tribes of <u>Compositae</u> as mentioned above.

iii. Diterpenoids

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Erdtman (1956, p. 453) has already pointed out the possible taxonomic importance of diterpenoids if it becomes feasible to distinguish specific patterns of their distribution in the different genera and sub-genera. In view of the difficulties involved in the isolation and separation of the diterpene hydrocarbons by classical methods, the gas-liquid

chromatographic (GLC) technique appeared to offer the most satisfactory method for investigation of the problem (Eglinton <u>et al.</u>, 1962a).

Aplin and co-workers (1963), using the GLC method, have applied their results to clarify anomalies connected with the occurrence and to determine the taxonomic value of the diterpene hydrocarbon content of twenty-eight species of the <u>Podocarpaceae</u> and nine related gymnosperms. They quote the example of <u>Podocarpus spicatus</u> to show that variation of the diterpene constituents could occur regardless of the geographical location, and suggest that the diterpenes of <u>Podocarpaceae</u> are of little value for taxonomy.

iv. Triterpenoids

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The common structural characteristic of triterpenes has been recognized as proceeding from squalene.

The genetic origin of the triterpenes may be explained by the action of specific enzymes that impose on the acyclic precursor a particular conformation that, upon cyclization, yields the observed basic carbon skeletons. Most triterpenes arise from further reactions of these skeletons such as rearrangement, degradation, oxidation and reduction.

Many genera of <u>Cucurbitaceae</u> contain bitter principles derived only from Cucurbitacin B, or only from Cucurbitacin E, but Rehm (1960) reported that both series are represented in <u>Cucurbita</u>. Ponsinet <u>et al</u>. (1968) considered that more primitive

species contain only <u>Cucurbitacin</u> B, whereas the more highly evolved species contain B and E, or only E. They said that

"derivatives of the related euphane and elemane skeletons are of great taxonomic interest since they are found in certain families considered to be closely related from a morphological point of view (Table 9)(...). Morover, among these substances there are a large number of bitter principles that possess great homogeneity in respect to degradation and oxidation patterns (Table 10)".

Examples of restricted distribution at sub-family, genus, or species level are discussed below. Dreyer (1966) found triterpenoids of interest at the sub-family level when examining plants of <u>Rutaceae</u> for limonoids. He detected limonin or related structures in 26 Citrus species, in 3 related genera, <u>Poncirus, Microcitrus</u> and <u>Fortunella</u> (all <u>Aurantioideae</u>), and in 6 genera of two other sub-families, <u>Toddalioideae</u> and <u>Rutoideae</u>. By contrast, they have not been reported in the four other sub-families, members of which are morphologically distinct from those mentioned above.

Steroidal sapogenins appear to be of interest at the genus level. Akahori (1965) analysed the steroidal sapogenins of 12 Japanese <u>Dioscorea</u> spp. and considered that the chemical composition of the sapogenins they contain corresponds to their morphological features.

At the species level, Ponsinet and Ourisson (see Ponsinet <u>et al.</u>, 1968), in studying the latex of more than 70 species of <u>Euphorbia</u>, found that various morphological types of Euphorbias produce different tetracyclic triterpenes, i.e.

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- 1) Herbaceous Euphorbias all contain cycloartenol (IV).
- Cactus-like Euphorbias contain euphol (V) and euphorbol (VI).
- Coral-like Euphorbias contain euphol and tirucallol (VII).

d. Alkanes

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Despite an early adverse prognostication by one of the pioneers in the field of chemotaxonomy (Erdtman, 1956), the universal presence of normal alkanes as constituents of leaf cuticular-waxes is well established and both their biogenesis and their value as a taxonomic criterion have been extensively discussed (Eglinton and Hamilton, 1967).

Herbin and Robins (1968b) have shown, in a survey of a large number of leaf cuticular waxes from a range of families in the Angiosperms, that in the homologous series of n-alkanes present, nonacosane (C_{29}) and hentriacontane (C_{31}) are the most frequent major components among the predominating odd carbon number constituents and the C_{28} and C_{30} are the most frequent major components among the, usually, less significant even carbon number constituents.

Castillo <u>et al.</u> (1967), in analysing the alkanes of thirty-two <u>Podocarpaceae</u> and other related species, considered that

> "the alkane constituents would appear to be a better guide to the botanical classification though there are a number of exceptions. Thus, of the twenty-one Podocarpus species only three cannot be grouped with

the others by their alkane distribution. In a number of species, it was possible to note that both alkane and diterpene contents were unusual, for example, <u>Libocedrus plumosa</u> and <u>Podocarpus lat-</u> <u>ifolius</u>. It would again appear that the alkane distribution in plant waxes would not be sufficiently diagnostic by itself."

The constituents of plant cuticular waxes have been shown by Eglinton and co-workers (1962a) to have significance in the study of interrelationships within a group of closely related genera in the sub-family <u>Sempervivoideae</u> of the <u>Crassulaceae</u>. In Eglinton's study gas-liquid chromatographic analysis of the alkane fraction of leaf cuticular waxes gave distribution patterns of normal and branched chain alkanes which could be correlated with the accepted taxonomy based on morphological characters.

In a further study of a number of New Zealand species drawn from different families, Eglinton and co-workers (1962b) found that in four species of the genus <u>Hebe</u> (<u>Scrophulariaceae</u>) a much wider variation in alkane pattern existed within a single genus than had been found by the earlier work within a sub-family. This variation within a genus indicates that, while alkane distribution patterns may be valid criteria for distinguishing related plants, difficulties might be encountered in any attempt to correlate less closely related groupings.

Herbin and Robins (1968a) confirmed, in studying the alkanes of cuticular waxes from 6 species in the genus <u>Aloe</u> (<u>Liliaceae</u>), the species specificity in composition. They subsequently examined (1968b), on the basis of large numbers of

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leaf wax alkanes from many orders, Hutchinson's sub-division of the Angiosperms into '<u>Herbaceae</u>' and '<u>Lignosae</u>'. Their results showed that, with the limited samples employed, there is no apparent distinction between the '<u>Lignosae</u> and the '<u>Herbaceae</u>' on the basis of leaf alkane pattern.

e. Sulphur Compounds

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The secondary sulfur compounds of plants have been considered in general articles by Kjaer (1963, 1966) and Ettlinger and Kjaer (1968). Harborne (1968), in his review of Biochemical Systematics, says:

> "of the various groups of natural products containing the element sulphur, only two, thioglucosides and sulphides, are at present of taxonomic interest".

Thioglucosides appear to be of interest at the order level, i.e. Kjaer (1963) has reported them in 300 of the 1500 species of the <u>Cruciferae</u> and in all species examined of the <u>Capparidaceae</u>, <u>Resedaceae</u>, and <u>Moringaceae</u>. They are, by contrast, clearly absent from the <u>Papaveraceae</u>, a family sometimes placed in the same order (Wettstein, 1935).

Saghir <u>et al</u>. (1966), using the gas chromatographic technique, determined the amount of methyl, n-propyl, and allyl sulphides in the volatile fractions of 25 North American species of <u>Allium</u> and compared the results with the sectional groupings of species based on cytology and morphology. Unfortunately, they found that a classification based entirely on chemical characters would not only place clearly unrelated species such as <u>Allium cernuum</u> and <u>A. haematochiton</u> together but would separate otherwise very similar taxa like <u>Allium campanulatum</u> and <u>Allium</u> <u>membranaceum</u>.

2. <u>Macromolecules</u>

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In some cases, the macromulecular data support the micromolecular data in confirming generic separation. In 1968, Harborne said

"that macromolecular data is more 'fundamental' or more phyletically significant than data obtained from studying secondary constituents. ... One real advantage of macromolecular data is that they will provide a means of chemically comparing widely separated taxa, an operation which is rarely possible by any other means. They may also, by means of amino acid sequences in proteins or DNA-DNA hybridization data, provide taxonomists with an objective means of drawing lines between the larger units of classification (family, order, class, etc.)".

At the present time, some macromolecules in plants have received attention with respect to taxonomic significance and these are discussed briefly below:

a. Polysaccharides

The distribution of polysaccharides in plants has been reviewed by Percival (1966). From the view point of chemotaxonomy, the more complex polysaccharides may eventually yield characters of use in systematic work. However, complete structures are not available and data on the sugar units present within complex polysaccharides are only available for relatively few plants. Therefore, only a few examples of their contribution to taxonomy follow:

Whistler and Gaillard (1961), in studying the xylans from the hemicellulose-A fractions of several annual plants, found the species of Leguminosae, unlike those of the Gramineae,

contained no arabinose at all. The uronic acid content of the polymers, on the other hand, is higher in the hemicellulose-A fraction from the Leguminosae. Later Gaillard (1965), in examining the composition of corresponding linear and branched polymers from the hemicellulose-B fraction of three members of the Gramineae and three of the Leguminosae, found the corresponding polymers from the former were very similar as were those from the latter. However, there was a distinct difference between the two plant families. The linear polymers of the Gramineae contained more arabinose and less glucuronic acid than those from the Leguminosae. The greatest difference between the two plant families was found in the branched polymers, those from the Gramineae containing a high percentage of xylose and rather small amounts of arabinose, galactose and uronic acid, whereas those from the Leguminosae contained relatively large amounts of uronic acid, galactose and arabinose and little xylose. In the branched polymers from the Gramineae the uronic acid was linked to xylose, whereas in those from Leguminosae it was linked to arabinose. However, it is still not clear whether or not the differences in hemicellulose composition between the Gramineae and the Leguminosae are a reflection of differences between Monocotyledons and Dicotyledons, rather than between the families.

b. Lignins

Lignins are found in vascular plants such as lycopods, ferns, gymnosperms and angiosperms, whereas it is absent from

plants such as fungi, and from all other organisms. The mosses are an exceptional group which do not have the cells characteristic of xylem tissue but which do contain lignin-like materials. These "moss lignins" have been discussed by Freudenberg (1968), and some doubts have been expressed as to whether or not they are true lignins.

There are, in general, differences between the lignins of gymnosperms and angiosperms. The woods from angiosperms give a rose-red color, whereas gymnospermous woods usually give only a brown colour with Maüle's test. A suggestion by Gibbs that the red coloration of the Maüle test is correlated with the presence of syringyl groups in lignin was verified by the work of Creighton, Gibbs and Hibbert (1944); Towers (1951); and Towers and Gibbs (1953).

That the angiosperms universally have lignins containing the syringl group is strongly suggested by the work of Gibbs (1958, and unpublished), he has carried out Maüle's test and got positive reactions from 207 families of dicotyledons. Only a few have given negative or doubtful results and these were mostly aquatics or very lightly lignified plants. At least 35 families of monocotyledons also gave positive results, the few exceptions being again largely aquatics.

c. Serology

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Alston and Turner (1963) reviewed the contribution of serology to systematics and concluded that, while catalytic to positive thinking in some instances, the extravagant claims

made for it apparently created more disinterest in "chemistry" as an approach to plant taxonomy that it did enthusiasm.

However, quantitative serological methods have been applied for many years to taxonomic investigations and are useful for the differentiation of plant species since they may offer an overall picture of the similarity of the proteins (Leone, 1964). In recent years the more qualitative methods of immunoelectrophoresis, which offer the possibility of distinguishing different proteins without the need of previous separation, are recognized to be exceptionally well suited for comparison of protein patterns from different origins, and have been successfully applied to selected problems. For instance, Klozova and Kloz (1964) have used immunochemical methods to detect the hybrid Phaseolus vulgaris x P. coccineus, the F₁ possessing a complementation of the distinctive parental protein lines, much as was found in the flavonoids in hybrid But Turner (1967) says: Baptisias.

"The detection of interacting macromolecular bands by immunogenetic techniques reflects the activity of relatively few genes and, viewed in this light, it is doubtful that this approach, taken alone, will contribute significantly to problem of plant phylogeny, although it has high value for distinguishing among presumably homologous proteins."

d. Proteins

In recent years, a number of investigators have shown a correlation between protein composition and systematics in higher plants. Using electrophoretic techniques, Johnson and

Hall (1965) investigated relationships in the <u>Triticineae</u> (<u>Gramineae</u>) and Boulter <u>et al</u>. (Fox <u>et al</u>., 1964; Boulter <u>et al</u>., 1967; and Thurman <u>et al</u>., 1967) examined separately the systematic relationship of albumin and globulin fractions in seeds of certain <u>Legumes</u>, and of two dehydrogenase enzymes within the same family, while Vaughan and Denford (1968) surveyed the albumin and globulin fractions of the seeds of a number of species of <u>Brassica</u> and <u>Sinapis</u>, correlating the results with the established taxonomy. Other workers (Gell <u>et al</u>., 1960) have used immunochemical techniques to demonstrate taxonomic affinities between species.

More recently, Crowden (1969), using acrylamide gel electrophoresis, surveyed soluble proteins and the enzymes peroxidase and esterase, present in the seed of selected species from all tribes in the <u>Apioideae</u> (<u>Umbelliferae</u>) and found distinct differences in patterns to be present at the tribal and generic levels.

It has been suggested (Wilson and Kaplan, 1962) that enzymes are better suited for taxonomic investigation than other proteins, and that a comparison of enzymes may allow assessment of genetic relations in and between taxa.

Numerous investigations have been performed which have demonstrated that isoenzyme staining patterns are dependent upon several variables. In interspecific comparisons, many patterns are reported to be species-specific and of diagnostic

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utility (Schwartz <u>et al.</u>, 1964; Clements, 1966; Bhatia <u>et al.</u>, 1967). Many authors (Shaw, 1965; Boulter <u>et al.</u>, 1966) have expressed the desirability of a more complete knowledge of the extent of genetically-based protein variations in natural populations in order to insure proper usage of isoenzyme pattern data for systematic purposes. However, Scogin (1969), in surveying natural populations of three species of <u>Baptisia</u> (<u>Leguminosae</u>), found no species-specific patterns and suggested that there is no way to predict, <u>a priori</u>, the possible taxonomic or physiological implications of a given isoenzyme pattern until possible intraspecific variation has been evaluated.

EXPERIMENTAL

A. <u>Material</u>

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Mainly, fresh leaves and stems of mature plants were used in this investigation. Specimens were obtained from the McGill University greenhouse, Montreal Botanical Gardens, and some important botanical gardens all over the world. Imported specimens were flown to Montreal, packed in polyethylene bags to ensure freshness.

B. Methods

As mentioned in the Introduction, there are many methods which have been well developed for chemotaxonomic study. They may be said to belong either to an intensive or to an extensive approach. Although most people prefer the intensive approach, they can with this study only a few plants and chemical constituents in years of work. We have chosen the extensive approach, which means doing as many <u>simple tests</u> as possible on as many species as possible in the time available. It precludes detailed chemical work, except at rare intervals.

Gibbs has spent many years, using simple tests as devised by others or modifications of these (Gibbs, 1962, and MSS). I have used these tests and in addition have done chromatography for phenolics as outlined below.

1. Cigarette and Hot-Water Tests

Dagmar Dykyj-Sajfertová (1958), in a paper on respiration pigments, described two simple tests which probably reveal the presence or absence of <u>polyphenolases</u> and their <u>substrates</u>. These tests were adopted by Gibbs and they are described below. I have included Miss Dykyj-Sajfertova's results, those of Gibbs (MSS), and my own in Appendix I.

Fresh leaves (mature, but not senescent) are used for the tests.

a. <u>Hot-Water Test</u>: The leaf is dipped halfway into hot water (85-90°C) for five seconds and then removed. Rapid darkening along the water-line (and sometimes of the whole dipped portion) is designated by I. If, after a minute or more, a dark 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 IV. In addition Dykyj-Sajfertova noted an "oxalis-reaction" (because first seen in Oxalis) -- a yellowing of the dipped portion. This particular result is perhaps due to highly-acid cell-sap; however, it has also been obtained in young leaves of other plants. It was observed doubtfully in two or three of our species.

b. <u>Cigarette Test</u>: A lighted cigarette is pressed gently against the underside of the leaf for three seconds. The results are classified as follows:

- I. -- an immediate reaction (formation of a dark ring around the heated area)
- II. -- a slower reaction
 III. -- a very slight reaction
 IV. -- no reaction

o.r.-- "oxalis reaction"

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The results of both tests are similar, but the reaction in the cigarette test is more rapid. These tests have been found to be good chemotaxonomically. Some groups give constantly positive (I-II) reactions, others are negative (IV). While yet others are mixed.

2. Syringin (1:1 H₂SO₄/H₂O) Test

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Freshly-cut transverse-sections of stems, or sometimes petioles, are used for this test. Two sections are placed on separate microscope slides. To one, a drop of 50% aqueous sulphuric acid is added and the preparation is examined under the microscope at intervals during about thirty minutes. A <u>blue color</u> in the xylem, lignified fibres, etc. is recorded as a <u>positive</u> "Syringin test". It is said to be due to the presence of <u>syringin</u> (Tunmann, 1931). Syringin, which was first found in Syringa, is the glucoside of 5-methoxy-coniferyl alcohol.

.OCH 3 _0-glucose HOH2C-HC=CH

Syringin

Other colors may appear in the xylem and fibres. A yellowish color often appears. The development of a pink to red color in the lignified tissue is closely correlated with positive HCl/ methanol and positive leuco-anthocyanin reactions. Purpling,

or darkening, especially in the cortex, is associated with the presence of aucubin or aucubin-like substances.

The other section, in a drop of water, is used as a control. The presence or absence of <u>raphides</u> and/or other crystals is noted in this section too. Most plants are negative to the syringin test.

3. Raphides

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These are bundles of needle-shaped crystals of calcium oxalate, occurring in special raphide-sacs (Gibbs, 1962). He, in 1963, says that the taxonomic importance of raphides was recognized by Robert Brown who considered its presence or absence as a diagnostic character. Gulliver (1866) remarks that raphides are restricted in distribution and that they may be used as taxonomic characters. Metcalfe and Chalk (1950) write:

"Other types of crystalline secretion such as raphides and crystal-sand are more restricted in distribution and therefore of still greater taxonomic importance."

We have included our own observations, made on control sections when doing the syringin test (above), observations of Gibbs (MSS), and others from the literature.

4. Ehrlich Tests (A and B)

Gibbs got this test from G.H.N. Towers but we do not know the original source of it.

About 0.5 gm of fresh leaf-blade material is chopped, placed in a test tube, and extracted with a little boiling 50% aqueous ethanol. The extract is then concentrated by evaporating it to a small volume.
Three spots of the concentrated extract are built up on a 9 cm. filter paper (Whatman No. 1) and allowed to dry. To the second spot is added a drop of acid alcohol (5 ml conc. HCl in 200 ml of 95% ethanol) as a control, to the third spot is added a drop of Ehrlich's reagent (1 gm p-dimethylaminobenzaldehyde; 5 ml conc. HCl: 200 ml 95% ethanol), and again the spots are allowed to dry. Often no marked change of colour occurs but the third spot may become <u>blue</u> (a <u>positive</u> reaction) or <u>magenta</u>. The filter paper is then placed in a preheated oven $(100^{\circ}C)$ for one minute. This sometimes causes the development of a <u>blue</u> color where it has not previously appeared.

A <u>blue</u> (positive) reaction is caused by the presence of aucubin or aucubin-like substances.



Aucubin

A <u>magenta</u> colour is very closely correlated with the red colour observed in the syringin test, with a magenta (positive) HCl/methanol reaction, and with a "carmine" (positive) leucoanthocyanin reaction. In the absence of a blue or magenta colour, yet other colours -- such as yellow -- may be observed. To the first spot a drop of dilute aqueous (10%) ammonia

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is added (Test B). Usually a pale yellow colour is produced, but sometimes a bright yellow colour appears, and although this has not been tested, this colour may be indicating flavonoids. Rarely a red or other off-beat reaction is developed. Such colours are recorded, since they may prove to be of taxonomic significance.

5. The HCl/Methanol (or Isenberg/Buchanan) Test

This test seems first to have been described by Isenberg and Buchanan (1945), and they found it to have some taxonomic significance. Gibbs verified this, and has adopted and modified it. I include, again, my own and his (MSS) observations. It is carried out as follow:

Fresh chips of wood (usually sapwood, and often obtained with a pencil-sharpener from pencil-sizedtwigs) are placed in a test tube and covered by a few millilitres of HCl/methanol solution (25 ml conc. HCl : 1000 ml methanol) and left for some hours, usually overnight.

A <u>magenta</u> colour (positive test) may develop, or no colour to a pale yellow colour may result (negative test). Using Ridgway's "Color Standards and Color Nomenclature' (1912), the specific colour is recorded. The positive reactions are also rated:

Purple 1 very pale purple

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2 pale purple

3 magenta

4 darker than magenta

Adler (1951) concluded that this magenta colour (positive) is due to the presence of catechol tannins. These condensed tannins, according to Swain (1965), are formed by the condensation of two or more molecules of flavan-3-ols, such as catechin, or flavan-3,4-diols, such as leucocyanidin, or mixtures of the two.



Catechin : R=H Leucocyanidin : R=OH

and he writes:

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"The hydroxylation patterns of the monomers vary depending on the source, but are generally related to the commonly occurring flavonols and anthocyanins."

Thus they are very closely correlated with the leucoanthocyanin test. Gibbs has observed a very rare <u>orange</u> reaction with the HCl/Methanol test (Gibbs, <u>et al.</u>, 1967). We have not seen it in any of our material.

6. Leucoanthocyanin Test A

This useful test is due to Bate-Smith. He demonstrated it to Gibbs, who has adopted and used it extensively. It is carried out as follows. Freshly-chopped leaves (ca. 0.5 gm) are placed in a 150 x 25 mm test tube which is etched at the 5 and 10 ml points. 2N HCl is added to the 5 ml level; the test tube is immersed in a boiling water bath for 20 minutes; after which it is removed and cooled. Isoamyl-alochol is then added to the 10 ml mark, and the solution is vigorously shaken.

On standing, the mixture separates into two layers. A <u>red colour</u> in the upper (isoamyl-alcohol) layer is a <u>positive</u> reaction, while a yellow, buff or green colour is negative. We usually match the colour against Ridgway and record it.

In this test, a <u>positive</u> reaction is considered to be due to the presence of leucoanthocyanins, which are colorless and water-soluble, but which are hydrolysed and oxidized to the corresponding isoamyl-alcohol-soluble <u>anthocyanidins</u> (Bate-Smith, 1954).



A leucoanthocyanidin

An anthocyanidin

The results of this test are usually distinctly positive or negative. Sometimes, however, because of the small amounts

of leucoanthocyanin or the presence of interfering substances (such as aucubin or aucubin-like compounds which give black or bluish colours) the results are doubtful. Catechins also obscure the test, especially in acid solution. They may form red-brown polymers known as phlobaphenes which are isoamyl alcohol soluble.

According to Bate-Smith (1965), the leucoanthocyanins are prevalent in woody plants, and the presence of these compounds are regarded as being a less advanced character.

7. HCN Test A

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About one gm of fresh leaf and stem material, including young tips if possible, is ground in a mortar with a few drops of water, a speck of emulsin (which hydrolyses cyanogenic glycosides and releases HCN), and a drop or two of chloroform. The mixture is poured into a glass-stoppered tube (ca. 150 x 25 mm). To the glass stopper has been fixed with wax an almost triangular piece of picric acid paper freshly dipped in 10% sodium carbonate (Na₂CO₃) and blotted.

A <u>strongly positive</u> reaction is one in which the yellow sodium picrate paper turns a deep rust colour within minutes to hours; while a <u>weakly positive</u> reaction is one in which the colour change may not show up clearly for several days and even then is not deep. In the absence of HCN the paper remains yellow (<u>negative</u> reaction). The test tubes are left in a rack for at least a week before being discarded.

Bohm (1803) first reported the presence of HCN or prussic acid in plants. Later, various authors, Lindley (1830), Dillemann (1958), Gibbs (1963), and others considered that HCN is taxonomically interesting.

