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Mallotus species from Vietnamese mountainous areas: phytochemistry and pharmacological activities

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Abstract The genus *Mallotus* belongs to Malphighiales order and Euphorbiaceae family. *Mallotus*, commonly known as "Ba bet" in Vietnam, is one of the most diverse and richest genera of the Euphorbiaceae family in Vietnam, where about 40 *Mallotus* species may be found. Some *Mallotus* species are used in traditional medicine in Vietnam for different indications. They are concentrated in mountainous areas with an altitude below 1,000 m, but some species can grow at an altitude of 2,000 m, such as *Mallotus oreophilus* Müll. Arg. Some *Mallotus* species are known to contain different natural compounds, mainly diterpenoids, triterpenoids, steroids, flavonoids, coumarinolignoids, phloroglucinol derivatives or benzopyrans, and to exhibit interesting biological activities such as antimicrobial, antioxidant, antiviral, or cytototoxic ones. Some of these properties may be explained by their chemical composition as, for example, benzopyrans accounting for the cytotoxicity of *Mallotus apelta* extracts. However, although these species seem to have a great medicinal potential, the existing knowledge about most *Mallotus* species is still in most cases very limited. This review underlines the interest to continue the study of this genus of the Euphorbiaceae.

Keywords Mallotus · Euphorbiaceae · Vietnam · Natural compounds · Biological activities

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Introduction

The genus *Mallotus*, commonly known as "Ba bet" in Vietnam, is one of the most diverse and richest genera of the Euphorbiaceae family in Vietnam where about 40 *Mallotus* species may be found among which six species and one variety are endemic. These endemic species, *Mallotus canii* Thin, *Mallotus chuyenii* Thin, *Mallotus eberhardtii* Gagnep., *Mallotus hanheoensis* Thin, *Mallotus poilanei* Gagnep., *Mallotus*

sathavensis Thin, *Mallotus cuneatus* Ridl. var. *glabratus* Thin, have been recently found, distributed in several regions in Vietnam and there is still a lack of information about them. Species belonging to the *Mallotus* genus are usually shrubs or small trees and grow in rainy, ever green primary or secondary forests. They can be also found in deciduous forests. Some species are considered as "first-coming plants" of forests recycling. Naturally, species are chiefly propagated from seeds. They are concentrated in mountainous areas with an altitude below 1,000 m, but some species can grow at an altitude of 2,000 m, such as *Mallotus oreophilus* Müll. Arg. (Thin 2003).

The genus *Mallotus* belongs to the Malphighiales order, the Euphorbiaceae family, Acalyphoideae subfamily, Acalypheae pro parte, Rottlerinae subtribe (Nowicke and Takahashi 2002). This genus includes approximately 150 species distributed in tropical and sub-tropical regions in Asia (Cambodia, China, India, Laos, Malaysia, Sri Lanka, Thailand, Vietnam). A few species are found in the North and East of Australia and the Pacific-Ocean Archipelago (the East of Fiji). Only two species are found in Africa and Madagascar (Schatz 2001). *M. oppositifolius* (Geiseler) Müll. Arg. is distributed in different African countries (Central Africa, Ghana, Nigeria, Tanzania) and Madagascar. *M. baillonianus* Müll. Arg. is endemic to Madagascar. The genus *Mallotus* is richer in Vietnam than in China, where 28 species are described of which seven are endemic. Sixteen species are common in Vietnam and China. In general, these species are distributed in higher altitude in China than Vietnam (Qiu and Gilbert 2008). Some species of the genus *Mallotus* (*M. apelta*, *M. barbatus*, *M. floribundus*, *M. glabriusculus*, *M. macrostachyus*, *M. oblongifolius*, *M. paniculatus*, *M. philippinensis*, *M. poilanei*) are used as medicinal plants in the traditional medicine in Vietnam and the South-East Asian countries for the treatment of various ailments ranging from minor infections such as gastrointestinal disorders to dysentery, hepatic diseases, cutaneous diseases, fever and malaria, and a series of other indications. The researched parts of the *Mallotus* species include aerials parts, bark, heartwood, leaves, roots, seeds, stem bark and whole plants. Some *Mallotus* species are known to contain different natural compounds, mainly terpenoids, polyphenols and benzopyrans. The compounds isolated from the *Mallotus* genus and extracts show many different biological activities including antioxidant, antiviral, antimicrobial, anti-inflammatory or cytotoxic. Some of these properties are attributed to the presence of specific classes of natural compounds, for example, benzopyrans accounting for the cytotoxicity of *Mallotus apelta* extracts (Van Chau et al. 2005a; Van Kiem et al. 2004) or polyphenols accounting for the antiradical activity of *Mallotus metcalfianus* extracts (Rivière et al. 2009).

In this review paper, we will summarize the data of the literature concerning the phytochemistry and the pharmacological activities of *Mallotus* species, described over the past few decades (Table 1; Fig. 1).

Phytochemistry

For some *Mallotus* species, studies were published on their chemical composition, especially for *M. apelta*, *M. metcalfianus*, *M. philippinensis*, *M. paniculatus*, *M. repandus*. These *Mallotus* species are known to contain different natural compounds, mainly diterpenoids, triterpenoids, steroids, benzopyranes, flavonoids, coumarinolignoids or phloroglucinol derivatives. The existing knowledge about the other investigated plants is in most cases very limited. However, some data underline the isolation of a novel furanocarboxamide from *M. cuneatus* (Groweiss et al. 1994), scopoletin from *M. resinosus* (Ma et al. 2004), phloroglucinol derivatives from *M. pallidus* (Supudompol et al. 2004; Likhitwitayawuid and Supudompol 2005) or triterpenoids and casbane-type diterpenoid lactones from *M. hookerianus* (Hui and Li 1976; Bai et al. 2006).

Terpenoids and steroids

Diterpenoids and diterpenic lactones (Table 2)

Cheng et al. (1999) and Cheng and Chen (1999) isolated five new diterpenoids (**1–5**) from the petroleum ether fraction of the ethanolic extract of *M. apelta*. Three highly oxidized casbane-type diterpenoids with unique α,β-unsatured γ-lactones, named hookerianolides A, B, and C (**6–8**), were isolated from the methylene chloride extract of *M. hookerianus* (Bai

Table 1 Vietnamese *Mallotus* species

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus anisopodus Airy Shaw	ND	Ruoikhe	T	V (South province AG), Ca, L	100–500	ND	ND	ND	ND
Mallotus apelta (Lour.) Müll. Arg.	*Ricinus apelta* Lour.	Babet trang, Buc trang, Bui bui, Bai bai, Bum bup, Bang bac, Cay ruong	Sm T, Sh	V (N to S), Ch	100–700	100–1,000	B: dh, ga, gy, hem, hep, oe L: co, cu, dh, gy, hep, oe, ot R: ai, dh, ga, gy, hep, oe	L: benzopyrans[1,2,3,4], pentacyclic triterpenoids and steroids[5,6,7,8], flavonoids[6,9,10], carotenoids[11], anthraquinone, coumarin and nicotinic acid[12] R: pentacyclic triterpenoids and steroids[13], pyridine type alkaloid and ellagic acid derivative[14] S: pentacyclic triterpenoids and steroids[15], iridoid (mussaenoside)[15], coumarinolignoids[16,17] Wp: diterpenoids[18,19]	Antiviral D-HBV[20] Bacteriostatic: triterpenoids and benzopyrans[1,13] Cytotoxic: benzopyrans[4] Hepatoprotective: coumarinolignoids[17,21] Inhibitory effect of reverse transcriptase and cellular DNA polymerase[22] Inhibitory effect of NFAT transcription and NF-κB activation[11]
Mallotus barbatus (Wall.) Müll. Arg.	*Rottlera barbata* Wall.	Bung buc, Bup bong gai, Bong bet, Bung buc gai, Ba bet long, Ruoi cau, Cam lon, Nhung dien rau	Sm T, Sh	V (N to S), Ch, Ind, L, Mal, Mya, Th	100–1,100	200–1,300	B: ga L: ac, hem, oe, sc R: an, fe, diu, cho, hea	L: polyprenols[23]	ND
Mallotus canii Thin	ND	Babet gialai	T	Endemic V (South province GL)	100–500	ND	ND	ND	ND
Mallotus chrysocarpus Pamp.	*M. contubernalis* var. *chrysocarpus* (Pamp.) Hand.-Mazz., *M. repandus* var. *chrysocarpus* (Pamp.) S.M. Hwang	Babet qua vang, Ruoi trai vang	Sm Sh	V (North province HT) Ch	100–500	500–1,000	ND	ND	Antiviral HIV[24]

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus chuyenii Thin	ND	Babet hoabinh	T	Endemic V (North province HB)	100–500	ND	ND	ND	ND
Mallotus clellandii Hook.f.	ND	Ruoi clelland, Ruoi tron, Ruoi khong long, Nhung dien clelland, Nhung dien khong long	Sm T, Sh	V (South provinces), Ca, L, Mya, Th	100–500	ND	ND	ND	ND
Mallotus contubernalis Hance	*M. repandus* var. *repandus*, *M. repandus* var. *scabrifolius* (A. Juss.) Müll. Arg.	Babet, Don xuong, Canhkien la bac, Buctruong ba ngan, Rem ban day	Sm T, Sh	V (North provinces), Ch, L	100–500	100–600	ND	ND	ND
Mallotus cuneatus Ridl.	*M. resinosus* var. *cuneatus* (Ridl.) N.P. Balakr. & Chakrab.	Duoi rung, Ruoi rung	Sm T, Sh	V (N to S), Ca, Ind, Mal, Phi, Th	100–500	ND	ND	L, T: furanocarboxamide[25]	ND
Mallotus cuneatus Ridl. var. *glabratus* Thin	ND	Babet nhan	T	Endemic V (North provinces)	100–500	ND	ND	ND	ND
Mallotus dispar (Blume) Müll. Arg.	*Rottlera dispar* Blume	Ruoikhong deu, Nhung dien khong deu	Sm T, Sh	V, Indo, Mal, Phi, Th	ND	ND	ND	ND	ND
Mallotus eberhardtii Gagnep	ND	Dodot	Sm T, Sh	Endemic V (Central and South provinces, TTH and KG)	100–500	ND	ND	ND	ND
Mallotus esquirolii H. Lév.	*M. grossedentatus* Merr. & Chun	Babet esquirol	T	V (North provinces LS and HB) Ch	100–500	300–1,500	ND	ND	ND

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus floribundus (Blume) Müll. Arg.	*Adisca floribunda* Blume	Babet nhieu hao, Bach dan, Ruoi trung bo, Ba bet hoa nhieu	Sm T, Sh	V (N to S), Austr, Ca, L, Indo, Mal, NG, Phi, Th	100–500	ND	R: dh, fe, gy Wp: sc	ND	ND
Mallotus glabriusculus (Kurz) Pax & K. Hoffm.	*Coelodiscus glabriusculus* Kurz	Babet nhan, Chiet canh, Kien canh, Nhung dien coudere, Ruoi khong long	Sm T, Sh	V (South provinces), Ca, L, Mya	100–500	ND	R: cou	ND	ND
Mallotus hanheoensis Thin	ND	Babe thon heo	T	Endemic V (South province KH)	100–500	ND	ND	ND	ND
Mallotus hookerianus (Seem.) Müll. Arg.	*Hancea hookeriana* Seem.	Babet cuong long, Ba bet long dung, Nhot vang, Chua nga, Choi moi nep, Nhung dien hooker, Ruoi hooker	T	V (N to S), Ch, NG	100–500	100–900	ND	L, S: pentacyclic triterpenoids[26], casbane-type diterpenoid lactones[27]	ND
Mallotus lanceolatus (Gagnep) Airy Shaw	*Coelodiscuslanceolatus* Gagnep	Babet thon, Ruoi thon, Nhung dien thon	Sm T, Sh	V (N to S), Ca, L, Th	100–500	ND	ND	ND	ND
Mallotus luchenensis F.P. Metcalf.	*M. barbatus* var. *barbatus*, *M. barbatus* var. *wui* H.S. Kiu	Camlon, Bum bup, Ruoi luchen	Sm T, Sh	V(N), Ch	100–800	200–1,300	ND	ND	ND
Mallotus macrostachyus (Miq.) Müll. Arg.	*Rottlera macrostachya* Miq.	Babet chum to, Bum bup bong to, Buc chum to, Ruoi duoi to, Ruoi trang, Nhung dien duoi to, Nhung dien trang	T	V (N to S), Ind, Indo, Mal, Phi, Th	100–500	ND	L: ac, hem, wo	ND	ND

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus metcalfianus Croizat	ND	Babet do, Ba bet mecalf, Ruoi mecalf	Sm T	V (N to S), Ch	100–1,000	100–1,900	ND	Wp: flavonoids, policosanol, flavonolignanes, pentacyclic triterpenoids, phenolic acids, megastigmane[28]	ND
Mallotus microcarpus Pax & K. Hoffm.	ND	Babet qua nho, Ruoi trai nho	Sm T, Sh	V (N to C), Ch	100–500	200–1,000	ND	ND	ND
Mallotus mollissimus (Vahl ex Geiseler) Airy Shaw	*Croton mollissimus* Vahl ex Geiseler	Bucnau, Babet nau, Ruoi mem, Buc qua thau dau	Sm T	V (N to S), Austr, Ca, Indo, L, Mal, NG, Phi	100–500	ND	ND	ND	ND
Mallotus nanus Airy Shaw	ND	Ba bet lun, Ruoi thorel	Sm T, Sh	V(C), Ca, L	100–500	ND	ND	ND	ND
Mallotus oblongifolius (Miq.) Müll. Arg.	*Rottlera oblongifolia* Miq., *Hancea muricata* Benth., *M. alternifolius* Merr., *M. columnaris* Warb., *M. furetianus* Müll. Arg., *M. helferi* Müll. Arg., *M. maclurei* Merr., *M. oblongifolius* var. *helferi* (Müll. Arg.) Pax & K. Hoffm., *M. odoratus* Elmer, *M. proterianus* Müll. Arg., *M. puberulus* Hook. f.	Choc mon, Choc moc, Choc mot, Cam heo, Ruoi tron dai	Sm T, Sh	V (N to S) Austr, Ca, Ind, Indo, L, Mal, Mya, NG, Phi, Th	100–800	ND	R: ma Wp: dh, ga	ND	ND
Mallotus oreophilus Müll. Arg.	*M. japonicuss* var. *oreophilus* (Müll. Arg.) S.M. Hwang	Babet nui cao	Sm T, Sh	V (Lao Cai province), Ch, Ind	700–2,000	600–2,000	ND	ND	ND
Mallotus pallidus (Airy Shaw) Airy Shaw	*M. philippensis* var. *menglianensis* C.Y. Wu ex S.M. Hwang, *M. philippinensis* var. *pallidus* Airy Shaw	Babet tai	Sm T, Sh	V (N to C), Ch, Th	100–500	1,200–1,400	ND	L: phloroglucinol derivatives[29,30,31]	Antiviral HIV-1, HSV-1, HSV-2: phloroglucinol derivatives[31]

