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A CHEMOSYSTEMATIC ANALYSIS OF TRIBES OF ASTERACEAE INVOLVING SESQUITERPENE LACTONES AND FLAVONOIDS*

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Key Word Index—Asteraceae; sesquiterpene lactones; *O*-glycosylflavonoids; *O*-methylflavonoids; biochemical evolution.

Abstract—The correlation of oxidation levels and skeletal specializations of sesquiterpene lactones reported to occur in genera of the Asteraceae suggest the distribution of the corresponding tribes along chemical gradients. The evolutionary polarity of these gradients was established by concomitant consideration of the evolutionary replacement of *O*-glycosylflavonoids by *O*-methylflavonoids. Information on structure and occurrence of sesquiterpene lactones as well as of flavonoids was obtained in a literature survey up to 1984.

INTRODUCTION

Previous work [2, 3] on the oxidation value (O) and the skeletal specialization (S) of sesquiterpene lactones led to the representation of evolutionary advancement parameters for tribes of Asteraceae (EA_O, EA_S) along two gradients (Fig. 1). However, as is common in chemosystematic work, the evolutionary polarity adopted within the gradients remained hypothetical. The present work aims to apply or even to extend existing methodology in order to verify if and how flavonoid-based data can be used in the solution of the problem.

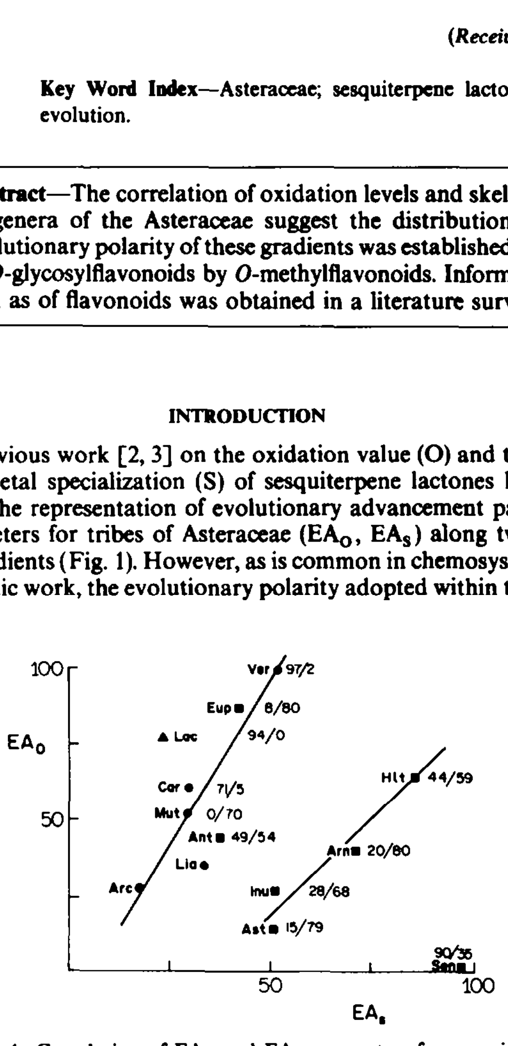

Fig. 1. Correlation of EA_O and EA_S parameters for sesquiterpene lactones from tribes of Asteraceae, indicated by the three initials of their names given in full in Table 2 and classified into Lactuceae (triangle), Asteroideae group 1 (circles) and Asteroideae group 2 (squares). Also given are EA_G/EA_M values.

*Part 32 in the series 'Plant Chemosystematics and Phylogeny'. For part 31 see ref. [1].

DEFINITIONS AND METHODS

Evolutionary Advancement Indices for selected taxa (here tribes) with respect to oxidation values of rings A and B of flavonoids (EA_{OA}, EA_{OB}) were determined as described for other classes of compounds [4]. Another form of presentation of flavonoid data concerns the Δ_{OA} and Δ_{OB} values, i.e. the differences between the highest and the lowest oxidation values of rings A and B of flavonoids isolated from the chosen taxa (here tribes). In order to facilitate comparisons, the EA_{OA} and EA_{OB} values are given in percentages of the highest observed value and the Δ indices are given in percentages of the largest difference between highest and lowest observed values for a tribe. In the case of sesquiterpene lactones, as in the previous paper [3], not only EA_O values (based on the oxidation levels of the compounds) but also EA_S values (based on the skeletal specialization of the compounds) are used.

