

## FLAVONOID PIGMENTS IN FLOWER AND LEAF OF THE GENUS *LINARIA* (SCROPHULARIACEAE)\*

B. VALDÉS

Departamento de Botánica, Facultad de Ciencias, Sevilla, Spain

(Received 11 November 1969)

**Abstract**—The pigments bracteatin-6-glucoside, aureusidin-6-glucoside, cyanidin-3-glucoside, cyanidin-3-rutinoside, aucubin and several cinnamic acids have been identified from *Linaria* flowers. From leaves of *Linaria* spp., the following pigments have been characterized: luteolin-7-glucoside, luteolin-7-diglucoside, an unidentified glycoside of eriodictyol, four glycosides of acacetin, and a glycoside of a scutellarein methyl ether. The study of flavonoids from leaves of twelve species of *Linaria* has proved to be useful for solving taxonomic problems at the specific and infraspecific level, particularly in such taxa as *L. tristis*, *L. aeruginea* and *L. nevadensis*.

**Resumen**—A partir de flores de *Linaria* se han identificado los siguientes pigmentos: bracteatin-6-glucosa, aureusidin-6-glucosa, cianidin-3-glucosa y cianidin-3-rutinoso, así como aucubina y varios ácidos cinámicos. A partir de hojas de varias especies de este género se han aislado los pigmentos: luteolín-7-glucosa, luteolín-7-diglucosa, un glicósido no identificado del eriodictiol, cuatro glicósidos de la acetina y un glicósido de la escutelareina. El estudio de flavonoides en hojas de 12 especies de *Linaria* ha resultado muy útil para ayudar a resolver varios problemas taxonómicos a nivel específico e infraespecífico en taxa tales como *L. tristis*, *L. aeruginea* y *L. nevadensis*.

### INTRODUCTION

AS A PART of an experimental taxonomic study on the genus *Linaria* (Scrophulariaceae), a survey has been made of the pigments present in the genus. This study was undertaken in an attempt to discover further characters which might correlate with the morphology of the taxa, and also because of the intrinsic interest in the nature of flower pigmentation, which is so striking and variable in this genus.

### RESULTS

#### *Flower Pigments*

Using two-dimensional chromatography, six aurones, seventeen flavones, two flavonols and two anthocyanins were detected.

Two of the aurones have been identified as aureusidin-6-glucoside and bracteatin-6-glucoside. These two pigments have the colour reactions of aurones,<sup>1</sup> and the  $R_f$ s agree with those given by Harborne<sup>2</sup> for these two pigments extracted from flowers of *Antirrhinum majus*. Harborne<sup>3</sup> found both pigments in flowers of *Linaria maroccana* and subsequently<sup>4</sup> also in flowers of *L. vulgaris*. In the present study, these pigments have been found in the

\* Part II in the series "Experimental Taxonomy of *Linaria*"; for part I, see B. VALDÉS, *Trab. Depart. Bot. Fisiol. Veg. Madrid* 1: 131-142 (1969).

<sup>1</sup> J. B. HARBORNE, *J. Chromatog.* 2, 581 (1959).

<sup>2</sup> J. B. HARBORNE, *Phytochem.* 2, 327 (1963).

<sup>3</sup> J. B. HARBORNE, *Phytochem.* 5, 111 (1966).

<sup>4</sup> J. B. HARBORNE, *Comparative Biochemistry of the Flavonoids*, Academic Press, London and New York (1967).

following taxa: *L. vulgaris* (L10, L14, L15), *L. supina* (L11, L20, L24), *L. anticaria* (L4), *L. genistifolia* (L6), *L. dalmatica* (L25), *L. hirta* (L7), *L. spartea* (L9) and *L. viscosa* (L2). In *L. reflexa* (L16) and *L. incarnata* (L13), aureusidin-6-glucoside is absent and only traces of bracteatin-6-glucoside have been detected (see Table 3).

