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# **REVEIW ARTICLE**

## A REVIEW: PATENT LITERATURE ON INVENTIONS CONTAINING ESSENTIAL OILS WITH MOSQUITO REPELLENCY

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ARTICLE INFO	ABSTRACT	
Article History: Received 18 <sup>th</sup> December, 2018 Received in revised form 29 <sup>th</sup> January, 2019 Accepted 18 <sup>th</sup> February, 2019 Published online 30 <sup>th</sup> March, 2019	Mosquitoes belonging to the general Aedes Meigen, Anopheles Meigen, Culex L., Haemagogus L. are a general problem and are responsible for the transmission of important tropical diseases such as malaria, hemorrhagic dengue and yellow fevers and filariasis. Traditional sources of mosquito repelling essential oils are plants, glyceridic oils and repellent and synergistic chemicals. A Chemical Abstracts search on mosquito repellent inventions containing plant based essential oils revealed 144 active patents mostly from Asia Chinese. Japanese and Korean Japanese patents and those of India written in English	
<i>Key words:</i> Mosquito repellent, Patent, Inventions, Essential oils, Chemical components. * <i>Corresponding author:</i> Pawankumar Rai	accounted for roughly $3/4$ of all patents. 67 essential oils and 9 glyceridic oils were individually cited in at least 2 patents. About 25% of essential oils containing inventions were made to be used with synthetic insect control agents having mosquito repellent properties such as pyrethroids, N,N-diethyl- m-toluamide (DEET), (±) p-menthane-3,8-diol (PMD) and dialkyl phthalates. Synergistic effects involving one or more essential oils and synthetic and/or natural components were claimed in about 10% of all patents. Scientific literature sources provide evidence for the mosquito repellency of many of the essential oils and individual chemical components found in the same to be used in inventions containing patented repellent.	

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### **INTRODUCTION**

In developed countries the urban population is expected to increase to more than  $70\%^1$  resulting in the increasing importance of urban pests. Similarly, for developing countries, the pests still remains as mosquitoes followed by others. Mosquitoes are the most important and abundant pest in urban, sub-urban and rural environment<sup>2</sup>. According to World Health Organization (WHO) Media Centre. Malaria;<sup>3</sup> malaria, dengue, yellow fevers and filariasis are severe human tropical diseases transmitted by the bites of infected hematophagous female mosquitoes belonging to the general Aedes Meigen, Anopheles Meigen, Culex L. and Haemagogus L. (Diptera: Culicidae). For example, about 3.3 billion people -1/2 of the world's population - are at risk of malaria. There were more than 247 million cases and more than 1 million deaths caused by malaria majorly in African children in 2008. Human malaria is caused by infections by unicellular protozoan parasites Plasmodium falciparum Welch, P. vivax Grassi and Feletti, P. malariae Feletti and Grassi and P. ovale Stephens which are transmitted by about 20 Anopheles species. According to World Health Organization (WHO) Media Centre. Dengue<sup>4</sup> Dengue hemorrhagic fever is another important disease threatening an estimated 2.5 billion people which is a viral infection caused by several *Flavivirus* spp. (*Flaviviridae*) whose most important vector is Aedes (Stegomvia) aegypti L. Around 500000 dengue patients, require hospitalization each

year and around 2.5% of those affected. From the studies of Burfield and Reekie<sup>5</sup>, Filariasis is another serious tropical disease which threatens about 1 billion people in 80 countries. This disease already affects an estimated 120 million people and severely incapacitates 40 million people worldwide. Infection caused by several roundworm species of which Wuchereria bancrofti Cobbold (Filariidea: Onchocercidae) is the most important and is transmitted by the bites of the common house mosquito Cx. pipiens L. complex, Cx. quinquefaciatus Say, Aedes and Anopheles spp. results in Filariasis which is also signified by WHO Media Centre. Filariasis<sup>6</sup>. Yellow fever is an arbovirus of the Flavivirus genus (Flaviviridae) which are transmitted from monkeys in the jungle to humans and then from human to human by mosquitoes. There are an estimated 200000 cases of yellow fever and approximately 30000 deaths attributed to this disease each vear despite the presence of numerous effective vaccines shown by WHO Media Centre. Yellow Fever<sup>7</sup>. Public health initiatives which works to limit or eradicate these and other tropical diseases, mosquito vector control methods such as repellence figure prominently among those which are employed.

**Synthetic mosquito repellents:** According to Brown and Hebert<sup>8</sup>; the synthetic compound DEET (N, N-diethyl- m-toluamide), has been the most effective single repellent for mosquito species and is the basis for many commercial

repellent products on the market from many years. Despite reports of severe toxic properties which can dramatically affect adults and especially young children including dermatitis, allergic reactions, neurological (seizures, coma) and cardiovascular toxicity, the risk of serious toxic effects from DEET is considered slight. Whereas, DEET should always be used at the lowest effective dose possible. Also, effective mosquito repellents such as dimethyl and di-n-butyl phthalates (DMP and DBP, respectively) were widely used in the last century, are no longer generally recommended for use as mosquito repellents due to their toxicity.

Mosquito preventive agents derived from plant sources: Burfield and Reekie<sup>5</sup> denoted in their study that plants have historically been valuable sources of agents for the control of insects. They are the sources of the natural insecticidal and larvicidal substances nicotine (Nicotiana L. spp.), quassin (Quassia amara L.), rotenone and rotenoids (Derris Lour. spp. and Lonchocarpus Kunth spp. roots), pyrethrins like chrysanthamic acid and its derivatives present in pyrethrum [extracts of Chrysanthemum cinerariifolium (Trevir.) Vis. flowers] and azadirachtin (Azadirachta indica A. Juss. seed kernel). Studies of Nerio *et al.*<sup>9</sup> also revealed that these and other natural insect control agents have served as the basis for the development of the structurally-related synthetic pyrethroid, nicotinoid and rotenoid classes of insecticides and piperonyl butoxide synergist. Also, pyrethroids and piperonyl butoxide synergism are the basis for a number of commercially available mosquito control products in use today.

Repellents from botanical source: From the studies carried out by Burfield and Reekie<sup>5</sup>, Citronella essential oils (Essential oils) are obtained mainly from varieties of Cymbopogon nardus (L.) Rendle (Ceylon citronella) and C. winterianus Jowitt ex Bor (Java citronella). They have been used in mosquito repellency for more than a century in much of the world and are the most widely used natural repellents today.Batish DR et al.<sup>10</sup>in their study signified that essential oils of Eucalyptus L' Hér. spp. are widely used to repel insects, including mosquitoes, and contain insecticidal and repellent components p menthane-3,8-diol (PMD), 1,8-cineole,  $\alpha$ - pinene, p-cymene and  $\gamma$ -terpinene among other active compounds. According to a recent review of the scientific literature by Nerio et al,<sup>9</sup> the most frequently studied repellent Essential oils are those obtained from species belonging to the general Cymbopogon Spreng., Ocimum L. and Eucalyptus L' Hér. spp. and a number of mosquito repellent essential oils have been identified in recent years having known active repellent chemical components. Mosquito repellency is believed to be due to the synergistic interactions of the chemical components in essential oils. Furthermore, strong synergistic effects between Essential oils and isolated natural or synthetic substances have been reported. The United States Environmental Protection Agency<sup>11</sup> has registered citronella oil, eucalyptus oil and other plant oils as safe and effective ingredients for use in topical insect repellents. However, caution is recommended by Nerio LS et al.9 in the use of essential oils in general due to a number of potential toxic effects. Among the important toxic effects of essential oils are mutagenicity and genotoxicity. Another toxic effect is the allergenicity of essential oils chemical components which are controlled in the European Community and elsewhere. According to Bakkali *et al*,<sup>12</sup> a number of repellent and insecticidal plant essential oils and their chemical constituents have been evaluated using a variety of methods and are believed to be non-mutagenic. A number of commercial repellent products have been developed over the past decades which utilize derivatives of plants such as essential oils, fractions and their isolated chemical components and synthetic components. Thus, it is important to have a comprehensive knowledge of the commercially significant uses of essential oils in mosquito repellent inventions and to the best of our knowledge the patent literature on this topic has not been reviewed. The aim is to analyze the scientific bases and relevancy of the use of plant essential oils and chemical components of these oils in patented mosquito repellent formulations. The another approach of the present review is to explore and analyze patent literature on mosquito repellent inventions which make use of or are based wholly on plant essential oils.