Evolutionary Advancement Indices for selected taxa (here tribes) with respect to *O*-glycosylflavonoids (EA_G), *O*-methylflavonoids (EA_M) and flavonoids with some of their hydroxyls protected (EA_P) are calculated by Equations 1, 2 and 3 respectively. *N* represents the total number of flavonoids (irrespective of the presence and the nature of the substituents) isolated from each of the species *a*, *b* . . . *z* of a given tribe; *n* represents the number of flavonoids substituted by at least one *O*-glycosyl unit (Eq. 1), at least one *O*-methyl unit (Eq. 2) and possessing specific *P* values (*P* = number of protected OH groups/total number of oxy-groups) (Eq. 3) isolated from each of

$$EA_G = 100\frac{\sum_a^z n_G}{\sum_a^z N} \quad \text{(Eq. 1)} \qquad EA_M = 100\frac{\sum_a^z n_M}{\sum_a^z N} \quad \text{(Eq. 2)} \qquad EA_P = 100\frac{\sum_a^z n_P}{\sum_a^z N} \quad \text{(Eq. 3)}$$

the species $a, b \ldots z$ of the same tribe. The determination of these flavonoid parameters is exemplified in Table 1.

In the above calculations, tribes are considered to be collections of species. The same applies to 'number of occurrences' (Table 2) i.e. a particular flavonoid, reported for x species of the tribe is counted x times in the calculation of this 'number'. Flavonoids of all types (aurones, chalcones, flavones, flavonols, dihydro-

Table 1. Determination of flavonoid parameters exemplified by the composition of species (A–P) belonging to Lactuceae

Flavonoid substitution 3	5	6	7	2′	3′	4′	5′	Occurrence in genera	Species	n_G	n_M	n_P
OH	OH		OH	OG		OH	OH	*Heywoodiella*	a	1	0	1/6
OH	OH		OH	OG		OH	OH	*Heywoodiella*	b	1	0	1/6
OH	OH		OH	OG		OH	OH	*Heywoodiella*	c	1	0	1/6
OH	OH		OH		OH	OG		*Hieracium*	d	1	0	1/5
OH	OH		OH	OH	OH		OH	*Hieracium*	e	0	0	0/6
OH	OH		OG		OH	OH		*Hieracium*	f	1	0	1/5
OH	OH		OG			OH		*Hieracium*	g	1	0	1/4
OH	OH		OG	OH		OH		*Hispidella*	h	1	0	1/5
OG	OH		OH		OH	OH		*Lactuca*	i	0	0	1/5
OH	OH		OG		OH	OH		*Sonchus*	j	1	0	1/5
OH	OH		OG		OH	OH		*Sonchus*	k	1	0	1/5
OH	OH		OG		OH	OH		*Tragopogon*	l	1	0	1/5
OH	OH		OG		OH	OH		*Tragopogon*	m	1	0	1/5
OH	OH		OG		OH	OH		*Tragopogon*	n	1	0	1/5
	OH	OG				OH		*Launea*	o	1	0	1/3
	OH	OG		OH	OH	OH		*Launea*	p	1	0	1/4

N for tribe LAC = 16; $EA_G = 100(15/16) = 94$; $EA_M = 0$; $EA_P = 3.13/16 = 0.195$.

Table 2. Chemical parameters in percentages for flavonoids from tribes of Asteraceae

Tribe	EA_{OA}	EA_{OB}	Δ_{OA}	Δ_{OB}	EA_G	EA_M	EA_P	Number of occurrences
Lactuceae*	36.5	95.4	50.0	80.0	94.0	0	31.0	16
	(0.15)	(0.31)	(0.66)	(0.88)			(0.19)	
Vernonieae†	38.7	76.7	50.0	40.0	97.0	2.0	58.0	238
	(0.16)	(0.27)	(0.66)	(0.44)			(0.37)	
Cardueae†	64.9	46.4	75.0	60.0	71.0	5.0	67.0	80
	(0.26)	(0.11)	(1.00)	(0.66)			(0.46)	
Mutisieae†	34.3	0	50.0	60.0	0	70.0	53.0	13
	(0.14)		(0.66)	(0.66)			(0.34)	
Anthemideae‡	56.2	59.4	100.0	80.0	49.0	54.0	60.0	195
	(0.23)	(0.16)	(1.33)	(0.88)			(0.38)	
Arnicineae†	90.4	30.5	25.0	40.0	20.0	80.0	63.0	10
	(0.37)	(0.04)	(0.33)	(0.44)			(0.40)	
Astereae†	35.3	42.8	100.0	100.0	15.0	79.0	81.0	109
	(0.14)	(0.09)	(1.33)	(1.11)			(0.52)	
Calenduleae‡	40.9	100.0	0	0	100.0	100.0	62.0	3
	(0.17)	(0.33)	(0)	(0)			(0.40)	
Eupatorieae‡	37.7	45.4	100.0	100.0	8.0	80.0	100.0	180
	(0.15)	(0.11)	(1.33)	(1.11)			(0.64)	
Heliantheae‡	30.8	22.4	100.0	80.0	44.0	59.0	69.0	289
	(0.13)	(0.01)	(1.33)	(0.88)			(0.44)	
Inuleae‡	62.4	27.1	100.0	80.0	28.0	68.0	80.0	109
	(0.25)	(0.03)	(1.33)	(0.88)			(0.51)	
Senecioneae‡	58.5	57.9	25.0	40.0	90.0	35.0	57.0	22
	(0.24)	(0.16)	(0.33)	(0.44)			(0.37)	
Tageteae‡	100.0	61.5	25.0	20.0	33.0	33.0	20.0	8
	(0.41)	(0.17)	(0.33)	(0.22)			(0.13)	

*Subfamily Cichoriodeae; subfamily Asteroideae, †group 1; ‡group 2 [7].