Two anthocyanins, cyanidin-3-glucoside and cyanidin-3-rutinoside, were isolated from *L. reticulata* var. *aureopurpurea* (L17) and *L. tristis* (L18) and identified by co-chromatography with authentic pigments. Cyanidin-3-rutinoside occurs in *L. tristis* (L18) and *L. reticulata* var. *aureopurpurea* (L17), and traces of it have been detected in chromatograms of *L. anticaria* (L4), *L. alpina* (L12), *L. triphylla* (L21), *L. repens* (L8, L19) and *L. nivea* (L5). Cyanidin-3-glucoside has been detected in *L. tristis* (L18), *L. anticaria* (L4), *L. alpina* (L12), *L. repens* (L3, L8, L19, L23), *L. nivea* (L5) and traces in *L. triphylla* (L21) (see Table 3).

Aucubin occurs in flowers and leaves of all the species studied. It appears as dark spots visible in daylight in chromatograms exposed to the light for several hours. Several cinnamic acids have been observed in chromatograms of extracts from *Linaria* flowers. These appear as blue or blue-green spots in u.v. light. The presence of cinnamic acids in *Linaria* was earlier reported by Harborne.<sup>5</sup>

#### *Intraspecific Variation in Flower Colour*

In a sample of *L. repens* (L3), specimens with cream flowers appeared among specimens with normal pale-violet flowers. It has been found that specimens with pale-violet flowers have acacetin glycosides in the leaves and cyanidin-3-glucoside and an unidentified flavone ( $R_f$  0.48 in BAW) as main pigments in the flowers. Specimens with cream flowers do not have acacetin derivatives in the leaves, nor the flavone with  $R_f$  0.48 in BAW in flowers, and only have traces of cyanidin-3-glucoside; however, they have bracteatin-6-glucoside in flowers, which is absent in the normal pale-violet flowers.

From hybridization experiments which were undertaken with these plants, it appears that variation in flower colour is controlled by two pairs of factors, and thus, according to the pigment pattern, these must control several different reactions.

#### *Leaf Pigments*

In *Linaria*, flowers are very rich in pigments and, as a consequence, their use for taxonomical purposes is complicated. In the present limited study, the leaves, with their simpler pigmentation patterns, have been found of greater value.

In leaves, flavonoids, aucubin, orobanchin and several other cinnamic acid derivatives have been found. Cyanidin-3-glucoside and cyanidin-3-rutinoside have been detected only in *L. glauca* (H13) and *L. saturejoides* (H22, H23). Aurones have not been detected in leaves.

A number of the leaf pigments were isolated and studied in more detail. Following extraction from leaves of *L. spartea* (L9), luteolin-7-glucoside and luteolin-7-diglucoside were identified by co-chromatography with authentic samples. In Table 1, the colour reactions,  $R_f$ s and spectral maxima are given. In addition to occurring in five samples of *L. spartea* (L9, H24, H25, H26, H27), these luteolin derivatives occur in *L. heterophylla* subsp. *tartessiana* (H14, H15), and *L. viscosa* var. *flava* (H40, H41); both pigments are absent from *L. viscosa* var. *viscosa* (H37, H38, H39).

A glycoside isolated from *L. spartea* (L9) gave an aglycone which co-chromatographed with an authentic sample of eriodictyol without separation. It occurs in *L. spartea* (L9, H24, H25, H26, H27), *L. heterophylla* subsp. *tartessiana* (H14, H15) and *L. viscosa* var. *flava*

<sup>5</sup> J. B. HARBORNE, Z. *Naturforsch.* 21, 604 (1966).

TABLE 1. PROPERTIES OF *Linaria* FLAVONES

Components	Colour reactions			$R_f$ values ( $\times 100$ ) in				
	In visible	In u.v.	U.v. + $\text{NH}_3$	BAW	Forestal	BEW	$\text{H}_2\text{O}$	15% AcOH
Luteolin (aglycone)	Colourless	Dark	Yellow	77	47	—	—	—
Luteolin-7-glucoside	Colourless	Yellow	Bright yellow	36	—	37	1	10
Luteolin-7-diglucoside	Colourless	Yellow	Bright yellow	30	—	35	5	26
Eriodictyol glycoside	Colourless	Bluish	Blue-green	50	—	50	35	58

  

Components	$\Delta\lambda$ in					
	$\lambda_{\text{max}}$ in EtOH (nm)		EtOH-NaOAc		EtOH-H <sub>3</sub> BO <sub>3</sub>	
	Band I	Band II	Band I	Band II	Band I	Band II
Luteolin (aglycone)	256	351	+15	+34	+6	+25
Luteolin-7-glucoside	257	352	0	0	+4	+13
Luteolin-7-diglucoside	257	352	0	0	+5	+12
Eriodictyol glycoside	228	291	—	+30	—	0
		325 (infl.)				

