

## **The potential use of natural essential oils in the fumigation of stored agricultural products – (review)**

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### **Abstract**

The authors give an overview of the concentrations of essential oils to control insect pests of stored grain, analyze the current prices of essential oils on the market and the cost of the fumigation, and discuss the potential of the introduction and the use of essential oil to fumigate stored grain. As with other groups of insecticides, the potential use of the natural essential oils (EO) in stored grain insect pest management depends on many barriers. Some of the barriers that may greatly prevent the adoption and use of the natural EO in stored grain fumigation are their relatively high concentrations needed for the effective protection of stored grain, a great difference in the sensitivity of various insect species, significant effect of different quantity of grain on the effectiveness and the current prices of natural essential oils on the market. Very high prices of essential oils, considering other characteristics (scent, sorption, penetration, aeration, etc.), may be really a very serious limiting factor for the application of natural essential oils in practice. There are two possible solutions to overcome the mentioned limiting factor; significant reduction of the prices of natural EO, or the production of the active components of natural EO synthetically.

**Keyword:** essential oils, fumigation, cost price, stored agricultural products

### **Introduction**

During the past few decades application of synthetic pesticides to control agricultural pests has been a standard practice. However, with the growing evidence regarding detrimental effects of many of the conventional pesticides on health and environment, require for safer means of pest management has become very crucial<sup>[1]</sup>. Despite of the numerous and ongoing research that have been conducted with new grain protectants, synthetic and natural ones, only a few have been adopted to be use as grain protectants. Daghli (2006)<sup>[2]</sup> discussed the barriers under biological, technical, legal and commercial categories why the adoption of new grain protectants is not widespread.

At the beginning of the new millennium, only two fumigants were in wider use in the world; fosfine and methyl bromide. Methyl bromide was already phased out, although the critical uses still allow for some consumption awaiting alternatives, with an exception for quarantine and pre-shipment treatment. There are various reasons for the disappearance of dozen of fumigants. First of all there were health reasons (suspected or alleged carcinogens), no food registration, flammability, lack of interest, strict limitations on fumigant re-registration, etc. The restrictions on the use of fumigants have posse new global challenges to food and chemical industry and have resulted in effort to develop and register new fumigants as an alternative, primary to methyl bromide<sup>[3] [4]</sup>. There are several new developed fumigants or new fumigant formulations such as sulfuryl fluoride<sup>[5] [6] [7]</sup>), carbonyl sulphide<sup>[3] [4]</sup>), propylene oxide<sup>[8] [9] [10]</sup>), methyl iodide<sup>[11]</sup>), ozone<sup>[12]</sup>), ethyl formate<sup>[13]</sup>), cyanogen<sup>[14]</sup> and ethanDiNitrile<sup>[3] [4]</sup>. Some of these fumigants suffer from the limitation and may be used only for treatment of the particular type of the commodity or for application in a specific situation only. Sulfuryl fluoride is a promising candidate for the fumigation of stored food commodities, food processing facilities and as a quarantine fumigant, propylene oxide for dry

and shelled walnut, spices, cocoa powder and nutmeats<sup>[15]</sup>), ethyl formate can be suitable for dried fruits, carbon disulfide (an old fumigant still in use) for seed materials, carbonyl sulphide for grain fumigation<sup>[3][4]</sup>. The global challenges in the research and development of new fumigants and technology of fumigation are in the development of the fumigants that will successfully replaced highly effective and pretty cheap phosphin and methyl bromide<sup>[3][4]</sup>.