Values of EA_{OA}, EA_{OB} [4], Δ_{OA}, Δ_{OB} [3] and EA_P (in parentheses) were calculated by the direct application of the respective equations; and transformed into percentages as explained in the text.

flavonols) are counted indiscriminately. The data bank for flavonoids of Asteraceae was compiled from surveys of the literature given in two books [5, 6] and completed up to 1984 in the style adopted in the latter of these compendia (Tables 3–6).

RESULTS

The evolutionary histories of rings A and B of flavonoids are not necessarily connected. Indeed both the biosynthetic pathways, respectively the polyketide and

Table 3. Flavones and flavonols in Asteraceae, reported between 1981 and 1984

Substitution	Plant source *Tribes	Genera	Ref.
5,6,7,4′-Tetra-OH			
6,7,4′-tri-Me	Ant	*Artemisia*	10
		Ursinia	11
6-Me		*Artemisia*	12, 13
6,7-di-Me		*Artemisia*	14
5,6,7,3′,4′,5′-hexa-OH			
6,3′-di-Me		*Artemisia*	12
6,3′,5′-tri-Me		*Artemisia*	12, 14
6,3′,4′-tri-Me		*Artemisia*	15, 12
6,7,3′,5′-tetra-Me		*Artemisia*	13
5,6,7,3′,4′-penta-OH			
6,3′,4′-tri-Me		*Artemisia*	12, 17
6,7,3′,4′-tetra-Me		*Artemisia*	14
6,7,3′-tri-Me		*Artemisia*	14, 18
6,7-di-Me		*Artemisia*	12, 19, 20
		Tanacetum	21
6-Me		*Artemisia*	12, 19
7-G		*Artemisia*	22
5,7,3′,4′-tetra-OH		*Chrysanthemum*	23
		Artemisia	12
3′,4′-di-Me		*Artemisia*	12
7-G		*Artemisia*	12
		Chrysanthemum	24, 25
7,3′-di-Me		*Artemisia*	20
5,7,4′-tri-OH		*Artemisia*	12
		Chrysanthemum	23
7-G		*Chrysanthemum*	23, 24
		Matricaria	26
		Otanthus	26
6,8-di-C-G		*Artemisia*	27
7,4′-di-Me		*Artemisia*	20
4′-Me		*Artemisia*	20
8-C-G		*Lemoanthemum*	28
7-di-G		*Chrysanthemum*	24
6,7,3′,4′,5′-penta-OH			
6,5′-di-Me		*Artemisia*	15
6,3′,5′-tri-Me		*Artemisia*	15
5,6,7,2′,4′,5′-penta-OH			
6,5′-di-Me		*Artemisia*	15
7,3′,4′,5′-tetra-OH			
5′-Me		*Artemisia*	15
5,7,3′,4′,5′-penta-OH			
3′,5′-di-Me		*Artemisia*	14
3,5,6,7,3′,4′-hexa-OH			
3,6,3′-tri-Me		*Tanacetum*	21
		Matricaria	29
		Achillea	30
3,6,7,3′,4′-penta-Me		*Artemisia*	31
3,6-di-Me		*Tanacetum*	32
3,6,7,3′-tetra-Me		*Artemisia*	18
3,6,7,3′,4′-penta-OH		*Artemisia*	33, 34
6-Me		*Matricaria*	29
3,6,7-tri-Me		*Matricaria*	29

Table 3. (*Continued*)