(H40, H41), but is absent in *L. viscosa* var. *viscosa* (H37, H38, H39). The colour reactions,  $R_f$  and spectral values of this eriodictyol glycoside are given in Table 1.

Four unidentified glycosides of acacetin have been extracted from *L. tristis* (L27) and *L. repens* (L1). Two of these (27F1 and L1F1)\* have yielded, after hydrolysis, acacetin, glucose and rhamnose; a third (27F2)\* yielded acacetin and glucose; insufficient quantity of the fourth pigment (L1F2)\* was available to permit a study of the sugars, but it also yielded acacetin after hydrolysis. The colour reactions,  $R_f$  and spectral values of these four glycosides and their aglycone are listed in Table 2.

According to Hattori,<sup>6</sup> seven acacetin glycosides are known, three of which are rutinosides, which can be separated by their physicochemical properties, but since only the spectral values of linarin and acaciin are known,<sup>4</sup> the four pigments isolated in the present study could not be identified. However, comparison (Table 2) with an authentic sample of linarin showed that none is identical with this substance. It has been observed that, with the exception of sample L10 of *L. vulgaris* with yellow flowers, acacetin derivatives only occur in leaves of those species the flowers of which contain anthocyanins.

Orobanchin, a complex ester of caffeic acid, was isolated from *L. sparteae* (L9) and identified by comparison with an authentic sample. Harborne<sup>5</sup> found this pigment in *Digitalis* (Scrophulariaceae); in the present work, it was found in almost all the samples of *Linaria* studied.

Finally, a flavone has been isolated from *L. aeruginea* (L26) which yields, on hydrolysis, an aglycone, probably a scutellarein monomethyl ether, and glucose and rhamnose as sugars. The spectral values of this glycoside ( $\lambda_{\text{max}}$  283 and 330;  $\Delta\lambda$  in NaOEt, +9 (band I) and +30 (band II);  $\Delta\lambda$  in  $\text{AlCl}_3$ , +14 and +17;  $\Delta\lambda$  in NaOAc, -6 and -5;  $\Delta\lambda$  in  $\text{H}_3\text{BO}_3$ , +9 and -5) are typical of scutellarein derivatives. The colour reactions of the glycoside are also those effected for scutellarein glycosides (e.g. dark in u.v. light); its  $R_f$ s were 0.5 in BAW,

\* 27F1 and 27F2, flavones isolated from *L. tristis* (L27); L1F1 and L1F2, flavones isolated from *L. repens* (L1).

<sup>6</sup> S. HATTORI, in *The Chemistry of Flavonoid Compounds* (edited by T. A. GEISSMAN), p. 317, Pergamon Press, Oxford (1962).

TABLE 2. PROPERTIES OF ACACETIN GLYCOSIDES IN *Linaria*

Glycosides*	Colour reactions			$R_f$ values ( $\times 100$ ) in				
	In visible	In u.v.	U.v. + $\text{NH}_3$	BAW	BEW	5% AcOH	15% AcOH	$\text{H}_2\text{O}$
27 F <sub>1</sub>	Pale yellow	Dark green	Dark green	68	63	70	66	54
27 F <sub>2</sub>				50	47	42	—	—
L <sub>1</sub> F <sub>1</sub>				65	—	51	49	36
L <sub>1</sub> F <sub>2</sub>				48	54	38	—	23
Linarin				43	—	—	22	7

  