Guideline for Chemical Abstracts Search: A Chemical Abstracts search of the patent literature for the period of 1991 through present was performed using Sci Finder Scholar<sup>®</sup>.<sup>13</sup>Combinations of the key words "repellent", "mosquito" and "essential oil" generated a primary set of approximately 160 patents. These sets were further refined. Patents describing mosquitocidal/ larvicidal inventions, but having no stated claim or use as mosquito repellents were not selected. Also, patented mosquito repellent inventions which did not make use of at least one commercially obtained, plant derived oil [a volatile (essential) oil, a concrete, or a pressed oil] or which did not describe the preparation of an essential oil by physical means for use within the patent were eliminated. This approach eliminated patents presenting solvent extraction performed on mixtures of several plant materials followed, for example, by distillation and evaporation as a means of acquiring the mosquito repellent invention. Also, by this criterion, incense and other smokegenerating inventions made from mosquito repellent plants but having no essential oil in the composition were also not selected. On these basis, data set of 144 patents describing plant oil containing mosquito repellent inventions which was the basis for the analysis.

Trends of patenting countries: There has been a significant contribution by Asian countries to the overall number of patented essential oils containing mosquito repellent inventions, contribution of China, Japan, Korea and India accounted for, respectively, 37, 15, 13 and 8% (73%) of all patents. An interesting trend is that all 53 Chinese, 17 of 18 Korean, 20 of 22 Japanese and 9 of 11 Indian patents have only been deposited in their countries of origin and in general have not been followed up by later patents. Also, all Belgian, Brazilian, Polish and German patents followed the rule of single deposits without follow up patents. In contrast, industrial, academic and other patents originating in Australia, Canada, the United Kingdom and the United States gave rise to higher patent families, made greater use of the World Patent System and led to deposits of patents on essential oil containing repellents in multiple countries as a rule. Finally, more than 75% of all patented repellent inventions are indexed as agrochemical bio regulators and over 80% describes chemical compositions (often together with all preparation details).

**Patented mosquito repellent inventions containing essential oils:** From Tropicos.org.<sup>14</sup> Missouri botanical garden; Plant taxonomic information was collected, whereas extraction methods, chemical composition, literature sources in all other references [8-117] and frequency of use for 67 essential oils and 9 glyceridic oils which were individually cited in at least 2 (1.4%) of the total number of patents are presented in following table. Approximately 60% of all patents named just one plant EO. One patent claimed to use almost 44 plant oils in formulations. Citronella (34.7%) and eucalyptus (30.6%) essential oils were each named in about 1/3 of all patented repellent inventions followed by (species, % of patents): lavender (Lavandula angustifolia Mill., 21.5%), peppermint (Mentha piperita L., 16.7%), clove [Syzygium aromaticum (L.) Merr. and L.M. Perry, 15.3%], lemongrass [Cymbopogon citratus (DC.) Stapf, 14.6%], cinnamon (Cinnamomum zeylanicum Blume, 12.5%), geranium (Pelargonium graveolens L'Her., 11.8%), camphor [Cinnamomum camphora (L.) J. Presl, 11.1%] and lemon [Citrus Limon (L.) Osbeck, 11.1%] essential oils. Many Essential oils were cited in only one patented repellent invention and were not included in above shown Table. Due to the given the importance of potentially novel sources of mosquito repellent botanicals it is important to mention the following Essential oils:

Almond [Prunus dulcis (Mill.) D.A. Webb], Australian yuzu (Citrus junos Siebold ex Tanaka), black pine (Pinus nigra Arnold), Blumea lacera (Burm. f.) DC., Calamus (Acorus calamus L.), Canada fleabane [Conyza canadensis (L.) Cronquist], Cardamom [Elettaria cardamomum (L.)Maton], cork tree (Phellodendron amurense Rupr.), Flos lonicerae japonicae (Lonicera japonica Thunb.Ex Murray flower), fructus forsythiae [Forsythia suspensa (Thunb.)Vahl], Galbanum (Ferula galbaniflua Boiss.& Buhse), Herba schizonepetae [Schizonepeta (Benth.)Briq.sp.], Hibiscus (Hibiscus L. sp.), Larch (Larix Mill. sp.), Lemon balm (Melissa officinalis L.), Lovage (Levisticum officinale W.D. J. Koch), Lilac (Syringa vulgaris L.), Limnanthes Alba Hartw.Ex Benth.Seed, linaloe wood (Bursera delpechiana Poiss. ex Engl.), Michelia Alba DC.Leaf, myrtle (Myrtus communis L.), Ocimum canum Sims, onion (Allium cepa L.), oregano (Origanum vulgare L.), peach [Prunus persica (L.)Batsch], pennyroyal (Mentha pulegium L.), pepper (Capsicum annuum L. var. annuum), petitgrain (Citrus aurantium L. var. amara), pimento [Pimenta dioica (L.)Merr.], pine needle (Pinus L. sp.), Rhodomyrtus tomentosa (Aiton) Hassk; Rue (Ruta graveolens L.), Stephania sinica Diels, tansy (Tanacetum L. sp.), Torreya grandis Fortune ex Lindl.and valerian (Valeriana L. sp.).Nevertheless, we will not systematically go into details of the composition and repellency of these essential oils in this review. Generally, patent literature on mosquito repellent inventions treats fragrant extracts as essential oils whether these extracts are produced using an initial solvent extraction step, by steam distillation, hydrodistillation and direct distillation or pressing resulting in proper essential oils. Mainly, fragrant oils and partially volatile balsamic oils which have volatile and non-volatile (Glyceridic) chemical components in their compositions and mosquito repellent properties, such as evening primrose (Oenothera biennis L.), perilla [Perilla frutescens (L.) Britton], Ligusticum (Ligusticum chuanxiong Hortorum ex Qiu, et al. in Qiu) and Copaiba (Copaifera L. spp.) oils are used in mosquito repellent patented formulations together with plant Essential oils.<sup>118-171</sup>

Patented Mosquito Repellent Inventions containing Glyceridic oils: Several plant glyceridic oils which are known to possess mosquito repellency were used in patented formulations as carriers or active ingredients which were associated with prolonged repellent action. The most used glyceridic oils in patented essential oils -containing repellent inventions were Neem/Margarosa (*Azadirachta indica* A. *Juss.*, 8.3%), castor (*Ricinus communis* L., 4.9%), Soybean [*Glycine max* (L.) Merr, 4.2%] and sesame (*Sesamum indicum* L., 3.5%) oils.<sup>172-181</sup>

Scientific Basis for Mosquito Repellency of Essential oils in Patented Inventions

