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## AN UPDATE REVIEW ON *ZANTHOXYLUM ARMATUM DC*

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### ABSTRACT

*Zanthoxylum armatum* is used mainly in the Indian system of medicine as carminative, stomachic, toothache, fever, dyspepsia and expelling roundworms. Essential oils of *Z. armatum* exhibited good antibacterial, antifungal and anthelmintics activities. A wide variety of chemical compounds including alkaloids, flavonoids, sterols, triterpenoids have been found in this plant. It contains volatile oil with active constituents such as linalool, limonene and ligan. Various studies indicated that it possesses antilarvicidal, antifungal, hepatoprotective, and allopathic properties. This Review has basic information about *Z. anthoxylum armatum* and its biological activities for further development in this field.

**Keywords:** *Zanthoxylum armatum*, *Zanthoxylum alatum*, Toothache Tree, Tejppal, Biological Activities

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## INTRODUCTION

*Zanthoxylum armatum* belongs to family Rutaceae is a sub deciduous aromatic shrub or small tree up to 5 m high. Bark pale brown rough croky; leave alternate imparipinnate, flowers small pale yellow and bisexual. The family Rutaceae comprises about 30 species in temperate and tropical parts of the world, of which 10 species were reported in India<sup>1</sup>

*Zanthoxylum armatum* DC. (Syn. *Zanthoxylum akztrnn* Roxb.) (Rutaceae) is an erect shrub or a small tree commonly occurring in hot valleys of the Himalayas. While its bark has been reported to possess active alkaloids, the fruits and seeds are also extensively used in the indigenous system of medicine. The fruits have been reported to yield an essential oil with linalool as the major constituent. The *seeds* have, earlier, been found to contain cis-9-hexadecenoic, eicosenoic, and palmitic acids along with tambuletin<sup>2</sup> Distribution Area found in the hot valleys of the Himalayas from Jammu to Bhutan at altitudes of 1,000-2,100 m<sup>3</sup>.



**Figure 1: *Zanthoxylum armatum***

### Nomenclature

Domain: *Eukaryota*

Kingdom: *Plantae*

Subkingdom: *Viridiaeplantae*

Phylum: *Tracheophyta*

Subphylum: *Euphylllophytina*

Infraphylum: *Radiatopses*

Class: *Magnoliopsida*

Subclass: *Rosidae*

Superorder: *Rutanae*

Order: *Sapindales*

Suborder: *Rutineae*

Family: *Rutaceae*

Subfamily: *Rutoideae*

Tribe: *Zanthoxyleae*

Genus: *Zanthoxylum*

Specific epithet: *armatum* - DC.

**Botanical name:** - *Zanthoxylum armatum* DC<sup>4</sup>

### **Vernacular Information**

Bengali - Gaira

Hindi - tejphal, tumru, darmar, trimal

Kannada - dhiva, tumburudu, jimmi

Malayalam - tumpunal, tumpuni

Marathi - chirphal, naepaali dhane

Mizoram - arhrikreh

Oriya - arhrikreh, ranabelli

Sanskrit - saurabha, tejovati, tumberu, vanaja

Tamil - tumpunalu

Telugu - gandhalu, konda-kasimi, kondakaasimanda

### **Other Names**

Burmese - Gawra Kha Nan Nan, Teza Bo

Chinese - Ci Zhu Ye Hua Jiao, Qin Jiao (Taiwan), Zhu Ye Jiao

English - Bamboo-Leaved Prickly Ash, Nepal Pepper,

Prickly Ash, Toothache Tree, Winged Prickly Ash,

Winged Prickly-Ash, Wingleaf Prickly Ash

German - Nepalpfeffer

Japanese - Fuyu Zanshou, Fuyu-Sansh

Korean - Gae San Cho

Nepalese - Timbur, Timur

Thai - Mak Kak

Laotian – Mad

**Distribution**

China, Japan, Korea, Taiwan, Bangladesh, Bhutan;  
India - Andhra Pradesh, Himachal Pradesh, Jammu and Kashmir,  
Manipur, Meghalaya, Nagaland, Orissa, Uttar Pradesh;  
Nepal, Pakistan, Laos, Myanmar, Thailand, Vietnam,  
Indonesia, Philippines<sup>5</sup>