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus paniculatus (Lam.) Müll. Arg.	*Croton paniculatus* Lam., *Echinus trisulcus* Lour., *M. chinensis* Müll. Arg., *M. cochinchinensis* Lour., *M. formosanus* Hayata, *M. paniculatus* var. *formosanus* (Hayata) Hurus., *Rottlera paniculata* (Lam.) A Juss.	Buc bac, Bong bet, Bai dai, Bum bup nau, Bung buc nau, Ba bet nam do, Bach thu	Sm T, T	V (N to S), NE Austr, Ca, Ch, Ind, Indo, L, Mal, Mya, NG, Phi, Th	100–1,650	100–1,300	F: co, oe R: gy Wp: fe, hea, wo	L: pentacyclic triterpenoids and steroids[32] SE: cardenolides[33,34], fatty acids[35] S: pentacyclic triterpenoids and steroids[26,32]	ND
Mallotus peltatus (Geiseler) Müll. Arg.	*Aleurites peltata* Geiseler, *Rottlera oblongifolia* Miq., *Hancea muricata* Benth., *M. furetianus* Müll. Arg., *M. maclurei* Merr., *M. oblongifolius* (Miq.) Müll. Arg.	Babet long, Ruoi long	Sm T	V (LC and South provinces), Ch, Ind, Indo, Mal, Mya, NG, Phi, Th	1,000–1,500	200–1,000	ND	L: tannins, triterpenoids and steroids, saponins, reducing sugars[36,37]	Antibacterial[36,38] Anti-inflammatory[36,38] Antipyretic[39] Neuropharmacological[37]
Mallotus philippinensis (Lamk.) Müll. Arg.	ND	Canhkien, Mot, Rum nao, Ba chia, Thuoc san, Tho khang sai, Rum hao	Sm T, Med T	V (N to S), Austr, Ca, Ind, Indo, L, Mal, Mya, Phi, Th, SL	100–500	ND	B: an, antis, cu, fe, hem, wo F: antis, cu, fe, ga, hem, oe, pa, sy L: cu, dh, diu, dy, wo R: antic, antis, dh, dy, fe, hem SE: diz, ve	B: phenolic compounds, condensed tannins[40,41], triterpenoids[42] F: dimeric chalcones derivatives[43,44], phenolic compounds[40] HW: triterpenoids[42], isocoumarin (bergenin)[42] L: tannins[45], bergenin[42] SB: pentacyclic triterpenoids and steroids[46]	Antioxidant[40,41] Antibacterial and antifungal[51,52] Bactericidal (*Helicobacter pylori*): rottlerin[53] Anti-inflammatory, immunoregulatory: chalcones[44] Antifertility[54]: rottlerin[55] Proteine kinase inhibitor PKCδ: rottlerin[50,56] Anthelminthic in ruminants[57] Anticestodal in beetal goats[58]

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
								SE: cardenolides[47], kamala oil (kamlolenic acid and hydroxy acids)[48]	
								Wp: flavonoids, phloroglucinol derivatives (rottlerin)[49,50]	
Mallotus pierrei (Gagnep) Airy Shaw	*Coelodiscus pierrei* Gagnep	Nhungdien pierre, Ruoi pierre	Sm T	V (South provinces DN, BRVT), Th	100–600	ND	ND	ND	ND
Mallotus poilanei Gagnep.	ND	Sita, Sito	Sm Sh	V (Central provinces)	100–500	ND	L: hea	ND	ND
Mallotus repandus (Rottler) Müll. Arg.	*Croton repandus* Willd.	Buc buc truon, Buc buc leo, Bum bup truon, Bum bup leo, Ruoi tran, Nhung dien bai	Sm T, Sh	V (N to S), N Austr, Ca, Ch, Ind, Indo, L, Mal, Mya, NG, Phi, SL, Th	100–500	100–1,000	L: sc, pimp R: fe, infl Wp: co, se	AP: iso-coumarin (bergenin)[59], cyano-γ-pyridone (mallorepine)[59] L: hydrolyzable tannins[60] S: triterpenoids[61], bergenin[61,62], D: A friedo-oleanane lactones[63] RB: triterpenoids, bergenin[61] Wp: diterpenic lactones[64,65], triterpenoids[66]	Antiradical[67] Antiviral HIV-1: hydrolyzable tannins[68]Antiulcerogenic: bergenin[59] Antitumor[69] Uterus muscle stimulant[70] Antihepatotoxic[71]
Mallotus resinosus (Blanco) Merr.	*Adelia resinosa* Blanco	Nhungdien mut, Ruoi resin	T	V (N to S), Ca, Ind, Indo, Mal, NG, Phi, SL	100–500	ND	ND	R: coumarins (scopoletin)[72]	DNA cleavage[72]
Mallotus sathayensis Thin	ND	Babet sa thay	T	Endemic V (Central province KT)	100–500	ND	ND	ND	ND

Table 1 continued

Botanical name	Synonyms[a]	Vietnamese vernacular names	Plant[b]	Distribution[c]	Altitude Vietnam (m)	Altitude China (m)	Traditonal uses[d,e]	Phytochemistry[f,g]	Pharmacological activities[g]
Mallotus spodocarpus Airy Shaw	ND	Babet set, Ruoi trai set kem	Sm Sh	V (South province NT), Th	100–500	ND	ND	ND	Anti-inflammatory, analgesic[73]
Mallotus thorelii Gagnep.	ND	Nhungdien thorel, Ruoi thorel	Sh, Sm T	V (South province KG), Ca, Ch, L, Th	100–500	1,200–1,300	ND	ND	ND
Mallotus tsiangii Merr. & Chun	*Macaranga lowii* King ex Hook. f.	Ruoi tsiang	Sm Sh	V (North province VP), Ch	500–1,000	100–500	ND	ND	ND
Mallotus ustulatus (Gagnep.) Airy Shaw	*Coelodiscus ustulatus* Gagnep.	Babet lua, Ruoi cui	Sm Sh	V (C to S), Ca	100–500	ND	ND	ND	ND
Mallotus yunnanensis Pax & K. Hoffm.	*Mallotus hainanensis* S.M. Hwang	Babet van nam, Ruoi van nam	Sm Sh	V(N), Ch	100–500	100–1,400	ND	ND	ND

ND no data

[a] Synonyms: Missouri Botanical Garden website: http://www.tropicos.org/ (basionyms, synonyms or accepted names)

[b] Plant: *T*, tree; *Sm T*, small tree; *Med T*, medium tree; *Sh*, shrubs, *Sm Sh*, small shrubs

[c] Distribution: *AG*, An Giang province; *Ba*, Ria-Vung Tau province; *Aust*, Australia; *C*, Center; *Ca*, Cambodia; *Ch*, China; *DN*, Dong Nai province; *GL*, Gia Lai province; *HB*, Hoa Binh; *HT*, Ha Tai province; *Ind*, India; *Indo*, Indonesia; *KG*, Kien Giang province; *KH*, Khanh Hoa province; *KT*, Kon Tum; *L*, Laos; *LC*, Lao Cai province; *Mal*, Malaysia; *Mya*, Myamar; *N*, North; *NG*, New Guinea; *NT*, Ninh Thuan; *Phi*, Philippines; *S*, South; *SE A*, South-East Asia; *SL*, Sri Lanka; *Th*, Thailand; *TTH*, Thua Thien Hue province; *V*, Vietnam; *VP*, Vinh Phuc

[d] Parts used: *B*, bark; *C*, branches; *F*, fruits; *L*, leaves; *R*, roots; *SE*, seeds; *Wp*, whole plant

[e] Vietnamese traditional usage: *ac*, acne; *ai*, antiinflammatory; *an*, analgesic; *antic*, anticonvulsivant; *antis*, antiseptic; *cho*, cholera; *co*, contusions and traumatic injuries; *cou*, cough; *cu*, cutaneous diseases; *dh*, diarrhea; *diu*, diuretic; *diz*, dizziness; *dy*, dysentery; *fe*, fever; *ga*, gastrointesinal disorders; *gy*, gynecological infection; *hea*, headache; *hem*, hemostatic; *hep*, hepatic diseases; *infl*, influenza; *ma*, malaria; *oe*, oedema; *ot*, otitis; *pa*, parasiticid; *pimp*, pimple; *sc*, scabies; *se*, sedative; *sy*, syphilis; *ve*, vertigo; *wo*, wounds

[f] Parts studied: *A*, aerial parts; *B*, bark; *C*, branches; *F*, fruits; *HW*, heartwood; *L*, leaves; *R*, roots; *RB*, root bark; *S*, stems; *SB*, stem bark; *SE*, seeds; *T*, twigs; *Wp*, whole plant

[g] Sources: [1] An et al. (2001), [2] An et al. (2003), [3] Van Chau et al. (2005a), [4] Van Kiem et al. (2005), [5] Van Kiem et al. (2004), [6] Van Chau et al. (2004), [7] Van Chau et al. (2005b), [8] Van Chau et al. (2005c), [9] Wu et al. (2006), [10] Zhu et al. (2007), [11] Van Chau et al. (2005d), [12] Kang and Lu (2007), [13] Shan et al. (1985), [14] Cheng et al. (1998), [15] Qi et al. (2005), [16] Cheng and Chen (2000), [17] Xu et al. (2008), [18] Cheng et al. (1999), [19] Cheng and Chen (1999), [20] Xu et al. (2006), [21] Zhao et al. (2002), [22] Ono et al. (1989), [23] Sasak and Chonjnacki (1973), [24] Nguyen et al. (1997), [25] Groweiss et al. (1994), [26] Hui and Li (1976), [27] Bai et al. (2006), [28] Rivière et al. (2009), [29] Supudompol et al. (2004), [30] Likhitwitayawuid et al. (2005), [31] Likhitwitayawuid and Supudompol (2005), [32] Hui et al. (1969), [33] Roberts et al. (1966), [34] Roberts et al. (1967), [35] Yu et al. (1991), [36] Chattopadhyay et al. (2002a), [37] Chattopadhyay et al. (2003), [38] Chattopadhyay et al. (2006), [39] Chattopadhyay et al. (2002b), [40] Arfan et al. (2007), [41] Arfan et al. (2009), [42] Bandopadhyay et al. (1972), [43] Tanaka et al. (1998), [44] Daikonya et al. (2004), [45] Saijo et al. (1989b), [46] Nair and Rao (1993), [47] Roberts et al. (1963), [48] Gupta et al. (1953), [49] Lounasmaa et al. (1975), [50] Gschwendt et al. (1994), [51] Kumar et al. (2006), [52] Moorthy et al. (2007), [53] Zaidi et al. (2009), [54] Thakur et al. (2005), [55] Gujral et al. (1960), [56] Liao et al. (2005), [57] Jabbar et al. (2006), [58] Akhtar and Ahmad (1992), [59] Hikino et al. (1978), [60] Saijo et al. (1989a), [61] Huang et al. (1999), [62] Tomizawa et al. (1976), [63] Sutthivaiyakit et al. (2001), [64] Nakatsu et al. (1981), [65] Kawashima et al. (1976a), [66] Hui and Li (1977), [67] Lin et al. (1995), [68] Ogata et al. (1992), [69] Kawashima et al. (1976b), [70] Kawashima et al. (1975), [71] Yang et al. (1987), [72] Ma et al. (2004), [73] Intahphuak et al. (2004)

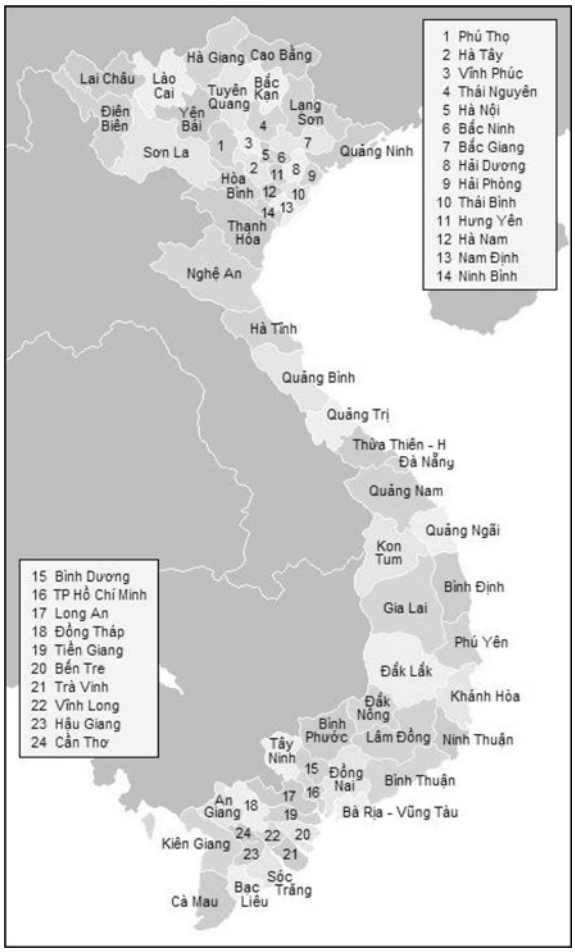


Fig. 1 Vietnamese provinces. http://commons.wikimedia.org/wiki/Image:VietnameseProvincesMapTiengViet.png(GNU_Free_Documentation_License)

et al. 2006). In 1976, two diterpenic lactones named mallotucin A and B (**9–10**) were obtained from *M. repandus* (Kawashima et al. 1976a). In 1981, Nakatsu et al. reported the isolation of three diterpenic lactones of which two were new from *M. repandus*: mallotucin B, C, and D (**10–12**).

Cardenolides (Table 3)

The seeds of *M. paniculatus* and *M. philippinensis* contain cardenolides. From the seeds of *M. paniculatus*, after fermentation, seven cardenolides were isolated, of which four were genins: two known (**18–19**), two new (**13–14**), and three were glycosides (**15–17**) (Roberts et al. 1966, 1967).

The seeds of *M. philippinensis* were found to contain after fermentation four cardenolides (**19–22**), of which two were new: corotoxigenin L-rhamnoside and coroglaucigenin L-rhamnoside (Roberts et al. 1963).

Carotenoids (Table 4)

β-Carotene and lutein (**23–24**) were isolated from the methanolic extract of the dried leaves of *M. apelta* (Van Chau et al. 2005b).

Iridoids (Table 5)

An iridoid, mussaenoside (**25**), was obtained from the ethyl acetate extract of the stems of *M. apelta* (Qi et al. 2005).

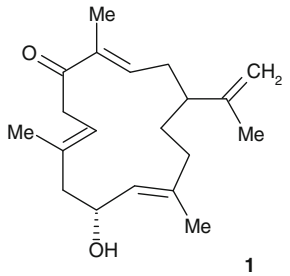
Polyprenols

In 1973, polyprenols were isolated from the leaves of *M. barbatus* (Sasak and Chonjnacki 1973). They were of 14–20 isoprene residues chain-length and they occurred in the form of acetic acid esters. The presence of long-chain polyprenols is frequent in leaves. It has been observed that the content of polyprenols in leaves increases with the age of the leaf and that in some species the age-dependent accumulation of polyprenols may attain extremely high values (Ranjan et al. 2001). In 2005, Van Chau et al. (2005d) reported the isolation of betulaprenol from *M. apelta*.