The scientific literature on the mosquito repellence of Essential oils used in patents provides important insights (Table 2). Firstly, citronella (Cymbopogon nardus, C. winterianus), eucalyptus (Eucalyptus spp.) and lemon eucalyptus (E. citriodora Hook.) Essential oils which were cited in many patented inventions have been the subject of a number of studies in which repellency against species of Culex, Anopheles and Aedes in some cases comparable to DEET have been reported for these oils alone or in formulations [Tawatsin *et al.;* Thorsell *et al.;* Moore *et al.*]. <sup>140,142,145</sup> Furthermore as stated by Gibson<sup>139</sup> many essential oils used in patented inventions have quite significant repellent properties according to published studies such as: bay laurel (Laurus nobilis L.), Camphor (Cinnamomumcamphora) [Yang et al.]<sup>134</sup>, Cassia (Cinnamomum cassia) [Yang et al.]<sup>134</sup> Essential oils against Ae. aegypti; Lemon (Citrus limon) EO [Amer *et al.*]<sup>130, 154</sup>against An. stephensi Liston, catnip (Nepeta cataria L.) EO [Amer and Mehlhorn]<sup>130</sup>, Lemongrass (Cymbopogoncitratus) [Amer and Mehlhorn H]<sup>130</sup>, May chong/litsea (Litseacubeba (Lour.) Pers.) [Amer A, Mehlhorn H; Noosidum *et al.*]<sup>130</sup>, tagetes (Tagetesminuta L.) [Amer A, Mehlhorn  $H_{1}^{130}$ , violet (Viola odorata L.) EOs [Amer A, Mehlhorn  $H_{1}^{130}$  against Ae. aegypti, An. stephensi and Cx. quinquefasciatus; peppermint (Mentha piperita) EO on human skin against An. annularis van derWulp, An. culicifacies Giles and Cx. Quinquefasciatus [Ansari MA et al.]<sup>162</sup>; Sandalwood (Santalum L. spp.) EO formulations against a Culex sp. [Ritchie SA]<sup>179</sup>; Geranium (Pelargonium graveolens) oil formulations [Gibson A; Kim S I *et al.*; Osmani Z *et al.*; Qualls WA, Xue R D]<sup>139,146,149,150,169,</sup> against species of Culex, Anopheles and Aedes; thyme (Thymus vulgaris L.) EO against Cx. Quinquefasciatus [Amer A, Mehlhorn H; Gibson A.; Pavela  $R_1^{130,139164}$ , Marjoram (*Origanummajorana* L.) and Juniper (Juniperus communis L.) Essential oils against Cx. Pipienspallens Coquillet [Kang et al.]<sup>153</sup>; and wintergreen (Gaultheria procumbens) EO against species of Culex and Aedes. A broad-scale screening of plant oils against Ae. aegypti, An. Stephensi and Cx. quinquefasciatus evaluated protection periods and percent of repellence on human skin as compared to 20% DEET [Amer A, Mehlhorn H]<sup>130</sup>. In general, Ae.aegypti was the most difficult species to repel, followed by An. stephensi and finally Cx. Quinquefasciatus for both oils and controls. The control (DEET) exhibited a protection period (PP) of 6 h and percent repellency (R%) of 46% against Ae. whereas against An. stephensi aegypti, and Cx. Quinquefasciatus protection was for 8 h at 100% repellency [Amer A, Mehlhorn H]<sup>130</sup> (Table 2). While the most active Essential oils against all three mosquito species were cited above, the following oils were active against An. Stephensi and Cx. quinquefasciatus, but not significantly active against Ae. Aegypti: Chamomile (Chamaemelum nobile), cinnamon (Cinnamomum verum), Galbanum (Ferula galbaniflua), Jasmine (Jasminum grandiflorum), lavender (Lavandula angustifolia), Pepper (Piper nigrum), Rosemary (Rosmarinus officinalis), Sandalwood (Santalum album) and Soybean (Glycine max) [Amer A, Mehlhorn H]<sup>130</sup>.

#### Table 1. Plant information, major chemical components, literature sources and (%) use of plant essential and glyceridic oils in patented mosquito repellent inventions

Oils	Part	Major components	Literature	% Patents
Ambrette	whole seed	2E,6E-farnesyl acetate, Z-7-hexadecen-16 olide, β-farnesene	Rout PK et .al <sup>15</sup> & Barik KC et.al <sup>16</sup>	1.4
Angelica	root	ligustilide, α & β-pinene, carvacrol, 3-carene, limonene, β-phellandrene, 15-pentadecanolide	Nivinskiene O et.al <sup>17</sup> & Wu M et.al <sup>18</sup>	1.4
Anise	fruit, seed	trans-anethole	Arslan N et. al <sup>19</sup>	4.2
Artemisia	leaf	germacrene D, α-phellandrene, α-myrcene, 1,8-cineole, borneol, terpinol, spathulenol	Wenqiang G et.al 20 & Zheng X et.al 21	4.9
Basil	leaf, flower top	estragole, limonene, fenchone, linalool, eugenol E-methyl cinnamate, 1,8-cineole	Inouye S et.al <sup>22</sup> ; Vina A, Murillo E <sup>23</sup> , & Chalchat JC, Ozcan MM <sup>24</sup>	4.9
Bay laurel	leaf	1,8-cineole, sabinene, α-terpinyl acetate, Linalool	Sangun MK et.al <sup>25</sup> & CosimiS et.al <sup>26</sup>	1.4
Bergamot	fresh or dried peel	limonene, linalyl acetate, B-pinene, y-terpinene, linalool	CosimiS et.al <sup>26</sup>	1.4
Camphor	wood, bark, leaf	1,8-cineole, $\alpha$ -terpineol, $\alpha$ -pinene, linalool, camphor, sabinene	Bakkali F et.al <sup>12</sup>	11.1
Cassia	leaf, bark, stalk	bark: E-cinnamaldehyde, methyl o-salicylate; leaf: 3-methoxy-1,2-propanediol, E-cinnamaldehyde, o-methoxycinnamaldehyde	Wang R et.al <sup>27</sup> & Du H, Li H <sup>28</sup>	2.1
Catnip, catmint	dry leaf, stem	nepetalactone, 1,8-cineole, $\alpha$ -humulene, $\alpha$ -pinene, E-geraniol, $\beta$ caryophyllene, citronellol	Inouye S et.al <sup>22</sup> ; Gilani AH et.al <sup>29</sup> & HeuskinS et.al <sup>30</sup>	4.9
Cedar	wood	thujopsene, eudesmol, E-(+)- $\alpha$ -atlantone; $\alpha,\beta \& \gamma$ -himachalenes; $\alpha$ - & $\beta$ -cedrenes; limonene, $\beta$ -phellandrene, $\alpha \& \beta$ -pinene, 3 carene; p-methyl- $\Delta$ -3-tetrahydro & p-methyl acetophenones; hinokitiol, carvacrol	Burfield T <sup>31</sup> .	9.7
Chamomile	seed, leaf, flower	Roman: isobutyl, isoamyl & 2-methylpentyl angelates, α-pinene German: E-β-farnesene, E,E-α farnesene,α-bisabolol, α-bisabolol oxides A & B	Inouye S et.al <sup>22</sup> ; HeuskinS et.al <sup>30</sup> & Lawrence BM <sup>32</sup>	2.1
Chrysanthemum	dry flower	verbenol, 2-(2,4-hexadiynylidene)-1,6-dioxaspiro[4,4]non-3-ene, 1,8-cineole, α-pinene, camphor, borneol, bornyl acetate	Wu LY et.al <sup>33</sup> & Zhu S <sup>34</sup>	2.8
Cinnamon	bark, leaf	eugenol, cinnamaldehyde	Wang R et.a <sup>27</sup> l; Fichi G et.al <sup>35</sup> & Singh G et.al <sup>36</sup>	12.5
Citronella	leaf	citronellal, geraniol, citronellol, geranylacetate	Nakahara K et.al <sup>37</sup> & Sethi ML et.al <sup>38</sup>	34.7
Clove	flower bud	eugenol, caryophyllene, eugenyl acetate	Guan W <sup>39</sup>	15.3
Coriander	fruit, seed	linalool, geraniol, geranyl acetate, 2-decenal, 3-dodecenal	Inouye S et.al <sup>22</sup> ; Msaada K et.al <sup>40</sup> & Zheljazkova VD et.al <sup>41</sup>	2.1
Cypress	needle, twig	sabinene, a-pinene, terpinen-4-ol, limonene	Tapondjou AL et.al <sup>42</sup> & Sacchetti OG et.al <sup>43</sup>	4.2
Dill	seed, leaf, stem	carvone, limonene, a-phellandrene, a-pinene, cis-dihydrocarvone	Bailer J et.al <sup>44</sup> & Callan NW et.al <sup>45</sup>	1.4
Eucalyptus	leaf	1.8-cineole, p-menthane-3.8-diol, α-pinene, p-cymene, γ-terpinene, eucamalol, allo-ocimene, citronellol, α-terpineol	Batish DR et.al <sup>10</sup>	30.6
Fennel	fruit	E-anethole, (+)-fenchone, α-phellandrene, (±)-limonene, estragole	CosimiS et.al <sup>26</sup>	4.9
Garlic	bulb	diallyl disulfide, diallyl trisulfide, methyl allyl trisulfide	Kimbaris AC et.al <sup>46</sup> & Shaath NA et.al <sup>47</sup>	6.3
Geranium	leaf, stem	2-phenylethanol, geraniol, citronellol, geranyl acetate	Seo SM et.al <sup>48</sup>	11.8
Ginger	rhizome	geranial, $\alpha$ -zingiberene, E,E- $\alpha$ -farnesene, neral, ar-curcumene, geraniol	Singh G et.al <sup>49</sup> & Sakamura F <sup>50</sup>	1.4
Grapefruit	peel	limonene, geranial, neral	ChutiaM et.al <sup>51</sup>	2.1
Guaiac wood	wood	bulnesol, guaiol, a-bulnesene	Bates RB et.al <sup>52</sup>	1.4
Hiba	wood	sabinene, 4-terpineol, thujopsene, hinokitiol, $\alpha$ -thujaplicine, carvacrol	YatagaiM et.al53; Burfield T54 & Nozoe T et.al55	2.1
Ho leaf	leaf	1,8-cineole, α-terpineol, linalool, camphor, safrole, sabinene, nerolidol	Bakkali F et.al <sup>12</sup> ; Zhang G et.al <sup>56</sup> & Nguyen XD et.al <sup>57</sup>	1.4
Hyssop	leaf, flower	sabinene, pinocamphene, isopinocamphene, isopinocamphone, pinocarvone, cis & trans-pinocamphones, β-pinene, 1,8-cineole, camphor, linalool	Kazazi H et.al <sup>58</sup> ; Langa E et.al <sup>59</sup> & Chalchat JC et.al <sup>60</sup>	1.4
Jasmine	flower	linalool, benzyl acetate, methyl & benzyl benzoates, methyl anthranilate, Z-jasmone, eugenol	Rath CC et.al <sup>61</sup> & Sandeep, Paarakh PM <sup>62</sup>	3.5
Juniper	fruit	a-pinene, myrcene, sabinene, germacrene D	Angioni A et.al <sup>63</sup>	1.4
Lady of the night	flower	phenylethyl alcohol, benzyl alcohol, eugenol	Al-Reza SM et.al <sup>64</sup>	2.1
Lavender	flower	linalool, linalyl acetate, lavandulyl acetate,α-terpineol, geranyl acetate, terpinen-4-ol, 1,8-cineole	Salehi FARP et.al65 & Daferera DJ et.al66	21.5
Lemon	peel	limonene, $\beta$ -pinene, $\gamma$ -terpinene	Flamini G et.al <sup>67</sup>	11.1
Lemon eucalyptus	leaf, twig	citronellal, citronellol	Batish DR et.al <sup>10</sup> & El-Zalabani SM et.al <sup>68</sup>	8.3
Lemongrass	leaf	geranial, neral, myrcene	Sacchetti OG et.al <sup>43</sup>	14.6
Lemon tea tree	leaf	neral, geranial, $\gamma$ -terpinene, geraniol, geranyl acetate, $\alpha$ -pinene, citronellal, terpinolene	Brophy JJ et.al <sup>69</sup>	1.4
Lime	peel	D-dihydrocarvone, D-limonene, $\alpha$ -terpineol	Patil JR et.al <sup>70</sup>	2.1
Manuka	leaf, stem	leptospermone, trans-calamenene, flavesone, 1,8-cineole, $\alpha$ -pinene	Douglas MH et.al <sup>71</sup> & Perry NB et.al <sup>72</sup>	1.4
Marjoram	leaf, flower	p-cymene, γ-terpinene, terpinen-4-ol, linalool, cis-sabinene hydrate	Vagi E et.al <sup>73</sup> & Vera RR et.al <sup>74</sup>	2.1
May chang/ Litsea	fruit	neral, R-(+)-limonene, geranial, citronellal	Seo SM et.al <sup>48</sup> ; Lalko J, Api AM <sup>75</sup> & Lawrence BM <sup>76</sup> .	4.9
Melaleuca/ Tea tree	leaf	terpinen-4-ol, γ-terpinene, α-terpinene, 1,8-cineole	Brophy JJ et.al <sup>77</sup>	9.7
Mint, mentha	leaf, flower	menthone, menthol, 1,8-cineole, 4-terpineol	Marengo E et.al <sup>78</sup>	9.7
Orange	Peel	limonene,myrcene	Tao NG et.al <sup>79</sup>	4.2
Palmarosa	dry leaf	geraniol, geranyl acetate, geranial	Raina VK et.al <sup>80</sup>	4.9
Parsley (Curl leaf)	leaf, stem, seed	$\beta$ -phellandrene,myristicin, $\alpha \& \beta$ -pinene, myrcene	Petropoulos SA et.al <sup>81</sup>	1.4
Patchouli	dry, fermented leaf	(-)-patchoulol, $\alpha$ -guaiene, seychellene, $\beta$ -caryophyllene, $\alpha \& \beta$ -patchoulenes, selinene, $\alpha$ -bulnesene, norpatchoulenol, pogostol	Nikiforov A et.al <sup>82</sup>	4.2
Pepper	Fruit	E-β-caryophyllene, limonene, β-pinene	Orav A et.al <sup>83</sup> & Kapoor IPS et.al <sup>84</sup>	2.1
Peppermint	Aerial part	isomenthol, p-menthone, isomenthyl & menthyl acetates	Kowalski R, Wawrzykowski J; <sup>85</sup> & Aziz EE, Craker LE <sup>86</sup>	16.7

