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CHEMISTRY OF SOME SPECIES GENUS *LANTANA*

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Abstract

This review presents the currently known phytochemical constituents of the genus *Lantana*; a total of 160 natural compounds have been included. *Lantana* is free from diterpenoids and rich in essential oils. Monoterpenes, triterpenes, flavones coumarin, steroids, iridoid glycosides, and caffeic acid derivatives are reported from *Lantana*. Triterpenes and flavones are the more common secondary metabolites in *Lantana*. *Lantana* plants are used in folk medicine in many parts of the world. The genus *Lantana* also produces a number of metabolites in high quantities and some have been shown to possess useful biological activities. The ethnopharmacology, phytochemistry and toxicity of *Lantana* are considered. The biological activities exhibited by some of the metabolites are discussed and the results of recent studies on the triterpene inhibitors of human α-thrombin discovered in this plant are presented.

Introduction

The genus *Lantana* (Verbenaceae), as described by Linnaeus in 1753, contained seven species, six from South America and one from Ethiopia (Munir, 1996). *Lantana* (from .the Latin *lento*, to bend) probably derives from the ancient Latin name of the genus *Viburnum*, which it resembles a little in foliage and inflorescence. *Lantana* is mostly native to subtropical and tropical America, but a few taxa are indigenous to tropical Asia and Africa. It now occurs in approximately 50 countries, where several species are cultivated under hundreds of cultivar names. The recorded number of *Lantana* species varies from 50 to 270 specific and subspecific entities, but it appears that a better estimate is 150 species (Atkin, 2004). The genus is a difficult one to classify taxonomically since species are not stable and hybridisation is widespread, the shape of inflorescence changes with age, and flower colours vary with age and maturity (Munir, 1996). In this review, the ethnopharmacology, phytochemistry and toxicity of *Lantana* species are considered. The biological activities exhibited by some of the metabolites are discussed and the results of recent studies on the triterpene inhibitors of human α-thrombin discovered in this genus are presented.

***Lantana indica*:** *L. indica* Roxb. is a shrub native to India, where it has been used as a sudorific, intestinal antiseptic, diaphoretic and in the treatment of tetanus, rheumatism and malaria in the Ayurvedic system of medicine (Singh *et al*., 1991). Steam distilled essential oil obtained from the leaves of *L. indica* was analyzed by capillary gas chromatography (GC) and gas chromatography mass spectrometry (GC/MS). These chemical compounds identified from the oil were trans-caryophyllene (1), α-selinene (2), globulol (3), trans-caryophyllene oxide (4), α-guaiene (5), valencene (6), humulene (7), and β-eudesmene (8) (Fig. 1) (Akhtar *et al*., 2006). Antifungal activity of the following triterpenoids from *L indica* root was studied: oleanolic acid (9), 3-ketooleanolic acid (10), (+)-24-hydroxy-3-oxoolean-12-en-28-oic acid (11), 3β,24-dihydroxyolean-12-en-28-oic acid (12), and 3,24-dioxo-olean-12-en-28-oic acid (13) (Fig. 1) (Verma *et al*., 1998; Singh *et al*., 1990; Singh *et al*., 1991). They concluded that these compounds had no antifungal activity.

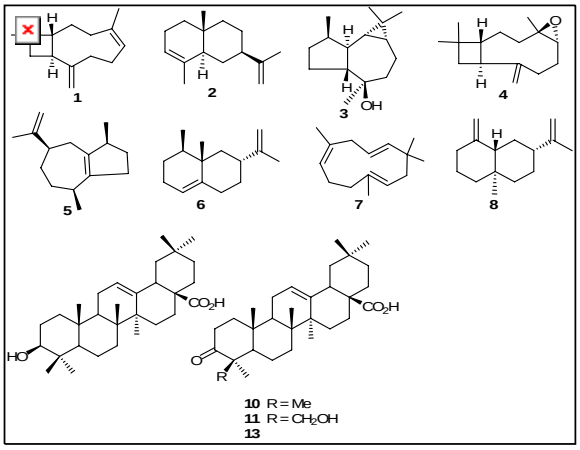

Fig. 1. Compounds 1-13 isolated from of *L. indica*.

***Lantana involucrata*:** In a study of novel antitumor agents from rainforest plants, three new isopropenylfurano-β-naphthoquinones, designated lantalucratins A (14), B (15), and C (16), and three new isoprenyl-α-naphthoquinones, designated lantalucratins D (17), E (18), and F (19), were isolated from *L. involucrata* (Fig. 2). Lantalucratins A (14) and B (15) showed cytotoxic activities against various human tumor cell lines, including drug-resistant variants, with IC_{50} values of 1.0-4.9 μM (Hayashi *et al*., 2004).

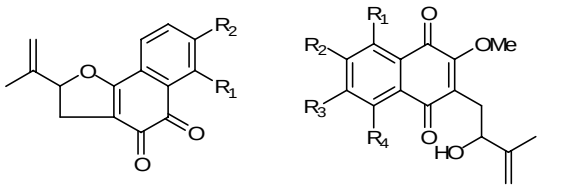

Fig. 2. Compounds 14-19 isolated from *L. involucrata.*

Lantana montevidensis: The flavonoid fraction from the leaves of *L. montevidensis* (Spreng.) Briq., showed antiproliferative activity against human gastric adenocarcinoma (MK-1, GI50: 12 μg/ml), human uterus

carcinoma (HeLa, 5 µg/ml), and murine melanoma (B16F10, 5 µg/ml) cells *In vitro*. Bioactivity-guided chemical investigation of the fraction resulted in the isolation of apigenin (20) and ten 5, 6, 7-oxygenated flavones: cirsilineol (21), eupatorin (22), 5, 4'-dihydroxy-6, 7, 3' ,5'-tetramethoxyflavone (23), 5, 6-dihydroxy-7, 3', 4'-trimethoxyflavone (24), 5, 6, 4'-trihydroxy-7, 3', 5'-trimethoxyflavone (25), 5, 6, 3'-trihydroxy-7, 4'-dimethoxyflavone (26), 5, 3', 4'-trihydroxy-6, 7, 5'-trimethoxyflavone (27), cirsiliol (28), hispidulin (29), and eupafolin (30) (Nagao *et al*., 2002). Similarly, Wollenweber *et al*., (1997) reported another three flavones, luteolin (31) 7, 3'-dimethoxy- (32) and 7, 3', 4'-trimethoxyluteolin (33), from *L. montevidensis* (Fig. 3).

Lantana tiliifolia: *L. tiliifolia* Cham. is the most common *Lantana* in Brazil, but it is not considered a weed because it is controlled by a complex of natural predators such as insects and fungi (Rwangabo *et al*., 1988). A new triterpene, 24-hydroxy-3-oxours-12-en-28-oic acid (34), was reported together with the known triterpenes ursonic acid (35), oleanonic acid (10), ursolic acid (36), and oleanolic acid (9) (Johns *et al*., 1983a) (Fig. 4).

