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Review Article

Review on Bioactive Potential of Indonesian Forest Essential Oils

Yelfi Anwar[1,*], Gunawan Pasaribu[2], M. Nazari V[1]

[1]*School of Pharmacy, University August 17, 1945, Jakarta, INDONESIA.*

[2]*Research Center for Biomass and Bioproducts, National Research and Innovation Agency, Jakarta, INDONESIA.*

Correspondence

Yelfi Anwar

School of Pharmacy, University August 17, 1945, Jakarta, INDONESIA.

E-mail: yelfi.anwar@uta45jakarta.ac.id

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ABSTRACT

Essential oils are composed of some natural, volatile, as well as aromatic compounds extracted from plants. In recent years, several studies have shown that some of their benefits can be attributed to their antimicrobial, antioxidant, anti-inflammatory, and also immunomodulatory properties. So, essential oils have been proposed as a common elective to anti-microbials or for utilize in combination with other anti-microbials against multidrug-resistant microscopic organisms. Most of the potential data about essential oils were obtained with *in vitro* and *in vivo* studies. Several types of essential oils are available in Indonesia which are reported to have biological activity such as antioxidants and antiaging are essential oils of pine (*Pinus merkusii*), kilemo (*Litsea cubeba*), agarwood (*Aquilaria* spp), eucalyptus (*Eucalyptus citriodora*), and cinnamon (*Cinnamomum burmanii*). The major chemical compound of pine oil is alpha pinene. Kilemo oil contains geranial. chemical marker compound of agarwood is chromone and sesquiterpenes. Eucalyptus contains citronella. The chemical component of cinnamon oil is cinnamaldehyde. Some essential oils have shown remarkable antioxidant activities when used at specific concentrations which can be due to their richness in phenolic compounds. However, toxicological studies are needed before any recommendation for application can be given. Preformulating and formulation studies will be needed to develop suitable dosage forms in order to introduce optimized pharmaceuticals (high active, low toxic) as alternative of current pharmaceutical dosage forms

Key words: Pharmaceutics, Bioactivity, Chemical compound, Essential oil, Indonesian forest.

INTRODUCTION

Indonesia as one of tropical country has different range of biodiversity, and also, is one of the producers of different essential oils that have many health benefits and advantages. In the regulation of the Indonesian Ministry of Forestry no. 35 there are twenty kinds of Non Timber Forest Products (NTFPs) which are classified as essential oils extracted from the forest.[1] Forestry-based essential oils are generally traded and exported in the form of raw materials. There are some gaps to research about how to increase the value of these essential oils. Various studies in other countries have shown that the bioactivity of these essential oils can be used in different aspects, such as application of them as antioxidants or even and antiaging agent.

Pine (*Pinus merkusii*) is a native tree in Indonesia which has main distribution in islands of Sumatra and Java. It belongs to the conifer group with the Pinaceae family. The pine oil is obtained from the distillation process which produces the solid part in the form of gum rosin and the liquid part, namely turpentine oil. This oil is used as a raw material for cosmetics, paint oil, solvent mixtures, antiseptics, camphor and medicines, especially as a thinner in paint industry. While gum rosin is used in variety of related industries, such as paper, ceramics, plastic, paint, batik, soap, pharmaceuticals and cosmetic industries.[2]

Kilemo (*Litsea cubeba* Lour. Pers) is in the family of Lauraceae. Litsea spreads from Asia, South and North America, tropical and subtropical Australia and New Zealand. This genus consists of approximately 622 species. Kilemo grows wild in Southeast Asia, including India, China, Bhutan, Nepal, Myanmar, Vietnam, Korea, Taiwan and Indonesia. In Indonesia, *Litsea cubeba* is known as kilemo or antarasa which grows naturally in North Sumatra and mountains of West Java.