8. Juglone Tests A-C

This old test is thus named because it was first described for juglone itself. Other naphthaquinones also give colour reactions with it. Only Test A, however, is for juglone ' and other <u>naphthoquinones</u>. Tests B and C are for other compounds but are conveniently included here.

"Juglone Test A" is carried out as follows. A little finely-chopped material (from leaves, root or stem, but preferably the root, and where possible bark material) is placed in a test tube. It is then covered in chloroform and left, with some shaking, for hours or overnight. The chloroform extract is evaporated to dryness on a water-bath, the residue is taken up in a few ml of ether and a few ml of 10% aqueous ammonia (NH₄OH) added.

On shaking, a brilliant <u>purple colour</u> is a <u>positive</u> reaction and is due to the presence of juglone or some closely-related naphthaquinone. An <u>orange</u> or <u>wine colour</u> may be indicative of other naphthoquinones also.

In the absence of naphthoquinones, it was seen that not infrequently the <u>ammonia</u> layer is <u>yellow</u>. This probably indicates the presence of flavonoids and is recorded as "<u>Juglone</u> <u>Test B</u>".

By using a long wave ultraviolet lamp any notable fluorescence of the ammonia layer is recorded as "Juglone <u>Test C</u>." Bright blue or green fluorescence is probably due to <u>coumarins</u>.

Gibbs (1965) finds that some plants develop, in the ammonia layer, slowly a green to blue-green colour from above. He suggests that it may prove to be of definite taxonomic interest. In order to be sure that one does not miss this slow reaction, the tubes from "Juglone Test A" are allowed to stand for several days before discarding.

Juglone (below) has been known to be present in the walnut for over a century (Thomson, 1957). I have included with my results some from Gibbs (unpub'd.).



9. Tannin Test A

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Gibbs first met this test in a thesis by Miss Harney. He has adopted it and carries it out as follows:

The reagent for tannins is 2.5% aqueous ferric ammonium citrate, freshly-prepared or kept under refrigeration. A small (we use 7 cm) Whatman No. 1 filter paper is dipped into the solution and blotted. A piece of clean leaf material is

placed on the filter paper which is folded around it. It is then squeezed with a pair of pliers.

A <u>positive</u> reaction is one in which a grey to purple colour is seen at once on the filter paper. The intensity of the colour is rated by positive (+, ++, +++) signs.

A <u>negative</u> reaction is one in which no such colour develops.

As a control we squeeze another piece of the same leaf with the filter paper dipped only in water. This enables us to distinguish colour reactions due to "Tannins" from those which may develop otherwise.

10. Saponin Test A

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Following the methods of Amarasingham and his coworkers (1964), a small amount (0.5 gm \pm) of finely-chopped fresh leaves is placed in a 150 x 15 mm glass-stoppered tube which is marked at 5 ml and 10 ml levels. Distilled water is added to the 5 ml mark. The mixture is boiled briskly for 1 minute and cooled. It is then vigorously shaken, and left standing for five minutes. The amount of foam is then noted. We record our results as follow:

If a deep (72 cm) layer of foam has persisted it is assumed that saponins are present (+); if rather less foam persists we record the result as doubtfully positive (+?); if a little foam remains we class the result as probably negative (-?); and if there is no foam, as negative (-). The tubes with their contents are then used for the test below.

11. "Saponin Test B (NH₃)"

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This is not a test for saponins but is conveniently included here since we use the tubes with their contents from Saponin Test A (above). Ammonia (10% aqueous) is added to the 10 ml mark. The test tube is shaken and the initial colour is recorded. Some samples give almost no colour, others a pale yellow or a very deep yellow." On standing for 3 days, the colour may remain unchanged or may even fade somewhat, or may deepen to orange-brown or even almost black. At present, it is not known why aqueous ammonia causes darkening of the solution, but darkening is closely correlated with occurrence of tannins. The colour deepens from above, thus indicating that oxygen is necessary for the reaction. Therefore, we remove the stoppers and shake gently from time to time to facilitate oxidation. Using Ridgway we record our results.

0 -- no change in colour, or fading

- 1 -- about "yellow ocher" to "ochraceous orange"
- 2 -- "tawny" to "hazel"
- 3 -- "liver brown"
- 4 -- deeper than "liver brown"

This test has evidently some taxonomic value (Gibbs, 1965).

12. Paper Chromatography for Phenolic Constituents

The application of chromatographic techniques to the analysis of phenolic mixtures in higher plants has opened a new

era in the study of these substances and their distribution in plant tissues. By means of chromatographic methods a large number of plants have been screened for phenolic constituents , and many phenolic substances infrequently detected in the past, such as chlorogenic acid, have been shown to be almost ubiquitous in higher plants.

Paper chromatography was first employed for the separation of phenolic pigments by Bate-Smith (1948) and has been widely used since with all types of phenolic compounds. Many methods have been developed and data for the separation of various phenolic substances have been summarized by Block <u>et al.</u> (1958) and Harborne (1961, 1967). Lists of R_f values in several solvent systems of most of the known, naturally-occurring phenols and related compounds are available (Harborne, 1958, 1959; Block <u>et al.</u>, 1958; Reio, 1960).

a. <u>Preparation of plant extracts</u>

Phenolic acids occur in plants largely as esters and glycosides (Bate-Smith, 1962a). The common practice for their extraction from plant tissue involves the use of diethyl ether, hot aqueous ethanol, methanol or isopropanol. Boiling <u>N</u> NaOH has also been used (Pearl <u>et al.</u>, 1957). The method described by McCalla and Neish (1959) has been found to be very satisfactory by other workers (Ibrahim and Towers, 1960).

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The present writer followed this method (slightly modified) for phenolic acid analysis. The fresh plant material is homogenized during extraction with aqueous ethanol and the ethanolic extract is evaporated to dryness. The residue thus obtained is re-extracted in a small volume of boiling water and washed with petroleum ether. The filtrate is extracted with ether, to remove the free acids, and then subjected to either alkaline or acid hydrolysis to release the phenolic acids bound in either ester or glycosidic linkages. The acids released on hydrolysis are subsequently extracted with ether under acidic conditions. The detailed extraction procedure is shown in Fig. 1.

b. Chromatographic Techniques and Equipment

i. <u>Paper</u>: Large sheets (18 1/2" x 22 1/4") of Whatman's No. 1 chromatography grade filter paper were used for two-dimensional separation of plant extracts fractionated as mentioned under a.

ii. <u>Tanks</u>: Air-tight, chromatographic chambers containing glass troughs suitable for development of large chromatograms by the descending methods were employed.

iii. <u>Solvents</u>: One of the best solvent systems for chromatographing the phenolic aglycones is that of Ibrahim and Towers (1960), but we used aqueous formic acid (2% by volume) (Bohm and Towers, 1962) instead of their mixture of Sodium Formate: Formic Acid: Water (10 : 1: 200) for the second direction.

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Two solvents were prepared with analytical grade reagents and distilled water as follows:

(1) <u>Benzene-Acetic Acid-Water</u> (10:7:3 by volume). This is prepared by shaking for a few minutes in a separatory funnel and then allowed to stand for at least 1 hour (and often overnight).

This two-phase solvent was used for the first direction in the chromatographic separation of the phenolic acids. In a tank (well-saturated by leaving for 4 hours with the aqueous phase) 70 ml of the organic phase were found adequate to irrigate two chromatograms at 21.1° C in 4 to 4.5 hours. Complete removal of the solvent from chromatograms required from 2 to 3 hours in the fume hood with the fan running.

(2) <u>Aqueous formic acid</u> (2% by volume) (Bohm and Towers, 1962). This solvent was used for the development of the chromatograms for phenolic acids in the second direction. Development time was 3 to 3.5 hours using 65 ml of solvent for irrigation of two chromatograms at 21.1°C. Chromatograms were dried subsequently for 4 to 6 hours.

c. Identification of Phenolic Acids

The identification of the phenolic acids was carried out by examining the paper chromatograms under ultraviolet lamps, both longwave and short wave, before spraying. The fluorescence was examined again after exposure to ammonia vapor. Chromatograms were then sprayed with one of the following reagents. i. <u>Diazotized p-nitroaniline</u> (Bray <u>et al.</u>, 1950)

The following stock solutions were prepared:

- (1) p-Nitroaniline; (2) 5% Sodium Nitrite solution;
- (3) 20% Sodium Acetate solution; (4) 5% Sodium

Hydroxide solution.

Chromatograms were sprayed with a mixture of stock solutions 1, 2 and 3 in a 5:1:15 ratio by volume, followed by overspraying with solution 4.

ii. Diazotized Sulfanilic Acid Spray (Evans et al., 1949)

The spray reagent was prepared from the following stock solutions:

(1) Sulfamilic acid solution prepared by dissolving
 9 gm of sulfamilic acid in 90 ml conc. HCl and diluting to one
 litre with distilled water.

(2) 5% Sodium Nitrite solution; (3) 20% Sodium Hydroxide solution.

The spray reagent was freshly prepared prior to use by mixing the 3 stock solutions in a 2:1:2 ratio by volume.

iii. 1% Ferric chloride solution

Since reference compounds had been run by earlier workers, and a chart had been prepared giving the colour and position of each known compound, the spots on the chromatograms were thus identified.

RESULTS

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For convenience in comparing and contrasting the systematic relationships of Rutales, all results are represented by tables 2-10.

The data have been collected (1) through work done in the laboratory by myself, (2) a survey of literature, up to September, 1969* and (3) the information cards of Dr. R. D. Gibbs (most unpublished).

Classes of chemical constituents as delimited in this thesis are those of Gibbs (MSS). For the writer's summary of her results, consult the tables in the appendix.

*A few later items are included.

						TE	STS					
F.		H.W. and CIG.	SYRINGIN	RAPHIDES	EHRLICH	HCN (TEST A)	HC1/METHANOL	L.A. (TEST A)	JUGLONE (TEST A)	FLUORESCENCE	TANNIN (TEST A)	SAPONIN (TEST A)
	Rutaceae	(∓.).	(-)(NR)	(-)	-(^{ot} ⁄M)	(-)	(Ŧ)	(Ŧ)	-	(+)	(±)	-
	Cneoraceae	(∓)	- (NR)	-	-ot	-	-	-	-	-	-	-
	Simaroubaceae	(±)	- (NR)	-	-(ot)	-	(-)	(-)	-	(ᆂ)	+	-
	Picrodendraceae	-	-(R)	-	-M	-	+		-	+		
	Burseraceae	(∓)	- (NR)	-		-	(∓)		-	±	÷	
	Meliaceae	(∓)	– (R)	-	-(^M Ot)	(-)	(+)	±	-	+	(±)	(∓)
	Akaniaceae	+	- (R)	-	-M	?	+	+	-	+	+	
•	Malpighiaceae Trigoniaceae	(∓)	-(R)	_	- (M)	(-)	(+)	(+)	_	(±)	(±)	-
	Vochysiaceae						±		-	. – .	1 -	
•	Tremandraceae	-	- (R)	-	-(^M ⁄6t)	(+)	+		_	+	+	
)	Polygalaceae	(∓)	-(NR)	-	-(ot)		(∓) .	(∓)	(-)	(∓)	. ±	(-)

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

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	TANNIN (TEST A)	SAPONIN (TEST A)	"SAP. TEST B" (NH ₃)	PHENOLIC ACIDS Benzoic	Phenylacetic	Cinnamic	Ellagic	COUMARINS	Simple coumarins	6,7-Furocoumarins	7,8-Furocoumarins	Chromanocoumarin	FLAVONOIDS	Anthocyanins	Leucoanthocyanins	Flavanoes	Flavanonols
⊦}	(±)	-	(∓)	(+) (+)	(-)	(+)	(+)	(+)	(+)	(+)	(-)	(+)	+	(-)	(∓)	(∓)	(-
-	-	-	-	(+) (+)	-	(+)	-	(+)					+		-		
=)	+	-	+	(+) (+)	-	(+)	(+)						+		-		
F																	
F	+			(+) (+)	-	(+)	(+)						+		(+)		
۲	(±)	(∓)	±	(+) (+)	(-)	(+)	(+)	(+)	(∓)	-	(-)	(-)	÷		(-)		
۲	+			(+) (+)	-	(+)	(+)										
±)	(±)	-	±	(+) (+)	-	(+)	(+)	(-)					+		(+)	<u> </u>	
- .	1 -	· .															
+	+			(+) (+)		(+)	(+)						+		_		
Ŧ)	. ±	(-)	±	(+) (+)	-	(+)	(+)						+		-		
	ve in es on be			Key	to	sym	bols:	- + (- + +) (- + +) (+ +) (+) R o) = = = = = = = = = = = = = = = = = = =	all maj mor hal mor no red oth	ne jori jori e p f p re n red	ty a osit osit egat than		egat han hal han	ive ne f no pos	egat	:i1

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TABLE 2

Chemical Characters of "Rutales"

	Ø]		1					-)	•	
		tive tive n neç lf ne n pos			(+)	ł	Ŀ	(+)	I	I	(†	Leucoanthocyanins	-0 8 =
		ive ive negative f negative positive									÷	Flavanoes	
		ve ive				}					î	Flavanonols	
											(-) (∓)	Flavones	
			(† (†		1+	1	Ŧ	(+	£	n		Flavonols	{
					• •		J	Ţ	ť	[+	î.		
						{	+	+	+		+	TERPENOIDS	
•			.			}	+	(+) (+)	I		(+) (±)	Monoterpenoids	1
							1+	(+)	1		î†	Sesquiterpenoids	
		• •	t	{ 		{ }	1+	(+)	I		, L		{
											÷	Diterpenoids	ļ
			÷				(+)		+		(+) (:	Triterpenoids	· ·
							I	1	1		(†	Tetraterpenoids	}
			+ +	• [(†)	+	(Ŧ)	-	1+		(+	ALKALOIDS	
							I		I		I+	Acridines	
							I		I.		H	Alkaloidal amines	
				}		{	+'		ı		Ŀ	Imidazoles	
							I		1+) +	Indoles	
							ı		1		I+	Isoquinolines	
						1	I		1		Î	Oxazoles	
							, I		ı		(-) (-)	Pyridines	{
							ı		1		<u>.</u>	Pyrrolidines	
					•		1		• •		·) (-)	Quinazolines	
· .						{	1		1+		+	Quinolines	
		•											с.
			I.			1						1	•

Fzmilies, etc. No. of genera			H.W.	& Ci	g. Te	ests			Sy	ringin Te	st
bracketed		I	-	II	III		IV	or	+	_	3
Rutaceae (204	+)		$4/4 \rightarrow 2$	6/7 /2 🛶	15/19	$\frac{2}{2}$	20/30	•	1/1	{29/46NR 18/10R (16/26NR	2/2R 1/1N
Rutoideae (142 Dictyolomatoid	leae (4		1,	/1	7/7	4	4/6		1/1	3/3R	272R
Flindersioidea Spathelioideae)	<u>1/1</u>							1/1R (5/6NR	
Toddalioideae		1/3	<u>1/1</u>	2/3			3/5			2 /3R	1/1N
Citroideae	(30)	1/1	<u>2/2</u>	<u>4/4</u>			6/14			8/14NR 2/2R	1 1
Rhabdodendroid	leae					_ •				1/1NR	
Cneoraceae	(2)					1/1				1/1NR	
						the second secon				(1/1R	1
Simaroubaceae	(26)	1/1	1,	/1 +++	1/1					5/5NR	
Surianoideae	(4)		1,	/1	-					1/1R	1
Simarouboideae										3/3N R	
Kirkioideae	(1)						1/1			1/1NR	
Irvingioideae	(4)										,
Picramnioideae		1/1								1/1NR	
Alvaradoideae	(1)										
Picrodendraceae	(7)						1/1			1/1R	
					- 1-					{2/2NR	
Burseraceae	(20)				2/2					1 /1R	
							c lc			{4/5R	
Meliaceae	(50)	4/6					6/6			{2/2NR	1
							á (a			1/1R) Y
Cedreloideae	(4)	1/1					2/2			1/1NR	
Swieteniodeae	(8)	1/1					1/1			2/2R	
Melioideae	(28)	211					2/2			$\int \frac{1}{2R}$	
Akaniaceae	(20)	2/4	· 1	/1			3/3			(1/1)	
AKaiilaceae	(1)		т,	/ 1						1/1R (2/2NR	
Malpighiaceae	(65)	1/1					3/4			4/8R	1
Trigoniaceae	(4)	1/1					5/4			(4/ ØK	
Vochysiaceae	(6)										;
vourystaceae	(0)									(1/1NR	
Tremandraceae	ુ (૩)						1/4			$\frac{1}{3R}$	
Polygalaceae	(13)			<u>1/</u>	1		1/3			2/6NR	
Xanthophylleae							J (-			• <i>16</i>	
Polygaleae	(8)			1/	<u> </u>		1/3			2/6NR	
Moutabeae	(4)										

Key to symbols: NR = no red; R = red; ot(in Ehr.) = color other than magenta or M = magenta; bk = bark blue

*Fraction represent numbers of genera/species

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TABLE 3

A list of plants tested (by families and subfamilies) in "Rutales"

Test	e			Ehrlich	+	HCN	~	нс1 +	/Meth.	2		L.A.	~
NR	? 2/2R	+	- +	- 16/28ot						?			?
)R	1/1NR	3/3	34/44	16/23M	2/4	34/53	1/1	9/12	29/46	4/6	13/17	21/30	2/
NR				7/80t	-		-, -		-				•
	2/2R	3/3	19/29	11/18M	1/3	18 /2 7		3/5	18/27	2/3	8/12	12/15	
			1/1	1/1M		1/2		1/1			1/1		
	1 /1		F /0	3/5ot				2/2	6 /0	1 /1		A 16	21
R	1/1NR		5/8	2/2M 6/15ot		6/8		3/3	6/9	1/1		4/6	2/
			9/16	2/2M	1/1	9/16	1/1	2/3	5/10	1/2	4/4	5/9	1/
			1/1	1/1M		1/1							
			1/1	1/10t		1/1			1/1			1/1	
			-	4/4ot		·			-				
			6/6	1/1M		5/5		1/1	4/4 ·			6/6	
			1/1	1/1M		1/1		1/1	a /a			1/1	
•	•		3/3	2/20t		1/1			3/3			3/3	
	1		1/1	1/lot		1/1						1/1	
L			1/1	1/1ot		1/1 1/1			1/1			1/1	
	-		1/1	1/1M		1/1		1/1					
2			2/3			2/2		2/3				1/1	
			•	3/3ot				-					
2	1		7/7	4/5M 1/1 ot	1/1	9/11		7/10	1/1		4/5	4/4	
Ł	1		2/2	1/1M		2/2		2/2			1/1	2/2	
			$\frac{1}{2}/2$	2/2M		$\frac{1}{2}/2$		2/2			2/2	•	
			,	2/20t				•					
ર			3/3	1/2M	1/1	5/7		3/6	1/1		1/2	2/2	
			1/1	1/1M			1/1	/1/1			1/1		
2			7/10	1/1 ot		A /7	1/2	7/10	2/2		4/6	1/1	
			7/10	4/6M		4/7	1/2	-	-		4/0	-/-	
	1			1/1				1/1	1/1				
•			2/5	1/1 ot 1/2M	2/2	1/3		2/5				1/1	
			2/5	1/4ot	2/2	1/5		-, -					
٤			2/6	1/1M		3/5		1/1	2/4		1/1	1/2	
٤			2/6	1/4ot 1/1M		3/5		1/1	2/4		1/1	1/2	

or

utales"

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•A•		Juglone	Fluor	escenc	e	Tan	nin			Sapo Test		"Sap. (N ^H	Test B" 3)
	?	+ –	+		?	+		?	+		?	<u>+</u>	
21/30	2/3	27/36 11/14 13/19	27/42	7/10	1/1	13 /20	8/11	4/4		13/4	2/2	4/4	10/15
12/15		6/7¥	16/26	1/1		9/15	7/9	3/3		4/6		2/2	3/5
	•	1/1	1/1			1/1				1/1		· -	1/1
4/6	2/2	5/5 1/1PY	4/4	1/3		3/4	1/2	1/1		3/5	1/1	1/1	2/4
5/9	1/1	8/11 4/6Y	6/11	5/6	1/1	5/6	5/6	1/5		5/9	1/1	1/1	4/5
1/1		1/1Y 1/1Y		1 1/1						1/1			1/1
6/6 1/1	:	5/5 1/1	4/4	2/2 1/1		3/3 :				2/2 1/1	1/1	1/1	
3/3 1/1		3/3 1/1	3/3 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		2/4	1/2	1/2		1/1				•			
4/4	•	8/9	7/8		1/1	5/6	2/2		2/2	2/2	2/2	4/4	4/4
2/2	6 	2/2 2/2	2/2 1/1		1/1	1/1 2/2	1/1		1/1 1/1	(bk)	2/2	2/2 2/2	2/2
2/2	÷	4/5 1/1	4/5 1/1			2/3 1/1	1/1			2/2			2/2
1/1		5/7 1/1¥	5/5	3/3		5/6	3/3			3/4		2/2	2/2
	•	2/2		2/2									
1/1	ı	· 1/1	1/1			2/2							
1/2	!	1/1											
1/2		1/1 1/2 2/2¥	1/1	2/ 2	1/1	1/1	1/1		2/3	1/2	1/1	1/1	1/1

.

TABLE 4

Occurrence of groups of phenolic acids by families and

subfamilies in "Rutales" (see Appendix II)*

		Benzoic	Phenyl- lacetic	Cinna- mic		
		acids	acids	acids	Coun	arins
Rutaceae	(204)	27/33	2/2	30/27	27/33	20/20**
Rutoideae Dictyolomatoideae Flindersioideae	(142) (1) (2)	15/20		17/22	14/18	- 10/10**
Spathelioideae Toddalioideae Citroideae Cneoraceae Simaroubaceae Simarouboideae	(3) (24) (30) (2) (26) (15)	5/5 7/8 1/1 2/2 1/1	2/2	5/6 8/9 1/1 2/2 1/1	5/6 8/9 1/1 2/2 1/1	4/4** 6/6** 1/1**
Kirkioideae Picrodendraceae Burseraceae	(1) (1) (20)	ī/ī 1/1		1/1 1/1	ī́/ī 1/1	
Meliaceae Cedreloideae	(50) (4)	4/5	1/1	6/7 1/1	6/7 1/1	1/1**
Swietenoideae Melioideae Akaniaceae	(8) (28) (1)	2/2 2/3 1/1	1/1	2/2 3/4 1/1	2/2 3/4 1/1	
Malpighiaceae Trigoniaceae Vochysiaceae	(65) (4) (6)	1/2		2/3	1/2	1/1**
Tremandraceae Polygalaceae	(3) (13)	2/2 1/2	1/1	2/2 1/2	2/2 1/1	

* Fractions represent numbers of genera/species

**Coumarins other than Ellagic Acid

A list of plants (by genera) tested by wr

(see Appendix I)

	· · · · · · · · · · · · · · · · · · ·	
	Hot Water and Syrin- Cig. test gin	Raph- ides Ehrlich
	I II III IV or + -	
RUTACEAE		
I. Rutoideae		
<u>Choisya</u> (7)		
Evodia (120)	1 1 2NR	-2 1
<u>Geijera</u> (7)	1 1NR	-1 1M
Melicope (50)	1 INR	-1 1
Orixa (1) Zanthoxylum (15)	2 1NR 2 2NR	-1 -2
Cneoridium (1)	2 ZNR 1 IR	1M
Dictamnus (2)	III?-IV-1 INR	-1 1
$\frac{D_{1}}{Ruta}$ (60)	3 3NR	-3 2
Acradenia (1)	1 1?R 5NR	-1 IM
Boronia (20)	1 2? 1R	-5 5M 3
Correa (11)	1 1?R 1NR	- l lm
<u>Chorilaena</u> (3)	1 INR	-1 1M
Crowea (4)	1 1NR	-1 1M
Diplolaena (8)	1? 1NR	-1 1
Eriostemon (30)	1 2NR	-2 2M -1 1M
Nematolepsis (2) Phebalium (36)	1 1NR 1 1? 1R	
Barosma (20)		-1 1M -1 1M
Calodendron (2)	1 1NR 1 1R	-1 1
Coleonema (6)	1 2NR	-2 -2 <u>2</u> M
Diosma (15)		
<u>Pilocarpus</u> (20)	1 1NR	-1, 1
Galipea (8)		+1.
Erythrochiton(5)	III-IV-1 1	+1 1M
Raputia (5) II. Dictyolomatoidea		+1
III. Flindersioideae	8	ŧ
Flindersia (20)	I-II- I lR	-1. 1M
IV. Spathelioideae		
V. Toddalioideae		
<u>Acronychia</u> (40)	II-III-1 1 1NR	
Amyris (20)		•
Casimiroa (60	I-II-l INR	-1 1
Halfordia (4)	1	lm
Key to symbols:	NR = no red $Sd = seed$	1
	$R = red \qquad Bk = bark$	
	M = magenta Rt = root	:
	PY = pale yellow Ot = other Y = yellow Tr = trace	÷
	Y = yellow Tr = trace	

15.