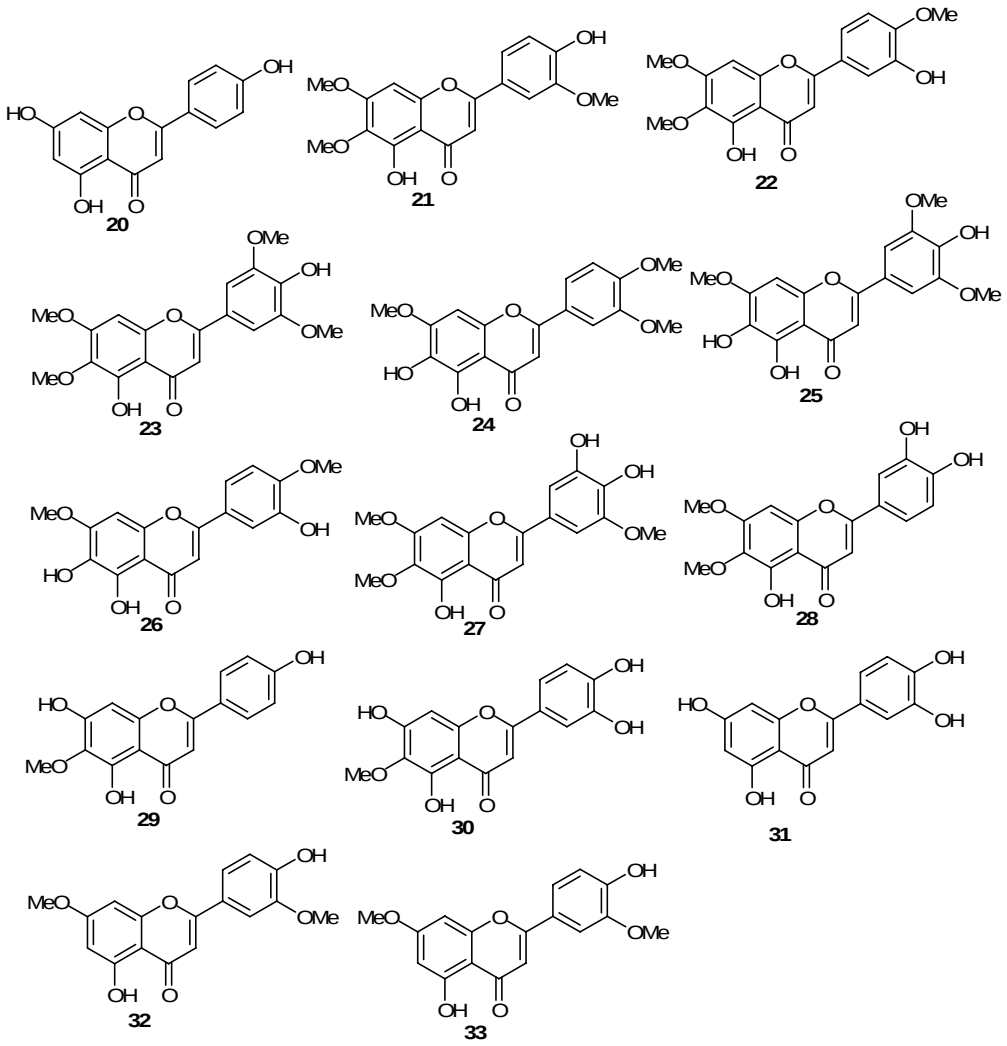

Fig. 3. Compounds 20-33 isolated from *L. montevidensis.*

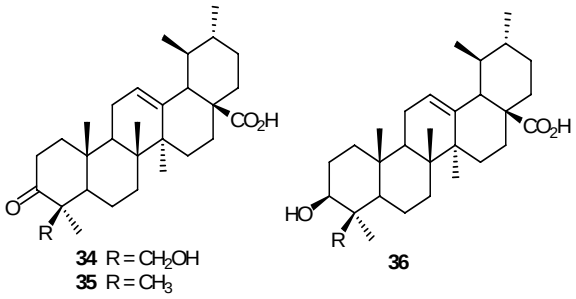

Fig. 4. Compounds **34-36** isolated from *L. tiliifolia.*

***Lantana* × *hybrida*:** A compound, 1-caffeylrhamnose (37), was isolated from the flowers of *L. hybrida* and after alkaline hydrolysis yielded caffeic acid and L-rhamnose (Imperato *et al*., 1975). Another sugar derivative, 1-(3-glucosyloxy-4-hydroxycinnamyl)glucose (38) was isolated from *L.* × *hybrida* (Imperato, 1976) (Fig. 5).

Lantana achyranthifolia: Aerial parts of *L. achyranthifolia* Desf. produced two flavonoids (Dominguez *et al*., 1983), penduletin (39) and chrysosplenetin (40), and the roots produced a new furano-1,4-naphthoquinone named diodantunezone (41) (Abeygunawardena *et al*., 1991) (Fig. 6).

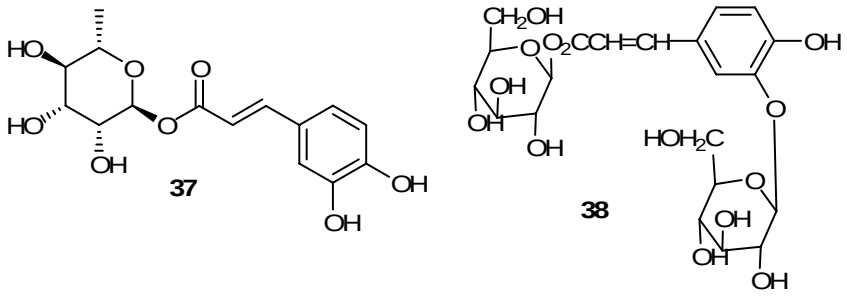

Fig. 5. Compounds 37 and 38 isolated from *L.* × *hybrida.*

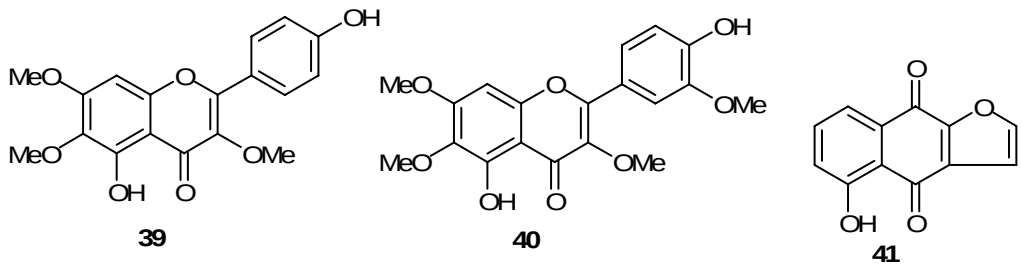

Fig. 6. Compounds 39-41 isolated from *L. achyranthifolia.*

Lantana lilacina: A new acyclic monoterpene glucosyl ester named β-D-glucopyranose, 1-[(2*E*, 6*S*)-6-hydroxy-2, 6-dimethyl-2, 7-octadienoate] (42) was isolated from the leaves of *L. lilacina* Desf. (Takeda *et al*., 1998) (Fig. 7).