Agarwood-producing trees consist of six genera, which include Aquilaria, Wikstroemia, Enkleia, Aetoxylon, Gonystylus and Gyrinops. Agarwood is used as an herbal perfume, soothes (relieves) stomach pain, cough medicine, rheumatism medicine, antitumor, anticancer, and antimicrobial agent.[3]

Eucalyptus is one of the Myrtaceae family which is known as a source of aromatics. Several types of eucalyptus that grow in Indonesia include *Eucalyptus globulus, Eucalyptus urophylla, Eucalyptus pellita*, and *Eucalyptus citriodora*. Two types of *E. globulus* and *E. citriodora* were the most widely developed plants because the quality of the essential oil was shown the best quality compared to others. Their bioactivity has been researched in different researches which showed the efficacy in analgesic, antifungal, anti-inflammatory, inhibition of bone resorption and natural repellent.[4] Cinnamon oil is produced by distillation of cinnamon (*Cinnamomum burmanii*) bark. The main distribution of cinnamon plants is in Jambi and West Sumatra. The main chemical compound of its essential oils is cinnamaldehyde and eugenol compounds. Other research reports another component in the form of alcohol: Methanol, cyclohexane. Aldehydes: Trans-cinnamaldehyde. Acetate: Trans-cinnamyl acetate; bornyl acetate; acetate; bornyl acetate. Terpinene: (-)-Spathulenol; caryophyllene; D-borneol; eucalyptus; guaiole.[5]

This review contains related study about chemical and bioactivity effects of five forest's essential oils. The Information was collected from scientific journals, books, and reports.

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ESSENTIAL OIL PLANTATION

In general, NTFP harvesting in Indonesia is not implemented as a sustainable system, which results in uncontrolled resources. Likewise, the cultivation system is still traditional.[6] Indonesian Pine (*Pinus merkusii*) is one of Sumatra endemic tree species which originally from Aceh, North Sumatra and Kerinci (Jambi).[7] This species is developed for plantation by Perum Perhutani in Java. *Pinus merkusii* belongs to conifer group in the family of Pinaceae. In Minister of Forestry Regulation No. 35 of 2007, it is stated that pine resin is one of a NTFPs in the resin group.[1] Pine resin is obtained through the process of tapping the pine tree. Based on Forestry Statistic data in 2018, it was stated that the realization of pine resin production in 2018 was 103,324.24 tons that spread in 15 provinces in Indonesia. This production is dominated by Perhutani's production, which reached to 90,000 tons with an area of approximately 800,000 hectares of pine stands.[8]

Different with kilemo (*Litsea cubeba*) plants, it has not been planted on a large scale. The current stands are generally natural, both in West Java and in North Sumatra. The diameter of stem is 6–20 cm and the height of tree is 5–12 meters.[9]

Agarwood-producing tree species, especially *Aquilaria* and *Gyrinops,* which are six and seven species respectively, grow naturally in almost all of region in Indonesia from Sumatra to Papua, mainly in mineral soils at an altitude of 0-1000 m above sea level as co-dominant and mixed with various tropical rainforest tree species. The trees are also found in India, Bangladesh, Sri Lanka, Indo-China, Thailand, Malaysia, and Papua New Guinea. Most of the agarwood came from natural forests, but some valuable commodities are managed on private lands or community forests to protect assets or security reasons in the long term.[6] Since 2000s, around 3.5 million agarwood-producing tresses, including Aquilaria and Gyrinops, have been planted in different forests or gardens in Indonesia (Figure 1).

