African Journal of Pure and Applied Chemistry Vol. 5(12), pp. 412-416, 30 October, 2011
Available online at http://www.academicjournals.org/AJPAC
ISSN 1996 - 0840 ©2011 Academic Journals

Review

Chemical constituents and biological activities of the genus *Zanthoxylum*: A review

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Accepted 4 June, 2011

Zanthoxylum **known as "Timoor" is used as mouth fresh, tooth care, spice and in intoxicating of the fishes. The present study deals with documentation of isolated chemical constituents and biological activities of the genus *Zanthoxylum*. The chemical constituents and essential oil have been isolated and characterized by several analytical methods. Alkaloids and cumarins are the main constituents of the genus. The presence of alkaloids and essential oil, *Zanthoxylum* possessed several types of biological activities such as larvicidal, anti-inflammatory, analgesic, antinociceptive, antioxidant, antibiotic, hepatoprotective, antiplasmodial, cytotoxic, antiproliferative, anthelminthic, antiviral and antifungal.**

Key words: *Zanthoxylum*, alkaloids, essential oil, biological activities.

INTRODUCTION

Zanthoxylum are deciduous shrubs and trees of the family Rutaceae which comprise 250 species that are native to warm temperate and subtropical region of the world. The genus has much ethanobotanical importance and is used as sources of pharmaceutical and cosmetics raw material. Traditionally, leaves and fruits are used for mouth fresh and tooth care while bark is used for intoxicating the fishes (Gaur, 1999). Leaves, fruits and barks are used as spice (Samant and Dhar, 1997). Alkaloids were isolated from *Zanthoxylum* (Rodriguez-Guzman et al., 2010; Yang et al, 2008; Liang et al., 2006; Martin et al., 2005). Twenty microelements were also determined in the root, stem and leaf of traditional Chinese herbs *Zanthoxylum nitidum* by ICP-AES (Qin et al., 2009). Due to presence of nitrogen containing compound, *Zanthoxylum* has wide spectrum of biological activities. The purpose of this review to collect all the possible information regarding the chemical constituents and biological effects of the genus *Zanthoxylum*, thus this will help to researchers and pharmaceutical companies to take action in this discipline.

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CHEMICAL CONSTITUENTS

2,4-dihydroxypyrimidine, syringic acid, 2,6-dimethoxy-1,4-benzoquinone, 4-hydroxybenzoic acid, ethylparaben, (Z)-3-(2,3,4-trimethoxyphenyl) acrylic acid, 5,6,7-trimethoxycoumarin, stigmast-9(11)-en-3-ol, daucosterol and beta-sitosterol were isolated and characterized from *Z. nitidum* (Hu et al., 2006). 6-(2',3'-dihydroxy-3'-methyl-butyl)-7-hydroxy-5-methoxy-2H-1-benzopyran-2-one,6-(2',3'-dihydroxy-3'-methyl-butyl)-7-methoxy-8-(3'-methyl-but-2'-enyl)-2H-1-benzopyran-2-one, 6-(2',3'-dihydroxy-3'-methyl-butyl)-7-hydroxy-2H-1-benzopyran-2-one, 6-(2',3'-oxiranyl-3'-methyl-butyl)-7-methoxy-8-(3-methyl-but-2-enyl)-2H-1-benzopyran-2-one, 7-methoxy-8-(3'-methyl-but-2'-enyl)-2H-1-benzopyran-2-one were also isolated from *Zanthoxylum* (Tao et al., 2001, 2003, 2005).

Coumarins and alkaloids such as bergarpten, umbelliferone, skimmianine and schinifoline were isolated from *Zanthoxylum* (Liu et al., 1991, Liu et al., 2008). A Novel amide N-(4'-methoxyphenyl ethyl)-3,4-methylenedioxy cinnamoyl amide was isolated and characterized from *Z. armatum* (Kalia et al., 1999) and norchelerythrine, magnoflorine and (-)(S)-O-methylbalfourodinium cation from *Z. scandens* bark (Nguyen et al., 2002). Aliphatic acid amides have been isolated from the pericarp of *Z. piperitum* fruits (Hatano et