Lantana trifolia: *L. trifolia* L. is a species used in the folk medicine of Rwanda (Johns *et al*., 1983a). From the MeOH extract of the dried leaves of *L. trifolia*, a new, antimicrobially active, polymethoxylated flavone was isolated and named umuhengerin (43) after the Rwandese name of the plant. The structure was established as 5-hydroxy-6, 7, 3', 4', 5'-pentamethoxyflavone (Rwangabo *et al*., 1988). Umuhengerin (43) exhibited *In vitro* antibacterial and antifungal properties in concentrations up to 200 µg/ml against various pathogens, including *Staphylococcus aureus*, *Salmonella typhimurium*, *Candida tropicalis*, *Aspergillus niger*, *Aspergillus fumigatus*, *Trichophyton mentagrophytes*, and *Mucrosporum canis* (Rwangabo *et al*., 1988) (Fig. 8).

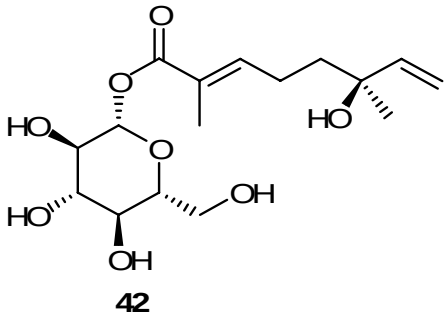

Fig. 7. Compound **42** isolated from *L. lilacina.*

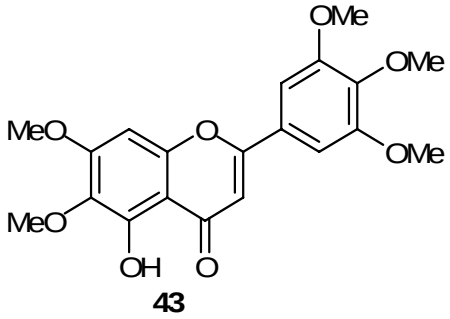

Fig. 8. Compound **43** isolated from *L. trifolia.*

Lantana camara: The plant *L. camara* L. is native to tropical and subtropical America. Dutch explorers introduced it into the Netherlands from Brazil in the late 1600s and later explorers from other countries brought seeds to Europe, Great Britain and North America. Following its introduction into Hawaii as a garden flower, it soon spread to the islands of the Pacific, Australia and southern Asia. In a similar way, from Natal it was rapidly spread by birds to the warmer areas of South Africa. In the 18th and 19th century, nurserymen commercialised and popularised many colourful forms and it is now cultivated world-wide as an ornamental plant. Of the 650 cultivar names in the genus, the majority are associated with the *L. camara* complex. The plant is an aggressive, obligate outbreeder weed that has invaded vast expanses of pastures, orchards and forest areas in many tropical and subtropical regions. It has been estimated that 4 million hectares in Australia (Munir, 1996; Ross, 1999) and 160,000 in Hawaii (Ross, 1999), have been invaded by this plant. Apart from its popularity as a garden plant, *L. camara* is said to form a useful hedge and to provide a good preparation for crops, covering the ground with fine leaf mulch (Munir, 1996). It improves the fertility of rocky, gravelly, or hard laterite soils, enriches the soil, and serves to retain humus in deforested areas and checks soil erosion. It can nurse parasitic sandalwood seedlings and in the Pacific islands has been used as a support for yam vines. *Lantana* leaves and twigs are often used in India as

green mulch. The ash is rich in potassium and manganese, which is useful in manuring coconut trees. The plant is not readily eaten by cattle unless pasturage is very scarce. In tropical countries, the ripe blue-black berries are eaten, but ingestion of the green berry has led to human fatalities (Morton, 1994; Ross, 1999).

Medicinal importance: *Lantana camara* has been used in many parts of the world to treat a wide variety of disorders (Ross, 1999). *Lantana camara* has been used in folk remedies for cancers and tumors. A tea prepared from the leaves and flowers was taken against fever, influenza and stomach-ache. In Central and South America, the leaves were made into a poultice to treat sores, chicken pox and measles. Fevers, colds, rheumatism, asthma and high blood pressure were treated with preparations from the plant (Irvine, 1961). In Ghana, an infusion of the whole plant was used for bronchitis and the powdered root in milk was given to children for stomach-ache (Irvine, 1961). In Asian countries, leaves were used to treat cuts, rheumatism, ulcers and intestinal worms. Decoctions were applied externally for leprosy and scabies. It has been claimed that a steroid, lancamarone, from the leaves, exhibited cardiotonic properties (Sharma & Kaul, 1959) and that lantamine, an alkaloid from the stem, bark and roots showed antipyretic and antispasmodic properties comparable to those of quinine (Sastri, 1962), but the validity of these claims has not been confirmed. An alkaloid fraction which lowered blood pressure, accelerated deep respiration and caused shivering in dogs was isolated from the leaves, and it was suggested that it may be useful in reducing fevers and as a treatment for asthma and hypertension (Sharaf & Naguib, 1959). Six phenolic compounds in a *L. camara* extract were identified by HPLC: salicylic acid (44), gentisic acid (45), β-resorcylic acid (46), coumarin (47), ferulic acid (48), and 6-Me coumarin (49) (Yi *et al*., 2006). Three new pentacyclic triterpenoids, camarin (50), lantacin (51), and camarinin (52), were isolated from the aerial parts of *L. camara*. The structures of the new constituents were elucidated by chemical transformation, HR-EI mass spectrometry, and NMR spectroscopy, including 1D (^{1}H- and ^{13}C-NMR) and 2D (^{1}H,^{1}H-COSY, NOESY, ^{1}H,^{1}H-TOCSY, J-resolved, HMQC, and HMBC) experiments (Begum *et al*., 2006) (Fig. 9).

Lantanilic acid (53), camaric acid (54) and oleanolic acid (9), all of which possess nematocidal activity, were isolated from the methanolic extract of the aerial parts of *L. camara* through bioassay guided fractionation. These compounds exhibited 98%, 95% and 70% mortality respectively against the root-knot nematode *Meloidogyne incognita* at 0.5% concentration (Barua *et al*., 1976; Qamar *et al*., 2005). Two triterpenoids named lantadene A (55) and lantadene B (56) were isolated from the chloroform extract fraction of leaves of *L. camara* (Hart *et al*., 1976a; Sharma *et al*., 1987; Pattabhi *et al*., 1991; Mirsa *et al*., 1997; Ma *et al*., 2004) (Fig. 9).