Pine	twig bud	3-carene a & B-ninene a-cadinal Camphene	Judzentiene A Kuncinskiene E <sup>87</sup> & Ustun O et al <sup>88</sup>	83
Rose	Petal	2-nhondry d chi principal citronella certania linalon nonadecane	Lawrence BM <sup>32</sup> & Almasirad A et al <sup>89</sup>	6.3
Rose	Flower	2-printed yr ateolof, etforeior, geranof, manoo, nonaceane	Sacchetti OG et $a^{143}$ & Gillii VG et $a^{190}$	9.0
Roseniary	leaf flower	complex 18 citation of the second sec	Geneva MP et al <sup>91</sup> : Kamatou GPP et al <sup>92</sup> : Loizzo MP	9.0
Salvia (Sage)	aerial part	earning, rossinger and and an and a second a	et al $^{93}$ . Cardile V et al $^{94}$	1.4
Sandalwood	heartwood	a R & eni-R-santalane: a R & eni-R-santalal: ansantalal: spiro R E-R & eni-R santalals trans-a-hergemotene trans-a-hergemotel	Nikiforov A et $al^{82}$ & Jones CG et $al^{95}$	3.5
Sour (hitter) orange	Peel	a, per op-p-sanacies, a, per op-p-sanacies, a sanator, spito, p. 2-per op-p-sanation, aans-a-oerganioten, italis-a-oerganioten	Lawrence BM <sup>96</sup>	3.5
Sour (bitter) brange	Flower		Kiran II. Patra DD <sup>97</sup> Wnag Z at al <sup>98</sup> .	2.8
Tagatas	loof stelly flower	Catvone, initiate impanance a taminaal dibudratagatanana. Z tagatanana E $k$ Z tagatananaa	$D_{2}$ Nilal O, Falla DD, Wildg Z et.al ,	2.0
Tagetes	leal, stark, nower	E & Z-p-ocimenes, milorene, u-terpineor, umyarotagetenone, Z-tagetone, E & Z-tagetenones		2.1
Tarragon	lear, nower	sabinene, elemicine, metnyl eugenol	Khodakov GV et.al	1.4
Thyme	flower, leaf	p-cymene, geraniol, cis-thujone, thymol, carvacrol	Sacchetti OG et.al <sup>45</sup>	7.6
Turmeric	rhizome	$\alpha$ -phellandrene, 1,8-cineole, terpinolene, zingiberene, $\beta$ -sesquiphellandrene, $\alpha \& \beta$ -turmerones	Sacchetti OG et.al <sup>43</sup>	2.8
Verbena	leaf	geranial, neral, limonene, geraniol	Argyropoulou C et.al <sup>102</sup>	1.4
Vetiver	root	khusimol, isonootkatool, $\beta$ -vetivenene, $\alpha \& \beta$ -vetivones	Champagnat P et.al <sup>103</sup>	2.1
Violet	leaf, flower	nona-2,6-dienal, cis-hex-3-en-1-ol, hexadec-1-ene, pentadec-3-enal, octadec-1-ene, hexadecanoic & octadeca-9,12-dienoic acids	Cu JQ et.al <sup>104</sup>	1.4
Wintergreen	leaf	methyl salicylate	Tisserand R <sup>105</sup>	6.3
Ylang-Ylang	flower	linalool, benzyl acetate, benzyl benzoate, benzyl salicylate	Sacchetti OG et.al43	2.8
	•	Glyceridic Oils	·	
Castor	seed	ricinolic, linoleic, oleic acids	Martin C et.al <sup>106</sup>	4.9
Evening primrose	seed	linalool; palmitic, stearic, oleic, linoleic & γ-linolenic acids	Bicchi C et.al <sup>107</sup> & Christie WW <sup>108</sup>	1.4
Ligusticum/ Chuanxiong	root	butylphthalide, 2-propenyl phenoxyacetate, 3-isobutyliden phthalide, palmitic & octadecenoic acids	Jeong JB et.al <sup>109</sup>	1.4
Mustard	seed	erucic, oleic, linoleic, linolenic, palmitic & stearic acids	Nabloussi A et.al <sup>110</sup>	2.1
Neem	seed	salanin, nimbinin, nimbidiol, azadirachtin; palmitic, stearic, oleic, linoleic acids	Martín C et.al <sup>106</sup> ; Sidhu OP et.al <sup>111</sup>	8.3
Olive	fruit	oleic, palmitic, linoleic & stearic acids	PatumiM et.al <sup>112</sup>	2.1
Perilla	seed	oleic, linoleic & linolenic acids, S-limonene, perillaldehyde, elsholzia ketone, perillaketone, myristicin, dillapiol, elemicin, isoegomaketone,	Nitta M et.al <sup>113</sup> ; Longvah T et.al <sup>114</sup> ; Koezuka Y	1.4
		perillene, egomaketone, shisofuran	et.al <sup>115</sup>	1.4
Sesame	seed	linoleic, oleic, palmitic & stearic acids	Lee DS et.al <sup>116</sup>	3.5
Soybean	seed	linoleic, oleic, palmitic, α-linolenic & stearic acids	Ribeiro APB et.al <sup>117</sup>	4.2

