Towards a scientific status for micromolecular Systematics

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Abstract

While different biosynthetic groups of secondary metabolites (micromolecules) rarely accumulate in the same plant species, one such group may replace another in morphologically related taxa. The use of micromolecules as general systematic markers of the plant kingdom thus requires unifying postulates concerning their evolution patterns. Two such postulates — contraction of the shikimate pathway and blocking of oxidative enzymes — are illustrated with the aid of systematic considerations on the genera **Aniba** (Lauraceae) and **Derris-Lonchocarpus** (Leguminosae) which involve besides chemistry, morphology, ecology and geography. Extrapolation of the principles applied in these examples to the entire plant kingdom seems possible, an important fact, due to the ecological implications of micromolecules. In this sense, the paper opens the way, rather than simply to a more "natural" classification, to an information retrieval device of ecologically relevant facts about plants.

Introduction

The evolutionary classification of plants is endowed with predictive value (Cronquist, 1968), i.e. a better than random chance exists for information on certain characters of as yet unstudied taxa to fall into the pattern which has been established for these characters by the study of a relatively limited number of taxa. The closer the evolutionary relationship of the taxa, the higher the chance for reasonable predictions. So far, so good; unless you have recognized the x-part of the equation I just mounted, namely the pattern-concept. It is, of course, not possible to extrapolate experimentally determined chemical characters until the patterns of chemical evolution are announced, until the chemical phenomena which accompany development of lineages become known.

We know by now about patterns of macromolecular evolution and appreciate that nucleic acid and protein (cytochrome c, ferredoxin, plastocyanin, hemoglobin) data (Smith, 1976) are the best, if not sole, unifying themes bridging the diversity of organisms. But can micromolecules be used in a similar way? The answer to this question is *no* if we think of each biogenetic group of metabolites in isolation. To quote only examples from our own work, benzylisoquinolines (Rezende *et al.*, 1975), indoles (Cagnin *et al.*, 1977), quinolizidines (Salatino & Gottlieb, 1980), quinolones (Gottlieb *et al.*, 1980) and a few other classes of alkaloids (Gomes & Gottlieb, 1980), as well as coumarins (Gottlieb *et al.*, 1980), flavonoids (Cagnin & Gottlieb, 1978; Gomes *et al.*, 1980), iridoids (Kaplan *et al.*, 1980), xanthones (Rezende & Gottlieb, 1973; Kubitzki *et al.*, 1978) are usually employed as systematic markers for particular plant taxa. It has been established, however, that different groups rarely accumulate, i.e. are subject to structural variation, in the same species (Gottlieb, 1980) and that, indeed, one group may replace another in morphologically related taxa (Gottlieb & Kubitzki, 1980a). Let us accept this as evidence that these groups perform anologous functions, say defence, attraction of pollinators, and set out to find a common characteristic capable of measuring evolutionary advancement of a metabolite irrespective of the biogenetic group to which the metabolite belongs. This means, of course, that we will have to shift emphasis from the structure of accumulated molecules to alterations of biosynthetic pathways. Although this requirement has been clearly stated years ago (Birch, 1963), its recognition was not im-

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mediately helpful. No procedures existed to assess the significance of such alterations in connection with plant evolution (Birch, 1973).

The natural products chemist knows of only one way to deduce biosynthetic correlations: inspect the structure of micromolecules and analyse their common features in relation to natural occurrence. In doing this we stumbled upon several conspicuous trends of micromolecular evolution. Interpreted in terms of basic principles they may prove of value in elevating micromolecular systematics from an old art into a scientific discipline.

The first principle (Fig. 1, top) states that evolution of the primary precursors (from which biogenetic groups of secondary metabolites are derived) proceeds by blocking of reaction steps. Such blocking leads to new chemical lines. Within each line evolution of the metabolites belonging to biogenetic groups proceeds by diversification ([1]). The second principle (Fig. 1, bottom) states that evolution of micromolecules proceeds by oxidation. The relatively highly oxidized compounds characterize new chemical lines. Within each line evolution proceeds by deoxygenation.

In the following sections, the relevance of these principles is discussed with the aid of two examples, the systematics of *Aniba* and of *Derris-Lonchocarpus*, genera of our south America flora. We have commented on these topics in papers (resp. Gottlieb & Kubitski, 1980a, b; Gomes *et al.*, 1980) which should be consulted inclusively for references to the voluminous literature on the chemical composition of species.

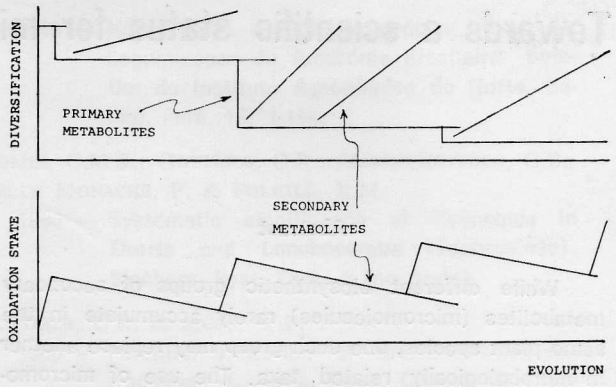


Fig. 1 — Basic concepts of micromolecular evolution.

Micromolecular evolution in Aniba

The chemistry of 18 out of the 41 recognized *Aniba* species (family Lauraceae) (Kubitzki, 1980) is fairly well known. In essence, their trunk wood contains propenylphenols (1) and allylphenols (2), precursors to neolignans belonging to several constitutional and configurational types which may be biosynthetically simple (3 — 6) or derived (7 — 9) (Fig. 2); pyrones, again biosynthetically simple (10, 12) or derived (11, 13), and benzophenones (14) (Fig. 3); benzyl benzoates (15), benzylisoquinoline alkaloids (16) and sporadically flavonoids, a queer alkaloid (17) and linalool (18) (Fig. 4). With the exception of this monoterpene, all these biogenetic groups of micromolecules stem from a primary biosynthetic pathway linked to shikimic acid (Fig. 5). The benzylisoquinolines and the benzyl benzoates are not only ubiquitous, but also practically invariant in all species, and hence of value only in revealing ancestry, i.e. in situating the genus within the family Lauraceae, a topic which was discussed elsewhere (Gottlieb, 1972, 1980; Ferreira *et al.*, 1980). Others, however, such as the neolignans, the pyrones and the benzophenones are restricted to particular species listed in Table 1 in groups according to chemical composition. A neat dichotomy is seen to exist in *Aniba* between neolignan *vs.* pyrone containing species. Not once have both types of compounds yet been reported for a single species.

In my experience such switch-overs are reasonably general phenomena and chemists who tried to classify taxons by presence/absence of chemical characters have often seen their results under fire from morphologists (cf. e.g. Mabry, 1974). This is where our principles of micromolecular systematics come in. According to the first principle, blocking of reaction steps in the shikimate

([1]) — This corollary of the first principle was formerly designated second principle (Gottlieb, 1980).

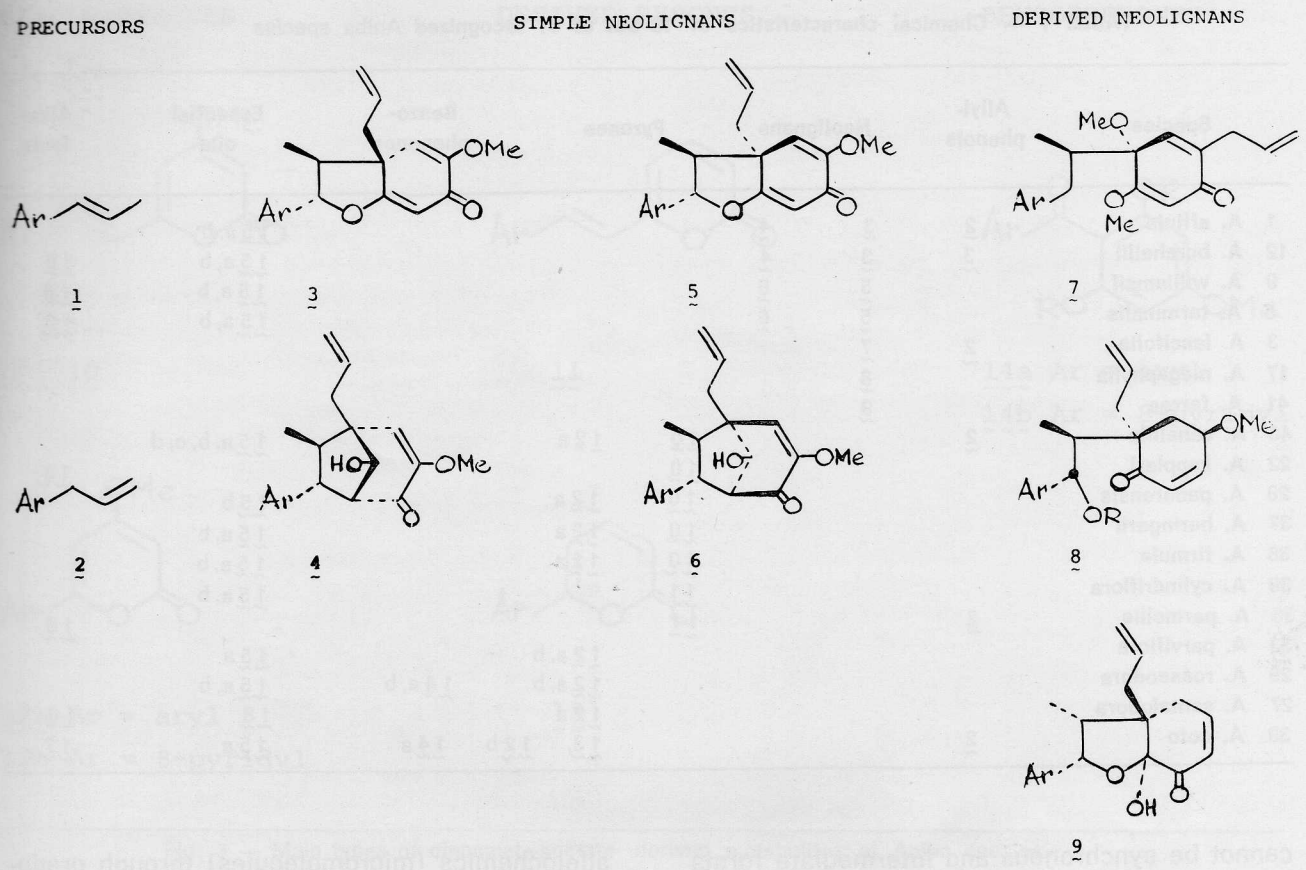


Fig. 2 — Main types of neolignans of **Aniba** species

pathway would imply the use of cinnamates rather than of propenylphenols as biosynthetic starting materials. Application of the principle to the case of *Aniba* (Fig. 5) thus suggests the evolution of the propenylphenol derived neolignans into the cinnamate derived pyrones, benzophenones and flavonoids, and assigns a more primitive position in the evolutionary history of the genus to the neolignan than to the pyrone containing species.

