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A CHEMICAL APPRAISAL VIA LEAF FLAVONOIDS OF DAHLGREN'S LILIIFLORAE

CHRISTINE A. WILLIAMS, JEFFREY B. HARBORNE and *BRIAN MATHEW

Department of Botany, School of Plant Sciences, University of Reading, Whiteknights, Reading, RG6 2AS, U.K.; *Royal Botanic Gardens, Kew, Richmond, Surrey, U.K.

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IN MEMORY OF TONY SWAIN, 1922–1987

Key Word Index—Liliiflorae; flavones; flavonols; flavone *C*-glycosides; chemotaxonomy.

Abstract—Leaf flavonoids have been identified in 91 species of the new superorder Liliiflorae. The results have been combined with those obtained earlier on 181 species so that data are now available on 29 of the 52 families. The flavonols quercetin and kaempferol occur in about half the sample, while the flavones apigenin and luteolin are present in about a fifth. *O*-Methylated flavonoids (isorhamnetin, tricin, diosmetin, etc.) are uncommon, as are other classes of flavonoid. The rare flavonol, 3-*O*-methyl-8-*C*-methylquercetin, previously reported in *Dasylirion acrotrichum*, was found in three other members of the family Nolinaceae and seems to characterize it. Otherwise, the 29 families fall into two groupings: 23 with flavonols in the leaves and six with flavonols and flavones. The limited diversity in flavonoid patterns and in other chemical features within these plants argues for a more conservative treatment at the family level.

INTRODUCTION

In the most recent systematic treatment [1] of the monocotyledons, based on a complete re-evaluation of extant data [2] and using in part a cladistic approach [3], new families have been created and old families have been reinstated. This is especially true from within the old Liliaceae complex and several new affinities are proposed at the ordinal level. Thus the new superorder Liliiflorae now contains 52 families divided into five orders (Table 1). Some chemical results have been used in this reevaluation but flavonoid data have not been compared to any extent. Earlier surveys of leaf flavonoid patterns in most of the other monocotyledonous families have indicated that these compounds, particularly their frequencies of occurrence, are characteristic at the family level [4]. We decided, therefore, to examine the leaf flavonoids within these plants to see if they provided any useful support for the creation or resurrection of so many families.

In this new classification [1] the orders and families are generally rather narrow and consequently the number of families is very numerous compared with previous systems. Five orders are recognised: Dioscoreales, Asparagales, Melanthiales, Burmanniales and Liliales and some 52 families are contained in the superorder. The Asparagales with 30 families includes the Alliaceae and Amaryllidaceae, while the Liliales is extended to include the Iridaceae and Orchidaceae. By contrast, the Liliaceae is reduced to a family of 13 genera and 385 species with many of the more traditional members transferred to other families, i.e. to become the Hyacinthaceae, Funkiaceae, Asphodelaceae in the order Asparagales and the Alstroemeriaceae, Colchicaceae and Uvulariaceae in the order Liliales. Hence, when compared with previous treatments this new system has an extremely narrow family concept within the group, based largely on Huber [5]. Recent serological results [6, 7] give some support for the new system in that there is generally little or no serological reaction between taxa, which have now been referred to different orders, while there is a strong reaction between at least some families within each order. Additionally, families (or orders) characterized by a combination of starchy endosperm, paracytic or tetracytic stomata, *Strelitzia* type epicuticular wax and UV fluorescent compounds in the cell walls have been excluded from the Liliiflorae.

Some previous chemical and anatomical investigations for the Liliaceae *sensu latu* and other families now included in the Liliiflorae are described by Hegnauer [8, 9] and are summarized in Table 1. The most characteristic chemical constituents of the group are chelidonic acid and steroidal sapogenins. A number of the smaller families have not been surveyed for these compounds so it is difficult to comment on their distribution in the new superorder. However, chelidonic acid is notably absent from the Uvulariaceae, Calochortaceae and Liliaceae in the Liliales, from five of the seven families of the Dioscoreales and at least eight of the 30 families of the Asparagales. Again, in the Liliales steroidal sapogenins are absent from the Uvulariaceae and Calochortaceae and also from the Colchicaceae. Alkaloids have been recorded from only seven families but are represented in all orders except the Burmanniales. Cardiac glycosides are known only from the Convallariaceae and Hyacinthaceae, while anthraquinones occur in the Asphodelaceae and Xanthorrhoeaceae and the non-protein amino acid, azetidine 2-carboxylic acid in 12 genera of the Asparagaceae.

The superorder Liliiflorae is characterized anatomically by the presence of raphides in all orders except the greater part of the Burmanniales and Liliales, while silica bodies are restricted to some members of the Orchida-

Table 1. The non-flavonoid chemical constituents of the Liliiflorae*

Order, Family	Chelidonic acid	Steroidal sapogenins	Alkaloids	Raphides present	Plants mucilaginous	Others†
Dioscoreales						
Trichopodaceae	—	—	—	+	—	ceryl alcohol
Dioscoreaceae	+	+	+	+	—	
Taccaceae	—	+	—	+	—	
Stemonaceae	—	—	+	+	—	
Trilliaceae	+	+	—	+	—	
Smilacaceae	+	+	—	+	—	
Petermanniaceae	—	—	—	+	—	
Asparagales						
Philesiaceae	—	+	—	—?	—	
Luzuriagaceae	—	—	—	+	—	
Convallariaceae	+	+	—	+	—	
Asparagaceae	+	+	—	+	—	
Ruscaceae	+	+	—	+	—	
Herreriaceae	+	+	—	—	—	
Dracaenaceae	+	+	—	+	+	
Nolinaceae	—	+	—	pseudo	—	
Asteliaceae	—	+	—	+	—	
Hanguanaceae	nd	nd	nd	silica bodies	—	
Dasypogonaceae	+	nd	nd	+	—	
Calectasiaceae	nd	nd	nd	—	—	
Blandfordiaceae	nd	nd	nd	—	—	
Xanthorrhoeaceae	—	—	—	+	—	Acoroid resin
Agavaceae	+	+	—	pseudo	+	Ascorbic acid
Hypoxidaceae	+	—	—	+	—	
Tecophilaeaceae	nd	nd	nd	—	—	
Cyanastraceae	—	—	—	—	—	
Eriospermaceae	nd	nd	nd	—	?	Aleurone but no starch
Ixioliriaceae	+	nd	—	+	—	
Phormiaceae	+	nd	nd	+	—	
Doryanthaceae	nd	nd	nd	+ (in tepals)	—	
Hemerocallidaceae	nd	nd	nd	+	?	
Asphodelaceae	+	—	—	+	+	Fructans but no starch
Anthericaceae	+	+	—	+		Cyanogens in *Chlorophytum*
Aphyllanthaceae	—	+	—	+	—	
Funkiaceae	nd	+	nd	+ fruit of *Hosta*	—	
Hyacinthaceae	+	+	—	+	+	Fructans and starch. Salicylic acid in *Hyacinthus*
Alliaceae	+	+	—	+ some genera	+	Allylic sulphide
Amaryllidaceae	+	—	+	+	+	
Melanthiales						
Melanthiaceae	+	+	+ some genera	+	—	
Campynemaceae	nd	nd	nd	+	—	
Burmanniales						
Burmanniaceae	nd	nd	nd	—?	—	γ-Methylene glutamic acids
Thismiaceae	nd	nd	nd	+	—	
Corsiaceae	nd	nd	nd	—	—	
Liliales						
Alstroemeriaceae	+	+	—	+	—	Tuliposides starch
Colchicaceae	+	—	+	—	—	Starch

Table 1. *Continued*

Order, Family	Chelidonic acid	Steroidal sapogenins	Alkaloids	Raphides present	Plants mucilaginous	Others†
Uvulariaceae	—	—	—	—	—	
Calochortaceae	—	—	+	—	—	
Liliaceae	—	+	(+)	—	—	Tuliposides γ-methylene glutamic acids
Geosiridaceae	nd	nd	nd	—	—	
Iridaceae	(+)	+	(+)	+	+	Carbohydrates
Apostasiaceae	nd	nd	nd	+	?	
Cypripediaceae	nd	nd	nd	+	?	
Orchidaceae	nd	nd	nd	+	?	

*Classification according to Dahlgren *et al.* [1].

†Azetidine-2-carboxylic acid is recorded in Asparagaceae, anthraquinones in Asphodelaceae and Agavaceae and cardiac glycosides in Convallariaceae and Hyacinthaceae.

ceae, Apostasiaceae and Cypripediaceae.

There have been three major leaf flavonoid surveys of the Liliaceae *sensu latu* and related families [10–12], in which most of the major flavonoid classes were found to be represented. Thus, Williams [10] found that flavonols (quercetin and kaempferol) were the most characteristic constituents of the Liliaceae *sensu* Melchior [13] being present in some 40 and 42% of the sample, respectively, while simple flavones (luteolin and apigenin) were found in only 24 and 20% of taxa. On the other hand, methylated flavonoids such as diosmetin, tricin and isorhamnetin were of rare occurrence and flavone *C*-glycosides were found in only three and flavonoid sulphates in only one species. Proanthocyanidins were detected in 17 species and anthraquinone pigments identified in a number of *Aloe*, *Asphodeline* and *Asphodelus* species. In the related families Amaryllidaceae, Agavaceae and Xanthorrhoeaceae flavonols were found to be the major leaf constituents.