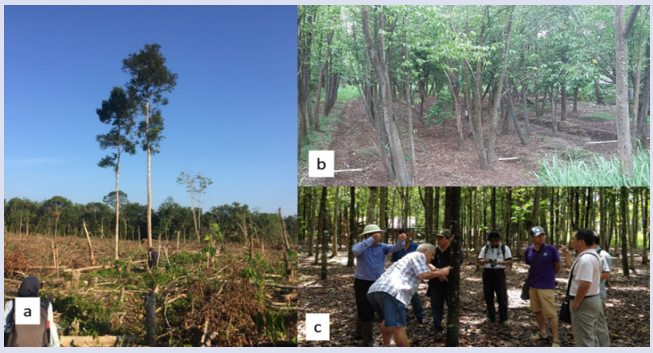

Figure 1: Natural *Aquilaria malaccensis* in South Sumatera (a); *Aquilaria malaccensis* plantation in Banten Province (b); *Aquilaria microcarpa* plantation in West Kalimantan (c)[6].

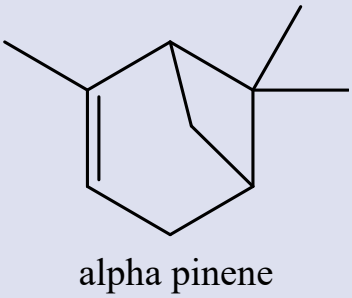

Figure 2: Major component of pine oil.

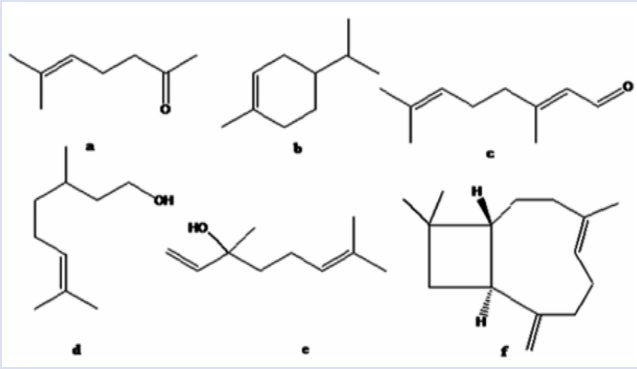

Figure 3: Chemical compound in fruit part: a. 6-methyl– 5-hepten, b. D-limonene, c. Z-citral, d. alpha-citronellol, e. l-linalool, f. trans-caryophyllene[20].

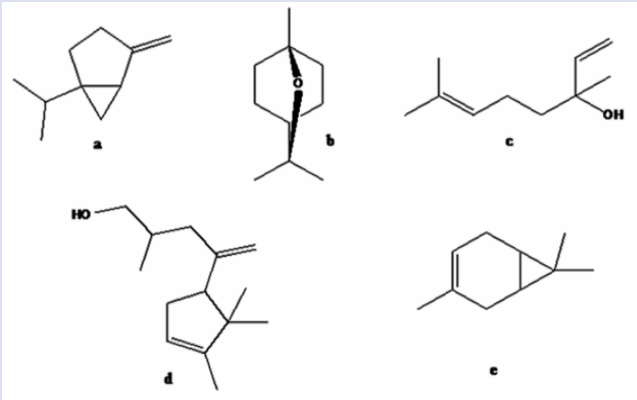

Figure 4: Chemical compound in leaves part: a. sabinine, b. eucalyptol, c. linalool, d. cyclohex-3-en-1-ylmethane, e. δ-3-carene[20].

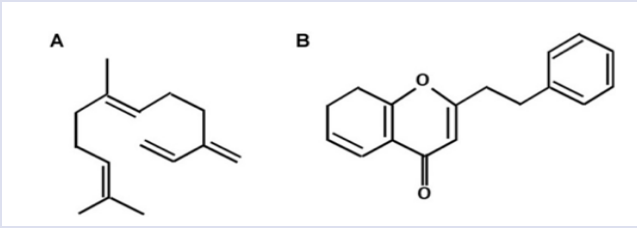

Figure 5: Sesquiterpene (A) and 2-(2-phenylethyl) chromones (B) from agarwood.