According to the corollary of the first principle, evolution of micromolecules in a biogenetic group involves substitutional, configurational and/or skeletal diversification Replacement of the neolignans of types 3 — 6 by 7 — 9, as well as of the pyrones of types 10 by 11 and of types 12a by 13 or addition of anibine (12b) and benzophenones (14), requires skeletal diversification through expansion of reaction pathways. Application of the corollary to the case of *Aniba* thus warrants the listing of the species within each of the three groups, the neolignan and the two pyrone ones, in order of evolutionary advance as shown in Table 1.

How does this chemical classification correlate with morphology (Table 2)? Floral structure suggests a subdivision of the genus in two groups according to the opening modes of the valves of the anther cells of the stamens. The structure of the lower leaf epidermis and wood anatomy also seem to be characters of classificatory importance.

What we have learnt so far is that the evolutions of chemical, floral, vegetative and histological characters proceed as general trends, in parallel. Since the differentiation of these characters is largely independent, it

TABLE 1 — Chemical characteristics of 18 out of 41 recognized Aniba species

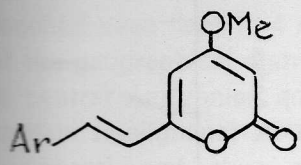
	Species	Allyl-phenols	Neolignans		Pyrones		Benzo-phenones	Essential oils	Alka-loids
1	**A. affinis**	2	3	4				15a, b	
12	**A. burchellii**	3	3	4				15a, b	16
9	**A. williamsii**		5	6				15a, b	16
5	**A. terminalis**		5	6				15a, b	16
3	**A. lancifolia**	2	7						
17	**A. megaphylla**		8						
41	**A. ferrea**		9						
40	**A. canelilla**	2			10	12a		15a, b, c, d	
22	**A. kappleri**				10				16
28	**A. panurensis**				10	12a		15b	
37	**A. heringerii**				10	12a		15a, b	
38	**A. firmula**				10	12a		15a, b	
39	**A. cylindriflora**				11			15a, b	
36	**A. permollis**	2			11				16
33	**A. parviflora**					12a, b		15a	
29	**A. rosaeodora**					12a, b	14a, b	15a, b	
27	**A. santalodora**					12a		18	16
39	**A. coto**	2				13, 12b	14a	15a	17

cannot be synchronous and intermediate forms with some primitive and some advanced characters must occur in all taxons. The assumption that morphology will correlate with chemistry has thus only a reasonable chance (in *Aniba* 13/18) to be correct, and it would be wise to look for other, again independent, evidence to strengthen the predictive value of the analysis.

That such evidence may be inherent to ecogeographical phytochemistry, a discipline for which we recently formulated basic tenets (Gottlieb *et al.*, 1980), can be inferred from the distribution of *Aniba* species registered in Table 2. Surely you recognize the accumulation of central Amazonian species on top of the list, the existence of extra Amazonian species towards the middle and the incidence of relatively widespread species towards the botton as an indication for the existence of general distributional trends. According to the firt tenet, plant populations possess a center of irradiation where they are endowed with a biosynthetically simple chemistry. Conquest of new regions is conditioned by the constitutional or configurational diversification of allelochemics (micromolecules) through gradual acquisition of new biosynthetic pathways. In the case of *Aniba*, central Amazonian stock appears to have spread towards the periphery of Amazonia diversifying its neolignan chemistry and into the regions beyond replacing it by pyrone chemistry. In this sense then the outward spread is postulated to have involved *A. affinis, A. burchellii* (Fig. 6) → *A. terminalis, A. williamsii* (Fig. 7) → *A. megaphylla* (Fig. 8). At some point of this evolutionary dispersal, the switch-over from neolignans to pyrones must have taken place and resulted in the development of two parallel, peripheral lines. One, with pyrones of type 10, is represented by *A. kappleri* and *A. heringerii* → *A. firmula* (Fig. 9). The other, with pyrones of type 12, is represented by *A. rosaeodora sensu* Ducke (i.e. from Guiana) and *A. coto* (Fig. 10).

According to the second tenet, the geographical development of natural products chemistry is directional. At the fringes of a species habitat, slightly different metabolites may be produced, together with the old ones, adapting the species to the new conditions. Dispersal from the fringe back into the territory occupied

SIMPLE PYRONES



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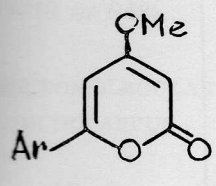
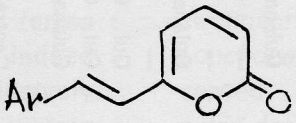


12a Ar = aryl
12b Ar = β-pyridyl

DERIVED PYRONES



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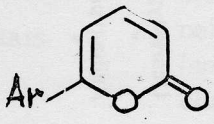

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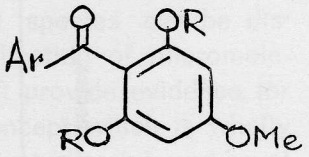
BENZOPHENONES

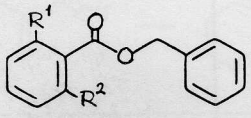


14a Ar = aryl
14b Ar = β-pyridyl

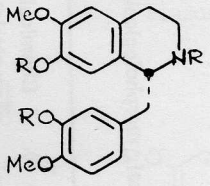
Fig. 3 — Main types of cinnamate-acetate derived metabolites of **Aniba** species

by the old and very similar species, however, is not favoured since for ecologically associated sympatric species chemical diversity is advantageous if not mandatory, and directionality of migration results. In the case of *Aniba*, chemically similar species are indeed allopatric (Figs. 6, 11). Once the chemical composition of the lineage, however, has been altered significantly (by acquisition of pyrones), possibly at or beyond the limits of the Amazon basin, reversal of the migratory direction would have been an allowed process since it leads to the co-existence of species characterized by considerably diverse chemical compositions. The neolignan-free chemistry of peripheral *Aniba* species thus made their re-entry into Amazonia possible. This was achieved in the 10-pyrone lineage by *A. panurensis* (Fig. 11) and *A. canelilla* (Fig. 12) and climaxed by the again central Amazonian *A. cylindriflora* and *A. permollis* (Fig. 13) with pyrones of type 11; and in the 12-pyrone lineage by *A. parviflora* and *A. duckei* Kosterm. (now included in *A.* rosaeodora) and climaxed in the again central Amazonian *A. santalodora* (Fig. 14).

Thus, although in 5 out of the 18 experimental cases of Table 2 (12, 17, 41, 40, 22) the



	R^1	R^2
15a	H	H
15b	OH	H
15c	OMe	H
15d	OMe	OMe



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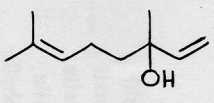

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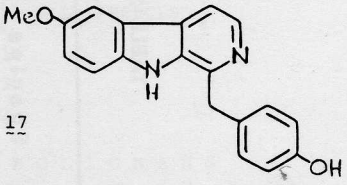

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Fig. 4 — Selected types of metabolites of **Aniba** species

TABLE 2 — Chemical, morphological, histological and geographical characteristics of 18 out of 41 recognized Aniba species.

Species	Chemical characteristics	Anther cells of outer stamens	Leaf under-surface	Wood anatomy	Geographical distribution Amazonia	Periphery
1 A. affinis	3 4	introrse-latrorse	smooth	Aniba-type	Negro	—
12 A. burchelli	3 4	"	papillose	"	Mad., Ama.	—
9 A. williamsii	5 6	"	smooth	"	Lower Negro	Guiana
5 A. terminalis	5 6	"	"	"	Sol., Mad., Ama.	—
3 A. lancifolia	7	"	"	"	Lower Negro	—
17 A. megaphylla	8	strictly introrse	"	"	Neg., Sol., Ama.	Guiana
41 A. ferrea	9	"	"	Licaria-type	Lower Negro	—
40 A. canelilla	11a 12a	"	"	"	Mad., Neg., Ama.	Gui., Orinoco
22 A. kappleri	11a	"	"	Aniba-type	—	Gui., Orinoco
28 A. panurensis	11a 12a	"	papillose	"	Mad., Sol., Negr., Tap., Ama.	Guiana
37 A. heringerii	11a 12a	"	"	"	—	Central Plat.
38 A. firmula	11a 12a	"	"	"	—	Atlantic Coast
35 A. cylindriflora	11b	"	"	"	Mad., Sol., Negr.	—
36 A. permollis	11b	"	"	"	Solimões	Guiana
33 A. parviflora	12a 13	"	"		Mad., Tap., Ama.	Guiana
29 A. rosaeodora	12a 13 15	"	"	"	Sol., Neg., Ama., Toc.	Guiana
27 A. santalodora	12a	"	"	"	Lower Negro	—
39 A. coto	12b 13 15	"	"	"	—	Andes

three morphological characters give conflicting evidence, a reasonable decision concerning the probable chemical composition can be reached even here in 4 cases by consideration of the geographical distribution (e.g. *A. ferrea* is central Amazonian and thus should indeed contain neolignans; *A. canelilla* is inclusively peripheral and thus should indeed contain pyrones). A wrong forecast would be given only for *A. megaphylla* (a significant result in view of the high specialization of its neolignan chemistry). The reliability of the chemical inventory in the *Aniba* case thus would attain 17/18 or 94%.

