Chemotaxonomy of Flowering Plants

Volume II

R. Darnley Gibbs

CHEMOTAXONOMY
OF
FLOWERING PLANTS

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JUNE, 1974 NOTES, QUERIES AND ERRATA

Professor G. Ourisson has very kindly pointed out two errors in the figure included in the promotional leaflet for this book. I am very grateful to him since his action has prompted me to recheck virtually all the formulae in the 189 figures of the text. This hasty check has revealed what is to me a shocking number of errors (mostly of a minor nature), as well as many cases in which I am unsure of the formulae. I have therefore prepared the following list, which I hope is substantially complete.

It is but a slight consolation to report the detection of several errors in the reference books available to me, and to note that *chemists* (I am a *botanist*) seem as liable to get plant names wrong as I am to err in my formulae.

Fig. 146 (p. 686) *Opuntiol* —CH_2OH, not —OH_2OH

Fig. 149 (p. 694) *Jacareubin* Should have —OH at 1. *Maclura-xanthone* should have —OH at 1. Have I the correct chain at top? *Mangiferin* =O missing

Fig. 150 (p. 698) *Embelin* Add —OH at 5. Plastoquinone, not plastaquinone

Fig. 154 (p. 711) *Cryptotanshinone* Should have 2 —CH_3 groups at top of left-hand ring?

Fig. 159 (p. 723) *Cycloartenol* Should have 2 —CH_3 groups (not 1) at bottom left (as in *butyrospermol*)? *Elemolic acid* —COOH, not —CH_3, at α

Fig. 161 (p. 745) Oleandrigenin, not Oldeandrigenin. *Periplogenin* —CH_3 omitted (compare other genins)

Fig. 162 (p. 747) *Digacetigenin* I find a formula somewhat different from mine

Fig. 166 (p. 766) *Sinalbin* Is the non-sinapine part of my formula correct?

Fig. 169 (p. 790) *1,8-Cineole* —O— should link to 8 position (not 4)

Fig. 173 (p. 797) *Mansonone-A* Should there be a second double bond (common to the rings)?

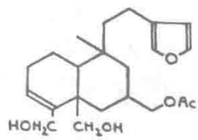
Fig. 174 (p. 798) *Drimenol* —CH_3 missing (compare with other formulae). *Cinnamodial* Is an —OH group omitted from top of right-hand ring? *Farnesiferol-C* —CH_3 is missing from top right of left-hand ring?

Fig. 175 (p. 801) *Petasin* Double bond in ring at left?

Fig. 177 (p. 806) *Mexicanin-C* Should have $=CH_2$, not $—CH_3$, in ring at right?

Fig. 180 (p. 812) *α-Santonin* Is a double bond missing from ring at right? *Artemisin* Is an —OH missing?

Fig. 182 (p. 819) *Dodonaea-diterpene* Should this be?

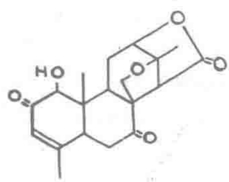


α-Camphorene Lower end should be $—CH{=}C(CH_3)_2$ (as top end)

Fig. 183 (p. 843) *Bacogenin-A* A formula proposed in 1973 transposes the $—CH_3$ and $—CH_2OH$ groups (middle level of my fig.) and has a terminal 5-membered heterocyclic ring

Panaxatriol The two $—CH_3$ groups at right should be on the C below (next to the O)

Fig. 184 (p. 856) *Samaderine-B* Should be?



Swietenine Should the chain at left be $—CH_2.CO.OCH_3$ or $—CH(OH).CO.OCH_3$?

Fig. 185 (p. 857) *Arnidiol* $=CH_2$, not $=O$, at top? *Limonin* Should double bond in ring at left be deleted? *Entandrophragmin* I am unsure of this formula. *Quassin* Is my formula correct?

MELANINS

GENERAL

The name 'melanin' has been variously used. It has been employed for any dark-brown or black substance (or mixture) occurring in plant or animal. Because there is abundant evidence that a group of substances so named would be chemically and biosynthetically unnatural, attempts have been made to use the term *melanins* only for those dark-coloured polymeric indole derivatives that involve *tyrosine*, *dihydroxy-phenylalanine* (DOPA), and *dopamine*, and *tyrosinase* and *oxygen*.

