

· 药学前沿 ·

2017 年中国天然产物化学研究进展(上)

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摘要 本文综述我国国内学者于 2017 年在天然产物化学领域所取得的主要原创性研究成果。文中所选取的天然化合物具有较新颖的结构和/或较显著的生物活性。据此,共选取本年度发表在国内外相关刊物共计 122 篇文献中的 139 个具有代表性的化合物,对它们的来源、结构特征和生物活性进行了介绍。

关键词 天然产物;新结构;生物活性;研究进展

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2017's advanced natural products chemistry researches in China (1)

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Abstract The advanced natural products chemistry researches accomplished by Chinese domestic scholars in 2017 have been reviewed in this survey. Selected naturally occurring compounds generally have extraordinary frameworks and/or significant bioactivities. A total of 139 distinctive compounds reported in 122 peer-reviewed publications were highlighted. Their origins, structural characteristics and promising bioactivities are introduced and illustrated herein mainly based on their structural classes and novelties.

Key words natural products; novel structures; bioactivities; progress

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随着综合国力的日趋增长,我国在科技创新等方面的投资也大幅增加,部分研究领域近年得到了长足发展。就天然产物化学研究而言,我国科技工作者已成为该领域的中坚力量。从发表论文来看,中国学者的论文数量在本专业领域多个国际权威期刊中处于领先地位,如 *Journal of Natural Products*、*Phytochemistry*、*Organic Letters*、*Tetrahedron* 和 *Planta Medica* 等。以 *Journal of Natural Products* 为例,中国学者 2017 年在该刊上发表论文数量持续保持领先地位,共计 116 篇。可见中国学者近几年(特别是 2015 至 2017 年)每年在该刊发表论文数

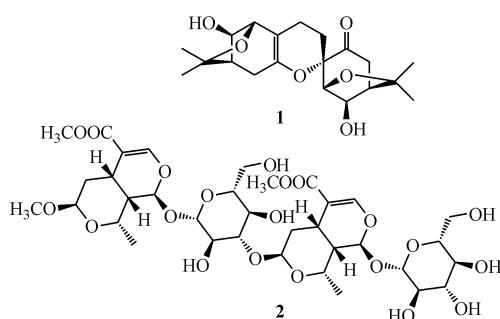
超过该刊所发表论文总数目的四分之一,论文数量连年排名第一。本文所选取的天然化合物均具有较新颖的结构和/或较显著的生物活性。现按照化合物的类型,对 2017 年度中国天然产物化学工作者的原创性研究进行综述,帮助读者了解该领域这一年的主要进展。

1 莘类

1.1 单莘

药用藤本植物香青藤(*Illigera aromatica*)与根围真菌 *Clonostachys rogersiana* 一起发酵得到薄

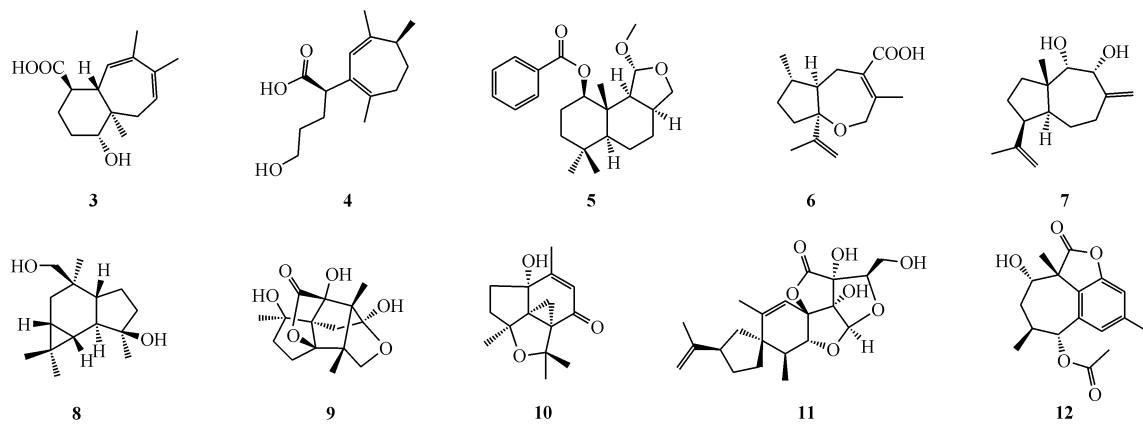
荷烷型单萜二聚体 dimericilligerate E (1)。化合物 1 对肝癌细胞 SMMC-7721 具有显著的细胞毒作用, IC_{50} 为 $(3.9 \pm 0.2) \mu\text{mol/L}$ ^[1]。从山茱萸 (*Cornus officinalis*) 的果实中得到裂环环烯醚萜苷二聚体 cornuside C (2)。研究发现化合物 2 对 HepG2 细胞中 IL-6 诱导的 STAT3 的激活具有抑制作用, IC_{50} 为 $11.9 \mu\text{mol/L}$ ^[2]。