Micromolecular evolution in Derris-Lonchocarpus

All flavonoids which have so far been reported for species of the tribe Tephrosieae (family Leguminosae), to which the genera *Derris* and *Lonchocarpus* belong (Polhill, 1980), were classified into structural types, arranged and codified according to their biosynthetic relationship (Fig. 15). Considering the composition of the species, characterized by the number of representatives of each structural type expressed percentually in relation to the total number of flavonoids, several neatly circunscribed clusters of *Derris* (Table 3) and *Lonchocarpus* (Table 4) species can be discerned. The direct application of micromolecular data thus does not provide evidence for infra-generic unity (a concept which is wholly derived from morphological criteria), and must be even less reliable in the differentiation or amalgamation of the two genera (a controversial botanical problem, cf. e.g. Polhill, 1971, *vs.* Geesink, 1980). There is, furthermore, no clearcut correspondence between the traditional morphological subgeneric groups *(Phacelanthus* and *Lonchocarpus)* and these chemical clusters. So either secondary metabolites are poor systematic markers or we overlooked information contained in the biogenetic map (Fig. 15).

This indicates formation, from an identical cinnamatetriacetate precursor, initially of stilbenes (*1*) and chalcones (*2*). From here on the reactions are mostly oxidative processes.

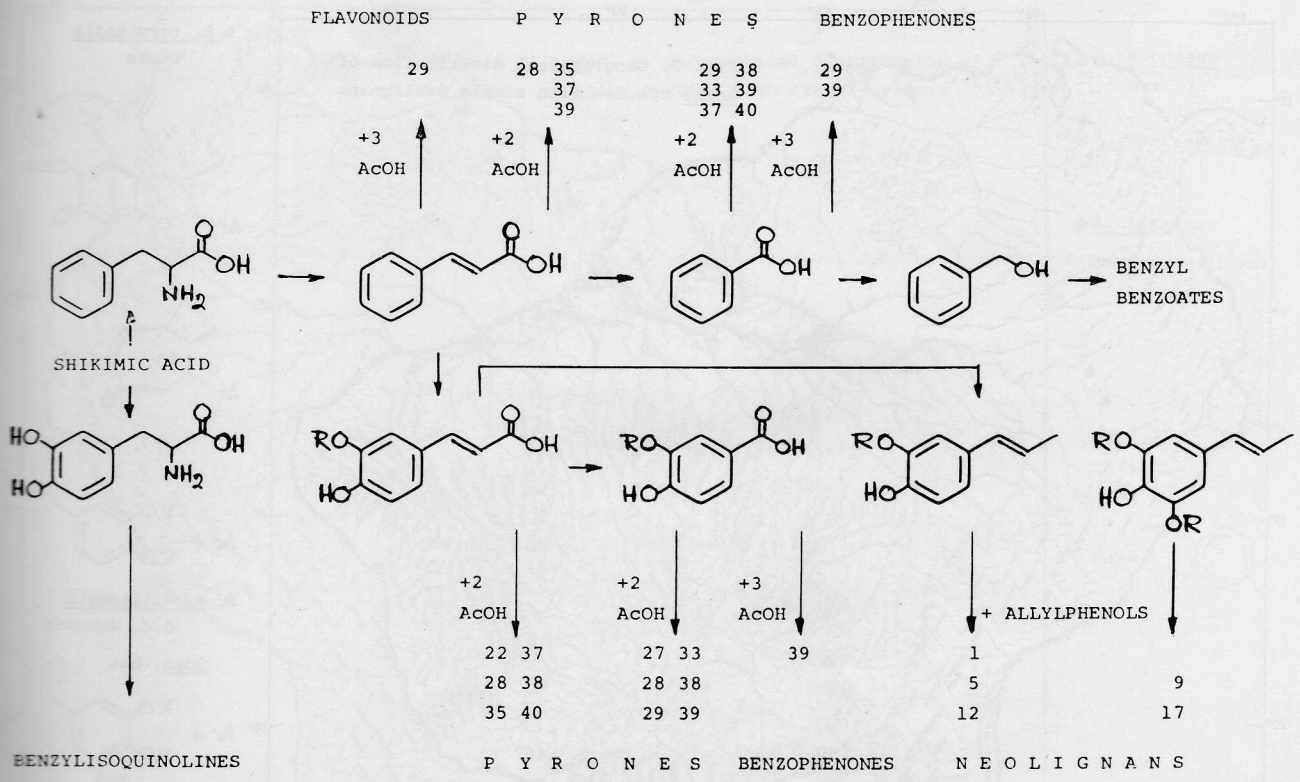

Fig. 5 — Metabolic pathways to classes of compounds of **Aniba** species indicateb by numbers (for key see Table 1)

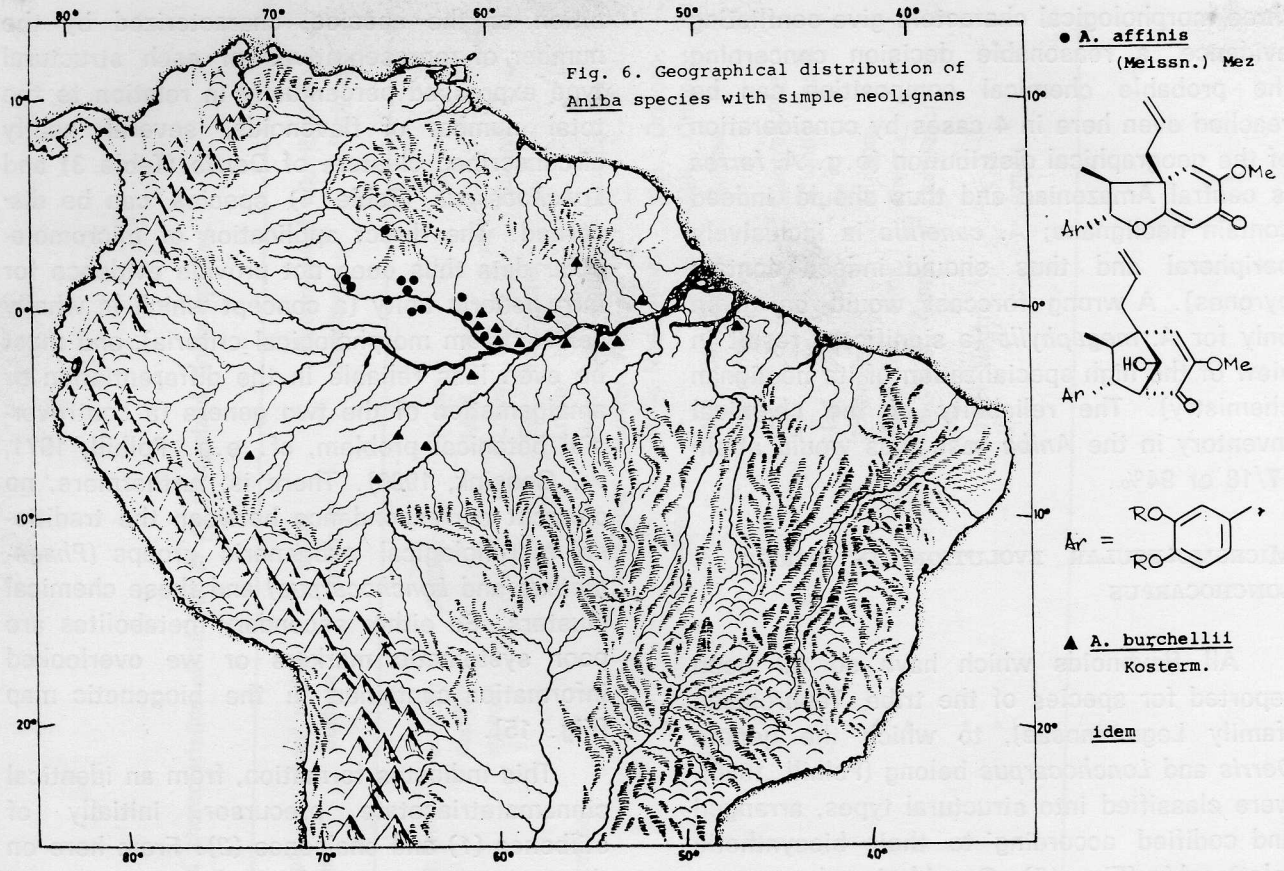

Fig. 6. Geographical distribution of Aniba species with simple neolignans

● A. affinis
(Meissn.) Mez

▲ A. burchellii
Kosterm.