1 ab - 6-1 : 11 4

lich	н +	CN _	HC1/ +	'Meth	ь. +	A.	Juglone + -	Flu +	ores.	Tan +	nin	Sar Tes +	oonin st A	"Sar +). Test (NH3) -	B"
1 1M 1 2 1M 1M 1M 1M 1M 1M 1M 1M 1M	3	2211 113132121112112	1 1? 1	2 115 13 21 112 11 12 11 2	1 1 1 4 2 1 1	1 1 1 1 3 1 1 1 2	2 1PY 1 2/3Y 1 1/3PY 4 1 1 1 1 1 1 1 1 2	3 1 3 1 3 4 1 1 1 1 1 1 2	1	1 1(1 1 1 2 1 2 1 2	1 (r) 1 3 1	· · · · ·	1 1 3 1	1	1 3	
1		1		1		1	ly	1		1			1		1	
1 <u>M</u>				1	1		1	1		1?						
lm		2	1		1		1	1		1			1		1	
		2	2	1									1			
l lm		1 1		1 1		1	1	1		1		1		1		

		Hot Water and Cig. test I II III	. IV	Syr g: or +	in- in -	Raph- ides	Ehrlic + -
RUTACEAE (cont'd.	.)						
Oricia	໌ (8)						•
Phellodendron	(10)	3 II-III-2		1?NR	2NR	-4	3
Ptelea	(3)		1		1NR	-1	-
Skimmia	(10)		ī	1?	. 1NR	-1	· 1
Teclea	(25)		1		1R	-1	lm
Toddalia	(1)						
VI.Citroideae	• •						
Aegle	(1)	1 II-III-1			1 NR	-1	lm
Atalantia	(30)	II-III-1	1		1nr	-1	1
Citrus	(60)		8		6NR	-6	6
Clausena	(30)	I-II-III-III-1			2 R	-2	2
Eremocitrus	(1)						
Feronia	(1)	II-III-l			1NR	-1	1M
Fortunella	(6)		1		1nr	-1	1
Glycosmis	(40)		1		1NR	-1	1
Limonia	(1)						
Murraya	(9)	I-II-1	2		$\begin{cases} 1R?\\ 1NR \end{cases}$	-2	3
Poncirus	(1)				INR	-1	
Triphasia	(2)		1	•	1NR	-1	1
VII. Rhabdodendroid	leae						
<u>Rhabdodendron</u> CNEORACEAE	(2)				1nr	-1	lm
Cneorum	(2)	III-1	1-77		1.NR	-1	1
SIMAROUBACEAE	(/				7.017		*
I.Surianoideae							
Suriana	(1)	1		•	1R	-1	lM
II. Simarouboideae	·/	_				-	
Ailanthus	(10)	II-III-1	1		1NR	-1	1
Hannoa	(4)		_		1NR	-ī	-
Picraena	(2)					-	
Picrasma	(17)		1		1NR	-1	1
III. Kirkioideae	、 — · <i>)</i>		-			-	-
Kirkia	(5)		1		1NR	-1	1
IV. Irvingioideae	• • •					_	-
Irvingia	(5)						
V. Picramnioideae		-			-	_	-
<u>Picramnia</u> FICRODENDRACEAE	(40)	1			1nr	-1	1
Picrodendron BURSERACEAE	(3)		1		1 R	-1	lm
	(100)	1-					
Bursera	(100)	1?					

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yri gi +		Raph- ides	Ehrlich + -	HC +	н_	, HC1, +	Meth		A.	Juglone + -	Fluc +	ores.	Tai +	nnin
-						·							······	·····
NR	2NR	-4	3	•	2	1	3	1?	2	2/3PY		3	2	
	1NR	-1	5				3 1? 2	.	ĩ		1	5	2 1	
	. 1NR	-1	· 1	1	1		2		2	1 1	ī		-	2
	1R	-1	1 <u>M</u> .		1 1	1		1?		1	1		lTr	
	ï NR	-1	1 M 1T	п?			1 1 ·	1		1Y	l		1	
	1nr	-1	1		1				1	1		1	_	1
	6NR	-6	6		1 6 2	2? 2	4	1 1	3	2/6Y	5 1?	2	1 2	1 1 5?
	2R	-2	2		2 1	2		1	1?	1/2Y	1?	1		•••
	1NR	-1	1 M	•	- 1		1 1	1		1	1		1	
	$1 \mathrm{NR}$	-1	1 1	1 ?			1		1	1Y		1 1		1
	1nr	-1	1		1	1	•		1	1		1		1
	$\left\{ \begin{array}{c} 1 \\ 1 \\ 1 \\ N \\ R \end{array} \right\}$	-2	3	J.	3		3		3	2/3Y	2 1	1	1	2
	⁻ lnr	-1			3 1 1		_			. 1	1	_		_
	1NR	-1	1		1		1		1	14		1		1
	1nr	-1	lm	ł	1							1		
	1nr	-1	1		1		1		1	ly		1		
	lr	-1	lm		1	1			1	1		1		
	1NR	-1	1		1		1		1	1	1			
	1NR	-1	-		-		ī		ī	ī	1 1		1	
	1NR	-1	1				1		1	1	1			
	1NR	-1	1		1				1	1	1		1	
			-		(1 Sc	i.)								
	1nr	-1	1		1	-	1		1	l "carmino	e"	1	1	
	lR	-1	lm		1	1				lpy				
										-		•		
				ł	1	2				2		2		

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				معريدة المراقف					· · · · · · · · · · · ·		
L/Meth	L. +	A.	Juglone + -	Fluc +	ores.	Tai +	nnin -	Saponi Test	.n "Sa A	np. Test (NH3)	. B "
3 1? 1 2	1?	2 1 2	2/3PY 1 1 1	1 1 1	3	2 1 lTr	2	3		3 1	
1 1 • 4	1 1 1	1 3 1?	1Y 1 2/6Y 1/2Y	1 5 1?	1 2 1	1 1 2	1 1 5?	1 4 1	?		
1 1	1	1 1	1 1Y 1	1	1 1	1	1 1	1	1	1	
3		3	2/3Y 1	2 1	1	1	2	2		2	
1		1	1Y	T	1		1	1		1	s. T
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1		1	ly		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	l "carmine lPY		1	1		1?	"cari	mine"	
			2		2					•	

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		Hot water and Cig. test	· · · · ·	Syrin- Raj gin ide
		I II III	IV or	+ -
<u>Commiphora</u> Pachylobus	(100) (21)	II-III-1		lnr lr - lnr -
MELIACEAE I. Cedreloideae				
<u>Cedrelea</u> Toona	(7) (15)	III-IV-1	1	1R -
<u>Ptaeroxylon</u> II. Swietenoideae	(1)	1		lnr -
Entandrophragma Khaya	(20) (10)		1	1R -
<u>Swietenia</u> III. Melioideae	(5)	I-II		1R -
<u>Carapa</u> Cipadessa	(15) (4)	2	1	2r - 1nr -
<u>Melia</u> Dysoxylum	(9) (100)	III-I ⊽-1 2	, 1 , · ·	lnr -
<u>Owenia</u> <u>Sandoricum</u> <u>Trichilia</u>	(5) (10) (20)			
AKANIACEAE		_		
<u>Akania</u>	(1)	1		lR -
MALPIGHIACEAE <u>Tristellateia</u> <u>Acridocarpus</u> <u>Gaudichaudia</u> Heteropterys	(22) (20) (15) (90)	1-11-1	1? 1 1?	lnr · lr · lnr · 2nr ·
Hiptage Malpighia Byrsonima Galphimia	(25) (30) (105) (12)	III-IV-1	1	4R 1R
Stigmaphyllum Thryallis TRIGONTACEAE	(12)		1	1R · . 2R ·
VOCHYSIACEAE <u>Vochysia</u> <u>Salvertia</u>	(100) (1)			1?R
TREMANDRACEAE				
<u>Platytheca</u> Tetratheca	(1) (25)		4	lnr 3r
POLYGALACEAE I. Xanthophylleae II. Polygaleae				
<u>Polygala</u> <u>Securidaca</u> Bredemeyera	(500) (30) (60)	III-IV-1	2	5nr
III. Moutabeae	(1)	II-III-l		1NR

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Tab-6-3

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IV or	Syrin- gin + -	Raph- ides	Ehrlich + -	HCI +	N _	HCl, +	/Meth	L +	• A• –	Juglon + -	€ F +
	lnR lR lNR	-2 -1			1	1			1	2	2
1	lr	-1	lm		1	1 1		1	1	1	1
	lnr	-1	1		1				1	1	1
1	lr	-1	lm		1 1	1		1		1	1 1:
1	lR 2R lNR lNR	-1 -2 -1 -1	1M 2M 1M 1 1	lTr	1 1 1 2 1	1 2 1 2 2	1	1 2 1	1	1 2 1 1 1	1 2 1 1 1
	lr	-1	lm	1?		1		1		1	1
1? 1 1? 1	lNR lR lNR 2NR 4R lR 1R 2R	-1 -1	1M 1M 1 3M 1M 2M	2?	1 1 3 2 2	1 1 3 1 2 3 1	1	1 1 3 2	1	1 1 1 3 1 3 1	1
	1?R	-1				1				1	
4	lnr 3r	-1 -4	1 2м	1 1	3	1 4				1	1
2	5 NF	4 -1	4	lTr.	3		3 1		2	1/ 1Y	3¥ 1
	lnf	t −1	M		1	1		1		1	1

				·		2003) 1992 1993				2			
/Meth	L.	.A.	Jug] +	Lone	Flu +	ores.	Tai +	nnin -	Saj Te: +	ponin st A _	"Sap (NH +	. Test	B"
		1	• .	2	2		1						
	1	1		1	1		1T)	c. ·					
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	2			1 3 1	1	3 1	1 2			2	2		
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				1	1		1? 1?						
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-	1		1		1?		11	?	2 (P]	lt)		1917	

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			Simple Coumarin	6,7-Furo- Coumarin	7,8-Furo- Coumarin	Chromano- Coumarin
RUTAC	EAE					
I.	Rutoideae					
	Evodia	(120)	1/1	2/1		
	Fagara	(200)	3/5	2/2		2/2
	Geijera	(17)	3/2	•		
	Melicope	(50)		1/1		1/1
	Zanthoxylum	(15)	5/3	3/3		3/1
	Cneoridium	(1)	1/1	3/1		
	Dictamnus	(2)	1/1	2/1		
	Ruta	(60 ⁻)	9/5	9/4		2/2
	Thamnosma	(6)			1/1	
	Phebalium	(36)		3/1		
III.	Flindersioide					
	<u>Flindersia</u>	(20)	3/3	1/1		2/2
	Chloroxylon	(1)	1/1			2/1
V.	Toddalioideae					
	<u>Casimiroa</u>	(6)	1/1	4/1		•
	Halfordia	(4)		2/2		1/1
	Helietta	(1)	- •	1/1		
	Ptelea	(3)	1/1	9/2		- •-
	Skimmia	(10)	3/3	2/2		1/2
	Toddalia	(1)	3/1			
VI.	Citroideae					••
	<u>Aegle</u>	(1)	5/1	4/1		
	Aeglopsis	(5)	1/1	3/1		
	Citrus	(60)	10/7	11/7		1/1
	Clausena	(30)		1/1		4/2
	Limonia	(1)		- 1-		1/1
	Luvunga	(12)	1/1	2/1		
	Micromelum	(10)	1/1			
	Murraya	(9)	4/3	o /2		
	Poncirus	(1)	7 /7	3/1		o /1
	Severina	(1)	1/1	3/1		2/1
MELIA	CEAE					
I.	Cedreloideae					
	Cedrelopsis	(7)	1/1			
	Ptaeroxylon	(1)	1/1		2/1	2/1
III.	Melioideae		-			-
		1765	7 /7			

TABLE 7Coumarins of "Rutales" (see Appendix III)*

* Fractions represent numbers of compounds/species

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<u></u>		1		·····	 l	ß	
		Anthano- cyanins	Leuco- antho- cyanins	Flavan- ones	Flavano- nols	Flavones	Flavonols
		ran	Leuco- antho- cyanin	laval ones	ol	av	av
		ςγ	c y n c y		н с н	4 1	
		I	II	III	IV	V	VI
RUTACEAE							
I. Rutoideae <u>Choisya</u>	(7)		-				2/1
Evodia	(120)		2/1				3/1
Fagara	(200)			1/1		1/2	
<u>Melicope</u> Zanthoxylum	(50) (15)		1 /2	1/4			7/3
Boenninghausen			1/2	1/4			3/5 1/1
Ruta	(60)						$\frac{1}{2}/2$
Boronia	(60)		1/1				3/1
<u>Correa</u> Crowea	(11) (4)		1/1 2/1 2/1				3/5 1/1 2/2 3/1 3/1 3/1
Eriostemon	(30)		2/1 _				-
Coleonema	(6)		_ 1/1				3/1
III. Flindersioideae			·				
<u>Flindersia</u> V. Toddalioideae	(20)						1/1
Phellodendron	(10)		-		1/1		2/2
Ptelea	(3)		-		•		2/2 2/2
Skimmia	(10)		-			a /1	
<u>Teclea</u> Casimiroa	(25) (6)					4/1 6/1	
7. Citroideae	(0)					0/1	
Citrus	(60)	2/1	-	14/16		13/15	5/2
Glycosmia	(40)		-			7 /7	-
<u>Murraya</u> Poncirus	(9) (1)		-	4/1		$\frac{1}{1}$	-
Fortunella	(6)			7/1		1/1 1/3	
NEORACEAE						,	
Cneorum	(2)		-				2/1
SIMAROUBACEAE Quassia	(40)		_				_
Ailanthus	(10)		1/1?				2/3
URSERACEAE							
<u>Protium</u> ÆLIACEAE	(60)		1/2				2/2
Cedrea	(7)		1/2				2/2
Ptaeroxylon	(1)		-, -				1/1
Melia	(9)		~				2/1 1/1
Aitonia	(1)		-				1/1
MALPIGHIACEAE Heteropteris	(9 0)		-				2/2
Hiptage	(25)		1/1				-/- -
Malpighia	(30)		1/1 1/1 1/1				_ 1/1
Tristellatlia	(22)		1/1				1/1
REMANDRACEAE Platytheca	(1)		~				2/2
Tetratheca	(25)		-				1/1
							, 2/5
POLYGALACEAE <u>Polygala</u>	(500)						2/5

Flavonoids of "Rutales" (see Appendix IV and IX)*

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*Fractions represent numbers of compounds/species.

TABLE 8

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		T	erpend	oids of	"Rutal	es" (see A	ppend:	ix V)*			
	I. Monoterpenoids	II. Sesquiterpenoids	l. Bsiaboiene Group	2. Cadienene Group	3. Eremophilone Group	4. Guaianolide Group	5. Selinene Group	III. Diterpenoids	IV. Triterpenoids	 Triterpenoid saponins and sapo- genins 	 Triterpenoids other than Saponins and Sapogenins 	V. Tetraterpenoids
RUTACEAE I. Rutoideae Evodia (120) Zanthoxylum (15) Medicosma (1) Dictamnus (2) Ruta (60) Boronia (60) Eriostemon (30) Phebalium (36) Zieria (15) Agathosma (170)	1/1 7/3 1/1 3/1 +2/2 +2/2 +2/1 +1/1	+1/1 +1/1 +1/1				+1/1 +1/1 +1/1			+3/3 +2/4 +3/1	+2/4	+3/3 +3/1	
Barosma(20)Calodendrum(2)Empleurum(2)III. FlindersioideaeFlindersia(20)V. ToddalioideaeAcronychia(40)Amyris(20)Casimiroa(6)Phellodendron(10)Skimmia(10)Vepris(20)	+5/4 +1/1 +1/2 +1/1	+1/1		+1/1					+2/1 +3/4 +1/1 +2/2 +2/1 +2/1 +1/1	+1/1 +1/1	+2/1 +2/3 +1/1 +2/2 +2/1 +2/1	
VI. Citroideae <u>Aegle</u> (1) <u>Atalantia</u> (20) <u>Citrus</u> (60) <u>Clausena</u> (30) <u>Fortunella</u> (6) <u>Glycosmia</u> (40) <u>Luvunga</u> (12) <u>Microcitrus</u> (5) <u>Murraya</u> (9) <u>Poncirus</u> (1) <u>Triphasia</u> (2)	+4/1 +3/2 +25/10 +1/1 +1/1		+1/1 +1/1	+2/1	+2/2		+1/1		+1/1 +9/14 +2/1 +2/1 +1/1 +2/1 +4/1	+1/1 +2/2	+7/13 +2/1 +2/1 +1/1 +2/1 +4/1	+5/3 +2/1 +2/1
SIMAROUBACEAE II. Simarouboideae <u>Eurycoma</u> (4) <u>Quassia</u> <u>Simaba</u> <u>Simarouba</u> <u>Samadera</u> <u>Ailanthus</u> (10) <u>Brucea</u> (10-12 <u>Castela</u> (12) <u>Perriera</u> (1))								+1/1 +2/1 +5/2 +4/1 +8/4 +1/1 +1/1		+1/1 +2/1 +5/2 +4/1 +8/4 +1/1 +1/1	
BURSERACEAE Boswellia (24) Bursera (100) Commiphora (100) Canarium (75)	+5/1 +8/2 +1/1 +2/1	+1/1 +4/2	+1/1				+4/2	+1/1 +1/1	+1/2 +1 /1 +8/4	+ 2/ 1	+1/2 +1/1 +6/4	
MELIACEAE I.Cedreloideae <u>Cedrela</u> (7) <u>Ptaeroxylon</u> (1) II.Swietenoideae <u>Entandrophregma</u> (2 <u>Khaya</u> (10) <u>Pseudocedrela</u> (1) <u>Swietenia</u> (5)	0)	+2/1		+2/1				+2/1	+11/ -1/5 +1/1 +6/9 +16/5 +5/1 +3/1	+1/1	+11/ -1/5 +6/9 +16/5 +5/1 +3/1	
III.Melioideae <u>Carapa</u> (15) Xylocarpus (5) <u>Turraeanthus</u> (6) Melia (9)		+1/1 +1/1	• • •	+1/1	+1/	′1		+2/1	+3/1 +6/2 +1/1 +1/1 +11/2 +1/1	. .	+3/1 +6/2 +1/1 +1/1 +1/1 +11/2 +1/1	
Guarea (160)		• -/ -		₩⊥ /⊥					+6/2		+6/2	

TABLE 9

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· · ·	I. Monoterpenoids	II. Sesquiterpenoid	l. Bsiaboiene Grou	2. Cadienene Group	3. Eremophilone Gr	4. Guaianolide Gro	5. Selinene Group	III. Diterpenoids	IV. Triterpenoids	l. Triterpenoid saponins and s genins	 Triterpenoids than Saponins Sapogenins 	V. Tetraterpenoid
RUTACEAE		н 					<u></u>	<u> </u>				
I. Rutoideae Evodia (120)	1/1								+3/3 +2/4	+2/4	+3/3	
Zanthoxylum (15) Medicosma (1)	7/3 1/1								+3/1	12/1	+3/1	
Dictamnus (2) Ruta (60) Boronia (60) Eriostemon (30) Phebalium (36) Zieria (15) Agathosma (170) Barosma (20)	3/1 1/1 +2/2 +2/2 +2/1 +1/1 +5/4	+1/1 +1/1 +1/1				+1/1 +1/1 +1/1					. 2 / 2	
Calodendrum (2) Empleurum (2)	+1/1								+2/1		+2/1	
III. Flindersioideae <u>Flindersia</u> (20)									+3/4	+1/1	+2/3	
V. Toddalioideae <u>Acronychia</u> (40) Amyris (20)		+1/1		+1/1					+1/1		+1/1	
VI. Citroideae	+1/2 +1/1			·					+2/2 +2/1 +2/1 +1/1		+2/2 +2/1 +2/1	
<u>Aegle</u> (1)	+4/1 +3/2	• •			-				+1/1	+1/1	•	
<u>Atalantia</u> (20) <u>Citrus</u> (60) Clausena (30)	+3/2 +25/10 +1/1	+4/2	+1/1		+2/2		+1/1		+9/1	4 +2/2	+7/13	+5/3
Fortunella (6) Glycosmia (40) Luvunga (12) Microcitrus (5)	+1/1		. 1 /1	. 0. /1					+2/1 +2/1 +1/1 +2/1		+2/1 +2/1 +1/1 +2/1	+2/1
<u>Murraya</u> (9) <u>Poncirus</u> (1) <u>Triphasia</u> (2)		+3/2	+1/1	+2/1					+4/1		+4/1	+2/1
SIMAROUBACEAE II. Simarouboideae <u>Eurycoma</u> (4) <u>Quassia</u> <u>Simaba</u> <u>Simaba</u> <u>Simarouba</u> (40) <u>Simarouba</u> <u>Ailanthus</u> (10) <u>Brucea</u> (10-1: <u>Castela</u> (12) <u>Perriera</u> (1)	2)								+1/1 +2/1 +5/2 +4/1 +8/4 +1/1 +1/1		+1/1 +2/1 +5/2 +4/1 +8/4 +1/1 +1/1	
BURSERACEAE Boswellia (24)	+5/1							+1/1	+1/2		+1/2	
	+1/1		+1/1				+4/2	+1/1	+1/1 +8/4		+1/1 +6/4	
<u>Canarium</u> (75) MELIACEAE	+2/1	+4/2					+++/2		. 07-4	· •/ ±	, - <u>.</u>	
I.Cedreloideae <u>Cedrela</u> (7) <u>Ptaeroxylon</u> (1) II.Swietenoideae		+2/1		+2/1				+2/1	+11 -1/5 +1/1		+11/ -1/5	
Entandrophregma (10) Khaya (10) Pseudocedrela (1 Swietenia (5)								+6/9 +16/ +5/1 +3/1	5	+6/9 +16/5 +5/1 +3/1	
III.Melioideae <u>Carapa</u> (15 Xylocarpus (5									+6/2 +1/1		+6/2 +1/1	
Turraeanthus (6))	+1/1			+1	./1		+2/1	+1/1 +11/ +1/1	2	+1/1 +11/2 +1/1	
Dysoxylum (100) Guarea (160)		+1/1	• • • • ·	+1/1					+6:/2		+6/2	
<u>Lansium</u> (7) <u>Trichilia</u> (230		+2/1	+1/1		+1/1				+1/1 +6/4		+1/1 +6/4	
POLYGALACEAE Xanthophyllum(40) Monnina (80) Polygala (500)					Vancaio				+1/1 +1/1 +8/4		+1/1 +1/1 +8/4	