The results of Skrzypczakowa's [14] survey of 60 species were similar to those of Williams [10] but quercetin (in 67% of taxa), isorhamnetin (in 25%) and apigenin (in 30%) all appeared more frequently. The different sampling of the two surveys could account for the differences in recorded frequencies. The facts that Skrzypczakowa recorded apigenin instead of flavone *C*-glycosides in *Polygonatum latifolium* and apigenin instead of tricin in *Hyacinthus orientalis* are more difficult to explain. Bate-Smith [12] in a survey of 121 Liliaceae for phenolic constituents reported the frequent occurrence of flavonols in the subfamilies Allioideae, Lilioideae and Asparagoideae and their absence from the Asphodeloideae, Melanthioideae, and Smilacoideae *sensu* Engler [14]. He also commented on the regular occurrence of what he thought were glycosylflavones but which in retrospect were almost certainly ordinary flavones.

Some previous detailed flavonoid examinations of individual species are summarized in Table 2. Amongst the most recent findings are the new aglycone, 3-*O*-methyl-8-*C*-methylquercetin from *Dasylirion acrotrichum* (Nolinaceae) and *Xanthorrhoea hastilis* (Xanthorrhoeaceae) [18], six homoisoflavanones from *Muscari neglectum* (Hyacinthaceae) [30], four reports of homoisoflavonoids from *Ophiopogon japonicus* [24–27] and a flavonoid alkaloid from *Lilium candidum* [32].

Anthocyanins have been identified in flowers of nine genera: *Allium*, *Alstroemeria*, *Colchicum*, *Fritillaria*, *Hyacinthus*, *Lilium*, *Scilla*, *Tulipa* and *Urginea* [37]. They are mostly of a 'simple' non-methylated type, i.e. pelargonidin, cyanidin and delphinidin glycosides. Methylated pigments are less common, although peonidin has been found as the 3-arabinoside in the scales of *Allium cepa* [38], while acylated anthocyanins have been found in *Hyacinthus* [39] and *Urginea* [40] species. More recently the blue pigment of bluebell and of *Scilla peruviana* L. [incorrectly designated as *S. pensylvanica* in this paper] has been identified as delphinidin 3-(6-*p*-coumaroylglucoside)-5-(6-malonylglucoside) [41]. A unique anthocyanidin, 6-hydroxycyanidin, has been found as the 3-glucoside and 3-rutinoside in *Alstroemeria* flowers [42].

RESULTS AND DISCUSSION

The results of the present leaf flavonoid survey of some 91 species combined with those of a previous survey of 181 taxa [10], together representing 29 of the 52 families of Dahlgren's Liliiflorae [1] are presented in Table 3 according to that system. Both fresh and herbarium material was used and, whenever possible, more than one sample of a species from different sources was examined. The data in Table 3 refer to flavonoid aglycones, which were identified from leaf tissue after acid hydrolysis by means of R_f and colour reaction in UV light when compared with authentic markers. Both two dimensional paper chromatography of 80% methanolic leaf extracts and some detailed characterisation of individual glycosides were used to confirm the aglycone results. Flavonoid sulphates were detected by paper electrophoresis of direct leaf extracts at pH 2.2 and flavone *C*-glycosides confirmed by their resistance to four hour acid hydrolysis.

The distribution of leaf flavonoids in the Liliiflorae

First, we should point out that although Dahlgren *et al.* [1] include both the Iridaceae and Orchidaceae in their Liliiflorae the flavonoid results for these families have not been included in Table 3 as they have been published previously [43, 44] but they will be referred to

Table 2. Flavonoid glycosides and some rarer aglycones reported from the Liliiflorae*

Family, genus, species	Glycoside or aglycone	Organ examined	Reference
Alliaceae			
Allium ascalonicum L.	Qu 4′-glucoside Qu 3,4′-diglucoside Qu 7,4′-diglucoside	Bulb	[15]
A. cepa L.	Qu 4′-glucoside Qu 3,4′-diglucoside Qu 7,4′-diglucoside	Bulb	[16]
A. tuberosum Rottler	Km 3-sophoroside-7-(2-feruloyl)glucoside Km 3,4′-diglucoside-7-(2-feruloyl)glucoside Km 3-(2-feruloyl)glucoside-7,4′-diglucoside Km 3,4′-diglucoside Qu 3,4′-diglucoside Km 3-sophoroside	Leaf	[17]
Agavaceae			
Dasylirion acrotrichum Zucc.	3-*O*-Methyl-8-*C*-methyl quercetin	Leaf	[18]
Colchicaceae			
Colchicum byzantinum Ker Gawl.	Ap 7-diglucoside Lu 7-diglucoside Dios 7-diglucoside Dios 7-glucoside	Leaf	[10]
C. autumnale L.	Ap 7-diglucoside Ap, Lu, Lu 7-Glc Ap 7-Glc, Lu 7-DiGlc	Flower	[19, 20]
C. speciosum Steven	Lu 7-laminaribioside Ap 7-xylosylglucoside	Flower	[21]
Convallariaceae			
Convallaria majalis L.	Qu & Isorh 3-Gal Rha Isorh 3-GalRha-4′-Rha Qu 3-GalRha-4′-Ara	Flower	[22]
C. keiskei Miq.	Isorh 3-RhaGal	Flower	[23]
Ophiopogon japonicus Ker Gawl. var. *genuinus* Maxim.	Homoisoflavonoids	Root	[24, 25]
O. japonicus Ker Gawl.	Homoisoflavones and homoisoflavanones	Root	[26, 27]
Polygonatum multiflorum (L.) All.	Ap 8-*C*-xylosylglucoside Ap 6-*C*-rhamnosylglucoside-7-glucoside	Leaf	[28]
Hyacinthaceae			
Hyacinthus orientalis L. cv 'Queen of the Pinks'	Tricin 7-FruGlc & 7-RG-4′-Glc 6 Tricin tri-Glcs† & a tricin diGlc†	Leaf	[10]
Lachenalia uniflora L.	Lu 3′-SO_4 and 7,3′-diSO_4 Dios 3′-SO_4 and 7,3′-diSO_4 Tricetin 3′-SO_4† and 7,3′-diSO_4†	Leaf	[29]
Muscari neglectum Guss.	Four 3-benzyl-4-chromanones Six homoisoflavanones	Bulb	[30]
Liliaceae			
Lilium auratum Lindley	Isorh 3-RG	Pollen	[31]
L. candidum L.	3,5,7,4′-Tetrahydroxy-8-(3-methyl-2-oxo-5 pyrrolidinyl)flavone	Aerial parts	[32]

Table 2. *Continued*

Family, genus, species	Glycoside or aglycone	Organ examined	Reference
L. lancifolium Thunb.	Isorh 3-RG Qu 3-RG	Pollen	[33]
L. leucanthum Baker	Km 3-Glc Km 3-Diglc	Petal	[34]
L. regale E. H. Wilson	Km 7-Rha Km 3-Glc-7-Rha	Leaf and petal	[35]
Tulipa cvs	Qu 3-RG Qu 3-RG-7-Glur Km 3-RG	Leaf and petal	[36]
Xanthorrhoeaceae			
Xanthorrhoea hastilis R. Br.	3-*O*-Methyl-8-*C*-methylquercetin	Leaf	[18]

*Classification according to Dalgren *et al.* [1] but excluding the Iridaceae and Orchidaceae.
†Incompletely identified.
Key: Qu = quercetin, Km = kaempferol, Isorh = isorhamnetin, Ap = apigenin, Lu = luteolin, Dios = diosmetin.

in the Discussion. Details of eight taxa from some of the smaller families, which were available in too small an amount to give good flavonoid results are listed in the Experimental instead of in Table 3. Of the remaining 46 families of the Liliiflorae some 29 have been examined for their leaf flavonoids. In agreement with the previous survey of the group [10], quercetin and kaempferol were the most frequent leaf constituents both being found in 48% of the sample. Isorhamnetin was detected in 12 species but because an easy screening method was not available for this compound when the first survey was carried out a percentage figure here would not be very meaningful. Flavones were present in far fewer species than flavonols: luteolin and apigenin in 20 and 18% of the sample, respectively, while tricin and diosmetin occurred in only four taxa and tricetin, chrysoeriol, 6-hydroxyluteolin and flavonoid sulphates were each detected in only one species. The unusual aglycone, 8-*O*-methyl-8-*C*-methylquercetin, previously reported in *Dasylirion acrotrichum* was confirmed in that species and also identified in *D. glaucophyllum* and *D. longissimum* and in *Calibanus hookeri*, another member of the Nolinaceae. Anthraquinones, in 6% of the sample, were only detected in the two subfamilies of the Asphodelaceae, although they have been reported previously from the Xanthorrhoeaceae [8].