We may cite first of all the brief review by Thomas (1955), who points out that the 'melanins' of higher plants *may* include indole derivatives—though no such melanin had at that time been isolated and analysed—and other non-nitrogenous 'melanins' such as the *phytomelanes* of the *Compositae*, Japanese lac, and many oxidation products of phenols.

The true N-containing melanins, he says, may be variable. Some may contain sulfur, and some (all ?) may exist as *melanoproteins*.

The animal (true) melanins are insoluble in hot strong acids, but more or less soluble in alkalis. He gives, as a possible unit of such melanins, the dimer of fig. 144.

In 1958 Thomas has a further article on *melanin* (he uses the singular form). He says that the first real clue as to the nature of melanin came from the work of Bertrand and Bourquelot (1894–6) who noted that in *Rhus* spp., the sources of lac, there seems to exist the system:

$$\text{laccol} \xrightarrow{\text{(laccase)}} \text{lac}$$

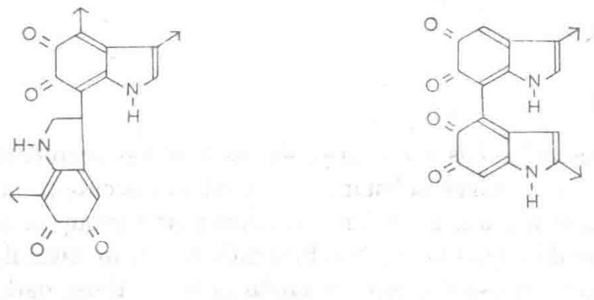
while in other plants one may have:

$$\text{Tyrosine} \xrightarrow{\text{(tyrosinase)}} \text{black pigment}$$

Thomson (1965) says that true melanins are probably produced by the legumes *Vicia faba*, *Cytisus nigricans* and *Sarothamnus scoparius*, and the banana. He gives a melanin unit (fig. 144) that differs slightly from that of Thomas.

Andrews and Pridham (1967) investigated the 'melanins' of some higher plants which have DOPA (β-(3,4-*dihydroxy-phenyl*)-L-*alanine*) or dopamine (β-(3,4-*dihydroxy-phenyl*) *ethylamine*), and compared them with enzyme-synthesized 'melanins'. They got the following results for nitrogen content, etc.

Astragalus cicer (pod), N 1·2%
Baptisia australis (pod), N 1·6%



Thomas (1955) Thomson (1965)

Fig. 144. Possible melanin units.

Lupinus polyphyllus (pod), N 1·3%; alkali fusion gave *catechol, protocatechuic acid* and 5,6-*dihydroxy-indole*.
Vicia angustifolia (pod), N 1·4%; alkali fusion as above
V. faba (fl.), N 2·1%; alkali fusion as above
V. faba (pod), N 1·3%
Musa sp. (epicarp), N 1·5%
Tyrosine-melanin (synthesized with phenolase), N 7·1%
Dopamine-melanin (synthesized with phenolase), N 6·8%

They concluded: 'The melanins from plants which contain DOPA and related compounds have been examined and shown to be largely composed of the catechol-type pigment. Some indole units also appear to be present, however.'

The book *Melanins* by Nicolaus (1968) contains little more about the *melanins* of higher plants.

The co-called *phytomelanes* of the *Compositae*, which blacken fruit-walls, and sometimes other parts of many species, seem not to contain nitrogen, or to have very little of it, and are not true *melanins*. Hegnauer (1964) lists work, largely by Hanausek, on the distribution of these substances in the tribes of the *Compositae*:

Vernonieae *absent* (5 genera)
Eupatorieae *present* in fruits of many (19 ?) genera.
Astereae *absent* (26 genera)
Inuleae rare; but *present* in *Caesulia axillaris* (frt), *Sphaeranthus* sp., *Ammobium* sp., *Inula helenium* (rhiz., rt).
Heliantheae *present* in spp. of 56 genera
Helenieae *present* in *Jaumeinae*, *Heleniinae* and *Tagetinae*; but *absent* from *Riddelliinae*.
Anthemideae *absent* from spp. of 10 genera.
Senecioneae rare; *absent* from spp. of 20 genera; but *present* in *Arnica*.