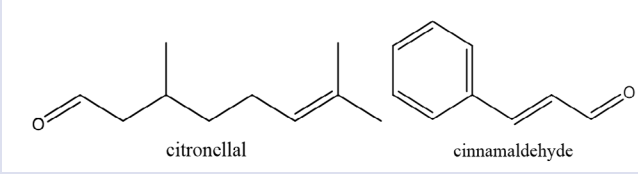

Figure 6: Citronellal and cinnamaldehyde compound.

However, agarwood plantation faced some challenges, including the inoculation process, that requires effective fungal species of agarwood formation.[10-12] In addition, inoculum practice on large scale requires high-quality agarwood-forming inoculants and skillful labors. As a semi-tolerant species, agarwood-producing trees need shade at the beginning of their growth. In this case, an agroforestry system that combines tree planting with annual crops such as bananas,

Table 1: Bioactivity of several essential oil.

No	Species	Bioactivity	References
1	Pine oil	Antibacterial: Resin extract from *Pinus oocarpa* has the diameter inhibition from concentration of 500 µg/mL, while the *Pinus insularis* and *Pinus merkusii* has diameter inhibition from 1000 µg/mL. This phenomenon show is that the resin extract of *Pinus oocarpa* was the most active as antibacterial against Staphylococcus aureus than all samples.	37
		Antioxidant: IC_{50} of n- hexane *P. oocarpa* 154.500; n- hexane *P. insularis* 99.328; n- hexane P. *merkusii*; 60.203 n- hexane; Turpentine *P. insularis* 359.687; Turpentine *P. oocarpa* 1194.250; Turpentine *P. merkusii* 1119.960.	37
		Anticancer: the MTT test showed the IC_{50} 57.92 ± 1.08 µg/mL, causing accumulation in the G0-G1 phase (64.52% to 70.45%) and increasing p53 expression (0.66% to 61.98%).	23
		The other research showed that Litsea essentisl oil from fruit were create having IC_{50} 72.86 ± 1.03 µg/mL This oil fruits have cytotoxicity effect by inhibited cycle inhibition and induction apoptosis.	22
2	*Litsea cubeba* oil	Antioxidant: · The percentage of inhibition vary from 6.18-40.35%. · The methanol extract, chloroform fraction and buthanol fraction showed inhibition value 9–90% of lipid peroxidation with TBA method. · Antioxidant activity of kernel oils originated from China have IC_{50} value about 7.20 ± 1.91 mg/mL.	24, 38
		· Anti-inflammatory: The inhibition activity in fruit and leaf oil up to 82.21% on concentrations 0.20 mg/mL and 0.25 mg/mL respectively. The IC50 values on fruit and leaf is better than diclofenac standard.	20
		Antibacterial: The MIC of Litsea oil against MRSA was 0.5 mg/mL and the MBC was 1.0 mg/mL, which showed a high inhibitory activity of Litsea oil against MRSA. *Litsea cubeba* essential oil is an effective bacterial inhibitor and bactericide against MRSA.	39
		Antimicrobial: · The minimal microbicidal concentrations (MMCs) for *Streptococcus sanguinis*, 375µg/mL, *Streptococcus mutans* 750µg/mL and *Streptococcus sobrinus* 1500µg/mL. Difference with the MMC for each strain with the biofilms increased twice in its resistance to bacteria. · The MMCs of Litsea oil at Vibrio spp. in oysters was 3000 µg/g, L. plantarum in orange–milk beverage was 6000 µg/g, and *H. anomala* in soy sauce was 375 µg/g. Generally, the food systems exhibited marked matrix effects on diminishing the antimicrobial activity of the Litsea oil. · The suppression zones against *Bacillus cereus*, *Staphyloccus epidermidis* and *Escherichia coli* is not good enough. Fruit oil, on the other hand, at 2 µL/disc, exhibited better. The fruit oil showed excellent antimicrobial activities.	40-42
		Antidiabetic: By inhibition alpha glucosidase, the major compounds in the leaves of *Aquilaria sinensis*, exhibited very strong inhibition with IC_{50} values are 126.5 ± 17.8 and 143.7 ± 10.6 lg/mL, respectively.	43
		Hepatoprotector: Aquilaria a agallocha leave extract with 400 mg/kg/day was comparable to that of standard drug silymarin 100 mg/kg/day.	44
3	Agarwood oil	Anticancer: *Aquilaria crassna* essential oils showed effective cytotoxic activity against MIA PaCa-2 cells with an IC_{50} (11 ± 2.18 lg/ml). The cell migration was effectually inhibited at (10 lg/ml).	45
		Anti-inflammatory: Isolated compounds from *Aquilaria malaccensis* showed the potential activities. The nitric oxide inhibitory effects with an IC_{50} value of 18.8 µM. For other compounds exhibited moderate inhibition of Nitric oxide production.	46
4	*Eucalyptus citriodora* oil	Antioxidant: the DPPH radical scavenging activity of the oil and BHT were found to be 92.1 and 91.5%, respectively.	27
		Antimicrobial: The zone of inhibition of E. citriodora oil against *Staphylococcus aureus* (31 mm), *Bacillus subtilis* (28 mm), *Escherichia coli*, *Aspergillus niger* (29 mm) and *Rhizopus solani* (18 mm). These results indicated that all the tested microorganisms were affected by potential ingredients of the essential oil	27
		Antidiabetic: On in vivo testing, the oil increased significantly glucose uptake by 3T3-L1 cells and repressed digestion of starch, glucose absorption, DPP-IV enzyme and glycation of protein. Dosage in 250 mg/5 ml/kg enhanced glycemic control and β-cell function.	47
		Antiproliferative: Its major 6-[1-(p-Hydroxy-phenyl) ethyl]-7-O-methyl aromadendrin (HEMA), a flavanol derivative, and the resin showed high antiproliferative effects on B16F10 cells.	48
5	Cinnamon oil	Antioxidant: DPPH 3.45 µg/mL; potency inhibit the free radicals at concentration 25 – 100 ppm.	35, 49
		Antidiabetic: α-glucosidase inhibition; IC50=0.50 µg/mL	35
		Antimicrobial: zone of inhibition of *Staphylococcus aureus* is 18.5 mm and *Candida albicans* 10.7 mm. Cinnamon stick inhibition *Staphylococcus aureus* was the most sensitive (15.7 mm)	49, 50

