藏药余甘子与诃子化学和药理作用比较*

亓 旗, 崔雅萍, 梁文仪, 陈文静, 李 师, 吴玲芳, 张兰珍**

（北京中医药大学中药学院 北京 100102）

摘 要:余甘子和诃子均为常用藏药, 藏药中余甘子、毛诃子、诃子 3 种药物配伍使用称为大三果, 是大多数常用方剂的基础方。本文对余甘子和诃子化学成分与药理作用进行归纳总结和比较, 为两种常用藏药的应用提供依据。

关键词:藏药 诃子 余甘子 化学成分 药理作用

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余甘子是大戟科叶下珠属植物余甘子 *Phyllanthus emblica* L. 的干燥成熟果实, 又名油柑、庵摩勒、余甘、油甘子、滇橄榄等[1], 主要分布于印度、马来西亚等热带和亚热带国家以及中国的海南、福建、广东、广西、云南等地。余甘子归肺、胃经, 性味甘、酸、涩、凉, 具有清热凉血, 消食健胃, 生津止咳之功效, 临床用于血热血瘀, 消化不良, 腹胀, 咳嗽, 喉痛, 口干等症[2]。

诃子是使君子科榄仁树属植物诃子 *Terminalia chebular* Retz. 及其变种绒毛诃子 *Terminalia chebula* Retz. var. *tomentella* Kurt. 的干燥成熟果实, 名诃黎勒、诃梨, 蒙药名为“阿如拉”, 泰语称“麻菜果”, 原产于印度、缅甸, 在中国主要分布于云南、广东、广西、西藏等地。诃子归肺, 大肠经, 具有涩肠止泻、敛肺止咳、降火利咽之功效[2]。

余甘子和诃子在藏药中应用广泛, 余甘子、诃子和毛诃子组成的大三果常作为基础方与其它藏药配伍使用, 藏药标准 205 种复方中含有三果的复方有 45 种, 占总数的 21.9%, 其中三果作为主药的复方有 6 种。在印度天竺阿育吠医药体系和藏医药体系中, 三果医药利用率均列为第一[3]。藏医药中诃子的使用频率与汉医方剂中甘草相当, 故有“众药之王”之称。

目前, 对余甘子和诃子的化学、药理作用研究较多, 但没有对余甘子和诃子的化学成分、药理作用进行比较系统的报道。本文对近年来两者的化学成分与药理作用进行总结, 以期为余甘子与诃子的深入研究提供参考。

1 化学成分

余甘子和诃子所含化学成分的主要类别相近, 均主要含有鞣质、酚类、苯丙素和黄酮类和三萜类化学成分, 在生物碱、挥发油和维生素等化学成分的分布方面有不同之处。

1.1 诃子、余甘子中共有化学成分

①多酚类成分:Methyl gallate, gallic acid 和 ethyl gallate, chebulic acid, ellagic acid;②可水解鞣质类:Chebulanin, corilagin 和 chebulagic acid;③甾醇类:*β*-sitosterol 和 daucosterol;④黄酮类成分:Quercetin;⑤苯丙素类成分:Cinnamic acid[4-17]。

1.2 余甘子中的化学成分

1.2.1 酚类

余甘子中酚类化合物主要有 mucic acid 1,4-lactone 3,5-di-*O*-gallate, mucic acid 1,4-lactone 3-*O*- gallate, mucic acid 1,4-lactone 5-*O*-gallate, mucic acid 1,4-lactone 6-methyl ester 2-*O*- gallate mucic acid 1,4-lactone 6-methyl ester 5-*O*-gallate,