Substitution	*Tribes	Plant source Genera	Ref.
3,6-di-Me		*Matricaria*	35
6,7-di-Me		*Matricaria*	35
6,3′-di-Me		*Matricaria*	29
3,5,6,7,4′-penta-OH			
6,7-di-Me		*Artemisia*	36
3,5,7,4′-tetra-OH			
3-Me		*Artemisia*	12
		Chrysanthemum	24
		Artemisia	18
7-Me		*Artemisia*	36
3,5,7,3′,4′-penta-OH		*Chrysanthemum*	10
3,3′-di-Me		*Artemisia*	12
3,4′-di-Me		*Tanacetum*	32
6,7,3,3′-tetra-OH			
3,6-di-Me		*Artemisia*	15
3,6,7,3′,4′-penta-OH			
3,6-di-Me		*Artemisia*	15
3,5,6,7,8,4′-hexa-OH			
3,6,3′,4′-tetra-Me		*Artemisia*	15
3,5,6,3′,4′-hexa-OH		*Artemisia*	22
3,6,3′,4′-tetra-Me		*Artemisia*	12, 31
6,7,4′-tri-Me		*Artemisia*	12, 31
5,4′-di-OH		*Matricaria*	38
5,6,7,4′-tetra-OH			
6,7-di-Me		*Artemisia*	36
6-Me		*Tanacetum*	32
5,7,4′-tri-OH			
4′-Me	Arn	*Arnica*	39
5,6,7,4′-tetra-OH			
6,4′-di-Me		*Arnica*	39
6-Me		*Arnica*	39
5,6,7,3′,4′-penta-OH			
6,3′-di-Me		*Arnica*	39
3,5,6,7,4′-penta-OH			
6-Me		*Arnica*	39
6,4′-di-Me		*Arnica*	39
3,5,6,7,8,3′,4′,5′-octa-OH			
3,6,7,8,4′-penta-Me	Ast	*Gutierrezia*	40
3,5,6,7,8,4′-hexa-Me		*Gutierrezia*	40
3,6,8,4′-tetra-Me		*Gutierrezia*	40
3,5,7,3′,4′,5′-hexa-OH			
3′,4′-di-Me		*Haplopappus*	40
3,3′,4′-tri-Me		*Haplopappus*	40
3,5,7,3′,4′-penta-OH		*Haplopappus*	41, 42, 43
3,3′-di-Me-4′-*i*-Mebut		*Gutierrezia*	40
3,3′-di-Me-4′-*i*-Val		*Gutierrezia*	40
3′-Me		*Haplopappus*	41, 42, 43
3-G		*Haplopappus*	41, 42, 43
7,3′-di-Me		*Haplopappus*	41
3,3′-di-Me		*Haplopappus*	41
3,7-di-Me		*Haplopappus*	41
3-Me		*Haplopappus*	41, 42, 45
3-G-3′-Me		*Haplopappus*	43, 45
3,5,6,7,3′,4′-hexa-OH			
3,6-di-Me		*Haplopappus*	42, 46
3,6,4′-tri-Me		*Haplopappus*	46
3,5,7-tri-OH			
3-Me		*Haplopappus*	41
3,5,7,4′-tetra-OH			
3,5,7,4′-tetra-OH			
3-Me		*Haplopappus*	46

Table 3. (*Continued*)

Substitution	*Tribes	Plant source Genera	Ref.
7-Me		*Haplopappus*	43
3,5,7,3′,4′,5′-hexa-OH		*Haplopappus*	46
3,5,6,7,4′-penta-OH			
3,6-di-Me		*Haplopappus*	42
5,6,7,8,4′-penta-OH			
6,7,8-tri-Me		*Baccharis*	47
5,6,7,4′-tetra-OH			
7,4′-di-Me		*Baccharis*	47
6-Me		*Haplopappus*	46
6-G		*Haplopappus*	46
5,7,4′-tri-OH		*Baccharis*	48, 49
7,4′-di-Me		*Baccharis*	48, 49
4′-Me		*Baccharis*	48, 49
7-Me		*Baccharis*	48, 49
6,8-di-C-G		*Haplopappus*	43
5,7,3′,4′-tetra-OH			
3′,4′-di-Me		*Baccharis*	49
3′-Me		*Haplopappus*	42
4′-Me		*Haplopappus*	46
3,5,6,7,8,3′,4′-hepta-OH			
3,6,7,3′-tetra-Me		*Gutierrezia*	50
3,6,7,8,3′-penta-Me		*Baccharis*	51
3,5,6,7,3′,4′,5′-hepta-OH			
3,6,7,3′-tetra-Me		*Gutierrezia*	50
3,5,7,4′-tetra-OH			
3-Me		*Haplopappus*	15
7-Me		*Haplopappus*	15
3-G		*Haplopappus*	45
3,6,7,8,3′,4′-Hexa-OH			
6,7,8,3′-tetra-Me		*Baccharis*	52
5,6,7,3′,4′-penta-OH			
6-Me		*Haplopappus*	43
5,7,3′,4′-tetra-OH			
7-G	Car	*Centaurea*	53
3,5,7,3′,4′-penta-OH		*Centaurea*	53
3,5,6,7,3′,4′-hexa-OH			
6-Me		*Centaurea*	53
6-Me-7-G		*Centaurea*	53
6-Me-7-G		*Centaurea*	53
3,5,7,4′-tetra-OH			
7-Me		*Centaurea*	53
5,7,4′-tri-OH		*Centaurea*	53
7-G		*Centaurea*	53
5,6,7,3′,4′-penta-OH			
6,7-di-Me		*Centaurea*	53
6-Me		*Centaurea*	53
6,3′-di-Me		*Amberboa*	55
6,7,3′,4′-tetra-Me		*Amberboa*	56
5,7,3′,4′-tetra-OH		*Amberboa*	54
5,6,7,4′-tetra-OH		*Amberboa*	54
6,7,4′-tri-Me		*Amberboa*	54
6,7-di-Me		*Amberboa*	54
6-Me		*Cirsium*	57
		Saussurea	58
6,4′-di-Me		*Saussurea*	58
5,7,4′-tri-OH		*Cirsium*	57
7-G-4′-Me		*Cirsium*	59
5,7,8,4′-tetra-OH			
8-Me		*Centaurea*	60
5,7,4′-tri-OH			
7-G	Cic	*Launea*	61, 62