fruits, rubber trees, citronella, and vegetables can also be carried out, especially on limited land.[13] The trees are also possibly planted in yard (home garden). Planting in gardens around the residence is easy to monitor for safety, inoculation, and harvesting purposes.[14]

Eucalyptus species have been widely developed in Indonesia, especially for pulp and paper production. But still the production of some essential oils such as Eucalyptus citriodora, has not been widely developed in Indonesia. In fact, eucalyptus oil is widely used in the production of aromatherapy and rubbing oils in Indonesia.[15]

In Indonesia, West Sumatra and Jambi are the center for cinnamon production, and is one of the commodities superior. Cinnamon cultivation is not the main livelihood of farmers. Farmers' income is only 26.93% of their farming results, or 16.03% of total farmers' income.[16]

PHYTOCHEMICAL AND CHEMICAL COMPOUND

The main compound and their distribution in pine were α-pinene from73.3 to 87.2% and δ-3 carene, from 7.3 to 19.3%. The minor compound (< 3%) were β-pinene, camphene, myrcene, sabinene and limonene. Sabinene was not found in Java land race and Blangkejeren, while limonene was not found in Takengon.[17] The major components of pine resin are turpentine and rosin.[18] After distillation process, the liquid part of the pine resin is turpentine which is volatile. Formed by monoterpenes (alpha pinene, beta pinene, limonene, -3-carene and myrcene) - and sesquiterpenes (caryophyllene and farnesene). The

solid part is gum rosin which is formed by diterpenes (abietic acid, dehydroabiic acid, and isopimaric acid).[2]