Triterpenoids (Tables 6, 7, 8)

Some pentacyclic triterpenoids with a 6/6/6/6/5 ring system (Table 6) were reported in some *Mallotus* species. A known triterpenoid, hennadiol (**26**) and a new, malloapelta A (**28**), were isolated from the methanolic extract of the dried leaves of *M. apelta* (Van Kiem et al. 2004; Van Chau et al. 2005d), whereas 3β,29-dihydroxylupane (**27**) was obtained from the roots of *M. apelta* (Shan et al. 1985). In 1976, Hui and Li reported the isolation of 29-nor-21αH-hopane-3,22-dione (**29**) from the stems of *M. paniculatus*. The petroleum ether extract of the heartwood of *M. philippinensis* yielded triterpenoids: betulin-3-acetate (**30**) as a major compound, lupeol acetate (**31**) and lupeol (**32**) (Bandopadhyay et al.

Table 2 Diterpenoids and diterpenic lactones



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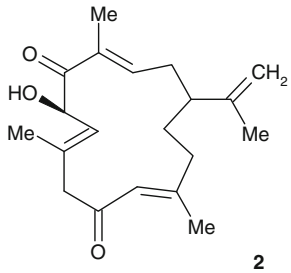

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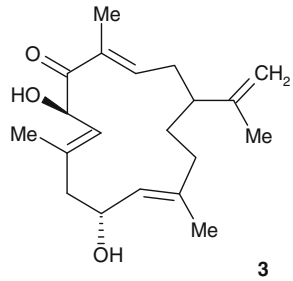

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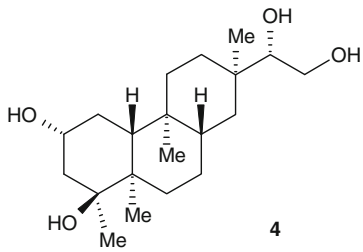

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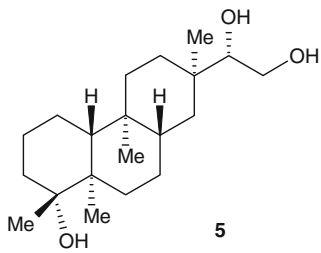

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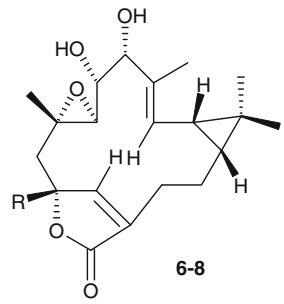

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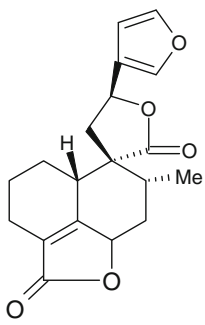

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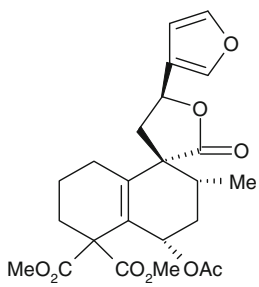

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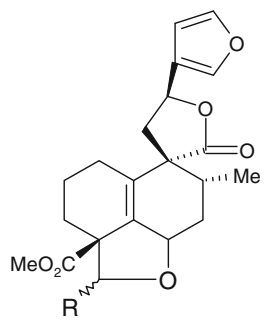
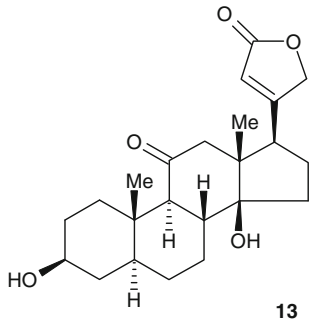


11-12

No.	Name	R	Plant	Ref.
1	10-Hydroxycembren-5-one		*M. apelta*	Cheng et al. (1999) and Cheng and Chen 1999)
2	6-Hydroxycembrene-5,10-dione		*M. apelta*	Cheng et al. (1999) and Cheng and Chen 1999
3	6,10-Dihydroxycembrene-5-one=malloapeltene		*M. apelta*	Cheng et al. (1999) and Cheng and Chen 1999
4	2α,4β,15,16-Tetrahydroxydolabradane		*M. apelta*	Cheng et al. (1999) and Cheng and Chen 1999
5	4α,15,16-Tetrahydroxydolabradane=malloapeltin		*M. apelta*	Cheng et al. (1999) and Cheng and Chen 1999
6	Hookerianolide A	OH	*M. hookerianus*	Bai et al. (2006)
7	Hookerianolide B	H	*M. hookerianus*	Bai et al. (2006)
8	Hookerianolide C	OC_2H_5	*M. hookerianus*	Bai et al. (2006)
9	Mallotucin A		*M. repandus*	Kawashima et al. (1976a)
10	Mallotucin B		*M. repandus*	Kawashima et al. (1976a)
				Nakatsu et al. (1981)
11	Mallotucin C	β-OH	*M. repandus*	Nakatsu et al. (1981)
12	Mallotucin D	α-OH	*M. repandus*	Nakatsu et al. (1981)

Table 3 Cardenolides



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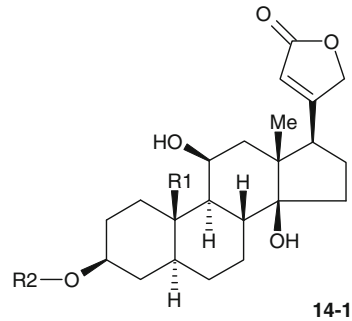
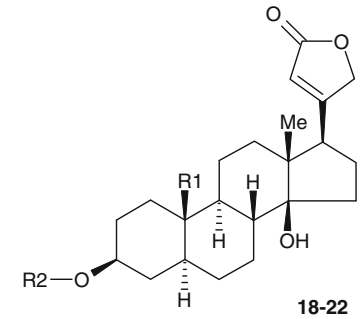


14-17



18-22

No.	Name	R1	R2	Plant	Ref.
13	5-Desarogenin			*M. paniculatus*	Roberts et al. (1966, 1967)
14	Mallogenin	CH_3	H	*M. paniculatus*	Roberts et al. (1966, 1967)
15	Malloside	CH_3	L-rham	*M. paniculatus*	Roberts et al. (1966, 1967)
16	Panoside	CH_2OH	L-rham	*M. paniculatus*	Roberts et al. (1966, 1967)
17	Glucopanoside	CH_2OH	Glc	*M. paniculatus*	Roberts et al. (1966, 1967)
18	Uzarigenin	CH_3	H	*M. paniculatus*	Roberts et al. (1966, 1967)
19	Coroglaucigenin	CH_2OH	H	*M. paniculatus*	Roberts et al. (1966, 1967)
				M. philippinensis	Roberts et al. (1963)
20	Coroglaucigenin L-rhamnoside	CH_2OH	L-rham	*M. philippinensis*	Roberts et al. (1963)
21	Corotoxigenin	CHO	H	*M. philippinensis*	Roberts et al. (1963)
22	Corotoxigenin L-rhamnoside	CHO	L-rham	*M. philippinensis*	Roberts et al. (1963)

1972). Lupeol was also obtained from *M. repandus* (Hui and Li 1977).

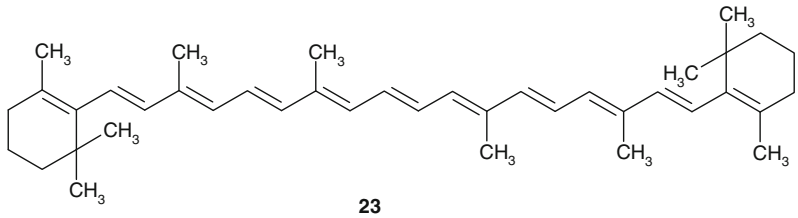
Pentacyclic triterpenoids with a 6/6/6/6/6 ring system are more often mentioned (Tables 7, 8). Friedelane-type triterpenoids are common in *Mallotus* species. Friedelin (**33**) was obtained from several *Mallotus* species: from the leaves of *M. apelta* (Van Kiem et al. 2004; Van Chau et al. 2005d), from the leaves and stems of *M. hookerianus* (Hui and Li 1976), from the leaves of *M. paniculatus* (Hui et al. 1969), from the stem bark of *M. philippinensis* (Nair and Rao 1993) and from *M. repandus* (Hui and Li 1977). Friedelin is common to many genera of Euphorbiaceae such as *Drypetes* (Wansi et al. 2006) or *Celaenodendron* (Castenada et al. 1993) and is also found in plants from other orders.

Friedelinol (**34**) was isolated from the leaves of *M. apelta* (Van Kiem et al. 2004; Van Chau et al. 2005d) and from *M. metcalfianus* (Rivière et al. 2009), whereas epifriedelanol (**35**) was isolated from the leaves of *M. apelta* (Van Kiem et al. 2004; Van Chau et al. 2005d), from the leaves and stems of *M. hookerianus* (Hui and Li 1976) and from the leaves of *M. paniculatus* (Hui et al. 1969). Three new D:A-friedo-oleanane lactones (**36–38**) were isolated from the stems of *M. repandus* (Sutthivaiyakit et al. 2001).

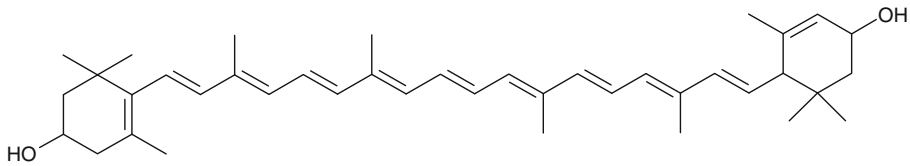
Other known pentacyclic terpenoids were detected in different *Mallotus* species: taraxerone (**39**), taraxerol (**40**) and epitaraxerol (**41**) in the leaves of *M. apelta* (Van Kiem et al. 2004; Wu et al. 2006; Van Chau et al. 2005d), erythrodiol-3-acetate (**42**) in the roots of *M. apelta* (Shan et al. 1985), acetylaleuritolic acid (**43**) in the stems of *M. apelta* (Qi et al. 2005) and in the petroleum ether and ether extracts of bark of *M. philippinensis* (Bandopadhyay et al. 1972). The first olean-18-ene triterpene oxidized at C-22 (**44**) was isolated from the stem bark of *M. philippinensis* (Nair and Rao 1993).

Several ursane-type triterpenoids were also isolated from *Mallotus* species: α-amyrin (**45**) from the petroleum ether and ether extracts of bark of

Table 4 Carotenoids



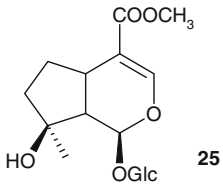
23



24

No.	Name	Plant	Ref.
23	*β*-Carotene	*M. apelta*	Van Chau et al. (2005b)
24	Lutein	*M. apelta*	Van Chau et al. (2005b)

Table 5 Iridoids



25

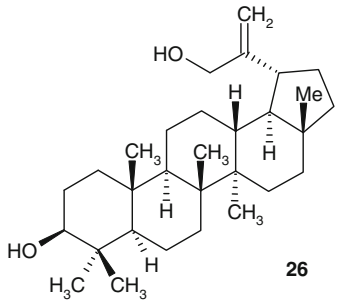
No.	Name	Plant	Ref.
25	Mussaenoide	*M. apelta*	Qi et al. (2005)

M. philippinensis (Bandopadhyay et al. 1972) and from *M. repandus* (Hui and Li 1977), ursolic acid (**46**) from the ethyl acetate extract of the stems of *M. apelta* (Qi et al. 2005), from *M. peltatus* (Chattopadhyay et al. 2002a, 2003) and from the stems and root bark of *M. repandus* (Hui and Li 1977; Huang et al. 1999), ursolic acid acetate (**47**) from the roots of *M. apelta* (Shan et al. 1985), 12-ursen-3-one (**48**) and 3-hydroxy-12-ursen (**49**) from the ethyl acetate extract of the stems of *M. apelta* (Qi et al. 2005). In 1976, Hui and Li reported the isolation of two new triterpene acids (**50–51**) from the ethanolic extract of the leaves of *M. hookerianus* (Hui and Li 1976). In 1977, the new triterpenes 3α-hydroxy-13α-ursan-28,12*β*-olide (**52**), 3*β*-hydroxy-13α-ursan-28, 12*β*-olide (**54**) and its benzoate (**55**) were isolated from *M. repandus* (Hui and Li 1977). In 1999, Huang et al. reported the isolation of three new triterpenoids, 3α-hydroxy-13α-ursan-28,12*β*-olide 3-benzoate (**53**), 3α-hydroxy-13α-ursan-28-oic acid (**56**) and 3α-hydroxy-28*β*-methoxy-13α-ursan-28,12*β*-epoxide 3-benzoate (**57**) from the stems and root bark of *M. repandus* (Huang et al. 1999).

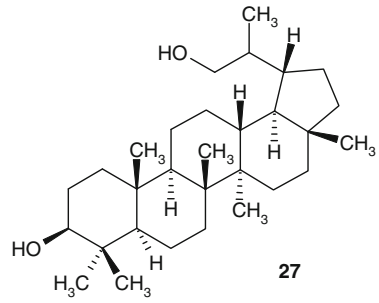
Steroids (Table 9)

Common steroids are mentioned in *Mallotus* species. *β*-Sitosterol (**58**) was isolated from the roots, the stems and the leaves of *M. apelta* (Shan et al. 1985; Qi et al. 2005; Wu et al. 2006), from the leaves and stems of *M. hookerianus* (Hui and Li 1976), from the stems of *M. paniculatus* (Hui et al. 1969), from *M. peltatus* (Chattopadhyay et al. 2002a, 2003, 2006), from the petroleum ether extracts of the heartwood and bark of *M. philippinensis* (Bandopadhyay et al. 1972). Daucosterol (**59**) was obtained from the leaves and the stems of *M. apelta* (Van Chau et al. 2004; Qi et al. 2005) and from the ether extract of the bark of *M. philippinensis* (Bandopadhyay et al. 1972).