Extracts of the leaves, twigs, stems, and roots of *L. camara* were solvent-partitioned and screened for activity using the brine shrimp lethality test. The active fractions yielded known oleanonic acid (10), lantadene A (55), and oleanolic acid (9), which were very toxic to brine shrimp larvae (Fatope *et al*., 2002). These compounds were not lethal to *Spodoptera littoralis* Boisduval (Lepidoptera: Noctuidae), *Clavigralla tomentosicollis* Stål (Hemiptera: Coreidae), and *Aphis craccivora* Koch (Homoptera: Aphididae) when tested at 5000 μg/ml. Lantadene A (55), however, suppressed the fecundity of *C. tomentosicollis* at 5000 μg/ml.

Three new pentacyclic triterpenoids, camaryolic acid (57), methylcamaralate (58) and camangeloyl acid (59), were isolated from the aerial parts of *L. camara* (Begum *et al*., 2003a). A new ursane derivative was isolated from the leaves of *L. camara* and its structure elucidated as 3,24-dioxo-urs-12-en-28-oic acid (60) by spectral analysis (Yadav & Tripathi, 2003). Three new pentacyclic triterpenes, ursoxy acid (61), methyl ursoxylate (62), and ursangilic acid (63), along with oleanolic acid acetate (64), were isolated from the aerial parts of *L. camara* (Begum *et al*., 2002). A new pentacyclic triterpene, ursethoxy acid (65), was isolated from the aerial parts of *L. camara*. Its structure has been elucidated as 3,25-epoxy-3α-ethoxy-urs-12-en-28-oic acid through extensive NMR studies (Begum *et al*., 2002). Chemical analysis of *L. camara* yielded 5,7-dihydroxy-4',6-dimethoxy flavone (66) as a minor constituent (Yadav & Tripathi, 2000). Similarly, *L. camara* L. var. *aculeata* yielded a triterpenoid, pomonic acid (67) (Misra & Laatsch, 2000a) (Fig. 10).

Mirsa *et al*., (1997) isolated two novel triterpenoids from the roots of *L. camara*. Their structures were determined as 3β,19α-dihydroxyursan-28-oic acid (68) and 21,22β-epoxy-3β-hydroxyolean-12-en-28-oic acid (69) (Misra & Laatsch, 2000b). Two new constituents, lantanoside (70) and lantanone (71), and the known compounds linaroside (72) and camarinic acid (73), were isolated from the aerial parts of *L. camara* (Fig. 11). Compounds 70, 72, and 73 were tested for nematicidal activity against the root-knot nematode *Meloidogyne incognita* and showed 90, 85, and 100% mortality, respectively, at 1.0% concentration (Verma *et al*., 1997; Begum *et al*., 2000). Two new constituents, camarolide (74) and lancamaric acid (75), have been isolated from the aerial parts of *L. camara*. Structures of the new compounds were established as 3-keto-urs-11-en-13β(28)-olide and 3,25-epoxy-3α-ethoxy-olean-12-en-28-oic acid, respectively (Siddiqui *et al*., 2000). The triterpenoid lantic acid (76) was isolated from *L. camara* plants cultivated in Egypt. The antibacterial activity of lantic acid (76) was assessed with bioautography assays for Gram-positive and Gram-negative bacteria (Barua *et al*., 1969; Barua *et al*., 1972; Siddiqui *et al*., 1995; Saleh *et al*., 1999). Lantic acid (76) was found to possess strong antibacterial activity against *Escherichia coli* and *Bacillus cereus*, with 0.08 and 0.1 μg the minimum inhibition doses, respectively, compared to 0.05 and 0.005 μg for chloramphenicol, respectively. The results indicate that lantic acid (76) has broad spectrum antibacterial activity and may hold potential as a non-selective antimicrobial agent (Saleh *et al*., 1999) (Fig. 11).

The known triterpenoids icterogenin (77), betulonic acid (78), and betulinic acid (79), the known flavonoid, β-sitosterol 3-O-β-D-glucoside (80), and a mixture of campesterol (81), β-sitosterol (82) and stigmasterol (83) were isolated from the stems of pink-flowering *L. camara* (Lai *et al*., 1998) (Fig. 11).