1.2 倍半萜

从南极洲的曲霉属真菌 *Aspergillus ochraceope-taliformis* SCSIO 05702 得到 9 个蛇麻烷型倍半萜。其中, ochracenes A (3) 和 B (4) 分别具有甲基位移和环开裂形成的新碳骨架类型。此外, 化合物 4 对脂多糖 (LPS) 诱导 RAW 264.7 细胞中一氧化氮 (NO) 的生成具有抑制作用, IC_{50} 为 $(14.6 \pm 0.5) \mu\text{mol/L}$ ^[3]。从多形炭角菌 (*Xylaria polymorpha*) 发酵物的乙酸乙酯提取物中分离到补身烷 (drimane) 型倍半萜 polymorphine B (5), 该化合物具有乙酰

胆碱酯酶 (AChE) 抑制作用及 α -葡萄糖苷酶抑制作用^[4]。Daphnauranin A (6) 具有罕见的 5/7 氧杂环庚烷体系, 分离自橙花瑞香 (*Daphne aurantiaca*) 的根部。它在 $1 \mu\text{mol/L}$ 浓度对果蝇拒食率为 $(46.2 \pm 7.1)\%$ ^[5]。Forkienin A (7) 是从福建柏 (*Fokienia hodginsii*) 枝叶中分离得到的一个罕见的胡萝卜型 (carotane) 倍半萜^[6]。Plebein A (8) 是从荔枝草 (*Salvia plebeia*) 全草中发现的新骨架倍半萜^[7]。Illisimonin A (9) 具有全新的三环 [5.2.1.0^{1,6}] 壁烷骨架, 是从野八角 (*Illicium si-monsii*) 的果实分离得到。同时, 化合物 9 对缺糖缺氧所致 SHSY5Y 细胞损伤具有保护作用, EC_{50} 为 $27.7 \mu\text{mol/L}$ ^[8]。Aromaticane A (10) 为独特的同时具有环丙烷和呋喃环的新型愈创木烷型倍半萜, 是从郁金 (*Curcuma aromatica*) 的根中分离得到。该化合物对 H_2O_2 引起的 PC12 细胞氧化应激损伤具有显著保护作用^[9]。Nicotabin A (11) 具有 5/6/5/5/5 驳合环体系, 分离自烟草 (*Nicotiana tabacum*) 叶。化合物 11 可以抑制 LPS 诱导 RAW264.7 细胞中 NO 的生成, IC_{50} 为 $22.1 \mu\text{mol/L}$ ^[10]。Commiphorane C (12) 分离自俗称没药的植物树脂中, 具有独特的 5/6/7 三环骨架结构。生物活性研究显示化合物 12 可以显著减少 TGF- β 1 引起的大鼠近端肾小管上皮细胞中纤连蛋白、I 型胶原和 α -平滑肌肌动蛋白 (α -SMA) 的过量生成^[11]。

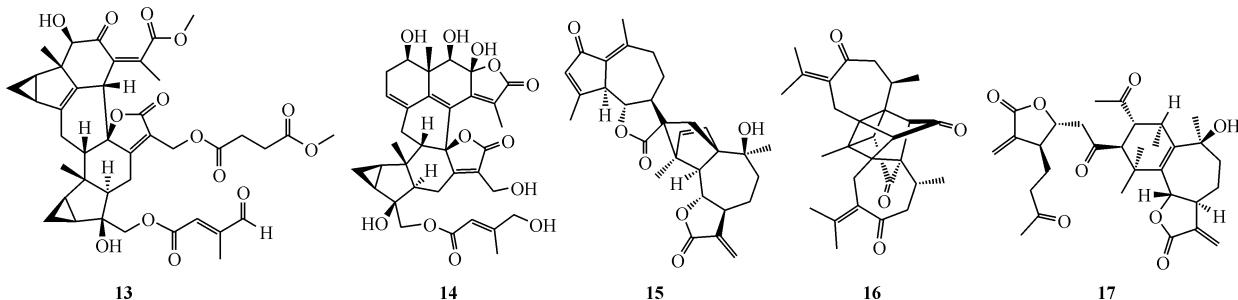


1.3 倍半萜二聚体

Fortunilide A (13) 是从丝穗金粟兰 (*Chloranthus fortunei*) 中得到的乌药烷 (lindenane) 型倍半萜二聚体。化合物 13 在纳摩尔级浓度即对氯喹耐药的恶性疟原虫显示活性, IC_{50} 为 $(5.2 \pm 0.6) \text{ nmol/L}$, 活性与青蒿素相当 ($IC_{50} = 4.0 \pm 4.2 \text{ nmol/L}$)^[12]。For-

tunoid B (14) 是从上述同一植物中发现的首例乌药烷型和桉烷型倍半萜杂合体, 显示中等抗疟活性^[13]。从萎蒿 (*Artemisia selengensis*) 中得到的 selengsisnin A (15) 是一个愈创木烷型倍半萜二聚体^[14]。Xylopiana A (16) 则是从木瓣树 (*Xylopia vielana*) 叶子中得到的又一愈创木烷二聚体^[15]。