The flavonoid results will now be considered together with other chemical characters at the different levels of classification in the Dahlgren system [1]. In order to do this the flavonoid data have been summarised for the orders and families of the Liliiflorae, excluding the Burmanniales, in Table 4. From this and the summary of other chemical constituents in Table 2 it may be seen that the chemical data are not very meaningful at ordinal level. Thus, chelidonic acid, steroidal sapogenins and flavonols are frequent in all the four orders studied. However, the Dioscoreales does differ from the other three orders in the apparent absence of flavones, although flavone *C*-glycosides may be present in the Trichopodiaceae. Otherwise flavone *C*-glycosides were detected only in a small number of species in the Asparagales and Liliales. Methylated flavones were also confined to these two orders. Both the Asparagales and Liliales are probably too large an assemblage for the chemical results to be taxonomically useful. However, within these groups there are some families which only produce flavonols and some which have both flavones and flavonols. The Iridaceae and Orchidaceae appear to be out of place both in the Liliales and in the Liliiflorae in that they both produce flavone *C*-glycosides and flavonols as major leaf constituents.

The flavonoid patterns of the families within each order will now be considered.

Dioscoreales

Of the seven families recognised by Dahlgren only four are represented in the flavonoid survey and these by too small a number of taxa to give a meaningful appraisal. However, in the Dioscoreaceae, Trilliaceae and Smilacaceae flavonols appear to be the characteristic leaf components with the exception of two *Ripogonum* species in the latter family. This absence of flavonols in *Ripogonum* supports its separation from *Smilax* and *Heterosmilax* (subfamily Smilacoideae) into the subfamily Ripogonoideae. *Ripogonum* also deviates from these genera in a number of morphological characters and in its distribution in the southern rather than the northern hemisphere. Indeed, in the new Kew system of herbarium arrangement, modified from Dahlgren, *Ripogonum* is being recognised at family level as Ripogonaceae.

Asparagales

Nineteen of the 31 families of the Asparagales were screened for their leaf flavonoids. These can be broadly divided into flavonol or flavonol and flavone producing families. In only three of 188 species surveyed were both flavones and flavonols detected in the same plant.

Families producing only flavonols. Of the 13 'flavonol' families the Amaryllidaceae is probably the most uniform in its flavonoid glycoside pattern. It can also be distinguished from the remaining families by the absence of steroidal sapogenins and the presence of characteristic alkaloids. The Nolinaceae, on the other hand, is unique in the present survey in the production of the unusual

Table 3. The leaf flavonoids of some Liliiflorae

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
Dioscoreales									
Trichopodiaceae									
Trichopus zeylanicus Gaertn.	—	—	—	—	—	→?			A. H. M. Jayasuriya 1325, Ceylon
Dioscoreaceae									
Stenomeridoideae									
Avetra sempervirens H. perr.	—	—	—	—	—	+		K	G. F. S. Elliot 2745 Madagascar
Dioscoreoideae									
Dioscorea quartiniana A. Rich	+	—	—	—	—	—	ProCy	MO	A. A. Bullock 2214 N. Rhodesia
D. transversa R. Br.	+	+	—	—	—	—		NSW	Church Point, NSW May 1964
Rajania hastata L.	+	—	—	—	—	—	ProCy	F	N. L. Britton & L. J. K. Brace 184, New Providence, Bahamas
Tamus communis L.	+	+	—	—	—	—		NY	H. C. Stutz 3005, Lebanon
Trilliaceae									
Medeola virginiana L.	+	+	—	—				MASS	L. H. Elwell 50643 Belchertown, Mass
Paris quadrifolia L.†	+	+	+	—	—	—		M	
	—	+	nd	—	—	—		RNG	Pobedimova & Ratnitsina 901, Kaliningrad, USSR
Scoliopus bigelovii Torr.	+	+	—	—	—	—	ProCy	K**	410–78.04059
Trillium erectum L.	—	+	—	—	—	—			M. Howard s.n. Belchertown, Mass.
T. cuneatum Raf.	+	+	—	—	—	—	—	M	
Smilacaceae									
Ripogonoideae									
Ripogonum album R. Br.	No aglycones detected						ProCy DK/DK compound high in BAW & immobile in 15% HOAc	NY	G. T. White 7488 Glenreagh, NSW
R. scandens Forst.	DK/Y glycoside no aglycones det.							MASS	M. Howard s. n. Cult.
Smilacoideae									
Heterosmilax gaudichaudiana Maxim.	+	+	—	—	—	—	ProCy		E. H. Wilson 674 H. Hupeh, China
Smilax aspera L.†	+	+	nd	—	—	—	—	RNG	V. H. Heywood *et al.* 19 Gibraltar
S. canariensis Willd.†	+	+	nd	—	—	—	—	RNG	K. Lems 2202 Tenerife
S. mauritanica Desf.†	+	+	nd	—	—	—	—	RNG	F. A. Ibanez, Cartagena, Spain
S. sarumami Ohwi†	+	+	—	—	—	—	ProCy	MO	M. Togashi, Kyoto, Japan
Asparagales									
Philesiaceae									
Philesia magellanica J. F. Gmel.	No recognisable aglycones						ProCy 1-ve B	K**	684-65.68407

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
P. magellanica J. F. Gmel.	No recognisable aglycones							NY	E. Wedermann 256 Chloe, Chile
Luzuriagaceae									
Behnia reticulata (Thunb.) Didr.	—	+	—	—	—	—		NY	L. J. Brass 16763 Nyasaland
Drymophila moorei Baker	—	—	—	—	—	—	Tricin	NY	C. J. White 7480 Dorrigo St. Forest, NSW
Eustrephus latifolius R. Br.	—	—	—	—	—	+		MASS	M. Howard s. n. Cult.
Luzuriaga erecta Kunth.	—	+	—	—	—	—		MO	H. Behn 10294 85 Quilineja, Chile
L. marginata (Gaertner) B. K.†	—	+	—	—	—	—		NY	N. Goodall 629 Tierra del Fuego
	+	+	nd	—	—	—		RNG	D. M. Moore 544, Port Stanley, E. Falkland
L. parviflora Skotts.	—	+	—	—	—	—		—	E. E. Collins 137
L. radicans R. & P.	No recognisable aglycones								Ph-Germain 639916 Concepcion, Chile
Convallariaceae									
Polygonatae									
Maianthemum bifolium (L.) F. W. Schmidt	+	+	nd	—	—	—		RNG	J. W. Reed 1896 Bernese Oberland
Oligobotrya henyri Oliv. var. *limprichtii* (Ling) Krause	+	—	—	—	—	—		F	H. Stevens 246 Szechwan, China
Polygonatum biflorum† Elliott	+	+	nd	—	—	—		K**	000-69.1985.M. Africa
P. hookeri Baker†	—	—	nd	—	—	—		K**	204-54.20402 Himalaya
P. latifolium Desf.	+	+	+	—	—	+		M	
P. multiflorum (L.) All.	+	—	nd	—	—	—		S	
P. aff. *stewartianum* Diels	+	+	nd	—	—	—		K**	631-68.00066
P. verticillatum (L.) All.	+	—	—	—	—	+		NY	R. R. Stewart 10434 Kashmir
Smilacina racemosa† Desf.	—	—	—	—	—	—		K**	000-69.19700 N. America
	+	+	—	—	—	—		M	
S. stellata (L.) Desf.	—	+	+	—	—	—		AC	A. S. Pease & A. S. Goodale 53829, Greenfield, Mass.
Convallarieae									
Convallaria majalis† L.	+	—	+	—	—	—		R	
	+	—	+	—	—	—		M	
Reineckia carnea Kunth.	+	+	+	—	—	—		M	E. Asia
Speirantha gardenii Baill.	+	—	—	—	—	—		K	000–69.17479
Aspidistreae									
Aspidistra lurida†	—	—	—	—	—	—		K**	000–69.16574