The use of botanical pesticides has been emerging as one of prime means to protect crops and their products and the environment from pesticide pollution, which is a global problem<sup>[16][17]</sup>). When extracted from plants, these chemicals are referred to collectively as “botanicals”. Botanical insecticides possess a spectrum of properties including insecticidal activity, repellence to pests, antifeedancy, insect growth regulation, toxicity to nematodes, mites, snail and slugs, and other pests of the agricultural importance. Also they possess antifungal, antiviral, and antibacterial properties against pathogens. Generally, botanicals degrade more rapidly than most conventional (synthetic) pesticides, and so are considered relatively environmentally benign and less likely to kill beneficial insects and mites than insecticides with longer residual activity. Since most of them generally degrade within a few days, and sometimes within a few hours, these insecticides must be applied more often. More frequent application, plus higher costs of production usually makes botanicals more expensive to use than synthetic insecticides<sup>[16]</sup>). Among botanicals the plant volatile essential oils (EO) are the most frequently studied as pesticides for pest and diseases management<sup>[18][17][19][20][21][22]</sup>.

However, the essential oils, beside a large scale demonstration of their efficacy and penetration, need a lot of research in order to determine their toxicological and safety data prior to the registration<sup>[21]</sup>). Also, as with other groups of insecticides, the potential use of the natural EO in stored grain insect pest management depends on many factors. Isman (1997)<sup>[23]</sup> tried to outline the challenges and barriers to the development and commercialization of new botanical insecticides and other natural insecticides. He believed, in spite of mostly favourable toxicology and minimal environmental impact and the efficacy, botanicals and other natural insecticides need to fulfil many other considerations for the successful commercialization and use. However, he believes that this group of insecticides may find a place in applications where there is a greater tolerance for the presence of insects and a focus is placed on environmental safety.

According to Rajendran and Sriranjini (2008)<sup>[24]</sup>, although in laboratory tests with adult insects some of the plant extracts have shown significant insect toxicity, their physical properties such as high boiling point, high molecular weight and very low vapor pressure are barriers for application in large-scale fumigations. The authors believe that plant products have the potential for small-scale treatments and space fumigations. Still there is lack of data for single or multiple components of essential oils on sorption, tainting and residues in food commodities. Also, the requirements for the registration of plant products may be another barrier<sup>[24]</sup>.

We believe that the other of factors that may greatly prevent the adaptation and use of the natural EO in stored grain fumigation are their relatively high concentrations needed for the effective protection of stored grain against insect pests, a great difference in the sensitivity of various insect species and the current prices of natural essential oils on the market.

The objectives of this review paper are:

- (a) to give an overview of the concentrations of essential oils to control insect pests of stored grain,
- (b) to analyze the current prices of essential oils on the market and the cost of the fumigation, and
- (c) to discuss the potential of the introduction and the use of essential oil to fumigate stored grain.

## An overview of the concentrations of essential oils to control insect pests of stored grain

The concentrations of natural EO and its active components needed for effective fumigation have been studied by many researchers. In order to enable the comparison of toxicity data we analyzed the reports that presented the doses of EO in the volume, mostly in  $\mu\text{g L}^{-1}$  or  $\mu\text{l L}^{-1}$ , published during the last 10 years.

Shaaya et al. (1997)<sup>[18]</sup> were assessed the fumigant activities of a large number of essential oils extracted from various spices and herb plants against *Tribolium castaneum* (Herbst) *Sitophilus oryzae* (L.), *Rhyzopertha dominica* (F.) and *Oryzaephilus surinamensis* (L.). The highly active *Labiatae* sp. oil ZP51, in a concentration of 1.4–4.5  $\mu\text{l L}^{-1}$  air and exposure time of 24 h caused 90% kill of all the insects in space tests. However, in columns 70% filled with wheat, a concentration of 50  $\mu\text{l L}^{-1}$  and 7 d exposure were needed to obtain 94–100% kill of the insects.

Liu and Ho (1999)<sup>[25]</sup> evaluated the fumigant activities of the essential oil extracted from *Evodia rutaecarpa* Hook f. et Thomas, against *Sitophilus zeamais* (Motsch.) adults and *T. castaneum* larvae and adults. *S. zeamais*  $\text{LC}_{50}$  was 41  $\mu\text{g L}^{-1}$  air and *T. castaneum*  $\text{LC}_{50}$  was 11.7  $\mu\text{g L}^{-1}$  air.

