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Chemical composition, biological activities and medicinal properties of genus *Zanthoxylum*: An updated review

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Abstract
Genus *Zanthoxylum* belongs to family Rutaceae. Rutaceae is one of the major Angiosperm families with 153 genera and 1975 species distributed worldwide. *Zanthoxylum armatum* DC. is common species found in Uttarakhand. It is widely spread in the hot valleys of Himalayas from Uttarakhand, Jammu to Bhutan, Nepal and Pakistan. *Zanthoxylum armatum* used as a medicine from ancient time for cure of various diseases such as toothache and problems related to tooth, asthma, used for gum bleeding, fever, dyspepsia, and tonics etc. Chemical composition, phytochemistry, pharmacological activities, traditional uses are explained in this review article.

Keywords *Zanthoxylum armatum*, ethno-medical, pharmacology, antiseptic, carminative, terpenoids, botanical, alkaloids

1. Introduction
Genus *Zanthoxylum* belongs to family Rutaceae. Rutaceae is one of the major Angiosperm families with 153 genera and 1975 species distributed worldwide. *Zanthoxylum armatum* is one of the most important medicinal plants in Indian medicinal literature. In India about ten species are grown. Almost all parts of this plant are used in Indian traditional system for the treatment of various ailments and the significant medicinal properties was further reported through scientific investigation. Plants containing active ingredients used to cure disease or relieve pain are called medicinal plants. Many species of genus *Zanthoxylum* like, *Z. acanthopodium* DC., *Z. americanum* Mill., *Z. armatum* DC., *Z. bungeanum* Maxim., *Z. beecheyanum* K. Koch, *Z. capense* (Thunb.) Harv., *Z. caribaeanum* Lam., *Z. clava-herculis* L., *Z. flavum* Vahl, *Z. gillettii* (de Wild) Waterm., *Z. piperitum* (L.) DC., *Z. rhetsa* (Roxb.) DC., *Z. simulans* Hance, *Z. xanthoxyloides* (Lam.) Zepernick & Tilmer etc. The members genus are trees, shrubs, lianas, or rarely herbs, often with spines or prickle and with secretary glands containing ethereal oils in many tissues and appearing as pellucid-punctate glands in the leaves and pericarp. The genus *Zanthoxylum* L. with 225 tropical species is a rich source of various phytochemicals such as amides, alkaloids, flavanoides, lignans, sterols and terpenes, etc. Many species of genus are of economic importance as source of edible fruits, essential oils, wood, ornamentals; raw materials are of economic importance as source of edible fruits, essential oils, wood, ornamentals and raw materials for industries, medicinal plants and culinary applications. Almost all the species of genus *Zanthoxylum* have great ability to produce tires which could be used as encapsulates in the pharmaceutical industry, diluents and emulsifying agents [1-5]. In Uttarakhand a well-known species and commonly found species is *Zanthoxylum armatum* DC. This is commonly called as 'Prickly ash' or 'Timur' or 'Kababe Tejal', is a shrub or small tree which predominately grows in well drained alluvial, black soil and have a strong aroma. The plants are armed scandent or erect, 6 m tall or more, with dense foliage [6]. In India, it is found in the warmer valleys of the Himalaya from Uttarakhand, Jammu and Kashmir to Assam and Khasi (1,000 to 2,100 m), in the Eastern Ghats in Orissa and Andhra Pradesh (1,200 m) and the lesser Himalayan regions in the northeastern part of India for example, Naga Hills, Meghalaya, Mizoram, and Manipur. These species which may use as medicinal plants have more effectiveness against the diseases and more curable without having any side effects. Out of these, four species are *Z. armatum* DC., *Z. acanthopodium* DC., *Z.*

oxyphyllum Edgew, and *Z. budrunga* are commonly found in India [7-10]. The main objective of this review is to focus on the essential oil components, biological activities, and other nutraceutical potentials of essential oils and major isolates of the genus *Zanthoxylum.*

2. Material and Methods

This current review focuses an update on recent studies performed on the chemical composition and biological studies on genus *Zanthoxylum*. The search engines Google Scholar, PubMed, ScienceDirect, and ResearchGate were used to access the literature. Relevant articles published in English were searched in different databases, specifically, Google Scholar, Scopus, Wiley Online Library, Semantic Scholar, and Pub Med. The following terms were used in combination within search strings: "*Zanthoxylum"*, "essential oils", and "anti-inflammatory", "antioxidant", "antibacterial", "antifungal", "antimicrobial", "antiviral". The search was restricted to English and experimental studies.