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** 通讯作者:张兰珍, 本刊编委, 研究员, 博士生导师, 主要研究方向:中药和民族药药效物质研究。

mucic acid 1-methyl ester 2-*O*-gallate, mucic acid 2-*O*-gallate, mucic acid 3-*O*-gallate, mucic acid 6-methyl ester 2-*O*-gallate, mucic acid di- methyl ester 2-*O*-gallate, flavogallonic acid bislactone, Gallic acid 3-*O*-(6′-*O*-galloyl)-*β*-D-glucoside, gallic acid 3-*O*-*β*-D-glucoside[12], (-)-epigallocatechin 3-*O*-gallate, 1-*O*-galloyl-*β*-D-glucose, L- malic acid 2-*O*-gallate, mucic acid 1,4-lactone 2-*O*-gallate, 2-(2-methylbutyryl) phloroglucinol 1-*O*-(6″-*O*-*β*-D-apiofuranosyl)-*β*-D-glucopyranoside, 2,6-dimethoxy-4-(2-hydroxyethyl)pheno l1-*O*-*β*-D- glucopyranoside, 2-carboxylmethylphenol 1-*O*-*β*-D- glucopyranoside, 4-hydroxy-3-methoxybenzaldehyde, methyl-4-hydroxybenzoate, 3,4,8,9,10 -pentahydroxy-dibenzo[b,d]pyran - 6-one, syringaldehyde, vanillic acid, 3-ethylgallic acid, 4-*O*-methylellagic acid-3′-*α*- rhamnoside, pyrogallol[18-23]。

1.2.2 鞣质

可水解鞣质有 phyllanthunin, 1,6-di-*O*-galloyl-*β*-D-glucose, carpinusnin, mallonin, neochebulagic acid, phyllanemblinin A, phyllanemblinin B, phyllanemblinin C, phyllanemblinin D, phyllanemblinin E, phyllanemblinin F, punicafolin, putranjivain A, tercatain, furosin, geraniin, isocorilagin, putranjivain B, isomallotusinin], isostrictinin, 1,2,4,6-tetra-*O*-galloyl-*β*-D-glucose[24]。缩合鞣质主要有 prodelphinidin B1, prodelphinidin B2, epicatechin-(4*β* → 8)-epigallocatechin, Phyllemtannin。

1.2.3 萜类

三萜类化合物有 lupeol, 3,20-dioxo-dinorfriedelane。倍半萜类有 phyllaemblic acid B, phyllaemblic acid C, phyllaemblicin D, phyllaemblinol[19, 20], phyllaemblicin A, phyllaemblicin B, phyllaemblicin C, phyllaemblic acid, phyllaemblicin E, phyllaemblicin F, 4′-hydroxyphyllaemblicin B, phyllaemblicin G1, phyllaemblicin G2, phyllaemblicin G3, phyllaemblicin G4, phyllaemblicin G5, phyllaemblicin G6, phyllaemblicin G7, phyllaemblicin G8, glochicoccin D, *β*-caryophyllene, *β*-bourbonene[25-28]。

1.2.4 黄酮类

黄酮类化合物主要有 (-)-epiafzelechin, (-)-epicatechin, (-)- epigallocatechin, prodelphinidin B-2 3′-*O*-gallate, (+)-gallocatechin, (S)-eriodictyol 7-*O*-(6″-*O*-(E)-*β*-coumaroyl)-*β*-D-glucopyranoside, (S)-eriodictyol7-*O*-(6″-*O*-galloyl)- β -D- glucopyranoside, Isoquercitrin, Kaempferol, kaempferol-3-*O*-*β*-D- glucopyranoside, quercetin-3-*O*-*β*-D-glucopyranoside, kaempferol-3-*O*-*α*-L-(6″-ethyl)-rhamnopyranoside, Avicularin, kaempferol-3-*O*-*α*-L-(6″-methyl)-rhamnopyranoside, apigenin-7-*O*-(6″-butyryl-*β*-glucopyranoside)[29-32]。

1.2.5 甾醇类

甾醇类成分主要有 5*α*, 6*β*-dihydroxysitosterol, 5*α*, 6*β*, 7*α*-trihydroxysitosterol, 6′-(stigmast-5-en-7-one-3-O-*β*-D- glucopyranosidyl) hexadecanoate, 7-ketositosterol, 7*α*-hydroxysitosterol, 7*α*- acetoxysitosterol, 7*β*-ethoxysiterol, 6′-(stigmast-5-en-3-*O*-*β*-D-glucopyranosidyl) hexadecanoate, stigmast-4-en-3-one, stigmast-4-en-3,6-dione, stigmast-4-en-6*β*-ol-3-onestigmast-4-ene-3*β*, 6*α*-diol。