The compositions of kilemo (*Litsea cubeba*) bark essential oil and their distributions are 1,8-Sineol (26,59 %), sitronelol (21,69 %), Linalool 9,93%, mircena 9,24%, sitronelal (8,68 %) dan α-terpenil asetat (8,12 %).[19] Furthermore, the kilemo's fruit and leaf essential oils of North-east India is different. Its major compound in oil part of fruit were methyl heptenone (30.9%), limonene (23.14%), Z-citral (14%), alpha-citronellol (6.52%), 3,7-dimethyl-octa-1,6-dien-3-ol (linalool) (2.26%) and the evaluated compounds in oil part of leaves were trans-caryophyllene (1.73%) and sabinene (25.22%), eucalyptol (16.8%), linalool (10.1%), 4-methyl-cyclohex-3-en-1-ol (8.7%), cyclohex-3-en-1-ylmethane (6.94%) and delta-3 carene (5.84%).[20]

Different from essential oils from China that the main oil chemical compound was identified as geranial (27.49%), Z-citral (neral) (23.57%), D-limonene (18.82%), β-thujene (3.34%), β-pinene (2.85%), α-pinene (2.57%), 6-methyl-5-hepten-2-one (2.40%) and linalool (2.36%).[21] On the other hand, *Litsea cubeba* oil contain phytochemistry compound such as alkaloids, flavonoids, steroids/triterpenoids, saponins, tannins and glycosides.[22]

Ethanol extract of *Litsea cubeba* fruit contain alkaloids, flavonoids, steroids, triterpenoids, saponins, tannins and glycosides.[23] Furthermore, the total phenols and total flavonoids of essential oils in *Litsea cubeba* fruit and leaf ranged from 2.019 mg per GAE (Gallic Acid Equivalent), 1.87 mg in quercetin equivalent (QE) and 7.99 mg GAE, 6.344 mg QE respectively. The total phenolic gallic acid found 1.06 mg mL^{-1} of quercetin is 0.972 mg mL^{-1} which is lower than the two samples tested. The total phenol and total flavonoid in fruit was higher than that of the leaves.[24]

The most compound from 154 compounds identified from Aquilaria plants are , 2-(2-phenylethyl)-4H-chromen-4-one derivatives (57%) and sesquiterpenes (35%).[25] *Aquilaria malacensis* and *Gyrinops versteegii* are the most commonly used species and are still found in Indonesian forests. Agarwood oils *A. malaccensis* contain numerous sesquiterpenes such as sesquiterpenes alcohols, oxygenated compounds, hydrocarbons, and acids. Six out of 43 compounds sensed in agarwood oil as remarkable compounds. Those compounds are 4-phenyl-2-butanone, valencene, curcumene, β-dihydroagarofuran, 10-epi-ϒ-eudesmol and α-guaiene, α-gurjunene, β-copeane, ϒ-elemene, aromadendrene, valencene, ϒ-gurjunene, elemol, β-vetivenene. Aromadendrene, β-agarofuran, 10-epi-ϒ-eudesmol and ϒ-eudesmol have been informed as important compounds in the *A. malaccensis* agarwood oil. Aromadendrene compounds can act as effective chemical distinguisher for agarwood. This chemical compound characterizes agarwood. Increasing the content of aromadendrene causes, some increase in the quality of agarwood oil.[26]

The higher content of essential oils in steam distilled *Eucalyptus citriodora* leaves was recorded to be up to 1.82%. It consists of citronellal, citronellol, patchoulene, germacrene D, α-terpinol, sabinene, γ-phellandrene, eugenol, α-pinene, β-citronellal, citrinyl acetate, geranial oxime, paraldehyde nitrile. The remaining 18 minor components were lower than 1% each.[27] The major compounds and quantity of *E. citriodora* from Espírito Santo State, Brazil were citronellal (29.31 %), geraniol (27.63 %), β-citronellol (14.88 %) and cadinene (6.32 %).[28]