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
Ker. Gawl.									China
Tupistra tupistroides (Kunth.) Dandy	—	—	—	—	—	—		K**	438–69.03674 India
Ophiopogoneae									
Liriope graminifolia Baker†	—	—	—	—	—	—		S	
L. spicata† Lour.	—	—	—	—	—	—		K**	000–69.10217
	(+)	—	—	—	—	—		M	
Ophiopogon jaburan† Loddiges	—	—	—	—	—	—		K**	No accession No. Japan
Asparagaceae									
Asparagus acutifolius† L.	+	+	nd	—	—	—		RNG	A. Rodriquez, E. Guinea 2735, Madrid, Spain
A. albus L.†	+	—	nd	—	—	—		RNG	V H. Heywood *et al.* 28, Gibraltar
A. arborescens Willd.†	+	—	nd	—	—	—		RNG	D. Bramwell 1693, Tenerife
A. pastorianus Webb† and Benth.	+	—	nd	—	—	—		RNG	D. Bramwell 993, Tenerife
A. plocamoides Webb† ex Svent	+	+	nd	—	—	—		RNG	D. Bramwell 1112, Gran Canaria
A. scoparius Lowe†	+	+	nd	—	—	—		RNG	D. Bramwell, Tenerife
A. umbellatus Link†	+	+	nd	—	—	—		RNG	D. Bramwell 1102, Gran Canaria
Ruscaceae									
Danae racemosa Moench	+	+	—	—	—	—		MO	P. Sintensis 1400 Persia
Ruscus aculeatus L.	(+)	(+)	—	—	—	—	Green/Y aglycone	K**	No Accession No.
R. hypoglossum L.†	+	—	nd	—	—	—		K**	000–69.19690
R. hypophyllum L.†	+	+	nd	—	—	—		RNG	V. H. Heywood *et al.* 57, Gibraltar
Herreriaceae									
Herreria interrupta Griseb.	—	(+)	—	—	—	—		NY	H. S. Irwin *et al.* Sierra do Espinhaco, Brasil
Dracaenaceae									
Sansevieria kirkii Baker var. *pulchra*	—	—	—	—	—	—		K**	370–03.37701
Nolinaceae									
Calibanus hookeri Trelease	+	+	—	—	—	—	3-OMe-8-*C*-Me-Qu?	K**	402–82.04536
Dasylirion acrotrichum Zucc.	—	—	—	—	—	—	3-OMe-8-*C*-Me-Qu	K**	000–73.131461
D. glaucophyllum Hook.	—	—	—	—	—	—	3-OMe-8-*C*-Me-Qu	K**	188–34.18803
D. longissimum Hemsl.	—	—	—	—	—	—	3-OMe-8-*C*-Me-Qu	K**	000–69.13485
Nolina recurvata (Lem.) Engl.	+	+	—	—	—	—		K**	495–36
Asteliaceae									
Astelia chathamica† (Skottsberg) L. B. Moore	—	—	—	—	—	—	ProCy	K**	038–70.00359 New Zealand

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
A. nervosa Banks & Soland.	+	—	—	—	—	—		K**	109–81.01435
A. pumila Banks & Soland.†	—	+	nd	—	—	—	ProCy	RNG	D. M. Moore 2050, Tierra del Fuego
Milligania stylosa F. Muell. ex Benth.	+	—	+	—	—	—		M	
Blandfordiaceae									
Blandfordia	+	+	—	—	—	—	DK/Y aglycone	K**	170–74.01753
punicea (Labill.) Sweet	+	+	nd	—	—	—		K**	014–68.01105 Tasmania
Xanthorrhoeaceae									
Xanthorrhoea† *australis* R. Br.	+	+	nd	—	—	—	ProCy	K**	253–67
Agavaceae									
Yuccoideae									
Yucca filamentosa L.†	—	+	nd	—	—	—		S	72/550
Phormiaceae									
Dianella caerulea† Sims	—	+	nd	—	—	—	Procy, Propel	K**	A. H. 15–5–52 No accession number
D. laevis R.Br.†	—	+	nd	—	—	—	ProCy, Propel	K**	343–64.34301
D. revoluta R.Br.†	—	+	nd	—	—	—	ProCy, Propel	K**	098–52.09801 Tasmania
D. tasmanica† Hooker fil.	—	+	nd	—	—	—	ProCy, Propel	K**	237.53 R. M. 2917
Phormium tenax Forst.	+	+	+	—	—	—	ProCy	K**	No accession number
Hemerocallidaceae									
Hemerocallis flava L.†	+	+	nd	—	—	—		RNG	Fiori and Beguinot 2825, Venetia, Italy
H. lilioasphodelus† L.	+	+	nd	—	—	—		R	4771
Hemerocallis sp.†	+	+	nd	—	—	—		K**	365–68.36509
Asphodelaceae									
Asphodeloideae									
Asphodeline lutea† (L.) Reichenb.	—	—	—	+	+	—	Anthraquinones	RNG	Davis 53109 Algeria
	—	—	—	+	+	—		K**	298–76.02659
Asphodelus acaulis† Desf.	—	—	—	+	+	—		RNG	H. Maurico, Sennen 8536, Morocco
	—	—	—	+	+	—		K**	263–82.02700
A. albus Miller†	—	—	—	+	—	—	Anthraquinones	RNG	R. M. Wadsworth *et al.* 282, Alicante, Spain
A. carasiferus Gay†	—	—	—	+	—	—	Anthraquinones	RNG	Fraser-Jenkins 46, Levida, Spain
A. fistulosus L.†	—	—	—	+	—	—	Anthraquinones	RNG	R.M. Wadsworth *et al.* 11, Valencia, Spain
A. mauritii Sennen	—	—	—	+	—	—	*Anthraquinones*	*RNG*	*Sennen* 9582
Bulbine frutescens Willd.	—	—	—	—	—	—	Anthraquinones	K**	381–57.38107
B. latifolia Baker	+	—	—	+	—	—	Anthraquinones	K**	286–73–02997
Eremurus elwesii† Micheli	—	—	—	+	+	—		K**	409–72.03744 Van Tubergen, Holland

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids								
	Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
E. himalaicus† Baker	—	—	—	+	+	—		K**	641–68.00160 Van Tubergen, Holland
E. stenophyllus† Baker var. *bungei*	—	—	—	+	+	—		K**	000–69.19582
Paradisia liliastrum (L.) Bertol†	—	—	—	+	+	+		RNG	G. Nicholson & I. W. Reed, July 1898, Switzerland
Simethis planifolia (L.) Gren.†	+	+	—	—	—	—	ProCy	RNG	E. Guinea 679, Asturias, Spain
Alooideae									
Aloe africana Miller†	(+)	—	—	—	—	—	Anthraquinones	K**	53.663
A. ballii Reynolds†	—	—	—	—	—	—		K**	69.985
A. bellatula Reynolds†	(+)	—	—	—	—	—		K**	68.812
A. cooperi Baker†	(+)	—	—	—	—	—		K**	68.875
A. cremnophila Reynolds†	(+)	(+)	nd	—	—	—	Anthraquinones	K**	72.162
A. dawei A. Berger†	—	—	—	—	—	—		K**	68.552
A. dolomitica Groenewald†	—	—	—	—	—	—	Anthraquinones	K**	49.368
A. elgonica Bullock†	(+)	(+)	nd	—	—	—		K**	68.1510
A. jacksonii Reynolds†	—	(+)	nd	—	—	—	Anthraquinones	K**	68.813
A. kedongensis Reynolds†	—	—	—	—	—	—	Anthraquinones	K**	72.747, Kenya
A. lateritia Engler†	—	—	—	—	—	—	Anthraquinones		B. & C. 15–136
	—	—	—	—	—	—	,,		B. & C. 8–61
	—	—	—	—	—	—	,,	K**	19–160
	—	—	—	—	—	—	,,		22–171
	—	—	—	—	—	—	,,		5–35
	—	—	—	—	—	—	,,		17–146
A. marlothii A.† Berger	—	—	—	—	—	—	Anthraquinones	K**	52–547
A. nyeriensis Christian†	—	—	—	—	—	—	Anthraquinones	K**	26–200
A. pretoriensis Pole Evans†	—	+	nd	—	—	—		K**	38157
A. rabaiensis Rendle†	—	—	—	—	—	—	Anthraquinones	K**	B. & C. 29–313
A. rauhii Reynolds†	—	—	—	—	—	—		K**	68436
A. saponaria (Aiton fil.) Haw.†	—	—	—	—	—	—		K**	52–547
A. speciosa Baker†	+	+	nd	—	—	—	Anthraquinones	K**	70.1695
Chamaealoe africana A. Berger†	+	+	nd	—	—	—		K**	68–1154
Gasteria caespitosa V. Poelln.†	—	—	—	—		—		K**	Type plant
G. candicans Haw.†	—	—	—	—	—	—		K**	68–541
G. carinata Duva†	—	—	—	—	—	—		K**	68–412
G. multipunctata† Hort.	—	—	—	—	—	—		K**	69–214
Haworthia cooperi Baker†	—	—	—	—	—	—		K**	68.291
H. reinwardtii Haw.† var. *adelaidensis*	—	—	—	—	—	—		K**	68.857