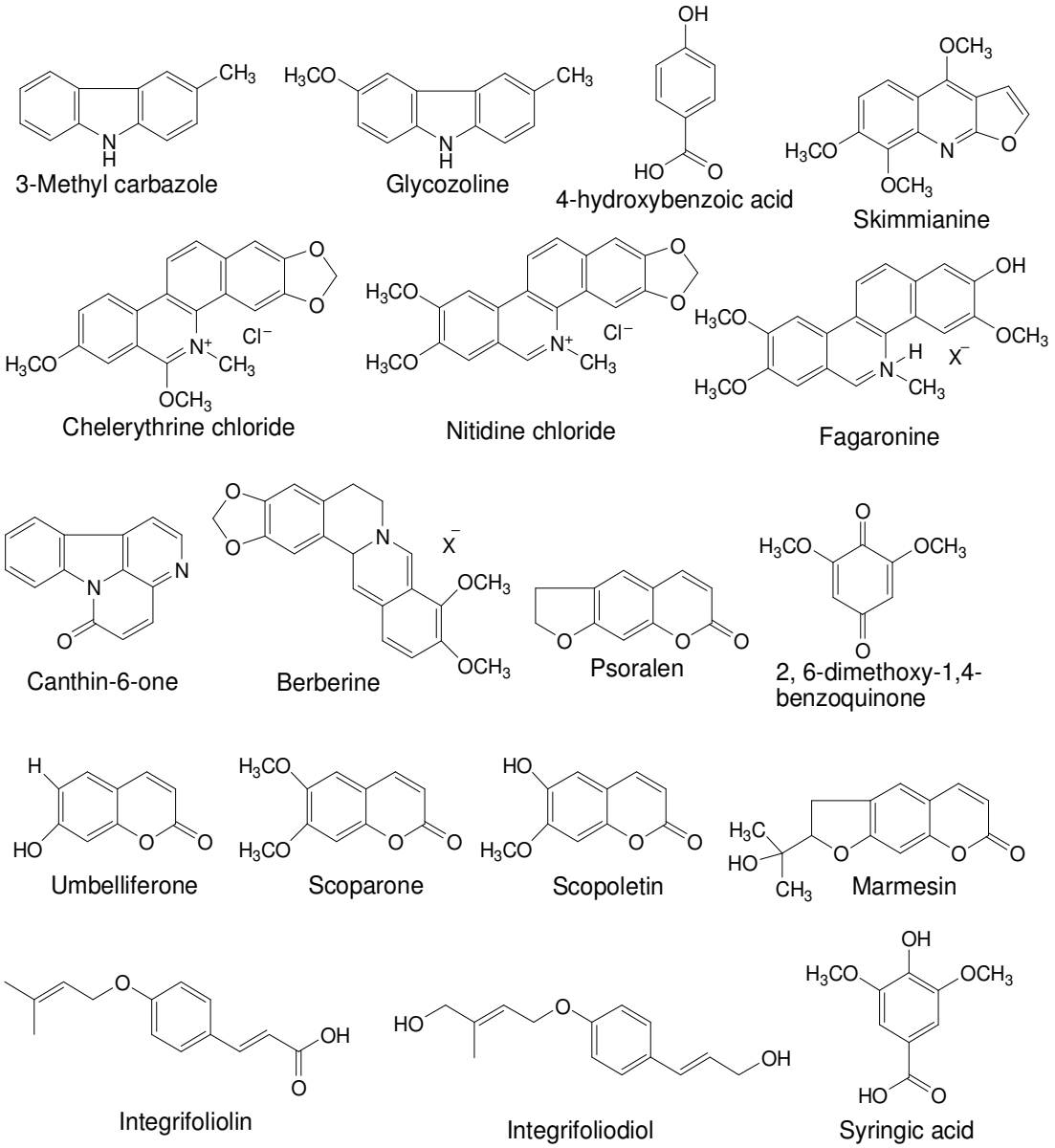

Figure 1. Structure of some isolated bioactives of *Zanthoxylum*.

al., 2004). Ahmad et al. (2003) isolated pseudophrynamine, lunacridine and 2-(2',4',6'-trimethyl-heptenyl)-4-quinozolone from the leaves of *Z. budrunga*. Quinoline alkaloids, (-)-R-geilbalansine and hyemaline were isolated from the stem barks of *Z. hyemale*. The antispasmodic activity of the crude ethanolic extract of *Z. hyemale* was studied in two different antispasmodic test models (De Moura et al., 2002). Canthin (fungicide) and pellitorine (insecticide) have been isolated from dichloromethane extracts of the roots and the bark of *Z. usambarense* (He et al., 2002) and 3-methoxy-3',4'-methylenedioxylignan-4,8,9,9'-tetraol from *Z. monophyllum*. Lupeol, ß-sitosterol, (+)-sesamin, *trans*-dimethylmatairesinol, hesperidin, (-)-cis-N-methylcanadine and sucrose were isolated from *Z. spruce* (Binutu and Cordell, 2000). L-sesamin and L-asarinin were determined by HPLC from different parts of *Zanthoxylum* (Roxb.) DC (Zhang et al., 2002). All the information regarding chemical and pharmaceutical aspect of Nigerian *Zanthoxylum* have been documented by Adesina (2005). The structures of some isolated bioactives of *Zanthoxylum* are depicted in Figure 1.