Calenduleae *absent* from spp. of 3 genera.
Arctoteae *absent* from spp. of 8 genera.
Cardueae (*Cynareae*) rare; *absent* from *Brotera*, *Carlina*, *Centaurea*, *Cirsium*, *Cynara*, *Galactites*, *Onopordon*, *Serratula*, *Silybum* (but see below), and *Xeranthemum*; but *present* in *Echinops*, *Carthamus*, and *Silybum marianum* (fruits of some).
Mutisieae rare; *present* in *Perezia* (rhiz., rt); but *not* in fruits of spp. of *Dicoma*, *Gerbera*, *Leuceria*, and *Moscharia*.
Cichorieae *absent* from spp. of 24 genera.

Much information on blackening of plants or their extracts is scattered through this book under *cigarette* and *hot-water tests*, the *aucubin-type glycosides*, *irritant plants* (*urushiol*, etc.).

NAPHTHALENE AND SOME OF ITS DERIVATIVES

GENERAL

Again we have a dilemma. *Naphthalene* itself (fig. 145) is a *hydrocarbon* and might be discussed with other members of that 'group'. Its derivatives include *aldehydes*, *alcohols*, *lactones* and *quinones*. We have sections for *naphthaquinones* (p. 699) and for *lactones* (p. 670) elsewhere. Some derivatives, such as those of *Ulmus*, are really *sesquiterpenes*, and are considered with them (p. 796).

It seems desirable to deal with *naphthalene* itself and yet other of its derivatives here.

According to Ruwet (1966) the cotyledons of *Impatiens balsamina* have a *naphtholglycosidase* which hydrolyses 1,2,4-*trihydroxy-naphthalene-4-glucoside* (fig. 145). The free 1,2,4-*trihydroxy-naphthalene* is then auto-oxidized to 2-*hydroxy*-1,4-*naphthaquinone* (*lawsone*; fig. 145). Ruvet says that the same or a similar enzyme from leaves of *Juglans regia* may be responsible for the formation of *juglone*.

List and Occurrence

4,5-Dihydroxy-2-methyl-naphthalene (fig. 145): see also *diospyrol*.
Eben. *Diospyros mollis* (frt)
4,5-Dimethoxy-6-hydroxy-2-methyl-naphthalene (Macassar-II; fig. 145)
Eben. *Diospyros celebica* (htwd—'macassar ebony')
4,5-Dimethoxy-6-hydroxy-2-naphthaldehyde
Eben. *Diospyros ebenum* (htwd)
Diospyrol (fig. 145) is a dimer of 4,5-*dihydroxy-2-methyl-naphthalene*.
Eben. *Diospyros mollis* (frt)

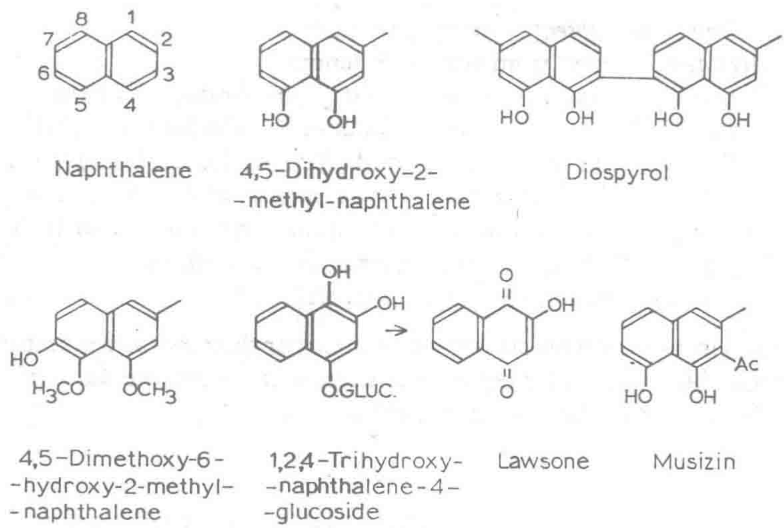


Fig. 145. Naphthalene and some derivatives.