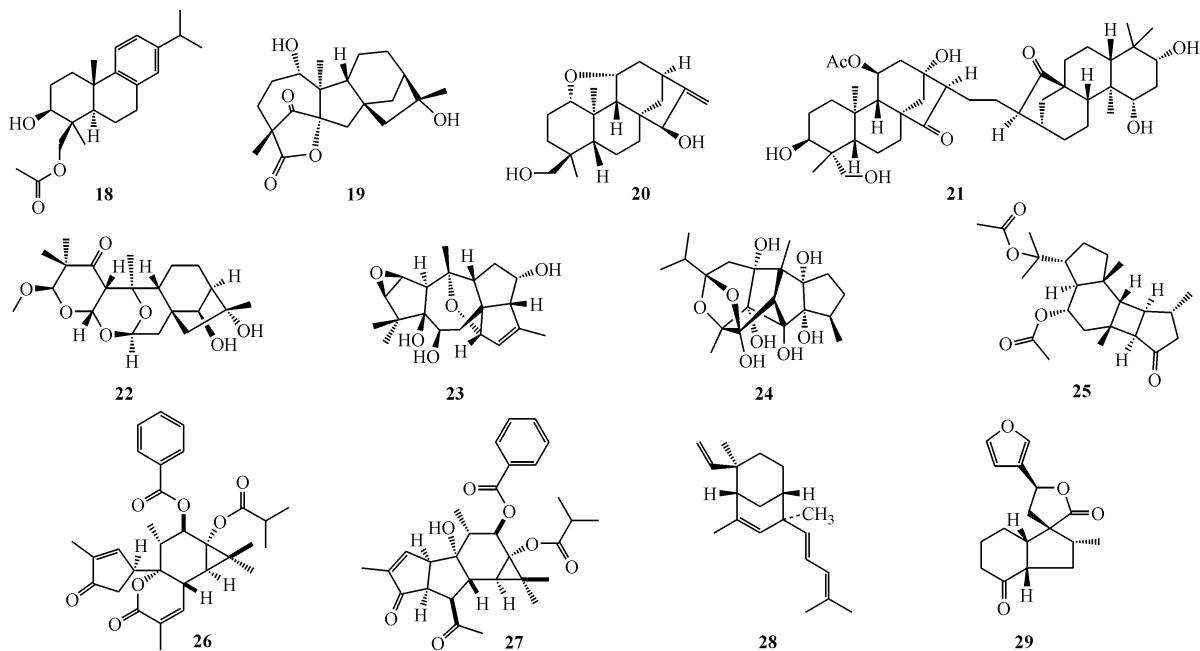
Artemisian B (**17**) 为首先罕见的推测可能由 1,10:4,5-二-断-愈创木内酯与愈创木内酯二烯经 [4+2] Diels-Alder 加成形成的二聚体, 是从传统中药艾蒿 (*Artemisia argyi*) 中分离而来。化合物 **17** 通过诱导细胞凋亡和引起 G2/M 期停止从而对乳腺癌细胞株 MDA-MB-468 显示抗增殖作用, IC_{50} 为 3.2 $\mu\text{mol/L}$ ^[16]。



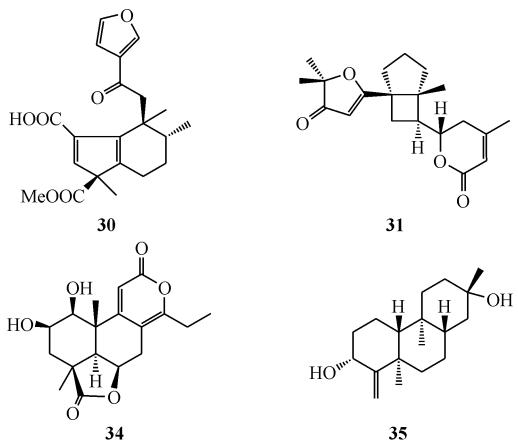
1.4 二萜

从单花莸 (*Caryopteris nepetaefolia*) 全草中分离到 nepetaefolin F (**18**)。经三磷酸腺苷-肿瘤体外药敏检测 (ATP-TCA), 发现化合物 **18** 对一种患者来源的非小细胞肺癌细胞显示比紫杉醇更强活性, IC_{50} 为 6.3 $\mu\text{mol/L}$ ^[17]。Pierisketolide A (**19**) 是从美丽马醉木 (*Pieris formosa*) 根中分离得到的 A-增-B-降-对映贝壳杉烷骨架二萜。其具有镇痛作用, 与空白对照组比较, 在 10.0 mg/kg 剂量时对醋酸引发的扭体抑制率达到 45%^[18]。Scopariusol A (**20**) 是第一个具有 1,11-醚桥特征的对映贝壳杉烷类化合物, 其与对映贝壳杉烷二聚体 scopariusol I (**21**) 同时从帚状香茶菜 (*Isodon scoparius*) 的地上部分分离得到^[19]。Rhodomollacetal A (**22**) 是从羊踯躅 (*Rhododendron molle*) 的树叶中分离得到的具有 2,