ESSENTIAL OIL CONTENT

The main components of *Zanthoxylum* oil are 38.91 oleic acid, 29.45 palmitic acid, 10.80 linoleic acid methyl ester

and 8.64% linolenic acid methyl ester (Li et al., 2001). Furthermore, Yang (2008) reported 15 linalyl acetate, 13 linalool and 12% limonene as major components. GC-FID and GC-MS analyses of the essential oil of *Z. schinifolium* and *Z. bungeanum* showed that limonene was the main component (21%), followed by 4-terpineol, gamma-terpinene, alpha-terpineol acetate, beta-pinene, alpha-terpineol and beta-linalool (Iseli et al., 2007). Itthipanichpong et al. (2002) had reported 31.09 limonene, 13.94 terpin-4-ol and 9.13% sabinene as major components of the essential oil of *Z. limonella* Alston. Chemical constituents of essential oil of *Z. bungeanum* were also determined by Guo et al. (2001). Bisht and Chanotiya (2011) analyzed the essential oil of *Z. armatum* DC leaf by capillary gas chromatography (GC-FID) and gas chromatography-mass spectrometry (GC-MS) and found 2-undecanone as dominate for the first time, although, linalool was reported as dominant content in several previous studies on *Z. armatum* oil (Yoshitito et al., 2000). The essential oil of *Z. gillettii* leaves were analyzed by GC-MS and olfactoric methods. Monoterpenes such as myrcene, trans-beta-ocimene, limonene, camphene, alpha- and beta-pinene are main constituents of the oil. Sesquiterpenes such as beta-caryophyllene, alpha- and beta-farnesene, beta-bisabolol, gamma-cadinene and nerolidol and alphatic components such as 2-undecanone and dodecanal were also identified from the essential oil of this species (Jirovetz et al., 1999). Bhattacharya and Zaman (2009) identified essential oil composition from the fruits and leaves of *Z. nitidum* by GC-MS and found monoterpenes in the fruit oil as linalool (23.3%), limonene (12.9%), aterpineot (8.3%), α-pinene (7.9%), γ-terpinene (6.6%), cis-β-ocimene (6.2%), terpinen-4-ol (4.7%) and isomenthone (2.0%), while, among the monoterpenes of leaf oil, the predominant was limonene (33.1%), followed by geraniol (10.6%), carvone (9.6%), 7-hydroxy-3,7-dimethyloctanal (4.3%), geranial (3.9%) and nerol (2.2%).

The essential oils from aerial parts of juvenile leaves, mature leaves, fruit and flowers of *Z. hyemale* were isolated by hydrodistillation and analyzed by GC, GC-MS and chiral phase gas chromatography (CPGC). The major constituent of the juvenile leaf essential oil was the sesquiterpene trans-nerolidol (51%), while the main constituent of mature leaf and flower oils was 31 and 22% hyemalol, respectively. In the fruit, the most abundant components were the monoterpenes β-pinene (25%) and α-pinene (10%). The essential oils of the leaves and the fruits of *Z. leprieurii*, *Z. macrophylla* and *Z. xanthoxyloides* of Cameroon were extracted by hydrodistillation. Extraction yields of 0.42 and 7.22%, 0.14 and 0.11%, 0.12 and 3.88% were obtained for leaves and the fruits of *Z. leprieurii*, *Z. macrophylla* and *Z. xanthoxyloides*, respectively. Essential oil from green fruit, ripe fruit and dried pericarp of Z. *piperitum* were also determined by using GC-MS (Jiang and Kubota, 2004). Zhang and Jiang (2008) developed a technique to produce biodiesel from *Z. bungeanum* seed oil.

BIOLOGICAL ACTIVITIES

The fruits are used as digestive appetizer, to cure asthma and bronchitis, eliminate pain, use to treat heart diseases, piles, diseases of mouth, teeth and throat disorder, also prescribed in dyspepsia and diarrhea (Kritikar and Basu, 1983). The roots bark and leaves of many species are used in various medicinal preparations for curing stomachache, tooth-ache, coughs, urinary and veneral diseases, leprous ulcerations, rheumatism and lumbago (Oliver-Bever, 1982). *Zanthoxylum* has been studied for several types of biological activities such as larvicidal, anti-inflammatory, analgesic, antinociceptive, antioxidant, antibiotic, hepatoprotective, antiplasmodial, cytotoxic, antiproliferative, anthelminthic, antiviral, anticonvulsant and antifungal (Tiwary et al., 2007; Lima et al., 2007; Chen et al., 2007, 2008; Guo et al., 2011; Rodrigures et al., 2010; Lee and Lim, 2008; Ross et al., 2004; Chou et al., 2011; Barnabas et al., 2010; Song et al., 2010; Yang and Chen, 2008; Islam et al., 2001a, b; Gansane et al., 2010; Amabeoku and Kinyua, 2010; Seal, 2011; Peneluc et al., 2009).

