Recent Developments of Natural Product Chemistry in Taiwan

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Geographically Taiwan is located at a latitude between the tropical and sub-tropical zones. The island has more than four hundred mountains with an altitude higher than 2000 meters, hence it also has the arctic climate. Rainfall is heavy, the fourth highest in the world. Forest land is abundant, thus Taiwan is a treasure island of plants. Here in this island the study on the constituents of plants especially the medicinal plants of the folk medicine is very popular.

As for the past progress of the studies on the chemistry of natural proucts in Taiwan, one can refer to the book "Abstract of Chinese Herb Medicine in Taiwan (1905~1972)"1), and the article "Studies on Natural Resources of Drugs in Taiwan"2) reported by Dr. H. Y. Hsu in the 3rd Asian Congress of Pharmaceutical Sciences, FAPA, held in Korea in 1968, and also the topic "Recent Studies on Chinese Drugs in Taiwan"3) reported by Dr. Hsu in the 5th Congress of FAPA, held in Taipei, Republic of China in 1974. Dr. H. Y. Hsu and I have already made a review titled "Studies on Chinese Drugs in Taiwan"4) in which the major studies on the chemical constituents relating to 40 kinds of Chinese herb drugs, and the 41 new chemical constituents isolated therefrom during the period from 1968 to 1974 were included.

In the present review, an outline on the major researches about plant constituents as done by the scientists in Taiwan during the recent five years will be introduced. The names of plants, parts used in the study and components isolated and determined are summarized as follows. (Where * denotes the new compound first isolated from the natural world.)

- (1) Six species of *Podocarpus* (family Podocarpaceae) grown in Taiwn were studied. The nor-and bisnor-diterpene lactones thus isolated from the trunks of these plants are shown below:
- P. macrophyllus (Thunb.)D. Don: inumakilactone A, inumakilactone A glucoside* (C₂₄H₃₀ O₁₃,mp. 296~300°), inumakilactone E*(C₁₉H₂₄ O₇,mp. 220~225°), nagilactone C
- P. philippinensis Foxworthy: inumakilactone A, inumakilactone A glucoside, inumakilactone E, nagilactone A
 - P. formosensis Dummer: nagilactone A
- P. nagi (Thunb.) Zoll. et Makino: nagilactone A
- P. polystachyus R. Br.: inumakilactone B, hallactone B, nagilactone A
 - P. nankoensis Hayata: nagilactone C

Among the above-mentioned components, inumakilactone A, inumakilatone A glucoside and nagilactone C exhibited anti-inflammatory and antipyretic activity in rats⁵⁾

Inumakilactone A glucoside

Inumakilacton E

(2) Alkaloidal constituents of the barks and the fruits of *Cephalotaxus wilsoniana* Hay., Ceph alotaxaceae were studied.

3-epi-schelhammericine, Base VI and 3-epiwilsonine of homoerythrina alkaloids, and cephalotaxine, acetylcephalotaxine, isoharringtonine and Alkaloid G of the cephalotaxus alkaloids were isolated.⁶⁾.

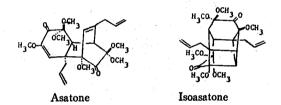
- (3) From the wood of Keteleeria davidiana Beissner, Pinaceae, α -pinene, β -pinene, limonene, α -copaene, bornyl acetate, β -elemene, caryophyllene, caryophyllene oxide, α -selinene, β -selinene, selin-11-en-4 α -ol, laevojunenol, α -cyperone, α -conidendrin, 15-hydroxydehydroabietic acid, hexacosanyl tetracosanoate, tetracosyltetracosanate, docosanyltetracosanoate, hexacosanol, tetracosanol, docosanol, β -sitosterol and stigmasterol were isolated.
- (4) From the wood of *Pinus luchuensis* Mayer, Pinaceae, 3β -methoxyserrat-14-en-21-one, serrat-14-en-3, 21-dione,* 3β -hydroxyserrat-14-en-21-one, 3β , 21α -dimethoxyserrat-14-ene, 3β -methoxyserrat-14-en-21 α -ol, methyl dehydroabietate, pinosylvin monomethylether, pimaric acid, dehydroabietic acid, α -pinene, β -pinene, camphon, 4-terpineol, α -terpineol, carvacrol, tetracosanol, hexacosanol, octacosanol, β -sitosterol and stigmasterol were isolated.⁸⁾
- (5) A new derivative of royleanone, 6β-acetoxy-7α-hydroxyroyleanone*, C₂₂H₃₀O₆, mp. 182~183°, [α]_D-52.5° (c=1.0, CHCl₃) besides 6,7-dehydroroyleanone were isolated from the barks of *Taiwania cryptomerioides* Hayata, Taxodiaceae.⁹⁾
- (6) A new phenolic diterpene, 6α-hydroxy-7-oxo-ferruginol*, C₂₀H₂₈O₃, mp. 207~208°, (α)_D²⁰ +59. 3° (c=1.0, EtOH) was isolated from the barks of *Libocedrus formosana* Florin, Cupressaceae.¹⁰⁾
- (7) Two novel neolignans, asatone*, $C_{24}H_{32}$ O_8 ,mp. $101\sim102^\circ$ $(\alpha)_D^{20}\pm0^\circ$ (MeOH) and isoa-