#### Table 2. Studies related plant derived oils with their respective mosquito repellency properties

Plant oil	Mosquito repellent properties	Source
Andiroba	patented candles commercialized in Brazil as mosquito fumigant-repellents; repellency in Amazonfield test of 1:1:1 Andiroba, Copaiba and Baby oil mixture	Gilbert B et.al <sup>126</sup> & Ribas J, Carreno AM <sup>127</sup>
Anise	good repellent against Cx. pipiens	Erler F. et.al <sup>128</sup>
Artemisia	repels mosquitoes	Lin X. <sup>129</sup>
Basil	Ae. aegypti (PP 2 h, R% 81), An. stephensi (PP 3.5 h, R% 67), Cx. quinquefasciatus (PP 8 h, R% 100)*;5% hairy basil mixed with 5% vetiver and 10% citronella EOs in nanoemulsion repel Ae. aegypti 4.7 h; repels Anopheles sp. in human bait test and Cx. pipiens	Amer A, Mehlhorn H <sup>130</sup> ; & Onanong N et.al <sup>131</sup>
Bay laurel	spatial repellence against Ae. aegypti, acceptable smell, definite mosquito repellence	Drapeau J et.al <sup>132</sup>
Bergamot	repellence against Culex & Aedes spp. is comparable to citronella oil	Rudolfs W. 133
Camphor	Ae. aegypti (PP 2.5 h, R% 32), An. stephensi (PP 8 h, R% 43), Cx. quinquefasciatus (PP 8 h, R% 57)*; EO (0.1 mg· cm-2 on human skin): repellency and duration against Ae. aegypti comparable to DEET	Amer A, Mehlhorn H <sup>130</sup> & Yang YC et.al <sup>134</sup>
Cassia	Bark extract (0.1 mg· cm-2): repels Ae. aegypti comparably to DEET (on human skin), 5% EO in cream provided 50 min of protection to humans against female Ae. Aegypti	Yang Y C et.al <sup>134</sup>
Catnip, catmint	Ae. aegypti (PP 8 h, R% 84), An. stephensi (PP 8 h, R% 100), Cx. quinquefasciatus (PP 8 h, R% 100)*;1–6 h protection against Ae. albopictus (23 & 468 µg· cm–2); safety pharmacology evaluation: N. cataria oil is safe compared to DEET, p-menthane-3,8-diol, etc. May cause minor skin irritation	Amer A, Mehlhorn H <sup>130</sup> ; Zhu J et.al <sup>135</sup> ; & Zhu JJ et.al <sup>136</sup>
Cedar	Ae. aegypti (PP 3 h, R% 38), An. stephensi (PP 8 h, R% 38), Cx. quinquefasciatus (PP 8 h, R% 100)*; repels An. stephensi	Lin X <sup>129</sup> ;Amer A, Mehlhorn H <sup>130</sup> ; & Curtis CF et.al <sup>137</sup>
Chamomile	Ae. aegypti (PP 4 h, R% 65), An. stephensi (PP 8 h, R% 76), Cx. quinquefasciatus (PP 8 h, R% 100)*	Amer A, Mehlhorn H <sup>130</sup>
Cinnamon	high repellency (RD95 mg·mat-1) against An. stephensi (49.6), Ae. aegypti (53.9), Cx. quinquefasciatus (44.2)	Prajapati V. et.al <sup>138</sup>
Citronella	repellency by 1:1 citronella & lavender EOs in 2 oz. castor oil; Ae. aegypti (PP 2 h, R% 76), An. Stephensi (PP 8 h, R% 52), Cx. quinquefasciatus (PP 8 h, R% 100)*; C. winterianus + vanillin repels Ae. aegypti, An. dirus, Cx. quinquefasciatus 8 h; + eucalyptus oil in floor cleaner, 6 h repellency; comparable to DEET & N,N-diethyl mandelic acid amide vs. Aedes spp.; 5% hairy basil + 5% vetiver + 10% citronella Eos in nanoemulsion with 4.7 h protection against Ae. aegypti; C. winterianus LC50 & LC95 = 0.5 & 0.9% for Cx. quinquefasciatus, 1.0 & 2.0% for Ae. aegypti	Guan W et.al <sup>39</sup> ; Amer A, Mehlhorn H <sup>130</sup> ; Onanong N et.al <sup>131</sup> ; Gibson A <sup>139</sup> ; Tawatsin A et.al <sup>140</sup> ; Bindra RL et.al <sup>141</sup> ; Thorsell W et.al <sup>142</sup> ; MakhaikM et.al <sup>143</sup>
Clove	Cx. pipiens pallens: isoeugenol & eugenol (from clove oil) > repellency than citronella. Clove bud oil + vanillin (long-term protection) > DEET; repels Ae. aegypti, An. dims, Cx. quinquefasciatus for 2–4 h; knockdown LC50 & LC95=0.5 & 0.9% for Cx. quinquefasciatus, 1.0 & 2.0% for Ae. Aegypti	Seo SM et.al <sup>48</sup> ; Angioni A et.al <sup>63</sup> ; MakhaikM et.al <sup>143</sup>
Copaiba	1:1:1 andiroba, copaiba & baby oils repels mosquitoes in Amazon field test	Ribas J, Carreno AM <sup>127</sup>
Dill	Ae. aegypti (PP 1.5 h, R% 78), An. stephensi (PP 3.5 h, R% 71), Cx. quinquefasciatus (PP 3 h, R% 57)*	Yang P. et al. <sup>144</sup> .
Eucalyptus	Ae. aegypti (PP 1 h, R% 57), An. stephensi (PP 5.5 h, R% 29), Cx. quinquefasciatus (PP 8 h, R% 100)*	Moore SJ et al. <sup>145</sup>
Fennel	5% in aerosol or 8% in cream repels Ae. aegypti comparably to citronella & geranium EOs; cream & EO repel Culex, Anopheles & Aedes spp. comparably to geranium & citronella EOs in field; hexane fraction (0.1 mg· cm-2 repels Ae. aegypti 99%) of fruit methanol extract contains repellent (+)-fenchone & E-9-octadecenoic acid	Kim S-I et.al <sup>146</sup> ; & Kim DH et.al <sup>147</sup>
Galbanum	Ae. aegypti (PP 2.5 h, R% 70), An. stephensi (PP 8 h, R% 100), Cx. quinquefasciatus (PP 8 h, R% 100)*	Amer A, Mehlhorn H <sup>130</sup>