Essential oil of *Z. armatum* showed strong inhibition of mycelial growth against the test fungus *Bipolaris sorokiniana* (Manandhar and Tiwari, 2005). The anti-tumor activity of the volatile oil of *Z. rhoifolium* Lam. leaves were investigated *in vitro* and *in vivo* using the *Ehrlich ascites* tumor model (Da Silva et al., 2007a; Da Silva et al., 2007b). Methyl-N-methylanthranilate and alpha-bisabolol represented the main compounds of essential oil of *Z. tingoassuiba* which showed antimicrobial activity (Detoni et al., 2009). Toothpaste containing *Z. nitidum* extract decreased the incidence of dental plaque and enhanced gingival health (Wan et al., 2005). An alkaloidal extract of the stem barks of *Z. chiloperone* exhibited antifungal activity against *Candida albicans*, *Aspergillus fumigatus* and *Trichophyton mentagrophytes* (Thouvenel et al., 2003). Bafi-Yeboa et al. (2005) investigated *Z. americanum* leaf, fruit, stem, bark and root for antifungal activity with 11 strains of fungi. All extracts exhibited a broad spectrum of antifungal activity. Alkamides isolated from the leaves of *Z. syncarpum* showed moderate antiplasmodial activity, with IC50 values of 4.2 and 6.1 mM against *Plasmodium falciparum* D6 and W2 clone (Ross et al., 2005). Ethanolic extracts of the trunk bark of *Z. fagara*, *Z. elephantiasis* and *Z. martinicense* showed antifungal activity (Dieguez-Hurtado et al., 2003). The petroleum ether, chloroform and methanol extracts of the leaves and barks of *Z. budrunga* have been evaluated for their antibacterial, antifungal and cytotoxic properties (Islam et al., 2001a). Benzophenanthrene alkaloids, 8-acetonyldihydronitidine and 8-acetonyldihydroavicine were isolated from *Z. tetraspermum* stem bark which

showed significant antibacterial activity (Nissanka et al., 2001). *Z. limonella* showed mosquito repellent activity (Trongtokit et al., 2005). Canthin isolated from *Z. chiloperone* possessed a broad spectrum of antifungal and leishmanicidal activities (Ferreira et al., 2006). The hepatoprotective activity of the ethanolic extract of *Z. armatum* leaves have been evaluated in CCl_4-induced hepatotoxicity in rats (Verma and Khosa, 2010). The antimicrobial activity of the essential oils of *Z. hyemale* was also reported (Simionatto et al., 2005). The antioxidant properties of the fruit samples of *Z. leprieurii* and *Z. xanthoxyloides* demonstrated an SC_{50} of 3.72 and 4.05 g/l, respectively, while the butyl hydroxy toluene reference gave an SC_{50} of 7.10 mg/l. The fruit essential oils of *Z. leprieurii* and *Z. xanthoxyloides* could be used as food supplements to protect against emergent diseases such as cardiovascular problems, cancer and diabetes (Dongmo et al., 2008).

Cytotoxic activity of essential oil of *Z. rhoifolium* was evaluated against HeLa (human cervical carcinoma), A-549 (human lung carcinoma), HT-29 (human colon adenocarcinoma), Vero (monkey kidney) cell lines and mice macrophages by Da Silva et al. (2007a). They observed that the essential oil is cytotoxic against tumoral cells (CD50 = 82.3, 90.7 and 113.6 µg/ml for A-549, HeLa, HT-29 cell lines, respectively), while noncytotoxic against non-tumoral cells (Vero and mice macrophages). Essential oil and methanolic extract of *Z. budrunga* showed significant hypnotic activity (Bhadoriya et al., 2009). Kamsuk et al. (2007) investigated protective effects of *Z. piperitum* for repellency against mosquitoes under laboratory and field conditions using synthetic repellent standard, N,N-diethyl-3-methylbenzamide.

CONCLUSION

The genus *Zanthoxylum* posses several types of biological activities such as larvicidal, anti-inflammatory, analgesic, antinociceptive, antioxidant, antibiotic, hepato-protective, antiplasmodial, cytotoxic, antiproliferative, anthelminthic, antiviral and antifungal, mainly due to the presence of alkaloids and essential oil.

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