**Description**

This shrub is found in North America, Nepal, India, and Bhutan<sup>6</sup>. Shrubs, woody climbers, or trees to 5 m tall, deciduous. Branchlets and leaflet blades abaxially on midvein usually with prickles. Young branchlets and inflorescence rachises glabrous or rust-colored pubescent. Leaves 3-9-foliolate; rachis glabrous or rust-colored pubescent, wings to 6 mm on each side; leaflet blades sub sessile, opposite, lanceolate, ovate, or elliptic, 3-12 cm long, 1-3 cm wide, base attenuate to broadly cuneate, secondary veins 7-15 on each side of midvein and generally faint, margin crenate or entire and often revolute when dry, apex acute to acuminate. Inflorescences terminal on short lateral branchlets and sometimes axillary, 1-7 cm, with less than 30 flowers. Perianth in 2 irregular series or 1 series, with  $6-8 \pm$  undifferentiated 0.3-1.5 mm tepals. Male flowers: stamens 4-6; anthers yellow prior to anthesis; connective apex with oil gland; disk pulvinate; rudimentary carpels lacking. Female flowers: carpels 2 or 3, abaxially often with a conspicuous oil gland; styles recurved; staminodes ligulate or lacking. Fruit follicles usually purplish red, 4-5 mm in diameter, with a few protruding oil glands. Seeds blackish brown, 3-4 mm in diameter. Minute yellow flowers arise in leaf axils. Flowers have 6-8 acute sepals. Petals are absent. Male flowers have 6-8 stamens, and large anthers because of which the flowers look yellow. Female flowers have 1-3 celled ovary, 3 mm in diameter, pale red, splitting into two when ripe. Seed are rounded, 3 mm in diameter, shining black. Flowering: March-April<sup>4</sup>.

**Parts Used**

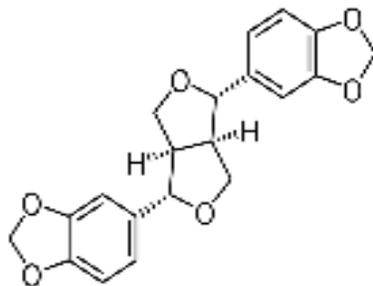
Bark, carpels, carpels of fruits, seeds

**Chemical constituents**

Bark: Alkaloids: g-fagarine, b-fagarine (-skimmianine), magnoflorine, laurifoline, nitidine, chelerythrine, tambetarine and candicine Coumarins: xanthyletin, zanthoxyletin, alloxanthyletin, Resin, tannin, volatile oil<sup>6</sup>.

Bark yields a bitter crystalline principle, identical to berberine, and a volatile oil and resin. The carpels yield a volatile oil, resin, a yellow acid principle, and crystalline solid body, xanthoxylin.

Carpels of the fruit yield an essential oil which is isomeric with turpentine had like eucalyptus oil in odor and properties. The bark contains berberine. The essential oil from the seeds consists entirely - over 85% - of the hydrocarbone 1- $\alpha$ -phellandrene and also a small quantity of linalool and an unidentified sesquiterpene. Bark yields active compounds: alkaloids (g-fagarine, b-fagarine, magnoflorine, laurifoline, nitidine, chelerythrine, tambetarine and cadicine), coumarins (xanthyletin, zanthoxyletin, alloxanthyletin), and resin, tannin and volatile oil <sup>7</sup>.



**Figure 2: Sesamin**

A volatile oil consisting mainly linalool<sup>8</sup> mono terpenetriol-3, 7-dimethyl 1-octane 3,6,7-triol, trans cinnemic acid, nevadensin umbelliferone,  $\beta$ -sitosterol and its glucoside, 3,5, dihydroxy-7, 8, 4'-trimethoxyflavone (tamblin) and tambulatin, 3-methoxy -11 -hydroxy -6, 8-dimethylcarboxylate biphenyl, 3, 5, 6, 7- tetrahydroxy-3', 4'-dimethoxyflavone -5 - $\beta$  -d -xylopyranoside, aramatamide, lignans, asarinin and fragesin,  $\alpha$  and  $\beta$ -amyrins lupeol, and  $\beta$  -sitosterol  $\beta$ -D-glycoside have been reported from the plant previously. Antihelmentic, antiprolifative, antifungal and anti-insecticidal activities have also been studied with different parts of the plant <sup>1</sup>.