1.2.6 苯丙素类

苯丙素类化合物主要有 Isolariciresinol, 4-ketopinoresinol, lirioresinol A, medioresinol, syringaresinol, 4,9,9′-trihydroxy-3,4′-dimethoxy-8-*O*-3′-neolignan, coniferyl aldehyde, methyl caffeate。

1.2.7 其他类

其他化合物包括有 5-hydroxymethylfurfural, mucic acid 1-methyl ester-6-ethyl ester, penicillide, purpactin A, methyl (2S)-1-[2-(furan-2-yl)-2-oxoethyl]-5-oxopyrrolidine-2- carboxylate, 1′S-11-dehydroxy penicillide, 2R-diethyl malate, 5-hydroxy-isoquinoline, mucic acid[33] 等。

1.3 诃子的化学成分

1.3.1 酚类

诃子中酚类成分主要有 phloroglucinol, Ferulic acid, pyragallol, p-coumaric acid, caffeic acid, vanillic acids, anthraquinone, protocatechuate, 3,6-di-*O*-galloyl-D-glucose, 6-*O*-galloyl-D-glucose, (-)-shikimide-4-*O*-gallate, (-)-shikimic acid 3-*O*-gallate+(-)-shikimic acid 5-*O*-gallate, 1,2,6-tri-*O*-galloyl-*β*-D-glucose[11], shikimic acid, 莽草酸甲酯, isoterchebulin, 1,3,6-tri-*O*-galloyl-*β*-

D-glucopyranose[18], galloylglucose, punicalagin[34, 35]。

1.3.2 萜类

三萜类成分有 arjunin, 2α-hydroxymicromeric acid, maslinic acid, 2α-hydroxyursolic acid, terminoic acid, Arjungenin, arjunolic acid, 诃五醇, chebuloside I, chebuloside II, arjunglucoside I, arjunglucoside II, arjunetin, bellericoside, arjunic acid, terminolic acid, 2α,19α- 二羟基 -3-O-12-en-28- 乌苏酸 -O-α-L- 鼠李糖 -O-β-D- 葡萄糖苷, eta-amyrin[36-40]。

1.3.3 鞣质类

鞣质类成分有 1,6-di-O-galloyl-D- glucose, 3,4,6-tri-O-galloyl-D-glucose, Casuarinin, 1,2,3,4,6-penta-O-galloyl- D-glucose, 1,3,4,6-tetra-O-galloyl-β-D-glucose, 1,3,6-tri -O-galloyl-β-D-glucose, Terchebin, chebulinic acid, 2,3,4,6-penta-O-galloyl-β-D-glucose, Methyl neochebulinate, neochebulinate, neochebulinic acid, terflavin A, terflavins B, terflavins C, terflavins D, punicalin, terchebulin, isochebulic acid, neochebulic acid 等。

1.3.4 其他

诃子中还包括有槲皮素 -3-O- 鼠李糖苷, 5,7,2-tri-O-methylflavanone-4-O-β-D- galactopyranosyl-(l-3) -β-D-glucopyranoside, Friedelin。

2 药理作用

2.1 余甘子、诃子共有的药理作用

2.1.1 抗氧化作用

研究发现, 余甘子、诃子均具有明显的抗氧化活性, 其抗氧化活性与其中的酚类化合物相关。余甘子果实中的槲皮素、5- 羟甲基糠醛、没食子酸、β- 胡萝卜苷和鞣花酸具有显著清除 DPPH 和 $ABTS^+$ 自由基的活性。另外, 余甘子中维生素 C 的含量特别高, 这也与其较强的抗氧化活性密不可分。吴士云[41]采用 FRAP 法和 DPPH 清除率评价诃子, 发现诃子乙酸乙酯部位抗氧化活性最强, 其强抗氧化活性主要与高含量总多酚类化合物有关。