Serrat-14-en-3, 21-dione

6β-Acetoxo-7αhydroxoroyleanone

6α-Hydroxy-7oxo-ferruginol

satone*, $C_{24}H_{32}O_8$, mp. 157~158°, $[\alpha]_0^{20}\pm0^\circ$, (MeOH), were isolated from the whole herb of Asarum taitonense Hayata, Aristolochiaceae, together with safrole, elemicin, esamin, borneol and β -sitosterol. From a biogenetic point of view, furthermore, thermal and photochemical reactions of these neolignans and their derivatives were carried out. In particular, asatone was photochemically converted into isoasatone. 11)



- (8) Form the whole herb of *Polycarpaea cory* mbosa Lam., Caryophyllaceae, camelliagenin A, A₁-barrigenol and stigmastanol were isolated.¹²⁾
- (9) From the whole herb of *Thalictrum urb-aini* Hayata, Ranunculaceae, S-(+)-oconovine and S-(+)-isocorydine were isolated.¹³⁾
- (10) From the roots of *Tinospora dentata* Diels, Menispermaceae, a diterpenoid, columbin and two alkaloids, palmatine and jatrorrhizine were isolated.¹⁴⁾
- (11) Two new schizandrin-type lignans, kadsurin*, $C_{25}H_{30}O_8$, mp. 157 ~158°, $[\alpha]_D^{25}$ -39° (c = 0.13, CHCI₃), kadsurarin*, $C_{30}H_{36}O_{11}$, mp.

255~256°, $[\alpha]_D^{25}$ -65° (c=0.10, CHCl₃), and a new A-secolanostane type triterpenoid, kadsuric acid*, $C_{30}H_{46}O_4$, mp. 185°, $[\alpha]_D^{20}+$ 77.3° (c=1.1, MeOH) were isolated from the stems of *Kadsura japonica* Dunal, Magnoliaceae. 15)

Kadsurin

Kadsurarin

Kadsuric aid

- (12) From the stem barks and leaves of Anona montana Macf., Anonaceae, five alkaloids were isolated. These include two major phenanthrene alkaloids, atherosperminine, argentinine and three minor bases, i.e. anonaine, oxoushinsunine and reticuline.¹⁶⁾
- (13) A new bisbenzylisoquinoline alkaloid, lindoldhamine*, $C_{34}H_{36}N_2O_6$, mp. $183\sim186^\circ$, $[\alpha]_D^{33}+35^\circ$ (c=1.0, EtOH),together with other seven alkaloids, namely, O-methylbulbocapnine* (O,N-dimetylnandigerine), $C_{20}H_{21}O_4N$, mp. $129\sim130^\circ$, $[\alpha]_D^{31}+248^\circ$ (CHCl₃), N-methylnandigerine (N-methylhernangerine), N-methylovigerine (N-methylhernovine), dicentrine, N-nordicentrine, dicentrinone and L-(+)-magnonocurarine were isolated from the trunks and leaves of Lindera oldhamii Hemsl, Lauraceae. 17)

Lindoldhamine

O-Methylbulbocapnine.