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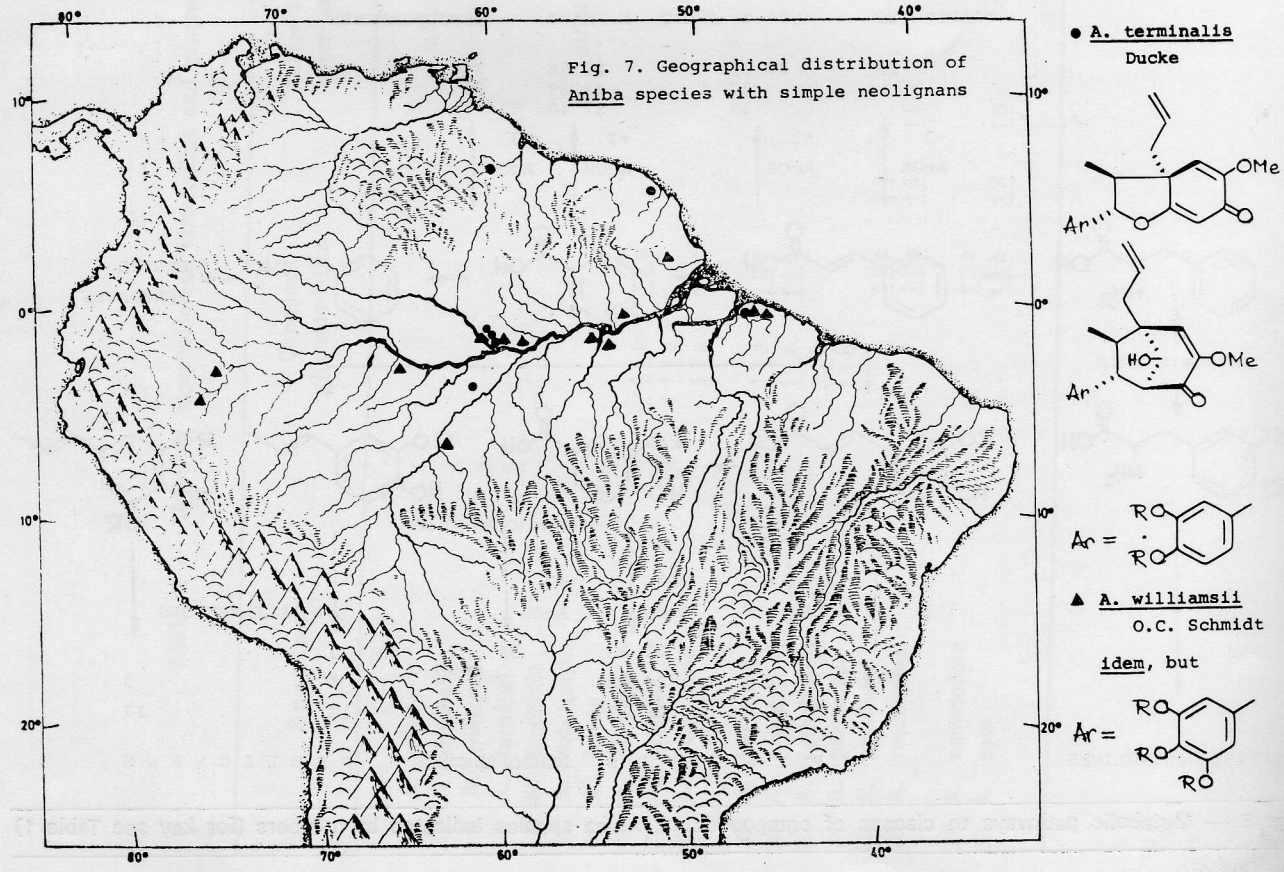

Fig. 7. Geographical distribution of Aniba species with simple neolignans

● A. terminalis
Ducke

▲ A. williamsii
O.C. Schmidt

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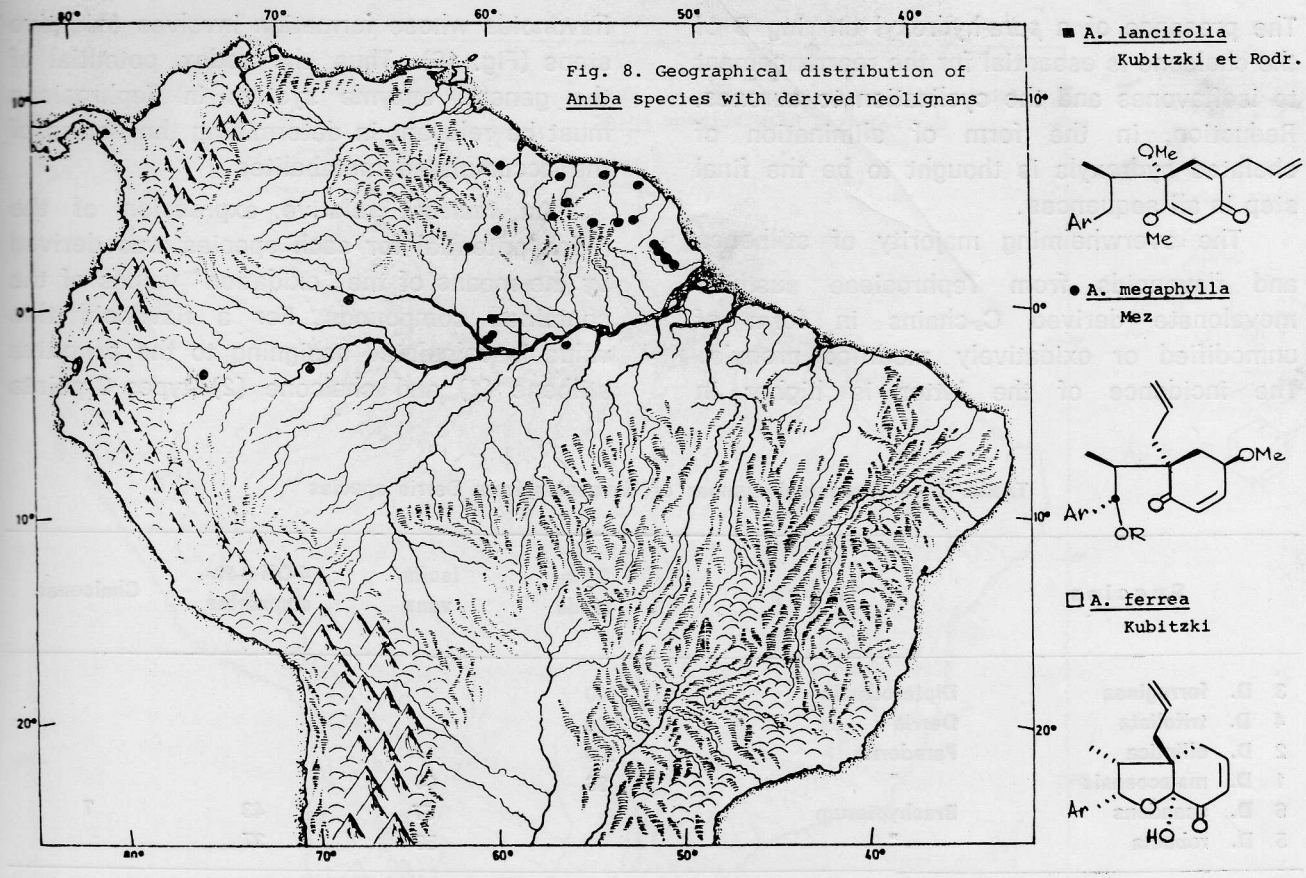


Fig. 8. Geographical distribution of Aniba species with derived neolignans

■ A. lancifolia Kubitzki et Rodr.