### Phenolic Constituents

Study isolated two new phenolic constituents from the seeds - 3-methoxy-11-hydroxy-6,8 dimethylcarboxylate biphenyl and 3, 5, 6, 7- tetrahydroxy-3', 4'- dimethoxyflavone- 5-  $\beta$ - d-xylopyranoside along with five known compounds<sup>7</sup>.

Chemical studies on the seeds of *Zanthoxylum alatum* Roxb. (Rutaceae) led to the isolation of two new phenolic constituents characterized as 3-methoxy-11-hydroxy-6,8-dimethylcarboxylate biphenyl and 3,5,6,7-tetrahydroxy-3',4'-dimethoxyflavone-5- $\beta$ -d-xylopyranoside along with the five known compounds, 1-methoxy-1,6,3-anthraquinone, 1-hydroxy-6,13-anthraquinone, 2-hydroxybenzoic acid, 2-hydroxy-4-methoxy benzoic acid, and stigmasta-5-en-3 $\beta$ -d-glucopyranoside, on the basis of spectral data and chemical analyses<sup>9</sup>.

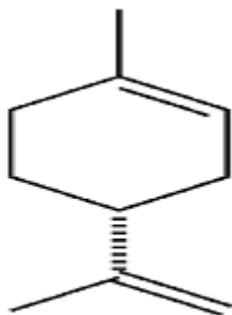


Figure 3: Limonene



Figure 4: Linalool

The essential oil of *Zanthoxylum armatum* was extracted through hydro distillation and analyzed by GC-MS

Hydrocarbon fraction (17.35%) of the oil was much lower and oxygenated compounds comprised fairly high portion of essential oil (39.21%). Percentages of monoterpenes and sesquiterpenes found were 47.33% and 10.83% respectively. Oxygenated monoterpenes comprised major profile of chromatogram of essential oil of *Zanthoxylum armatum* *i.e.* 37.23% where as monoterpenes hydrocarbons were 10.09%. Alcoholic percentage was much higher *i.e.* 26.76% and 15-hexadecanoloide (6.58%) the only cyclic ester was found in relatively high percentage<sup>10</sup>.

#### A New Amide from *Zanthoxylum Armatum*

A new amide designated as armatamide along with two lignans, asarinin and fargesin, alpha- and beta-amyrins, lupeol, and beta-sitosterol-beta-D-glucoside-has been isolated from the bark of *Zanthoxylum armatum*. The structure of the new compound was deduced by spectral and chemical analysis as N-(4'-methoxyphenyl ethyl)-3, 4-methylenedioxy cinnamoyl amide<sup>11</sup>.

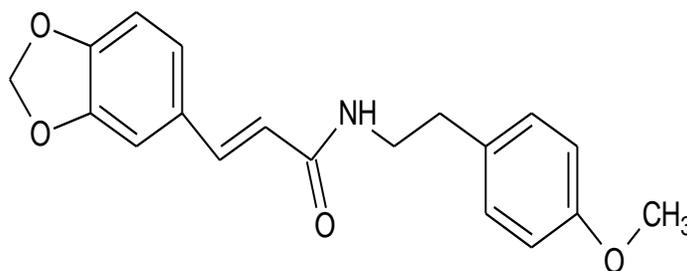


Figure 5: Armatamide

#### Therapeutic Uses

Asthma, Bronchitis, Cholera, Fever, Fibrosis's, Indigestion, Rheumatism, Skin diseases, Toothache, Varicose veins. Prickly Ash is used in many chronic problems such a rheumatism and skin diseases; chilblains, cramp in the leg, varicose veins and varicose ulcers. It is also used

for low blood pressure, fever, and inflammation. Externally it may be used as a stimulation liniment for rheumatism and fibrositis. It has a stimulating effect upon the lymphatic system, circulation and mucous membranes<sup>6</sup>.

The fruit and seeds are employed as an aromatic tonic in fever dyspepsia and cholera and the bark used for intoxicating fishes. The bark, fruits and seeds are extensively used in indigenous system of medicine as a carminative, stomachic and anthelmintic. The stem has exhibited hypoglycaemic activity in the preliminary trials. The bark is pungent and used to clean teeth. The fruits and seeds are employed as an aromatic tonic in fever and dyspepsia. An extract of the fruits is reported to be effective in expelling roundworms. Because of their deodorant, disinfectant and antiseptic properties, the fruits are used in dental troubles, and their lotion for scabies. The essential oil is said to possess antiseptic, disinfectant and deodorant properties<sup>3</sup>.