3. Traditional uses of *Zanthoxylum* spp.

Zanthoxylum armatum DC. (syn. *Zanthoxylum alatum* Roxb.) (Rutaceae) The Bhots people of Spiti Valley, Himachal Pradesh, India, use the bark use to relieve toothache [11]. The people of Baitadi and Darchula districts of far-western Nepal use the fruits used to treat colds, coughs, toothaches; the bark is used to stupefy fish [12]. Local people in the Rasuwa district of central Nepal take the pickled fruits for stomach ache and indigestion [13]. In Newer community of Kathmandu, Nepal, the fruit used for antileech, indigestion, spice and flavorant [14].

Essential oils compositions

There are number of essential oil reports on *Zanthoxylum* species from worldwide. Linalool (55.3%), limonene (22.5%), methyl cinnamate (8.8%) as a major compounds reported from fruit essential oil of *Zanthoxylum armatum* from district Pithoragarh, Uttarakhand [15]. Also fruit pericarp essential oil of *Z. armatum* from Uttar Pradesh reported, linalool (72%), methyl cinnamate (12.2%), limonene (6.2%), β-phellandrene (5.3%) [16]. Leaf essential of *Z. armatum* from Kumaon, Uttarakhand reported, 2-undecanone (55.7%), linalool (11.5%), β-caryophyllene (4.6%), 1, 8-cineole (4.3%) [17]. Leaf essential oil *Z. alatum* from Mandi, Himachal Pradesh, reported linalool (30.6%), 2-decanone (20.9%), 2-tridecanone (8.9%), β-fenchol (9.4%), β-phellandrene (6.0%) [18]. The leaf essential oils of *Z. rhoifolium* and *Z. setulosum* (Rutaceae) from Monteverde, Costa Rica have been obtained by hydrodistillation and analyzed by gas chromatography-mass spectrometry. The principal constituents of *Z. rhoifolium* leaf oil were germacrene D (14.6%), limonene (12.5%), trans-2-hexenal (11.3%), beta-elemene (9.2%), 2-undecanone (9.2%), myrcene (7.9%), bicyclogermacrene (7.5%), and germacrene A (5.2%). The leaf oil of *Z. setulosum* was composed largely of beta-phellandrene (37.5%), beta-caryophyllene (13.7%), alpha-pinene (11.9%), germacrene D (10.9%), myrcene (5.9%), and nerolidol (5.4%). [19].The seasonal variation in the chemical composition of the leaf essential oil of *Z. clava-herculis* has been analyzed by GC-MS. The leaf essential oils were made up of 25 components, largely menthane monoterpenoids, dominated by limonene (44-73%) and 1,8-cineole (16-43%), with lesser amounts of alpha-thujene, linalool, gamma-terpinene, and alpha-terpineol. The ratio of oxygenated monoterpenoids to monoterpene hydrocarbons generally increased during the season, largely reflected in the 1, 8-cineole/limonene ratio [20]. The leaf essential oils from five species of *Zanthoxylum* (Rutaceae) from Monteverde, Costa Rica, have been obtained by hydrodistillation and analyzed by gas chromatography-mass spectrometry. The species examined include *Z. fagara*, *Z. acuminatum*, *Z. melanostictum*, *Z. monophyllum*, and an undescribed species. The most abundant classes of compounds found in Zanthoxylum leaf oils are acyclic and menthane monoterpenoids as well as simple alcohols, aldehydes, and ketones. In terms of molecular diversity, menthane and acyclic monoterpenoids, cadinane and mesocyclic sesquiterpenoids, and simple alcohols, aldehydes, and ketones dominate the essential oils of *Zanthoxylum* species. Monoterpenoids make up the majority of the mass of the leaf oils of *Z. monophyllum*, *Z. acuminatum*, *Z. fagara*, and *Zanthoxylum* sp. nov. Linalool, 4-terpineol, alpha-terpineol, and trans-2-hexenol, are found in all of the *Zanthoxylum* species examined in