Rahman and Schmidt (1999)<sup>[26]</sup> examined the toxic effects of vapors of essential oils of *Acorus calamus* (L.) rhizomes obtained from three countries; India, Russia, and Former Yugoslavia on the adults and eggs of *Callosobruchus phaseoli* (Gyllenhal) reared on seeds of *Lablab purpureus* (Medik.). Significant reduction of oviposition was found in oils vapours at 5 and 10  $\mu\text{L}$  oil per 400 ml jar (12.5 to 25  $\mu\text{L}$  oil per 1000 ml jar) after 24 h exposure. Newly-laid eggs were more susceptible than older ones.

Tunç et al. (2000)<sup>[27]</sup> tested the ovicidal activity of essential oil vapours distilled from anise *Pimpinella anisum* (L.), cumin *Cuminum cyminum* (L.), eucalyptus *Eucalyptus camaldulensis* (Dehnh.), oregano *Origanum syriacum* (L.) var. *bevanii* and rosemary *Rosmarinus officinalis* (L.) against the confused flour beetle, *Tribolium confusum* (du Val.), and the Mediterranean flour moth, *Ephestia kuehniella* (Zeller). The exposure to vapours of essential oils from anise and cumin resulted in 100% mortality of the eggs. At a concentration of 98.5  $\mu\text{l L}^{-1}$  of anise essential oil the  $\text{LT}_{99}$  values were 60.9 and 253.0 hours for *E. kuehniella* and *T. confusum*, respectively. For the same concentration of the essential oil of cumin, the  $\text{LT}_{99}$  value for *E. kuehniella* was 127.0 h.

Sánchez-Ramos and Castañera (2000)<sup>[28]</sup> found out that the vapor of natural monoterpenes pulegone, eucalyptol, linalool, fenchone, menthone,  $\alpha$ -terpinene and  $\gamma$ -terpinene at the concentration of 14  $\mu\text{l L}^{-1}$  or below generated 90% mortality of mobile stages of *Tyrophagus putrescentiae* (Schrank).

Lee et al. (2001)<sup>[29]</sup> examined the fumigant toxicity of different essential oils towards the rice weevil, *S. oryzae*. The essential oil from eucalyptus contained 1,8-cineole (81.1%), limonene (7.6%) and  $\alpha$ -pinene (4.0%). The oil generated  $\text{LD}_{50}$  = 28.9  $\mu\text{l L}^{-1}$  air. 1, 8- cineole was more active ( $\text{LD}_{50}$  = 23.5  $\mu\text{l L}^{-1}$  air) than limonene and  $\alpha$ -pinene. Benzaldehyde ( $\text{LD}_{50}$  = 8.65  $\mu\text{l L}^{-1}$  air) occurring in peach and almond kernels had also a potent fumigant toxicity towards the rice weevils.

Papachristos and Stamopoulos (2002)<sup>[30]</sup> assessed the toxicity of vapours of the essential oils from *Lavandula hybrida* (Reverch.), *R. officinalis* and *Eucalyptus globulus* (Lab.) against the larvae and pupae of *Acanthoscelides obtectus* (Say.). The essential oil vapours were toxic to all immature stages tested with  $\text{LC}_{50}$  values ranging between 0.6 and 76  $\mu\text{l L}^{-1}$  air, depending on oil and development stages.

Lee et al. (2003)<sup>[20]</sup> evaluated the fumigant toxicity of twenty naturally occurring monoterpenoids against *S. oryzae*, *T. castaneum*, *O. surinamensis*, the house fly, *Musca*

*domestica* L., and the German cockroach, *Blattella germanica* L. Cineole, *l*-fenchone, and pulegone at 50 µg ml<sup>-1</sup> air caused 100% mortality in all five species tested.