### **Dosage**

Bark-infusion, decoction

Infusion: Pour a cup of boiling water onto 1-2 teaspoonfuls of the bark and let infuse for 10-15 minutes. Drink this three times a day.

Tincture: take 1-2 ml of the tincture three times a day<sup>6</sup>.

### **Ayurvedic Preparation**

Tejowatyadya Grita,

Tumbawardi Churna<sup>6</sup>

## **BIOLOGICAL ACTIVITIES**

*Zanthoxylum armatum* shows different biological activities according to literature some of them as summarized below.

### ***Zanthoxylum armatum* in gut, airways and cardiovascular disorders**

This study describes the gut, airways and cardiovascular modulatory activities of *Zanthoxylum armatum* DC. (Rutaceae) to rationalize some of its medicinal uses. The crude extract of *Zanthoxylum armatum* (Za.Cr) caused concentration-dependent relaxation of spontaneous and high K<sup>+</sup> (80 mM)-induced contractions in isolated rabbit jejunum, being more effective against K<sup>+</sup> and suggestive of Ca<sup>++</sup> antagonist effect, which was confirmed when pretreatment of the tissues with Za.Cr shifted Ca<sup>++</sup> concentration-response curves to the right, like that caused by verapamil. Za.Cr inhibited the castor-oil-induced diarrhea in mice at 300–1000 mg/kg. In rabbit tracheal preparations, Za.Cr relaxed the carbachol (1µM) and high K<sup>+</sup>-induced contractions, in a pattern similar to that of verapamil. In isolated rabbit aortic rings, Za.Cr exhibited vasodilator

effect against phenylephrine (1 $\mu$ M) and K<sup>+</sup>-induced contractions. When tested in guinea pig atria, Za.Cr caused inhibition of both atrial force and rate of spontaneous contractions, like that caused by verapamil. These results indicate that *Zanthoxylum armatum* exhibits spasmolytic effects, mediated possibly through Ca<sup>++</sup> antagonist mechanism, which provides pharmacological base for its medicinal use in the gastrointestinal, respiratory and cardiovascular disorders<sup>12</sup>

### **Insecticidal activity of essential oil from *Zanthoxylum armatum* fructification against two mosquito species**

Essential oil, extracted by steam distillation from *Zanthoxylum armatum* fructification, were tested for their insecticidal activity to the larvae and pupae of *Aedes albopictus* and *Culex pipiens quinquefasciatus* under laboratory conditions by means of steeping, and its fumigant activity to the adults of two mosquito species were evaluated by means of sealing conical flask. The volatile components of this oil were also analyzed by gas chromatography-mass spectrometry. Results are given as follows:(1)The 24 h LC50 values of *Z.armatum* essential oil for I,II,III,IV Wistar larvae and pupae of *Ae.albopictus/Cx.pipiens quinquefasciatus* were 25.634/61.472,31.763/76.431,52.356/110.172,258.497/121.884 and 198.263/162.048 mg·L<sup>-1</sup>,respectively;(2)The LC50 values for adults of *Ae.albopictus* and *Cx.pipiens quinquefasciatus* were 24.957 and 29.517  $\mu$ g·cm<sup>-3</sup>,respectively;(3)At the dosage of 147.52  $\mu$ g·cm<sup>-3</sup>,the death rate of adults of *Ae.albopictus* and *Cx.pipiens quinquefasciatus* both were 100%,and the KT50 values for adults of two mosquito species were 3.493 min and 2.993 min, respectively;(4)Eighteen chemical constituents were identified, including 10 kinds of terpenoids compounds which were the main components and accounted for 67.122% of total. This essential oil had a high and rapid poison activity on *Ae.albopictus* and *Cx .pipiens quinquefasciatus*, which had potential to develop natural insecticides against mosquitoes<sup>13</sup>.