- (14) Three species of *Neolitsea* (family Lauraceae) were studied. Following alkaloids were isolated.
- N. aurata (Hay.) Koidz., wood: laurolitsine, litsericine, N-methyllitsericine, (+)-anonaine, (-)-roemerine¹⁸⁾
- N. buisanensis Yamamoto et Kamikoti, wood: laurolitsine, litsericine¹⁸⁾
- N. daibuensis Kamikoti, root wood and root barks: S-(+)-reticuline¹⁹⁾
- (15) From the leaves and the trunks of *Deh* aasia triandra Merr., Lauraceae, and aporphine alkaloid, isocorydine and a bisbenzylisoquinoline alkaloid, obaberine were isolated, respectively.²⁰⁾
- (16) From the leaves and the stems of *Cinn amomum osmophloeum* Kanehira, Lauraceae, kaempferitrin, kaempferol-7-rhamnoside, coumarin and fumaric acid were isolated.²¹⁾
- (17) From the stems of *Illigera luzonensis* (Presl.) Merr., Hernandiaceae, an aporphine-type base, launobine was isolated.²²⁾
- (18) From the root barks of *Hernandia ovigera* L., Hernandiaceae, six alkaloids, namely, oxothalicarpine*, C₄₀H₄₀O₉N₂•2H₂O, mp. 219~220° (dec.), [α]_D²⁵+115° (c=0.1, CHCl₃), thalicarpine, dehydrothalicarpine, ovigerine, hernangerine, hernandonine and a phenyl-tetralin type lignan, desoxypodophyllotoxin were isolated.²³⁾

Desoxypodophyllotoxin and thalicarpine exhibited a distinctive cytotoxic activity against nasopharynx carcinoma was reported by S. M. Kupchan et. al.

Oxothalicarpine

(19) The alkaloidal constituents of Corydalis ochotensis Turcz., Papaveraceae was studied,

From the whole herb of this plant, two new spirobenzylisoquinoline akaloids, yenhusomine*, $C_{21}H_{23}NO_6$, mp. $127\sim128^\circ$, $(\alpha)_D^{19}+48^\circ$ (c=1.0, MeOH) and yenhusomidine*, $C_{21}H_{21}NO_6$, mp. $240\sim241^\circ$, $(\alpha)_D^{29}O^\circ$ (c=0.41, CHCl₃) and two tetrahydroprotoberberine alkaloids, corytenchine*, $C_{20}H_{23}NO_4$, mp. $257\sim258^\circ$, $(\alpha)_D^{30}-268^\circ$ (c=0.89, CHCl₃) and corytenchirine*, $C_{21}H_{25}NO_4$, mp. $246\sim247^\circ$, $(\alpha)_D^{24}-299^\circ$ (c=1.0, CH Cl₃) together with four known alkaloids, ochotensimine, adlumidine, protopine and didehydroc heilantifoline were isolated.²⁴⁾

- (20) From the stems of Capparis formosana Hemsl., Capparidaceae, scopoletin, scopolin, caffeic acid ethyl ester and a mixture of β -sitosterol, stigmasterol and campesterol were isolated.²⁵⁾
- (21) From the stems of *Spiraea formosana* Hay., Rosaceae, friedelin, glutinol, β -amyrin, 10-nonacosanol and a mixture of β -sitosterol and campesterol were isolated.²⁶
- (22) Two species of *Tephrosia* (family Leguminosae) were studied. Three new flavonoids named obovatin*, $C_{20}H_{18}O_4$, mp. 123-124°, $[\alpha]_D$ -93.8° (c=3.74, CHCl₃), obovatin methyl ether*, $C_{21}H_{20}O_4$, mp. 163°, $[\alpha]_D$ -50° (c=2.6, CHCl₃), and obovatachalcone*, $C_{21}H_{20}O_4$, mp. 105° were isolated from the whole herb of T.

obovata Merr. These three components displayed moderate piscicidal activity against loach fish.²⁷⁾ Obovatin methyl ether was also isolated from the whole plant of *T. candida* (Roxb.) DC.²⁸⁾

Obovatin R=H Obovatin methyl ether R=Me Obovatachalcone

(23) The flavonoids in the leaves of *Clitoria* ternatea L., Leguminosae was studied. Four kaempferol glycosides, namely, kaempferol-3-0-rhamnosyl-(1→2)-0-[rhamnosyl-(1→6)]-glucoside (named clitorin*), C₃₃H₄₀O₁₉, mp. 198°, kaempferol-3-glucoside (astragalin), kaempferol-3-rutinoside (nicotifolin) and kaempferol-3-neohesperidoside were isolated.²⁹⁾