Musizin (3-Acetyl-4,5-dihydroxy-2-methyl-naphthalene; fig. 145)

Rhamn. *Maesopsis eminii* (htwd—'musizi')

Lili. *Dianella laevis*

Naphthalene (fig. 145) is said to occur in a lichen and in

Myrt. *Eugenia caryophyllata* (cloves)

Comp. *Saussurea lappa* (rt-oil)

Irid. *Iris germanica*

Gram. *Oryza sativa* (sdlg, with *methyl-* and *p-dimethyl-naphthalene*)

1,2,4-Trihydroxy-naphthalene-4-glucoside (fig. 145) may be the precursor of *lawsone* (fig. 145, and see above).

Balsamin. *Impatiens balsamina* (cotyledons)

1,4,5-Trihydroxy-naphthalene-4-glucoside may be the precursor of *juglone* (fig. 151 and p. 70).

4,5,6-Trimethoxy-2-methyl-naphthalene (Macassar-III)

Eben. *Diospyros celebica* (htwd)

4,5,6-Trimethoxy-2-naphthaldehyde

Eben. *Diospyros ebenum* (htwd)

PYRONES

GENERAL

We may distinguish *α-pyrones*, of which *α-pyrone* (*coumalin*) itself (fig. 146) may be considered to be the 'parent'; and *γ-pyrones*, of which *γ-pyrone* (fig. 147) may be considered to be the parent.

I α-PYRONES

GENERAL

Only a few simple *α-pyrones* are known. These incluae the *kawa-pyrones* of *Piper*. They are *lactones* and some of them are named as such.

The *benzo-α-pyrones* include the *coumarins*, *isocoumarins*, *furocoumarins*, etc. They are dealt with in a separate section (p. 440).

Some *α-pyrones* are *phthalides* and some are derivatives of *naphthalene*.

List and Occurrence

Aparajitine (fig. 146) is the δ-*lactone* of *2-methyl-4-hydroxy-pentacosanoic acid*.

Legum. Clitoria maritima, ternata

Demethoxy-yangonin

Piper. Piper methysticum

Dihydro-kawain (Marindinin)

Piper. Piper methysticum (st., rt)

Dihydro-methysticin (Pseudo-methysticin)

Piper. Piper methysticum (rt)

Gentiopicroside (Erytaurine; Gentiamarin; Gentiopicrin; Sabbatin; Swertiamarin; fig. 146) has, says Paris (1963), the formula shown. It yields *mesogentiogenin* and *glucose*.

Gentian. Chlora (1), *Cicendia* (1), *Erythraea* (1), *Gentiana* (20), *Pleurogyna* (1), *Sabbatia* (at least 1), *Swertia* (3).

Hyptolide is like *massoilactone*.

Lab. Hyptis pectinata

Kawain (fig. 146)

Piper. Piper methysticum (rt)

Massoilactone (fig. 146)

Laur. Cryptocarya (*Massoia*) *aromatica* (bk-oil)

4-Methoxy-paracotoin

Laur. Aniba fragrans

Methysticin (fig. 146)

Piper. Piper methysticum (rt)

Mevalolactone (fig. 146) may be placed here. The biologically active form is *R*-(−)-*mevalolactone*.

Opuntiol (fig. 146)

Cact. Opuntia elatior

Paracotoin (fig. 146) is said to occur in Bolivian 'coto-bark' and 'Paracoto-bark'.