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
Anthericaceae									
Anthericum algeriense†(Boiss. & Reuter)	—	+	nd	—	—	—		RNG	Sennen and Mauricio, Sennen, 1932, Morocco
A. liliago L.†	—	—	—	+	—	—		K**	Whitehead 190 433–71.04261
A. suffruticosum (Baker) Milne-Redhead	—	—	—	—	—	+?		K**	307–70.02986
Arthropodium cirrhatum R. Br.	+	—	+	—	—	—		M	
A. minus R. Br.	+	+	+	—	—	—		K**	422–71.04089
Chlorophytum† *comosum* Baker form	—	—	—	—	—	—		K**	Temperate house No accession no.
C. elatum R. Br. var. *variegatum*	—	—	—	—	—	—	DK/DK unknown aglycone	K**	No accession no.
C. gallabatense Schweinf. ex Baker	+	—	—	—	—	—		K**	360–70.03547
C. macrophyllum Aschers.	—	—	—	—	—	—	No aglycone but 2 DK/Y glycs	K**	254–74.02188
C. undulatum Wall.†	—	—	—	—	—	—	Propel	K**	167–71.01652
Echeandia sp.	—	—	—	+	+	—		K**	480–77.04599
Aphyllanthaceae									
Aphyllanthes monspeliensis L.†	+	+	nd	—	—	—		RNG	L. Ercriche E. Guinea 2730 Tudela, Spain
	+	—	+	—	—		Diosmetin and acacetin	M	
Funkiaceae									
Hosta lancifolia† Tratt.	—	+	nd					K**	000–69.19612
H. plantaginea† Ascherson	—	+	nd					K**	000–69.19618
H. rupifragum Nakai	—	+	—	—	—	—		M	
H. ventricosa (Salisb.) Stearn†	—	+	nd					K**	000–69.19628
Hyacinthaceae									
Albuca melleri Baker	—	—	—	—	—	—		K**	429–74.03608
Bellevalia flexuosa† Boiss.	—	—	—	+	+	—		K**	000–72.10662
B. glauca Kunth†	—	—	—	+	+	—		K**	528–67
Bowiea volubilis Harv.	—	—	—	—	+	—		K**	12–12–85
Camassia esculenta Lindl.	+	+	nd	—	—	—		K**	641–68.00119
C. leichtlinii S. Watson†var. *alba*	+	+	nd	—	—	—		K**	238–52.23801 N. W. America
Chinodoxa cretica Boiss. & Heldr.†	—	—	nd	+	+	—		K**	000–72.10226
C. sardensis Barr & Sugden†	—	—	nd	+	+	—		K**	000–72.10239
Chlorogalum pomeridianum Kunth.	+	+	+	—	—	—		M	
	—	+	—	—	—	—		K**	311–82.03154
Dipcadi serotina (L.) Medicus†	+	+	nd	—	—	—		RNG	M. M. Martinel 2742 Madrid, Spain

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
D. viride Moench.	—	—	—	—		+		K**	254–70.02371
Hyacinthoides hispanica (Mill.) Rothm.†	—	—	—	+	+	—		RNG	E. Guinea 1921, Mayo, Spain
H. non-scripta (L.) Chouard ex Rothm.†	—	—	—	+	+	—		S	
Eucomis undulata Ait.	—	—	—	+	+	—		K**	440–64.44003
Galtonia candicans† Decne.	+	—	—	—	—	—		K**	000–69.19589
Hyacinthella millingenii† (Post) Feinbrun	—	—	—	+	+	—		K**	000–72.10288
Hyacinthus orientalis†L.	—	—	—	—	—	—	Tricin	R	
Lachenalia aloides Pers.	+	—	—	+	+	—	DK/DK-ve	K**	000–73.14513
L. orthopetala Jacq.†	—	—	—	+	+	—		S	72/369
L. unifolia Jacq.	—	—	—	—		—	Diosmetin, Tricetin as 3′-SO_4S & 7,3′-di-SO_4S	L	WFB/26/59
Massonia pustulata Jacq.	—	—	—	+	+	—		K**	296–76.02634
Muscari armeniacum† Leichtlin ex Baker	—	—	—	+	+	—		S	
M. comosum (L.)	—	—	—	+	+	—		S	
Miller†	—	—	—	+	+	—		RNG	V. H. Heywood *et al.* 441, Sevilla Spain
Ornithogalum juncifolium Jacq.	—	—	—	—	—	+		K**	567–69.05036
O. montanum Tenore	—	—	—	—	—	—	2DK/DK unknown aglycones	K**	245–69.02171
O. narbonense L.†	—	—	—	—	—	—		K**	000–69.19681 Mediterranean 000–69.19681
O. saundersiae Baker	—	+	—	—	—	—		K**	542–56.54205
O. tenuifolium Guss.†	—	—	—	+	+	—		RNG	Gunpriob 1939 USSR
O. umbellatum L.†	—	—	—	+	+	—		K**	N. Africa
	—	—	—	+	+	—		RNG	V. H. Heywood *et al.* 62, Prov. de Cadiz, Spain
Polyxena corymbosa (L.) J. P. Jessop	—	—	—	+	+	—		K**	285–75.02765 S. Africa
Puschkinia scilloides Adams†	—	—	—	+	+	—		S	
P. scilloides Adams†	—	—	—	+	+	—		K**	527–59
var. *libanotica*	—	—	—	+	+	—		K**	641–68.00186 Japan
Schizobasis intricata Baker	+	+	+	—	—	—		K**	000–69.13967
Scilla adlami Baker†	—	—	—	—	+	—		K**	No accession no. S. Africa
S. liliohyacinthus L.†	—	—	—	—	+	—		RNG	1–4–42, Bilbao, Spain, E. Guinea
S. natalensis Planch.	—	—	—	—	+	—		K**	657–63.65708
S. peruviana L.†	—	—	—	+	+	—		RNG	Fraser-Jenkins 116, Prov. Jaen, Spain

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
S. sibirica Andrews†	—	—	—	+	+	—		S	
S. socialis Baker	—	—	—	—	—	—	Tricin	K**	000–73.13510
S. verna Hudson†	—	—	—	—	+	—		S	
Urginea altissima Baker	—	—	—	—	—	+		K**	555–64.55506
U. maritima†Baker	—	—	—	—	—	+		K**	000–72.10347
Veltheimia deasii† Barnes	—	—	—	—	+	—		S*	
Alliaceae									
Agapanthoideae									
Agapanthus orientalis Leighton	+	+	—	—	—	—		K**	080–82.00540
Tulbaghia alliacea† L. fil.	—	+	nd	—	—	—		S	
T. violacea Harvey†	—	+	nd	—	—	—		S	67192
	+	+	—	—	—	—		K**	567–69.05037
T. violacea Harvey var. *tricolor*	+	+	—	—	—	—		K	307–81.03417
Allioideae									
Brodiaeae									
Brodiaea† *californica* Lindley	+	+	nd	—	—	—		K**	No accession no.
Allieae									
Allium cyathophorum Bureau et Franch.	+	+	—	—	—	—		M	
A. karataviense† Regel	+	+	nd	—	—	—		K**	No accession no.
A. mairei Leveille†	+	+	nd	—	—	—		K**	57–53.57003 China
A. narcissiflorum† Vill.	+	+	nd	—	—	—		S	
A. oreophilum† C. A. Meyer	—	+	nd	—	—	—		K**	No accession no.
A. paniculatum Viv. var. *pallens*	+	—	nd	—	—	—		K**	27–42.2701 Libya
Ipheion uniflorum Rafin. var. *violaceum*	+	—	—	—	—	—	DK/DK-ve	K**	000–73.15748
Nothoscordum euosum† (Link & Otto) Kunth	+	—	nd	—	—	—		RNG	H. Roivainen 492 Argentina
N. fragrans Kunth†	—	—	nd	—	—	—		S	73/161
Tristagma nivale† Poeppig	+	+	nd	—	—	—		RNG	D. M. Moore 2087 Tierra del Fuego
Gilliesioideae									
Gilliesia graminea Lindl.	+	+	—	—	—	—		M	
Amaryllidaceae									
Amaryllideae									
Amaryllis belladonna L.†	+	+	nd	—	—	—		S	
Amaryllis sp.	+	+	nd	—	—	—		So	
Crinum bulbispermum† (Burm.) Milne-Redhead & Schweickerdt	+	+	nd	—	—	—		S	72/524
Lycorideae									
Lycoris squamigera† Maxim.	—	+	nd	—	—	—		S	

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
Eucharideae									
Eucharis subedentata† Bentham & Hooker	—	+	nd	—	—	—		R	
Pancratieae									
Pancratium canariensis Ker-Gawler ex Webb & Benth.	—	+	nd	—	—	—		RNG	D. Bramwell 427 Tenerife
P. illyricum L.†	—	+	nd	—	—	—		RNG	C. J. Humphries & I. B. K. Richardson 133, Sardinia
Narcisseae									
Narcissus asturiensis†(Jordan) Pugsley	+	+	nd	—	—	—		RNG	B. Casaseca 1967 Salamanca, Spain
N. bulbocodium L.†	+	+	nd	—	—	—		RNG	E. Kjellquist & A. Love N517, Prov. Teruel, Spain
N. jonquilla L.†	+	+	nd	—	—	—		RNG	E. Guinea 1943 Granada, Spain
N. pallidulus L.† Graells	+	—	nd	—	—	—		RNG	B. Casaseca 1967 Salamanca, Spain
N. pseudo-narcissus L.†	+	—	nd	—	—	—		RNG	E. Guinea 1943 Fornes Granada, Spain
N. requienii Roemer†	+	+	nd	—	—	—		RNG	C. R. Fraser-Jenkins 117, Jaen, Spain
N. tazetta L.†	+	+	nd	—	—	—		RNG	E. F. Galiano & J. Novo 8–12–65 Prov. Cadiz, Spain
Galantheae									
Galanthus nivalis L.†	+	—	nd	—	—	—		R	
Leucojum aestivum L.†	+	+	nd	—	—	—		RNG	C. J. Humphries & I.B.K. Richardson 145, Sardinia
Melanthiales									
Melanthaceae									
Melanthieae									
Veratrum album L.†	—	—	—	+	+	—		RNG	E. Guinea 1242 Asturias, Spain
V. nigrum L.†	—	—	—	+	—	—		RNG	Nr Gadnen, 1–7 1896
Zigadenus fremontii Torr. ex S. Wats.	+	+	+	—	—	—		K**	730–67.73019
Narthecieae									
Heloniopsis orientalis (Thunb.) C. Tanaka	+	+	—	—	—	—		K**	478–79.04887
Narthecium ossifragum (L.) Hudson	—	—	—	—	—	—		RNG	E. Guinea 238, Spain