Clitorin cuspidiol

- (24) From the leaves of *Uraria crinita* Desv., Leguminosae, six C-glycosylflavones, namely, vitexin, vitexin 7-0-glucoside, orientin, orientin 7-0-glucoside, saponaretin 4'-0-glucoside and vicenin II were isolated.³⁰⁾
- (25) Five flavonol glycosides, kaempferol-3-arabinoside, kaempferol-3-xyloside, quercetin) and quercetin-3-galactoside were isolated from the leaves of *Leucaena glauca* (L.) Benth., Leguminosae.³¹⁾
- (26) From the leaves of Cassia fistula L., Leguminosae, three kaempferol glycosides, kaem

pferol-3-glucoside (astragalin), kaempferol-3-neohesperidoside and kaempferol-3-0-rhamnosyl- $(1\rightarrow 2)$ -0-[rhamonosyl- $(1\rightarrow 6)$ -glucoside(clitorin) were isolated.³¹⁾

(27) Kaempferol-3-rhamnoside (afzelin) and quercitrin were isolated from the leaves of Pithecolobium dulce Benth., Leguminosae. 31) Vicenin II, quercetin-3-xyloside (reynoutrin), quercetin-3-rhamnoglucoside(rutin), myricetin-3-rhamnoside (myricitrin) and kaempferol-3-robinoside-7-rhamnoside(robinin) were obtained from the leaves of Albizzia lebbeck Benth., Aeschynomene indica L., Canavalia lineata D.C., Flemingia congesta Roxb., and Vigna marginata Benth., Leguminosae, respectively. 31)

(28) The constituents of Xanthoxylum cuspid atum Champ. (Fagara cuspidata Engl.), Rutaceae, was investigated. From the wood a new phenylpropanoid, cuspidiol*, C14H20O3, mp. 65-67°, and β -sitosterol, besides six alkaloids, i.e. nitidine chloride, dictamine, γ-fagarine, skimmianine, robustine and haplopine were isolated.32) From the bark three new alkaloids, arnottianamide*, C21H19O6N, mp. 264-267°, isoarnottianamide*, C21H19O6N, mp. 254-257°, and des-N-290-295°, methylavicine*, C₁₉H₁₁O₄N, mp. together with eight known alkaloids, des-Nmethylchelerythrine, decarine, liriodenine, oxynitidine 4-methoxy-1-methyl-2-quinolone, nitidine chloride, 7-fagarine and skimmianine were obtained. Dihydro-p-coumaryl alcohol, cuspidiol and β-sitosterol were also isolated from the bark of this plant. 321 A new amide, integriamide*, mp. 302-304°, was isolated from the root-xylem of Xanthoxylum integrifoliolum Merr. 33)

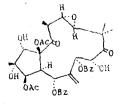
Cuspidiol Arnottianamide R₁=OMe, R₂=H Isoarnottianamide R₁=H, R₂=OMe

Des-N-methylavicine

Integriamide

- (29) From the peels of Citrus reticulata Blanco, Rutaceae, nobiletin, 5-demethylnobiletin, ponkanetin, 5, 7, 8, 4'-tetramethoxyflavone, 6, 7-dimethylesculetin, limonin, cholesterol, campesterol, stigmasterol and β -sitosterol were obtained. From the roots of Citrus maximus form hounyu, xanthyletin, stigmasterol and β -sitosterol were obtained.
- (30) From the leaves of *Phelloedndron wilsonii* Hayata et Kanehira, Rutaceae, phellamurin and amurensin were isolated and it was confirmed that the latter coincided with 8-isopentenylkaempferol-7-glucoside, i.e. epimedoside C.³⁶)
- (31) From the flowers and the fruits of Murraya paniculata(L.) Jack, Rutanceae, scopolin, scopoletin and glucose were obtained.³⁷⁾
- (32) From the toxic fractions of the dried roots of Euphorbia kansni Liou., Euphorbiaceae, six new diterpene derivatives were isolated. They were kansuinine A*, C₃₇H₄₆O₁₅, mp. 218-220° (α)²³_D+28° (c=0.25, MeOH), kansuinine B*, C₃₈H₄₂O₁₄, mp. 160-162° (α)²³_D+37 o(c=0.27, MeOH), 20-deoxyingenol-3-benzoate*, 20-deoxyingenol-5-benzoate*, ingenol-3-(2, 4-decadienoate)-20-acetate*, and 13-oxyingenol-13-dodecanoate, 20-hexanoate* C₃₈H₆₀O₈. Among them, kansuinine A and kansuinine B exhibited a strong anagesic and anti-writhing activity in mice.³⁸⁾