Table 6 6/6/6/6/5-Ring triterpenoids



26



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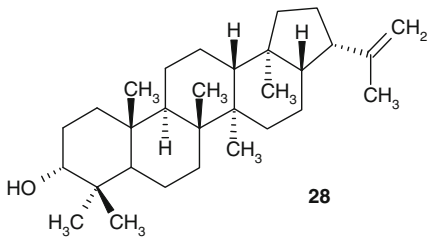

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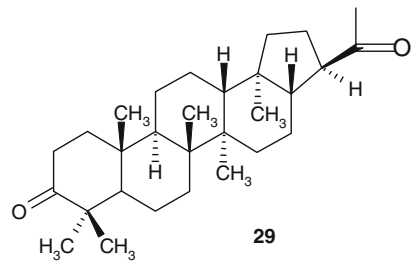

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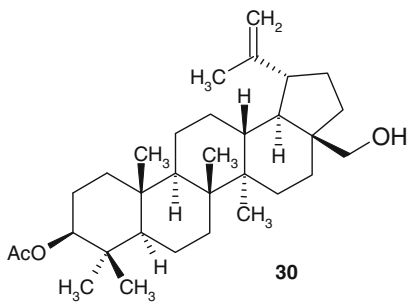
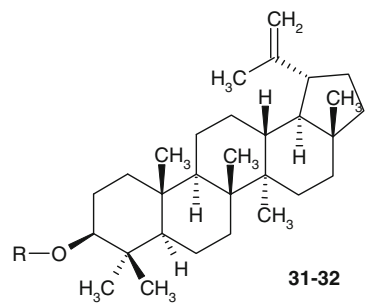


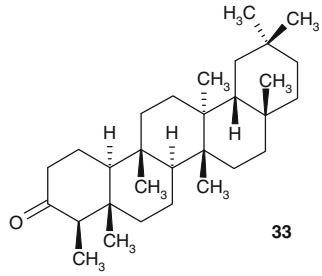
30

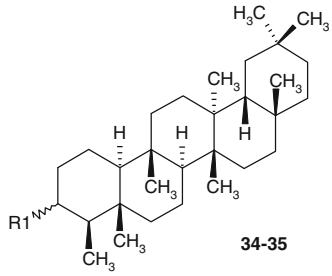


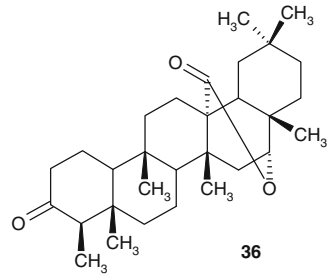
31-32

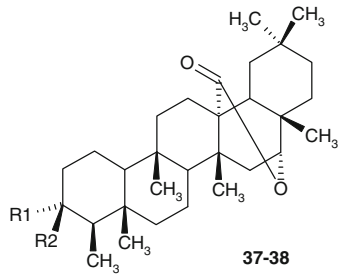
No.	Name	R	Plant	Ref.
26	Hennadiol		*M. apelta*	Van Kiem et al. (2004)
				Van Chau et al. (2005d)
27	3β,29-Dihydroxylupane		*M. apelta*	Shan et al. (1985)
28	3α,Hydroxyhop-22(29)-ene or malloapeltaA		*M. apelta*	Van Kiem et al. (2004)
29	29-Nor-21αH-hopan-3,22-dione		*M. paniculatus*	Hui and Li (1976)
30	Betulin-3-acetate		*M. philippinensis*	Bandopadhyay et al. (1972)
31	Lupeol-3-acetate	Ac	*M. philippinensis*	Bandopadhyay et al. (1972)
32	Lupeol	H	*M. philippinensis*	Bandopadhyay et al. (1972)
			M. repandus	Hui and Li (1977)

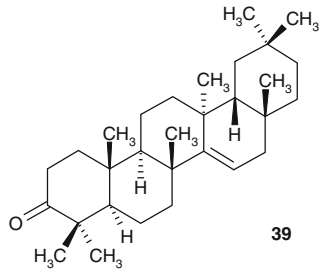
Table 7 6/6/6/6/6-Ring triterpenoids (1)

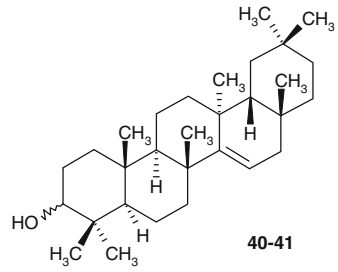


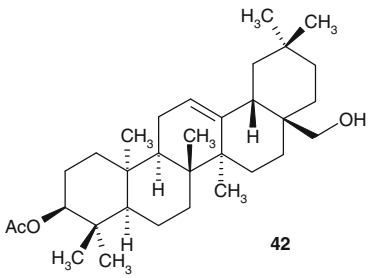


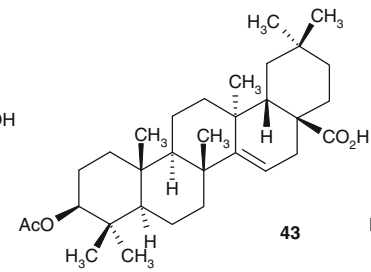


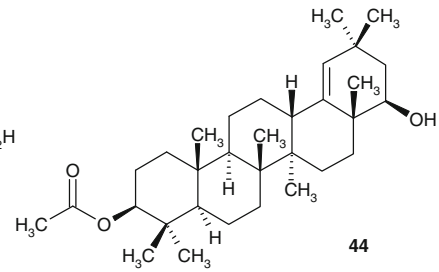












No.	Name	R1	R2	Plant	Ref.
33	Friedelin			*M. apelta*	Van Kiem et al. (2004) and Van Chau et al. (2005d)
				M. hookeriauns	Hui and Li (1976)
				M. paniculatus	Hui et al. (1969)
				M. philippinensis	Nair and Rao (1993)
				M. repandus	Hui and Li (1977)
34	Friedelinol or friedelin-3α-ol or friedelanol	α-OH		*M. apelta*	Van Kiem et al. (2004) and Van Chau et al. (2005d)
				M. metcalfianus	Rivière et al. (2009)
35	Epifriedelinol or friedelin-3β-ol or epifriedelanol	β-OH		*M. apelta*	Van Kiem et al. (2004) Van Chau et al. (2005d)
				M. hookerianus	Hui and Li (1976)
				M. paniculatus	Hui et al. (1969)
36	3-Oxo-D:A-friedo-oleanan-27,16α-lactone			*M. repandus*	Sutthivaiyakit et al. (2001)

Table 7 continued

No.	Name	R1	R2	Plant	Ref.
37	3α-Benzoyloxy-D:A-friedo-oleanan-27,16α-lactone	O–C(=O)Ph	H	*M. repandus*	Sutthivaiyakit et al. (2001)
38	3β-Hydroxy-D:A-friedo-oleanan-27,16α-lactone	H	OH	*M. repandus*	Sutthivaiyakit et al. (2001)
39	Taraxerone			*M. apelta*	Van Kiem et al. (2004)
					Van Chau et al. (2005d)
40	Taraxerol	β-OH		*M. apelta*	Wu et al. (2006)
41	Epitaraxerol	α-OH		*M. apelta*	Van Kiem et al. (2004)
					Van Chau et al. (2005d)
42	Erythrodiol-3-acetate			*M. apelta*	Shan et al. (1985)
43	Acetylaleuritolic acid or aleuritolic acid acetate			*M. apelta*	Qi et al. (2005)
				M. philippinensis	Bandopadhyay et al. (1972)
44	3β-Acetoxy-22β-hydroxyolean-18-ene			*M. philippinensis*	Nair and Rao (1993)

Ergosterol (**60**) was reported in the leaves of *M. apelta* (Van Chau et al. 2004), as well as stigmasterol (**61**). This last compound was also mentioned in the stems of *M. paniculatus* (Hui et al. 1969).

Other terpenoids (Table 10)

Squalene (**62**) and *trans*-phytol (**63**) were isolated from the methanolic extract of the leaves of *M. apelta* (Van Chau et al. 2004).

Phenolic compounds

Coumarins, isocoumarins and coumarinolignoids (Table 11)

Scopoletin (**64**), a simple coumarin, was detected in *M. resinosus* (Ma et al. 2004). Isoscopoletin (**65**) was obtained from the leaves of *M. apelta* (Kang and Lu 2007). Isopimpinellin (**66**), a furanocoumarin, was reported in the leaves of *M. apelta* (Van Chau et al. 2005d). Bergenin (**67**), an isocoumarin, was isolated in 1972, from the heartwood of *M. philippinensis*. This compound was also obtained from the bark and the leaves of *M. philippinensis* (Bandopadhyay et al. 1972). In 1976, Tomizawa et al. reported also the isolation of this same isocoumarin from *M. repandus*. Bergenin was also isolated in 1999 by Huang et al. (1999) from the stems and root bark of *M. repandus*.

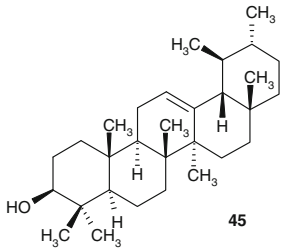
In 2000, three coumarino-lignoids, aquillochin (**74**), cleomiscosin A (**69**) and 5′-demethylaquillochin (**73**) were isolated from *M. apelta* (Cheng and Chen 2000).

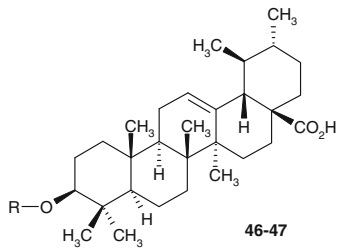
In 2008, three new coumarinolignoids, malloapelins A–C (**68**, **71**, **72**), together with three known coumarinolignoids (**69–71**), were isolated from the roots of *Mallotus apelta*. These compounds are three pairs of regioisomeric coumarinolignoids (Xu et al. 2008).

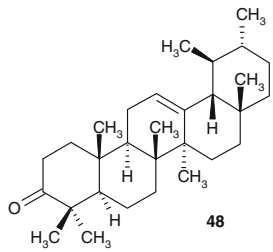
Flavonoids: flavonols, flavones, chalcones, flavonolignanes (Tables 12, 13)

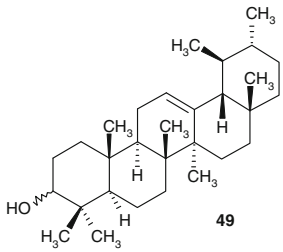
Flavonols glycosides such as quercitrin (**75**), were isolated from several *Mallotus* species: *M. apelta* (Van Chau et al. 2004), *M. metcalfianus* (Rivière et al. 2009), or identified, in a recent study conducted in our laboratory, in *M. nanus*, *M. cuneatus*, *M. paniculatus* (unpublished). Quercitrin was also obtained from other Euphorbiaceae genera: *Alchornea* (Manga et al. 2004), *Euphorbia* (Liu et al. 2007), *Phyllanthus* (Fang et al. 2008) and *Pedilanthus* (Abreu et al. 2008) but also in many plants from other families. Similarly, kaempferol glycosides have been described in some species of the Euphorbiaceae, for example in the genera *Euphorbia* (Saleh 1985) and *Acalypha* (Nahrstedt et al. 2006) but also in other families. Kaempferol 3-*O*-α-L-rhamnoside (**76**) was isolated from *M. metcalfianus* (Rivière et al. 2009) and identified in our laboratory in *M. barbatus* and several samples of *M. nanus* (unpublished). Glycoside dihydroflavonols such as astilbin (**79**) was isolated from *M. apelta* (Van Chau et al. 2004) and from *M. metcalfianus* (Rivière et al. 2009). To our knowledge, astilbin has not been described in other Euphorbiaceae, thus may have some

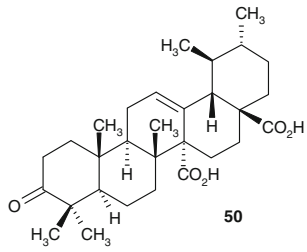
Table 8 6/6/6/6/6-Ring triterpenoids (2)

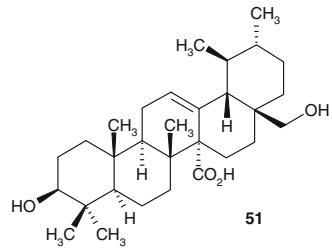


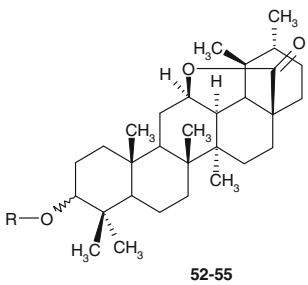


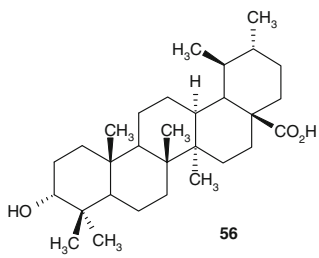


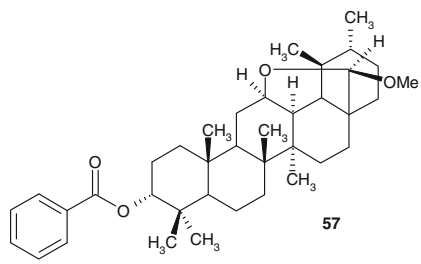










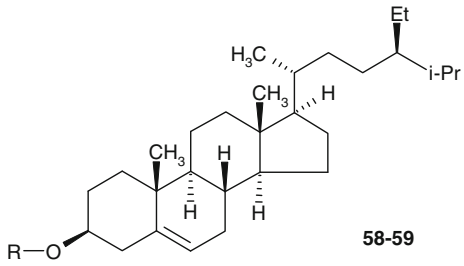


No.	Name	R	Plant	Ref.
45	α-Amyrine		*M. philippinensis*	Bandopadhyay et al. (1972)
			M. repandus	Hui and Li (1977)
46	Ursolic acid	H	*M. apelta*	Qi et al. (2005)
			M. peltatus	Chattopadhyay et al. (2002a, 2003)
			M. repandus	Hui and Li (1977) and Huang et al. (1999)
47	Ursolic acid acetate	Ac	*M. apelta*	Shan et al. (1985)
48	12-Ursen-3-one		*M. apelta*	Qi et al. (2005)
49	3-Hydroxy-12-ursen		*M. apelta*	Qi et al. (2005)
50	3-Oxours-12-ene-27,28-dioic acid		*M. hookerianus*	Hui and Li (1976)
51	3*β*,28-Dihydroxyurs-12-en-27-oic acid		*M. hookerianus*	Hui and Li (1976)
52	3α-Hydroxy-13α-ursan-28,12*β*-olide	α H	*M. repandus*	Hui and Li (1977)
53	3α-Hydroxy-13α-ursan-28,12*β*-olide 3-benzoate	α (C=O)Ph	*M. repandus*	Huang et al. (1999)
54	3*β*-Hydroxy-13α-ursan-28,12*β*-olide	*β* H	*M. repandus*	Hui and Li (1977)

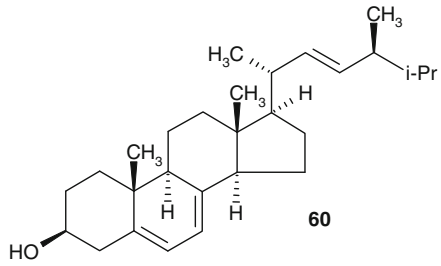
Table 8 continued

No.	Name	R	Plant	Ref.
55	3β-Hydroxy-13α-ursan-28,12β-olide 3-benzoate	β (C=O)Ph	*M. repandus*	Hui and Li (1977)
56	3α-Hydroxy-13α-ursan-28-oic acid		*M. repandus*	Huang et al. (1999)
57	3α-Hydroxy-28β-methoxy-13α-ursan-28,12β-epoxide 3 benzoate		*M. repandus*	Huang et al. (1999)

Table 9 Steroids



58-59



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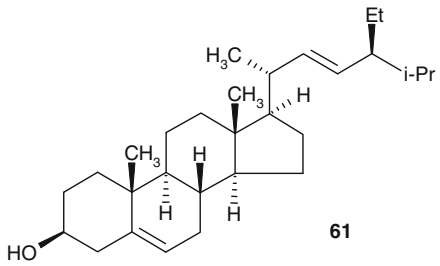
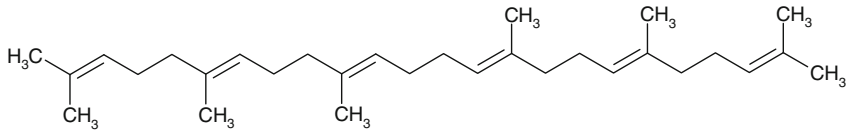