● A. megaphylla Mez

□ A. ferrea Kubitzki

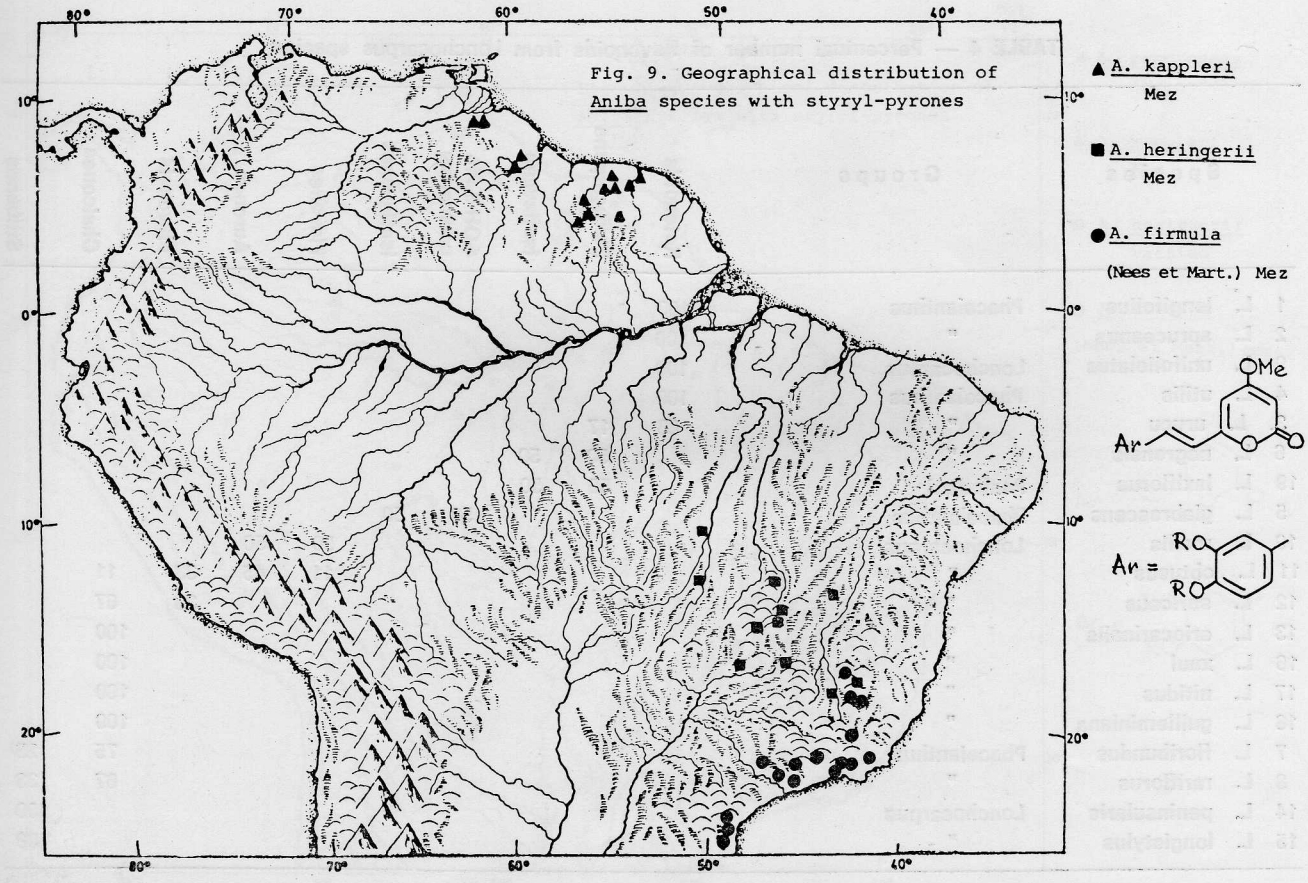


Fig. 9. Geographical distribution of Aniba species with styryl-pyrones

▲ A. kappleri Mez

■ A. heringerii Mez

● A. firmula (Nees et Mart.) Mez

The presence of a *para*-hydroxyl on ring B of the chalcone is essential for the rearrangement to isoflavones and the cyclization to aurones. Reduction, in the form of elimination of chelated hydroxyls is thought to be the final step in all sequences.

The overwhelming majority of stilbenes and flavonoids from Tephrosieae sustains mevalonate derived C_5-chains in form of unmodified or oxidatively modified prenyls. The incidence of the latter is higher in flavonoids whose formation involves oxidative steps (Fig. 16). Thus the redox potential of the general enzyme system in Tephrosieae must be relevant in determining the nature of the accumulated metabolites.

An indirect relative expression of the redox potential for each species was derived by the means of the "oxidation" values of the contained compounds. For a flavonoid this value is calculated assigning to the primitive stilbene (*1*) and chalcone (*2*) types 0 points

TABLE 3 — Percentual number of flavonoids from Derris species

	Species	Groups	Rotenoids	Isoflavans	4-OH-3-Ph-coumarins	Chalcones
3	D. ferruginea	Dipteroderris	100			
4	D. trifoliata	Derris	100			
2	D. elliptica	Paraderris	100			
1	D. malaccensis	"	86	14		
6	D. scandens	Brachypterum		57	43	7
5	D. robusta	"		56	37	

TABLE 4 — Percentual number of flavonoids from Lonchocarpus species

	Species	Groups	Rotenoids	Pterocarpans	Isoflavans	4-OH-3-Ph-coumarins	Isoflavones	Flavones	Aurones	Flavonols	Chalcones	Stilbenes
1	L. longifolius	Phacelanthus	100									
2	L. spruceanus	"	100									
9	L. unifoliolatus	Lonchocarpus	100									
4	L. utilis	Phacelanthus	100									
3.	L. urucu	"	83	17								
6	L. negrensis	"		50	50							
19	L. laxiflorus	Paniculati		50	50							
5	L. glabrescens	Phacelanthus				50	50					
10	L. mollis	Lonchocarpus						50	50			
11	L. obtusus	"						11	45	33	11	
12	L. sericeus	"								33	67	
13	L. eriocarinalis	"									100	
16	L. xuul	"									100	
17	L. nitidus	"									100	
18	L. guilleminiana	"									100	
7	L. floribundus	Phacelanthus									75	25
8	L. rariflorus	"									67	33
14	L. peninsularis	Lonchocarpus										100
15	L. longistylus	"										100

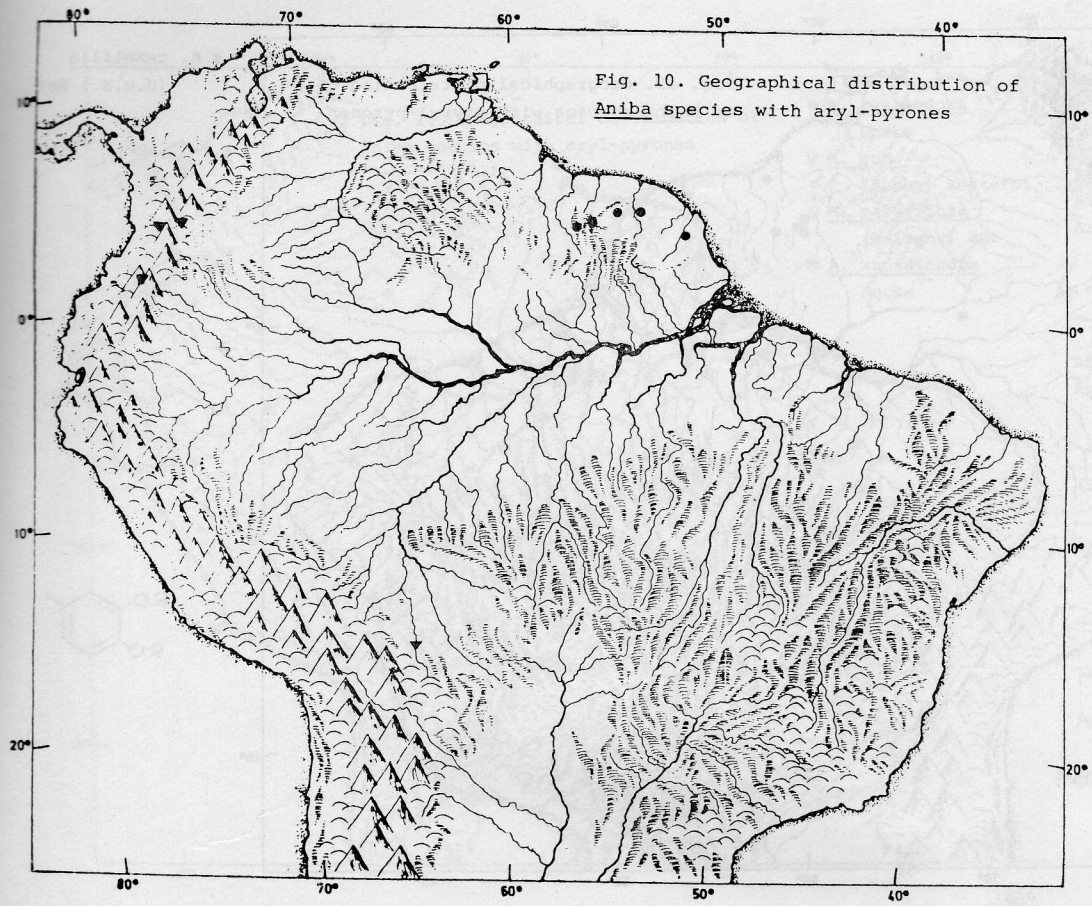
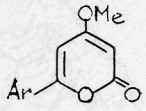


Fig. 10. Geographical distribution of Aniba species with aryl-pyrones

● A. rosaeodora Ducke

▼ A. coto (Rusby) Kosterm.



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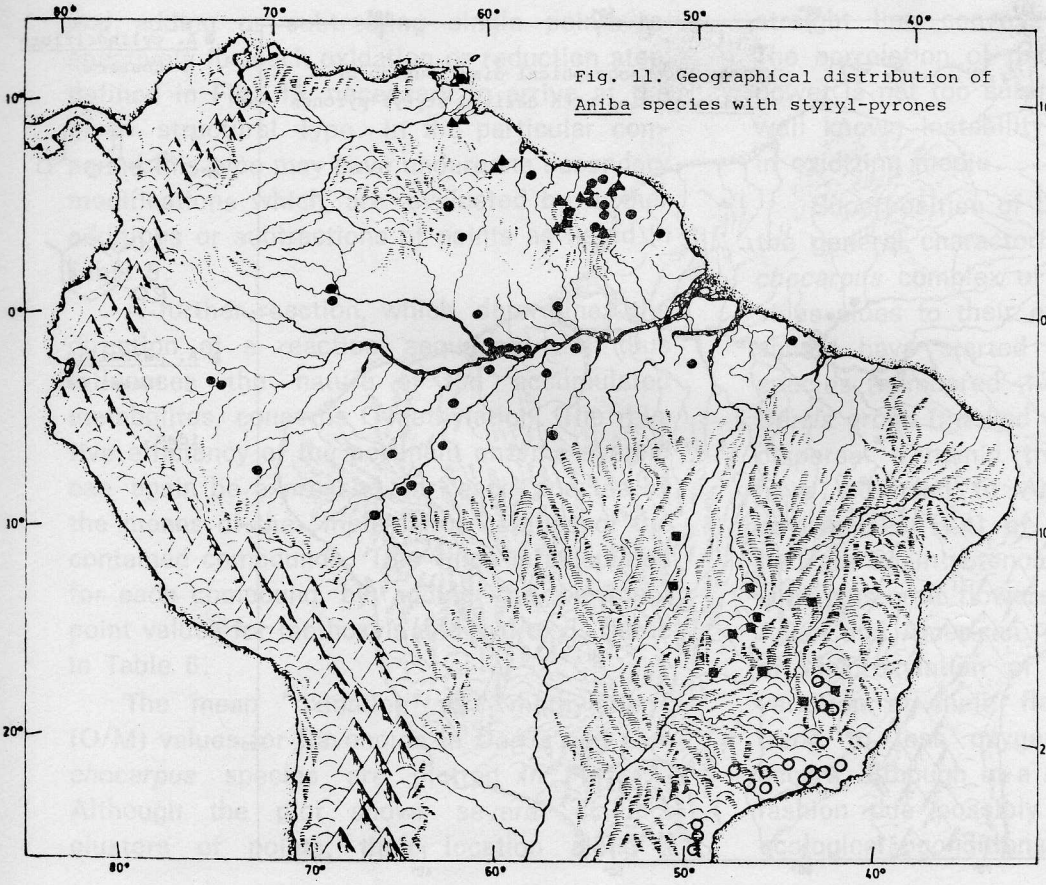