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids								Collector's name & number or accession number
	Q	K	I	L	A	C	Other	Source	
Tofieldieae									
Tofieldia calyculata† (L.) Wahlenb.	+	+	—	—	—	—		RNG	G. Halliday 324/72 Yugoslavia
T. yezoensis Miyabe & Kudo	+	+	—	—	—	—		M	
Xerophylleae									
Xerophyllum tenax Nutt.	+	+	+	—	—	—	DK/DK aglycone	M	
Liliales									
Astroemeriaceae									
Alstroemeria† *brasiliensis* Sprengel	—	+	nd	—	—	—		K**	109–57.10902
A. pelegrina L. var. *alba*†	+	(+)	nd	—	—	—		K**	836–59.83601 Chile
A. pulchella L. fil.†	—	+	nd	—	—	—		K**	109–57.10903 Brazil
Bomarea caldasii† (H.B.K) Asch. & Graeb.	—	+	nd	—	—	—		K**	No accession no. temp. house Guatemala
B. carderi Masters†	—	—	—	—	—	—		K**	No accession no. temp. house Guatemala
Colchicaceae									
Iphigenieae									
Gloriosa simplex† L.	—	—	—	+	+	—		S	71/319
Littonia lindenii Baker	+	—	—	+	+	—	ProCy chrysoeriol	K	N. Rhodesia Nov. 1951
Sandersonia	—	—	—	+	+	—		MO	J. Medley Wood
aurantiaca Hooker	—	—	—	+	+	—		M	10,413 Durban
Colchiceae									
Androcymbium† *punctatum* (Cav.) Baker	—	—	—	+	—	—		RNG	Hno. Jeronimo & F. Sennen 8175, Almeria, Spain
Colchicum byzantinum Ker Gawler†	—	—	—	+	+	—	Diosmetin	S	
C. speciosum Steven cv 'Album'	—	—	—	+	—	—		M	
C. speciosum Steven cv 'The Giant'†	—	—	—	+	+	—	Diosmetin	S	
C. speciosum Steven cv 'Lilac Wonder'†	—	—	—	+	+	—	Diosmetin	S	
C. triphyllum.† Kunze	—	—	—	+	+	—		RNG	E. Guinea 272 Madrid
Merendera sobolifera Fischer & Meyer	—	—	—	+	+	—		S	
Uvulariaceae									
Tricyrtideae									
Tricyrtis flava Maxim.	+	—	nd	—	—	—		K**	Woodruff 465/60
T. formosana Baker†	+	+	nd	—	—	—		K**	413–46–41301
	+	+	+	—	—	—		M	Formosa
T. formosana Baker† var. *stolonifera*	+	+	nd	—	—	—		K**	000–69.11425
T. hirta Hooker†	+	—	nd	—	—	—		K**	147–61.14701 Japan

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
T. macropoda Miq.†	+	+	nd	—	—	—		K**	106–68.10602
T. puberula Nikai & Kitagawa†	+	+	nd	—	—	—		K**	010–67.01001
Uvularieae									
Disporopsis luzoniense V. Kumer	+	—	—	—	—	—		K**	460–77.03498
Disporum menziesi Nichols.	—	+	—	—	—	—		F	Howell 22821 Western Oregon
D. sessile Don	—	—	—	+	—	—		F	L. M. Smith 773798 Seoul, Korea
D. smithii (Hooker) Piper	—	+	—	—	—	—		F	V. Duran 3413 Humbolt Co., California
Kreysigia multiflora Reichb.	—	—	—	+	+	—		NY	R. A. Campbell 10 1900 Murwillumbah, Australia
Schelhammera multiflora R. Br.	—	—	—	+	+	—	ProCy 6 OHLu	NY	L. J. Brass 5937 Oriomo River, Papua
Uvularia grandiflora† J. E. Smith	+	—	—	—	—	—		K**	000–69.19703
U. perfoliata L.†	—	—	—	—	—	—	Tricin	K**	557–56.55702
U. sessiliflora L.†	+	—	—	—	—	—		W	
Liliaceae									
Erythronium† *denscanis* L.	+	+	nd	—	—	—		RNG	G. Negrean, Ilfor, Romania
E. montanum S. Watson†	+	+	nd	—	—	—		K**	587–69.05263
Fritillaria hispanica† Boiss.	+	+	nd	—	—	—		RNG	A. Rodriguez & E. Guinea 2731, Madrid
F. meleagris L.†	—	+	nd	—	—	—		S	
F. pallidiflora Schrenk†	+	+	nd	—	—	—		K**	No accession no. S. Siberia
F. persica L.	—	+	—	—	—	—		M	
Gagea foliosa (Presl.) Schultes†	—	+	nd	—	—	—		RNG	Kjellquvist and Love, N120, Prov. Jaen, Spain
Lilium bulbiferum L.†	+	+	nd	—	—	—		RNG	C. J. Humphries L/2 W. of Brusane, Yugoslavia
L. davidii Duchartre†	+	+	nd	—	—	—		S	
L. hansoni Leichtlin†	+	+	nd	—	—	—		S	
L. henryi Baker†	+	+	nd	—	—	—		S	
L. martagon L. var. *album*†	+	—	—	—	—	—		K**	No accession no.
L. pardalinum† Kellogg	+	+	nd	—	—	—		S	
L. *pyrenaicum* Gouan†	+	—	—	—	—	—		K**	640–68.00113
L. regale E. Wilson	+	+	—	—	—	—		M	
L. szovitsianum† Fischer & Ave-Lall.	+	—	—	—	—	—		S	
L. tsingtauense Gilg†	+	+	nd	—	—	—		K**	718–67

Table 3. *Continued*

Order, Family Subfamily, Tribe, Genus, Species*	Flavonoids Q	K	I	L	A	C	Other	Source	Collector's name & number or accession number
Notholirion thomsonianum(Royle) Stapf	—	+	—	—	—	—		K**	377–53.37703
Tulipa australis† Link	+	+	nd	—	—	—		K**	No accession no.
T. batalinii Regel†	—	+	nd	—	—	—		K**	498–70.04683 Bokhara
T. greigii Regel†	+	+	nd	—	—	—		K**	447–67.44706
T. saxatilis Siebert† ex Sprengel	—	+	—	+	nd	—		—	No accession no. Crete
T. schrenkii Regel†	+	+	+	nd	—	—		K**	339–71.03081 Russia
T. tarda Stapf†	+	+	nd	—	—	—		K**	745–59.74515 Turkistan

*Classification according to Dahlgren *et al.* [1].

Chemical key: Q = Quercetin; K = Kaempferol; I = Isorhamnetin; A = Apigenin; C = *C*-glycosides. ProCy = Procyanidin; Propel = propelargonidin; 6-OHLu = 6-hydroxyluteolin; DK/DK = dark to dark in UV light with with and without ammonia vapour.

Plant origin key: K** = Plant growing at the Royal Botanic Gardens, Kew, for which accession numbers are given; L = Plant growing under glass at Department of Plant Sciences, University of Leeds; RNG = Reading University Herbarium; S = Plants growing at Shinfield Horticultural Station, University of Reading; W = Plants supplied and verified by C. D. Brickell, The Director, The Royal Horticultural Society's Garden, Wisley; R = Plants growing at the Botanic Garden, Plant Science Laboratories, University of Reading; SO = plants supplied by Mrs. R. M. Souster, Goring-on-Thames; US = United States National Herbarium, Department of Botany, Smithsonian Institution, Washington; K = The harbarium, Royal Botanic Gardens, Kew; MO = Herbarium, Missouri Botanic Garden, St. Louis, Missouri; NSW = National Herbarium of New South Wales, Sydney, Australia; F = John G. Searle Herbarium, Field Museum of National History, Chicago; NY = Herbarium, New York Botanical Garden; MASS = Herbarium, Department of Botany, University of Massachusetts, Amherst; AC = Herbarium, Amherst College, Amherst, Massachusetts, U.S.A.; H = Material from Margaret Howard, Botany Department, University of Massachusetts, Amherst 01003; M = material from the collection of Mr Brian Mathew.

†Results from Williams (1975).

C-methylated aglycone, 3-*O*-methyl-8-*C*-methylquercetin. However, this compound has been reported previously from one member of the Xanthorrhoeaceae [8] suggesting a possible close relationship between these two families. The Alliaceae, although restricted in its leaf flavonoids, are easily recognised by the presence of strong smelling allyldisulphides and propyl- and vinyl sulphides. Also, the bulbs of *Allium* species are characterised by complex mixtures of flavonol glycosides (see Table 2). The remaining small families have not shown any obvious chemical distinctions as yet.