this study [21]. The pungent principles and the essential oil from the pericarp of *Z. schinifolium* (Rutaceae) have been investigated and compared to those of *Z. bungeanum*, the primary source of the traditional Chinese drug Huajiao (Pericarpium Zanthoxyli). HPLC-MS and HPLC-NMR analyses revealed an alkylamide profile highly similar to that of *Z. bungeanum*, with hydroxy-alpha-sanshool and hydroxy-beta-sanshool being in both plants the major constituents of the alkylamide fraction. GC-FID and GC-MS analyses of the essential oil showed that limonene was, like in *Z. bungeanum*, the main component (21%), followed by 4-terpineol, gamma-terpinene, alpha-terpineol acetate, beta-pinene, alpha-terpineol and beta-linalool [22]. This study focused, for the first time, on the evaluation of the seasonal effect on the chemical composition and biological activities of essential oils hydrodistillated from leaves, trunk bark and fruits of *Z. leprieurii*, a traditional medicinal wild plant growing in Côte d'Ivoire. The essential oils were obtained by hydrodistillation from fresh organs of *Z. leprieurii* growing on the same site over several months using a Clevenger-type apparatus and analyzed by gas chromatography-mass spectrometry (GC/MS). Leaf essential oils were dominated by tridecan-2-one (9.00 ± 0.02-36.80 ± 0.06%), (*E*)-β-ocimene (1.30 ± 0.50-23.57 ± 0.47%), β-caryophyllene (7.00 ± 1.02-19.85 ± 0.48%), dendrolasin (1.79 ± 0.08-16.40 ± 0.85%) and undecan-2-one (1.20 ± 0.03-8.51 ± 0.35%). Fruit essential oils were rich in β-myrcene (16.40 ± 0.91-48.27 ± 0.26%), citronellol (1.90 ± 0.02-28.24 ± 0.10%) and geranial (5.30 ± 0.53-12.50 ± 0.47%). Tridecan-2-one (45.26 ± 0.96-78.80 ± 0.55%), β-caryophyllene (1.80 ± 0.23-13.20 ± 0.33 and tridecan-2-ol (2.23 ± 0.17-10.10 ± 0.61%) were identified as major components of trunk bark oils. Statistical analyses of essential oil compositions showed that the variability mainly comes from the organs [23]. Huajiao denotes the fruits of various species of *Zanthoxylum* in the plant family Rutaceare used for cooking. The two most commercially popular species are Z. *bungeanu*m (red huajiao) and Z. *schinifolium* (green huajiao). Fresh huajiao has a very high content of essential oil, up to 11%, which is described as having fresh, spicy, floral, cooling, and green aroma notes. A comprehensive analysis of the essential oils by GC-MS using advanced peak deconvolution and data processing software, revealed many overlapping components. In the essential oils, linalyl acetate (15%), linalool (13%), and limonene (12%) are the major components of red huajiao, whereas linalool (29%), limonene (14%), and sabinene (13%) are the major components of green huajiao [24]. The flavor and quality of *Z. armatum* DC. are mainly determined by the essential oil components in the

Chinese prickly ash peels. In this study, the correlation between climate change in different regions and the content of essential oils of *Z. armatum* was investigated using gas chromatography-mass spectrometry (GC/MS) and multivariate statistical analysis [25]. The Cellulose Synthase gene (CS) superfamily and COBRA-like (COBL) gene family are essential for synthesizing cellulose and hemicellulose, which play a crucial role in cell wall biosynthesis and the hardening of plant tissues from the *Z. bungeanum* (Zb) genome was also reported [26]. A new megastigmane sesquiterpenoid, which is referred to as schinifolenol A, was isolated from *Z. schinifolium* [27].