Glycosides	$\Delta\lambda$ in									
	$\lambda_{\text{max}}$ in EtOH (nm)		NaOEt		$\text{AlCl}_3$		NaOAc		$\text{H}_3\text{BO}_3$	
	Band I	Band II	Band I	Band II	Band I	Band II	Band I	Band II	Band I	Band II
27 F <sub>1</sub>	275	330	+22	+50	+23	+11	0	0	0	0
L <sub>1</sub> F <sub>1</sub>	275	330	+22	+50	—	—	0	0	0	0
L <sub>1</sub> F <sub>2</sub>	275	330	—	—	—	—	0	0	0	0
Linarin	270	330	—	—	—	—	0	-5	0	0

\* 27 F<sub>1</sub> and 27 F<sub>2</sub>, isolated from *L. tristis* (L27); L<sub>1</sub>F<sub>1</sub>, isolated from *L. repens* (L<sub>1</sub>). All gave acacetin on hydrolysis, which had  $R_f$ s 92 in BAW, 90 in Forestal and 95 in PhOH and a dark colour in u.v. changing to a slightly lighter colour with  $\text{NH}_3$ .

— = not determined.

0.54 in BEW and 0.28 in 5 per cent HOAc. The aglycone had almost identical spectral properties to scutellarein, but slightly higher  $R_f$  values in most solvents ( $R_f$ s 0.75 in BEW, 0.75 in Forestal and 0.84 in PhOH). The occurrence of this pigment is of systematic significance since it is present in all the samples of *L. aeruginea* studied (H1, H2, H3, H4, H5, H6)

TABLE 3. DISTRIBUTION OF FLOWER PIGMENTS IN *Linaria* SPECIES

Taxa	Flower pigments				
	Bracteatin-6-glucoside	Aureusin	Cyanidin-3-glucoside	Cyanidin-3-rutinoside	Aucubin
<i>L. vulgaris</i>	+	+	—	—	+
<i>L. supina</i>	+	+	—	—	+
<i>L. anticaria</i>	+	+	+	tr	+
<i>L. genistifolia</i>	+	+	—	—	+
<i>L. dalmatica</i>	+	+	—	—	+
<i>L. hirta</i>	+	+	—	—	+
<i>L. spartea</i>	+	+	—	—	+
<i>L. viscosa</i>	+	+	—	—	+
<i>L. reflexa</i>	tr	—	n.d.	n.d.	+
<i>L. incarnata</i>	tr	—	n.d.	n.d.	+
<i>L. reticulata</i> var. <i>aureopurpurea</i>	—	+	—	+	+
<i>L. tristis</i>	+	+	+	+	+
<i>L. alpina</i>	n.d.	n.d.	+	tr	+
<i>L. triphylla</i>	n.d.	n.d.	tr	tr	+
<i>L. repens</i>	+	—	+	tr	+
	—*		tr *		
<i>L. nivea</i>	n.d.	n.d.	+	tr	+

tr = trace; n.d. = not determined; \* = variable (see text).

TABLE 4. DISTRIBUTION OF LEAF PIGMENTS IN *Linaria* SPECIES

Taxa	Leaf pigments						
	Cyanidin-3-glucoside	Cyanidin-3-rutinoside	Luteolin-7-glucoside	Luteolin-7-diglucoside	Eriodictyol glycoside	Acacetin glycosides	Aucubin
<i>L. glauca</i>	+	+	n.d.	n.d.	n.d.	n.d.	n.d.
<i>L. saturejoides</i>	+	+	n.d.	n.d.	n.d.	n.d.	n.d.
<i>L. spartea</i>	—	—	+	+	+	—	+
<i>L. heterophylla</i> subsp. <i>tartessiana</i>	—	—	+	+	+	—	+
<i>L. viscosa</i> var. <i>viscosa</i>	—	—	—	—	—	—	+
<i>L. viscosa</i> var. <i>flava</i>	—	—	+	+	+	—	+
<i>L. tristis</i>	—	—	n.d.	n.d.	n.d.	+	+
<i>L. aeruginea</i> *	—	—	n.d.	n.d.	n.d.	+	+
<i>L. nevadensis</i> *	—	—	n.d.	n.d.	n.d.	+	+

n.d. = not determined; \* scutellarein derivative only detected in these species.

as well as in all the samples of *L. nevadensis* (H19, H20, H21), but it is absent in all the samples studied of *L. tristis* (H28, H29, H30, H31, H32, H33, H34, H35, H36) (see Table 4).