*Fraction represent numbers of compounds/species

<u>9</u>]	•		91				
	· · · · · · · · · · · · · · · · · · ·		I. Acridine Group	II. Alkaloid amines	III. Imidazole Group	IV. Indole Group	l. Simple indole base
	RUTACEAE I. Rutoideae <u>Choisya</u> <u>Evodia</u> Fagara <u>Geijera</u> <u>Medicosma</u> <u>Melicope</u> <u>Orixa</u>	(7) (120) (200) (7) (1) (50)	9/2 4/1	2/6		4/1	
	Platydesma Pentaceras Balfourodendron Lunasia Xanthoxylum Boenninghausenia Dictamnus Ruta	(1) (3) (1) (10) (15) (1) (2) (60)	1/1 1/1	1/1		3/1 3/3	
-10 tt	Thamnosma Haplophyllum Boronia Eriostemon Phebalium Geleznowia Cusparia Galipea Pilocarpus Ravenia	(6) (70) (60) (30) (36) (36) (3) (25) (8) (20) (18)	1/1	1/1	4/4		
9-10	II.Dictyolomatoideae Dictyoloma	(2)				1/1	1/1

Tab-10 - 2.24

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				1	1							
1/1 1/1	3/3	3/1	4/1	IV. Indole Group 1. Simple indole base								
 • • • • • • • • • • • • • • • • • • •				2.Carboline alkaloids								
	2/1		4/1	3. The Quinazoline carboline								
	1/2	3/1	•	4. Canthin-6-ones								
		•	•	5. The Carbazole group								
7	20/15		1/1 19/16	V. Isoquinoline group	Alkaloids							
	1/1		1/6	1. 1,1-Benzyliso- quinolines	oids of							
	6/8		8/6	2. The Aporphine group	TABLE 10 "Rutales"							
	4/5		1/1 2/1	3. Protoberberine								
	3/2		3/1	4. Protopine group	(see App							
TABLE		see Ap	opendix VI)*		· .	22				·		
---------------------------	-------------------	--------------------	--	-----------------------	----------------	--------------------	------------------	---	--	---	-------------------------------	--
2. The Aporphine group	3. Protoberberine	4. Protopine group	 Phthalide- isoguinalines The x-Naphtha- phenanthridines 	VI. The Oxazole group	VII. Pyridines	VIII. Pyrrolidines	IX. Quinazolines	X. Quinoline group	1. Simple Quinolones	2. Furoquino- lines	3. Quinolyl- quinuclidines	
9/8	1/1 2/1	3/1	4/6				•	4/1 9/5 4/4 4/1 1/1 2/1 8/1 6/1	1/1 2/2 1/1	4/1 8/4 2/2 4/1 1/1 2/1 8/1 5/1	:	
6/8	4/5	3/2	6/8		1/1			13/1 19/4 4/3 1/1 3/2 7/1 2/1 14/9 2/1 2/5 5/1 1/1 2/2 13/3 1/1	1/1 5/2 3/1 2/2 1/1 1/1 13/3	12/1 14/4 4/3 1/1 3/2 4/1 2/1 12/9 1/1 2/5 5/1 1/1 1/2 1/1		
		·										

	TABLE 10 (cont'd.)		92					
		I. Acridine	II. Alkaloid amines	III. Imidazole group	IV. Indole group	l. Simple indole base	2. Carboline alkaloids	3. The Quinaz oline carboline
]	RUTACEAE III. Flindersioideae Chloroxylen (1) <u>Flindersia</u> (20) V. Toddalioideae <u>Acronychia</u> (40)	7/3			~ <u>~</u>		/ <u></u>	
	Casimiroa(6)Phellodendron(3)Ptelea(3)Skimmia(10)Teclea(25)	2/2	1/1 1/1	3/1				: - -
	Toddalia(1)Vepris(20)Halfordia(1)Hortia(10)VI. Citroideae(10)Agele(1)		1/1		4/2			4/2
	Citrus(60)Clausena(30)Glycosmis(40)Murraya(9)Poncirus(1)	1/1	1/1 8/5 1/1		2/2 1/1 2/1 2/1	2/2		
	SIMAROUBACEAE II.Simarouboideae <u>Picrasma</u> (17) <u>Picrolemma</u> (3) MALPIGHIACEAE				3/2			•
	Banisteria (75) Cabi (1) Banisteriopsis(100)			3/1 1/1 3/3	2/2	3/1 1/1 1/1	

*Fraction represent numbers of compounds/species.

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cies.	3/1 1/1 3/3	3/2	2/1 2/1 2/1	4/2			[IV. Indole group
	2/2		2/2					l. Simple indole base
	3/1 1/1 1/1							2. Carboline alkaloids
	 .	· · · •		4/2				3. The Quinaz- oline carboline
		3/2						4. Canthin-6-ones
			1 2/1 2/1					5. The Carbazole group
			. 1/2		5/1	5/1		V. Isoquinoline group
						14		1. Isoquinoline group
						1/1		2. The Aporphone group
					1/1	4/1		3. Protoberberine
								4.
			1/2					5. Phthalide- isoquinalines
		·			4/1			6. The x-Naphtha- phenanthridines ,
				4/1				VI. The Oxazole group
								VII. Pyridines

		1. Isoquinoline group
	5	2. The Aporphone group
	4/1 1/1	3. Protoberberine
		4.
	1/2	5. Phthalide- isoquinalines
	4 /1	6. The x-Naphtha- phenanthridines ,
	4/1	VI. The Oxazole group
		VII. Pyridines
	5	VIII. Pyrrolidines
	2/1 4/1	IX. Quinazolines
1/1	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	X. Quinoline group
	1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	l Simple Quinoloneș
Ĭ/I	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	2. Furoquinolines Quinolyl- 3. quinuclidines

DISCUSSION

A. The individual families of "Rutales"

1. Rutaceae

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Many chemotaxonomists and phytochemists have intensively investigated the various chemical constituents which may prove useful for taxonomic study of the <u>Rutaceae</u> (e.g. Dreyer's series of "Chemotaxonomy of Rutaceae" (1966-1969)). However, there is still some disagreement as to which genera should belong to this family and which sub-families should be raised to familial rank. Let us first of all discuss the general distribution of substances within the family.

<u>Cigarette and Hot-Water Test</u> -- I-IV. <u>Polyphenolases</u>, as judged by this test, seem to be absent in most species tested but a few are found randomly present among all four sub-families species.

<u>Syringin</u> -- as indicated by the <u>Syringin (1:1 H₂SO₄/H₂O)</u> <u>Test</u> (p.64). Most members of this family are characterized by the absence of syringin. Only one member of the <u>Rutoideae</u> was recorded as being positive with syringin test. A few doubtfully positive species were found in the <u>Rutoideae</u>, <u>Toddalioideae</u> and <u>Citroideae</u> (<u>Aurantioideae</u>). Most of the species did not develop any red colour in the lignified tissue of the treated sections.

<u>Raphides</u> -- are probably absent from all the genera investigated except for a few members of the <u>Rutoideae</u> where

they are said to occur in the subtribe <u>Cusparineae</u>. These were not available to us.

The usual correlation of results of Ehrlich, HCl/Methanol, and Leucoanthocyanin tests (p.64) was inconsistent. Many species gave a negative HCl/Methanol result, but a magenta spot in the Ehrlich test, a positive Leucoanthocyanin result, and a non-red reaction in the syringin test (this could indicate presence of leucoanthocyanins in leaves but not in stems).

<u>Aucubin-like substances</u> -- were found to be completely absent from all the plants tested as judged by Ehrlich tests (p. 65)

<u>Cyanogenic glucosides</u> -- were usually absent, too, but one genus -- <u>Boronia</u> (<u>Rutoideae</u>) -- was recorded as having at least two species yielding HCN with HCN (Test A) (p. 70).

<u>Leucoanthocyanins</u> -- using L.A. (Test A) (p.68) our results were more often negative than positive. This is consistent with the findings of Bate-Smith (1957; 1962).

<u>HCl/Methanol test</u> -- results were largely <u>negative</u>. However, it was interesting to find that most of the <u>positive</u> results were correlated with positive Tannin tests; but the reverse is not always true, since tannins were found in some species, as Bate-Smith found (1957) which did not give a positive HCl/Methanol test.

<u>Naphthoquinones</u> -- as judged by the Juglone Test (p.71)-were absent, but most species showed blue fluorescence in the aqueous layer ("Juglone Test C"). These results are consistent with those found in the literature (Table 5 and 7) and with the results of paper chromato-

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graphy done in our laboratory (Table 4), i.e. <u>coumarins</u> (excluding <u>ellagic acid</u>) occurred in 20 genera of the plants analysed. It should be mentioned here, that although <u>ellagic</u> <u>acid</u> is structurally a <u>coumarin</u>, according to the biogenetic synthesis, it should be considered to be one of the benzoic acids.

<u>Saponin Test A</u> -- results, except for one recorded as doubtfully positive in <u>Casimiroa edulis</u> (<u>Toddalioideae</u>) and one doubtfully negative in <u>Clausena lansium</u> (<u>Citroideae</u>), were negative. However, Amarasingham <u>et al</u>. (1964) found several genera of the <u>Citroideae</u> (<u>Aurantoideae</u>) to contain saponins, and from Table 9, we know <u>Rutoideae</u>, <u>Flindersioideae</u>, and <u>Citroideae</u> to contain triterpenoid saponins and sapogenins.

<u>"Saponin Test B", (NH3)</u> -- results, from the limited data available, seem to correlate with tannin test A. This is consistent with Gibbs' (MSS) findings.

<u>Terpenoids</u> -- are, except for diterpenoids, widely distributed in <u>Rutaceae</u> (Table 9). To the best of my knowledge, the <u>limonoids</u> (triterpenoids) are characteristic of this family (see also Simaroubaceae, Burseraceae, and Meliaceae).

<u>Alkaloids</u> -- the <u>Rutaceae</u> are possibly the most versatile of all families from the point of view of alkaloid synthesis. At least ten structural classes of alkaloids have been found in <u>Rutaceae</u> (Table 10). <u>Acridine alkaloids</u> are prominent in the family, all but one being derivatives of <u>acridone</u> rather than

<u>acridine</u>. However, the most characteristic alkaloids are the <u>furoquinolines</u> such as <u>skimmiamine</u>, which have been found in 56 species of the 30 genera recorded (Appendix VI).

<u>Flavonoids</u> -- in <u>Rutaceae</u>, they show very beautifully the replacement of flavonol by flavone at the generic level (Table 8). There was one exception from <u>Citrus</u>, but only two out of 15 species have been surveyed (<u>Citrus limon</u> and <u>aurantium</u>) which contain both types of compound. Furthermore, in most cases, the flavonols were present together with leucoanthocyanins, and flavonanols; while flavones were present with flavanones. This suggests that they may have evolutionary significance in the family <u>Rutaceae</u>, as probably among the angiosperms generally (Bate-Smith, 1962; Harborne, 1967).

<u>Phenolic acids</u> -- were found to occur widely in <u>Rutaceae</u> (Appendix II). The major compounds included Group I: gentisic acid; Group II: <u>p</u>-coumaric, caffeic, ferulic and sinapic acids; Group IV: ellagic acid. However, this is not completely consistent with Bate-Smith's (1969) idea. He said:

"The occurrences of these phenolic acids, associated in the cases of caffeic and ellagic acid with the presence of flavonols and flavandiols, and in the cases of ferulic and sinapic acid with their absence, reinforce the indications provided by the flavonoid constituents (I-V) themselves regarding the evolutionary status of particular plants or families,".

The genera <u>Flindersia</u> and <u>Chloroxylon</u>, from a botanical viewpoint, have proved difficult for botanists to classify. Scholz (1964) included just these two genera in his third sub-

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family, <u>Flindersioideae</u>. Our results (Tables 3 and 5) show that there are three possibilities: the two genera may go into <u>Rutaceae</u>; they may go with <u>Cedrela</u>, etc. into <u>Meliaceae</u> (<u>Cedreloideae</u>); or they may constitute a family <u>Flindersiaceae</u>, intermediate between <u>Rutaceae</u> and <u>Meliaceae</u>.

In spite of the fact that Flindersia and Chloroxylon both contain the typical Rutaceous furoquinolines, they do not have the acridine alkaloids which are prominent in the Rutaceae, occurring widely in Rutoideae, Toddalioideae and Citroideae. Simple coumarins and chromano-coumarins, characteristic of the Rutaceae, are known to occur in these two genera, but they are also in three genera of the Meliaceae (Cedrelopsis, Ptaeroxylon and Ekebergia). Flavonoid data show that the Flindersioideae contain only flavonols, whereas the other Rutaceous sub-families (Rutoideae, Toddalioideae and Citroideae) have both flavones and flavonols. Flindissol, isolated from two species of Flindersia, is structurally midway between epoeuphol (a limonoid of the Rutaceae) and the meliacins of the Meliaceae. We feel, therefore, that the chemical evidence supports the placing of Chloroxylon and Flindersia in a small family Flindersiaceae between Rutaceae and Meliaceae.

Should <u>Zanthoxylum</u> and <u>Fagara</u> be maintained as distinct genera? The principal compounds so far isolated from these plants are very closely related <u>alkaloids</u>, <u>coumarins</u>, <u>terpenoids</u>, and even the phenolic aldehydes - <u>parvifloral</u> from (<u>Zanthoxylum</u>) and zanthoxylol (from Fagara). On the other hand, these two genera

have completely different types of <u>flavonoids</u>, <u>Fagara</u> has <u>flavone</u> and <u>Xanthoxylum</u> has <u>flavonols</u>. We suggest that these two genera are closely related but distinct.

Is the family <u>Rutaceae</u> homogeneous or heterogeneous? Paris and Etchepare (1968) concluded that it is heterogeneous for flavonoid pigments, since it contains not only <u>flavones</u> and <u>flavonones</u>, but also methylated derivatives, and some with hydroxymethyl groups and or isopreme chains. On the other hand, Price (1963) has concluded, from the distribution of <u>alkaloids</u> and <u>coumarins</u> in the <u>Rutaceae</u>, that the major sub-families constitute a highly homogeneous group. Later, this conclusion was supported by Dreyer (1966) who found that there was botanically uniform distribution of <u>limonoids</u> throughout the Rutaceae. He said:

"The uniformity is further emphasized by the fact that structural variation(s) of these limonoids that occur in the Rutaceae are rather slight (excepting flindissol which appears to occur only in a further removed botanical group) The homogeneity of the Rutaceae is attested to by the slight structural variation in the limonoids while the clear definition between subfamilies is attested to by the uniform difference in oxidation levels of their limonoids".

On the basis of the still limited data available, this family would be relatively homogeneous if the sub-family <u>Flindersioideae</u> were removed and made a family <u>Flindersiaceae</u>.

2. Cneoraceae

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This family (as we have seen) has been assigned to the <u>Rutales</u>, <u>Geraniales</u>, and <u>Sapindales</u>. It also has been made the type of an order <u>Cne</u>orales.

It has only three species belonging to two genera, and all our information is derived from but one of these -- <u>Cneorum</u> <u>tricoccon</u>.

<u>Cigarette and Hot-Water Tests</u> -- III-IV (negative?)

<u>Syringin</u> -- negative, no red in lignified tissue which agrees with the negative Ehrlich, HCl/Methanol and leucoanthocyanin tests.

Raphides -- absent.

Saponins -- absent.

<u>Naphthoquinones</u> -- presumably absent (negative with Juglone Test A), but blue fluorescence was observed. This is consistent with paper chromatographic results -- which showed that <u>scopoletin</u> is present.

<u>Phenolic Acids</u> -- the major phenolic acids are Group I: gentisic acid; Group II: <u>p</u>-coumaric, caffeic, ferulic and sinapic acids, but no ellagic acid.

<u>Flavonoids</u> -- the presence of flavonols but no flavone, has been reported.

3. <u>Simaroubaceae</u>

As mentioned in the review of literature, many authors have raised the sub-families and even some genera to familial rank. We may, therefore, expect it to be chemically heterogeneous.

<u>Polyphenolases</u> -- as judged by Cigarette and Hot Water Tests, occur in sub-families <u>Picramnioideae</u>, <u>Surianoideae</u>, and probably also in <u>Simarouboideae</u>, but <u>Kirkia acuminata</u> of the Kirkioideae gave a IV (negative) reaction with the hot water test. <u>Syringin</u> -- appears to be absent from the whole family. Only <u>Suriana</u> gave a red colour in the xylem. These results are consistent with those obtained from the Ehrlich, HCl/Methanol, and leucoanthocyanin tests.

<u>Raphides</u> -- no raphides were seen in the control sections of the syringin test, and none have been reported in the literature, but solitary and cluster crystals(Appendix XI) have been reported. The size and distribution of the cluster crystals in <u>Castela</u>, <u>Holacantha</u>, and <u>Picramnia</u> are said to be of value in the identification of the genera (Metcalfe and Chalk, 1950). Styloids have been reported in one genus -- <u>Alvaradoa</u>.

<u>Naphthoquinones</u> -- as judged by the Juglone test are absent. Blue fluorescence was noted in <u>Simarouboideae</u> and <u>Kirkioideae</u>, but no <u>coumarins</u> have been detected (ellagic acid is not fluorescent).

It was interesting to note, during the course of the Juglone test, that <u>Picramnia pentandra (Picramnioideae</u>) gave a "carmine" colour (Ridgway) in the ammonia layer under visible light. This may be due to the presence of a naphthoquinone, but, following the method of Chen and Bohm (1966), the unknown was compared with authentic samples of juglone, lapachol, lawsone, lematiol, 7-methyl-juglone, plumbagin and dunnione in different kinds of solvents. Unfortunately, this attempt to identify the compound proved unsuccessful.

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In "Saponin Test B" (NH₃), <u>Picramnia pentandra</u> also gave immediately a "carmine" colour reaction in the ammonia layer. Could the substance responsible be an aurone?

2,6-Dimethoxy-1,4-benzoquinones have been reported from genera of the <u>Simarouboideae</u> (<u>Ailanthus</u>, <u>Eurycoma</u>, <u>Picrasma</u> and <u>Quassia</u>.). Unfortunately, we have no records from these genera with the Juglone test comparing with genus <u>Picramnia</u>. A specimen of 2,6-dimethoxy-1,4-benzoquinone (from Dr. R. G. Cooke) gave a bluish colour changing to yellow in Juglone Test B.

<u>Tannin tests</u> -- tannins were present in <u>Kirkioideae</u> and <u>Picramnioideae</u>, and this is consistent with the results of "Saponin Test B" (NH₃).

<u>Saponins</u> -- were recorded as probably present in <u>Picramnia</u> pentandra.

<u>Phenolic acids</u> -- the major phenolic acids were: Group I: gallic acid and Group IV: ellagic acid. Compounds of Group III --<u>p</u>-coumaric, caffeic and ferulic acids were identified in <u>Ailanthus</u> but not in <u>Kirkia.</u>

<u>Flavonoids</u> -- <u>Ailanthus</u> is reported to have flavonols, too. This result is not consistent with the conclusions of Bate-Smith (1969).

<u>Triterpenoids</u> -- have been reported from several genera of this family, especially in the sub-family <u>Simarouboideae</u>. <u>Simarolide</u> is an acetate of a C₂₅ compound and its occurrence in the <u>Simaroubaceae</u> (<u>Simarouba amara</u>) may be of biogenetic significance, since the other bitter substances (C₂₀ compounds)

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of this family (quassin, chaparrin, glaucarubin. ... etc.) are closely related. Polonsky (1964) suggested that the biogenetic precursor of <u>simarolide</u> may be a tetracyclic triterpene of the tirucallol or possibly of butyrospermol (A-euphol) type, and that its biogenesis might then follow the path proposed for <u>limonin</u> and lead later to the basic skeleton of <u>simarolide</u>. Therefore, <u>Simaroubaceae</u> may have affinity with <u>Rutaceae</u> in this context.

<u>Alkaloids</u> -- have been reported in 50% of the species tested (Appendix VIII), but only two classes of alkaloids, <u>indole</u> and <u>quinoline</u>, are known to occur in this family.

<u>Fatty Acids</u> -- 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 composition of their seed fats as to prompt further inquiry into their botanical classification".

We have the following:

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Genus	Chief Fatty Acids
Ailanthus	Oleic, linoleic
Picrasma	petroselinic
Picramnia	tariric
Irvingia	myristic, lauric

These four genera are distributed among three sub-families. Each contains a different major fatty acid. Chemical evidence would seem to support the view, that the <u>Picramnioideae</u> should be excluded from the <u>Simaroubaceae</u>. The presence of <u>myristic</u> and <u>lauric acids</u> in <u>Irvingia</u> and other genera, links this family with <u>Vochysiaceae</u>.

4. Picrodendraceae

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This family is treated by Scholz as having <u>Picrodendron</u> only with 3 spp. Airy Shaw (in Willis, 1966) includes 5 other genera from the <u>Euphorbiaceae</u> of which we know nothing.

The information below comes from <u>Picrodendeon</u> <u>baccatum</u> only (Gibbs, MSS).

> <u>Cigarette & H.W. Tests</u> -- IV <u>Syringin--</u> -ve (red in xylem; no raphides). <u>HCl/Methanol_Test</u> -- positive (2-3) consistent with

red in xylem in syringin test (above) and magenta in Ehrlich test (below).

<u>Ehrlich Test</u> -- negative (Ehrlich spot magenta) <u>Juglone Tests</u> -- -ve, but with blue fluorescence. <u>Cyanogenic Glycosides</u> -- absent? (negative result with <u>HCN Test A</u>).

Tannins, flavonoids, alkaloids and saponins <u>may</u> be absent (Table 2).

5. Burseraceae

This family is said to have affinities with <u>Anacardiaceae</u>, <u>Meliaceae</u>, <u>Rutaceae</u>, and <u>Simaroubaceae</u>.

Few members of this family were available to us for testing.

Cigarette and Hot-Water Tests -- II-III

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Syringin (1:1 H_2SO_4/H_2O) Test -- Syringin appears to be absent, red color was seen in xylem of Commiphora trothai.

<u>Raphides</u> -- No raphides were seen, nor have they been recorded by others; however, solitary and cluster crystals were found in this family by Metcalfe and Chalk (1950).

<u>Juglone Tests</u> -- negative results from Juglone Test A suggest that naphthoquinones are absent. Blue fluorescence was observed in Commiphora and Pachylobus.

<u>HCN Test A</u> -- results negative, suggesting absence of cyanogenic glycosides.

<u>Tannin Test A</u> -- the leaf of <u>Commiphora merkeri</u> gave a strong positive result (+++) with tannin test A.

<u>Phenolic acids</u> -- members of this family are known to contain gallic, <u>p</u>-coumaric, caffeic, and ellagic acids. According to Bate-Smith (1969), these results might suggest that flavonols should be present in this family, and in fact <u>Protium</u> is reported to have flavonol (Bate-Smith, 1962). We have no other information on this point.

<u>Terpenoids</u> -- are widely present in this family. <u>Diter-</u> <u>penoids</u>, as in <u>Meliaceae</u>, are present. Acetates of triterpenes -derivatives of <u>euphone</u> and <u>elemane</u>, which occur in <u>Rutaceae</u>, <u>Meliaceae</u> and <u>Simaroubaceae</u>, are found in <u>Burseraceae</u>, too (Ponsinet et al., 1968).

Alkaloids -- have been reported in 20% of the plants tested.

The chemical evidence above, and the reported presence of stearic acid as a major fatty acid of the seeds, links this family with the Meliaceae.

6. Meliaceae

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As discussed in the review of literature there is no general agreement as to which genera should belong to this family, which sub-families should be raised to familial rank and which families it should be allied with. Hutchinson (1969) further complicated things by making this family the sole member of its order.

Cigarette and Hot-Water Tests -- variable, I-IV.

<u>Syringin test</u> -- only <u>negative</u> results were obtained. The majority of species showed some red colour in the xylem and/or fibres. These results are consistent with those obtained from the Ehrlich, HCl/Methanol, and leucoanthocyanin tests.

<u>Tannin Test</u> -- results were mostly positive and agreed with positive results of HCl/Methanol tests.

<u>"Saponin Test B" (NH3)</u> -- the few results were mostly consistent with the results of the tannin test.