2.1.2 抗病原微生物作用

余甘子、诃子具有广泛的抑菌以及抗病毒作用。余甘子的乙醇提取物对金黄色葡萄球菌、大肠杆菌、枯草芽孢杆菌、变形杆菌和嗜热脂肪芽孢杆菌有很强的抑制作用。乙酸乙酯提取物、甲醇提取物和水提物在体外对于单纯疱疹病毒 1 型和 2 型均有一定的抑制作用, 其中乙酸乙酯提取物的活性最强[42, 43]。

诃子对金黄色葡萄球菌、大肠杆菌、绿脓杆菌、白假丝酵母菌、铜绿假单胞菌、白色念珠菌和解脲脲原体等具有抑制作用, 其水提物对皮肤真菌也有一定的抑制作用, 诃子乙酸乙酯提取物的抗菌活性与邻苯三酚类成分相关。高浓度的诃子复方提取物可显著性降低 A 型流感病毒 H3N8 的感染率。诃子酸和石榴皮鞣素还能通过阻断 HIVrgp120 与 CD4 细胞的结合从而抑制病毒复制[44-46]。

2.1.3 抗肿瘤作用

余甘子、诃子均可抑制多种癌细胞的增殖。余甘子中的酚类化合物有显著的抗肿瘤活性。余甘子提取物可强烈抑制人胃腺癌细胞 MK-1、人子宫癌细胞 HeLa 细胞、鼠黑素瘤细胞 B16F10、乳腺癌细胞 MCF-7、人肝癌细胞 HepG2 和人肺癌细胞 A549 的增殖。余甘子果汁对 S180 荷瘤小鼠灌服不仅能抑制肿瘤生长, 还具有较好的免疫调节和免疫保护作用。诃子醇提物可抑制人乳腺癌细胞株 MCF-7、鼠乳腺癌细胞株 S115、人前列腺癌细胞株 PC-3、人骨肉瘤细胞株 HOS-1 和 PNT1A 人前列腺癌细胞的增殖[47, 48]。

2.1.4 保肝作用

余甘子醇提取物能减少 TNF-α、TGF-β、HA、P Ⅲ NP、LN、MDA 和 Hyp 含量, 提高 SOD 活性, 对肝纤维化大鼠有较好的抗肝纤维化作用。朱伟等发现余甘子可改善非酒精性脂肪肝形成过程中的肝脏损伤, 也可增强肝损伤大鼠肝细胞的恢复能力[49-51]。

诃子的 70% 乙醇提取物能显著改善肝损伤鼠体外铁螯合性能和体内铁过剩, 减低肝损伤鼠的肝毒性。诃子炮制品能够降低由 α- 萘异硫氰酸酯诱导肝损伤大鼠血清中 AST 和 ALT 水平, 对急性肝损伤具有降酶保肝作用, 且毒性较小[52]。

2.1.5 抗炎作用

余甘子、诃子对不同炎症模型均有一定的抑制作用。余甘子甲醇部位、乙酸乙酯部位、石油醚部位和正丁醇部位为余甘子抗炎镇痛的有效部位, 其中以甲醇部位与正丁醇部位的作用最强 ;还可显著降低毛细血管通透性, 抑制白细胞游出, 其抗炎作用成剂量依赖性[53, 54]。诃子水提物可显著抑制二甲苯所致小鼠耳肿胀的程度。阿育吠陀传统草本配方三果汤 (诃子、毛诃子和余甘子) 对弗氏佐剂诱导的 Wistar 大鼠关节炎模型有抗炎作用。

2.1.6 对血液系统的作用

余甘子、诃子均有降血糖、调血脂的作用。余甘子水提物对四氧嘧啶诱发的小鼠糖尿病模型具有明显的降血糖作用。二十五味余甘子丸对脂肪乳诱发大鼠高脂血症脂代谢具有调节作用[55,56]。