61

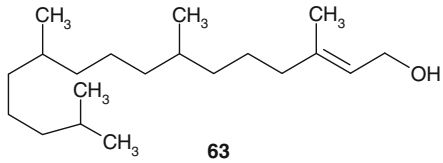
No.	Name	R	Plant	Ref.
58	β-Sitosterol	H	*M. apelta*	Shan et al. (1985), Qi et al. (2005) and Wu et al. (2006)
			M. hookerianus	Hui and Li (1976)
			M. paniculatus	Hui et al. (1969)
			M. peltatus	Chattopadhyay et al. (2002a, 2003, 2006)
			M. philippinensis	Bandopadhyay et al. (1972)
59	Sitosteryl β-D-glucose or daucosterol	Glc	*M. apelta*	Van Chau et al. (2004) and Qi et al. (2005)
			M. philippinensis	Bandopadhyay et al. (1972)
60	Ergosterol		*M. apelta*	Van Chau et al. (2004)
61	Stigmasterol		*M. apelta*	Van Chau et al. (2004)
			M. paniculatus	Hui et al. (1969)

chemotaxonomical interest. In a previous study, from *M. metcalfianus*, we isolated two other glycoside flavonols, quercetin 3-*O*-β-neohesperidoside (**77**) and kaempferol 3-*O*-β-neohesperidoside (**78**), but also a mixture of two pairs of new diastereoisomeric flavonolignans (±)-hydnocarpin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside)/(±)-hydnocarpin-D 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside) with a 2:1 ratio (**86**) (Rivière et al. 2009). The isolation of these products seems to have a chemotaxonomic interest as it is the first report of a flavonolignan in this family. Hydnocarpin not substituted by a

Table 10 Other terpenoids



62



63

No.	Name	Plant	Ref.
62	Squalene	*M. apelta*	Van Chau et al. (2004)
63	*Trans*-phytol	*M. apelta*	Van Chau et al. (2005d)

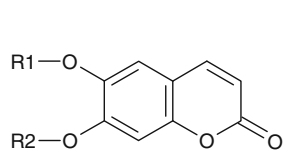
coumaric acid was also isolated from other families of the same order Malphigiales such as *Hydnocarpus wightiana* Blume (Flacourtiaceae) (Guz and Stermitz 2000) and from other families of different orders such as Ranunculales with *Meconopsis*, Papaveraceae or Sapindales with *Brucea*, Simaroubaceae (Shang et al. 2002; Pan et al. 2009). Lignans and neo-lignans are more common in Euphorbiaceae, such as in the genera *Phyllanthus* and *Trewia* (Bagalkotkar et al. 2006; Zhao and Shen 2004). We also isolated from *M. metcalfianus* two new flavones, luteolin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-*β*-glucopyranoside (**80**) and chrysoeriol 7-*O*-(4″-*O*-(*E*)-coumaroyl)-*β*-glucopyranoside) (**81**). Flavonoid *p*-coumaroylglucosides are commonly found in some genera of the Lamiaceae and they are generally considered as valuable markers in this family from a chemotaxonomic point of view (Sahpaz et al. 2002). The position of the coumaroyl substitution on the glucose is often described in position 3″ or 6″ but not in position 4″ (Sahpaz et al. 2002; Karioti et al. 2003). The substitution in position 4″ is more unusual. Apigenin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-*β*-glucopyranoside was described recently in the genus *Turnera* belonging to the family Turneraceae. This family belongs to the same order, Malphighiales, as the Euphorbiaceae family (Zhao et al. 2007). A few flavonoid *p*-coumaroylglucosides have been described in the Euphorbiaceae family (Zhang et al. 2002; Yuan et al. 2007). The substitution in position 4″ on glucose was also described in the genus *Cnidoscolus* with naringenin 7-*O*-(4″-*O*-(*Z*)-coumaroyl)-*β*-glucopyranoside or aromadendrin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-*β*-glucopyranoside (Yuan et al. 2007). From the leaves of *M. apelta*, apigenin (**82**), apigenin-7-*O*-*β*-D-glucoside (**83**), mallotusin (**84**) were isolated (Xu et al. 2006). Vicenin II (**85**) was obtained from the butanolic extract of *M. apelta* (Zhu et al. 2007).

Two new chalcone derivatives, kamalachalcone A and B (**87–88**) with a unique ring system caused by dimerization between a dimethylchromene ring and a phenoxyl group, were isolated from kamala (*Mallotus philippinensis*) (Tanaka et al. 1998). Three other novel chalcone derivatives, mallotophilippens C, D, and E (**89–91**), were isolated from the fruits of *M. philippinensis* (Daikonya et al. 2004; Li et al. 2006).

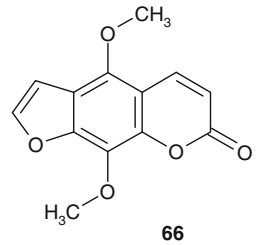
Phloroglucinol derivatives (Table 14)

Five phloroglucinol derivatives (**92–96**) were isolated from the leaves of *M. pallidus* (Supudompol et al. 2004). In 2005, a phytochemical investigation of an ethyl acetate extract of the leaves of the same species led to the isolation of a new phloroglucinol dimer, mallopallidusol (**97**) (Likhitwitayawuid and Supudompol 2005).

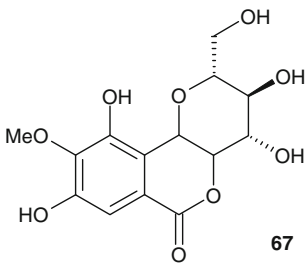
Table 11 Coumarins, isocoumarins and coumarinolignoids



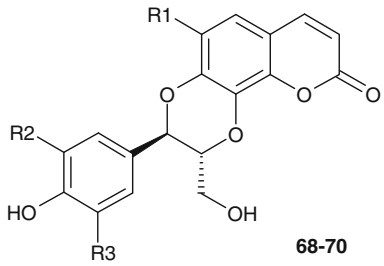
64-65



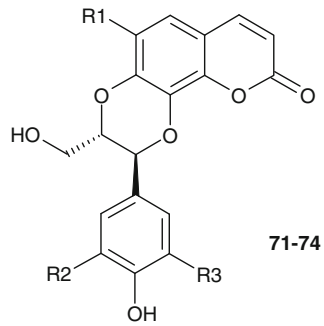
66



67



68-70



71-74

No.	Name	R1	R2	R3	Plant	Ref.
64	Scopoletin	CH_3	H		*M. resinosus*	Ma et al. (2004)
65	Isoscopoletin	H	CH_3		*M. apelta*	Kang and Lu (2007)
66	Isopimpinellin				*M. apelta*	Van Chau et al. (2005d)
67	Bergenin				*M. philippinensis*	Bandopadhyay et al. (1972)
					M. repandus	Tomizawa et al. (1976) and Huang et al. (1999)
68	Malloapelin A	OH	OH	OCH_3	*M. apelta*	Xu et al. (2008)
69	Cleomiscosin A	OCH_3	OH	OCH_3	*M. apelta*	Cheng and Chen (2000) and Xu et al. (2008)
70	Cleomiscosin B	OCH_3	H	OCH_3	*M. apelta*	Xu et al. (2008)
71	Malloapelin B	OH	OH	OCH_3	*M. apelta*	Xu et al. (2008)
72	Malloapelin C	OCH_3	OH	OCH_3	*M. apelta*	Xu et al. (2008)
73	5′-demethylaquillochin	OCH_3	H	OCH_3	*M. apelta*	Cheng and Chen (2000) and Xu et al. (2008)
74	Aquillochin	OCH_3	OCH_3	OCH_3	*M. apelta*	Cheng and Chen (2000)

Four phloroglucinol derivatives (kamalins) were isolated from *M. philippinensis*, including rottlerin (**98**) and isoallorottlerin (**99**) (Lounasmaa et al. 1975). Isorottlerin (**100**) was also mentioned in *M. philippinensis* (Zaidi et al. 2009).

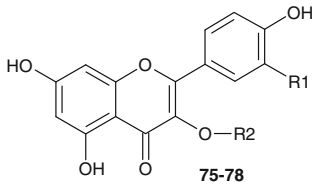
Quinones and phenolic acids (Table 15)

Chrysophanol (**101**), an anthraquinone, was isolated from the leaves of *M. apelta* (Kang and Lu 2007). Ferulic acid (**102**), a phenolic acid, was reported in *M. metcalfianus* (Rivière et al. 2009).

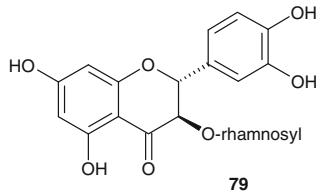
Tannins (Table 16)

A phytochemical examination of the leaves of *M. repandus* led to the isolation of four new hydrolyzable tannins, named repandusinin (**105**), repandusinic acids A and B (**106–107**) and mallotinin (**109**) together with eight other hydrolyzable tannins (**103, 108, 110–115**) and a phenolcarboxylic acid named brevifolin carboxylic acid (**104**) (Saijo et al. 1989a). 4,6,4′-Trimethyl-ellagic acid (**116**) was reported in the roots of *M. apelta* (Cheng et al. 1998).

Table 12 Flavonoids (flavonols, flavones, flavonolignans)



75-78



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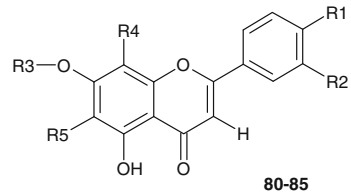

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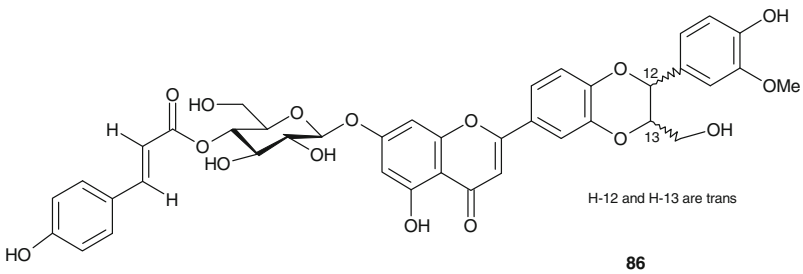

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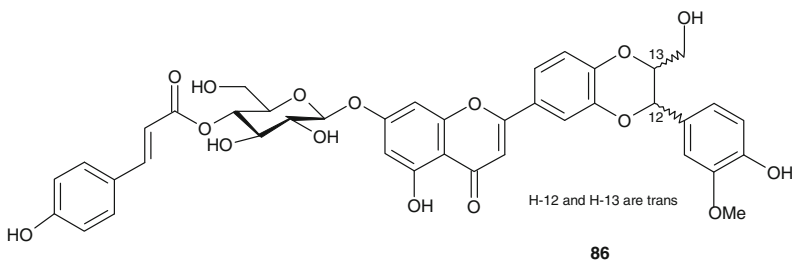


86

No.	Name	R1	R2	R3	R4	R5	Plant	Ref.
75	Quercitrin or quercetin 3-*O*-α-L-rhamnoside	OH	Rham				*M. apelta* *M. metcalfianus*	Van Chau et al. (2004) Rivière et al. (2009)
76	Kaempferol 3-*O*-α-L-rhamnose	H	Rham				*M. metcalfianus*	Rivière et al. (2009)
77	Quercetin 3-*O*-β-neohesperoside or quercetin 3-*O*-(2″-*O*-α-L-rhamnopyranosyl)-β-D-glucopyranoside	OH	Glc-Rham				*M. metcalfianus*	Rivière et al. (2009)
78	Kaempferol 3-*O*-β-neohesperoside or kaempferol 3-*O*-(2″-*O*-α-L-rhamnopyranosyl)-β-D-glucopyranoside	H	Glc-Rham				*M. metcalfianus*	Rivière et al. (2009)
79	Astilbin or dihydroquercetin 3-*O*-α-L-rhamnoside						*M. apelta* *M. metcalfianus*	Van Chau et al. (2004) Rivière et al. (2009)
80	Luteolin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside	OH	OH	Glc-coumaroyl	H	H	*M. metcalfianus*	Rivière et al. (2009)
81	Chrysoeriol 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside	OH	OCH_3	Glc-coumaroyl	H	H	*M. metcalfianus*	Rivière et al. (2009)

Table 12 continued

No.	Name	R1	R2	R3	R4	R5	Plant	Ref.
82	Apigenin	OH	H	H	H	H	*M. apelta*	Wu et al. (2006)
83	Apigenin-7-*O*-*β*-D-glucoside	OH	H	Glc	H	H	*M. apelta*	Wu et al. (2006)
84	Mallotusin or 5,7-dihydroxy-6-isopentenyl-4′-methoxy-flavanone	H	OCH_3	H	H	CH_2–CH=C(Me)$_2$	*M. apelta*	Wu et al. (2006)
85	Vicenin II	OH	H	H	C Glc	C Glc	*M. apelta*	Zhu et al. (2007)
86	Mixture of (±)-hydnocarpin 7-*O*-(4″-*O*-(*E*)-coumaroyl) *β*-glucopyranoside/(±)-hydnocarpin-D 7-*O*-(4″-*O*-(*E*)-coumaroyl) *β*-glucopyranoside						*M. metcalfianus*	Rivière et al. (2009)

Phytochemical study of the crude methanolic extract of *M. peltatus* leaves revealed the presence of tannins along with saponins, terpenoids, steroids and reducing sugars (Chattopadhyay et al. 2002a, 2003). Tannins were highlighted in the polar fractions of *M. metcalfianus*, partly responsible for the anti-radical activity of these fractions (Rivière et al. 2009). Phenolic compounds of which condensed tannins, responsible for the antioxidant activity, were quantified in several extracts of *M. philippinensis* fruits and bark and in the fractions obtained after separation from the methanolic extract of *M. philippinensis* bark on a Sephadex LH-20 column using ethanol and acetone–water as the mobile phases. The content of total phenolics in the bark extract was 541 mg/g. The content of total phenolics in the fractions ranged from 54 mg/g (fraction I) to 927 mg/g (fraction VI) and condensed tannins were detected in fractions II-VI (Arfan et al. 2007, 2009). In 1989, known tannins and related compounds were isolated from the leaves of *M. philippinensis* (Saijo et al. 1989b).

Other compounds

Unsatured fatty acids (Table 17)

Octadeca-9,12,15-trienoic acid (**117**) and octadeca-9,12,15-trienoic acid 1-*β*-D-glucopyranosyl ester (**118**) were isolated from the methanolic extract of the leaves of *M. apelta* (Van Chau et al. 2004). The seed oil of *M. paniculatus* contains long-chain fatty acids (Yu et al. 1991). Kamala (*M. philippinensis*) seed oil has been shown to contain the triply-unsatured hydroxy acid kamlolenic acid (**119**), different fatty acids and glyceride (Gupta et al. 1953).