#### TAXONOMIC INTERPRETATIONS

In *Linaria*, yellow shades of flowers are determined by the presence in the cell vacuoles of aurones, modified by the presence of several other flavonoids and cinnamic acids. Violet, purple, brown and white shades in flowers are determined by the presence of two anthocyanins: cyanidin-3-glucoside and cyanidin-3-rutinoside, these being modified by pH, several flavonoids and cinnamic acids. Linarin is a well-known flavone in the flowers of *L. vulgaris* but no pigment with the  $R_f$  values of linarin has been found in leaf extract of any species studied.

In some cases, the study of flavonoids in leaves has not added any data to strengthen the morphological separation of some species; such is the case of *L. verticillata*, *L. lilacina* and *L. anticaria*. Although these taxa are easily separable by their morphological characters, all the samples studied of these species (see Appendix II) show the same pigment composition.

However, the chemical data obtained from the study of leaf extracts has sometimes proved very useful to solve taxonomic problems. The morphological characters used to separate *L. viscosa* var. *viscosa* (with dense inflorescence) from *L. spartea* (with lax inflorescence) are reinforced by the presence of luteolin-7-glucoside, luteolin-7-diglucoside and a glycoside of eriodictyol in *L. spartea*, which are all absent from *L. viscosa* var. *viscosa*. These three pigments are present, however, in *L. viscosa* var. *flava*, and this supports the separation of this variety (with decumbent stems covered by glandular hairs) from *L. viscosa* var. *viscosa* (with erect stems and only inflorescence glandular-hairy). On the other hand, the taxon described by Vicioso<sup>7</sup> as *L. heterophylla* subsp. *tartessiana*, also possesses these three pigments, and this reinforces the morphological resemblance of this taxon with *L. spartea*, to which it should perhaps be more correctly referred.

The separation between *L. tristis* (with broader flat leaves and bigger flowers with broader tube) and *L. aeruginea* (with narrower and grooved leaves and flowers with narrower tube) is sometimes very difficult using only morphological characters, but the chromatograms of both species are very distinctive. Similarly, *L. nevadensis*, which has been considered as

<sup>7</sup> C. VICIOSO, *Anal. Inst. Bot. Cavanilles* 6 (2), 5 (1946).

separate species by several authors, and subordinate to *L. supina* by others, must be subordinate to *L. aeruginea*; the morphology of these taxa suggest this affinity and this view is reinforced by the chemical study of several samples of *L. nevadensis*, since the chromatograms are identical to those obtained from *L. aeruginea*.

It has been observed that within the same species, the samples show some differences as far as pigment content is concerned. Such differences are more marked in cultivated samples than in samples of wild origin. Generally, however, such differences affect only pigments of minor importance, and they have not been taken into consideration in the present study. Nevertheless, it should be noted that the two samples of *L. clementei* studied (H11, H12), although very similar morphologically, are quite distinct chemically.

## EXPERIMENTAL

### Plant Material

Plant material consisted of fresh samples for several species from plants in cultivation at The University of Liverpool Botanic Gardens (Ness, Cheshire), and herbarium material. The former are listed in Appendix I and are preceded by the letter L; the later are listed in Appendix II and are preceded by the letter H. In the text, the reference number of the samples of each species is given in brackets.

### Methods

Flavonoids were extracted from flowers or leaves with 70% or 95% ethanol; anthocyanins, with 1% MeOH-HCl. Chromatography was carried out on Whatman No. 1 paper using the solvents and techniques of Harborne.<sup>1</sup> Purification of pigments was carried out on Whatman No. 3 paper using BAW, 15% acetic acid and BEW. Pigments were identified by spectral and chromatographic comparison with authentic markers and by hydrolysis to aglycones and sugars.