Table 1: Different types of bioactivities shown by different species of *Zanthoxylum*

Sr. No.	Name of activity	Details	References
1.	Antioxidant activity	Methanolic and ethanolic fruit extract of *Z. alatum* and *Z. armatum* plant shows *in-vitro* activities in Wistar rats and due to presence of free radical scavanging it may shows the antioxidants activities.	[28-29]
2.	Antimicrobial activity	The essential oil composition of *Z. armatum* show the effect against microorganisms *Bacillus subtilis* and minimum bactericidal concentration also shows the fungicidal activities against *Alternaria brassicicola*.	[30]
3.	Anti-inflammatory activities	The fruit extract of *Z. armatum* used for inhibition of carrageenan and analgesic effect.	[31-32]
4.	Antitumor activity	The fruits and leaves *Z. armatum* reported for anticancer activity.	[33]
5.	Anti-diabetic activity:	The bark extract of *Z. armatum* show good result in diabetic activity. It shows the anti-diabetic activity against the streptozotocin induced in diabetic rats.	[34]
6.	Hepatoprotective Activity	Leaves of *Z. armatum* exhibits hepatoprotective activity with its ethanolic extract against the carbon-tetrachloride which damage the liver and it helps to normalizing the increase the level of hepatic enzymes.	[35]
7.	Anti-depressant activity	*Z. alatum* possess good anti-depressant activity. Mostly the hexane extract show good potency of anti-depressant activity. Molecular and biochemical studies give significant result.	[36]
8.	Memory enhancing property	*Z. alatum* was studied for memory enhancer by use of various hydroalcohlic extract.	[37]
9.	Cytotoxicity	Leaves of *Z. armatum* show good result in cytotoxicity and in anti-oxidant potential. The plant also useful to increase the efficiency of the chemotherapeutic drugs such as mitomycin C, cisplatin and camptothecin.	[38]
10.	Cardiovascular Activity	*Z. armatum* show antispasmolytic effect and arrythmic effect in crude extract. It may inhibit by calcium antagonistic mechanism which may use for Gastrointestinal effect, respiratory and cardiovascular disorders.	[39]
11.	Mosquito repellant activity	The mosquito repellent property of the essential oil of *Z. armatum* was performed against mosquitoes in mustard and coconut oil base. The results were compared with synthetic repellent dimethyl phthalate (DMP) which was used as standard. The oil has shown better protection in both the bases at all the concentrations.	[40]
12.	Leech repellant activity	Volatile oil of *Z. armatum* has leech repellent action. Investigations on perseverance of anti-agents properties of N, N diethyl phenyl acetamide (DEPA) N, N diethyl phenyl m-toluamide (DEET), 3-acety 2 (2, 6-dimethyl-5-heptenyl) oxazolidine (citronyl), Dimethyl phthalate (DMP) and N-benzoyl piperidine (NBP) on material were tried against land leeches in evergreen rain and deciduous backwoods of Assam.	[41-43]

4. Biological activities

Essential oils are secondary metabolites in plants with a variety of biological activities. Leaf essential oils of *Z. rhoifolium* and *Z. setulosum* were tested for antibacterial activity against *Bacillus cereus*, *Staphylococcus aureus*, *Pseudomonas aeruginosa*, and *Escherichia coli* and for *Artemia salina* (brine shrimp) lethality. Neither *Z. rhoifolium* nor *Z. setulosum* leaf oils exhibited cytotoxicity or antibacterial activity. Both oils showed activity against *A. salina* [19]. Biological activities of the produced essential oils were also investigated. Essential oils of *Z. leprieurii* exhibited high insecticidal activities against *Sitophilus granarius*, as well as antioxidant, anti-inflammatory and moderate anti-plasmodial properties [23]. Some other activities are summarized in given Table 1.

5. Conclusions

This review shows that the genus *Zanthoxylum* is very rich in beneficial chemical components in essential oils and in different extracts. Essential oils and extracts exhibit significant biological and pharmacological activities. Several species are very useful in the present scenario for daily life in fragrance and perfumery. As well as in aromatherapy and many other purposes, nowadays aromatic plants and essential oils are used in cosmetic industries. The plant can be a successful option for therapeutic and agro-industries application. We can conclude that the genus *Zanthoxylum* is very useful in many ways in daily life so more studies and commercial production of essential oils will be useful to society, perfume, cosmetics, and pharma industries.

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Conflict of Interest: There is no any conflict of interest.

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