Table 3. (*Continued*)

Substitution	Plant source *Tribes	Plant source Genera	Ref.
5,7,3′,4′-tetra-OH			
7-G		*Launea*	62
5,6,7,8,2′,3′,4′,5′,octa-OH			
5,6,7,8,2′,3′,4′,5′,octa-Me	Eup	*Ageratum*	63
5,6,7,2′,3′,4′,5′-hepta-OH			
5,6,7,2′,3′,4′,5′-hepta-Me		*Ageratum*	63
5,6,7,8,2′,4′,5′-hepta-OH			
5,6,7,8,2′,4′,5′-hepta-Me		*Ageratum*	63
5,6,7,8,3′,4′-hexa-OH			
5,6,7,8,3′,4′-hexa-Me		*Eupatorium*	64
5,6,7,8,3′-penta-Me		*Eupatorium*	64
5,6,7,3′,4′-penta-OH			
6,7,4′-tri-Me		*Lasioloena*	65
6-*Me*		*Brickellia*	66
6,7-di-Me		*Brickellia*	66
6,7,4′-tri-Me		*Brickellia*	66, 67
6,4′-di-Me		*Brickellia*	67
6,3′-di-Me		*Eupatorium*	68
5,6,7,4′-tetra-OH			
6-Me		*Brickellia*	67
6,7,4′-tri-Me		*Brickellia*	66, 67
6,7-di-Me		*Aristeguetia*	69
6,4′-di-Me		*Brickellia*	66
5,7,3′,4′-tetra-OH			
7-Me		*Eupatorium*	70
		Chromolaena	70
7-G		*Stevia*	71
5,6,7,2′,4′,5′-hexa-OH			
6,5′-di-Me		*Brickellia*	72
3,5,7,3′,4′-penta-OH		*Brickellia*	73
7-Me		*Chromolaena*	70
		Eupatorium	70
3-G		*Brickellia*	66
3-G-3′-Me		*Brickellia*	66
3-Me		*Brickellia*	73
3,7-di-Me		*Brickellia*	73
3,5,6,7,3′,4′-hexa-OH			
6,7,4′-tri-Me		*Brickellia*	67
3,6,4′-tri-Me		*Brickellia*	67, 71
3,6,7,3′,4′-penta-Me		*Symphyopappus*	74
		Brickellia	67
3,6,7,4′-tetra-Me		*Brickellia*	67
3-G-3-Me		*Brickellia*	67
3-G-6,7-di-Me		*Brickellia*	67
3-SO_3-6-Me		*Brickellia*	67
3-SO_3-6,7-di-Me		*Brickellia*	67
6,7-di-Me		*Eupatorium*	70
		Chromolaena	70
3,6,7-tri-Me		*Brickellia*	73
3,6-di-Me-7-G		*Brickellia*	73
3,6,3′,4′-tetra-Me		*Brickellia*	73
6,7-di-Me		*Brickellia*	73
3,6,7,3′,4′-penta-OH			
6-Me		*Brickellia*	66
3,6,7,4′-tetra-Me		*Brickellia*	66
3,6,7,3′-4′-penta-Me		*Brickellia*	66
3-G-6,7-di-Me		*Brickellia*	66
3-G-6,7,3′-tri-Me		*Brickellia*	66
6,7,4′-tri-Me		*Brickellia*	66
3-SO_3-6,7,4′-tri-Me		*Brickellia*	66
3,7,3′-tri-Me		*Chromolaena*	75

Table 3. (*Continued*)