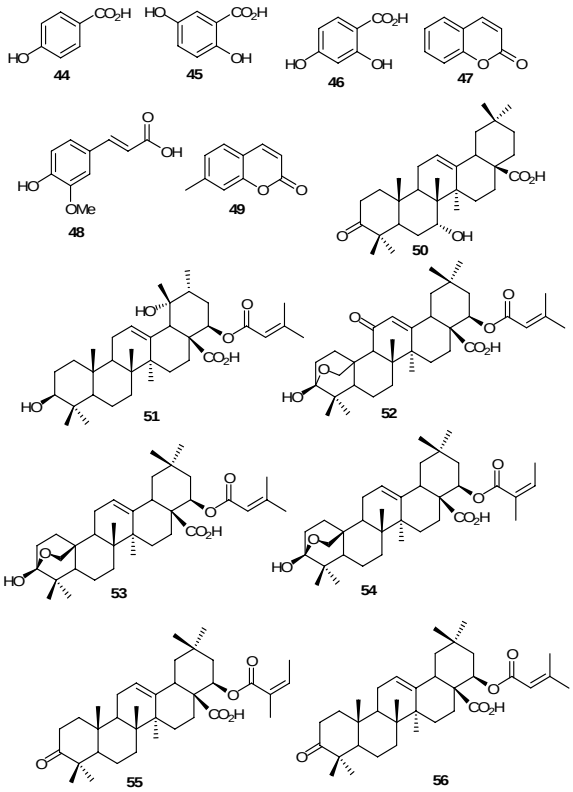

Fig. 9. Compounds 44-56 isolated from *L. camara.*

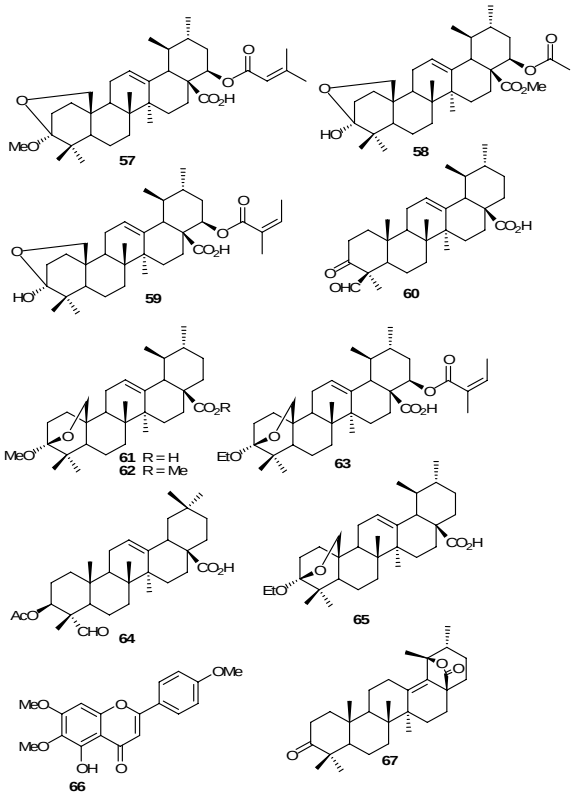

Fig. 10. Compounds 57-67 isolated from *L. camara.*

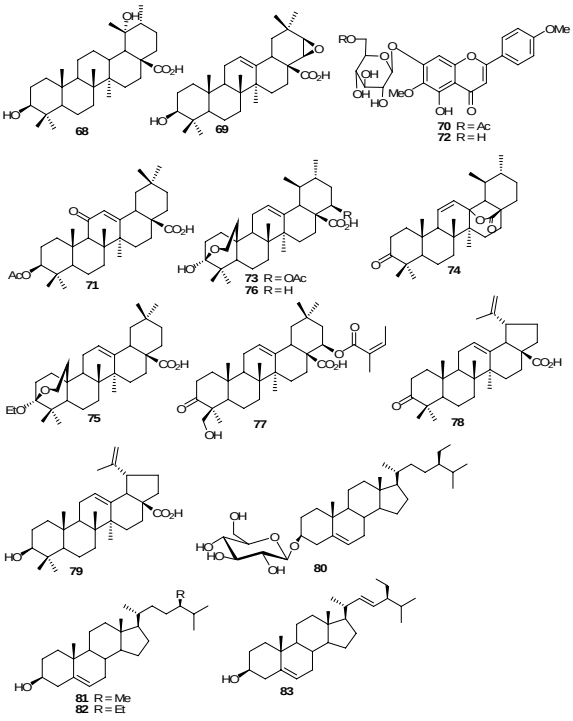

Fig. 11. Compounds 68-83 isolated from *L. camara.*

An investigation of the methanolic extract of *L. camara* revealed a suite of euphane triterpene lactones. The presence of these metabolites, which occur in trace quantities (0.00004-0.0002%) (O'Neill *et al.,* 1998), was detected by using an assay in which thrombin activity was measured as a function of clot formation from fibrinogen. In all, five active compounds (84–88) were isolated. The structure of these compounds was determined by means of spectroscopic methods and confirmed by single crystal X-ray crystallographic studies on 84. All compounds were potent inhibitors of human thrombin (IC_{50} 18-130 nM) and showed comparable activity to hirudin (IC_{50} 12 nM), a dried and refined extract of leeches (*Hirudo medicinalis*) (O'Neill *et al*., 1998). Verbascoside (89), a widespread phenylethanoid, has been isolated from *L. camara* (Herbert *et al*., 1991). The *Z*-isomer of verbascoside, lantanaside (90), has also been found to co-occur with verbascoside in *L. camara* (Mahato *et al.*, 1994), but it is useful to note that *trans-cis* isomerisation of cinnamoyl esters can occur during manipulation of the sample in the light. Isoverbascoside (91), which often co-occurs with verbascoside, martynoside (92), derhamnosylverbascoside (93), isonuomioside A (94) and calceolarioside E (95) (Fig. 12), has been isolated from *L. camara* (Taoubi *et al*., 1997).

Known triterpenoids, a mixture of α-amyrin (96), β-amyrin (97) and pomolic acid (98), were isolated from the stems of *L. camara* (Lai *et al.,* 1996). Wollenweber *et al*., (1997) isolated three quercetin derivatives named contained 3-methoxy- (99), 3,7-dimethoxy- (100), and 3,7,4'-trimethoxyquercetin (101) from *L. camara*. A novel triterpene, 22β-acetoxylantic acid (102), and the known triterpenes lantic acid (76) and lantanolic acid (103) (Fig. 13), were also isolated from *L. camara* (Barua *et al*., 1966; Barau *et al*., 1971; Barre *et al*., 1997).

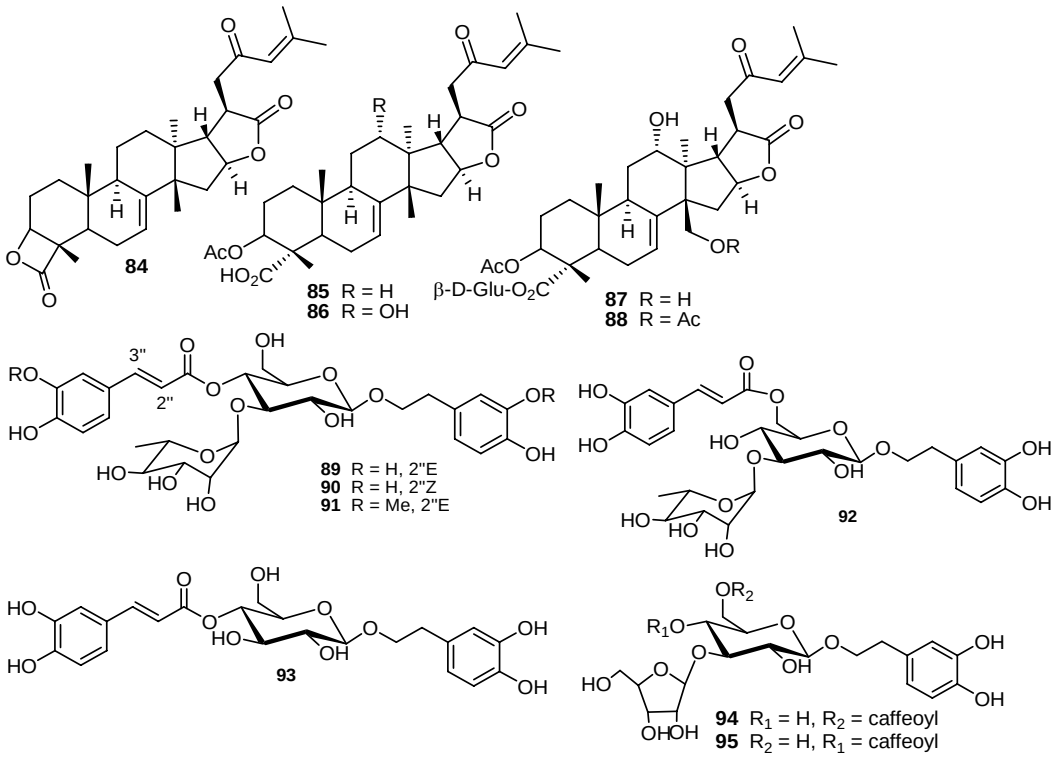

Fig. 12. Compounds 84-95 isolated from *L. camara.*

Singh *et al*., (1996) isolated two additional triterpenes, 25-hydroxy-3-oxoolean-12-en-28-oic acid (104) and hederagenin (105), from *L. camara*. Begum *et al*., (1995) reported a new Δ12-oleanane triterpenoid and a new Δ12-ursane type triterpenoid, camarilic acid (106) and camaracinic acid (107), respectively, from the aerial parts of *L. camara* collected in Pakistan. A number of triterpenoids were isolated by Pan *et al*., (1993a), from the roots of *L. camara* and they were identified as lantanolic acid, 22β-hydroxy-oleanonic acid (108), and lantaiursolic acid (109) (Fig. 13).