R=Benzoyl×1, Acetyl, ×3 Kansuinine A



Kansuinine B

OH 20 CH₃ OR CH₂OOCCCH₃

R₁=COPh, R₂=H 20-Deoxyingenel-3benzoate R₁=H, R₂=COPh

R₁=H, R₂=COPh 20-Deoxyingenol-5benzoate R=CO(CH=CH)₂ (CH₂)₄CH₃ Ingenol-3-(2, 4-decadienoate)-20-acetate

R₁=CO(CH₂)₄CH₃ R₂=Dodecanoyl

13-Oxyingenol-13-dodecanoate-20-hexanoate

(33) The pigments in the seed-kernels of wax-tree, Rhus succedanca L., Anacardiaceae were investigated. Three new biflavanones, i.e. rhusflavanone* (6,8"-binaringenin), $C_{30}H_{22}O_{10}$. 1.5 H_2O , mp. 204-206°, $(\alpha)_D^{20}-29^\circ$ (c=1.8, MeOH), succedancaflavanone*(6,6"-binaringenin), $C_{30}H_{22}O_{10}$, mp. 318-322° (dec.), $(\alpha)_D^{20}-13^\circ$ (c=2.15, C_5H_5N), neorhusflavanone*(8,8"-binaringenin), $C_{30}H_{22}O_{10}$, mp. 274-280°, $(\alpha)_D^{28}-357^\circ$ (c=0.2, MeOH), and a new flavanoflavone, i.e. rhusflavone* (6,8"-naringeninylapigen in), $C_{30}H_{20}O_{10}$, mp. 236-238°, $(\alpha)_D^{25}-163^\circ$ (c=

Rhusflavanone

Succedaneaflavanone

Neorhusflavanone

Rhusflavone

0.39, EtOH) besides other four biflavones, agathisflavone, robustaflavone, amentoflavone and hinokiflavone were isolated.³⁹⁾

(34) The constituents of the heartwood of Garcinia multiflora Champ, Guttiferae was studi ed. A new biflavanone glucoside, GB-la-7"-O- β -glucoside* (3,8"-binaringenin-7"-O- β -glucoside), mp. 218-221°, $[\alpha]_D^{20}-31.68^\circ$ (c=2.5, EtOH) was isolated together with three known biflavanoid glucosides, spicataside, fukugiside, xanthochymuside, and seven phenolic compounds, i.e. apigenin, 1,3,6,7-tetrahydroxyxanthone, (-)-GB-la, (+)-GB-2a, (+)-volkensiflavone, (+)-morelloflavone [i.e. (+)-fukugetin], and (\pm)-morelloflavone, ⁴⁰⁾

GB-la-7"-O-β-glucoside

- (35) From the leaves of Garcinia spicata Hook, friedelin was isolated.⁴¹⁾
- (36) From the roots of *Elaeagnus oldhami* Maxim., Elaeagnaceae, maslinic acid (crataegolic acid), arjunolic acid, sitosteryl glucopyranoside and β -sitosterol were obtained.⁴²⁾
- (37) From the roots of *Glehnia littoralis* Fr. Schmidt, Umbelliferae, xanthotoxin and bergapten were isolated.⁴³⁾
- (38) Bergapten was isolated from the roots of *Peucedanum formosanum* Hay., Umbelliferae.
- (39) A new quinone, ardisianone*, C₂₄H₃₈O₅, mp. 49-54°, and a new phenol, ardisianol*, C₂₆ H₄₂O₆·1/3H₂O, mp. 82-84 were isolated from the stems and the leaves of *Ardisia quinquegona* Blume, Myrsinaceae.⁴⁴⁾