Raphides -- appeared to be absent,

<u>Cyanogenic Glycosides</u>. except for <u>Dysoxylum fraseranum</u> (<u>Melioideae</u>), all species investigated contained no cyanogenic glycosides. <u>Saponin Test A</u> -- <u>Amarasingham et al</u>. (1964) found the majority to give a negative result, but <u>Chisocheton</u> and Dysoxylum of the Melioideae gave positive. tests.

<u>Juglone Tests</u> -- results were negative, but <u>blue fluores</u>-<u>cence</u> was observed. So far, however, <u>coumarins</u> are known only from three genera belonging to <u>Cedreloideae</u> (<u>Cedrelopsis</u> and <u>Ptaeroxylon</u>) and <u>Melioideae</u> (<u>Ekebergia</u>). These two sub-families, again, were reported to contain flavonols but no flavones.

<u>Terpenoids</u> -- <u>Diterpenoids</u>, to the best of my knowledge, have been isolated, in the <u>Rutales</u>, only from two genera of the <u>Burseraceae</u> and from <u>Cedrela</u> (<u>Cedreloideae</u>), <u>Melia</u> and <u>Aphanomyxis</u> (both of <u>Melioideae</u>). <u>Triterpenoids</u> occur here, as in the <u>Rutaceae</u>, <u>Simaroubaceae</u>, and <u>Burseraceae</u>. Dreyer (1966) wrote:

> "Limonoids have been found in genera belonging to each of the three subfamilies of Meliaceae but in this case there seems to be no general correlation between the limonoid structure and the botanical distribution within the family. ... The known compounds of this series in the Meliaceae show, in general, much wider structural variation than those in Rutaceae. These variants range from members whose position is fairly low on the biogenetic ladder, such as, gedunin, cedrelone, and anthothecol, to those with extensive structural alterations, for example, andirobin, swietenine, nimbin, mexicanolide and methyl angolensate".

However, recently, three structurally close relatives of flindissol -- aphanomixin, melianone and turraeanthin -- were isolated from the <u>Melioideae</u>. These seem to be useful as taxonomic markers on a sub-family level and perhaps even as phylogenetic markers for the whole family, as in <u>Rutaceae</u>.

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<u>Alkaloids</u> -- are said to be present in 40% of the plants tested (Li and Willaman, 1968).

The monotypic genus <u>Ptaeroxylon</u>, which has given great difficulty to systematists, has been variously placed in <u>Sapindaceae</u>, <u>Rutaceae</u> and <u>Meliaceae</u>, but it is now considered to form (with <u>Cedrelopsis</u>) a separate family, the <u>Ptaeroxylaceae</u>. Its chemistry has recently been shown to be distinct from the usual pattern both of the <u>Meliaceae</u> and the <u>Rutaceae</u> (e.g. absence of degraded <u>triterpenes</u> in the timber), and in the remarkable range of <u>chromones</u>, and some unusual <u>coumarins</u>. The results of our simple tests (Appendix I) strongly support this new classification. Unfortunately we did not have <u>Cedrelopsis</u>.

<u>Cedrelopsis</u> and <u>Ptaeroxylon</u> both contain <u>coumarins</u> and <u>ptaeroxylin</u> which has an unusual seven-membered ring-structure not encountered elsewhere. There would thus seem to be good evidence for removing <u>Cedrelopsis</u> with <u>Ptaeroxylon</u> into the new family, <u>Ptaeroxylaceae</u>.

The sub-family <u>Swietenioideae</u>, in many ways, seems chemically to be very distinct from the <u>Cedreloideae</u> and <u>Melioideae</u>, as it is on the basis of its anatomy and morphology. However, it is unsafe to draw definite taxonomic conclusions at this junction.

7. Akaniaceae

Although many authors placed this family in the <u>Sapindales</u>, Scholz considered it to be a family of the <u>Rutales</u>. The sole member is <u>Akania hillii</u> and Gibbs (MSS) obtained most of the following results.

Cigarette and hot-water tests -- II.

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Syringin Test 4:1 H_2SO_4/H_2O) -- negative, but a red color was present in the lignified tissues. Correspondly, it gave positive HCl/Methanol and leucoanthocyanin reslts. The positive HCl/Methanol result was also consistent with positive tannin tests.

<u>HCN Test A</u> -- doubtfully <u>positive</u> reaction was obtained. <u>Raphides</u> -- no raphides present.

Juglone Tests -- negative, suggesting that naphthoquinones are absent; but blue fluorescence was observed.

<u>Phenolic Acids</u> -- the major phenolic acids according to Galang (thesis, unpub'd. Material described as "<u>A</u>. <u>lucens</u>") are: gentisic, <u>p</u>-coumaric, caffeic, ferulic and ellagic, but Bate-Smith (1962) lists of these only <u>p</u>-coumaric as present in "<u>A.hillii</u>". Are these, in fact, the same plant?

<u>Alkaloids</u> -- are present in this family (Appendix VIII).

More information is required, and particularly, knowledge about <u>terpenoids</u> and <u>flavonoids</u>, before the affinities of this family can be decided.

8. Malpighiaceae

Only Scholz (1964) placed this family in <u>Rutales</u>. If it is properly placed by him *it* should have many chemical characteristics in common with the <u>Rutaceae</u>.

<u>Cigarette and Hot-Water Tests</u> -- mostly IV, but <u>Stigmaphyllon</u> tomentosum was recorded as I. <u>Syringin Test $(1:1 H_2SO_4/H_2O)$ </u> -- all plants gave a <u>negative</u> syringin test, but in contrast to <u>Rutaceae</u>, the majority of the species showed some red colour in the lignified tissue. These results correlated with those obtained with leucoanthocyanin, HCl/Methanol, and Ehrlich tests.

<u>HCN Test A</u> -- the majority of the plants seem to contain no cyanogenic substances. <u>Heteropterys</u> (2 spp.) gave doubtfully positive results with <u>HCN (Test A)</u>.

Raphides -- are absent from the species investigated.

Tannin Test A -- most of the few plants tested were tanniniferous. Thus results were consistent with "Saponin Test B" (NH_3) and also some of the results correlated with positive HCl/Methanol tests.

<u>Juglone Tests</u> -- were negative, so <u>naphthoquinones</u> probably are absent. Blue fluorescence was observed. This is in keeping with the finding that <u>umbelliferone</u> is present in <u>Malpighia</u> (paper chromatography),

<u>Phenolic Acids, etc</u>. -- major phenolic acids are: gentisic, p-coumaric, caffeic, ferulic and ellagic.

<u>Flavonoids</u> -- two classes of flavonoids, <u>leucoanthocyanins</u> and flavonols, have been found in this family.

<u>Alkaloids</u> -- of indole type are reported to occur in <u>Malpighiaceae</u>.

9. <u>Trigoniaceae</u>

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No material of this family was available to us, and we have found no information as to its chemistry from the literature.

10. Vochysiaceae

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Very little information was found in the literature as to the chemistry of this family. Fortunately, two species were available for investigation.

<u>HCl/Methanol Tests</u> -- gave one strong positive result (Vochysia) and one negative (Salvertia).

<u>Juglone Tests</u> -- naphthoquinones seem to be absent and no blue fluorescence was observed.

<u>Raphides</u> __ none have been reported, but solitary and cluster crystals have been seen (Appendix XI).

<u>Fatty Acids</u> -- we learn from the literature that the major fatty acids of the seeds are <u>myristic</u> and <u>lauric acids</u>. Many genera of the <u>Simaroubaceae</u> also contain these two acids.

11. Tremandraceae

This family has been placed in various positions. The relationships of this family to others remains unestablished.

A few species belonging to two genera, <u>Tetratheca</u> and Platytheca, were tested by Gibbs (unpub'd.)

Cigarette and Hot-Water Test -- IV for 4 spp. of Tetratheca.

<u>Syringin Tes</u>t (1:1 H_2SO_4/H_2O) -- negative results were obtained. A red colour has been seen only in <u>Tetratheca</u>. These results correlate with those obtained for the related tests.

<u>Raphides</u> - no raphides were seen in the Syringin Test Controls. Solitary and cluster crystals have been reported to occur. <u>HCN Test A</u> -- Gibbs got positive <u>HCN Test A</u> results for Platytheca and one species of Tetratheca.

Juglone Tests ... naphthoquinones seem to be absent. Blue fluorescence was recorded.

<u>Tannins</u> -- doubtfully present (Galang, thesis, unpub'd.) HCl/Methanol Test -- positive (1-4) in all tested.

12. Polygalaceae

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The <u>Polygalaceae</u> is said to be a very natural family. Many genera, however, such as <u>Xanthophyllum</u>, <u>Krameria</u>, and <u>Diclidanthera</u> have been doubtfully placed in this family. Unfortunately, material of these genera were not available for investigation.

<u>Cigarette and Hot-Water Tests</u> -- were recorded as IV for <u>Polygala</u>, but as II-III for Mundtia.

<u>Syringin Test</u> -- the species of <u>Polygala</u> tested gave negative with no red reactions in the lignified tissue.

<u>Ehrlich Test</u> -- <u>Polygala</u> negative. <u>Mundtia</u> negative (but a magenta Ehrlich spot).

<u>HCl/Methanol Test</u> -- <u>Polygala</u> and <u>Securidaca</u> negative. <u>Mundtia</u> positive (2).

Tannin Test A -- Polygala negative; Mundtia positive (++).

Leucoanthocyanin Test -- Polygala negative; Mundtia positive.

Juglone Tests -- Polygala negative (some fluorescence);

Securidaca negative; Mundtia positive.

Cyanogenic Glycosides -- as judged by <u>HCN Test A</u> -- seem to be absent.

Saponins -- have been recorded in some species. Amarasingham

et al. (1964) reported that one species of <u>Xanthophyllum</u> contains saponin. Four genera (<u>Xanthophyllum</u>, <u>Monnina</u>, <u>Bredemeyera</u> and <u>Polygala</u>) are reported to contain triterpenoid saponins and sapogenins. Two tests made by us with <u>Saponin Test A</u> were recorded as negative or doubtfully positive.

Raphides -- none have been recorded or observed by us.

<u>Flavonoids</u> -- many species of <u>Polygala</u> are reported to have flavonols.

<u>Phenolic Acids</u> -- the major phenolic acids appear to be gentisic, p-coumaric, caffeic, and ferulic.

<u>Alkaloids</u> -- are said to be present in 50% of the species tested, but none have been identified.

<u>Mundtia spinosa</u>, on the basis of the chemical evidence, seems to be out of place in this family, but there is the possibility that our material was wrongly labelled. Obviously this should be checked. Obviously, too, we need to know much more about the family as a whole.

B. The order Rutales

This is obviously a very diverse and unnatural order. Thus the chemical reactions and other results obtained were mixed (Table 2,3,4,and 5).

<u>Polyphenolases</u> (as judged from Cig. & H.W. Tests) were present in <u>Akaniaceae</u>; absent from both <u>Picrodendraceae</u> and <u>Tremandraceae</u>; but mixed results were obtained from members of the other families.

Except for one subtribe (<u>Cusparineae</u>) of the <u>Rutaceae</u>, raphides as well as syringin are absent. Other calcium oxalate crystals, for example, the widely distributed solitary and cluster crystals, are present. These crystals varied in their distribution, and are possibly familial rather than ordinal characters. In the syringin test, the presence or absence of a red colour in lignified tissues was mixed. Some families usually showed no red while others usually did so.

The results of HCl/Methanol tests were mixed. In most families, the majority of the species were positive in their response. The results were usually closely correlated with the red or magenta colours in syringin, leucoanthocyanin and Ehrlich tests. But, as previously mentioned, this correlation failed in the <u>Rutaceae</u>. The positive results of the HCl/Methanol test were also consistent with positive tannin tests in most of the plants tested.

Mixed results were obtained for the tannin test. Many were positive. These results were also correlated with those of "Saponin Test B" (NH_3).

<u>Saponins</u> -- as judged from Saponin Test A were generally absent, being present only in a few members of the <u>Meliaceae</u>. However, from the literature reported, there are saponins in <u>Polygalaceae</u>, <u>Meliaceae</u>, <u>Rutaceae</u>, and <u>Burseraceae</u>.

HCN was absent from or rare in most families. <u>Boronia</u> (<u>Rutaceae</u>), <u>Platytheca</u> and <u>Tetratheca</u> (<u>Tremandraceae</u>) appear to

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contain cyanogenic glycosides. Three further genera **Dysoxylum** (<u>Meliaceae</u>), <u>Akania</u> (<u>Akaniaceae</u>) and <u>Heteropterys</u> (<u>Malpighiaceae</u>) gave doubtfully positive <u>HCN Test A</u> results. Obviously more testing is needed.

The sole positive Juglone test was obtained in the case of <u>Mundtia spinosa</u> (<u>Polygalacea</u>e) but see above. Naphthoquinones seem to be absent from all families. Some blue fluorescence was observed in most of the families investigated (not in <u>Cneoraceae</u> and <u>Vochysiaceae</u>). Paper chromatographic results, however, showed that coumarins (excluding ellagic acid)(Table 4) are present in <u>Rutaceae</u>, <u>Meliaceae</u>, <u>Malpighiaceae</u>, and strikingly enough also in <u>Cneoraceae</u>. From the literature, we have reports of their occurrence only in <u>Rutaceae</u> and <u>Meliaceae</u>.

The number of triterpenes characteristic of members of the <u>Rutales</u> is forever increasing. The biogenetic and close structural relationships of elemolic acid, the limonoids and the simaroubolides parallel the close taxonomic relationships of the <u>Rutaceae</u>, <u>Burseraceae</u> and <u>Meliaceae</u> with the <u>Simaroubaceae</u>. In these four families other terpenoids also occur widely.

Flavonoids, especially the flavonols, are also widely spread in most families of the <u>Rutales.</u> The co-occurrence of flavons and flavanones has been found only in <u>Rutaceae</u>. This family also shows the replacement of flavones by flavonols.

Some of the families are alkaloid-containing, especially the family <u>Rutaceae</u>. However, there are no recorded alkaloids from <u>Cneoraceae</u>, <u>Picrodendraceae</u>, <u>Trigoniaceae</u> and <u>Vochysiaceae</u>, families which have been little investigated.

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The fatty-acid compositions of the seeds vary, but those of the <u>Simaroubaceae</u> and <u>Vochysiaceae</u> are similar, and so are those of the <u>Burseraceae</u> and <u>Meliaceae</u>. This could link them taxonomically.

Group II phenolic constituents were rarely found in the families. The other compounds of Group I and III were found in varying degrees. While ellagic acid was very widely distributed in the families of this order.

SUMMARY AND CONCLUSION

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The <u>Rutales</u> seems to be a homogeneous order, if we consider it to contain the families <u>Rutaceae</u>, <u>Simaroubaceae</u>, <u>Burseraceae</u> and <u>Meliaceae</u>. These appear to be correctly grouped by Scholz in his suborder <u>Rutineae</u>. We are less sure about the correctness of placing of the remaining families of his suborder, the <u>Cneoraceae</u>, <u>Picrodendraceae</u>, and <u>Akaniaceae</u>.

We know too little as yet of the chemistry of the <u>Trigoniaceae</u> and <u>Vochysiaceae</u> to say whether or not they constitute with the <u>Malpighiaceae</u> a homogeneous group (Scholz's <u>Malpighiineae</u>) within the <u>Rutales</u>. The chemistry of the <u>Malpighiaceae</u>, however, is consistent with a position near the Meliaceae.

The two families <u>Tremandraceae</u> and <u>Polygalaceae</u> (Scholz's suborder <u>Polygalineae</u>) seem to differ greatly from each other in their chemistry, but the inclusion of <u>Mundtia</u> (see above) contributes to this. We do not feel in a position, at this stage of our knowledge, to pass judgement on the placing of these families.

There are some systematists who would have the chemotaxonomist deposit in a herbarium a voucher specimen of every plant tested. This is not practicable in extensive investigations, but it would obviously be very worth while to have a voucher

specimen of the "<u>Mundtia spinosa</u>" referred to above, for comparison with any later material that becomes available. Unfortunately we do not have this.

The creation of two more small families <u>Flindersiaceae</u> (<u>Flindersia</u> and <u>Chloroxylon</u>) and <u>Ptaeroxylaceae</u> (<u>Ptaeroxylon</u> and <u>Cedrelopsis</u>) is supported by chemical evidence. They should be placed, perhaps, between <u>Rutaceae</u> and <u>Meliaceae</u>.

The data also support the separation of <u>Picramnia</u> (<u>Picramnioideae</u>) from the <u>Simaroubaceae</u>; <u>Mundtia</u> (if our material was genuine) from the <u>Polygalaceae</u>; and <u>Fagara</u> from Zanthoxylum.

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Appendix I A list c

		H.W. I II	III	T17	Or	Ciq I		II
	RUTACEAE	<u></u>	<u> </u>	<u> </u>		<u> </u>	<u></u>	
	I. Rutoideae							
	Zanthoxylum americanum	+				+		
	Zanthoxylum martinicence	+						
	Zanthoxylum simulans	•						
	Evodia Danielli							
	Evodia Henryi	+						
	Evodia micrococea	-0		+				
	Orixa japonica			•	•			
	Malicopa ternata			+				
	Ruta Bracteosa			+				
	Ruta chalepensis			+				
				- -				
	<u>Ruta graveolens</u>			+				
	Dictamnus albus			т				
	<u>Dictamnus</u> albus var.		III-					
	turkestanicus		***-					
	Cneoridium dumosum		•	+				
	<u>Boronia denticulata</u>				+?			
	<u>Boronia lanaqmusa</u> ?							
	<u>Bornnia pinnata</u>			+				
	<u>Boronia purdiana</u>				_			
	<u>Boronia viminea</u>				+?			
	Eriostemon spicatum							
	Eriostemon myoporoides	+						
	Phebalium billordierii			+?				
	Phebalium phylicifolium	+						
	<u>Crowea</u> <u>dentata</u>			+				
	<u>Correa harrisii</u>							+7
	Correa laurenciana							
	Correa turnbullii							
	Diplolaena angustifolia		+?					
•	<u>Calodendron</u> <u>capense</u>			+				
	Barosma Scoparia			+				
	Diosma appositifolia			•				
				+				
	<u>Pilocarpus</u> pennatifolius	-	TTT 2-					
	Erythrochiton brasiliensi	<u>.s</u>	111?-	- T A	•			
	II. Dictyolomatoideae							
	III.Flindersioideae							
	<u>Flindersia</u> <u>australis</u>	I-II						
	IV. Spathelioideae							
	V.Toddalioideae							
	Phellodendron amurense	II-	-III	+ 1		+		
	Phellodendron japonicum Phellodendron lavallei							
	Phellodendron lavallei		-III					
	Phellodendron sachalinens	se +						
	Ptelea trifoliata			+				
	Oricia eurynnertonii							
	Acronychia imperforata	II-	-III	+ .				
	Acronychia suberosa							
	Skimmia foremanii				+?			
	Skimmia japonica			+	• •			
	<u>Teclea</u> simplicifolia			+				

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Appendix I A list of all plants tested (by the writer, Gibbs and others)

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tested (by the writer, Gibbs and others)

Appendix I (cont'd.)

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	H.W. I II	III	IV	Or	Cig. I II	III	IV	Or
RUTACEAE (cont'd.)								
								:
VI. Citroideae (Aurantioideae)			ъ					
<u>Glycomis pentaphylla</u>			+					
<u>Murraya</u> <u>exotica</u>			+				+	
<u>Murraya koeniqii</u>	I-II							
Murraya paniculata			+					
Clausena lansuim	II-	III						
<u>Clausena</u> <u>lumulata</u>	I-II							
	1. L. L.		+			III-	T 17	
<u>Atalantia ceylaniea</u>			т			TTT _	τv	
Poncirus trifolia								
<u>Citrus aurantifolia</u>			+				+	
<u>Citrus Limetta</u>			+				+	
Citrus limoxia			+					
<u>Citrus mascina</u>			+				+	
	_		_				+	
<u>Citrus medica</u>	-		T				т	
<u>Citrus</u> nobilis			+					
<u>Citrus simensis</u>			+					
<u>Citrus sp. ("otasheite</u>			+					
orange")								
<u>Aegle marmelosa</u>	I	٠			II-I	тт		
	Ŧ				<u> </u>			
Fortunella margarita			+					
<u>Triphasia</u> trifolia			+					
VII.Rhabdodendroideae								
Rhabdodendron								
CNEORACEAE								
<u>Cneorum_tricoccon</u>		III-	τv				·	
SIMAROUBACEAE			- •					
	-							
I. Surianoideae								
<u>Suriana maritima</u>	+							
II. Simarouboideae								
<u>Hannoa klaineana</u>								
Ailanthus altissima	II-	III					+	
III.Kirkioideae								
			+					
<u>Kirkia</u> <u>acuminata</u>			т					
IV. Irvingioideae								
V.Picramnioideae								
<u>Picramnia pentandra</u>	+		•					
VI.Alvaradoideae								
PICRODENDRACEAE								
Picrodendron baccatum			+					
			т					
BURSERACEAE								
<u>Bursera simarouba</u>					•			
<u>Bursera simplifolia</u>		+						
<u>Commiphora</u> merkeri	II-	III						
Commiphora trothai								
Pachylobus klaineana								
MELIACEAE								
I. Cedreloideae								
Cedrela odorata		III (or I	V				
Toona ciliata			+					+
Ptaeroxylon obliguum	Ŧ		•					-
TT Chi abort address ODILQUM	+							
II.Swietenioideae								
Khava Avasica								
Khaya hyasica <u>Entandrophragma candatum</u>			+					
Swietenia Manogni	I-II							

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IV	Or	Syr.	Rap.	Ehr.	HCN	HC1/M	L.A.	J A	lug. B	C	т.т.	S.T.A	S.T.B
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		-NR	-	-YB	-	-	-	- '	· -	-	++	-	0
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v		-PK	-	-YB	-	-	-	-	-	-	-	-	0
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Appendix I (cont'd.)