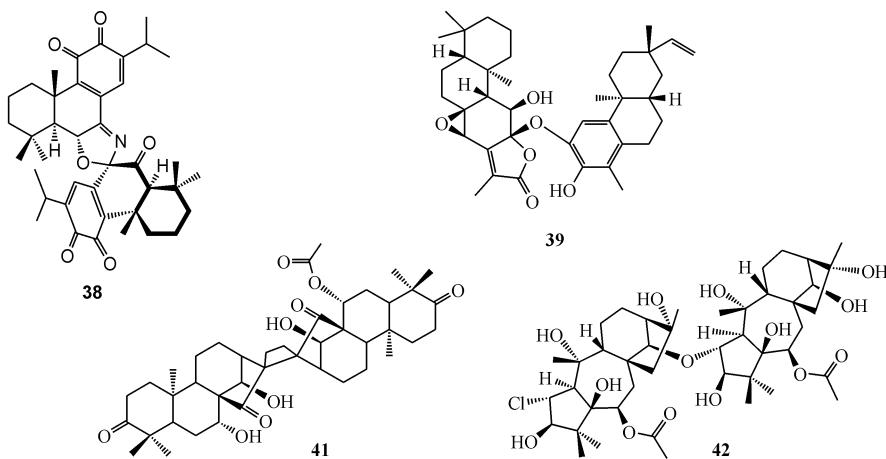
3:5,6-二-断-木藜芦烷 (grayanane) 碳骨架二萜^[20]。Rhodomollanol A (**23**) 则是从羊踯躅 (*Rhododendron molle*) 中得到的一个高度氧化的二萜, 显示中等 PTP1B 抑制作用^[21]。Cinnamomols A (**24**) 和 B 具有笼状、刚性的六环结构, 是从肉桂 (*Cinnamomum cassia*) 的树叶中分离而来。化合物 **24** 显示显著免疫刺激作用^[22]。自海绵中分离的 eleganstone A (**25**) 对大肠埃希菌、枯草杆菌和金黄色葡萄球菌显示弱抗菌作用^[23]。从蒿状大戟 (*Euphorbia dracunculoides*) 中得到的 euphordraculoates A (**26**) 和 B (**27**) 代表着两种碳骨架的巴豆烷 (tiglane) 型二萜。化合物 **27** 可以剂量、时间依赖性地抑制 Wnt 信号传导途径^[24]。Xishacorene A (**28**) 分离自西沙珊瑚 (*Sinularia polydactyla*), 它对刀豆蛋白 A (ConA) 诱导的 T 淋巴细胞增殖显示剂量依赖地提升作用^[25]。



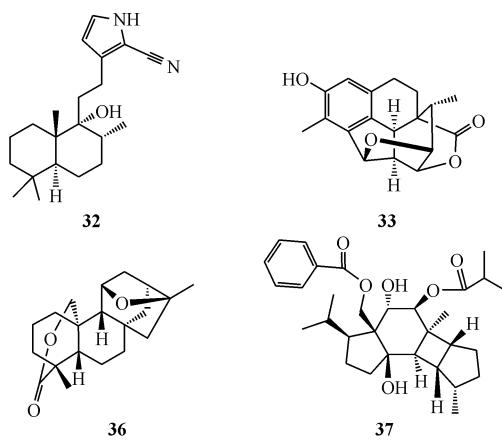
从鸡骨香(*Croton crassifolius*)根的70%丙酮提取物中得到8个新二萜成分。其中,crassin A(29)是B环重排的二萜,而crassin C(30)是A环重排的二萜^[26]。Aphapolin A(31)是从山棟(*Aphanamixis polystachya*)中分离到的nemoralisin型二萜^[27]。Vitepyrrolloid A(32)为蔓荆(*Vitex trifolia*)叶子中分离的含有2-氰基取代吡咯环结构的半日花烷型二萜,它对鼻咽癌细胞CNE1显示细胞毒活性,IC₅₀为8.7 μmol/L^[28]。Cephalanolide A(33)是从粗榧(*Cephalotaxus sinensis*)中分到的第一个A环缩环的cephalotane型降二碳二萜^[29]。2β-



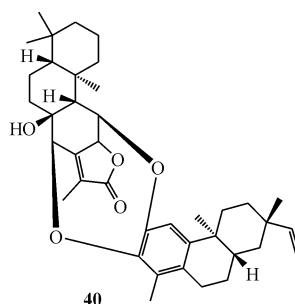
Taxodikaloid A(38)从池杉(*Taxodium ascendens*)的种子分离得到。该化合物具有全新的噁唑啉环连接两个松香烷单体的结构,并对A_B₂₅₋₃₅引起的人神经母细胞瘤细胞(SH-SY5Y)的细胞损伤具有保护作用^[33]。从月腺大戟(*Euphorbia ebracteolata*)的根中分离得到的eupractenoids A(39)和B(40)属于杂合二萜二聚体,均包含松香烷内酯单元和降玫瑰烷单元。化合物39对葡萄糖苷酶显示一定抑制



Hydroxynagilactone L(34)是从竹柏(*Podocarpus nagi*)的种子中得到的降二碳二萜,它在低于2.5 μmol/L浓度时对A2780和HEY两种肿瘤细胞显示细胞毒性^[30]。从蹄叶橐吾(*Ligularia fischeri*)全草中分离到几个二萜,其中,fischericin A(35)是具有C₁₈骨架的15,16-降二碳erythroxylane型二萜;而fischericin B(36)是第一个6/6/6/6/5/5-六环对映贝壳杉烷二萜^[31]。Vulgarisin B(37)是从药用植物夏枯草(*Prunella vulgaris*)中分得,它对A549细胞显示一定的细胞毒作用^[32]。

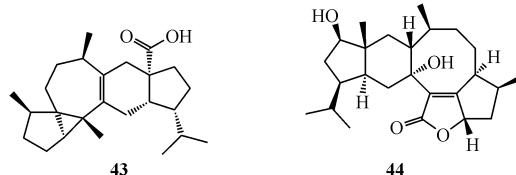


作用(IC₅₀=7.9 μmol/L),K_i为10.8 μmol/L;而化合物40可以抑制肺结核新治疗靶点结核杆菌GlmU(*Mycobacterium tuberculosis* GlmU)的乙酰基转移作用(IC₅₀=41.8 μmol/L)^[34]。Neoglaucocalyxin(41)为稀有的具有环丁烷结构的对映贝壳杉烷二聚体,是从毛叶香茶菜(*Rabdosia japonica*)的地上部分分离得到^[35]。Birhodomollein A(42)从羊踯躅的花中得到,其木藜芦烷片段出现少见的氯取代^[36]。