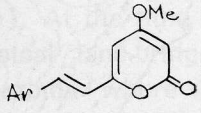
Fig. 11. Geographical distribution of Aniba species with styryl-pyrones

● A. panurensis (Meissn.) Mez

▲ A. kappleri Mez

■ A. heringerii Vattimo

○ A. firmula (Nees et Mart.) Mez



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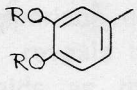


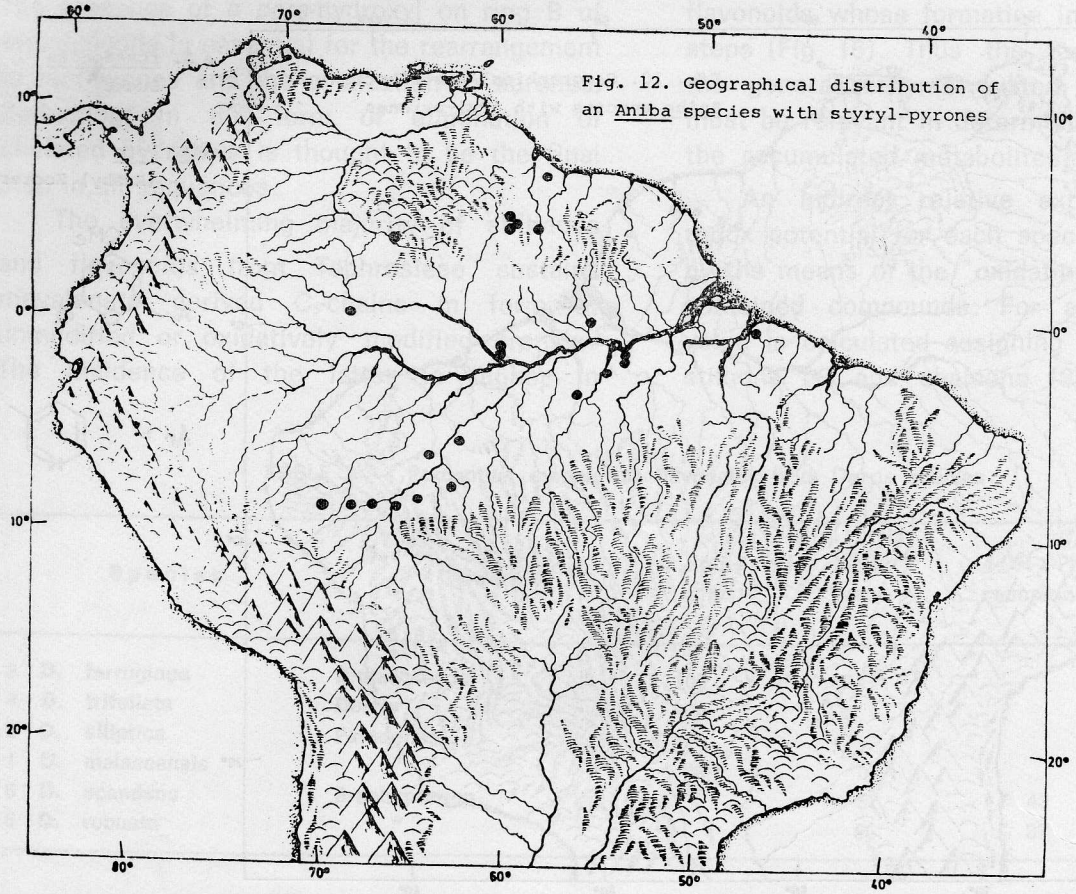
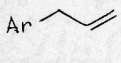
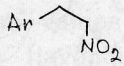
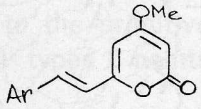


Fig. 12. Geographical distribution of an Aniba species with styryl-pyrones

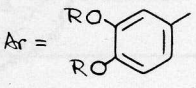
● A. canelilla
(H.B.K.) Mez







(traces)



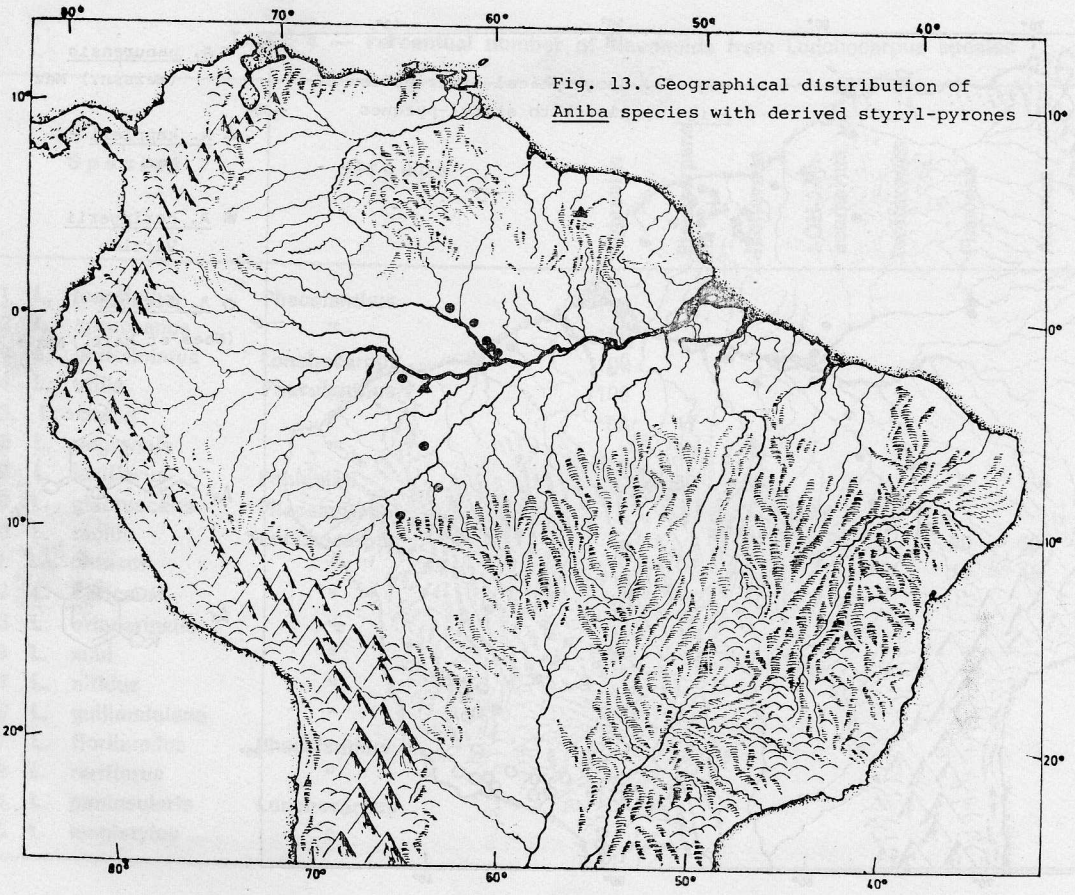
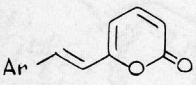
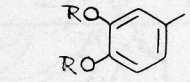


Fig. 13. Geographical distribution of Aniba species with derived styryl-pyrones

● A. cylindriflora
Kosterm.