Ardisianone

Ardisianol

- (40) From the fruits of *Diospyros discolor* Willd, Ebenaceae, betulinic acid, nonacosane, hentriacontane and tritriacontane were obtained.⁴⁵⁾
- (41) The constituents of Formosan gentianaceous plants were studied. Mangiferin from the aerial portion of *Tripterospermum taiwanense* Stake, and oleanolic acid from the whole herb of *Gentiana arisanensis* Hayata were isolated, respectively. From the whole herb of *Swertia* randaiensis Hayata, gentianine and oleanolic acid were obtained.⁴⁶⁾

- (42) Two flavones have been isolated from the seeds of *Vitex rotundifolia* Linn. f., Verbe naceae, and identified to be 5-hydroxy-3, 6, 7, 3', 4'-pentamethoxyflavone(artemetin) and 5, 3'-dihydroxy-3, 6, 7, 4'-tetramethoxyflavone (casticin).⁴⁷⁾
- (43) The roots of Scutellaria rivularis Wall, Labiatae, yielded three flavonoids. They were characterized as 2',5-dihydroxy-6',7,8-trimethoxyflavone, named rivularin*, C₁₈H₁₆O₇, mp. 257-259°, 5-hydroxy-7,8-dimethoxyflavone(7-0-methylwogonin*),mp. 180-182°, and 5,7-dihydroxy-8-methoxyflavone(wogonin).⁴⁸⁾

Rivularin $R_1=R_4=OH$, $R_2=R_3=R_5=OMe$ 7-O-Methylwogonin $R_1=OH$, $R_2=R_3=OMe$, $R_4=R_5=H$

- (44) The crystalline component obtained from the essential oil of the whole plant of *Pogostemon formosanus* Oliv., Labiatae, was identified as a 10-membered ring sesquiterpene ketone, germacrone. On the basis of the spectroscopic evidence and of the orbital symmetry rule, *trans*, *trans* configuration was assigned to germacrone.
- (45) The piscicidal constituents of the whole herb of Justicia hayatai var. decumbers Yamamoto, Acanthaceae, was investigated. The six fish poison constituents were identified to be lignans, justicidin A*, C₂₂H₁₈O₇, mp. 262°, justicidin B*, C₂₁H₁₆O₆, mp. 240°, justicidin

Justicidin A R=OMe, Justicidin B R=H

C (neojusticin B) justicidin D (neojusticin A), diphyllin and chinensinaphthol methyl ether. Four non-active constituents, stearic acid, βsitosterol, KCl and KNO₃ were also isolated.⁵⁰⁾

(46) A new flavonoide, glycoside kaempferol- $3-\beta$ -D-(6''-O-p-coumaroyl) monoacetyl glucoside (tiliroside monoacetate*), $C_{32}H_{28}O_4$ ·2. $5H_2O$, mp. 178-182°, was isolated together with kaempferol- $3-\beta$ -D-(6''-O-p-coumaroyl) glucoside (i.e. tiliroside), quercetin, apigenin and kaempferol, from the flowers of *Anaphalis contorta* Hooker, Compositae. 51)

Tiliroside

(47) From the whole herb of Siegesbeckia orientalis L., Compositatae, 3,7-dimethylquerc-

etin and potassium nitrate were isolated.⁵²⁾

(48) From the whole herb of Glossogyne tenu ifolia (Labill.) Cass., Compositae, luteolin-7-0- β -D-glucoside and luteolin were obtained.⁵³⁾

(49) The constituents of the Formosan Cirsium species, Compositae were investigated. From the leaves of C. arisanense Kitamura, linarin, pectolinarin and fumaric acid were isolated.⁵⁴,⁵⁵⁾

Pectolinarin and fumaric acid from the leaves of *C. ferum* Kitamura⁵⁵⁾, luteolin-7-glucoside from the aerial portion of *C. kawakami* Hayata⁵⁶⁾, and fumaric aci from the aerial portion of *C. wallichii* DC.⁵⁶⁾ were obtained.

(50) From the tops of Eupatorium fortunei Turcz, Compositae, taraxasteryl palmitate, taraxasteryl acetate, taraxasterol, β -amyrin palmitate, β -amyrin acetate, stigmasterol, β -sitosterol, octacosanol and palmitic acid were isolated. ⁵⁷⁾

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