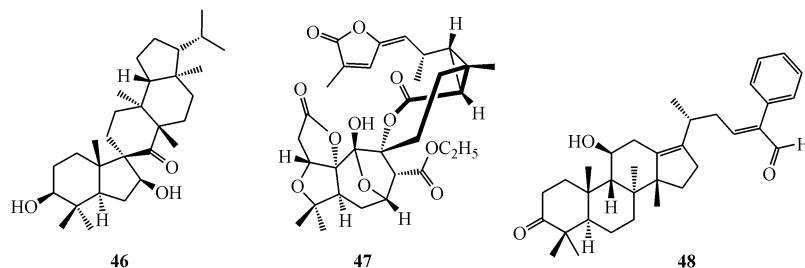
1.5 二倍半萜

从中药獐牙菜(*Swertia bimaculata*)中发现 aspterpenacid C(43)。该化合物显示一定抑制 NO 生成 ($IC_{50} = 16.1 \mu\text{mol/L}$) 及抑制 HIV-1 复制 ($EC_{50} = 1.4 \mu\text{mol/L}$) 的作用^[37]。Sesteralterin(44)是从海洋红藻 *Lomentaria hakodatensis* 表面分离到的链格孢(*Alternaria alternata* k21-1)的培养物中分离得

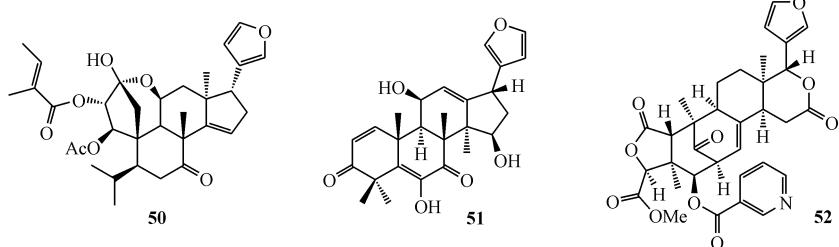


1.6 三萜

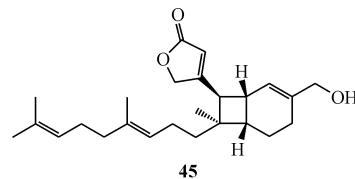
Euviscin A(46)分离自血见愁(*Teucrium viscidum*)的全草。它具有罕见的 7(8→9)-*顺*-9R-D: C-friedo-B': A'-*neo*-gammacerane 骨架^[40]。Spiroschinarin A(47)是从兴山五味子(*Schisandra incarnata*)的果实中分离到的化合物,具有独特 1-氧杂螺[6.6]十三烷主体结构^[41]。Alismanin A(48)是具



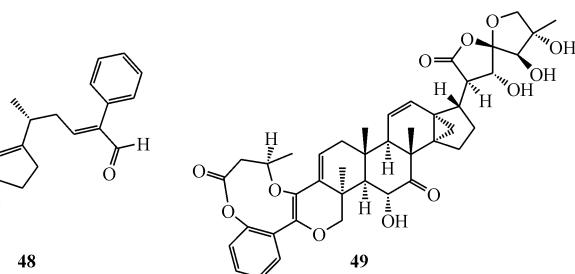
Walrobsin A(50)是从割舌树(*Walsura robusta*)的根皮中分离得到。它可显著抑制 LPS 诱导 RAW264.7 细胞中 NO 的生成, IC_{50} 为 $7.8 \mu\text{mol/L}$, 同时可以剂量依赖地抑制 iNOS 和 IL-1 β 地表达^[44]。Walsuronoid F(51)具有 18(13/14)-*顺*柠檬苦素特征,是从割舌树(*W. robusta*)的果实分离而来^[45]。Triconoids A(52)和 D(53)具有重排的柠檬苦素骨架,是从采集自尼泊尔首都地区的鹧鸪花(*Trichilia connaroides*)的枝叶中分离得到^[46]。



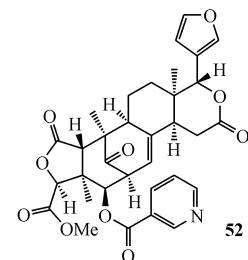
到。该化合物是第一个真菌产生的 nitidasane 型二倍半萜^[38]。(−)-Hippolide J(45)及其对映体是从中国南海采集的茸毛马海绵(*Hippopspongia lachne*)中分离而来。二者对医院来源的三种致病真菌(*Candida albicans* SC5314、*Candida glabrata* 537 和 *Trichophyton rubrum* CMCCFLA)显示强抗菌作用, MIC_{50} 为 $0.1 \sim 0.3 \mu\text{g/mL}$ ^[39]。