Substitution	*Tribes	Plant source Genera	Ref.
3,5,7-tri-OH		*Chromolaena*	76
3,5,6,7-tetra-OH			
6-Me		*Chromolaena*	76
3,4,7,8,3′,4′-hexa-OH			
3,7,8,3′-tetra-Me		*Chromolaena*	75
3,5,7,4′-tetra-OH			
3-G		*Stevia*	71
3-Me		*Acritopappus*	77
3,7-di-G		*Eupatorium*	78
7-G		*Eupatorium*	78
3,5,6,7,4′-penta-OH			
3-G-6-Me		*Brickellia*	66, 37
3-G-6,7-di-Me		*Brickellia*	66
3,6,7-tri-Me		*Brickellia*	73
5,6,7,8-tetra-OH			
5,6,7,8-tetra-Me-3′,4′-O_2CH_2		*Ageratum*	63
		Eupatorium	64
5,6,7,8,5′-penta-OH			
5,6,7,8,5′-penta-OH-3′,4′-O_2CH_2		*Ageratum*	63
		Eupatorium	64
		Conoclinium	79
5,7,4′-tri-OH		*Goyazanthus*	80
7-G		*Eupatorium*	78
4′-Me		*Eupatorium*	78
4′-G		*Stevia*	71
3,5,6,7,4′-penta-OH			
3,7-di-Me	Hlt	*Neurolaena*	81
7-Me-7-G		*Neurolaena*	82
3,5,6,7,3′,4′-hexa-OH		*Neurolaena*	81
3-Me-7-SO_3		*Neurolaena*	82
3,6-di-Me		*Neurolaena*	81, 82, 83
3,7-di-Me-6-G		*Neurolaena*	82
3-Me-6-G		*Neurolaena*	82, 83
7-G-6,3′-di-Me		*Clibadium*	84
6-Me-7-G		*Neurolaena*	82
		Desmanthodium	82
7-G		*Neurolaena*	81, 82, 83
		Desmanthodium	85
		Clibadium	84
3,7-di-Me		*Neurolaena*	82, 83
3,6-di-Me-7-G		*Neurolaena*	83
7-G-3′-Me		*Desmanthodium*	85
		Clibadium	84
3,5,6,7,4′-penta-OH			
3-Me		*Neurolaena*	82
3,7-di-G		*Verbesina*	86
7-G		*Clibadium*	84, 85
3-G		*Verbesina*	86
		Clibadium	88, 85
3-G		*Ichthyothere*	89
3-G		*Heliopsis*	90
3,5,7,3′,4′-penta-OH	Inu	*Helichrysum*	91
		Achyrocline	92
3-Me		*Achyrocline*	92
3-G		*Pulicaria*	93
3,3′-di-Me		*Ditrichia*	94
3-G		*Helichrysum*	95, 96
3,5,6,7,8-penta-OH			
6,7,8-tri-Me		*Achyrocline*	97
3,6,8-tri-Me		*Gnaphalium*	98
3,6,7,8-tetra-Me		*Gnaphalium*	99

Table 3. (*Continued*)

Substitution	*Tribes	Plant source Genera	Ref.
3,6,7-tri-Me		*Gnaphalium*	100
6,7,8-tri-Me		*Helichrysum*	101
6,8-di-Me		*Helichrysum*	101
3,7,8-tri-Me		*Achyrocline*	102
3,5,7,8-tetra-OH			
3,7,8-tri-Me		*Achyrocline*	97, 102
7,8-di-Me		*Achyrocline*	97
		Helichrysum	103
3,8-di-Me		*Gnaphalium*	104
3,5,7-tri-OH		*Achyrocline*	92
3-Me		*Achyrocline*	92
3-Me		*Helichrysum*	96, 101
3,5,6,7-tetra-OH			
3,7-di-Me		*Helichrysum*	105
3,6-di-Me		*Gnaphalium*	99
3,5,6,7,3′,4′-hexa-OH			
3,6,7,3′,4′-penta-Me		*Helichrysum*	106
		Pluchea	107
3,7-di-Me		*Pulicaria*	93, 108
3,5,7-tri-Me		*Pulicaria*	93
3,5,7,3′-tetra-Me		*Pulicaria*	93
3,7,4′-tri-Me		*Pulicaria*	108
3,5,6,7,4′-penta-OH			
3,7-di-Me		*Pulicaria*	108
3-Me-6-G		*Pulicaria*	108
3,6,7-tri-Me		*Pulicaria*	108
3,5,7,8,4′-penta-OH			
3,7-di-Me		*Pluchea*	107
5,6,7,8-tetra-OH			
6,7-di-Me		*Gnaphalium*	100
5,6,7,8-tetra-OH			
6,7-di-Me		*Gnaphalium*	100
5,7,4′-tri-OH		*Gnaphalium*	104
		Helichrysum	91
7-G		*Gnaphalium*	96, 104, 109
4′-G		*Gnaphalium*	96
5,7,3′,4′-tetra-OH		*Helichrysum*	96
		Gnaphalium	104
7-G		*Gnaphalium*	104
5,6,7,3′,4′-penta-OH			
6,3′-di-Me		*Gnaphalium*	104
5,7,8-tri-OH			
7,8-di-Me		*Achyrocline*	102
3,7,4′-tri-OH		*Helichrysum*	109
3,5,7,4′-tetra-OH	Mut	*Trixis*	110
7-Me		*Trixis*	110
3,7-di-Me		*Gochnatia*	111
3,5,7-tri-OH			
7-Me		*Trixis*	111
3,5,7,3′,4′-penta-OH			
3,3′-di-Me		*Gochnatia*	111
3,7-di-Me		*Gochnatia*	111
3′-Me		*Gochnatia*	111
3,5,7,3′,4′-penta-OH			
3-G	Sen	*Senecio*	112
3,5,7,3′-tetra-OH			
3-G-3′-Me		*Senecio*	112
3,5,7,4′-tetra-OH	Tag	*Tagetes*	113
7-G		*Tagetes*	113
3,5,6,7,4′-penta-OH		*Tagetes*	113
7-G		*Tagetes*	113

Table 3. (*Continued*)

Substitution	Plant source *Tribes	Genera	Ref.
3,5,6,7,3′,4′-hexa-OH		*Tagetes*	113, 114
5,6,7,3′,4′-penta-OH			
5-Me		*Tagetes*	115

*Here and in Tables 4–6 tribes are represented by the three initial letters of their names (see Table 2).