	<u> </u>								
	H.	w.				Ci	g.		
	I		III.	IV	or	I	II	III	IV Or
MELIACEAE (cont'd.)			فيأتليب فيجعل سابتتها		جعد بالرحفني عالاتهم				
III. Melioideae									
Carapa quianensis	+								
Carapa procera	+								
<u>Cipadessa cinerascens</u>	•			+					
Melia azodaroch				+				III	-IV
Trichilia emetica									
Dysoxylum fraseranum	÷								
Dysoxylum spectabile	+								
AKANIACEAE	•								
<u>Akania hillii</u>		+							
MALPIGHIACEAE		•							
Hiptage benghalensis									
Tristellateia australasi	ae	•		+?					+?
Heteropterys chrysophyll			•	••					
Heteropterys umbellata					+?				
Malpiqhia coccipera		-		+	••				
Malpiqhia cubensis				, 上					
Malpighia glabra			III-	- T 17					
Malpighia punicifolia			TTT-	-т, ,					
Byrsonima crassifolic									
Stigmaphyllon ledifolium									
Thryallis glauca	Ļ								
Stigmaphyllon ciliatum				+					
Stigmaphyllon tomentosum	. т			-					
TRIGONIACEAE	Γ								
VOCHYSIACEAE				•					
<u>Vochysia</u> sp. <u>Salvertia</u> convalloriodor	-								
	d								
TREMANDRACEAE									
<u>Platytheca</u> verticilliata									
<u>Tetratheca</u> ericifolia				+					
<u>Tetratheca</u> pilosa									
<u>Tetratheca</u> <u>setigera</u>				+					
<u>Tetratheca</u> thymifolia				+					
<u>Tetratheca viminoa</u>				+					
POLYGALACEAE									
<u>Polygala capitata</u>									
<u>Polygala dalmaisiana</u>				+					
<u>Polygala myrtifolia</u>			III-	-IV				┸┸┘	I-IV
<u>Polygala</u> sanguinea				~					
<u>Polygala senega</u>				+					
<u>Polvgala virgata</u>									
<u>Securidaca diversifolia</u>									
<u>Mundtia spinosa</u>		II-	III						
	. <u></u>						<u> </u>		
Key to symbols: NR = no red				= br					
R = red					ange				
PK = pink			YB	= ye	210w	-bro	wn		
C = crystal	0		an		n = n = 1		~		

C = crystals Y = yellow Gy = green-yellow PY = pale-yellow BY = brown-yellow

GB = green-brown
GB = pink-brown
M = magenta
DM = dull magenta
tr = trace

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	G	-						ug.	_		S.T.A.	C T B
IV Or	Syr.	Rap.	Ehr.		HC1/M	L.A.	<u>A</u>	В	C	T.T.	D. 1. D .	₽•±•₽;
Í	-R		-M	-	+4	+	_		+	+++		
	-R	-	-M	_	+	+	-		+	+++		
	-NR	-	-M	-	+3	+	-	-	+	++		0
I-IV	-NR		-Y	-	-	-	-	-	+	-	-	0
				_	+2							
Í.			vo	+?	+2 +3		-	-PY	+		-	
1			-YG		+3							
	-R	-		+?	+1-2	+	-	-	+	+++		
					+4		-	-	-			
+?	–NR –NR	-		- +?	-		-		+	tr.		
i	-NR	-	-Y	+?	+4	-	-	+Y	_	-		0
	-R	~	-M	-	+4	+	-	-	-	+?	-	1
	-R	-	M		+4	+	-	-		+++	-	4
1	-R	-	-M	-	-?	+?	-		+	-		
	-R				+1							
	-R				+4 +4		_	+Y	_			
i					+4		_		_			
	-R	-	-M	-	+1	+	-	_ 1	-	+++	-	4
	-R	-	-M	-	+4	+	-	-	-	+++	-	3
					+4		-	-				
					~			-	-			
	-NR	-	-Y	+	+1	-	-	-	+	+?		
	-R	-			+3							
				+	+4							
	-R -R		-M	-	+4							
	- R	-	-M	-	+4					+?		
			-14	-								
	-NR	-			~							
	-NR	-	-Y	-	-		-	+Y	-	-	-	1
II-IV	-NR	-	- B	-	-	-	-		-			
	-NR	-	-B -Y -Y									
	-NR	-	-Y	_		-	_		+		-?	0
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			-M	-	+2	+	+		tr	? ++		

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Key to a list of phenolic acids and coumarins:

- I. Benzoic acids (C_6-C_1)
 - 1. <u>p-Hydroxybenzoic</u>
 - 2. 2,3-Dihydroxybenzoic (6-pyrocatechuic)
 - 3. 3,4-Dihydroxybenzoic (protocatechuic)
 - 4. 2,5-Dihydroxybenzoic (gentisic)
 - 5. 2-Hydroxy-5-methoxy-benzoic
 - 6. 2-Hydroxy-6-methoxy-benzoic
 - 7. 2-Hydroxy-3-methoxy-benzoic
 - 8. 4-Hydroxy-3-methoxy-benzoic (vanillic)
 - 9. 4-Hydroxy-3,5-dimethoxybenzoic (syringic)
- 10. 3,4,5-Trihydroxy-benzoic (gallic)
- II. Phenylacetic acids (C6-C2)
 - 1. <u>p-Hydroxyphenylacetic</u>
 - 2. o-Hydroxyphenylacetic
 - 3. 3-methoxy-4-hydroxy-mandelic
- III. Cinnamic acids (C_6-C_3)
 - 1. <u>o-Hydroxycinnamic (o-coumaric)</u>

- 2. p-Hydroxycinnamic (p-coumarin)
- 3. 3,4-Dihydroxycinnamic (caffeic)
- 4. 4-Hydroxy-3-methoxycinnamic (ferulic)
- 5. 4-Hydroxy-3,5-dimethoxycinnamic (sinapic)
- 6. <u>o-Hydroxydihydrocinnamic</u> (melilotic)
- 7. <u>p-Hydrodihydrocinnamic</u> (phloretic)
- 8. 2, 3-Dihydroxycinnamic
- 9. 2-Hydroxy-3-methoxycinnamic
- 10. 2, 3-Dihydroxy-phenyl-proprionic
- 11. p-Hydroxy-phenyllactic
- IV. Coumarins

AP I -1

- 1. Ellagic
- 2. Umbelliferone
- 3. Aesculetin
- 4. Scopoletin

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						I				+ 14		,II							II			
	1	2	3	4	5	6		8	9	10	1	2	_3	1	2	_3		5	6	7	8	_9
CNEORACEAE																						
<u>Cneorum</u> tricoccon	-			+	-	-	-	-	-	-	-	-	-	.+	++	++	+	++	-	-	-	-
SIMAROUBACEAE																						
<u>Ailanthus</u> <u>altissima</u> <u>Kirkia</u> acuminata	-	-	-	-	-	-	-	-	- Tr	+ +++	-	-	-	-	+ -	+++	++ 	-	Tr -		-	-
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PICRODENDRACEAE BURSERACEAE <u>Commiphora</u> <u>merkeri</u>	-	-	-	-	-	-	_	-	-	+	-	-	_	-	+	++	Tr	-	_	-	-	-
MELIACEAE														l '								
<u>Ptaeroxylon</u> obliguum	_	-	-	-	_	-	-	-	-	-	<u>;</u>	_	_	-	+	+	+++	_	+?	_	_	_
Khaya nyasica	-	·	-	+?	-	-	-	-	-	+?	-		-	-	+	+	+	-	-	-	-	-
<u>Swietenia</u> ·macrophylla	+	-	+	+	_	_		+	-	+	+	-	_	_	++	++	+	+		-	_	_
<u>Carapa</u> guianensis		_	-	+	_	_	_	_	-	_	<u> </u> _	_	_	_	÷	+	Tr?	-	-	_	_	-
Carapa		_	_	·	_	_	_	m~ '	°	Tr	: :	_	_		•		Tr		<u> </u>		_	•
procera Cipadessa	-	-	-	-	-	-	-	TT (?Tr?	TT	ľ	-	-	-		+++		-	TT	-	-	-
<u>cinerascens</u> <u>Melia</u>	-	-	-	+?	-	-	-	-	+	-	-	-	-		+	+	Tr	-	+?	-	-	-
azedarach AKANIACEAE	-		. –	-	-	-	-	-	-	-		-	-	-	Tr'	?+	+		+	. –		-
Akania lucens	. +?	_	-	+	-	-	-	Tr:	?-	-	-	-	-	-	++	+	+	-	~	-	-	-
MALPIGHIACEAE <u>Gaudichaudia</u>	ŀ																					
Cyananchoide Malpighia	<u>s</u> -	-	-	-	-	-	-	-			-	-	-	_	+	-	Tr?	-	-	-	-	-
coccigera	+	_	-	+	-	-	-	-	+	-	-	-	_		+	+	+	+	-	-	-	-
<u>Malpighia</u> cubensis	-	_	_	Tr	?-		_	-	-	+?	_	_	_	_	-	+	Tr?	_	+	-	_	-
TRIGONIACEAE VOCHYSIACEAE TREMANDRACEAE																						
Platytheca																						
verticillia Tetratheca	<u>a</u> -	-	-	-	-	-	· •••	+	Tr?	+++	-	-	-	-	+	+?	Tr?	-	+?	-	-	-
<u>thymifolia</u>	· _	-	-	-	-	~	-	-	-	+	í -	-	-	-	+	-	-	-	-	-	-	-
POLYGALACEAE <u>Polygala</u> myrtifolia	_	-	_	+	_	_	-	_	_	_	_	-		-	-	+	Tr?	+	_	_	-	-
											L											

(cont'd.) II

,II IV III I 5 6 567 10 1 2 3 10 11 2 3 8 9 2 3 4 7 8 9 1 4 1 + + Tr +++ -Tr +++ Tr ++ + + Tr -++ . _ +? -2 +? -.? + ++ _ + _ + _ 4-4 Tr? -_ + Tr?Tr? Tr Tr Tr -++ + Tr Tr +? --+ + _ Tr Tr?+ + --Tr?--+ 22

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Key to a list of coumarins: (cont'd.)

III. 7,8-Furocoumarins

- 1. Meishoutin (Cyclo-obliquetin)
- 2. Nieshoutol
- 3. Thamnosimin

IV. Chromano-coumarins

- 1. Alloxanthoxyleton
- 2. Braylin
- 3. Clausenidin
- 4. Clausenin
- 5. Dentatin
- 6. Luvangetin

- 7. Nordentatin
- 8. Obliquin
- 9. Obliquol
- 10. Xanthoxyleton (5-Methoxyxanthyletin)
- 11. Seselin
- 12. Xanthyletin (2': 2'-dimethyl-pyrano-(5': 6'-6:7) coumarin)

AP II-3

Key to a list of Coumarins: (cont'd.)

II. 6-7-Furocoumarins:

- 1. Alloimperatorin
- 2. Bergapten (5-Methoxy-psoralen)
- 3. Bergamottin (5-Geranoyx-psoralen; 5-Geranloxy-6,7-furocoumarin-Bergaptin)
- 4. Bergaptol (5-Hydroxy-psoralen)
- 5. Byakangelicin (5-Methoxy-8-dihydroxy-isopentanoxypsoralen
- 6. Chalepensin
- 7. Chalepin
- 8. 5-(3,6-Dimethyl-6-formy-2-heptenyl) oxy)psoralen
- 9. 6,7-furocoumarin (Psoralen; Ficusin)
- 10. 5-Geranoxy-8-methoxypsoralen
- 11. 8-Geranoxy psoralen
- 12. Halfordin
- 13. Heliettin ((+)-3-(1,1-Dimethylallyl)-6,7-dihydro-7-(1-hydroxyl-methylethyl)2H-furo-(2,3-g)-1benzopyran-2-one)

- 14. Imperatorin
- 15. (-)-Imperatorin oxide
- 16. Isohalfordin
- 17. Isoimperatorin (5-r,r-Dimethylallyloxypsoralen; 5-Isopenteneoxypsoralen)
- 18. Isopimpinellin (5,8-Dimethoxypsora
- 19. Marmesin ((-)-Marmesin)
- 20. (+)-Marmesin
- 21. Marmesine
- 22. 5-Methoxy-8-geranyloxypsoralen
- 23. 5-(3'-Methyl-2',3'-dihydroxybutanyl) 8-methoxy-psoralen
- 24. Oxypeucedanin hydrate (5-Dihydroxyisopentanoxy-psoralen)
- 25. Phellopterin (5-methoxy-8-r,rdimethylallyloxy-psoralen
- 26. Kanthotoxin (8-Methoxy-psoralene)

Key to a list of Coumarins: (cont'd.)

- I. Simple coumarins:
 - 28. Meranzin (Auraptene (2); 7-methoxy-8-epoxyisopentenyl-coumarin)
 - 29. 7-Methoxy-8(2-formyl-2-methyl-propyl)-coumarin
 - 30. 7-Methoxy coumarin-6-aldehyde
 - 31. 7-Methoxy-5-geranoxy-coumarin
 - 32. 8-Methoxy-4-methylcoumarin
 - 33. Mexoticin (5,7-dimethoxy-8-(2',3'-dihydroxyisopentyl)-coumarin
 - 34. Micromelin
 - 35. Obliquetin
 - 36. Obliquetol

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- 37. Osthol (7-Methoxy-8-isopentenyl-coumarin)
- 38. Prenyletin (7-0-(3,3-Dimethylallyl) aesculetin)
- 39. Prenyletin-6-0-methyl ether

- 40. Scopoletin (Chrysatropic acid; 6-Methyl-aesculetin)
- 41. 7-0-(1,1-dimethylallyl) scopoletin
- 42. 7-0-(3,3-dimethylallyl) Scopoletin
- 43. Scopolin
- 44. Skimmin (Umbelliferone-7-glucoside)
- 45. Suberenol
- 46. Suberosin
- 47. Toddaculin (5,7-Dimethoyx-6-(2'isopentenyl)
- 48. Toddalo-lactone (Aculeatin-hydrate? 5,7-Dimethoxy-6(2,3-dihydroxyisopentenyl)-coumarin)
- 49. 6,7,8-Trimethoxycoumarin (Dimethylfraxetin)
- 50. Umbelliferone (Dichrin-A; Hydrangin; 7-Hydroxy-coumarin; Skimmetin)

Key to a list of Coumarins:

- I. Simple coumarins:
 - 1. Aculeatin
 - 2. Aurapten
 - 3. Auraptena (7-Geranuloxy-coumarin)
 - 4. Auraptenol (7-Methoxy-8 (2-hydroxy-3methyl-3-butenyl)-coumarin)
 - 5. Brayleanin
 - 6. Collinin (7-Geranoxy-8-methoxy-coumarin)
 - 7. Coumarins
 - 8. Coumarrayin (5,7-dimethoxy-8 (2-iso-pentenyl)coumarin)
 - 9. Cyclobisuberoidene
 - 10. Daphnoretin
 - 11. Dehydrogeijerin (6-(B,B-Dimethylacrylyl)-7methoxycoumarin)
 - 12. 7-Desmethyl-2', 3'-dihydroxy dihyrosuberosine
 - 13. 7-Demethylsuberosin
 - 14. 7-(6',7'-Dihydroxy-3',7'-dimethyl-2'-octenyl) oxy) coumarin

- 15. 5-Isopentenoxy-7-methoxy-coumarin(5-r,rdimethylallyloxy-7-methoxy coumarin)
- 16. 5,7-Dimethoxycoumarin
- 17. 5,7-Dimethoxy-8-(3'-methyl-2'-oxobutyl) coumarin
- 18. 6,7-Dimethoxycoumarin (Aesculetindimethyl ether; Scoparin (2); Scoparone)
- 19. 8-(Dimethyallyl)-7-hydroxy-6-methoxycoumarin
- 20. Geijerin (6-isovalery1-7-methoxycoumarin)
- 21. Geiparvarin (7-(4,7-epoxy-3,7-dimethyl-6-oxoocta-2,4-dienyloxy)coumarin)
- 22. Gravelliferone (3-(1,1-dimethylally1)-6-(3,3-dimethylally1)umberiferone)
- 23. Gravelliferone methyl ether
- 24. Herniarin (Ayapanin; 7-Methoxy-coumarin
- 25. 3-(1,1-Dimethyally) Herniarin
- 26. Limettin (5,7-Dimethoxycoumarin-Citropten)
- 27. Marmin (7-Dihydroxygeranoxy-coumarin)

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Appendix III Distribution of Coumarins (by genera (Numbering of Coumarins as in key; numbers

								ins a					_
.			1	2	3	4	5	<u> </u> 6	7	8	9	10	
RUTAC	EAE												
I.													
	Evodia	(120)											
	<u></u>	(200)											
		(200)											
	Fagara	(200)											
	Geijera	(7)											
	Melicope	(50)											
	Zanthoxylum	(15)									1		
	Cneoridium	(1)	•										
	Dictamnus	(2)		1									
				Ŧ					-			-	4
	Ruta	(.60).							1			1	
	Thamnosma	´ (6)											
•	Phebalium .	(36)											
	· · · · · · · · · · · · · · · · · · ·	•			-								
III.	Flindersioideae	2											
• • •	Flindersia	(20)					1	1					
	E TTUMET DTG	(20)	•				-						
								(BK)					
	Chloroxylon	(1)											
v.	Toddalioideae												
	Casimiroa	(6)											
	Halfordia	(4)											
	Helietta												
		(8)			-								
	Ptelea	(3)			1								
	Skimmia	(10)											
	Toddalia	(1)	1										
			(Rt.Bk)										
VI.	Citroideae					-							
	Aegle	(1)			1								
	VEATE	(1)											
					(Rt)								
	<u>Aeglopsis</u>	(5)						•					
	Citrus	(60)			2	1							
		-				(Ft,C))						
	Clausena	(30)					,						
	Limonia	(1)											
	Luvunga												
		(12)											
	Micromelum	(10)											
	Murraya	(9)								1			
	Poncirus	(1)											
	Severina	(1)											
		(-/											
MET	IACEAE												
	. Cedreloideae												
T		/=>											
	Cedrelopsis	_ (7)											
	Ptaeroxylon	(1)											
	Ekebergia	(15)											
													-
Key t	o symbols: Bk	= Bark	5		LV =	= Le	ave						
	- Ft				0 =	-							
	HW		lwood										
			MOOO		Rt =	= 76	ot						

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Appendix III (cont'd.)

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UTACEAE		26	27	28	29	30	31	32	33	34	
I. Rutoideae											1
Evodia	(120)										1
Fagara	(200)										
Geijera	(200)										:
Melicope	(50)										
Zanthoxylum	(15)				1						
	(10)				-						
Cneoridium	(1)										
Dictamnus	(2)										•
Ruta	(60)	1 :									•
Thamnosma	·(6)										
Phebalium	(36)										
	-			`							÷
III. Flindersioideae	(00)										
Flindersia	(20)										
Chloroxylon	(1)										•
7. Toddalioideae											
Casimiroa	(6)										
Halfordia	(4)										
Helietta	(8)										•
Ptelea	(3)										
Skimmia	(10)										
Toddalia	(1)		1								
											4 8 ¹
VI. Citroideae	(1)	1	7	1							
Aegle	(1)	1 Rt,Bk	1	$\frac{1}{(DTO)}$:
Aeglopsis	(5)	KL, DA)(E)	(PLD)							:
Citrus	(60)	3	1			3					•
Clausena	(30)	5	-								
Limonia	(1)										;
Luvunga	(12)					1					:
· ·					•	(0)					;
Micromelum	(10)						•			1	;
Murraya	(9)			•					1		-
									(Bk)		
Poncirus	(1)										i ;
Severina	(1)										
· · ·	•••					~					ŀ
MELIACEAE	(7)										ł
I. Cedrelopsis	(7)										
<u>Cedrelopsis</u> Ptaeroxylon	(7) (1)										
FLACELOXYTOIL	(1)										11
Ekebergia	(15)					1					1

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			1	2	<u>3</u>	4	5	6	7	8	9	10	1
RUTAC													
I.		(100)											
	Evodia	(120)											
	Fagara	(200)											
	Geijera	(7)											
	Melicope	(50)								1			
	Zanthoxylum	(15)								1			
	Cneoridium	(1)		1						• -			
	Dictamnus	(2)		-							1		
	Ruta	(60)		3			1	2	1		1 3		
	Thamno'sma	(6)		•			1 1	-	-	;	ĩ		
	Phebalium	(36)		1			-			•	1 1		
			116,000,000								· / · · · · · · · · · ·	•	
III.	Flindersioideae												
	<u>Flindersia</u>	(20)								•			
	Chloroxylon	(1)								÷			
										1			
v.	Toddalioideae			-									
	<u>Casimiroa</u>			1						1			
	· · · · · · · · · · · · · · · · · · ·									ł			
	Halfordia	(4)											
	<u>Helietta</u>	(8)											
	1101200000	(0)								•			
	Ptelea	(3)		1			1			•	2		
	Skimmia	(ÌO)											
	Toddalia	(1)								•			
							-			4			
VI.	Citroideae	•											
	Aegle	(1)	1										
			(Ft)										
•	<u>Aeglopsi's</u>	(5)											
				_	_	_	_						
	<u>Citrus</u>	(60)		2	4	2	2			1		_	-
		((P1,0)		2	2
	<u>Clausena</u>	(30)											
	Limonia	(1)											
	Luvunga	(12)								i			
	Micromelum	(10)								:			
	Murraya	(9)	-										
	Poncirus	(1)	1	-						:			
	Severina	(1)		1						4			
MELIA	OF AF	· · · · · · · · · · · · · · · · · · ·		(Ft)						į		•	
MELIA I.	CEAL Cedreloideae												
ہ علد	Cedrelopsis	(7)											
	Ptaeroxylon	(1)								+ -			
										-			
	<u>Ekebergia</u>	(15)											

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<u></u>						1	1 (Bk)		1					1	
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						1 1				•		and over Statistican V	1 ()	-	
2 (Bk)				1		1				1				1 (Cđ)	
(70)	1 (Bk)	1	1			1	1	1					2		
		1 (Ft) 1 (Ft)			1 (Ft)	1 (Ft)	1			• •				1	917 (1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 - 1999 -
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		1 1 (Ft)			 • •	1 (Ft)	••• .		1	• • • • • •		^с а.	• •	1	
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Appendix III (cont'd.)

				III		í			
			1	2	3	1 1	2	3	4
RUTAC	EAE					-			
I.	Rutoideae					ļ		l	
	Evodia	(120)]		ł	
	<u>Fagara</u> <u>Geijera</u>	(200) (7)				1		1	
	Melicope	(50)							
	Zanthoxylum	(15)				1			
						(Bk)			
	<u>Cneoridium</u> Dictamnus	(1) (2)							
	Ruta	(60)							
	· · · · · · · · · · · · · · · · · · ·								
	Thamnosma	(6)			1				
	Phebalium	(36)		•					
III.	Flindersioidea	е						•	
	Flindersia	(20)	,		ĺ		1		
							(Bk)		
	<u>Chloroxylon</u>	(1)			į			•	
v.	Toddalioideae								
	Casimiroa	(6)			•				
	Halfordia	(4)							
	<u>Helietta</u> Ptelea	(8)						÷	
	Skimmia	(3) (10)							
	Toddalia	(1)							
				-				•	-
VI.	Citroideae <u>Aegle</u>	(1)							
	Aeglopsis	(1) (5)							
	Citrus	(60)						-	
	Clausena	(30)						1]
	Limonia	(1)							
	Luvunga	(12)							
	Micromelum	(10)							
	Murraya	(9)							
	Poncirus	(1)							
	Severina	(1)		ì		,			
ELIACE									
I.	Cedreloideae	•-•							
	<u>Cedrelopsis</u> Ptaeroxylon	(7)	т [,]	-					
	PLACEOXYLON	(1)	1 (Hw)	1 (Hw)					
	Ekebergia	(15)	(1144)	(11)				•	
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APPENDIX IV (cont'd.)

- VI. Flavonols:
 - 1. Auranetin
 - 2. Datiscetin-2'-methyl ether-?-diglucoside
 - 3. Demethoxy-icaritin-7-glucoside (Amurensin)
 - 4. Flindulatin
 - 5. 5-Hydroxy-4',6,7,8-tetramethoxy-flavonol-3-methyl ether
 - 6. Isolimocitrol
 - 7. Limocitrin
 - 8. Limocitrol
 - 9. Melibentin
 - 10. Melisimplexin
 - 11. Melisimplin
 - 12. Meliternatin
 - 13. Meliternin
 - 14. Quercetin-3-rutinoside
 - 15. Quercitrin
 - 16. Tambuletin
 - 17. Tambuletin-4',7-dimethyl ether
 - 18. Ternatin
 - 19. Wharingin

APPENDIX IV

Key to a list of Flavonoids:

- I. Anthocyanins
 - 1. Cyanidin-3-glucoside
 - 2. Delphinidin-3-glucoside
- **II.** Leucoanthocyanins
 - 1. Leucopelargonidin
- III. Flavanones
 - 1. Citromitin
 - 2. Citronetin-7-rhamnoglucoside
 - 3. Demethylcitromitin
 - 4. Eriodictyol-7-rhamnoside
 - 5. Eriocitrin (Eriodictyol-7-rutinoside)
 - 6. Hesperetin-7B-neohesperidoside (Neohesperidin)
 - 7. Hesperidin (Hesperetin-7-rutinoside)
 - 8. Isosakuranetin
 - 9. Isosakuranetin-7-rutinoside
 - 10. Isosakuranetin-7-neohesperidoside (Poncirin)
 - 11. Naringenin
 - 12. Naringenin-7-neohesperidoside (Naringin)
 - 13. Naringenin-7-rutinodide
 - 14. Naringenin-4'-glucoside-7-rutinoside
- IV. Flavanonols
 - 1. Phellamurin
- V. Flavones:
 - 1. Acacetin-7-rutinoside
 - 2. Apigenin-7-neohesperidoside

- Apigenin-7-rutinoside 3. 3'-Demethoxy-sudachitin **'4**. 5,6-Dimethoxyflavone 5. 6. Diosmetin-7-rutinoside Diosmetin-6-C-B-D-glucoside 7. Diosmetin-8-C-B-D-glucoside 8. 9. 3,3',4',5,5',6,7-heptamethoxyflavone 10. Luteolin-7-rutinoside Nobiletin 11. 5-0-desmethylnobiletin 12. 13. Orientin 14. Tsoomientin 15. Ponkanetin 16. Sinensetin 17. Sudachitin 18. Tangeretin 19. 3', 5, 5', 6-tetramethoxyflavone 5,6,2'-trimethoxyflavone 20. 3', 5, 6-trimethoxyflavone 21. Vitexin-?-xyloside 22. 23. 0-D-xylosylvitexin
- 24. Vitexin
- 25. Isovitexin
- 26. Zapotin
- 27. Zapotinin

Appendix IV Distribution of Flavonoids (

(Compiled from literature survey) (Numbering of

(Numbers in table are numbers of si

		I	II		- <u></u>			
	1	2	1	1	2	3	4	5
RUTACEAE I. Rutoideae l. Zanthoxyleae <u>Fagara</u> (200) <u>Melicope</u> (50) <u>Zanthoxylum</u> (15)								
2. Ruteae <u>Boenninghaus-</u> <u>eni'a</u> (1) <u>Ruta</u>		·						-
III. Flindersioideae <u>Flindersia</u> (20)								-
V. Toddalioideae <u>Casmiroa</u> (6) <u>Phellodendron</u> (10) <u>Teclea</u> (25)	-							•
VI. Citroideae <u>Citrus</u> (60) <u>Fortunel'la</u> (6) <u>Murraya</u> (9) <u>Poncirus</u> (1)	ï	1		2	1	2	1	1
MELIACEAE								
<u>Ptaeroxylon</u> (I) MALPIGHIACEAE								•
<u>Malpighia</u> (30)			1					
Key to symbols: Bk = Bark F1 = Flowe Ft = Fruit		P	= Leav = Peel = Seed	_	• .			