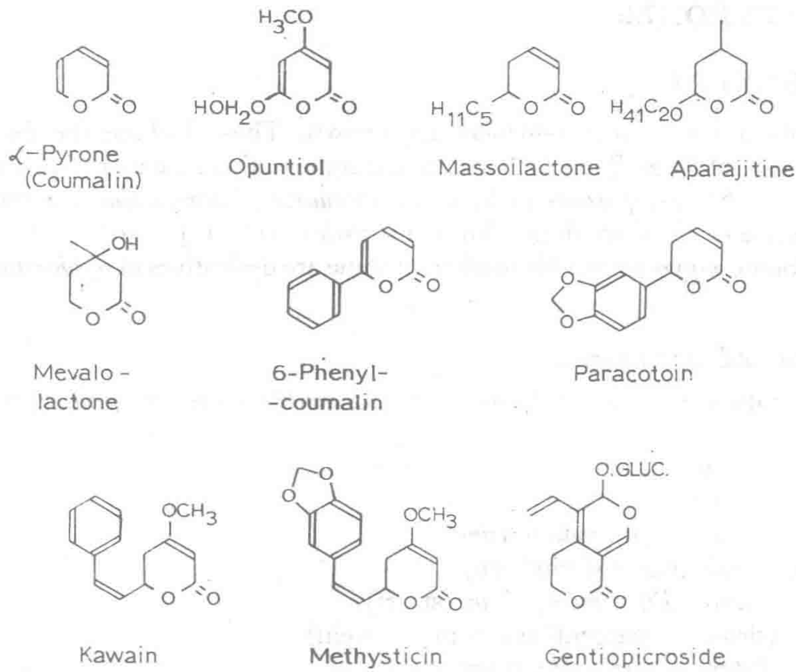


Fig. 146. Some α-pyrones.

Parasorbic acid
Ros. *Sorbus aucuparia* (unripe frt)
6-Phenyl-coumalin (fig. 146) is said to occur in 'true' coto-bark.
Yangonin
Piper. *Piper methysticum* (rt)

II γ-PYRONES

GENERAL

We may recognize three groups of *γ-pyrones*:

1. Simple γ-Pyrones.
2. Chromones, derivatives of *benzo-γ-pyrone* (fig. 148). See also *flavonoids*.
3. Xanthones, derivatives of *dibenzo-γ-pyrone* (*xanthone*; fig. 149).

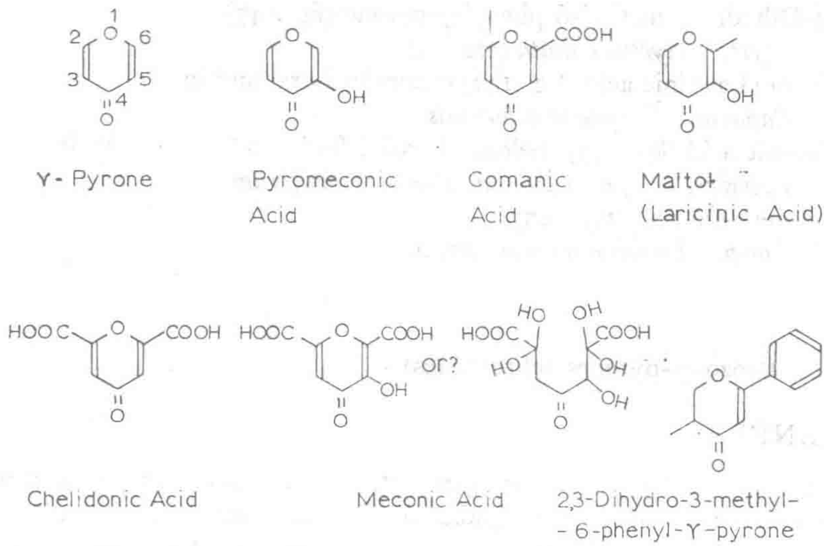


Fig. 147. Simple γ-pyrones.

II.1 Simple γ-Pyrones

GENERAL

Only a few simple *γ-pyrones* seem to have been recorded from higher plants. One of them—*chelidonic acid*—is, however, very widely distributed.

List and Occurrence

Chelidonic acid (fig. 147) was found in more than half of the more than 1100 species examined by Ramstad (1953). Kwasniewsky (1953) also lists many plants as containing the acid. We list only a few examples to show how widely spread it is.