### **Antifungal and Insect-Repellent Activity of Essential Oil of *Zanthoxylum alatum***

The essential oil of fruits of *Zanthoxylum alatum* (Rutaceae) proved repellent to the insect *Allacophora foveicollis* and fungistatic to 24 fungi, including aflatoxin-producing strains of *Aspergillus flavus* and *A. parasiticus* at a minimum dose of 2.0  $\times$  10<sup>3</sup>  $\mu$ l<sup>-1</sup>. The fungistatic property of the oil was not affected by high temperature, prolonged storage and increased inoculums<sup>14</sup>.

**Antifungal / Insect Repellent:** Essential oil of the fruits of ZA showed repellent activity against insect *Allacophora foveicollis* and fungistatic activity against 24 fungi, including aflatoxin-producing strains of *A flavus* and *A parasiticus*<sup>7</sup>.

### **Larvicidal activities**

*Zanthoxylum armatum* DC (Rutaceae) against three mosquito vectors larvicidal potential of the essential oil from the seeds of *Zanthoxylum armatum* DC [syn. *Z. alatum* Roxb] (Rutaceae) against three medically important species of mosquito vectors, *Aedes aegypti*, *Anopheles stephensi* and *Culex quinquefasciatus*. Study of essential oil yielded at least 28 compounds, consisting mainly of oxygenated monoterpenes and monoterpenes. The larvae of three mosquito species - *A. aegypti*, *A. stephensi*, and *C. quinquefasciatus* - were susceptible to the essential oil composition and presents a potential for the development of alternative plant based larvicides<sup>15</sup>.

### **Anti-Inflammatory and Antioxidant Activities of *Zanthoxylum Armatum* Stem Bark**

Anti inflammatory and antioxidant activities of ethanolic extract of steam bark of *Zanthoxylum armatum*. In vivo anti inflammatory activity was evaluated in Wistar species of rats by using Carrageenan induced paw edema, where as in vitro antioxidant activity was performed by DPPH free radical method. The plant extract exhibited significant anti-inflammatory and antioxidant activities<sup>16</sup>.

### **Hepatoprotective**

Study of the ethanolic extract of leaves of *Z armatum* on CCl<sub>4</sub>-induced hepatotoxicity in rats showed significant decrease in liver enzymes and liver inflammation, supported by histopatho studies on the liver. Results exhibited significant hepatoprotective activity<sup>17</sup>.

### **Insecticidal**

Study of the essential oil of *Zanthoxylum armatum* showed high and rapid poison activity on *Aedes albopictus* and *Culex quinquefasciatus*, showing a potential as natural insecticides against mosquitoes<sup>7</sup>.

### **Spasmolytic**

Crude extract of ZA caused concentration-dependent relaxation of spontaneous and high K<sup>+</sup> induced contractions in isolated rabbit jejunum. Results showed *Zanthoxylum armatum* exhibits spasmolytic effects, mediated possibly through Ca<sup>++</sup> antagonistic mechanism, which provides pharmacologic base for its medicinal use in the gastrointestinal, respiratory, and cardiovascular disorders<sup>7</sup>.

### **Inhibition of Keratinocyte Growth by Different Nepalese *Zanthoxylum* Species**

A total of 11 methanol extracts obtained from four different Nepalese *Zanthoxylum* species were screened for their antiproliferative activity against the growth of human keratinocytes (HaCaT cells). The extract obtained from *Z. armatum* barks was highly active with an IC<sub>50</sub> value of 11

micrograms/mL. Also, the extracts obtained from *Z. oxyphyllum* barks and roots with IC50 values of 53 and 57 micrograms/mL, respectively, showed potent activity. Their antiproliferative activity was not due to cytotoxic effects on cell membranes, as documented by the activity of lactate dehydrogenase released from the cytoplasm of keratinocytes, which did not exceed that of the control value. Rather, they also protected against radical-induced damage to model membranes stimulated with 2, 2'-azo-bis (2-amidinopropane) dihydrochloride<sup>18</sup>.