Flavonol and flavone producing families. These include the Luzuriagaceae, Asphodelaceae, Anthericaceae, Aphyllanthaceae, Hyacinthaceae and Convallariaceae. In the latter, although flavonols are the main constituents and simple flavones were not detected, flavone *C*-glycosides occur in two *Polygonatum* species. Also in this family there was notable variation at tribal level in that the Polygonatae and Convallarieae are very rich in flavonoid constituents, whereas flavonoids were not detected in the members of the Aspidistreae and Ophiopogoneae tested, with the exception of a trace of quercetin in one *Liriope spicata* sample.

In the Luzuriagaceae kaempferol was the main constituent in four *Luzuriaga* species and in *Behnia reticulata.* However, the finding of tricin as the only flavonoid aglycone in *Drymophila moorei* supports Dahlgren's indication that the genus *Drymophila* is a dubious member of the Luzuriagaceae. It would also support the suggestion that it be included in the Uvulariaceae, rather than its more usual placement in the Convallariaceae.

Although members of both subfamilies of the Asphodelaceae commonly produce anthraquinone pigments, they differ markedly in their flavonoid constituents. Thus, the Asphodeloideae produce largely flavone glycosides while in the Alooideae flavonoids were either not detected or flavonols were present in small amount. The only exceptions were in the Asphodeloideae, where both quercetin and luteolin were found in *Bulbine latifolia* and flavonols only in *Simethis planifolia.* This probably reflects the more varied geographical distribution of the genera in this subfamily. But it also lends some support to the more usual assignment of *Simethis* to the Anthericaceae. Flower structure and certain embryological details also agree with this placement [1]. Although anthraquinones have been reported previously [7] from leaf tissue of *S. planifolia* they were not found in the present sample.

Amongst the 11 taxa of the Anthericaceae surveyed a range of different flavonoid patterns were observed. Although the sampling is small, the chemical data support the view of Dahlgren *et al.* [1] that this is a heterogeneous group badly in need of further study. The systematic position of the monospecific *Aphyllanthes*, which has been included in the Anthericaceae by some taxonomists, is still uncertain [1]. However, the discovery of free

Table 4. A summary of the leaf flavonoid constituents of the Liliiflorae*

Order, family, tribe	No. of species	Qu	Km	Isorh	Lu	Ap	Flavone *C*-glycosides	Others
Dioscoreales								
Trichopodiaceae	1	—	—	—	—	—	1?	
Dioscoreaceae	5	3	1	—	—	—	—	ProCy (2)
Trilliaceae	5	4	5	1†	—	—	—	
Smilacaceae	7	5	5	—†	—	—	—	DK/DK aglycone in *Ripogonum*
Asparagales								
Luzuriagaceae	7	1	5	—#	—	—	1	Tricin (1)
Convallariaceae	19	12	7	3#	—	—	2	
Asparagaceae	7	7	4	nd	—	—	—	
Ruscaceae	4	4	3	—#	—	—	—	Green/Y aglycone in *Ruscus*
Herreriaceae	1	—	(1)	—	—	—	—	
Dracaenaceae	1	—	—	—	—	—	—	
Nolinaceae	5	2	2	—	—	—	—	3-*O*-methyl-8-*C*-methyl-quercetin(4)
Asteliaceae	4	2	1	1#	—	—	—	ProCy (2)
Blandfordiaceae	1	1	1	—	—	—	—	DK/Y aglycone
Xanthorrhoeaceae	1	1	1	nd	—	—	—	ProCy (1)
Agavaceae	1	—	1	nd	—	—	—	
Ixioliriaceae	5	—	5	1#	—	—	—	ProCy (5)
Hemerocallidaceae	3	3	3	nd	—	—	—	
Asphodelaceae	38	9	7	—	11	6	1	Anthraquinones (17)
Asphodeloideae	13	2	1	—	11	6	1	Anthraquinones (7)
Alooideae	25	7	6	—	—	—	—	Anthraquinones (10)
Anthericaceae	11	3	2	2	2	1	1?	
Aphyllanthaceae	1	1	1	1	—	—	—	'Free' Diosmetin & acacetin (1)
Funkiaceae	4	—	4	—#	—	—		
Hyacinthaceae	44	7	6	1#	22	28	5	Tricin (2) Diosmetin (1) Tricetin (1)
Alliaceae	16	13	12	—#	—	—	—	
Amaryllidaceae	16	12	13	—#	—	—	—	
Melanthiales								
Melanthaceae	8	5	5	2	2	1	—	DK/DK aglycone (1)
Liliales								
Alstroemeriaceae	5	1	4	—#	—	—	—	
Colchicaceae	11	1	—	—	11	9	—	Diosmetin (3) chrysoeriol (1), ProCy (1)
Uvulariaceae	15	3	2	—	3	2	—	Tricin (1), 6-OHLu (1), ProCy (1)
Liliaceae	24	18	21	—	—	—	—	
Iridaceae‡	255	62	45	25	6	8	168	6-Hydroxy flavones (3), tricin (21), Biflavonoids (2), Mangiferin (44), Plumbagin(10) ProCy (41)

Table 4. *Continued*

Order, family, tribe	No. of species	Qu	Km	Isorh	Lu	Ap	Flavone C-glycosides	Others
Orchidaceae§	142	52	31	—	2	—	75	6-Hydroxy flavones (8), Mangiferin (50), Tricin (1).

*Classification according to Dahlgren *et al.* [1]. Figures refer to the number of species in which each aglycone was found.
†Not determined in all species.
‡See ref. [43].
§See ref. [44].
Key: Qu = quercetin, Km = kaempferol, Isorh = isorhamnetin, Lu = luteolin, Ap = apigenin, 6-OHLu = 6-hydroxyluteolin, Pro-Cy procyanidin, nd = not determined, ( ) = trace constituent.

diosmetin and acacetin in leaf tissue are sufficiently distinctive characters to lend some support to its separation from the Anthericaceae as the family Aphyllanthaceae. Interestingly, *Aphyllanthes* is being recognised as a separate family in the new Kew system.

As only one of the three recognised genera of the Funkiaceae, *Hosta*, was available for study it is difficult to comment on the relationships of the family. However, the flavonoid patterns of the four *Hosta* species examined were distinctive in that they were all based on the aglycone, kaempferol, a pattern also found in *Yucca filamentosa*, the only member of the Agavaceae surveyed. Cytological evidence: a chromosome number of $n = 30$ in both *Hosta* and in Agavaceae, also suggests a possible close relationship between these groups. A wider flavonoid survey of the Agavaceae is obviously needed to confirm the chemical similarity.

The Hyacinthaceae is another chemically heterogeneous group (Table 3). However, in the majority of the 44 taxa surveyed the flavones, luteolin and apigenin are the major leaf flavonoids. Of the remaining taxa it has been suggested [1] that *Camassia* (flavonols only) and *Bowiea* (flavone *C*-glycosides) are not very closely allied to the majority of the genera in the family and that the position of *Chlorogalum* (flavonols only) needs reconsideration. The most distinctive flavonoid pattern encountered in the Liliiflorae is in *Lachenalia unifolia* (Hyacinthaceae), which contains diosmetin and tricetin 3′-monosulphates and 7,3′-disulphates in the leaves. This is also the only occurrence recorded during the present survey of flavonoid sulphates in the whole superorder. Other unusual patterns based on tricin glycosides were found in *Hyacinthus orientalis* [10] and *Scilla socialis*. The flavonoid evidence (regular presence of apigenin and luteolin glycosides) would also support the inclusion of *Chinodoxa* species within *Scilla* as suggested by Dahlgren *et al.* [1] but does not distinguish *Puschkinia* from these genera. Although the floral habit of *Massonia* resembles that of some species of *Haemanthus* (Amaryllidaceae) the present chemical evidence clearly refutes any idea of its placement in that family. In the present survey the distinctive flavonoid glycosides luteolin and apigenin 7-glucuronide and luteolin 7,4′-diglucuronide were identified in *Massonia pustulata*. This contrasts with the universal presence of flavonols in all members of the Amaryllidaceae that have been surveyed. This was confirmed in a spot check on leaf of *Haemanthus sanguineus* Jacq. and of the closely related *Scadoxus nutans* (Friis and Bjornstad) Friis and Nordal, which yielded quercetin from the former and kaempferol from the latter.

The Melanthiales

This is a small order of only two families of which only the Melanthaceae was surveyed for flavonoid constituents. In the small sample available the only exception to a basic flavonol pattern was *Veratrum*, where flavones were found instead. However, an interesting dark to dark (in UV light plus NH_3) aglycone was detected in *Xerophyllum tenax* which could possibly be a biflavonoid but its R_f values did not agree with any of the available markers so its identification remains uncertain.