诃子醇提物可降低糖尿病大鼠的血糖水平、还具有调节血脂的作用。诃子幼果的乙酸乙酯、正丁醇和水部位对 α- 葡萄糖苷酶均有明显的抑制作用，活性跟踪表明 5- 羟甲基糠醛、没食子酸甲酯为活性单体[57-59]。

2.1.7 对胃肠道的作用

余甘子的传统运用既有抗便秘作用，又有抗腹泻作用。现代药理研究采用科学方法证实余甘子的止泻作用主要是由于毒蕈碱受体和 Ca^{2+} 通道双重阻断，而抗便秘的活性可能通过激活毒蕈碱受体而介导。余甘子的双重调节作用可能是由于不同剂量导致的，低剂量兴奋毒蕈碱受体，高剂量抑制毒蕈碱受体[60,61]。

诃子具有抑制胃肠道蠕动的作用，其提取物的高、中剂量均可显著抑制正常小鼠胃排空与小肠推进运动，还可抑制家兔离体肠平滑肌的收缩，对氯化乙酰胆碱引起的肠平滑肌兴奋有明显的拮抗作用，均表明诃子醇提物对动物胃肠运动有抑制作用，其作用途径可能与 M 胆碱受体有关[62]。

2.2 余甘子与诃子不同的药理作用

2.2.1 余甘子药理作用

（1）抗突变：可明显降低骨髓细胞染色体的畸变，其中 Vc 是其主要抗突变成分；（2）抗主动脉粥样硬化：能抑制高脂血症家兔主动脉粥样硬化的形成；（3）抗疲劳：可延缓模拟高原环境下小鼠疲劳的产生，可加速疲劳消除[63,64]。

2.2.2 诃子药理作用

（1）镇咳：对豚鼠咳嗽有较强的镇咳作用且效果强于可待因；（2）镇痛：能够减少醋酸扭体模型中小鼠的扭体次数；（3）解毒：与甘草、草乌共煎后，可降低次乌头碱的含量，在相同配伍比例下，诃子 – 草乌共煎液解毒活性强于甘草 – 草乌共煎液[65,66]；（4）心脏保护：在离体蛙心脏模型中具有心脏保护作用；（5）细胞保护：对HEK-N/F细胞具有显著的保护作用；（6）促进伤口愈合：可促进大鼠皮肤伤口愈合，缩短皮肤愈合时间[68, 69]。

3 总结

余甘子和诃子均为常用藏药，是藏药大三果中的两个主药，具有很多相同的化学成分和药理作用，它们在抗氧化、抗病原微生物、抗肿瘤以及调节肠胃功能和血液系统等方面均有较好的作用。但是，它们的功效也有区别，余甘子侧重于抗疲劳、抗突变以及抑制主动脉粥样化的形成，诃子在镇咳、镇痛、解毒、促进伤口愈合以及心脏保护、细胞保护等方面更有优势，二者均未见两者毒副作用的记载与报道。因此，余甘子和诃子在疾病预防、治疗和保健品的开发等方面有广泛前景。

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The Phytochemical and Pharmacological Progress on Comparisons Between *Terminalia Chebula* Retz. and *Terminalia Chebula* Retz. in Tibetan Medicine

Qi Qi, Cui Yaping, Liang Wenyi, Chen Wenjing, Li Shi, Wu Lingfang, Zhang Lanzhen
(School of Chinese Materia Medica, Beijing University of Chinese Medicine, Beijing 100102, China)

Abstract: *Phyllanthus emblica* L. and *Terminalia chebula* Retz. were the most common Tibetan medicines. The combination of *Phyllanthus emblica* L., *Terminalia chebula* Retz. and *Term inalia bellirica* (Gaertn.) Roxb. was known as Triphala, which was the basis of the most frequently–used prescriptions. The present study summarized and made a further comparison between *Phyllanthus emblica* L. and *Terminalia chebula Retz*. over chemical constituents and pharmacological activities, which provided evidence for their clinical use and the basic theory.
Keywords: Tibetan medicine, *Phyllanthus emblica* L., *Terminalia chebula* Retz., chemical constitution, pharmacological action

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