Dnoids (by genera) in "Rutales" ering of Flavenoids as in key) ers of species)



	·		1	2	3	4	5	6	7	8	9	10	11	12
RUTAC I. F 1.	autoideae Zanthoxyleae	(200) (50) (15)						2						
2.	Ruteae <u>Boenning-</u> <u>hausenia</u> <u>Ruta</u>	(1) (60)										·		
III.	Flindersioidea <u>Flindersia</u>	ae (20)												
۷.	Toddalioideae <u>Casmiroa</u>	(6)					1	- 1						
	Phellodendro Teclea	on (10) (25)					(Rt, Bi	-)					·	
VI.	Citroideae <u>Citrus</u>	(60)		1 (Lv)		1			1	1		1 (P)	6 (Bk)	1
,	Fortunella	(6)	3 F1)	-	,							(=)	(DX)	
	<u>Murraya</u> Poncirus	(9) (1)	с т)	1							1			
MELIA	CEAE													
	<u>Ptaeroxylon</u>	(1)												
MALPI	GHIACEAE													
	Malpighia	(30)												

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12		V 14		16	 17	18	19	20		22	23	24	25	26	27	
*******		<u> </u>			· <u>-</u> -											
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1			1	1 (P)	1	6				1 (Ft)		·				
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Appendix IV (cont'd.)

			1	2	3	4	5	6	7	8	9	10
RUTAC I. R I.	utoideae Zanthoxyleae <u>Fagara</u>	(200)										
	<u>Melicope</u> Zanthoxylum	(50) (15)									1	3 (Bk)
2.	Ruteae <u>Boenning-</u> <u>hausenia</u> <u>Ruta</u>	(1) (60)	an a an	·								
III.	Flindersioideae <u>Flindersia</u>	(20)				1 (Lv)						
v.	Toddalioideae <u>Casmiroa</u> Phellodendron	(6) (10)	-		1							
	Teclea	(25)		نلا)	у., В]	s) 						
VI.	Citroideae <u>Citrus</u>	(60)	1				1 (P)	1 (Ft)	1 (P)	1		
	Fortunella Murraya Poncirus	(6) (9) (1)					(-)	()	x - <i>y</i>			
MELIA	CEAE							~ ~	-		-	
	Ptaeroxylon	(1)		1 (Bk)	•							
MALPI	GHIACEAE											
	Malpighia	(30)										

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7	8	9	10	, 11	л 12	13	14	15	16	17	18	19	
		1	3 (Bk)	3 (Bk)	3 (Bk)	I (Bk)			1 (Sd)	1 (Ft)	2 (Bk)	1 (Bk)	
	·· ~ ••		<u>.</u>		•		1 1	1			·		•
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1 t)(P)	T												
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									,				

B. Triterpenoids other than saponins and

sapogenins (cont'd.)

- 71. Melianodiol
- 72. Melianol
- 73. Melianone
- 74. Melianotriol
- 75. Methyl 6-acetoxy-angolensate
- 76. Methyl angolensate
- 77. Methyl 6-hydroxyangolensate
- 78. Mexicanol
- 79. Mexicanolide
- 80. Mexicanolide C
- 81. Neohavanensin
- 82. Neoquassin
- 83. Nimbin
- 84. Nimbinin
- 85. Nimbolide
- 86. Nomibin
- 87. Nyasin
- 88. Obacunone
- 89. Picrasmin
- 90. Pseudo-epitaraxastane-diol
- 91. Pseudo-taraxasterol
- 92. Pseudelone-A
- 93. Pseudocedrelone-B
- 94. Quassin
- 95. Rutaevin
- 96. Salannin
- 97. Samaderine-A
- 98. Samaderine-B
- 99. Samaderine-C
- 100. Simarolide
- 101. Swietenin
- 102. Swietenolide
- 103. Taraxeral
- 104. Taraxerone
- 105. Trichilenone

- 106. Turraeanthin
- 107. Utilin
- 108. Veprisone
- V. Tetraterpenoids
 - 1. -Carotene
 - 2. -Carotene
 - 3. B-Carotene
 - 4. Cryptoxanthin
 - 5. Mutatochrome
 - 6. Semi-B-carotene
 - 7. Zeaxanthin

B. Triterpenoids other than saponins and

sapogenins (cont'd.)

- 10. Anthothecol
- 11. Aphanamixin
- 12. Arborinol
- 13. Azadiracione
- 14. Azadirone
- 15. Bauerenol
- 16. «-Boswellic acid
- 17. Bourjotone
- 18. Bein
- 19. Bruceine A
- 20. Bruceine B
- 21. Bruceine C
- 22. Bussein
- 23. Canaric acid
- 24. Candollein
- 25. Carapin
- 26. Cedrelone
- 27. Cedronine
- 28. Cedronyline
- 29. Chaparrin
- 30. Chaparrinone
- 31. **B**-Citraurin
- 32. 7-Deacetoxy-3-deacety1-7oxo-khivorin
- 33. 7-Deacetoxy-7-oxo-dihydro--gedunin
- 34. 7-Deacetoxy-7-oxogedunin
- 35. 7-Deacetoxy-7-oxokhivorin
- 36. 7-Deacetylgedunin
- 37. 3-Deacetyl-khivorin

- 38. Deacetyl-nomilin
- 39. 3-Dehydromexicanol
- 40. Deoxy-limonin
- 41. 6-Deoxy-swietenolide
- 42. 6 d, 11B-Diacetoxy gedunin
- 43. Dihydrogedunin
- 44. Entandrophragmin
- 45. Epi-lupeol
- 46. Epoxy-malabaricol
- 47. Eurycoma-lactone
- 48. Fissinolide
- 49. Flindissol
- 50. Gedunin
- 51. Glaucarubin
- 52. Glaucarubinone
- 53. Glaucarubolone
- 54. Grandifoliolinone
- 55. Havanensin
- 56. Heudelottin
- 57. Hirtin
- 58. 6-Hydroxy-angolensic acid-methyl ether
- 59. 6-Hydroxycarapin
- 60. Ichangin
- 61. Isoarborinol
- 62. 11-Keto- a -amyrin
- 63. Khayanthone
- 64. Khivorin
- 65. Lansic acid
- 66. Limonin
- 67. Limonin diosphenol
- 68. Malabaricanedial
- 69. Malabarical
- 70. Meldenin

APPENDIX V (cont'd.)

- II. Sesquiterpenes (cont'd.)
 - C. Eremophitone group
 - 1. Nootkatone
 - 2. Valencene
 - D. Guaianolide group
 - 1. Aromadendrene
 - 2. *a-Chigadmarene*
 - 3. Cyclocolarenone
 - 4. Ledol
 - E. Selinene group
 - 1. Canarone
 - 2. **B**-Caryophyllene-epoxide
 - 3. Elemol
 - 4. (+)-Jujenol
 - 5. (+)-Junenol
- III. Diterpenoids
 - 1. Aphanamixol
 - 2. *a*-Camphorene
 - 3. Crocetin
 - 4. Geranyl-geraniol
 - 5. Incensole
 - 6. Nimbiol
 - 7. Sugiol

- IV. Triterpenoids
 - A. Triterpenoid saponins and sapogenins
 - 1. **<-**Amyrin
 - 2. **B**-Amyrin
 - 3. **B**-Citraurin
 - 4. Ifflaionic acid
 - 5. Lupeol
 - 6. Monninin
 - 7. Polygalic acid
 - 8. Polygala-prosapogenin
 - 9. Polygala-saponin-A
 - 10. Polygala-saponin-B
 - 11. Presenegenin
 - 12. Prosapogenin
 - 13. Sapteroxyloside
 - 14. Senegenin
 - 15. Senegin
 - 16. Xanthophyllum-Saponin
 - B. Triterpenoids other than saponins and sapogenins
 - 1. 11B-Aceroxygedunin
 - 2. Aglaiol
 - 3. Ailantholide
 - 4. Ailanthone
 - 5. Amarolide
 - 6. Amarolide-12-acetate
 - 7. **B**-Amyrin acetate
 - 8. Andirobin
 - 9. Angolensic acid

Key to a list of Terpenoids:

- I. Monoterpenoids
 - 1. d-Camphene
 - 2. **1**-Camphene
 - 3. $d-4^3$ -Carene
 - 4. $l \Delta^3$ -Carene-5, 6-epoxide
 - 5. Carvacrol
 - 6. 1,4-Cineole
 - 7. 1.8-Cineole
 - 8. Citral
 - 9. d-Citronellal
- 10. d-Citronellic acid
- 11. Cuminic aldehyde
- 12. p-Cymene
- 13. Diosphenol
- 14. Dipentene
- 15. Filifilone
- 16. Geranic acid
- 17. Geranial
- 18. d-Limonene
- 19. d-Linalool
- 20. **2**-Linalool
- 21. Linalool epoxide
- 22. *L*-Linalyl acetate
- 23. d-Menthone
- 24. Mullilam-diol
- 25. Myrcene
- 26. d-myrtenal
- 27. Nerol
- 28. Ocimene

- 29. Perilla alcohol
- 30. *l*-Phellandral
- 31. d- <- Phellandrene
- 32. d-**B**-Phellandrene
- 33. Phellandrinic acid
- 34. **1**-q-Pinene
- 35. *I***-B**-Pinene
- 36. Sabinene
- 37. *A*-Terpinene
- 38. Y-Terpinene
- 39. ℓ -Terpinenol-(4)
- 40. d-d-Terpineol
- 41. *I-d*-Terpineol
- 42. «-Thujene
- 43. d-Verbenol
- 44. Verbenone
- II. Sesquiterpenes
 - A. Bisaboiene group
 - 1. Bisabolene
 - 2. Bisabolol
 - B. Cadinene group
 - 1. Cadinene
 - 2. Cadinol
 - 3. Capaene
 - 4. Epi-khusinol

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All were											<u>.</u>	··			1						
					1	2	34	5	6	7 8	9	10	11	12	13	14	15	16	5	17	18
	RUTACI.	CEAE Rutoid e	ae																		
		<u>Evodia</u> Zantho	תור דיציא	(120) (15)	•				1			1	1								
		Medico	sma	(1)					-			Ŧ	-							•	
		Dictam Ruta	nus	((2) (60)	5					1			1								
×.		Boroni	a	(60)	5					-			-						Ì		
		<u>Eriost</u> Phebal	emon ium	(30) (36)						1									ļ		
		Zieria		(1)			1										1				
		Agatho Barosm		(170) (20)								1			1	1			i i		
		Calode	ndron	(2)																	
	III.	Empleu Flinders	<u>rum</u> ioideae	(1)																	
		Flinde	<u>rsia</u>	(20)																	
	` V.	Toddalic Acrony	id e ae chia	(40)																	
		Amyris		(20)																	
		Casima Phello	<u>roa</u> dendron	(2) (10)																	
	\$.	Skimmi	<u>a</u>	- (10)																	
	VI.	Vepris Citroide	ae	(20)																	
		Aegle		(1)							1		1	1							
		Atalan Citrus		(30) (60)	1	2	1?			2		1		1		1		1		1	1
•		Clause	na	(30)														1			
		Fortur Glycos		(1) (40)			•														
		Luvung Microc		(1) (5)						1	•										
•		Murray	a	(5) (9)	•																
<i></i>		Poncir	us	(1)																	
	SIMA	Tripha ROUBACEAE	sia	(2)																	
		Euryco	ma	(4)																	
		Quassi Simaba																			
		Simarc	uba 🌔	(40)																	
		Samade Ailant	ra J hus	(10)																	
		Brucea	Ĺ.	(10 - 12)																	
		Castel Perrie		(12) (12)																	
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(by genera) in survey)						
			II.	Sesquite	rpenes	
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		Appendix V	. (cont'd.)	
		whengry A	. (cont u.)	
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2 August - 1 - 1 - 1 - 1 - 1 - 1 - 1 - 1 - 1 -	III. Diterpenoid			
ing and Mines. An an Article Andreas and Artic Article Andreas and Article Andr		A. Triterpe	noid saponins and	sapogenins
	1234567	12345	6789101112	13 14 15 16
RUFACEAE (cont'd.)				
I. Rutoideae				
Evodia (120)		ł		
Zanthoxylum (15)		1 3		
Medicosma (1)			•	
Dictamnus (2				
Ruta (60)		ſ		
Boronia (60)		í		
Eriostemon (30)		}		
Phebalium (36)				
Zieria (1				
Agathosma (170)		ł		
Barosma (20)		1		
Calodendron (2)				
Empleurum (1)				
III. Flindersioideae	,			
Flindersia (20)		1		
V. Toddalioideae		-	-	
Acronychia (40)				
Amyris (20		}		
Casimaroa (2				
Phellodendron (ιό			
Skimmia (10)				
Vepris (20)				
VI. Citroideae	•]		
Aegle (1)		5		
Atalantia (30)				
Citrus (60)		1 1 1		
Clausena (30)				
Fortunella (1)				
Glycosmia (40)		i -		
Luvunga (1)				
Microcitrus (5)		1		
Murraya (9)			•	•
Poncirus (1)				:
Triphasia (2)				
SIMAROUBACEAE	·			
Eurycoma (4)				
Quassia	•			
Simaba				
Simarouba (40)		1		
Samadera		1		: :
Ailanthus (4)				
Brucea (10-	-12)	1		
Castela (12)				
Perriera (12				
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Appendix V. (cont'd.)

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Appendix V (cont'd.)	132					
			IV. 1	riterpenöl	(cont'd.)	
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RUTACEAE I. Rutoideae <u>Evodia</u> (120) <u>Zanthoxylum</u> (15) <u>Medicosma</u> (1) <u>Dictamnus</u> (2) <u>Ruta</u> (60)					3 2 1	
Boronia(60)Eriestemon(30)Pheballum(36)Zieria(1)Agathosma(170)Barosma(20)Calodendron(2)						
Calodendron(2)Empleurum(1)III. FlindersioideaeFlindersia(20)V. ToddalioideaeAcronychia(40)Amyris(20)						
Casimaroa(2)Phellodendron(10)Skimmia(10)Vepris(20)VI.CitroideaeAcgle(1)					1	
Atalantia(30)Citrus(60)Clausena(30)Fortunella(1)Glycosmia(40)Luvunga(1)Microcitrus(5)Poncitrus(1)Triphasia(2)	-		1		13 1 1 1 1	
SIMAROUBACEAE <u>Eurycoma</u> (4) <u>Quassia</u> <u>Simaba</u> (40) <u>Simarouba</u> <u>Samadera</u> <u>Ailanthus</u> (10) <u>Brucea</u> (10-12)					1	1
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BURS	ERACEAE													•	
	Boswellia	(24)										1			_
	Brusera Commiphora	(100) (100)													1
	Canarium	(75)										1			
	ACEAE													•	
I.	Cedreloideae Cedrela	(7)			•										
	Ptaeroxylon	(1)												•	
II.	Swietenioideae	(20)													,
•	Entandrophrag Khaya	(10)													
•	Pseudocedrela	(1)													
III.	<u>Swietenia</u> Melioideae	(5)												`	
•	Carapa	(15)													
	Xylocarpus	(5)													
	<u>Turraeanthus</u> Melia	(6) (9)													
	Aglaia	(300)				1	1						•		
•	<u>Aphanamixis</u> Dysoxylum	(23) (100)											- 1		
	Guarea	(160)												•	
	Lansium	(7)													
POLY	Trichilia GALACEAE	(230)													
	Xanthophyllum													•	
	<u>Monnina</u> Polygala	(80) (500)											1		
	rorygara	(500)											ł		

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pendix V	'. (c	ont'd.)

	III	Diterp	enoids									
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I. Cedreloideae Cedrela	(7)	1	1									
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Swietenia	(5)											
III. Melioideae				1								
Carapa	(15)									ì		
Xylocarpus Turraeanthus	(5) (6)			·						•		
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Guarea	(160)									:		
Lansium	(100)											
Trichilia	(230)							ì		;		
POLYGALACEAE Xanthophyllum	(40)											
Monnina	(40)					1		ſ				
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Appendix V- B-3

Appendix V (cont'd.)

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	53	54	55	56	57	58	59	60	61	62	63	64	65	66	67	,
BURSERACEAE															•	
<u>Boswellia</u> (24)																
Brusera (100)																
Commiphora (100)										_						
Canarium (75)	•									1					;	
MELIACEAE							_									
I. Cedreloideae							1									
<u>Cedrela</u> (7)																
Ptaeroxylon (1)																
II. Swietenioideae															ł	
Entandrophragma (20)		-				_					_					
Khaya (10)		1				2					1	3				
Pseudocedrela (1)																
Swietenia (5)															·	
III.Melioideae																
<u>Carapa</u> (15)									•							
Xylocarpus (5)																•
Turraeanthus (6)																
Melia (9)																
Aglaia (300)																
Aphanamixis (23)													-		1	
Dysoxylum (100)													1			
<u>Guarea</u> (160) Lansium (7)			_	-	-							-	_			
<u>Lansium</u> (7) Trichilia (230)			1	1	1							1	?			
POLYGALACEAE (230)																
Xanthophyllum (40)															i.	
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X. Quinoline Group

в.	Fur	oquinolines and Related Alkaloids		
		-	30.	Kokusaginine (6,7-Dimethoxy-dictamnine)
	2.	Acronydidine	31.	
	3.	Acrophyllidine		Lunacridine
		Acrophylline (9-(3-methylbut-2-enyl)-7-	33.	(-) Lunacrine
				Lunacrinol
		Choisyine	35.	(+)-Lunacrinol (Isobalfourodine)
				Lunasine
	7.	Dubinidine	37.	Lunidine
	8.	Dubinine	38.	Lunine
		Evodine	39.	Maculine (6,7-Methylenedioxy-dictamnine)
		Evolatine	40.	Maculosidine (6,8-Dimethoxy-dictamnine)
		Evolitrine (7-Methoxy-dictamnine)	41.	Maculosine
]	12.	Evoxine (Haploperine)		
]	13.	Evoxoidine		
-	14.		44.	Nor-r-fagarine
		/		"Nor-orixine"
-	15.			O-Methyl balfourodinium+
		8-methoxy-dictamine)	47.	O-Methyl-luninium cation
	_			Orixine
			-	Pilokeanine
7	18.	Haplophylline	50.	Platydesmine
		Haplopine (7-Hydroxy-8-methoxy-dictamnine)	51.	Platydesmine acetate
			52.	
		(-)-Hydroxy-lunacridine		a 2-alkoxy-4-quinoline)
			-	Ribalinine
	23.			Ribalinium
			55.	Robustine (8-Hydroxy-dictamine)
			56.	Skimmianine (7,8-Dimethoxydictamnine)
		Ifflaiamine		
				olyl-quinoclidine
2	28.	Khaplofoline	1.	Quinine
	29.	Kokusagine (7,7-Methylenedioxy-dictamine)		· · · · · · · · · · · · · · · · · · ·
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IX.	Ou	inazolines
•	ĩ.	Aegelenine (1-Phenyl-7-hydroxytetrahydro-
		quinazoline-4-one)
	2.	quinazolone)
	3.	
	4.	
	5.	Glycosminine (2-Benzyl-4-quinazolone; Glycasmine)
	6.	7-Hydroxy-l-phenyldihydroquinazol-4-one
		inoline group
А.	S	imple Quinolines
	1.	
		2-Amyl-quinoline
	3.	2-Amyl-4-methoxy-quinoline
	4.	2- [4 ⁻ -(3",4"-Methylene-dioxyphenyl)]-n-Butyl- quinolone
	5.	Casimiroine (1-Methyl-4-methoxy-7,8-methy-
		lene-dioxy-2-quinolone)
	6.	Casimiroitine (1-Methyl-4-0-ethyl-7,8-methy-
		lene-dioxy-2-quinolone)
	7.	Cuspareine
	8.	Cusparidine
	9.	Cusparine
	10.	1,2-Dimethyl-4-quinolone
	11.	3-Dimethylally-4-methoxy-2-quinolone
	12.	3-Dimethylallyl-4-dimethylallyloxy-2-quinolone
	13.	Dubamine
	14.	Eduleine (N-methyl-2-phenyl-7-methoxy-4-quinolone)
	15.	Eduline (N-Methyl-2-phenyl-6-methoxy-4-quinolone)
	16.	Edulinine

17. Edulirine (4,8-Dimethoxy-2-quin-

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olone
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- 18. Evocarpine 19. Fagaramide
- 20. Foliosidine
- 21. Galipine
- 22. Galipoidine
- 23. Galipoline
- 24. Graveoline [(N-Methyl-2-(3',4'-methylene-di-
- oxyphenyl)4-quinolone] 25. Graveolinine (2-(3',4'-Methylenedioxyphenyl)-4-methoxy-quin
 - oline)
- 26. 3-Isopentenyl-4-methoxy-7,8methylenedioxy-2-quinolone
- 27. Lunamarine (N-Methyl-2(3',4'methylenedioxyphenyl-7methoxy-4-quinolone)
- 28. 1-Methyl-2-quinolone
- 29. N-Methyl-2-quinolone
- 30. 2-Pentylquinoline
- 31. 4-Methoxy-2-pentyquinoline
- 32. 2-Phenyl-4-methoxy-quinoline
- 33. Quinaldine
- 34. Quinoline
- 35. Rutamine (Dimethyl-graveoline)

App. VI

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- D. Canthin-6-ones
- 1. Canthinone (Canthin-6-one)
- 2. 4,5-Dimethoxycanthin-6-one
- 3. Nigakinone (4-Methoxy-5-hydroxycanthin-6-one)
- 4. 5-Methoxycanthionone (5-Methoxy-canthin-6-one)
- 5. 4-Methylthio-cantin-6-one)
- E. The carbazole group
- 1. Girinimbine
- 2. Glycozolidine (5,7-Dimethyl-carbazole)
- 3. Glycozoline (3-Methyl-6-methoxy-carbazole
- 4. Heptaphylline
- 5. Murrayanine (1-Methoxy-3-formycarbazole)
- V. Isoquinoline group
 - A, 1, 1'-Benzylisoquinolines
 - 1. Tembetarine (N-Methyl-1-(+)-reticuline)
 - B. The aporphine group
 - 6-Hydroxy-2,3,5-trimethoxy-NN-dimethylaporphine
 - 2. Isocorydine (Artabotrine; Lauteanine)
 - 3. Isocorydine methiodide
 - 4. Laurifoline
 - 5. Magnoflorine
 - 6. Magnoflorine iodide
 - 7. (+) N-Methylcorydine
 - 8. N-Methyl-corydinium cation
 - 9. N-Methylisocorydine
 - 10. N-Methyl-isocorydinium cation
 - 11. Quaternary aporphine
 - 12. Xanthoplanine
 - C. Protoberberine
 - 1. Berberine
 - 2. Jatrorrhizine (Jaterorhizine)
 - 3. **d**-l-Canadine methochloride ((-)-N-Methyltetrahydroberberine chloride)]