Garlic	EO and chemical components have strong repellent properties	BhuyanM et.al <sup>148</sup>	
	Ae. aegypti (PP 2.5 h, R% 78), An. stephensi (PP 8 h, R% 62), Cx. quinquefasciatus (PP 8 h, R% 100)*; geranium added with lemongrass essential oils repel Aedes spp.; geranium added with		
	sandalwood EOs added with soybean oil in burned stick repellents; Geranium (25%geraniol) EO added with lemongrass extract product protect against bites of Ae. atlanticus and Ae.mitchellae for 4	G G M ( 1 <sup>48</sup> A A M I II II <sup>130</sup> C'I A <sup>139</sup> K' G I	
Geranium	hr; geranium added with sandalwood essential oils with soybean oil product repel Culex sp. & other mosquitoes for 3 hr in the field; geranium added with citronella essential oils(1: 1) in cream	Seo S M et.al ; Amer A, Meninorni :, Gibson A ; Kim S I	
	product repels Ae Aegypti and Culex Anopheles & Aedes spp in the field: in cold creams with repellency to Cx fatigans in lab &	et.al <sup>110</sup> ; Qualls WA, Xue R D; <sup>110</sup> Osmani Z et.al <sup>110</sup>	
	on humans: various forms of EO definite promise as repellents		
Ginko	Edular (isolated from leaf extract) renets Ae albonicitus	Matsumoto A et al <sup>151</sup>	
Glinko	Lucatan (Isolated from Fel Fedrav) (Nepolation) and a solution of the analysis of the solution	Musumoto A. et.ul	
Hibawood	For board	Kobayashi A, Mizutani T et.al <sup>152</sup>	
Icomino	Not commute (DD 4.5 h. D9/ 14) An stanhami (DD 9 h. D9/ 100) Cy. suinguefaceistic (DD 9 h. D9/ 100)*	Amer A. Mahlhorn H <sup>130</sup>	
Jasmine	Av. $acgypti (1 + z, t; K/t) (4)$ , At. stephenist (1 + t; K/t) (00), CX. (unique fascinus (1 + t); K/t) (00) Av. $acgypti (1 + z, t; K/t) (1 + z)$ , At. stephenist (1 + t); K/t) (00), CX. (unique fascinus (1 + t); K/t) (10))	Aller A, Mellilolii II	
Juniper	Ac. $deg(p)$ ( $r = 5.5$ ), $K/6$ ( $45$ ), Al. stephenis ( $r = 6.1$ , $K/6$ /0), A. (unique)asciatus ( $r = 6.1$ , $K/6$ 100) <sup>2</sup> , and results ( $r = 6.1$ , $r =$	Amer A, Mehlhorn H <sup>130</sup> ; & Kang S H <sup>153</sup>	
Lavandar	good repertency at $y_{12}$ cm <sup>2</sup> against remare CX. pipens parents	Lin V <sup>129</sup> & Amer A Mahlhorn H <sup>130</sup>	
Lavenuei	Ac acception ( $D1 \le b$ , $V/0.24$ ), All, stephenis ( $T \le 0$ , $V/0.05$ ), $V/0.01$ (unique facciatus ( $T \le 0$ , $V/0.00^{\circ}$ ), insignito rependint curvinent 1, lavenuet 1 & castor on 2 oz.	Lili A. & Alilei A, Melililofii H	
Lemon	Ac. $dgypti (r - 1.5)t, K/0.00)$ , Al. stephenis (r - 7)t, K/0.10), C. (unique lasciaus (r - 8)t, K/0.100), regularized to the stephenic Defect is a nine of the stephenic defect in a nine of the stephenic defect in a nine of the stephenic defect is a nine of the stephenic defect of the stephenic defect in a nine of the stephenic defect of t	Amer A, Mehlhorn H <sup>130</sup> & Oshaghi MAet.al <sup>154</sup>	
	rependency to Aut, stephenis ~ DEET in annual or nonnan costs	America Mahlham H <sup>130</sup> , Ovalla WA, Yua D D <sup>149</sup> , & Mana SI	
Lemongrass	Ac adgptu ( $r = 5$ in, $K^{*}_{0}$ ( $00$ ), An stepletist ( $r = 6$ in, $K^{*}_{0}$ 100), C. dunductastatus ( $r = 6$ in, $K^{*}_{0}$ 100), the desired 25% genation on a demonstrate frame language to the administrate administrat	Allel A, Mellioni H , Qualis WA, Aue K D , & Moore SJ	
T	and Ac intercented, institute containing tentoring ass EO + p-interlinated of (FMD) tepets Air. darling a other spp. 95–96% for 5–6 in the field (rependency > 15–20% DEE1)	CL.al	
Leptospermum	repenent: blocks ability official of perceive CO2 emitted by numans	Maguranyi SK et al , Molyneux wim	
Marjoram	S µg·cm-2 good repellent against female Cx. pipens pallens	Oshaghi MA et. al	
May chang/	Ae. aegypti (PP 8 h, R% 73), An. stephensi (PP 8 h, R% 100), Cx. quinquetasciatus (PP 8 h, R% 100)*; high contact and noncontact repellency to female Ae. aegypti in vitro in cages; tormulation to	Lalko J. Api AM <sup>75</sup> : Amer A. Mehlhorn H <sup>130</sup> : Yang Y C et.al <sup>134</sup> :	
Litsea	fix onskin with high repellency in human volunteers for up to 8 h & 100% repellency to An. stephensi, Cx. Quinquefasciatus, Ae. aegypti; greater repellence of night-biting mosquitoes An. dirus, Cx.	Tawatsin A et.al <sup>158</sup>	
	quinquetasciatus and Ae. albopictus than Ae. Aegypti		
Melaleuca	Limited repellence of Ae. aegypti, Cx. quinquefasciatus, Cx. annulirostris in lab	Brophy JJ et.al	
Mint, mentha	d-8-acetoxycarvotanacetone isolated mosquito repellent	Ding D, Sun H et.al <sup>159</sup>	
Myrtle	Ae. aegypti (PP 2.5 h, R% 57), An. stephensi (PP 6.5 h, R% 43), Cx. quinquefasciatus (PP 8 h, R% 86)*	Amer A, MehlhornH <sup>130</sup> .	
Palmarosa	High geraniol content mosquito repellent	Dubey VS <sup>160</sup>	
Patchouli	Partially repels Ae. aegypti, Cx. quinquefasciatus, An. dims	Trongtokit Y et.al <sup>161</sup>	
Pepper, black	Ae. aegypti (PP 1.5 h, R% 65), An. stephensi (PP 3 h, R% 62), Cx. quinquefasciatus (PP 8 h, R% 100)*;repels An. dirus, Cx. quinquefasciatus, Ae. albopictus (≥ 4.5 h)	Amer A, Mehlhorn H <sup>130</sup> & Tawatsin A et.al <sup>158</sup>	
n :.	Ac. acgypti (PP 2 h, R% 59), An. stephensi (PP 6.5 h, R% 57), Cx. quinquefasciatus (PP 8 h, R% 100)* on human skin strongly repels An. annularis (100%), An. culicifacies (92%), Cx.		
Peppermint	quinquefasciatus (85%); comparable to mylol oil (di-butyl & di-methyl phthalates); repels adult female Cx. pipiens	Erler F et.al <sup>10</sup> ; Amer A, Mehlhorn H <sup>10</sup> ; & Ansari MA et.al <sup>10</sup>	
Pine	200d mosquito repellency	Judzentiene A. Kupcinskiene E <sup>87</sup>	
Rose	moderate mosquito renellency	Lin X <sup>129</sup>	
	Ac acounti (PP 5.5 h R% 43) An stenhensi (PP 8.h R% 100) Cx quinquefasciatus (PP 8.h R% 100)* renels An stenhensi Ac Acounti&Cx Quinquefasciatus low renellency against Ac acounti	Gillii YG et al <sup>90</sup> . Amer A. Mehlhorn H <sup>130</sup> . Draneau Let al <sup>132</sup> .	
Rosemary	in the acception of the second s	Prajanati V et al <sup>138</sup>	
Sage	An accurate to initiate a main regime in potentiation of the depth (in potentiation)	Amer A Mehlhorn H <sup>130</sup>	
Sandalwood	As accurate $(P 2 \le h \le 10^{-5})$ , the dependence $(P 2 \le h \le 10^{-5})$ , the demonstration $(P 2 \le h \le 10^{-5})$ and $(P 2 \le h \le 10^{-5})$ .	Amer A Mehlhorn H <sup>130</sup>	
Sour (bitter)	Ne. usjpa (11 2.5 n, R/s 57), full stephens (11 5 n, R/s 100), CX quinquensentus (11 6 n, R/s 100)		
orange	limited protection (repellency) against mosquitoes	Gibson A <sup>139</sup> .	
Spearmint	renellent: nineritane avide from M. sniceta (var. viridis) FO renels An. stenhensi	Trinathi AK et al <sup>163</sup>	
Spearmint	Ac accurate (DD 1 h 00 k0 A) An a transport (DD 2 h 00 (100) Cr curate accurate (DD 2 h 00 (100)*)	Thpathi AK tiai	
Tagetes	Ac. $degphi$ ( $r = 1$ , $\kappa/\delta = \delta + \beta$ , All, stephens) ( $r = \delta + \eta$ , $\kappa/\delta = 100$ ), C. dunique lastialus ( $r = \delta + \eta$ , $\kappa/\delta = 100$ ), $r = 100$ ,	Gillij YG et.al <sup>90</sup> & Amer A, Mehlhorn H <sup>130</sup>	
	repers Ac. acgyput (avg 70 mm) renada lab razard adulf fanala Ac. albaniatus for 2 b: 0.010/ tatally ranala Cr. Ouinguefaceintus		
Thyme	repers rao-reared aduit remain Ac. anoppetus for 2 n, 0.01% totany repers CX. Quinquerascialus	Zhu JJ et.al <sup>136</sup> & Pavela R. <sup>164</sup>	
	Tomorie added with 60% willing much Age and the disconding interview in and 60 per some for 0 her however, since added with some in a data with an illing added with some it.		
Turmeric	turmenc added with 5/wannin repers Ac adgypt, An artis and Cx, duriduelascialus in cage & large room for 8 nr, turmenc, citronena and nairy basil essential ons added with vannin provide substitute for DEET, repeale DEET & D2352 reprint the accounting of a for the former of the substitute of of the s	Tawatsin A et.al (2001) <sup>140</sup> & Tawatsin A et.al (2006) <sup>164</sup>	
Varbana	substitute for DEE1; repeis DEE1 & IN3333-resistant Ac. acgypti for 4.3 if	Amer A Mahlham II <sup>30</sup>	
Verbena	Ae. aegypti (PP 2.5 n, K% /0), An. stepnensi (PP 5.5 n, K% 38), (X. quinquetasciatus (PP 8 n, K% 100)*	Amer A, Meninornh	
venver	station nanocimuision oi 3% inany basili, 3% vertifer & 10% citronelia EU: repeis Ae. acgypti for 4./ h	Sacchetti UG et.al	
Violet	Ae. aegypti (PP 6 h, K% 68), An. stephensi (PP 8 h, K% 100), Cx. quinquetasciatus (PP 8 h, K% 86)*	Amer A, Mehlhorn H	
Wild verbena	sources of perillaldehyde (repels An. gambiae) & perillic acid (repels An. arabiensis & Ae. aegypti)	Omolo MO et.al <sup>165</sup> & Dorfling EA, Mouton I. <sup>166</sup>	
(Lippie)		-130	
Wintergreen	strongly repels Culex and Aedes spp. in different tests	Amer A, Mehlhorn H <sup>130</sup>	
Wormwood	mosquito repellent properties	Tripathi AK et.al <sup>10</sup>	
Zanthoxylum	repel mosquitoes	Trongtokit Y et.al	
Glyceridic oils			
Castor	best carrier for pyrethrum extracts (long-lasting)	Gibson A <sup>139</sup> .	
Mustard	longer protection (up to 5 h with Zanthoxylum limonella or lime oils) than coconut (Cocus nocifera) oilagainst Ae. albopictus	Das NG et.al <sup>168</sup>	
	knockdown repellency against Ae. aegypti, Ae. albopictus, An. quadrimaculatus Say; repels femaleAn. stephensi (ED50 0.191–0.156 mg· cm-2) in lab; low repellency against Ae. albopictus & Cx.	Xue R D et. al <sup>169</sup> ; Vatandoost H, HanafiBojd AA <sup>170</sup> ; Barnard DR,	
Neem/	nigripalpus; 2% in coconut (Cocus nocifera) oil on exposed body parts of human volunteers provided complete protection for 12 hr from all Anopheles spp.; protection from Anopheles spp. (96-	Xue R D <sup>171</sup> ; Sharma VP et.al <sup>172</sup> ; Sharma SK et.al <sup>173</sup> ; Dua VK	
Margosa	100%), Aedes (85%), Culex sp. (61-94%); significant protection by neem cream against adult Ae. aegypti; 1% in kerosene lamps in preclinical & clinical safety evaluation is safe to humans; 1% in	et.al <sup>174</sup> ; Valecha N et.al <sup>175</sup> ; Sharma VP, Ansari MA <sup>176</sup> ; & Mishra AK	
ũ	kerosene burned in lamps effective in 2 field tests. Repellence: Anopheles > Culex; 1-4% in coconut oil on exposed body parts of humans: 81-91% protection for 12 hr	et.al <sup>177</sup>	
Olive	Ac. acgypti (PP 3.5 h, R% 68). An, stephensi (PP 8 h, R% 71). Cx. quinquefasciatus (PP 8 h, R% 71)* 1:1 w/nvrethrum renels mosquitoes for 4 h	Amer A. Mehlhorn H <sup>130</sup> & Gibson A <sup>139</sup>	
	Ac. acyvti (PP 3 h, R% 54), An. stephensi (PP 8 h, R% 76), Cx. quinquefasciatus (PP 8 h, R% 100)*; oil-based product provided 1.5 h (low) repellency. 24%DEET provided 5 hr protection: good	Amer A. Mehlhorn H <sup>130</sup> : Barnard DR. Xue R D <sup>171</sup> : Fradin MS Dav	
Sovbean	renellencyin product w/sandalwood added with geranium in burned sticks vs. DEET: commercial product containing 2% oil renels Ae. Albonicus, Cx. nigrinalous Ae. triceriatus for 5, 8,5, 8,> 73 h	JF <sup>178</sup> . Ritchie SA et al <sup>179</sup>	
	rementively	. ,	