▲ A. permollis
(Nees) Mez

idem

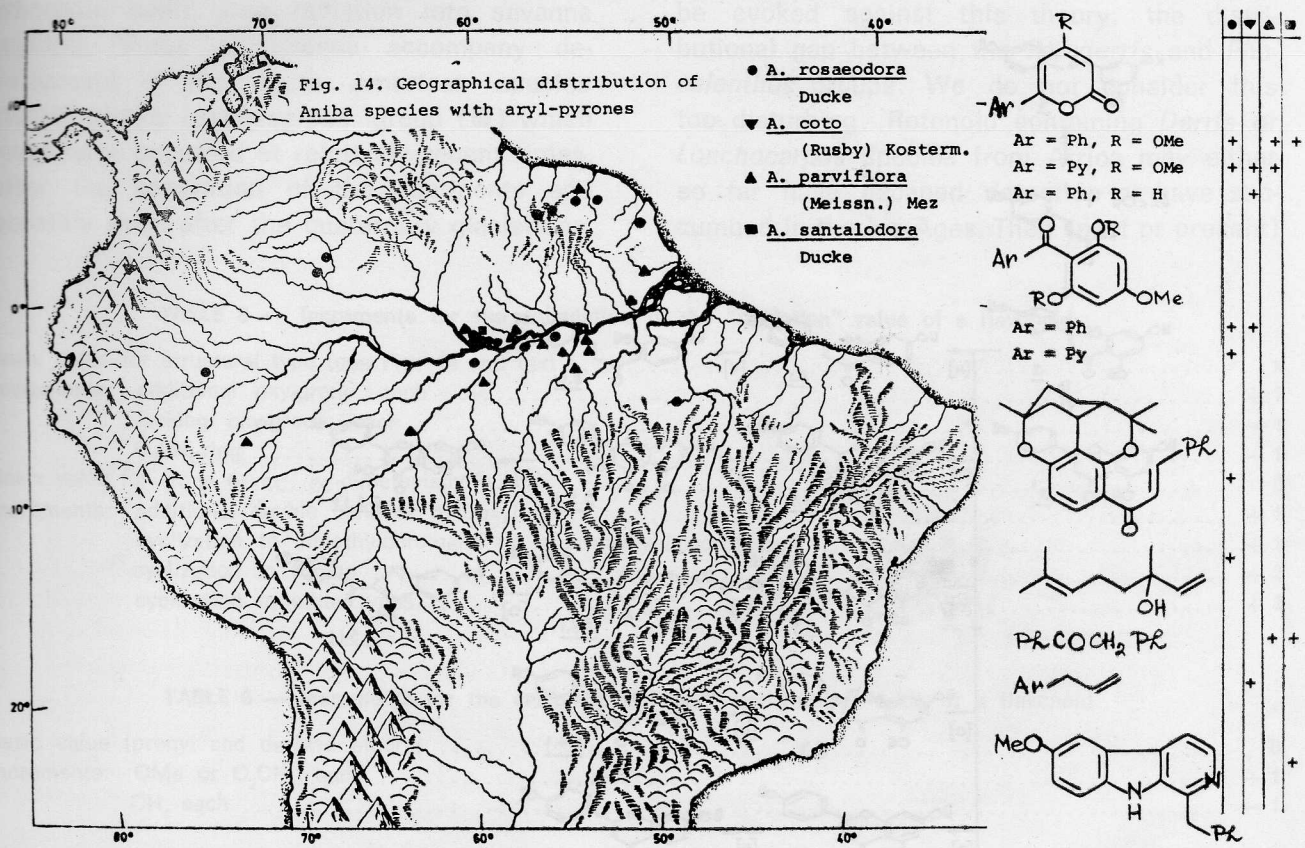


Fig. 14. Geographical distribution of Aniba species with aryl-pyrones

and adding or subtracting single points respectively for each oxidation or reduction step, defined in Fig. 15, necessary to arrive at the given structural type. In the particular compound this type may have undergone secondary modifications which are evaluated by further additions or subtractions of points as listed in Table 5.

A further reaction, which determines the direction of a reaction sequence and thus influences the nature of the accumulated metabolites, concerns O-methylation. The relative efficiency of the pertinent enzyme system can again be expressed for each species by the means of the "methylation" values of the contained compounds. This value is calculated for each compound by adding the pertinent point values for methoxyls and hydroxyls listed in Table 6.

The mean "oxidation" vs. "methylation" (O/M) values for flavonoids in *Derris* and *Lonchocarpus* species are plotted in Fig. 17. Although the plot shows several disjunct clusters of points, their location along a straight line conveys a feeling of continuity. The correlation of oxidizing and methylating power is not too surprising for reasons of the well known instability of phenolic substrates in oxidizing media.

Superposition of the chemical gradient on the general characteristics of the *Derris-Lonchocarpus* complex of species (Fig. 17) provides clues to their evolutionary history. This should have started in south-east Asia with species (numbered 1-4) associated with the *Derris* group (marked ○). At the time of their dispersal a humid tropical land-bridge must have extended to Amazonia where present day species (1-4) of the *Phacelanthus* group (●) are again rotenoid bearing forest lianes with clustered flowers. Comparison of these Asian and American groups reveals the loss of 5-hydroxylation of rotenoids as the sole variation of their flavonoid chemistry. The trend to less oxygenated metabolites also occurs, although in a much more pronounced fashion due possibly to dramatically altered ecological conditions, together with con-

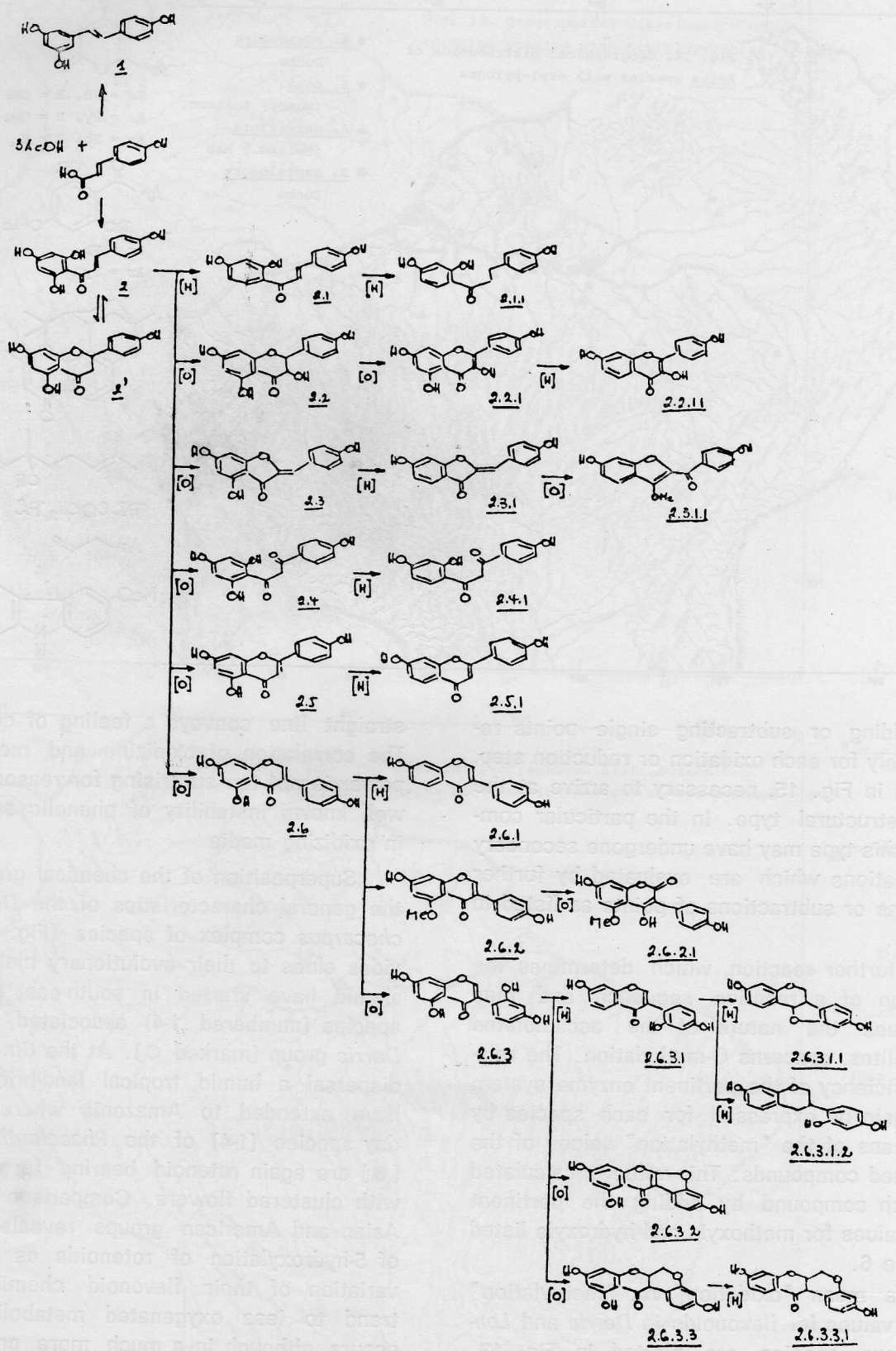


Fig. 15 — Biogenetic Classification and Codes for structural types of Flavonoids from Tephrosieae

traction of the inflorescence and change to arboreous habit, upon radiation into savanna regions. These phenomena accompany development of the south American species (10-18) of the *Lonchocarpus* group (■) which must have occurred at relatively recent times, after the separation of the continents and possibly even after the quaternary glaciations.

There is only one major fact which may be evoked against this theory: the distributional gap between the *Paraderris* and *Phacelanthus* groups. We do not consider this too disturbing. Rotenoid containing *Derris* or *Lonchocarpus* species from Africa may either so far have escaped detection or have succumbed in the Ice Ages. Their (past or present)

TABLE 5 — Increments for the calculation of the "oxidation" value of a flavonoid

Basic value for structural type (see Fig. 14 and text		x
Increments:	additional oxy-group, each	+ 1
	lacking oxy-group, each	— 1
	O_2CH_2-ring	+ 1
Basic value for γ, γ- or ∝, ∝-dimethylallyl		0
Increments:	additional double bond or hydroxyl	+ 1
	cyclization to dimethylchromene or isopropenyldihydrofurane	+ 1
	cyclization to furane	+ 2
	cyclization to other types	+ 3

TABLE 6 — Increments for the calculation of the "methylation" value of a flavonoid

Basic value (prenyl and derived ethers)		0
Increments:	OMe or O_2CH_2, each	+ 1
	OH, each	— 1