The major components of *Cinnamomum burmannii* oil from South Bandung, Indonesia was determined as cinnamaldehyde (68.3%-82%), cinnamyl acetate (2.5%- 16%), cinnamyl alcohol (2.25%-4.6%), and cinnamic acid (3%-8%).[29] Chemical content of cinnamon bark are an alkaloid, flavonoid, tannins and essential oils consisting of camphor, safrole, eugenol, cinnamaldehyde, cinamylacetate, terpene, cineol, citral, citronellal, polyphenols and benzaldehyde.[30] Cinnamon essential oil or oleoresin contains several chemical compounds such as cinnamal aldehyde, eugenol, methyl ketene, furtural, benzaldehyde, nonyl aldehyde, hydrocinnamic aldehyde, cuminaldehyde and coumarin.[16] Cinnamon essential oil contains biologically active compounds, such as cinnamon aldehyde, cinnamic alcohol, cinnamic acid, and cinnamate.[31]

BIOACTIVITY OF ESSENTIAL OIL

There are various ethnopharmacological applications and uses of essential oil in *Litsea cubeba* such as in pharmaceutical industry such as antimicrobial, antioxidant, antidiabetic, anti-inflammatory, antiasthma, diarrhea and arthritis.[32]

Furthermore, different types of agarwood are used, for the treatment of mothers after giving birth and reducing spasms in the digestive and respiratory systems. The other application was helping to control asthma attack, stomach pain, chest tightness, diarrhea, hiccups, nausea, nerves, colic, mental disorders, and cancer. Malaysians, Indians, and Chinese use agarwood oil mixed with coconut oil as a lotion to treat various skin diseases. In addition, agarwood oil is also helpful in rheumatism, lung disease, stomach tumors, carminative diuretics, and other types of body pains. Traditional Chinese and Japanese medicine use agarwood as a sedative, analgesic, and digestive agent.[6] Biological activity of agarwood compound and aquilaria trees are for neural activity, gastrointestinal regulation, antibacterial and antifungal, anti-Inflammatory, analgesic effect, antiasthma, cytotoxicity, anti-diabetes, antioxidation.[25,33]

The main compound of eucalyptus oil is 1,8-cineole, citronellil acetate. These compounds have bioactivity as antiseptic, anti-inflammatory, antibacterial, antifungal. Other forms of use are for the treatment of asthma, bronchitis, and sinus.[34]

In addition to the use of spices, cinnamon is also useful as a medicine, including antibacterial effects.[5] Its bioactivity includes antioxidant, anti-inflammatory, and antibacterial properties. Such essential oil is used for the treatment of diabetes and cardiovascular disease. Traditionally, cinnamon oil was used as a remedy for the respiratory and digestive disorders.[31,35] Furthermore, cinnamon also has the potential to be bioactivity as anti-diabetic, antitumor, anti-obesity, immunoregulation, insecticidal and acaricidal, cardiovascular protective, and cytoprotective.[36]

CONCLUSION

The results of the review show that quite a lot of research has been done related to forestry-based essential oils. Forest-based essential oils have a very wide range of bioactivity. The activities of essential oils include antibacterial, antifungal, antioxidant, anticancer, anti-inflammatory and antidiabetic activities. Next research is required due to increase benefits of Indonesian forest essential oils.