**Acknowledgements**—This work was made possible by an interchange scholarship between The Consejo Superior de Investigaciones Científicas and The British Council, which enabled the author to work at the University of Liverpool; to both organizations the author expresses his thanks, and also to Professor V. H. Heywood for the provision of facilities in the Department of Botany. The author thanks Dr. Harborne, in whose laboratory this study was carried out, for his advice and encouragement, and for the provision of authentic pigment samples. Dr. P. Gibbs kindly corrected the English of the manuscript.

## APPENDIX I

Origin of samples grown at the University of Liverpool Botanic Gardens (Ness, Cheshire).  
The number and origin of the samples are indicated.

L1.—*L. repens* (L.) Miller; Jardín Botánico (Madrid). L2.—*L. viscosa* (L.) Dum.-Courset; Chiclana (Cádiz, Spain), V-1961, *Borja & Rodriguez*. L3.—*L. repens* (L.) Miller; Jardín Botánico (Madrid). L4.—*L. anticaria* Boiss. & Reut. var. *angustifolia* Boiss. & Reut.; Priego (Córdoba, Spain); Sierra Halconera, VI-1960, *Borja*. L5.—*L. nivea* Boiss. & Reut.; Peñalara (Madrid), VIII-1957, *Rivas-Goday & Monasterio*. L6.—*L. genistifolia* (L.) Miller; Jardín Botánico (Madrid). L7.—*L. hirta* (L.) Moench; Priego (Córdoba, Spain), VI-1960, *Borja*. L8.—*L. repens* (L.) Miller; The Botanic Garden of the University (Uppsala). L9.—*L. sparteae* (L.) Hoffmanns. & Link; Aldeanueva de Atienza (Guadalajara, Spain), VIII-1965, *Silvestre*. L10.—*L. vulgaris* Miller; Hortus Medicinalis Lodziensis (Ludz, Poland). L11.—*L. supina* (L.) Chaz.; Jardin Botanique de Dijon (France). L12.—*L. alpina* (L.) Miller; Jardin Botanique de Dijon (France). L13.—*L. incarnata* (Vent.) Sprengel; The University Botanic Garden (St. Andrews, Scotland). L14.—*L. vulgaris* Miller; Institutum Plantarum Medicinalium (Budapest, Hungary). L15.—*L. vulgaris* Miller.\* L16.—*L. reflexa* (L.) Desf.\* L17.—*L. reticulata* Desf. var. *aureopurpurea*.\* L18.—*L. tristis* (L.) Miller.\* L19.—*L. repens*

\**Hortus botanicus hauniensis* (Copenhagen).

(L.) Miller.\* L20.—*L. supina* (L.) Chaz.\* L21.—*L. triphylla* (L.) Miller.\* L23.—*L. repens* (L.) Miller.† L24.—*L. supina* (L.) Chaz.† L25.—*L. dalmatica* (L.) Miller.† L26.—*L. aeruginea* (Gouan) Cav.; El Escorial (Madrid, Spain), VI-1966, *Getliffe, Gilbert & Valdés*. L27.—*L. tristis* (L.) Miller; Montes de Tolox (Málaga, Spain), V-1966, *Getliffe, Novo & Valdés*.

## APPENDIX II

Origin of herbarium material sampled for flavonoid studies. The name of the species, the number of the sample, locality, date of collection and collectors are indicated. All the localities are Spanish.