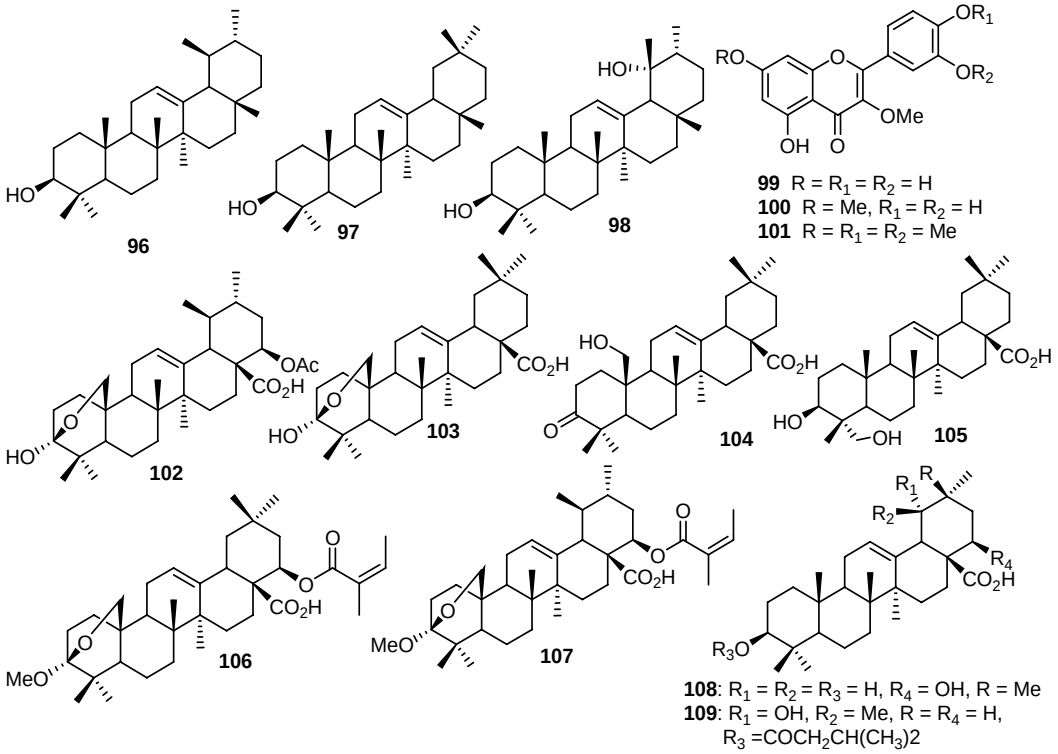

Fig. 13. Compounds 96-109 isolated from *L. camara.*

As part of a taxonomic study of *Lantana*, greenhouse-grown plants were shown to contain the sodium salt of theveside (110) in both leaves and roots, but theviridoside (111), the corresponding methyl ester, was only found in the roots (Ford & Bendall, 1980; Rimpler *et al*., 1986). From the roots of *L. camara*, geniposide (112), the biosynthetic precursor of theveside, has been isolated, together with 8-epiloganin (113), shanzhside methyl ester (114) and lamiridoside (115) (Pan *et al*., 1992a; Pan *et al*., 1992b). Six oligosaccharides, ajugose (116), stachyose (117), verbascotetraose (118), verbascose (119), and lantanose A (120) and B (121) (Fig. 14), were isolated from the roots of *L. camara* (Pan *et al*., 1992a).

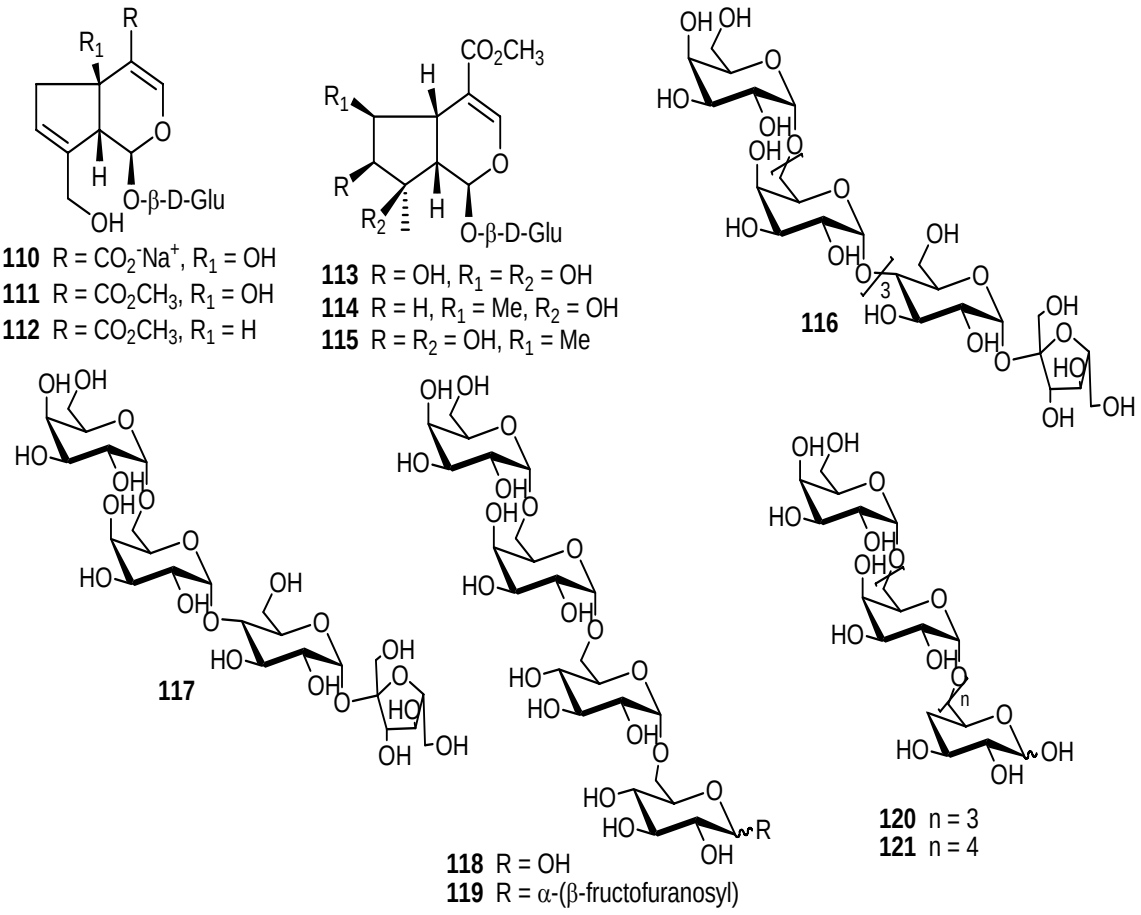

Fig. 14. Compounds **110-121** isolated from *L. camara.*

Pan *et al.*, (1993b) isolated another flavone named camaroside (122) and characterized it as 4',5-dihydroxy-3,7-dimethoxyflavone-4'-O-β-D-glucopyranoside.