Another group of oils in this study actively repelled only Cx. quinquefasciatus: cedar (Cedrus, Cupressus and Juniperus spp.), Citronella (Cymbopogon nardus, C. winterianus), Eucalyptus (Eucalyptus globulus), broad and narrow-leaved eucalyptus (E. dives and E. radiata, respectively), Geranium (Pelargonium graveolens), Juniper (Juniperus communis), Lemon (Citrus limon), Lemon eucalyptus (Eucalyptus citriodora), Myrtle (Myrtus communis), Peppermint (Mentha piperita), sage (Salvia sclarea), Thyme (Thymus serpyllum), Verbena (Lippia triphylla) and wild soybean (Glycine soja) [Amer and Mehlhorn]<sup>130</sup>. Differences in the species specificity of the repellence profiles of these and other EOs may explain their use in repellent mosquito products. This may have to do with differences in the local and regional profiles of mosquito species populations and explain the use of mixtures of these essential oils to generate broad spectrum formulations for simultaneous repellency of multiple mosquito species. While Ae.Aegypti and other adults may in general be difficult to repel using plant-based products or synthetic repellents, a number of plant essential oils have been identified which are effective against this species (Table 2). The following repellency effects of essential oils against adult Ae. aegypti have been observed: hairy basil (Ocimum basilicum) in stable nanoemulsions with vetiver (Vetiveria zizanioides) and citronella (Cymbopogon nardus and Winterianus) essential oils are a good repellent [Onanong]<sup>131</sup>, Bay laurel (Laurusnobilis) EO is an acceptable smelling, good spatial repellent [Drapeau J], Camphor (Cinnamomum camphora) EO and Cassia (C. cassia) bark extract on human skin are repellents comparable to DEET [Gibson A]<sup>139</sup>, Catmint (Nepetacataria) EO exhibited 8 hr of protection on humanskin, cinnamon (Cinnamomum zeylanicum, C. verum) EOs exhibit moderate to good repellency [Amer and Mehlhorn]<sup>130</sup>, Citronella (C. winterianus) EO exhibits knockdown repellency at 1–2% [Makhaik et al.]<sup>143</sup> and C. winterianus EO + vanillin exhibits 8 h of repellency [Tawatsin et al.]<sup>140</sup>, Fennel (Foeniculumvulgare) fruit extract/fractions offer complete repellency [Kim *et al.*]<sup>147</sup> and EO as an aerosol or cream has comparable repellency to other EO repellents [Kim *et al.*]<sup>146</sup>. Geranium (Pelargonium graveolens) added with citronella EO in a cream product exhibited good repellency in the field [Seo SM et al.]<sup>48</sup>, lemon eucalyptus (E. citriodora) has knockdown repellency/ adulticide activity [Seo et al.]48, may chang/litsea (Litseacubeba) EO exhibits contact and noncontact repellency and is a good/excellent repellent in formulations on the human forearm providing protection over 8 hr [Lalko et al., H.]<sup>75,130</sup>, Turmeric (Curcuma longa) EO added with 5% vanillin exhibits 8 hr of repellency and in formulations with other EOs added with 5% vanillin offers protection from DEET and IR3535- resistant strains of Ae. aegypti over 4.5 hr [Tawatsin A *et al.*, Tawatsin A. *et al.*]<sup>140,158</sup> and violet (Viola odorata) EO on human skin exhibited 8 hr protection at a good level of repellency [Amer and Mehlhorn]<sup>130</sup> (Table 2). Several essential oils from Zanthoxylum L. spp. (Z. piperitum DC., Z. armatum DC., Z. bungei Planch. and Linden ex Hance) were cited in patented mosquito repellent inventions. According to recent literature, Zanthoxylum L. spp. essential oils have mosquito repellent activity [Trongtokit Y et al., Choochote W et al.] 161,182. Also, besides A. argyi H. Lev. and Vaniot (artemisia) EO, which is known to repel mosquitoes [Lin  $X_{..}^{129}$ , the essential oils of several other Artemisia L. spp. were used in patented formulations such as A. annua L. (wormwood), A. vulgaris L. and A. apiacea Hance. This is interesting given that A. annua EO has proven insect repellent properties [Tripathi et al.].<sup>167</sup> The Amazon region is a source of plant-derived mosquito repellent oils. For example, Carapa guianensis Aubl. (andiroba) pressed fruit oils or extracts are formulated preferentially into candles during manufacturing as fumigant mosquito repellents [Gilbert B.et.al], burned in kerosene lamps or used in topical repellent formulations which are commercially available in Brazil. Copaifera L. spp. (copaiba) balsam oils or extracts are used in mosquito repellency in Brazil especially in the Amazon region (Table 2). Furthermore, a formulation of andiroba, copaiba and baby oils exhibited repellency to mosquitoes in an Amazon field study [Ribas *et al.*]<sup>127</sup>.