- L. aeruginea* (Gouan) Cav.: H1, El Escorial (Madrid), VI-1966, *Getliffe, Gilbert & Valdés*; H2, Beas de Segura (Jaén), VI-1954, *Galiano*; H3, Calamocha (Teruel), VIII-1951, *Galiano*; H4, Sierra Nevada (Granada): Las Vívoras, VI-1966, *Getliffe & Valdés*; H5, Sierra Nevada (Granada): between Peñones de San Francisco and Dornajo, VI-1966, *Getliffe & Valdés*; H6, Sierra Nevada (Granada): Monte Dornajo, VI-1966, *Getliffe & Valdés*.
- L. anticaria* Boiss. & Reut.: H7, Torcal de Antequera (Málaga), V-1966, *Getliffe, Novo & Valdés*; H8, Torcal de Antequera (Málaga), V-1966, *Getliffe, Novo & Valdés*; H9, Sierra Gorda (Málaga), V-1966, *Getliffe, Novo & Valdés*; H10, Sierra de Alhama (Granada), V-1966, *Getliffe, Novo & Valdés*.
- L. clementei* Haens. ex Boiss.: H11, between Coín and Ojén (Málaga), V-1966, *Getliffe, Novo & Valdés*; H12, Sierra de Carratraca (Málaga), V-1966, *Getliffe, Novo & Valdés*.
- L. glauca* (L.) Chaz.: H13, Aranjuez (Madrid), VI-1966, *Getliffe, Gilbert & Valdés*.
- L. heterophylla* Desf., subsp. *tartessiana* C. Vicioso: H14 and H15, Palos de Moguer (Huelva), II-1966, *Novo*.
- L. lilacina* Lange: H16, Sierra de Cazorla (Jaén): Quesada, V-1962, *Kjellqvist & Löve*; H17, Sierra de Cazorla (Jaén): El Chorro, V-1966, *Kjellqvist & Löve*; H18, Sierra de la Cabrilla (Jaén), VI-1948, *Heywood*.
- L. nevadensis* Boiss. & Reut.: H19, Sierra Nevada (Granada): Dehesa de San Jerónimo, VI-1966, *Getliffe & Valdés*; H20, Sierra Nevada (Granada): Hoya de la Mora, VI-1966, *Getliffe & Valdés*; H21, Sierra Nevada (Granada): Peñones de San Francisco, VI-1966, *Getliffe & Valdés*.
- L. saturojoides* Boiss.: H22, Sierra de Almijara (Granada): Sierra del Chaparral, V-1966, *Getliffe, Novo & Valdés*; H23, Sierra de Almijara (Málaga): Montes de Cómpeta, V-1966, *Getliffe, Novo & Valdés*.
- L. sparteae* (L.) Hoffmanns. & Link: H24, El Escorial (Madrid), VI-1966, *Getliffe, Gilbert & Valdés*; H25, between Madrid and Arenas de San Pedro (Madrid), VI-1966, *Getliffe, Gilbert & Valdés*; H26 and H27, between Arenas de San Pedro and Candeleda (Ávila), VI-1966, *Getliffe, Gilbert & Valdés*.
- L. tristis* (L.) Miller: H28, Gibraltar, IV-1965, *Gilbert & D. Wood*; H29, Montes de Tolox (Málaga), V-1966, *Getliffe, Novo & Valdés*; H30, Sierra Bermeja (Málaga), V-1966, *Getliffe, Novo & Valdés*; H31, Sierra de Grazalema (Cádiz): Cerro de San Cristobal, V-1966, *Getliffe, Novo & Valdés*; H32, between Ronda and San Pedro de Alcántara (Málaga), V-1966, *Getliffe, Novo & Valdés*; H33, Sierra Tejeda (Málaga), V-1966, *Getliffe, Novo & Valdés*; H34, Sierra de Carratraca (Málaga), V-1966, *Getliffe, Novo & Valdés*;

\* *Hortus botanicus hauniensis* (Copenhagen).

† *Hortus botanicus Universitatis* (Budapest).

H35, Sierra de Grazalema (Cádiz), V-1966, *Getliffe, Novo & Valdés*; H36, Sierra Bermeja (Málaga), V-1966, *Getliffe, Novo & Valdés*.

*L. viscosa* (L.) Dum.-Courset var. *viscosa*: H37, between Grazalema and Ronda (Cádiz), V-1966, *Getliffe, Novo & Valdés*; H38, Sierra Nevada (Granada): Peñones de San Francisco VI-1966, *Getliffe & Valdés*; H39, Sierra Nevada (Granada): Hoya de la Mora, VI-1966, *Getliffe & Valdés*.

*L. viscosa* (L.) Dum.-Courset var. *flava* (Boiss.): H40, Sierra de Almijara (Málaga): Montes de Cómpeta, V-1966, *Getliffe, Novo & Valdés*; H41, Sierra de Almijara (Granada): Sierra del Chaparral, V-1966, *Getliffe, Novo & Valdés*.