The Liliales

In this order 10 families are recognised of which six have been surveyed, some previously [10]. As it stands it is the largest and most heterogeneous group because it includes both the Iridaceae and Orchidaceae, which as indicated above do not fit chemically into this order (or in the Liliiflorae) since they predominantly contain flavone *C*-glycosides and flavonols in their leaf tissue [43, 44]. These families would therefore be better treated as superorders in their own right. Of the remaining four families examined, the Alstroemeriaceae and Liliaceae are the most uniform with consistent flavonol patterns, although the former may be distinguished by the predominance of kaempferol over quercetin glycosides. The Colchicaceae, on the other hand, is characterised by the flavones luteolin and apigenin with diosmetin present in three taxa. In this family *Littonia lindenii* is exceptional in that quercetin, luteolin, apigenin and chrysoeriol are all present in the leaves and this may be better placed in the Uvulariaceae as suggested by Hutchinson [45]. Dahlgren *et al.* divide the Uvulariaceae into two subfamilies: the Tricyrtideae with a single genus, *Tricyrtis* and the Uvularieae, which is a much more heterogeneous assemblage of genera. This is a very different treatment from that of Melchior [13], who places most of these taxa in his subfamily Melanthioideae. The chemical evidence supports the separation of *Tricyrtis* with its uniform flavonol pattern and highlights the variation within the Uvularieae. Even within *Uvularia* chemical diversity is evident in that *U. grandiflora* and *U. perfoliata* contain only tricin. Another member of the group, *Schelhammera multiflora* is so far unique in the Liliiflorae in synthesizing 6-hydroxyluteolin together with luteolin and apigenin. In two American *Disporum* species, *D. menziesii* and *D. smithii* only kaempferol was detected, while in *D. sessile* from Korea luteolin was the only aglycone. This chemical evidence gives preliminary support to the suggestion of Conover [46] that the American *Disporum* species are probably distinct from the Asiatic species and should be placed in a separate genus, *Prosartes*, which shows greater affinity with *Tricyrtis*, another flavonol producing genus, than *Disporum*. An extended flavonoid study of *Disporum* species from both continents is obviously desirable and could prove most rewarding.

CONCLUSIONS

On the whole, the recognition by Dahlgren *et al.* [1] of so many new families within their Liliiflorae cannot be justified from the presently available flavonoid data. However, certain groups appear to be more homogeneous than in previous treatments for example, the very much reduced Liliaceae and the newly formed Funkiaceae (*Hosta* species). But it is unfortunate that the Hyacinthaceae, which is equivalent to Melchior's [13] Scilloideae, should still include some chemically anomalous taxa such as *Camassia*, *Bowiea* and *Chlorogalum*, even though Dahlgren *et al.* [1] agree that the position of these three genera may need reconsideration; *Dipcadi* also seems to be out of place here. Similarly, the Aspidistreae and Ophiopogoneae do not fit well into the new family Convallariaceae, although Melchior [10] did include *Aspidistra* and *Convallaria* in his tribe Convallarieae of the subfamily Asparagoideae. But he grouped *Ophiopogon* with *Liriope* in the tribe Ophiopogonoideae. In fact, all six members of the Aspidistreae and Ophiopogoneae surveyed essentially lack flavonoids while 15 species of the remaining two tribes: the Polygonatae and Convallarieae are rich in flavonol glycosides. The Anthericaceae is another chemically heterogeneous group while the Amaryllidaceae and Alliaceae are probably the two most chemically and morphologically uniform families.

The Iridaceae and Orchidaceae are clearly distinguished from all other families in the Liliiflorae and it has already been suggested from the flavonoid data that they should be put into separate orders as in most previous systems. In both these families flavone *C*-glycosides, which are otherwise rare in the Liliiflorae, are major constituents. In fact the Iridaceae can be chemically distinguished from all other monocot families in the diversity of its phenolic profile and the production of several otherwise rare leaf components especially quinones, biflavonoids and the xanthone, mangiferin. Biflavonoids were found in two of the more primitive tribes of the Iridaceae. This could be a retained ancient feature in these plants and suggests considerable antiquity for the Iridaceae. Thus, although this family is specialised in many ways it may have diverged very early from the basal monocotyledon stock maybe even before the Liliiflorae. In the latter there has been one tentative identification of a biflavanone from *Lophiola americana* [47] in the Melanthiaceae [1]. Otherwise there have been no reports of biflavonoids in the Liliiflorae and none was characterised in the present survey. However, a few unidentified aglycones were noted on 2D PCs of methanolic extracts of some *Ripogonum*, *Chlorophytum*, *Ornithogalum* and *Xerophyllum* species, with similar colour reactions to biflavonoids. Most of these were present in too small amount to allow identification. The *Xerophyllum tenax* compound was isolated but its R_f and UV spectral data did not correspond to any of the common biflavonoid markers. If biflavonoids should be characterised in the Liliiflorae it would give some support for the inclusion of the Iridaceae but at present the evidence is very much against this.

From the viewpoint of leaf flavonoid chemistry, the 29 families of the super order Liliiflorae surveyed fall into two clear groups: those with flavonols alone (23 families) and those with flavonols and flavones, including *O*-methylated flavones (6 families). Such groupings, however, cut across the ordinal boundaries of the new system. The limited diversity in flavonoid structures within the Liliiflorae does not support a treatment in which many small families are recognised. The only flavonoid marker at the family level seems to be the occurrence of quercetin 3,8-dimethyl ether in Nolinaceae. However, the glycosidic variation of the flavonoids has yet to be explored on a systematic basis within these plants and it is possible that glycosidic patterns might be more characteristic for these new family groupings.

EXPERIMENTAL

Verified plant material was received from various sources, details of which are given in Table 3. The following taxa were examined for their flavonoid constituents but were available in too small amount to give a result and therefore are not included in Table 3: (1) *Stenomeris borneensis* Oliv. (Dioscoreaceae, Stenomeridoideae) US, R.S. Toroes 2011, Bila, Sumatra; (2) *Tacca leontopetaloides* (L.) O. Kuntz (Taccaceae) MASS., M. Howard *s. n.*; (3) *Croomia japonica* Miq. (Stemonaceae) US. T. Nanokawa 240497, Japan; (4) *Stichoneuron caudatum* Ridl. (Stemonaceae) NY, M. R. Henderson 22666, Malay Peninsula; (5) *Petermannia cirrosa* F. v. M. (Petermanniaceae) F, J. L. Boorman 287133, Dorrigo NSW, Australia and (6) *Lapageria rosea* R. & P. (Philesiaceae) NY, Skottsberg 1427, Concepcion, Chile.

Identification of flavonoids. Flavonoid aglycones were identified from acid hydrolysed leaf extracts using standard procedures and by comparison with authentic markers. Isorhamnetin was distinguished from quercetin and kaempferol by 2D TLC on cellulose in 50% HOAc and CAW 2:1 ($CHCl_3$–HOAc–H_2O, 2:1 saturated with H_2O). Direct 80% MeOH leaf extracts were chromatographed two dimensionally in BAW and 15% HOAc. Known glycosides were isolated and purified by standard procedures and identified from their R_f values, spectral data, acid hydrolysis to aglycone and sugar and were possible by direct comparison with authentic samples.

Identification of 3-O-methyl-8-C-methylquercetin from Dasylirion acrotrichum. 3-*O*-Methyl-8-*C*-methylquercetin was isolated from an 80% MeOH leaf extract by PC on 3MM paper in BAW and 15% HOAc. R_fs (compared with quercetin 3-methyl ether) are: BAW 91 (91), Forestal 66 (74), 50% HOAc 42 (58) and CAW (2:1) 53 (53) agreed with Lit. values [18]. UV λ_{max}^{MeOH} 260, 271, 364; +NaOAc 273, 283, 394; +H_3BO_3 265, 382; +$AlCl_3$ 280, 435 and +NaOH 278, 415 (lit. values: λ_{max}^{MeOH} 261, 272, 362; +NaOAc 276, 332', 386; +H_3BO_3 263, 301', 382 and +NaOH

276, 333′, 410). Demethylation gave a DK/DK product (8-*C*-methylquercetin) R_fs: BAW 73, Forestal 31, 50% HOAc 17 and CAW (2:1) 32 and some original compound.

Identification of flavonoid glycosides in Massonia pustulata. Three glycosides (**1–3**) were isolated from an 80% MeOH leaf extract by 3MM PC in BAW and 15% HOAc. They were provisionally identified as luteolin and apigenin 7-glucuronides (**1**, **2**) and luteolin 7,4′-diglucuronide (**3**). R_f values for **1–3** were: BAW 24, 40, 10; 15% HOAc 13, 26, 39 and H_2O 37, –, 46, respectively. Acid hydrolysis of **1** and **2** gave luteolin and apigenin, respectively with some unchanged 7-glucuronides and 3 gave luteolin 7-glucuronide but no aglycone. Compound **3** appeared DK/DK in UV + NH_3 suggesting that the second glucuronic acid moiety, which was indicated from its low mobility in BAW and high mobility in H_2O, was probably attached to the 4′-position of luteolin. UV spectral data: (**1**) λ_{max}^{MeOH} 255, 267, 350; + NaOAc 260, 376 and H_3BO_3 260, 376 (**2**) λ_{max}^{MeOH} 268, 335; + NaOAc 268, 345, 390 and H_3BO_3 268, 340 and (**3**) λ_{max}^{MeOH} 269, 325, + NaOAc 269, 320 and + H_3BO_3 269, 325 nm.

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