Table 4. Dihydroflavonols in Asteraceae, reported between 1981 and 1984

Substitution	Plant source Tribes	Genera	Ref.
5,7-di-OH	Ant	*Artemisia*	13
7-Me		*Artemisia*	13
5,7,4′-tri-OH		*Chrysanthemum*	23
7-Me		*Artemisia*	13
7-G		*Chrysanthemum*	23
3,5,7,4′-tetra-OH			
7-Me		*Artemisia*	13, 36
5,7,4′-tri-OH	Ast	*Baccharis*	116
7-Me		*Baccharis*	116
3,5,7,3′,4′-penta-OH			
3-Ac		*Baccharis*	116
5,7,3′,4′-tetra-OH		*Baccharis*	116
3,5,7,4′-tetra-OH			
3-Ac*		*Baccharis*	116
5,6,7-tri-OH			
5,7-di-Me-3′,4′-OCH_2	Eup	*Ageratum*	117
5,6,7,3′,4′-penta-OH			
5,7,3′,4′-tetra-Me		*Ageratum*	117
5,7,3′-tri-Me		*Ageratum*	117
5,7-di-OH		*Chromolaena*	76
3,5,7-tri-OH		*Chromolaena*	76
3,5,6,7-tetra-OH			
6-Me		*Chromolaena*	76
5,7,4′-tri-OH		*Mikania*	118
7,4′-di-Me		*Stevia*	68
7-Me		*Mikania*	119
4-Me		*Mikania*	118
3,5,6,7,4′-penta-OH			
6,4′-di-Me		*Eupatorium*	120
6,7-di-Me		*Eupatorium*	120
5,7,3′,4′-tetra-OH			
7,4′-di-Me	Ver	*Lychnophora*	121
7,3′-di-Me		*Piptocarpha*	122
5,7,4′-tri-OH			
7-G			

the shikimate routes leading to these rings, and the modification reactions are independent [4]. For this reason the chemosystematic parameters (Table 2) refer separately to EA_{OA} and Δ_{OA} and to EA_{OB} and Δ_{OB}. The correlation of Δ_{OA} and Δ_{OB} (Fig. 2) is instructive, chiefly if compared with Wagenitz's classification based on morphological criteria [7].

Wagenitz divides the Asteraceae into Cichorioideae (tribe Lactuceae) and Asteroideae, the latter subfamily being further divided into two groups of tribes. Lactuceae

Table 5. Chalcones in Asteraceae, reported between 1981 and 1984

Substitution	Plant source Tribes	Genera	Ref.
3,4',6',tri-OH			
4'-Me-4,5-O_2CH_2	Eup	*Ageratum*	117
2',4',6'-tri-OH		*Chromolaena*	76
4',5',6',4,5-penta-OH			
4'-G	Hlt	*Clibadium*	88
		Zinnia	123
4',6',4-tri-OH			
4'-G		*Clibadium*	88
4',6',4,5-tetra-OH			
4'-G		*Clibadium*	88
2',3',4',6',4-penta-OH			
5-Geranyl		*Helichrysum*	124
2',4',6'-tri-OH			
5'-Prenyl	Mut	*Pleiotaxus*	125
4'-Me-5'-Prenyl		*Pleiotaxus*	125
2',6'-di-OH			
4'-Prenyl		*Pleiotaxus*	125

Table 6. Aurones in Asteraceae, reported between 1981 and 1984

Substitution	Plant source Tribes	Genera	Ref.
4,6,3',4'-tetra-OH 6-G	Hlt	*Clibadium*	88
6,3',4'-tri-OH 6-G	Hlt	*Zinnia*	123

is said to be closer to group 1 and Anthemideae, although placed into group 2, is considered to occupy an intermediate position between both groups. The flavonoid data are consistent with this scheme (Fig. 2). Indeed the tribes Vernonieae, Cardueae and Mutisieae, all belonging to Asteroideae group 1, as well as the subfamily Cichorioideae (Lactuceae) contain flavonoids with smaller oxidative variation of both A and B rings; while the tribes Heliantheae, Inuleae, Eupatorieae and Astereae, all belonging to Asteroideae group 2, contain flavonoids with relatively greater oxidative variation of both rings. Only the tribe Senecioneae, considered by Wagenitz to belong to Asteroideae group 2, is exceptional with respect to the oxidative variation of its flavonoids. This is not surprising. Sesquiterpene lactone data for Senecioneae are also abberrant (Fig. 1).