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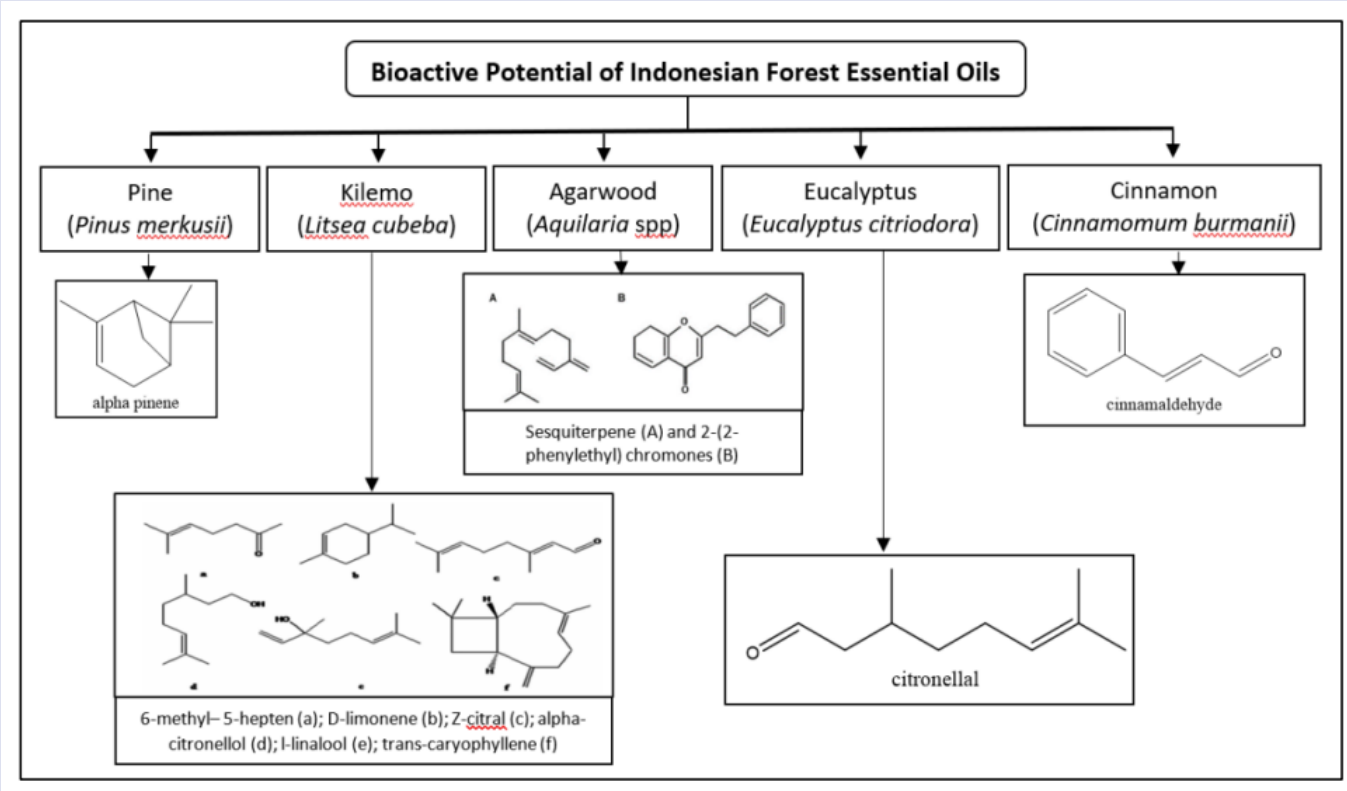
GRAPHICAL ABSTRACT



ABOUT AUTHORS

Yelfi Anwar (Ph.D) is a Lecturer and Researcher at School of Pharmacy, University August 17, 1945, Jakarta, Indonesia. She's current research are mainly on natural product, pharmacology, pharmacoeconomic, Cosmeceutical, Biotechnology / Biotransformation.

Gunawan Pasaribu (M.Sc) is a researcher at Research Center for Biomass and Bioproducts, National Research and Innovation Agency, Jakarta, Indonesia. He's current research are mainly on natural product chemical compound, isolation and its bioactivity for nutraceutical.

M. Nazari V (Ph.D) is a Lecturer and Researcher at School of Pharmacy, University August 17, 1945, Jakarta, Indonesia. She's current research are mainly on cancer research, bioinformatics and drug discovery of pharmaceutical compounds.

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