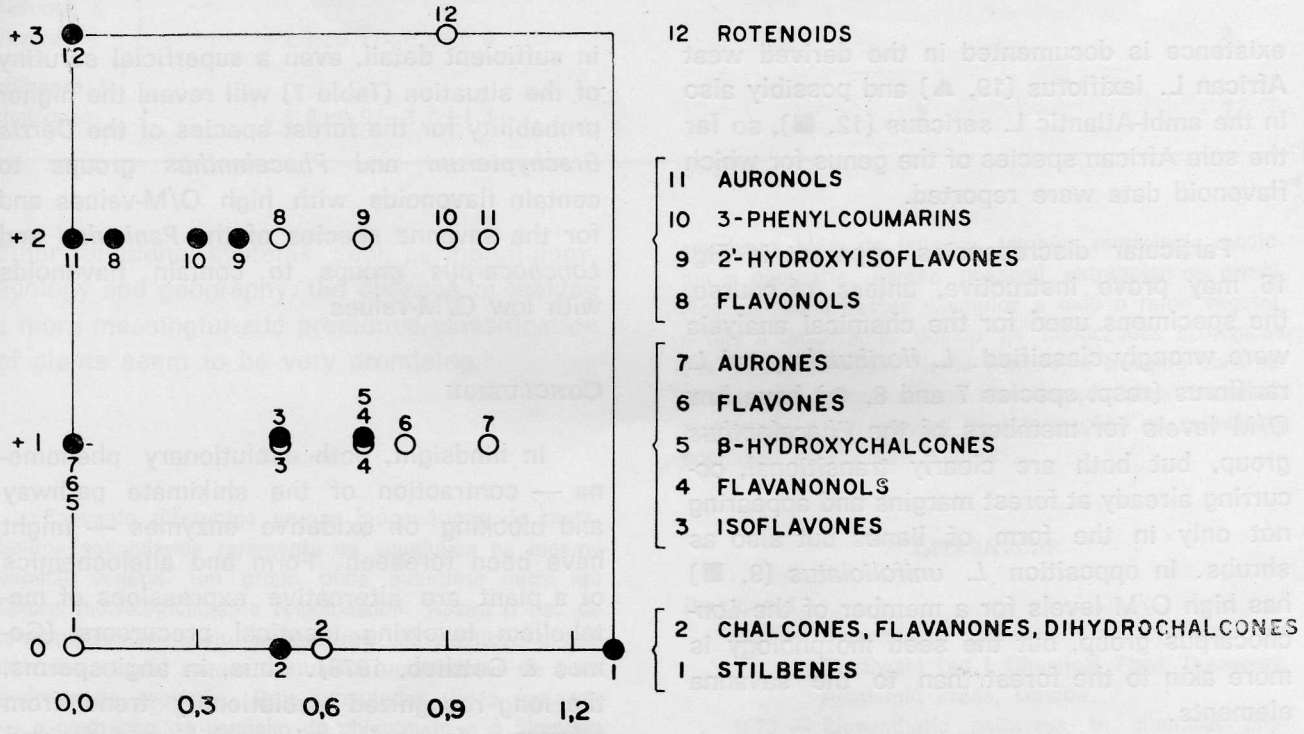
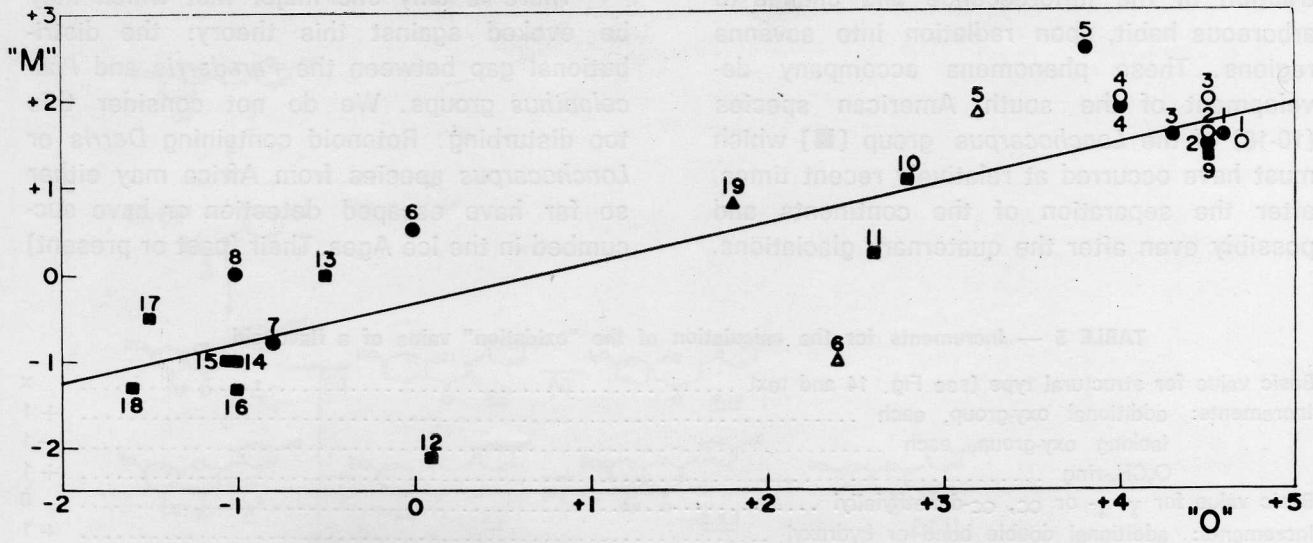


Fig. 16 — Correlation of "oxidation" values of the indicated Flavonoid skeletons (ordinate) with the number of unmodified prenyls (●) and oxidatively modified prenyls (○) per derivative (abscissa).



GENERA	GROUPS	SPECIES SYMBOLS	SPECIES NUMBERS	FLOWERS	HABIT	HABITAT	REGION
Derris	Dipterod., Parad., D.	○	1 - 4 (Table 3)	clustered	lianes	forest	Asia
"	Brachypterum	△	5, 6 "	"	"	"	"
Lonchocarpus	Phacelanthus	●	1 - 8 (Table 4)	"	"	"	America
"	Lonchocarpus	■	9 - 18 "	paired	trees	savanna	"
"	Paniculati	▲	19 "	single	"	"	Africa

Fig. 17 — Mean "oxidation"/"methylation" (O/M) values for Flavonoids from **Derris-Lonchocarpus** species.

existence is documented in the derived west African L. laxiflorus (19, ▲) and possibly also in the ambi-Atlantic L. sericeus (12, ■), so far the sole African species of the genus for which flavonoid data were reported.

Particular discrepancies apparent in Fig. 16 may prove instructive, unless, of course, the specimens used for the chemical analysis were wrongly classified. *L. floribundus* and *L.* rariflorus (resp. species 7 and 8, ●) have low O/M levels for members of the *Phacelanthus* group, but both are clearly transitional, occurring already at forest margins and appearing not only in the form of lianes but also as shrubs. In opposition *L. unifoliolatus* (9, ■) has high O/M levels for a member of the *Lonchocarpus* group, but the seed morphology is more akin to the forest than to the savanna elements.

As in the *Aniba* case, although thus all facts may be understandable only if examined in sufficient detail, even a superficial scrutiny of the situation (Table 7) will reveal the higher probability for the forest species of the *Derris Brachypterum* and *Phacelanthus* groups to contain flavonoids with high O/M-values and for the savanna species of the *Paniculati* and *Lonchocarpus* groups to contain flavonoids with low O/M-values.

Conclusion

In hindsight, both evolutionary phenomena — contraction of the shikimate pathway and blocking of oxidative enzymes — might have been foreseen. Form and allelochemics of a plant are alternative expressions of metabolism involving identical precursors (Gomes & Gottlieb, 1978). Thus, in angiosperms, the long recognized evolutionary trend from arboreous forms (based on shikimate derived lignin) to herbaceous forms should indeed be accompanied by the gradual replacement of

shikimate by acetate or mevalonate derived allelochemics. Should, furthermore, an increase in oxygen uptake be required to power the energy producing process which introduces the modifications associated with the development of a new lineage? If so, once the lineage is established, blocking of oxygen--transferring enzymes is to be expected for reasons of economy and concomitant activation of other, inherited enzyme systems should take place.

Whatever the acceptability of such interpretations, the principles point the way to a unified evolutionary scheme of secondary metabolites on which to base the appraisement of trends in plant evolution. If these trends were to be considered with changes of other functional systems, such as morphology ecology and geography, the chances of making a more meaningful and predictive classification of plants seem to be very promising.

TABLE 7 — Number of species of known flavonoid composition belonging to the Derris, Dipteroderris & Paraderris **(DER),** Brachypterum **(BRA),** Phacelanthus **(PHA),** Paniculati **(PHAN) and** Lonchocarpus **(LON) groups.**

Flavonoid composition	Variation of O/M	Number of species				
		Derris		Lonchocarpus		
		DER	BRA	PHA	PAN	LON
rotenoids	+4.4±0.4/+1.8±0.3	4		5		1
4-OH-3-phenyl coumarins	+3.1±0.7/+0.9±1.9		2	1		
pterocarpans & isoflavans	+0.9±0.9/+0.6±0.2			1	1	
flavones & flavonols	+2.7±0.1/+0.8±0.3					2
stilbenes & chalcones	+0.8±0.9/—1.0±1.1			2		7

Resumo

Enquanto diferentes grupos biogenéticos de metabolitos secundários raramente se acumulam na mesma espécie vegetal, um grupo pode substituir outro em taxons morfologicamente relacionados. Assim o uso de micromoléculas como marcadores sistemáticos gerais do reino vegetal requer postulados unificadores de seus padrões de evolução. Dois postulados desta natureza — a contração do caminho do chiquimato e o bloqueio de enzimas oxidativas — são ilustrados com ajuda de considerações sistemáticas acerca dos generos **Aniba** (Lauraceae) e **Derris-Lonchocarpus** (Leguminosae) que envolvem, além de química, também morfologia, ecologia e geografia. Parece possível extrapolar os princípios aplicados nestes exemplos a todo o reino vegetal, o que é importante, devido às implicações ecológicas das micromoléculas. Neste sentido, o trabalho abre caminho, não apenas a uma classificação mais "natural", mas ainda a um arquivo de informações de relevância ecológica acerca das plantas.

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