### **Allelopathic Influence of *Zanthoxylum armatum* D.C. on Important Field Crops Seeking its Sustainable Domestication in Existing Agroforestry Systems of Garhwal Himalaya, India.**

Allelopathic influence of *Z. armatum* was tested on some important winter field crops (*T. aestivum*, *H. vulgare*, *B. compastris*, and *L. culminaris*) of Garhwal Himalaya region, using different concentrations of leaf, bark, and fruit pulp aqueous extracts. Significant effects of these bioassays were noticed on germination and growth of all the test crops. Among all the tested crops, *B. compastris* followed by *T. aestivum* were found to be most sensitive, while *H. vulgare* and *L. culminaris* were the least sensitive in terms of germination. On the other hand, increased concentrations (5 and 10%) of leaf, bark, and fruit pulp aqueous extracts considerably reduced the radicle and plumule growth of all the test crops as compared with control. It is suggested that *H. vulgare* and *L. culminaris* may be cultivated in proximity of *Z. armatum*<sup>19</sup>.

### **Enhanced Repellency of Binary Mixtures of *Zanthoxylum armatum* Seed Oil, Vanillin, and Their Aerosols to Mosquitoes under Laboratory and Field Conditions**

The repellency of *Zanthoxylum armatum* seed oil (ZA-SO), alone or in combination with vanillin (VA), its six major constituents, and another four major previously known *Zanthoxylum piperitum* fruit oil constituents, as well as aerosol products containing 5 or 10% ZA-SO and 5% VA, was evaluated against female *Aedes aegypti* in laboratory and field studies. Results were then compared with those of *N, N*-diethyl-3-methylbenzamide (DEET) as a standard. Hand in cage laboratory tests showed that 0.2, 0.1, and 0.05 mg/cm<sup>2</sup> ZA-SO resulted in >92% protection through 30-min post exposure and was not significantly different than 0.05 mg/cm<sup>2</sup> DEET. Skin treated with linalool and limonene (from *Z. armatum*) provided >80% repellency to female *Ae. aegypti* at 10-min exposure, whereas cuminaldehyde, citronellal, geranyl acetate, and cuminyl alcohol (from *Zanthoxylum piperitum*) provided >90% protection during this same time period. Only cuminaldehyde and citronellal provided complete protection comparable to DEET at 10-min post exposure. After that time, repellency of all plant constituents to mosquitoes was

considerably decreased (65%). An increase in repellency and duration of effectiveness was produced by a binary 1:4 mixture of ZA-SO and VA (0.05:0.2 mg/cm<sup>2</sup>) that was significantly more effective than 0.05 mg/cm<sup>2</sup> DEET through 90 min. In field tests, an aerosol formulation containing 5 or 10% ZA-SO plus 5% VA gave 100% repellency at 60-min post exposure. Although these formulations were equal to the level of protection afforded by 10% DEET, repellency to the binary ZA-SO aerosol formulations at 90 min was significantly less effective than DEET. However, mixtures formulated from ZA-SO and VA merit further study as potential repellents for protection of humans and domestic animals from biting and nuisance caused by mosquitoes<sup>20</sup>.

### **Concentration Dependent Antioxidant Activity of *Zanthoxylum Armatum***

The essential oil of the fruits of *Zanthoxylum armatum* DC. was evaluated for its antioxidant activity. GCMS of the essential oil was also taken into consideration for major constituents. The oil showed concentration dependent DPPH scavenging activity. The major chemical constituents found in the oil were 1, 8 – cineole, linalool,  $\alpha$  – terpeniol and  $\beta$ - cubebene<sup>21</sup>.

### **Insect Pests on *Zanthoxylum Armatum* DC.**

Insect pests infesting *Zanthoxylum armatum* in District Rajouri of Jammu Division of J&K (India). *Zanthoxylum armatum* is used as a carminative, stomachic, circulatory stimulant and exhibits antispasmodic, antihelminthic, anti-inflammatory and antibacterial properties. Keeping in view the great medicinal value of the plant, a survey was conducted which revealed the occurrence of 7 insect pests belonging to 4 families in 3 insect orders<sup>22</sup>.

## **CONCLUSION**

It is concluded that medicinal plants have contributed hugely to the traditional and western medicines through providing ingredients for drugs or having played central roles in the drug discovery. The above review provides the update information regarding the *Zanthoxylum armatum*. Essential oils of *Z. armatum* exhibited good antibacterial, antifungal and anthelmintics activities. All are the essential oil of the seeds of *Zanthoxylum armatum* has *Antimicrobial potential* and drugs used for the treatment of microbial diseases. . Essential oils of *Z.armatum* exhibited good antibacterial, antifungal and anthelmintics activities.

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