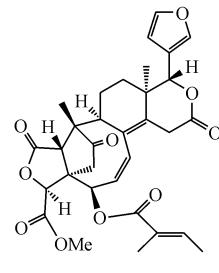


有 C_{34} 骨架的芳香萜,是从东方泽泻(*Alisma orientale*)中分离得到。该化合物在 10 nmol/L 对孕烷受体 X 具有显著激活作用^[42]。从海南叶下珠(*Phyllanthus hainanensis*)中得到几个高度修饰的三萜。其中, phainanolide A(49)对 HL-60 细胞显示显著的细胞毒作用, IC_{50} 为 $0.08 \mu\text{mol/L}$ ^[43]。

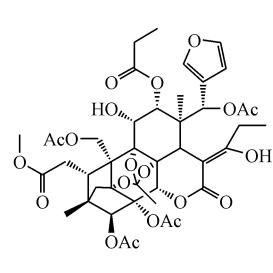


Chukvelutilide Y(54)为一个 C-15 烯醇酰化 phragmalin 型柠檬苦素原酸酯,分离自麻棟(*Chukrasia tabularis*)种子的乙醇提取物^[47]。Xylomexicanin I(55)为 B 环和 C 环桥连的降四碳三萜,是从中国红树木果棟(*Xylocarpus granatum*)的种子分离而来^[48]。Krishnadimer A(56)为一个 C_2 对称的柠檬苦素二聚体,属于一类轴向手性非联苯天然产物,是从中国红树木果棟属植物 *Xylocarpus moluccensis* 的种子中分离而来^[49]。

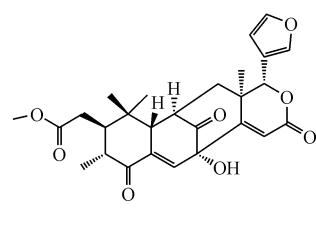




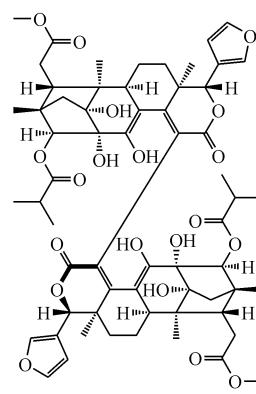
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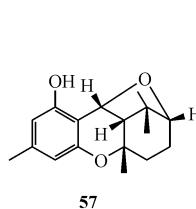
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1.7 杂 菇

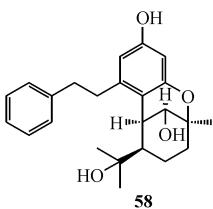
(+)-Rhodonoid C (**57**) 和 (-)-rhodonoid C 是从头花杜鹃(*Rhododendron capitatum*)的地上部分得到的一对杂萜对映体, 是首次发现的含有 6/6/6/5 四环体系的杂萜。化合物 **57** 对 1 型单纯疱疹病毒 (HSV-1) 显示出体外拮抗作用^[50]。(\pm)-rasumatranin A (**58**) 为一对以双苄基为基础的杂萜对映体, 是从附生的中国苔类植物 *Radula sumatrana* 中分离得到^[51]。从芒果球座菌(*Guignardia mangiferae*)中得到 7 个含有螺环体系的单萜-莽草酸酯结合而成的杂萜。其中, manginoid A (**59**) 为第 1 个具有桥连环己二酮片段的螺环杂萜; Manginoid E (**60**) 是具有 2,4-二氧杂三环[3.3.1.0^{3,7}]壬烷片段杂萜。而且, 化合物 **59** 对 1 型 11 β -羟基类固醇脱氢酶 (11 β -HSD1) 显示抑制作用, IC₅₀ 为 0.8 $\mu\text{mol/L}$ ^[52]。Frutescone A (**61**) 是从岗松(*Baeckea frutescens*)的地上部分得到的基于 tasmanone 的杂萜, 显示中等细胞

毒作用^[53]。Chrysogenolide A (**62**) 是从蛇足石杉(*Huperzia serrata*)内生菌产黄青霉(*Penicillium chrysogenum* MT-12)的固态培养物中分离得到^[54]。

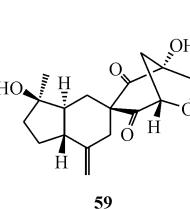
Ganoleucin A (**63**) 是从西藏灵芝(*Ganoderma leucocontextum*)的子实体中分离而来, 它对酵母菌和小肠黏膜来源的 α -葡萄糖苷酶具有强大非竞争性抑制作用^[55]。Spiroaspertrione A (**64**) 属于萜-聚酮杂合的螺环杂萜化合物, 是从曲霉属真菌 *Aspergillus* sp. TJ23 的培养物中分离而来。它可以使苯唑西林对耐甲氧西林金黄色葡萄球菌(MRSA)的最小抑制浓度 (MIC) 从 32 $\mu\text{g/mL}$ 降低到 1 $\mu\text{g/mL}$, 显示强再致敏作用^[56]。Chartarolide A (**65**) 为苯基螺环补身烷衍生的杂萜, 自海绵真菌 *Stachybotrys chartarum* WGC-25C-6 中得到分离。它对一小组人肿瘤细胞显示显著细胞毒作用, 同时对与人肿瘤细胞相关的蛋白激酶 FGFR3、IGF1R、PDGFRb 和 TrKB 均显示强抑制作用^[57]。