- 4. (-)-K-Canadine methoidide
- 5. N-Methyl- a -Canadine+
- 6. Palmatine (Calystegine; Gindarinine)
- 7. Phellodendrine
- D. Protopine group
- 1. Allocryptopine
- 2. **A**-Allocrytopine (B-homochelidonine; r-Fagarine)
- 3. **B**-Allocryptopine (r-Homochelidonine)
- 4. Fagarine II
- E. Phthalide-isoquinalines
- 1. (-)-&-Narcotine
- F. The **d**-Naphthaphenanthridines
- 1. Avicine
- 2. Chelerythrine (Toddaline)
- 3. Chelerythrine chloride
- 4. Chelerytrine
- 5. Dihydrochelerythrine
- 6. 7,8-Dimethoxy-2',3'-methylenedioxy-1,2benzophenanthridine
- 7. Nitidine
- 8. Oxynitidine
- VI. The oxizole group
 - 1. Halfordine
 - 2. Halfordinol
 - 3. Halfordinone
 - 4. N-Methyl-halfordinium chloride
- VII. Pyridines
 - 1. Trigonelline (Coffeorin)
- VIII. Pyridines
 - 1. Stachydrine

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APPENDIX VI

Distribution of Alkaloids (by genera) in "Rutales" (Compiled from literature survey)

Key to a list of Alkaloids:

- I. Acridine group
 - 1. 1, 3-Dimethoxy-10-methylacridone
 - 2. 1,2,3-Trimethoxy-10-methyl-acridone
 - 3. N-methylacridone
 - 4. Acronycine
 - 5. Arborinine
 - 6. Evoprenine
 - 7. Evoxanthidine
 - 8. Evoxanthine (1-Methoxy-2, 3-methyenedioxy-N-methyl-acridone)
 - 9. Melicopicine
 - 10. Melicopidine
 - 11. Melicopine
 - 12. Xanthevodine
 - 13. Xanthoxoline
- II. Alkaloid amines (Including the
 - **B**-Phenyl-ethylamine)
 - 1. Aegeline
 - 2. Candicine (**B**-p-Hydroxy-phenylethyl-trimethylammonium hydroxide)
 - 3. Coryneine (3-Hydroxy-candicine)
 - 4. Feruloputrescine
 - 5. Jaborandine
 - 6. Noradrenaline
 - 7. N-Benzaytyramine
 - 8.(±)-N-benzoyl(2-hydroxy-2-(4'-methoxyphenyl) ethylamine
 - 9. N-Methyl-anthranilic acid
 - 10. N-Methyl-anthranlic acid methyl ether
 - 11. N, N-Dimethyl-4-methoxy-phenethylamine
 - 12. Nor-adreneline (Arterenol; Nor-epinephrine
 - 13. **Q**-Octopamine (1-Nor-synephrine)
 - 14. O-Methyl-tyramine-N-methylcinnamide (Herclavin)
 - 15. 2-synephrine
 - 16. Tyramine

- III. Imidazole group
 - 1. Casimiroedine
 - 2. Isopilocarpine
 - 3 N, N-Dimethylhistamine
 - 4. Pilocarpidine (De-N-methyl-pilocarpine)
 - 5. Pilocarpine
 - 6. Pilosine
 - 7. Zapotidine
- IV. Indole group
 - A. Simple indole bases
 - 1. Indole
 - 2. 6-Methoxy-Nb-dimethyl-tryptamine
 - 3. 5-Methoxy-Nb, Nb-dimethyl-tryptamine
 - 4. Nb, Nb-Dimethyl-tryptamine (Nigerine)
 - 5. Tryptamine
 - B. Carboline alkaloids
 - 1. Hermaline (3,4-Dihydro-harmine; Harmidine)
 - 2. Harmine (?Banisterine; Passiflorine; Telepathine; ?Yageine)
 - 3. Tetrahydro-harmine (Leptoflorine)
 - C. The quinazoline carboline
 - 1. Evodiamine
 - 2. Hortiacine
 - 3. Hortiamine
 - 4. Hydroxy-evodiamine
 - 5. Rhetsine (dl-Evodiamine)
 - 6. Rhetsinine
 - 7. Rutaecarpine

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APPENDIX VI (cont'd.)

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<u>Glycosmis</u> (30) <u>Glycosmis</u> (40) <u>Murraya</u> (9) <u>Poncirus</u> (1) SIMAROUBACEAE II.Simarouboideae <u>Picrasma</u> (17) <u>Picrolemma</u> (3) MALPIGHIACEAE <u>Banisteria</u> (75) <u>Cabi</u> (1) <u>Banisteriopsis</u>(100)

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APPENDIX VII

The Distribution of Saponins in the Rutales (from Amarasingham <u>et al</u>, 1964)

Genus	sapo	nins
	Present	Absent
Rutaceae		
<u>Xanthoxylum</u> Glycosmis	+1	-2
Micromelum	+1 ,	-1
Luvunga Atalantia	+1	-1
Cneoraceae Simaroubaceae		
Eurycoma		-1
Picrodendr aceae Burseraceae		
<u>Triomma</u> <u>Canarium</u> <u>Dacryodes</u> Santiria	c	-1 -2 -2 -1
Meliaceae		
Walsura Aphanamixis Amoora? Aglaia? Chisocheton Dysoxylum	+2 +I	-1 -2 -1 -6 -2 -4
Akaniaceae Malpighiaceae		
Hiptage		-1
Trigoniaceae Vochysiaceae Tremandraceae		
Polygalaceae Xanthophyllum	+1	-1

APPENDIX VIII

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The Occurrence of Alkaloids in Rutales (from Li and Willaman, 1968)

	% of alkaloid plants in those	Approx. no. of species in	Species for Alk	
	tested	family	Positive	Negative
Rutaceae	60%	1,300	181	103
Cneoraceae				
Simaroubaceae	50%	200	14	13
Picrodendroacea	e			
Burseraceae	20%	600	2	7
Meliaceae	40%	800	20	29
Akaniaceae	100%	1	1	-
Malpighiaceae	30%	850	7	14
Trigoniaceae				
Vochysiaceae				
Tremandraceae	50%	30	1	1
Polygalaceae	50%	700	3	3

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N.
APPENDIX IX

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Distribution of Phenolic Constituents (by genera) in 1

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I. Choisya (7) -1 -1 -1 -1 Evodia (120) +1 +1 +1 Zanthoxylum (15) +2 +2,-1 -2 Boronia (60) +1 +1 +1 Correa (11) +1 +1 +1 Crowea (4) +1 +1 +1 Eriostemon (30) +1 +1 -1 Coleonema (6) +1 +1 +1 V. Phellodendron(10) -1 Ptelea (3) +1 -1 -2 Skimmia (10) -1 -1 -1 VI. Citrus (60) -1 -1 -1 Murraya (9) -1 -1 -1 CNEORACEAE -1/1 -1/1 -1/1 CNEORACEAE -1/1 -1/1 -1/1 SIMAROUBACEAE +1/1 +1/1 -2/4 Quassia (40) -1 Ailanthus (10) +1 +1 BURSERACEAE -2/2 +1/1, -1/1 -3/4 -2/2 Cedrea (7) -2 Swietenia (5) +1 +1 MaLPIGHIACEAE -1/1, +2/2 -1/1, +2/2 -4/4 -2/2	-1(
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	+4, -1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	+1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	+2
$\begin{array}{c} \hline \hline \text{Correa} & (11) & +1 & +1 & +1 & +1 \\ \hline \text{Crowea} & (4) & +1 & +1 & +1 & +1 \\ \hline \text{Eriostemon} & (30) & +1 & +1 & +1 & +1 \\ \hline \text{Eriostemon} & (30) & +1 & +1 & +1 & +1 \\ \hline \text{Coleonema} & (6) & +1 & +1 & +1 & +1 \\ \hline \text{V. Phellodendron(10)} & & -1 \\ \hline \text{Ptelea} & (3) & +1 & -1 & -2 \\ \hline \text{Skimmia} & (10) & -1 & -1 & -1 \\ \hline \text{Ptelea} & (3) & +1 & -1 & -2 \\ \hline \text{Skimmia} & (10) & -1 & -1 & -1 \\ \hline \text{VI. Citrus} & (60) & -1 & -1 & -1 \\ \hline \text{Glycosmis} & (40) & & -1 \\ \hline \text{Murraya} & (9) & -1 & -1 & -1 \\ \hline \text{Poncirus} & (1) & -1 & -1 & -1 \\ \hline \text{CNEORACEAE} & & -1/1 & -1/1 & -1/1 \\ \hline \text{CNEORACEAE} & & +1/1 & +1/1 & -2/4 \\ \hline \text{Quassia} & (40) & & & -1 \\ \hline \text{MELIACEAE} & & +2/2 & +1/1, -1/1 & -3/4 & -2 \\ \hline \frac{\text{Cedrea}}{\text{Swietenia}} & (5) & +1 & +1 \\ \hline \text{Melia} & (9) & +1 & -1 & -1 \\ \hline \text{Ailonia} & (1) & & & -1 \\ \hline \text{MALPIGHIACEAE} & & -1/1, +2/2 & -4/4 & -2 \\ \end{array}$	-1
$\begin{array}{c} \hline Crowea & (4) & +1 & +1 & +1 & +1 \\ \hline Eriostemon & (30) & +1 & +1 & +1 & -1 \\ \hline Coleonema & (6) & +1 & +1 & +1 & +1 \\ \hline V. & Phellodendron(10) & & -1 \\ \hline Ptelea & (3) & +1 & -1 & -2 \\ \hline Skimmia & (10) & -1 & -1 & -1 \\ \hline VI. & Citrus & (60) & -1 & -1 & -1 \\ \hline VI. & Citrus & (60) & -1 & -1 & -1 \\ \hline Murraya & (9) & -1 & -1 & -1 \\ \hline Poncirus & (1) & -1 & -1 & -1 \\ \hline CNEORACEAE & & -1/1 & -1/1 & -1/1 \\ \hline CNEORACEAE & & -1/1 & -1/1 & -1/1 \\ \hline Quassia & (40) & & -1 \\ \hline MurserACEAE & & +1/1 & +1/1 & -2/4 \\ \hline Quassia & (40) & & -1 \\ \hline MELIACEAE & & +2/2 & +1/1, -1/1 & -3/4 & -2 \\ \hline Cedrea & (7) & & -2 \\ \hline Swietenia & (5) & +1 & +1 \\ \hline Melia & (9) & +1 & -1 & -1 \\ \hline MALPIGHIACEAE & & -1/1, +2/2 & -1/1, +2/2 & -4/4 & -2 \\ \hline \end{array}$	+1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	+1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	-1
V. Phellodendron(10) -1 -1 <u>Ptelea</u> (3) +1 -1 -2 <u>Skimmia</u> (10) -1 -1 -1 VI. Citrus (60) -1 -1 -1 <u>Glycosmis</u> (40) -1 <u>Murraya</u> (9) -1 -1 -1 <u>Poncirus</u> (1) -1 -1 -1 <u>CNEORACEAE</u> -1/1 -1/1 -1/1 <u>Cneorum</u> (2) -1 -1 -1 <u>SIMAROUBACEAE</u> +1/1 +1/1 -2/4 <u>Quassia</u> (40) -1 <u>Ailanthus</u> (10) +1 +1 BURSERACEAE <u>Protium</u> (2) <u>MELIACEAE</u> +2/2 +1/1, -1/1 -3/4 -2 <u>Cedrea</u> (7) -2 <u>Swietenia</u> (5) +1 +1 <u>Melia</u> (9) +1 -1 -1 <u>Aitonia</u> (1) -1/1, +2/2 -4/4 -2	?
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	-1
VI. $\overrightarrow{\text{Citrus}}$ (60) -1 -1 -1 -1 $\overrightarrow{\text{Glycosmis}}$ (40) -1 -1 -1 $\overrightarrow{\text{Murraya}}$ (9) -1 -1 -1 -1 $\overrightarrow{\text{Poncirus}}$ (1) -1 -1 -1 -1 CNEORACEAE -1/1 -1/1 -1/1 $\overrightarrow{\text{Cneorum}}$ (2) -1 -1 -1 -1 SIMAROUBACEAE +1/1 +1/1 -2/4 $\overrightarrow{\text{Quassia}}$ (40) -1 $\overrightarrow{\text{Ailanthus}}$ (10) +1 +1 BURSERACEAE +2/2 +1/1, -1/1 -3/4 -2 $\overrightarrow{\text{Cedrea}}$ (7) -2 $\overrightarrow{\text{Swietenia}}$ (5) +1 +1 $\overrightarrow{\text{Melia}}$ (9) +1 -1 -1 $\overrightarrow{\text{Aitonia}}$ (1) -1/1, +2/2 -1/1, +2/2 -4/4 -2	-2
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	-1
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	-1
$\begin{array}{c cccc} \hline Poncirus & (1) & -1 & -1 & -1 & -1 \\ \hline CNEORACEAE & & -1/1 & -1/1 & -1/1 \\ \hline Cneorum & (2) & -1 & -1 & -1 \\ \hline SIMAROUBACEAE & & +1/1 & +1/1 & -2/4 \\ \hline Quassia & (40) & & -1 \\ \hline Ailanthus & (10) & +1 & +1 \\ \hline BURSERACEAE & & & & & \\ \hline Protium & (2) & & & \\ \hline MELIACEAE & & +2/2 & +1/1, -1/1 & -3/4 & -2 \\ \hline Cedrea & (7) & & & -2 \\ \hline Cedrea & (5) & +1 & +1 \\ \hline Melia & (9) & +1 & -1 & -1 \\ \hline Aitonia & (1) & & & & \\ \hline MALPIGHIACEAE & & -1/1, +2/2 & -1/1, +2/2 & -4/4 & -2 \\ \hline \end{array}$	-1
$\begin{array}{cccc} \underline{Cneorum} & (2) & -1 & -1 & -1 \\ \text{SIMAROUBACEAE} & & +1/1 & +1/1 & -2/4 \\ \underline{Quassia} & (40) & & -1 \\ \underline{Ailanthus} & (10) & +1 & +1 \\ \end{array}$ $\begin{array}{cccccccccccccccccccccccccccccccccccc$	-1
$\begin{array}{cccc} \underline{Cneorum} & (2) & -1 & -1 & -1 \\ \text{SIMAROUBACEAE} & & +1/1 & +1/1 & -2/4 \\ \underline{Quassia} & (40) & & -1 \\ \underline{Ailanthus} & (10) & +1 & +1 \\ \end{array}$ $\begin{array}{cccccccccccccccccccccccccccccccccccc$	-1
$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	-1,
$\begin{array}{cccc} \underline{\text{Quassia}} & (40) & & -1 \\ \underline{\text{Ailanthus}} & (10) & +1 & +1 \\ \\ \text{BURSERACEAE} & & & & \\ \underline{\text{Protium}} & (2) \\ \text{MELIACEAE} & & & +2/2 & +1/1, -1/1 & -3/4 & -2 \\ \underline{\text{Cedrea}} & (7) & & & -2 \\ \underline{\text{Cedrea}} & (5) & +1 & +1 \\ \underline{\text{Melia}} & (9) & +1 & -1 & -1 \\ \underline{\text{Aitonia}} & (1) & & & -1 \\ \end{array}$	-1
$\begin{array}{c cccc} \hline Ailanthus & (10) & +1 & +1 \\ BURSERACEAE & & & & \\ \hline Protium & (2) \\ \hline MELIACEAE & & +2/2 & +1/1, -1/1 & -3/4 & -2 \\ \hline \underline{Cedrea} & (7) & & -2 \\ \hline \underline{Swietenia} & (5) & +1 & +1 \\ \hline \underline{Melia} & (9) & +1 & -1 & -1 \\ \hline \underline{Aitonia} & (1) & & & -1 \\ \hline MALPIGHIACEAE & & -1/1, +2/2 & -1/1, +2/2 & -4/4 & -2 \\ \hline \end{array}$	-2,
BURSERACEAE Protium (2) MELIACEAE $+2/2$ $+1/1$, $-1/1$ $-3/4$ -2 <u>Cedrea</u> (7) -2 <u>Swietenia</u> (5) $+1$ $+1$ <u>Melia</u> (9) $+1$ -1 <u>Aitonia</u> (1) -1 -1 MALPIGHIACEAE $-1/1$, $+2/2$ $-1/1$, $+2/2$ $-4/4$ -2	-1
MELIACEAE $+2/2$ $+1/1, -1/1$ $-3/4$ -2 Cedrea (7) -2 Swietenia (5) $+1$ $+1$ Melia (9) $+1$ -1 -1 Aitonia (1) -1 -1 MALPIGHIACEAE $-1/1, +2/2$ $-1/1, +2/2$ $-4/4$ -2	1?
MELIACEAE $+2/2$ $+1/1, -1/1$ $-3/4$ -2 Cedrea (7) -2 Swietenia (5) $+1$ $+1$ Melia (9) $+1$ -1 -1 Aitonia (1) -1 -1 MALPIGHIACEAE $-1/1, +2/2$ $-1/1, +2/2$ $-4/4$ -2	+1, +2
$\begin{array}{ccc} \underline{\text{Cedrea}} & (7) & -2 \\ \underline{\text{Swietenia}} & (5) & +1 & +1 \\ \underline{\text{Melia}} & (9) & +1 & -1 & -1 \\ \underline{\text{Aitonia}} & (1) & -1 \\ \underline{\text{MALPIGHIACEAE}} & -1/1, +2/2 & -1/1, +2/2 & -4/4 & -2 \\ \end{array}$	
$\begin{array}{c cccc} & & & \\ \hline Swietenia & (5) & +1 & +1 \\ \hline Melia & (9) & +1 & -1 & -1 \\ \hline Aitonia & (1) & & -1 \\ \hline MALPIGHIACEAE & -1/1,+2/2 & -1/1,+2/2 & -4/4 & -2 \\ \hline \end{array}$	
Melia (9) $+1$ -1 -1 Aitonia (1) -1 MALPIGHIACEAE $-1/1, +2/2$ $-1/1, +2/2$ $-4/4$ -2	+2
Aitonia(1) -1 MALPIGHIACEAE $-1/1, +2/2$ $-1/1, +2/2$ $-4/4$ -2	-1
MALPIGHIACEAE $-1/1, +2/2, -1/1, +2/2, -4/4, -2$	-1
	-1
$\frac{\text{Heteropterrs}(50)}{\text{Hiptage}} (25) +1 +1 -1$	+1
$\frac{\text{Inpugge}}{\text{Malpighia}} (30) +1 +1(\text{L.P}) -1$	~1
Tristellateia(22) -1	+1
TREMENORACEAE $-1/1$ $-2/2$	-2
Platytheca (1) -1 -1	-1
Tetratheca (25) -1	-1
POLYGALACEAE $-1/1$ $-1/1$ $-1/1$	-1
Polygala (500) -1 -1 -1	-1

(From Bate-Smith, 1957; 1962)*

*Fractions represent numbers of compounds/species Key to symbols: D = Leuco-delphinidin K = kaempfe: Cy = Leuco-cyanidin E = ellagic M = myricetin Caff.= caffeic

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 $\tilde{M} = myricetin$ Q = Quercetin

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y genera) in Rutales

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		Chem	ical	data				
on	Leuco D	anthocya Cy	nins M	Fiavonols Q	к	Phenolic : E	acids Caff.	
<u>2n</u>	-10/12 +5/5 -1 +1 -2 +1 +1 +1 +1 +1 -1 +1 -1	Cy -10/11 +4/10 -1 +1 +2 -1 +1 +1 +1 -1 ? -1 -2	M +5/5 -1 +1 -2 +1 +1 +1 +1 +1 -1 +1 -1	Q -4/4 +9/10 +1 +1 +1 +1 +1 +1 +1 +1 +1 +1 +1 +1	K -4/4 +8/8 +1 +1 +1,-1 +1 +1 +1 +1 +1 +1 ? +1	E -12/13 -1 -1 -2 -1 -1 -1 -1 -1 -1 -1 -1		-
1.		-2 -1 -1 -1 -1 -1/1 -1 -1/1 -1 -1/1 -1 -1/1 -1 -1/1 -1/2 -1/2 +1/2 +2 +2	-1 -1 -1/1	-1 -1 $+1/1$ $+1$ $-1/1,+1/3$ -1 $+3$ $+1/2$ $+2$? -1 -1 + $1/1$ +1 +1/3 -1 -1 +3 + $1/2$ +2	$ \begin{array}{c} -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\$	-1 -1 +1 -1 +1/1 -1 +1/2,-2/2 -1 -1,+2 +1/1,-1/1 -1,+1 +2/2,-1/1 +1	
ŗ	$ \begin{array}{c} -1 \\ -1 \\ -4/4 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1$	$ \begin{array}{c} -1 \\ -1 \\ /2,+2/2 \\ -1 \\ +1 \\ -1 \\ +1 \\ -2/2 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1$	$-1 \\ -1 \\ -1 \\ -1 \\ -1 \\ -1 \\ +2/2 \\ +1 \\ +1 \\ -1/4 \\ -4$	$ \begin{array}{r} +1 \\ -3/3,+1/1 \\ +1 \\ -1 \\ -1 \\ -1 \\ +1/1 \\ +1/1 \\ +1 \\ ? \\ -1/1,+1/3 -1 \end{array} $	+1 -1 +1 -2/2 1, -1 +1	2 -4/4 -1 -1 -1 1,+1/1 -1 +1 2 -1/4	$ \begin{array}{c} +1 \\ -1 \\ -2/2 \\ +1 \end{array} \\ \begin{array}{c} -1 \\ -1 \\ -1 \\ -1/1 \\ -1 \end{array} \\ +2/2, -2/2 \\ -2, +2 \end{array} $	

s/species

K = kaempferol E = ellagic Caff.= caffeic acid

APPENDIX X

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Cyanogenesis in Rutales (from Gibbs (MSS))

Family	()thers ? -	G	ibbs ? -
Rutaceae	4/12	18/25	2/3	11/15
Cneoraceae		1/1		1/1
Simaroubaceae				2/3
Picrodendraceae				1/1
Burseraceae	1/2	1/1		
Meliaceae	5/5	2/3		1/1 2/2
Akaniaceae		1/1		1/1
Malpighiaceae				2/3 2/3
Trigoniaceae				
Vochysiaceae				
Tremandraceae		1/1		2/2 1/3
Polygalaceae		2/4		2/4

* Fraction represent numbers of Genera/Species

APPENDIX XI

The	Oco	urrence	of	Ca	lcium	Oxalate
(fı	com	Metcalfe	e ar	nd	Chalk,	1950)

	Solitary crystals	Cluster crystals	Styloids	Raphides	Crystal sand		
Rutaceae	xx	xx	(x)	(x)	(x)		
Cneoraceae	(x)						
Simaroubaceae	x	x	(x)				
Picrodendraceae							
Burseraceae	x	x					
Meliaceae	x	x					
Akaniaceae	(x)	x					
Malpighiaceae	x	x	x				
Trigoniaceae	x	x					
Vochysiaceae	x	x					
Tremandraceae	x	x					
Polygalaceae	×	x					

Key to symbol: ()= limited in distribution

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Appendix II Paper chromatographic results of phenolic acids and coumarins

in "Rutales" (by writer and Galang) (Numbering as in Key)

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