Benzopyrans (Table 18)

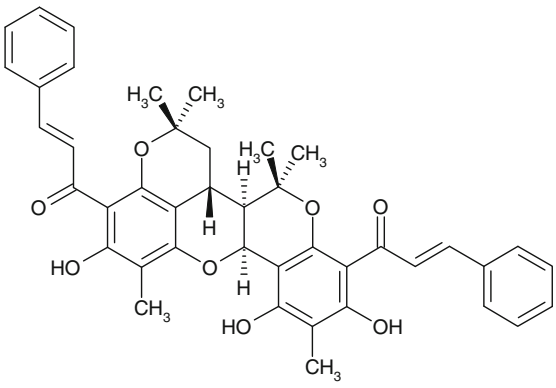
From the leaves of *M. apelta*, seven benzopyran compounds (**120–126**) were obtained in 2001 (An et al. 2001) and two new (**127–128**) in 2003 (An et al. 2003).

In 2005, two new benzopyrans (**133–134**) were isolated from the leaves of *M. apelta* by Van Kiem et al. 2005, as well as four other benzopyrans (**129–132**) by Van Chau et al. 2005a. The compound (**135**), 6-methoxy-benzopyran-4-one, was obtained from the ethyl acetate extract of the stems of *M. apelta* (Qi et al. 2005).

Various compounds (Table 19)

α-Tocopherol (**136**) was isolated from the leaves of *M. apelta* (Van Chau et al. 2005d). In 2007, nicotinic acid (**140**) was isolated from the leaves of *M. apelta* (Kang and Lu 2007). From *M. apelta*, one new pyridine type alkaloid, named malloapeltine (**142**) was isolated and structurally elucidated (Cheng et al. 1998). The methanolic extract of the aerial parts of *M. repandus* was fractionated monitored by the antiulcerogenic activity to give mallorepine (**143**), a cyano-*γ*-pyridone, together with bergenin as one of the active principles. Mallorepine may be an

Table 13 Flavonoids (chalcones)



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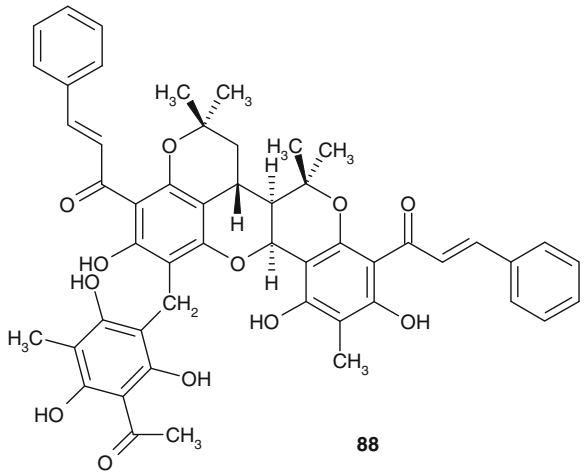

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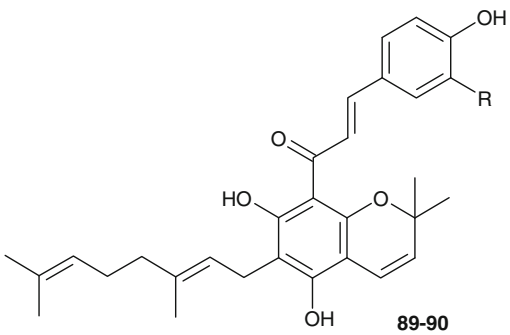

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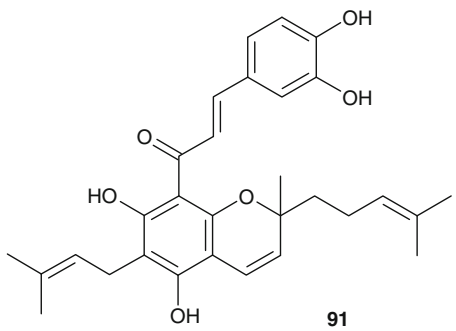
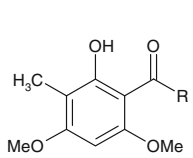


91

No.	Name	R	Plant	Ref.
87	Kamalachalcone A		*M. philippinensis*	Tanaka et al. (1998)
88	Kamalachalcone B		*M. philippinensis*	Tanaka et al. (1998)
89	1-[6-(3,7-Dimethyl-octa-2,6-dienyl)-5,7-dihydroxy-2, 2-dimoethyl-2H-chromen-8-yl]-3-(4-hydroxy-phenyl)-propenone or Mallotophilippen C	H	*M. philippinensis*	Daikonya et al. (2004), Li et al. (2006)
90	3-(3,4-Dihydroxy-phenyl)-1-[6-(3,7-dimethyl-octa-2,6-dienyl)-5, 7-dihydroxy-2,2-dimethyl-2H-chromen-8-yl]-propenone or Mallotophilippen D	OH	*M. philippinensis*	Daikonya et al. (2004), Li et al. (2006)
91	1-[5,7-Dihydroxy-2-methyl-6-(3-methyl-but-2-enyl)-2-(4-methyl-pent-3-enyl)-2H-chromen-8-yl]-3-(3,4-dihydroxy-phenyl)-propenone or Mallotophilippen E		*M. philippinensis*	Daikonya et al. (2004), Li et al. (2006)

intermediate in the biosynthetic pathway from nicotinamide to ricinine (Hikino et al. 1978). Moreover, *trans*-2-carboxy-4-hydroxytetrahydrofuran-*N*,*N*-dimethylamide (**141**), a novel furanocarboxamide, was reported in *M. cuneatus* (Groweiss et al. 1994). In 2009, we reported the isolation of a fattyl alcohol named *n*-hexacosanol (**137**), a megastigmane named blumenol-*C*-glucoside (**138**) and methyl-2-*O*-*β*-D-glucopyranosylbenzoate (**139**) from *M. metcalfianus* (Rivière et al. 2009).

Table 14 Phloroglucinol derivatives



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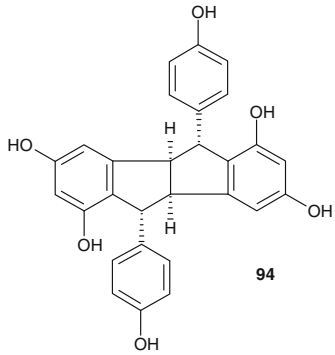

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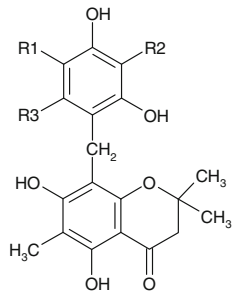

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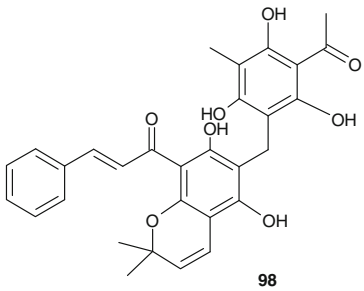

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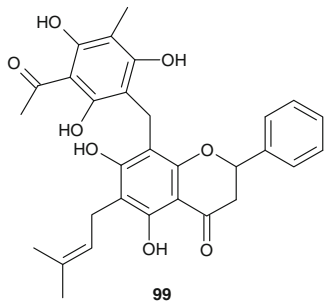

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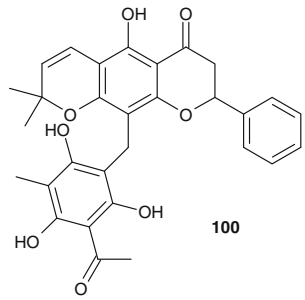


100

No.	Name	R1	R2	R3	Plant	Ref.
92	Pallidusol	Bu-i			*M. pallidus*	Supudompol et al. (2004)
93	Dehydropallidusol	$CH{=}C(Me)_2$			*M. pallidus*	Supudompol et al. (2004)
94	Pallidol				*M. pallidus*	Supudompol et al. (2004)
95	Mallopallidol	CH_3	C(=O)–Pr-i	OCH_3	*M. pallidus*	Supudompol et al. (2004)
96	Homomallopallidol	CH_3	C(=O)–CH(Me)Et	OCH_3	*M. pallidus*	Supudompol et al. (2004)
97	Mallopallidusol	C(=O)–Pr-i	CH_3	OH	*M. pallidus*	Likhitwitayawuid et al. (2005)
98	Rottlerin				*M. philippinensis*	Lounasmaa et al. (1975)
99	Isoallorottlerin				*M. philippinensis*	Lounasmaa et al. (1975)
100	Isorottlerin				*M. philippinensis*	Zaidi et al. (2009)

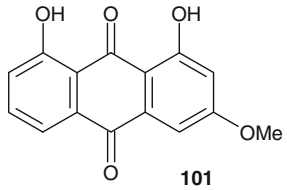
Pharmacological activities

Anti-inflammatory and immunoregulatory activities

The anti-inflammatory activity of the methanolic extract of *M. peltatus* leaves against carrageenan (acute model) and dextran-induced (subacute model) rat paw oedema and cotton pellet-induced granuloma (chronic model) in rats were studied using indomethacin as standard. The methanolic extract of this species at 200 and 400 mg/kg, and two *n*-butanolic fractions (A and B) at 25 mg/kg, exhibited significant anti-inflammatory activity in Albino rats, compared with indomethacin. Further study with fractions showed that the anti-inflammatory activity is due to either fraction A, ursolic acid (**46**), alone or the combination of fractions A and B, *β*-sitosterol (**58**) and fatty acids (Chattopadhyay et al. 2002a). The methanolic extract of *M. peltatus* showed also a significant dose-dependent anti-inflammatory and antioxidant activity at nontoxic concentrations (Chattopadhyay et al. 2006).

The chalcones isolated from the fruits of *M. philippinensis*, mallotophilippens C, D and E (**89–91**)

Table 15 Quinones and phenolic acids



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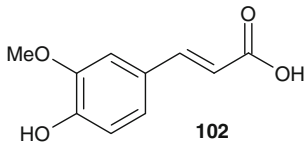


102

No.	Name	Plant	Ref.
101	Chrysophanol	*M. apelta*	Kang and Lu (2007)
102	Ferulic acid	*M. metcalfianus*	Rivière et al. (2009)

inhibited nitric oxide (NO) production and inducible NO synthase (iNOS) gene expression by a murine macrophage-like cell line (RAW 264.7), which was activated by lipopolysaccharide and recombinant mouse interferon-γ (IFN-γ). Furthermore, they down-regulated cyclooxygenase-2 gene, interleukin-6 gene and interleukin-1β gene expression. These results suggest that these chalcones have anti-inflammatory and immunoregulatory effects (Daikonya et al. 2004).

The chloroform extract from the roots of *M. spodocarpus* was investigated for anti-inflammatory and analgesic activities in animal models. The results obtained suggest marked anti-inflammatory and analgesic activity of the extract. In acute inflammatory models, the extract significantly inhibited ethyl phenylpropiolate-induced ear oedema and carrageenin- and arachidonic acid-induced hind paw oedema in rats. In the chronic inflammatory model using the cotton pellet-induced granuloma in rats, the extract exhibited inhibitory activity on the formation of granuloma. The extract also elicited pronounced inhibitory effect on acetic acid-induced writing response in mice in the analgesic test (Intahphuak et al. 2004).

Antifertility activity

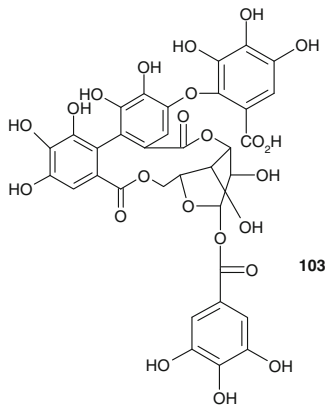
The Kamala (*M. philippinensis*) seeds extract presents adverse effects on various reproductive parameters of female rats. The data indicate that Kamala reduced serum FSH and LH levels probably by affecting hypothalamic/pituitary axis in treated animals. Thus, reduced levels of FSH and LH and estradiol might have affected the follicular development, quality of ovulated eggs, corpora lutea formation, estrus cycle, establishment and maintenance of pregnancy in treated rats (Thakur et al. 2005). The antifertility effect of this species seems to be caused by rottlerin (**98**); a phloroglucinol derivative. Acetylrottlerin is also active, but isorottlerin (**100**) is either inactive or only slightly active (Gujral et al. 1960).

Antimicrobial activity

Among seven benzopyrans obtained from the leaves of *M. apelta*, one compound (**120**) showed moderate antibiotic activity against *Micrococcus lutens* (An et al. 2001). Moreover, erythrodiol-3-acetate (**42**), β-sitosterol (**58**), 3β,29-dihydroxylupane (**27**) and ursolic acid acetate (**47**) isolated from the roots of *M. apelta* possess some bacteriostatic activities on *Staphylococcus aureus*, *Bacillus subtilis*, *Escherichia coli*, and *Bacillus pyocyaneum* (Shan et al. 1985).

The antimicrobial activity of several fractions of *M. metcalfianus* was evaluated on 20 strains. This activity was moderate: fractions were not active on some Gram negative bacteria at the highest concentrations tested (1,000 μg/ml) but were effective on at least eight strains at 500 μg/ml (MAC, minimal active concentration, the minimal concentration reducing the growth of the microorganism as compared to controls), i.e., on Gram positive bacteria (*Staphylococcus aureus* ATCC 25923, *Enterococcus faecalis* ATCC 29212), on Gram negative bacteria (*Escherichia coli* ATCC 25922, *Pseudomonas aeruginosa* ATCC 27853, *Morganella morganii* 180, *Yersinia enterocolitica* E 170/98, *Yersinia enterocolitica* E 169/98) and on Saccharomycetes fungi (*Candida albicans*). Some MAC were as low as to

Table 16 Tannins



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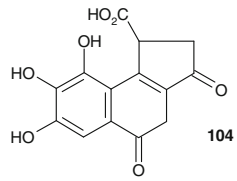

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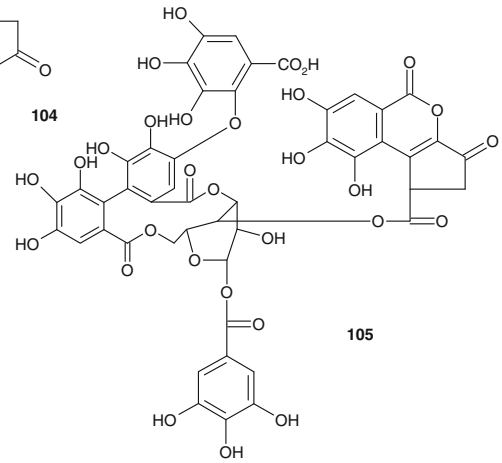

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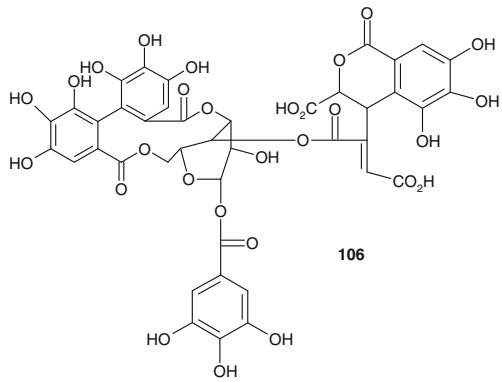