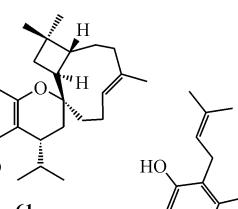
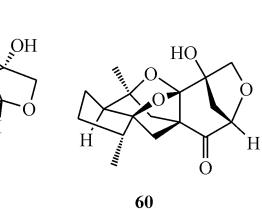
57



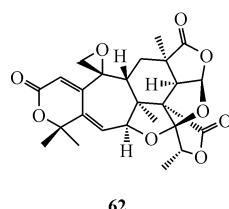
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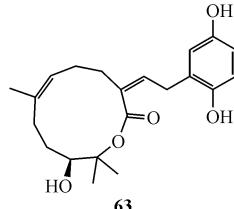
60



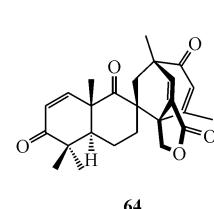
61



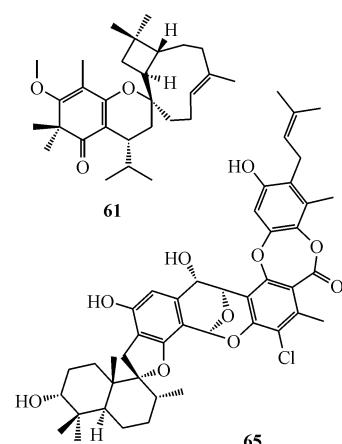
62



63



64



65

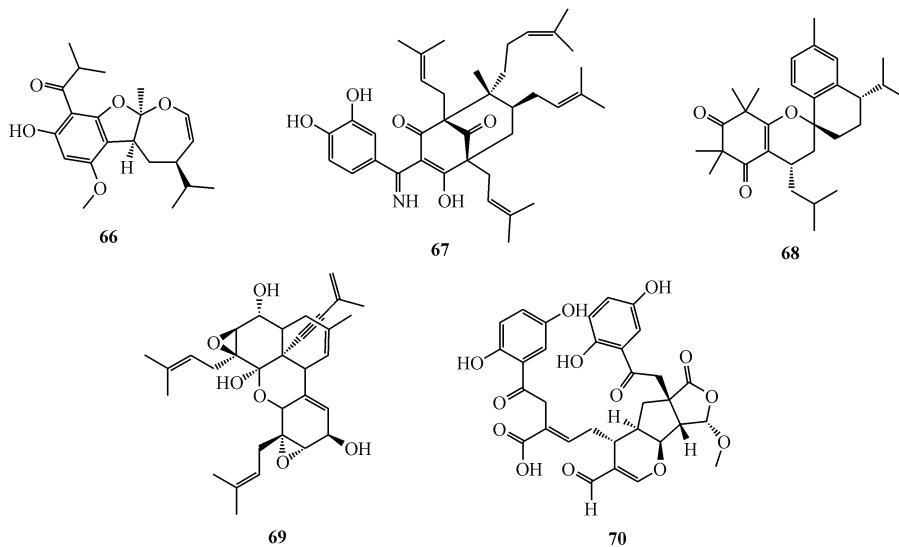
1.8 间苯三酚-萜加成物

Viminalin F (**66**) 是分离自串钱柳(*Callistemon*

viminalis) 果实中的酰基间苯三酚和-水芹烯的杂合体, 具有中等细胞毒作用^[58]。Garciyunnanimine A

(**67**)为多环多异戊烯基化的酰基间苯三酚亚胺,是从云南藤黄(*Garcinia yunnanensis*)中分离而来^[59]。从桃金娘(*Rhodomyrtus tomentosa*)的叶子中得到数个杂萜。其中,tomentodione E(**68**)是首例基于 β -calacorene的杂萜^[60]。Dimericbiscognienyne A(**69**)为少见的二异戊烯基化的环己烷杂萜二聚体,是从苔藓*Usnea mutabilis*分离出的真菌

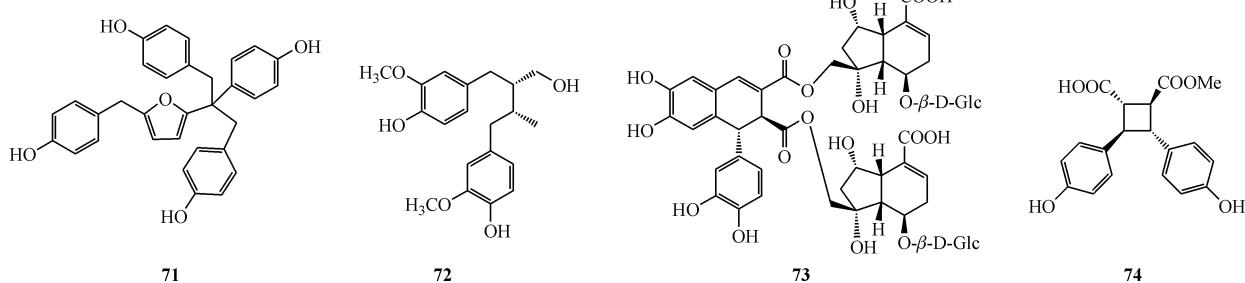
Biscogniauxia sp. (No. 71-10-1-1)中分离而来^[61]。从平盖灵芝(*Ganoderma applanatum*)的子实体中分离到三对光学对映杂萜二聚体,全部证实为JAK3的抑制剂。此外,光学对映体applanatumine B(**70**)对盘状结构域受体1(DDR1)显示抑制作用,IC₅₀分别为8.2和6.9 $\mu\text{mol/L}$ ^[62]。