Lantadene C (123) isolated from the leaves of the hepatotoxic plant *L. camara* was identical with dihydrolantadene A, reported earlier (John *et al*., 1983; Sharma *et al*., 1992). The molecular structure of lantadene C (123) (Fig. 15) has been deduced from single crystal x-ray diffraction analysis. It resembles lantadene A in the pentacyclic portion of the molecule but differs in the side chain region. Atom C-34 is *cis* to C-35 in lantadene C but is *trans* in lantadene A (55) (Sharma *et al.*, 1992). Sharma *et al.*, (1990) reported another novel triterpene named lantadene D (124) from *L. camara*. Lantoic acid (125), a new triterpene, was also isolated from the leaves of *L. camara* (Roy & Barua, 1985). Another pentacyclic triterpene named lantabetulic acid (126) (Fig. 15) was isolated from *L. camara* (Hart *et al*., 1976b).

Many essential oils have been reported from *L. camara* (e.g. Mahmud *et al*., 1979; Avadhoot *et al*., 1980; Da Silva *et al.*, 1999; Ngassoum *et al*., 1999; Weyerstahl *et al*., 1999; Deena & Thoppil, 2000; Fathy, 2000; Jose & Thoppil, 2000; Weyerstahl *et al*., 2001; Khan *et al*., 2002; Kasali *et al*., 2002; Sefidkon, 2002; Oyedeji *et al*., 2003; Pino *et al*., 2004; Randrianalijaona *et al*., 2006) These essential oils are rich in sesquiterpenes such as aromadendrene (127), cubenol (128), davanone D (129), epicubenol (130), germacrene D (131), humulene oxide (132), δ-elemene (133), δ-cadinene (134), γ-muurolene (135), (±)-γ-cadinene (136), β-elemene (137), l-β-bisabolene (138), α-cubebene (139), aglaiene (140), l-α-cadinol (141), himbaccol (142), espatulenol (143), uncineol (144), β-bourbonene (145), B1-cadinene (146), β-cubebene (147), α-ylangene (148), germacrene B (149), α-calacorene (150), l-alloaromadendrene (151), germacrene A (152), β-gurjunene (153), (syn-anti-anti)-helifolen-12-oic acid (154), (anti-anti-anti)-helifolen-12-oic acid (155), (anti-syn-syn)-helifolen-12-oic acid (156), (syn-syn-syn)-helifolen-12-oic acid (157), β-patchoulene (158), ε-muurolene (159), and oplopanon (160) (Fig. 16).

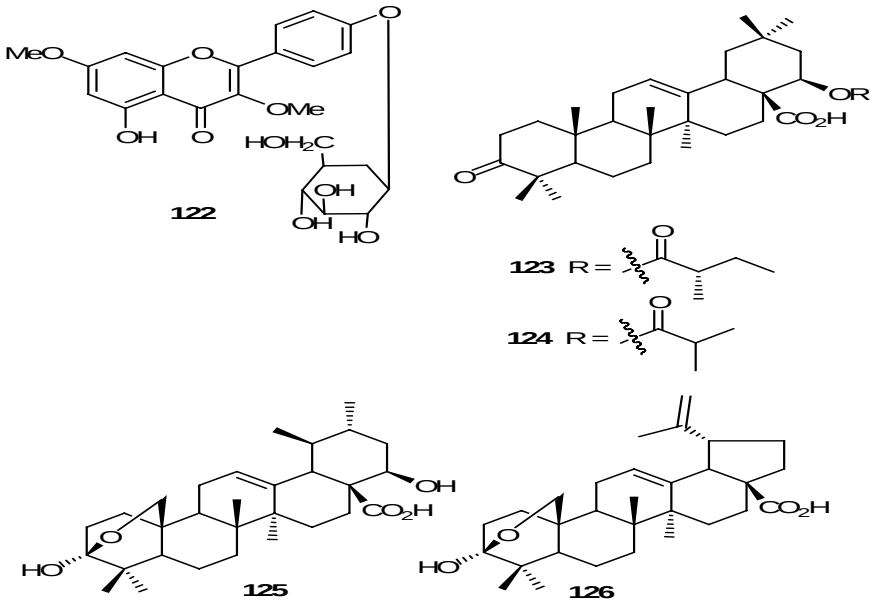

Fig. 15. Compounds 122-126 isolated from *L. camara.*

Bioactivity of *Lantana* metabolites: Lantadenes A (55), B (56), and C (123) inhibited Epstein-Barr virus activation in Raja cells induced by 12-O-tetradecanoylphorbol-13-acetate (TPA). Compounds 56 and 123 were active even at 10 mol triterpenoid/1 mol TPA, so they could be considered valuable inhibitors of tumour promoters *In vivo* (Inada *et al*., 1995). Lantadenes A (55) and B (56) were shown to have inhibitory effects on the two-stage carcinogenesis of mouse skin papillomas, using 7,12-dimethylbenz[*a*]anthracene as an initiator and TPA as a promoter. Lantadene B (56) (47 µg), applied before each treatment of TPA, delayed the formation of papillomas on mouse skin, reduced the rate of papilloma-bearing mice (by 15% at 20 weeks) and reduced the average number of papillomas per mouse (50% at 20 weeks) (Inada *et al*., 1997).

Considerable interest has been shown in the anti-inflammatory action of some triterpenes (Safayhi & Sailer, 1997). For example, oleanolic acid (9) and ursolic acid (36) have significant activity (IC_{50} 2-4, 6 µM) as inhibitors of human leucocyte elastase (HLE). This enzyme participates in the destruction of elastin and plays a role in chronic disorders such as pulmonary emphysema, cystic fibrosis, hepatitis and rheumatic arthritis.

Oleanolic acid (9) and ursolic acid (36) possess inhibitory effects on inflammation and on various stages of tumour development (Hsu *et al*., 1997). In a recent study, ursolic acid was shown to have COX-2 inhibitory effect with an IC_{50} value of 130 µM and a COX-2/COX-1 selectivity ratio of 0.6. Oleanolic acid showed IC_{50} 295 µM and a ratio of 0.8 (Ringbom *et al*., 1998).