Papaver. *Chelidonium majus*, *Stylophorum diphyllum* (rt, bound to an *alkaloid*)
Berberid. *Berberis vulgaris*
Campanul. *Lobelia inflata* and other spp.
Rubi. *Uragoga* (*Cephaelis*)
Lili. *Asparagus* (plt, frt), *Colchicum*, *Convallaria* (lvs), *Gloriosa* (lvs), *Polygonatum* (2), *Schoenocaulon* (sd), *Veratrum* (2)

Comanic acid (fig. 147)
Occurrence?

2,3-Dihydro-3-methyl-6-phenyl-γ-pyrone (fig. 147)
Myrt. *Myrtus bullata* (ess. oil)
Maltol (Laricinic acid; fig. 147) occurs in *Larix* and in
Papaver. *Corydalis ochotensis*
Meconic acid (fig. 147): belongs here? I find two formulae for it.
Papaver. *Papaver dubium, rhoeas, somniferum*
Pyromeconic acid (fig. 147)
Comp. *Erigeron annuus* (lvs, fl.)

II.2 Benzo-γ-pyrones (chromones)

GENERAL

Many of the *benzo-γ-pyrones* have other rings too. Thus some *furo-chromones* are known. Other substances which might be included here, such as *deguelin*, *elliptone*, and *rotenone*, are essentially *isoflavanone* derivatives and are treated with them (p. 609). Some of the other groups of *flavonoids* are also *chromone* or near-chromone derivatives.

The occurrence of a unique group of *benzo-γ-pyrones* in *Ptaeroxylon* and *Cedrelopsis* is of interest. These genera are removed from the *Meliaceae* by some taxonomists and placed in a little family *Ptaeroxyl-aceae* (q.v.). I know of no *benzo-γ-pyrone* in other members of the *Meliaceae*.

List and Occurrence

Ammiol (2-Oxymethyl-5,8-dimethoxy-furo-4′,5′,6,7-chromone)
Umbell. *Ammi visnaga* (sd)
Angustifolionol (fig. 148)
Myrt. *Backhousia angustifolia* (ess. oil)
Chellol (fig. 148) is the aglycone of *khellinin* (*chellol-2-glucoside*). Does it occur free?
Eugenin (fig. 148)
Myrt. *Eugenia aromatica, caryophyllata* ('cloves')
Eugenitin (6-Methyl-eugenin)
Myrt. *Eugenia caryophyllata*
Heteropeucenin (fig. 148)
Meli. *Ptaeroxylon obliquum* (htwd)
Heteropeucenin-dimethyl ether
Meli. *Ptaeroxylum obliquum*
Heteropeucenin-7-methyl ether
Meli. *Ptaeroxylon obliquum*

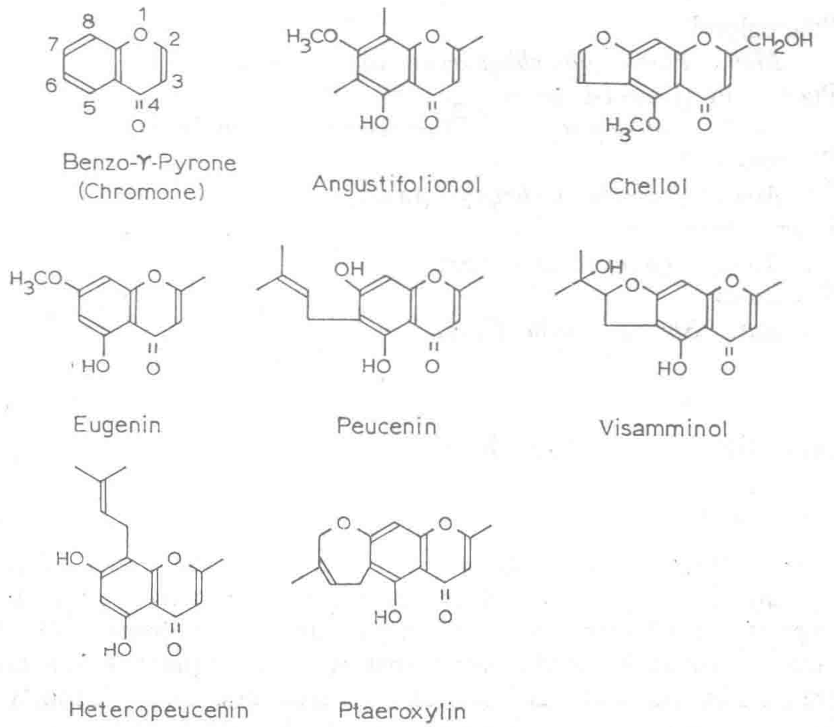


Fig. 148. Some benzo-γ-pyrones (chromones).