2 木脂素

Gastradefurphenol(**71**)是从天麻(*Gastrodia elata*)根茎的水提物中得到的具有新骨架的9,9'-新木脂素^[63]。Schibitubin B(**72**)是从瘤枝五味子(*Schisandra bicolor* var. *tuberculata*)的果实中分离得到。与阴性对照物相比,化合物**72**对A β_{25-35} 诱

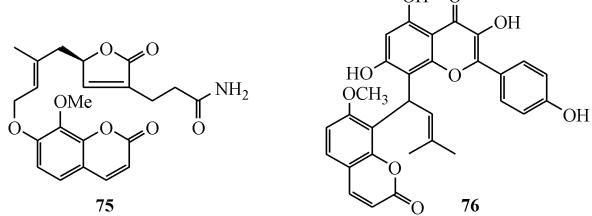
导的SH-SY5Y细胞损伤具有显著的保护作用^[64]。从南烛(*Vaccinium bracteatum*)叶子中得到的化合物**73**是一个木脂素-裂环环烯醚萜糖苷二酯类化合物^[65]。化合物anisumic acid(**74**)为 δ -truxinate衍生物,是从细叶黄皮(*Clausena anisum-olens*)中分离得到^[66]。



3 香豆素

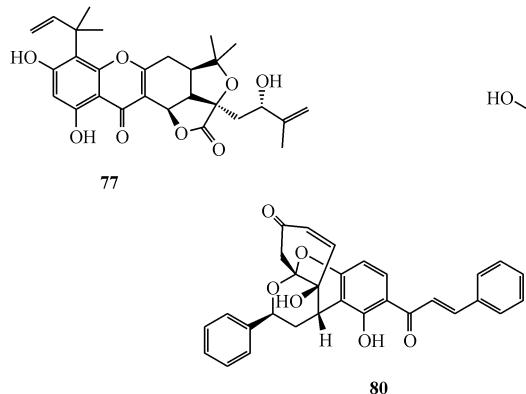
Anisucoumaramide(**75**)是从细叶黄皮(*Clausena anisum-olens*)中分得。它对人单胺氧化酶的同工酶显示高度选择性,在纳摩尔浓度即显示抑制活

性^[66]。从蛇床(*Cnidium monnierii*)的果实中分离到消旋混合物(\pm)-enidimonin A(**76**)。与对应的光学对映体相比,其消旋体对HSV-1显示更强的抗病毒作用,IC₅₀为1.2 $\mu\text{mol/L}$ ^[67]。



4 黃 酮

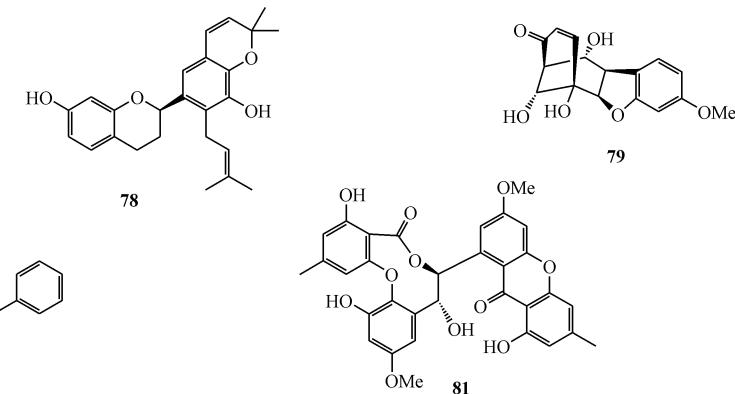
Neobraclactone A (77) 从大苞藤黃 (*Garcinia bracteata*) 的叶子中得到分离, 其对白血病细胞 HL-60 和 K562 具有显著抑制作用, IC_{50} 分别为 0.5 和 0.4 $\mu\text{mol}/\text{L}$ ^[68]。从黄瑞香 (*Daphne giraldii*) 的茎和根皮中得到 4 对异戊烯基化的黄烷对映异构体。分离物对肝癌细胞均显示选择性细胞毒作用。其



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中, (2R)-kazinol B (78) 比其消旋体对 Hep3B 细胞显示更强的抑制作用^[69]。Sophopterocarpan A (79) 是从苦参 (*Sophora flavescens*) 的根中分离而来。它具有一定自吞噬激活作用并对 MCF-7 细胞显示细胞毒性^[70]。Caesalpinnone A (80) 为黄烷和查尔酮的杂合体, 是从九羽见血飞 (*Caesalpinia enneaphylla*) 的茎和叶中得到分离。化合物 80 对 HL-60、SMMC-7721、A549、MCF-7 和 SW-48 5 种人肿瘤细胞显示较强细胞毒作用, IC_{50} 范围为 0.5 ~ 0.9 $\mu\text{mol}/\text{L}$ ^[71]。(+)-Ascomlactone A (81) 和 (-)-ascomlactone A 为一对光学对映聚酮二聚体, 是从红树内生菌 *Ascomycota* sp. SK2YWS-L 分离而来。两者对-葡萄糖苷酶都有强抑制作用, IC_{50} 分别为 63.7 和 27.9 $\mu\text{mol}/\text{L}$ ^[72]。



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