Based on their Mosquito Repellency scientific basis for the use of Glyceridic Oils:

Neem or margarosa oil is obtained by pressing the fruit of the neem tree (Azadirachta indica). Neem oil is burned in 1% compositions in kerosene lamps as indoor mosquito emitters of chemical repellent-fumigant deterrents which have been evaluated and are considered to be safe [Valecha N et al.;Sharma and Ansari].<sup>175,176</sup> Also, neem oil in mixtures with coconut (Cocos nucifera L.) oil provides good protection forvery long periods against Anopheles and Aedes spp. [Sharma *et al.*; Sharma *et al.*; Mishra *et al.*].<sup>172,173,177</sup> Castor (Ricinuscommunis L.), Mustard (Brassica spp.), Olive (Olea europaea) and other glyceridic oils have important roles in several patented repellent compositions containing pyrethrum extracts and essential oils where they act as carriers and can extend the duration of repellent effects for several hours perhaps by slowing the release or evaporation of essential oils from surfaces. There are mixed scientific reports on the effective mosquito repellency of several glyceridic oils. Thus, for soybean (Glycine max) oil low repellency was observed as compared to 24% DEET formulations [Fradin and Day]<sup>178</sup> and good repellency was observed for the smoke generated from burned sticks which contained soybean oil. Especially interesting is a report from a United States Department of Agriculture laboratory where 4 well-known synthetic mosquito repellents based on 10% KBR3023 [1-piperidinecarboxylic acid 2-(2-hydroxyethyl)-1-methylpropyl ester], 7.5% IR3535 [3-(N-butyl-N-acetyl)-aminopropionic acid ethyl ester], 15% and 7% DEET and 8 natural product-based repellents based on 2% soybean oil, 10% citronella (Cymbopogon spp.) EO, neem oil (Azadirachta indica) and others were tested in the lab against Ae. Albopictus Skuse, Cx. nigripalpus Theobald and Ae. triseriatus Say [Barnard D R, Xue R D].<sup>171</sup> The 2% soybean oil formulation exhibited mosquito repellency comparable to both 10% KBR3023 and 15% DEET based products each of which provided estimated mean protection time (eMPT) responses averaged over all three mosquito species of  $\geq 7.2$  h [Barnard D R, Xue R D]<sup>171</sup>. This study is evidence for the potential of soybean oil as a stand-alone repellent and as a component oil of repellent formulations.

Chemical Composition of Essential oils used in Patented **Inventions:** Information on the major chemical components of plant essential oils used in mosquito repellent inventions is presented in Table 1. Among these are essential oils which are concentrated sources of proven mosquito repellent monoterpenes and phenylpropanoids such as the essential oils of angelica (Angelica archangelica), Artemisia (Artemisia argvi), Basil (Ocimum basilicum), Bergamot (Citrus bergamia), Camphor (Cinnamomum camphora), Cassia (Cinnamomum cassia), Catnip (Nepeta cataria), Chrvsanthemum (Chrysanthemum indicum), Cinnamon

(Cinnamomum zeylanicum), Citronella (Cymbopogon nardus, C. winterianus), Coriander (Coriandrum sativum), cypress (Cupressus sempervirens), Dill (Anethum graveolens), (Eucalyptus spp.), Geranium (Pelargonium Eucalyptus graveolens), Grapefruit (Citrus reticulata), Ho leaf (Cinnamomum camphora), Hyssop (Hyssopus officinalis), Juniper (Juniperus communis), Lavender (Lavandula angustifolia), Lemon (Citrus limon), Lemon eucalyptus (Eucalyptus citriodora), Lemongrass (Cymbopogon citratus), Lemon tea tree (Leptospermum petersonii), Lime (Citrus aurantifolia), Marjoram (Origanum majorana), Mav chang/litsea (Litsea cubeba), Melaleuca/tea tree (Melaleuca alternifolia), Mint/mentha (Mentha), Orange (Citrus sinensis), Palmarosa (Cymbopogon martini), Curl leaf parsley (Petroselinum crispum), Pepper (Piper nigrum), Pine (Pinus sylvestris), Rose (Rosa damascena, R. Centifolia), Rosemary (Rosmarinus officinalis), salvia/sage (Salvia spp.), Sour (bitter) Orange (Citrus aurantium), spearmint (Mentha spicata), thyme (Thymus vulgaris), verbena(Lippia triphylla) and wintergreen (Gaultheria procumbens). In these essential oils, proven mosquito repellent volatile components camphor, 1,8-cineole, citronellol, eugenol, geranial, geraniol, limonene, linalool, myrcene,  $\alpha$  and  $\beta$ -pinenes,  $\gamma$ -terpinene, terpinen- 4-ol and  $\alpha$ -terpineol are well represented among the major components.<sup>180-184</sup>

Synergism versus Suppression/Dilution of Synthetic essential oils:

In some cases, patented formulations in effect improve upon the natural repellency of an essential oil by mimicking or synthesizing an oil which ideally contains components which together contribute to the repellent effect and eliminates those which counteract (attract mosquitoes), make no contribution or suppress the repellency of other components. Work done by Odalo et al.<sup>185</sup> nicely illustrates this process. An initial observation was that major components of 6 essential oils when tested singlywere less repellent than the natural essential oils in which these components are found. Synthetic essential oils were prepared by mixing pure major components in the same proportion in which they occur in the natural essential oils. Repellencies of synthetic essential oils ranged from comparable to up to three times the activity of the corresponding natural essential oils against An. gambiae in the human-bait test based on RC75 values [Odalo et al.].<sup>185</sup> The activity of synthetic essential oils substantiates the additive and/or synergistic nature of the interaction of blended EO components (and also suppressive or diluting/repellency diminishing effects of nonactive components). The same principles of addition/ synergism and suppression/dilution are operational in the process of formulating Essential oils and isolated components into patented repellent inventions.

#### Conclusion

The identification of these potential repellent plant essential oils and volatile components from different literature reviews and patents will stimulate local efforts to enhance public health and make use of scientifically substantiated claims to mosquito repellency based on lab and field experiences. The studies from the different sources revealed that mosquito repellents and other insect control products must be evaluated in the environments and settings in which they are to be used. Repellents must be developed based on plant oils and isolated chemical components by targeting Anopheles, Aedes, Culex and other mosquito species which are found locally and regionally. The approaches for the repellency should be multipronged and use of sticks, candles, fast cards, sprays, paints and varnishes, etc. should be encouraged for individual protection. The formulations will prove to be much effective, devoid of side effects, ecological and economical, if made from the reviewed and supportive mosquito repellent essential oils.

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