Isoeugenitin
Myrt. *Eugenia caryophyllata*
Isoeugenitol
Myrt. *Eugenia caryophyllata*
Karenin
Meli. *Ptaeroxylon obliquum* (htwd)
Khellin (Kellin; Visammin)
Umbell. *Ammi visnaga* (sd)
Khellinin (Chellol-2-glucoside)
Umbell. *Ammi visnaga* (sd)
Khellinol (5-Norkhellin)
Umbell. *Ammi visnaga* (sd)
Peucenin (fig. 148)
Meli. *Ptaeroxylon obliquum* (htwd)
Umbell. *Peucedanum ostruthium* (rhiz.)
Ptaerochromenol
Meli. *Ptaeroxylon obliquum* (htwd)
Ptaerocyclin
Meli. *Ptaeroxylon obliquum* (htwd)

Ptaeroglycol
Meli. *Ptaeroxylon obliquum* (htwd)
Ptaeroxylin (Deoxy-karenin; fig. 148)
Meli. *Cedrelopsis grevei, Ptaeroxylon obliquum* (htwd)
Ptaeroxylinol
Meli. *Ptaeroxylon obliquum* (htwd)
Ptaeroxylone
Meli. *Ptaeroxylon obliquum*
Sorbifolin
Rut. *Spathelia sorbifolia* (rt)

II.3 Dibenzo-γ-Pyrones (Xanthones)

GENERAL

These plant constituents are derivatives of *dibenzo-γ-pyrone* (xanthone; fig. 149). Karrer (1958) says that only a few *xanthones* have been found in plants but I have records of about 70, many of them from a paper by Gottlieb (1968). A useful review is that by Roberts (1961). Mostly they are free, but some at least occur also as glycosides. They are found in fungi, in lichens, and in at least one fern. In the higher plants they may occur in all parts. All are yellow to red-yellow in colour.

Xanthone itself may not occur in plants, at least I have no record of it. The naturally occurring *xanthones* are hydroxy-, methoxy-, and other derivatives of *xanthone*.

We have records of them from the following dicotyledonous families: *Anacardiaceae* (a few), *Flacourtiaceae*, *Gentianaceae* (several), *Guttiferae* (many: see discussion under that family), *Hippocrateaceae*, *Leguminosae*, *Moraceae*, *Polygalaceae* and *Sapotaceae*. From the monocotyledons we have: *Liliaceae* and *Iridaceae*.

List and Occurrence

Alvaxanthone
Mor. *Maclura pomifera*
Bellidifolin (1,5,8-Trihydroxy-3-methoxy-xanthone; fig. 149)
Gentian. *Gentiana bellidifolia*
Celebixanthone (3,4,8-Trihydroxy-2-methoxy-1-(3-methyl-2-butenyl)-xanthone)
Gutt. *Cratoxylon celebicum*
Corymbiferin (4,5-Di-*O*-methyl-corymbin)
Gentian. *Gentiana bellidifolia* (rt), *corymbifera* (as glycoside)