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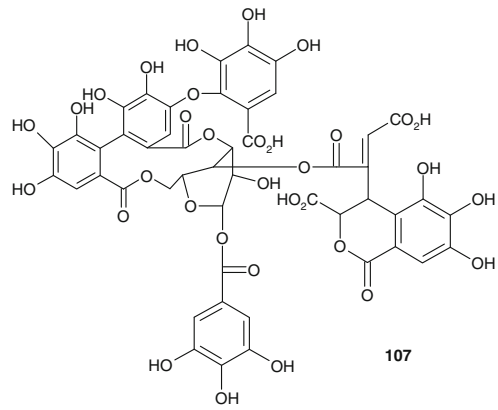

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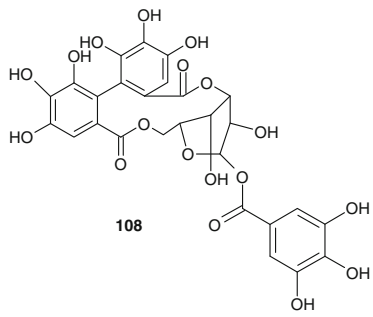

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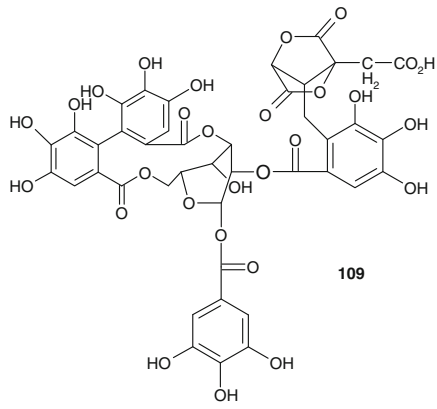

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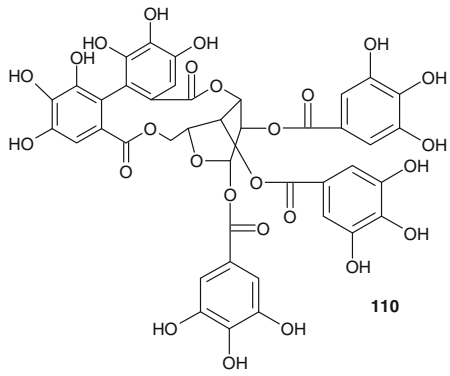

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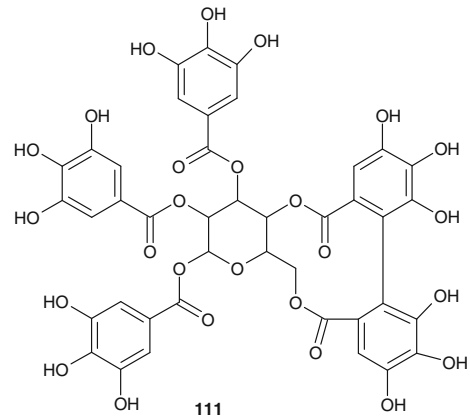

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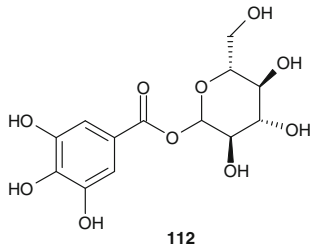

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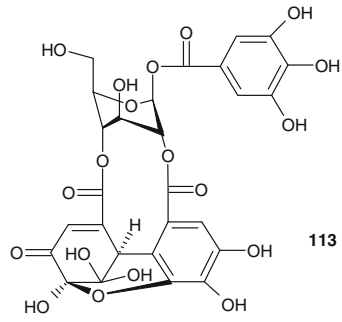

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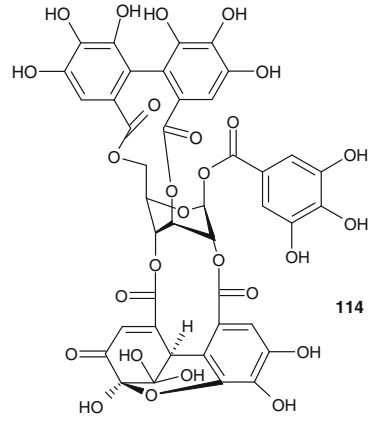

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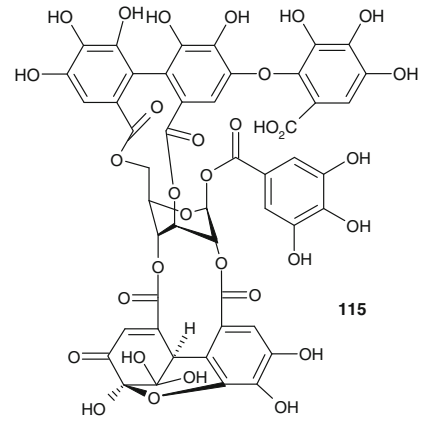

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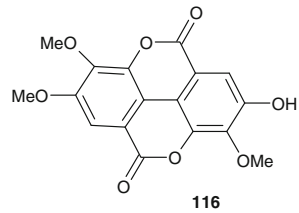


116

Table 16 continued

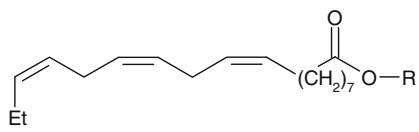
No.	Name	Plant	Ref.
103	Mallotinic acid	*M. repandus*	Saijo et al. (1989a)
104	Brevifolin carboxylic acid	*M. repandus*	Saijo et al. (1989a)
105	Repandusinin	*M. repandus*	Saijo et al. (1989a)
106	Repandusinic acid A	*M. repandus*	Saijo et al. (1989a)
107	Repandusinic acid B	*M. repandus*	Saijo et al. (1989a)
108	Corilagin	*M. repandus*	Saijo et al. (1989a)
109	Mallotinin	*M. repandus*	Saijo et al. (1989a)
110	Punicafolin	*M. repandus*	Saijo et al. (1989a)
111	Eugeniin	*M. repandus*	Saijo et al. (1989a)
112	Glucogallin	*M. repandus*	Saijo et al. (1989a)
113	Furosin	*M. repandus*	Saijo et al. (1989a)
114	Geraniin	*M. repandus*	Saijo et al. (1989a)
115	Mallotusinic acid	*M. repandus*	Saijo et al. (1989a)
116	4,5,4′-Trimethyl-ellagic acid	*M. apelta*	Cheng et al. (1998)

200 μg/ml. This activity in most cases (polar extracts) may be explained at least partly by the presence of tannins as minimal inhibitory concentration (MIC) increases after their removal. Hexanic and some chloroformic fractions show also an interesting activity. Pure isolated major flavonoids, quercitrin (**75**), kaempferol 3-*O*-α-L-rhamnoside (**76**) and astilbin (**79**), have a moderate activity (MIC = 128 μg/ml on some strains) (Rivière et al. 2009).

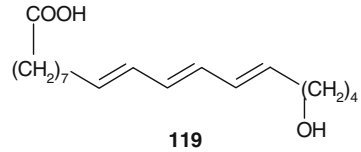
The crude methanolic extract of *M. peltatus* leaves was found to be active against *Staphylococcus aureus*, *Staphylococcus saprophyticus*, *Streptococcus faecalis*, *Bacillus subtilis*, *Escherichia coli*, and *Proteus mirabilis* and the dermatophytic fungi *Microsporum gypseum*. The minimum inhibitory concentration (MIC) ranges from 128 to 2,000 μg/ml for bacteria and 128 mg/ml for fungi, while the minimum bactericidal concentration (MBC) was twofold to fourfold higher than MIC. The methanol–water fraction of the extract showed similar activity against *Staphylococcus*, *Streptococcus*, *Bacillus*, and *Proteus* isolates. The fraction A, ursolic acid (**46**), alone or the combination of fractions A and B, β-sitosterol (**58**) and fatty acids, are responsible for the antimicrobial and anti-inflammatory activities (Chattopadhyay et al. 2002a). The methanolic extract of *M. peltatus* showed also an antibacterial activity at 64–1,000 μg/ml (Chattopadhyay et al. 2006).

A series of 61 Indian medicinal plants belonging to 33 different families used in various infectious disorders, were screened for their antimicrobial properties. On the basis of the results obtained, the crude extract of *M. philippinensis* exhibited significant antimicrobial activity (Kumar et al. 2006). *M. philippinensis* var. *tomentosus* was tested against *Escherichia coli*, *Klebsiella pneumoniae*, *Pseudomonas aeruginosa*, *Salmonella typhi*, and *Bacillus subtilis*. From the results obtained, the chloroformic fractions and the methanolic extract showed zones of inhibition comparable to the standard drug used. However, the hexanic extract did not show any appreciable activity. The results of the study may justify the use of the plant against bacterial pathogens (Moorthy et al. 2007). Moreover, in the quest for potent anti-*Helicobacter pylori* agents, ethanolic extract of *M. philippinensis* showed a strong bactericidal activity at the concentration of 15.6–31.2 mg/l against eight *H. pylori* strains. Further fractionation and purification of this extract led to the isolation of five compounds. Among the isolated compounds, rottlerin (**98**), exhibited the most potent bactericidal activity with a minimal bactericidal concentration (MBC) value of 3.12–6.25 mg/l against several clinical *H. pylori* isolates including Japanese and Pakistani strains, nine clarithromycin resistant (CR), and seven metronidazole resistant (MR) strains. This

Table 17 Unsatured fatty acids



117-118



119

No.	Name	R	Plant	Ref.
117	9,12,15-Octadecatrienoic acid or linolenic acid	H	*M. apelta*	Van Chau et al. (2004)
118	9,12,15-Octadecatrienoic acid 1β-D-glucopyranosyl ester	Glc	*M. apelta*	Van Chau et al. (2004)
119	Kamlolenic acid		*M. repandus*	Gupta et al. (1953)

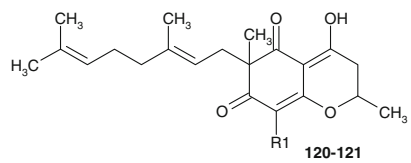
study thus revealed the potent in vitro anti-*H. pylori* activity of the ethanolic extract and of rottlerin, specially against CR and MR strains, which could be gainfully utilized for the development of novel antimicrobials to prevent *H. pylori* related disorders (Zaidi et al. 2009).

Antioxidant, antiradical activity

From our phytochemical results, *M. metcalfianus* is rich in flavonoids and phenolic compounds. These flavonoids were mainly present in the ethyl acetate extract while the aqueous fraction and the residue were richer in tannins. Concerning the antiradical activity, for the ethyl acetate fraction which was the most active, we observed that tannins were only responsible for a small part of the activity which seems to be mainly due to flavonoids. In fact, the elimination of tannins in this fraction only slightly decreased the antiradical properties. On the contrary, tannins seem to be responsible for a large part of the antioxidant activities of the residue and the aqueous fraction: their elimination greatly decreased the activity. We tested the different pure compounds isolated from *M. metcalfianus* and different reference samples (flavonoids and cinnamic acid derivatives) for their antiradical activities in order to discuss about structure–activity relationships of these products. We observed that quercetin 3-*O*-β-neohesperidoside (**77**) shows about 50% of the activity of rutin. This decrease in activity can be due to the different position of rhamnose on glucose. Kaempferol 3-*O*-β-neohesperidoside (**78**), having an OH less on the B ring, shows a very moderate activity. The new flavonolignans (**86**) were not very active. This lack of activity could be explained by the cyclization of the catechol group of the B ring of the flavone. Indeed, by comparison with luteolin, luteolin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside) (**80**) was found to be moderately active in the DPPH assay. The substitution of the flavone by a coumaric acid could explain this decrease of activity, as coumaric acid does not show a real antioxidant activity unlike caffeic acid. Chrysoeriol 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside) (**81**) is less active than luteolin 7-*O*-(4″-*O*-(*E*)-coumaroyl)-β-glucopyranoside), probably because of the loss of phenol function in position 3′, replaced by a methoxy group. *n*-Hexacosanol (**137**), blumenol C glucoside (**138**), methyl 2-*O*-β-D-glucopyranosylbenzoate (**139**) and friedelinol (**34**) were found to have only a low activity. Friedelin (**33**) (hydroxyl function of friedelinol in position three replaced by a ketone) was not more active than friedelinol (Rivière et al. 2009).

The total antioxidant activity (TAA), antiradical activity against DPPH and reducing power of several extracts of *M. philippinensis* fruits and bark and of the fractions, obtained after separation of the methanolic extract bark on a Sephadex LH-20 column using ethanol and acetone–water as the mobile phases, were evaluated. The extract of the bark showed the strongest antiradical activity and reducing power; its TAA was 5.27 mmol Trolox equiv./g. The TAA of other extracts ranged from 0.05 to 1.79 mmol Trolox equiv./g. The TAA of phenolic fractions of *M. philippinensis* bark extract ranged from 0.58 mmol Trolox/g (fraction I) to 6.82 mmol Trolox/g (fraction IV). Fraction IV also showed the strongest antiradical activity against DPPH and reducing power (Arfan et al. 2007, 2009).

Table 18 Benzopyrans



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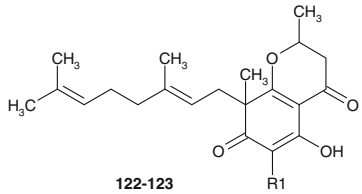
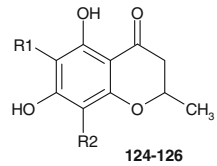
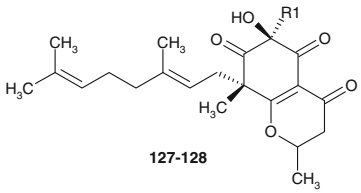