Thrombin inhibitory activity was found to be associated with the euphane lactone triterpenes (84-88), which inhibit the blood-clotting cascade via acylation of the active site Ser 195 residue of thrombin. This acylating activity is generic towards other serine proteases (Weir *et al*., 1998). The lactones 84-88 are potent inhibitors of α-thrombin and, to a lesser extent, of α-chymotrypsin and other serine proteases. α-Thrombin is a serine protease that belongs to the trypsin family and has a central role in the hemostatic process, where it displays both coagulant and anticoagulant activities. Tight-binding reversible competitive inhibition was shown by compounds 85 and 87. Protease inhibition involves the opening of the lactone ring and acylation of the active-site serine 195. The IC_{50} for α-thrombin, α-chymotrypsin and trypsin was 0.004, 0.07 and 0.07 for 85 and 0.004, 0.01, 0.12 µM for 87. X-Ray crystallographic studies of the α-thrombin (85) and a-thrombin (87) complexes showed the inhibitor in the ring-opened form. The hydroxyl group that attacks the seryl ester probably occupies the position normally taken by water during deacylation of peptide substrates. Model compounds incorporating 5,5 trans-fused indane lactones have been tested as inhibitors of thrombin (Pass *et al*., 1999a). Although some of these showed significant activity as HLE, chymotrypsin and human α-thrombin inhibitors, they were relatively unstable in plasma (Pass *et al*., 1999a). Model compounds containing a lactam had much enhanced plasma stability compared to their lactone counterparts and showed appreciable *In vitro* anticoagulant activity (Pass *et al*., 1999b).

Of the iridoids so far isolated from *L. camara*, only geniposide (112) has been studied in detail. Geniposide (112) inhibited hepatoxicity and the DNA repair synthesis induced by aflatoxin B1 in rat primary hepatocytes. The possible mechanism involves enhancement of levels of glutathione S-transferase and GSH peroxidase (Wang *et al*., 1992). Intraduodenal administration of geniposide showed a delayed but potent choleretic action in rats. It was observed that the aglycone genipin is the active agent since geniposide had no effect on intraportal administration (Aburada *et al*., 1978). Iridoid glycosides are known to be hydrolysed in the gastrointestinal tract. In mice, geniposide (112) was metabolised to the aglycone genipin, which was found in the entire gastrointestinal tract, especially the cecum and the colon (Yamouchi *et al*., 1976). Geniposide (112) is hydrolysed by β-D-glucosidases from intestinal bacteria (Yang *et al*., 1996). Diodantunezone (41) was tested for cytotoxicity against KB epidermoid nasopharynx, K562 human leukaemia and P388 lymphocytic leukaemia cell lines and found to be active (IC_{50} 6.76, 9.2 and 7.94 mmol/l, respectively). The corresponding methyl ether showed greater activity (IC_{50} 1.3, 1.32 and 1.86 mmol/l, respectively) (Perry *et al*., 1997).

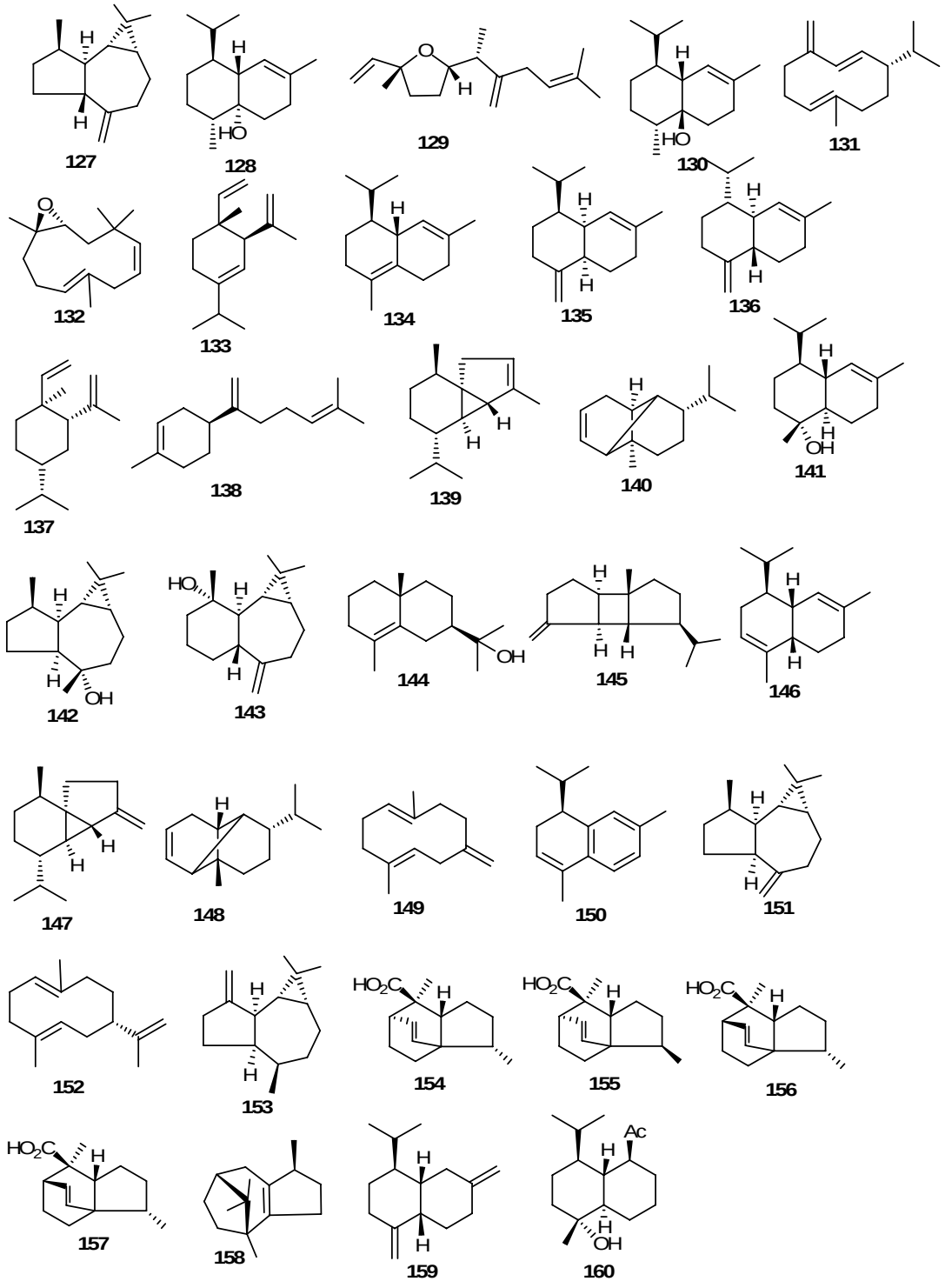

Fig. 16. Compounds 127-160 isolated from *L. camara.*

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