Decussatin (8-Hydroxy-1,3,7-trimethoxy-xanthone)
Gentian. Swertia decussata
Dehydrocyclo-guanandin
Gutt. Calophyllum brasiliense
6-Dehydroxy-jacareubin
Gutt. Calophyllum brasiliense, scriblitifolium (htwd); *Kielmeyera ferruginea* (bk), *speciosa*
Demethyl-bellidifolin (Demethyl-swertianol)
Gentian. Gentiana bellidifolia, Swertia tosaensis
6-Deoxy-jacarubin
Gutt. Calophyllum inophyllum (htwd), *scriblitifolium* (htwd); *Kielmeyera speciosa*
Deoxy-morellin
Gutt. Garcinia morella
Dihydro-isomorellin
Gutt. Garcinia morella
1,3-Dihydroxy-5-methoxy-xanthone
Gutt. Calophyllum brasiliense
1,7-Dihydroxy-8-methoxy-xanthone
Gutt. Kielmeyera excelsa, ferruginea (bk), *petiolaris*
5,6-Dihydroxy-7-methoxy-xanthone
Gutt. Kielmeyera corymbosa
6,7-Dihydroxy-8-methoxy-xanthone
Gutt. Kielmeyera speciosa
1,5-Dihydroxy-xanthone (fig. 149)
Gutt. Mammea americana, Mesua ferrea
4,5-Dimethoxy-bellidin (4,7-Di-*O*-methyl-bellidin)
Gentian. Gentiana bellidifolia (rt)
4,7-Dimethoxy-bellidin
Gentian. Gentiana bellidifolia (rt)
2-(3,3-Dimethylallyl)-1,3,5,6-tetrahydroxy-xanthone
Gutt. Calophyllum inophyllum (htwd)
2-(3,3-Dimethylallyl)-1,3,5-trihydroxy-xanthone
Gutt. Calophyllum scriblitifolium (htwd)
2-(3,3-Dimethylallyl)-1,3,7-trihydroxy-xanthone
Gutt. Calophyllum scriblitifolium (htwd)
Euxanthone (Purrenone; 1,7-Dihydroxy-xanthone) seems to be more widely spread than most *xanthones*.
Anacardi. Mangifera indica
Gutt. Calophyllum sclerophyllum, Kielmeyera excelsa, Mammea americana, Mesua ferrea, Platonia insignis, Symphonia globulifera
Gambogic acid
Gutt. Garcinia hanburyi, morella

Gentioside (Gentiin; Isogentisin-3-primeveroside?)
Gentian. *Gentiana lutea*
Gentisin (Gentianin (1); 1,7-Dihydroxy-3-methoxy-xanthone; fig. 149)
Gutt. *Calophyllum brasiliense*
Gentian. *Gentiana lutea, Swertia japonica*
Globuxanthone
Gutt. *Symphonia globulifera*
Guanandin
Gutt. *Calophyllum brasiliense*
1-Hydroxy-3,7-dimethoxy-xanthone
Gutt. *Calophyllum brasiliense*
1-Hydroxy-7,8-dimethoxy-xanthone
Gutt. *Kielmeyera petiolaris*
5-Hydroxy-1,3-dimethoxy-xanthone
Gutt. *Kielmeyera coriacea, corymbosa, ferruginea* (bk), *speciosa*
5-Hydroxy-6,7-dimethoxy-xanthone
Gutt. *Kielmeyera coriacea, corymbosa, ferruginea* (bk), *rupestris* (wd), *speciosa*
6-Hydroxy-5,7-dimethoxy-xanthone
Gutt. *Kielmeyera speciosa*
6-Hydroxy-7,8-dimethoxy-xanthone
Gutt. *Kielmeyera rupestris* (wd), *speciosa*
1-Hydroxy-7-methoxy-xanthone
Gutt. *Kielmeyera corymbosa, excelsa*; *Mesua ferrea*
7-Hydroxy-8-methoxy-xanthone
Gutt. *Kielmeyera excelsa, speciosa*
5-Hydroxy-6,7-methylenedioxy-xanthone
Gutt. *Kielmeyera corymbosa, speciosa*
3-Hydroxy-1,5,6-trimethoxy-xanthone
Gutt. *Kielmeyera rupestris*
5-Hydroxy-xanthone (fig. 149)
Gutt. *Calophyllum brasiliense, Mammea americana*
7-Hydroxy-xanthone
Gutt. *Kielmeyera excelsa, speciosa*; *Mammea americana*
Isobellidifolin (1,3,8-Trihydroxy-5-methoxy-xanthone)
Gentian. *Gentiana bellidifolia*
Isogentisin (1,3-Dihydroxy-7-methoxy-xanthone)
Gentian. *Gentiana lutea*
Isoguanandin
Gutt. *Calophyllum brasiliense*
Isomorellic acid
Gutt. *Garcinia morella*