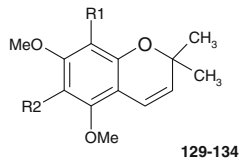
122-123



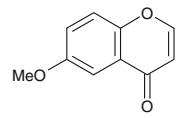
124-126



127-128



129-134



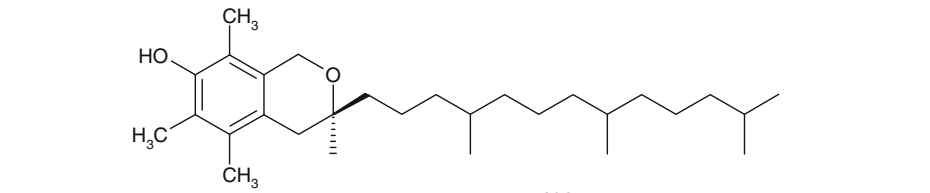
135

No.	Name	R1	R2	Plant	Ref.
120	4-Hydroxy-2,6-dimethyl-6-(3,7-dimethyl-2,6-octadienyl)-8-(3-methyl-2-butenyl)-2H-1-benzopyran-5,7(3H,6H)-dione	CH_2–CH=C(Me)$_2$		*M. apelta*	An et al. (2001)
121	4-Hydroxy-2,6,8-trimethyl-6-(3,7-dimethyl-2,6-octadienyl)-2H-1-benzopyran-5,7(3H,6H)-dione	CH_3		*M. apelta*	An et al. (2001)
122	5-Hydroxy-2,8-dimethyl-6-(3-methyl-2-butenyl)-8-(3,7-dimethyl-2,6-octadienyl)-2H-1-benzopyran-4,7(3H,8H)-dione	CH_2–CH=C(Me)$_2$		*M. apelta*	An et al. (2001)
123	5-Hydroxy-2,6,8-trimethyl-8-(3,7-dimethyl-2,6-octadienyl)-2H-1-benzopyran-4,7(3H,8H)-dione	CH_3		*M. apelta*	An et al. (2001)
124	2,3-Dihydro-5,7-dihydroxy-2,6-dimethyl-8-(3-methyl-2-butenyl)-4H-1-benzopyran-4-one	CH_3	CH_2–CH=C(Me)$_2$	*M. apelta*	An et al. (2001)
125	2,3-Dihydro-5,7-dihydroxy-2,8-dimethyl-6-(3-methyl-2-butenyl)-4H-1-benzopyran-4-one	CH_2–CH=C(Me)$_2$	CH_3	*M. apelta*	An et al. (2001)
126	2,3-Dihydro-5,7-dihydroxy-2,6,8-trimethyl-4H-1-benzopyran-4-one	CH_3	CH_3	*M. apelta*	An et al. (2001)
127	6-Hydroxy-2,6,8-trimethyl-8-(3,7-dimethyl-2,6-octadienyl)-2H-1-benzopyran-4,5,7(3H,6H,8H)-trione	CH_3		*M. apelta*	An et al. (2003)
128	6-Hydroxy-2,8-dimethyl-6-(3-methyl-2-butenyl)-8-(3,7-dimethyl-2,6-octadienyl)-2H-1-benzopyran-4,5,7(3H,6H,8H)-trione	CH_2–CH=C(Me)$_2$		*M. apelta*	An et al. (2003)
129	8-(1′-Oxo-2′-en-butyl)-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran or malloapelta B	CO–CH=CH–CH_3	H	*M. apelta*	Van Chau et al. (2005a)
130	8-(1′-Oxo-3′(*R*)-hydroxy-butyl)-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran	CO–$CH_2CH(CH_3)OH$	H	*M. apelta*	Van Chau et al. (2005a)
131	8-(Acetic acid 1′-oxo-3′(*R*)-hydroxy-butyl ester)-5,7-dimethoxy-2,2-dimethyl-2H-1- benzopyran	CO–$CH_2CH(CH_3)OAc$	H	*M. apelta*	Van Chau et al. (2005a)
132	6-(1′-Oxo-2′-en-butyl)-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran	H	CO–CH=CH–CH_3	*M. apelta*	Van Chau et al. (2005a)
133	6-(1′-Oxo-3′(*R*)-hydroxy-butyl)-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran	H	CO–$CH_2CH(CH_3)OH$	*M. apelta*	Van Kiem et al. (2005)

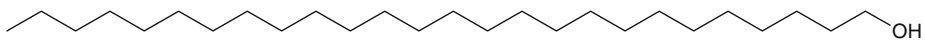
Table 18 continued

No.	Name	R1	R2	Plant	Ref.
134	6-(1′-Oxo-3′(*R*)-methoxy-butyl)-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran	H	CO–$CH_2CH(CH_3)OCH_3$	*M. apelta*	Van Kiem et al. (2005)
135	6-Methoxy-benzopyran-4-one			*M. apelta*	Qi et al. (2005)

Table 19 Various compounds



136



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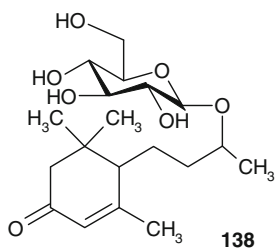
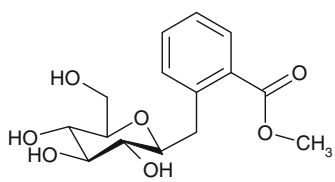
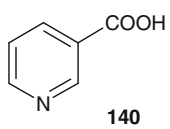


138



139



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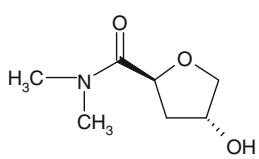

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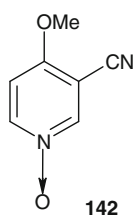
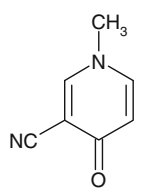


142



143

No.	Name	Plant	Ref.
136	α-Tocopherol	*M. apelta*	Van Chau et al. (2005d)
137	*n*-Hexacosanol	*M. metcalfianus*	Rivière et al. (2009)
138	Blumenol C glucoside	*M. metcalfianus*	Rivière et al. (2009)
139	Methyl-2-*O*-*β*-D-glucopyranosylbenzoate	*M. metcalfianus*	Rivière et al. (2009)
140	Nicotinic acid	*M. apelta*	Kang and Lu (2007)
141	Trans-2-carboxy-4-hydroxytetrahydro furan-*N*,*N*-dimethylamide	*M. cuneatus*	Groweiss et al. (1994)
142	4-Methoxy-3-cyano-pyridine 1-oxide or malloapeltine	*M. apelta*	Cheng et al. (1998)
143	Mallorepine	*M. repandus*	Hikino et al. (1978)

The ethyl acetate fraction of *M. repandus* stems showed the greatest superoxide-scavenging activity, and the hexanic extract of stems and roots had the greatest hydroxyl-scavenging activity (Lin et al. 1995).

As phenolic compounds have been identified in several *Mallotus* species and because of the health interest of antioxidant extracts or compounds (prevention of cancers, anti-inflammatory properties) (Tapiero et al. 2002; Soobrattee et al. 2006), we determined the antiradical activity of 33 samples (methanolic extracts) of seventeen *Mallotus* species from Vietnam by the DPPH assay. Some species were collected in different provinces. For some species, different parts of the plant were studied. The most effective methanolic extracts come from *Mallotus barbatus* MA29, *Mallotus cuneatus* MA17, *Mallotus floribundus* MA15, *Mallotus hookerianus* MA22, *Mallotus nanus* MN37R, MN37L, and MN39C, *Mallotus oblongifolius* MA14, *Mallotus paniculatus* MP35R, and *Mallotus philippinensis* MA28. According to the literature and what is known about their chemical compositions, antioxidant activities of *Mallotus nanus*, *M. paniculatus*, *M. philippinensis* could be explained by the presence of flavonoids and tannins. We noted that some extracts have an antiradical activity similar to tocopherol. They thus represent valid alternative sources of antioxidant agents, as we also showed that they did not show cytotoxicity on cultured cells. Combining fingerprint technology with data-handling techniques allows indicating the peaks potentially responsible for given activities. We indicated from chromatographic fingerprints the peaks potentially responsible for the antioxidant activity of these *Mallotus* species. Relevant information was extracted using linear multivariate calibration techniques (Nguyen Hoai et al. 2009; Tistaert et al. 2009).

Antipyretic activity

The leaf extract of *M. peltatus* showed a potential anti-pyretic effect in rats. At oral doses of 100, 200, and 300 mg/kg, the extract showed significant reduction in normal body temperature and yeast-provoked elevated temperature in a dose-dependent manner and the anti-pyretic effect was comparable to that of standard anti-pyretic agent paracetamol (150 mg/kg). The effect also extended up to 5 h after the drug administration (Chattopadhyay et al. 2002b).

Antiulcerogenic activity

The methanolic extract of the aerial parts of *M. repandus* was fractionated monitored by the antiulcerogenic activity to give mallorepine (**143**), together with bergenin (**67**) as one of the active principles. Mallorepine was shown to be inactive in inhibiting the formation of the stress-induced gastric ulcers (Hikino et al. 1978).

Antiviral activity

In 1989, 40 preparations of extracts from 28 kinds of Asian herbs were tested for their ability to inhibit the activities of murine retroviral reverse transcriptase and human DNA polymerases. Among the 40 extracts, very strong inhibitions were observed with the extract from *M. apelta* as shown by its low IC_{50} values for reverse transcriptase (0.4–0.5 μg/ml) and DNA polymerase-α (0.9–1.4 μg/ml). The mode of inhibition of reverse transcriptase by this extract was competitive with respect to the template-primer [poly(rA)-oligo(dT)] and noncompetitive with respect to dTTP substrate. Besides reverse transcriptase and DNA polymerase-α, DNA polymerase I and RNA polymerase from *Escherichia coli* were inhibited by this extract (Ono et al. 1989). In 2002, a massive screening of natural products showed also that *M. apelta* has significant anti-HIV activity. From this species, thirty compounds have been isolated and structurally elucidated. The most interesting and promising compounds for further study were terpenoids, pyridine type alkaloids; cerebrosides and three coumarinolignoid compounds. The coumarinolignoids have been proved to be the most active compounds against HIV (Cheng and Chen 2002). The root of *M. apelta* has therapeutic effect on duck hepatitis B virus (D-HBV). It can restrain the duplication of D-HBV in vivo. Although this effect is weaker than that of lamivudine, it lasts longer (Xu et al. 2006).

M. chrysocarpus is reported to have potential anti-HIV activity (Nguyen et al. 1997).

Five phloroglucinol derivatives isolated from *M. pallidus* were studied for their inhibitory effects against herpes simplex virus HSV-1, HSV-2, and human immunodeficiency virus HIV-1. The data obtained in this study suggest the bis-hydroxyphenyl structure as a potential lead for anti-HSV

and anti-HIV drugs development (Likhitwitayawuid et al. 2005).

The inhibitor of human immunodeficiency virus type-1 reverse transcriptase (HIV-1-RT) isolated from an aqueous extract of *Phyllanthus niruri* was purified and identified as repandusinic acid A monosodium salt (**106**), an hydrolyzable tannin, which was originally isolated from *Mallotus repandus*. The 50% inhibitory doses (ID_{50}) of this compound on HIV-1-RT and DNA polymerase-α (from HeLa cells) were 0.05 and 0.6 μM, respectively, representing approximatively a ten-fold higher sensitivity for HIV-1-RT compared to DNA polymerase α. This tannin was shown to be a competitive inhibitor with respect to the template-primer while it was a noncompetitive inhibitor with respect to the substrate (Ogata et al. 1992).

Cytotoxic and antitumor activities

In 2005, two benzopyrans isolated from the leaves of *M. apelta* showed a cytotoxic activity. The benzopyran, 6-[1′-oxo-3′(*R*)-hydroxy-butyl]-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran (**133**), was found to have a strong cytotoxic effect against two human cancer cell lines, human hepatocellular carcinoma (Hep-2, $IC_{50} = 0.49$ μg/ml) and rhabdosarcoma (RD, $IC_{50} = 0.54$ μg/ml), while the benzopyran, 6-[1′-oxo-3′(*R*)-methoxy-butyl]-5,7-dimethoxy-2,2-dimethyl-2H-1-benzopyran (**134**), showed moderate activity against the Hep-2 cell line ($IC_{50} = 4.22$ μg/ml) by in vitro assay (Van Kiem et al. 2004). In searching for bioactive compounds from natural products by analyzing their cytotoxic effects against various cancer cell lines, 22 compounds isolated from *M. apelta* were tested for their cytotoxic effects against various cancer cell lines, such as KB (human epidermoid carcinoma), FL (fibrillary sarcoma of the uterus), and Hep-2 (human hepatocellular carcinoma) cells in an in vitro assay system. Malloapelta B (**129**), a benzopyran, showed strong cytotoxic effect against the three cancer cell lines, while the other compounds did not show inhibitory activities and had IC_{50} values over 50 μM (Van Chau et al. 2005a).

Two antitumor agents, AK-3A [62534-39-8] and AK-3B [62534-40-1] were isolated from the leaves, bark and xylem of *M. repandus* (Kawashima et al. 1976b).

DNA cleavage activity

A crude extract prepared from roots of *M. resinosus* exhibited significant Cu^{2+}-dependent DNA strand scission activity and was thus selected for bioassay-guided fractionation. Scopoletin (**64**), a simple coumarin, was identified as the active principle responsible for the DNA cleavage activity of the crude extract (Ma et al. 2004).

Hepatoprotective activity

An extract from the roots of *M. apelta* could reduce the progression of liver fibrosis, having a capacity of anti-oxidation (Zhao et al. 2002). Malloapelin C (**72**), a coumarinolignoid isolated from *M. apelta*, showed promising hepatoprotective activity against D-galactosamine-induced toxicity in WB-F344 rat hepatic epithelial stem-like cells (Xu et al. 2008).

One hundred twenty-nine samples of Taiwanese plants were screened for antihepatotoxic activity in primary cultured hepatocytes, against cytotoxicity produced by carbon tetrachloride and D-galactosamine. *M. repandus* belongs to the plants which disclosed significant antihepatotoxic activity in both methods (Yang et al. 1987).

Inhibition of proteins implicated in cancer process

In searching for inhibitory components from natural products on NFAT transcription factor and NF-κB activation, the methanolic extract from the leaves of *M. apelta* has been investigated. Fourteen compounds were isolated. Of these compounds, malloapelta B (**129**) exhibited also a strong activity against the NFAT transcription factor and inhibition of NF-κB activation (Van Chau et al. 2005d).

Rottlerin (**98**), a compound isolated from *M. philippinensis*, is shown to inhibit protein kinases with some specificity for PKC. To some extent, the novel inhibitor is able to differentiate between PKC isoenzymes, with IC_{50} values for PKC δ of 3–6 μM, PKC α,β,γ of 30–42 μM and PKC υ,η,ζ of 80–100 μM. Inhibition of PKC appears, at least in part, to be due to a competition between rottlerin and ATP. Among the protein kinases tested, only CaM-kinase III is suppressed by rottlerin as effectively as PKC δ. The chemical structure of rottlerin might serve as a

basis for the development of novel inhibitors with improved selectivity for a distinct PKC isoenzyme, such as PKC δ, or for CaM-kinase III (Gschwendt et al. 1994; Liao et al. 2005).

Neuropharmacological activity

The methanolic extract and different fractions of *M. peltatus* leaves showed several neuropharmacological effects in rats and mice. The results revealed that the crude extract at 200–300 mg/kg and its fractions A and B at 50 mg/kg caused a significant reduction in spontaneous activity, remarkable decrease in exploratory behavioral pattern, a reduction in muscle relaxant activity and also a significantly potentiated phenobarbitone sodium-induced sleeping time. Further fractionation and purification yielded two major fractions A, ursolic acid (**46**), and B, β-sitosterol (**58**) with some fatty acids, as major compounds. The psychopharmacological activity of the crude leaf extracts appeared to be either due to fraction A (50 mg/kg) or a combination of fractions A and B (50 mg/kg) along with some fatty acids present in the *n*-butanolic part of methanolic extract of *M. peltatus* leaf (Chattopadhyay et al. 2003).

Uterus muscle stimulant

A compound stimulating the uterus muscles was isolated from the methanolic fraction of *M. repandus* (Kawashima et al. 1975).

Veterinary applications

A survey was conducted in southern Punjab, Pakistan, in order to document existing ethnobotanical knowledge by the herdsmen/key respondents about anthelmintics in ruminants. *M. philippinensis* is one of the main plants used (Jabbar et al. 2006). The fruits of *M. philippinensis* showed a gastrointestinal anticestodal activity in Beetal goats (Akhtar and Ahmad 1992).

Conclusions

The results of this review confirm the great potential of *Mallotus* species. For many of them still only very limited information is available. It leads us to continue studies on certain *Mallotus* species which showed interesting pharmacological properties, to identify the compounds responsible for these activities.

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