Jointly with Δ_O values, evolutionary advancement parameters concerning hydroxyl protection for flavonoid containing tribes (EA_P) also increase upon passing from tribes of Asteroideae group 1 to tribes of Asteroideae group 2 (Fig. 2). Under the term protection both forms of hydroxyl stabilization, *O*-glycosylation and *O*-methylation, are considered together. It is even more instructive to consider each of these protective devices separately. To this end EA_G and EA_M values are calculated for each tribe. A graphical correlation of these parameters (Fig. 3) shows a surprisingly clear negative correlation. In a general way, upon passing from tribes of Cichorioideae and Astero-

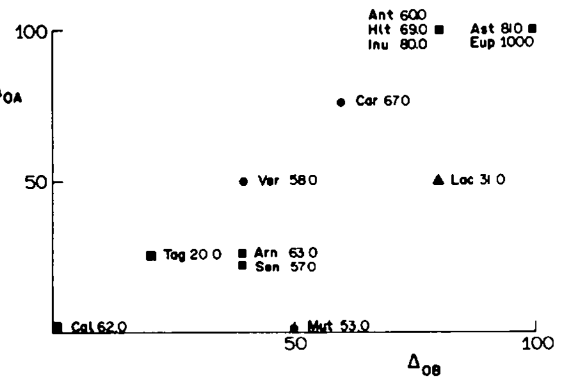

Fig. 2. Correlation of Δ_{OA} and Δ_{OB} parameters for flavonoids from tribes of Asteraceae (for meaning of symbols see Fig. 1). Also given are EA_P values.

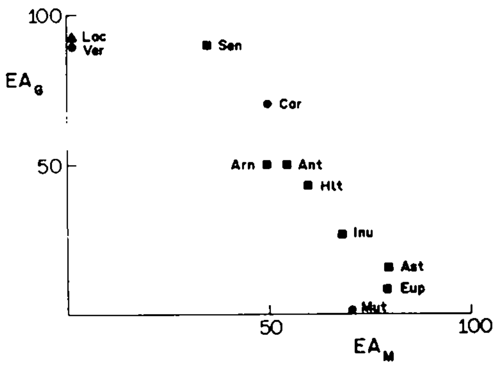

Fig. 3. Correlation of EA_G and EA_M parameters for flavonoids from tribes of Asteraceae (for meaning of symbols see Fig. 1).

ideae group 1 to tribes of Asteroideae group 2, *O*-glycosylation diminishes in favour of *O*-methylation. Again Senecioneae show an aberrant result, producing more *O*-glycosylflavonoids and less *O*-methylflavonoids than expected for a tribe of Asteroideae group 2.

Upon superimposing, for tribes of Asteraceae, flavonoid data (EA_G/EA_M) on the sesquiterpene lactone EA_O/EA_S correlation (Fig. 1) some striking regularities are observed. Thus Cichorioideae ($EA_G/EA_M = 94/0$) are also, with respect to flavonoid protection, close to Vernonieae (97/2), a tribe of Asteroideae group 1 in which *O*-glycosylflavonoids predominate vastly over *O*-methylflavonoids. From Vernonieae onwards along gradient 1 a decrease in *O*-glycosylation and an increase in *O*-methylation of flavonoids is observed: Cardueae (71/5), Mutisieae (0/70). To our knowledge no flavonoids have yet been reported from Liabeae and Arctoteae. An analogous decrease of *O*-glycosylflavonoids vs increase of *O*-methylflavonoids is observed for the tribes of gradient 2: Anthemideae (49/54), Heliantheae (44/59), Arnicineae (20/80), Inuleae (28/68) and Astereae (15/79). Again considering EA_G-/EA_M values, Eupatorieae (8/80) should follow Astereae. This is a relevant fact since Eupatorieae have been placed by Wagenitz into Asteroideae group 2. On sesquiterpene lactone EA_S (skeletal specialization based) data Eupatorieae are indeed closely related to Inuleae and Astereae of gradient 1. On sesquiterpene lactone EA_O (oxiation value based) data Eupatorieae are closely related to Vernonieae of gradient 1.

DISCUSSION

Plant antiherbivore defence is only favoured by natural selection when the cost of production is less than the benefit of enhanced protection [8]. If glycosylation of phenolic hydroxyls involves more energy commitment than methylation, a rationale exists for the switch from glycosylation to methylation. The observation, reported for *Parthenium* species (Asteraceae) [9], is consistent with previous opinions on the nature of primitive and advanced flavonoid characters, such as glycosylation and *O*-methylation respectively [5, 6]. The frequently mentioned ubiquity in the plant kingdom, ease and speed of identification and wealth of data would make characterization of plant taxa along the lines indicated in the present paper a valuable chemosystematic tool. Especially EA_G/EA_M ratios may well serve as the long sought for yardstick for the determination of evolutionary polarity of other classes of micromolecular markers besides sesquiterpene lactones in tribes of Asteraceae.

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