Lee et al. (2004)<sup>[21]</sup> studied the potent fumigant toxicity of 42 essential oils and found out that six of them extracted from *Eucalyptus nicholi* (Maiden & Blakely), *E. codonocarpa* (Blakely & McKie), *E. blakely* (Maiden), *Callistemon sieberi* (F.Muell.), *Melaleuca fulgens* (R.Br.) and *M. armillary* (R.Br.) were toxic to *S. oryzae*, *R. dominica* and *T. castaneum*. The LD 50 and LD 95 against the adults of *S. oryzae* were between 19.0 to 30.6 and 43.6 to 56.0 µg ml<sup>-1</sup> air, respectively. The LD95 of 1,8-cineole was for *S. oryzae* 47.9, for *R. dominica* 30.4 and for *T. castaneum* 21.0 µg ml<sup>-1</sup> air. The fumigant toxicity of five oils in the space 50% filled up with wheat was 3 to 5 times lower in 50% filled up the space than in an empty space and in a case of EO extracted from *E. codonocarpa* in 50% filled up the space with wheat, even 9 times less toxic.

Prajapati et al. (2005)<sup>[31]</sup> were evaluated the insecticidal, repellent and oviposition-deterrent activity of essential oils extracted from 10 medicinal plants against *Anopheles stephensi*(Liston), *Aedes aegypti* (L.) and *Culex quinquefasciatus* (Say.). The essential oil of *Pimpinella anisum* (L.) showed toxicity against 4th instar larvae of *A. stephensi* and *A. aegypti* with equivalent LD<sub>95</sub> values of 115.7 µg ml<sup>-1</sup>, whereas it was 149.7 µg ml<sup>-1</sup> against *C. quinquefasciatus* larvae. Essential oils of *Zingiber officinale* and *Rosmarinus officinalis* were found to be ovicidal and repellent, respectively towards the three mosquito species.

Ketoh et al. (2005)<sup>[32]</sup> studied the effectiveness of the essential oil extracted from *Cymbopogon schoenanthus* (L.) against all development studies of *Callosobruchus maculatus* (Fab.). At the highest concentration tested (33.3 µl L<sup>-1</sup>) all adults of *C. maculatus* were killed within 24 h of exposure to the oil and the development of newly laid eggs and neonate larvae was also inhibited.

Ketoh et al. (2006)<sup>[33]</sup> assessed the insecticidal activity of crude essential oil extracted from *Cymbopogon schoenanthus* (L.) and of its main constituent, piperitone, on different developmental stages of *C. maculatus*. Piperitone was more toxic to adults with a LC<sub>50</sub> value of 1.6 µl L<sup>-1</sup> vs. 2.7 µl L<sup>-1</sup> obtained with the crude extract.

Tapondjou et al. (2005)<sup>[34]</sup> investigated the toxicity of cymol and essential oils of *Cupressus sempervirens* (L.) and *Eucalyptus saligna* (Sm.) against *S. zeamais* and *T. confusum*. *Eucalyptus* oil was more toxic than *Cupressus* oil to both insect species (LD<sub>50</sub>=0.36 µl cm<sup>-2</sup> for *S. zeamais* and 0.48 µl cm<sup>-2</sup> for *T. confusum*) on filter paper discs, and was more toxic to *S. zeamais* on maize (LD<sub>50</sub>=38.05 µl per 40 g grain).

Wang et al. (2006)<sup>[35]</sup> investigated repellent and fumigant activity of essential oil from mugwort *Artemisia vulgaris* (L.) to *T. castaneum*. At 8.0 µl mL<sup>-1</sup>, mortality of adults reached 100%, but with 12-, 14- and 16-day larvae, mortalities were 49%, 53% and 52%, respectively. At dosages of 10, 15 and 20 µl L<sup>-1</sup> air and a 96 h exposure period, mortality of eggs reached 100%. No larvae, pupae and adults were observed following a 60 µl L<sup>-1</sup> dosage.

Choi Won-Sik et al. (2006)<sup>[36]</sup> determined the toxicity of volatile components of thyme, sage, eucalyptus, and clove bud against the mushroom sciarid, *Lycoriella mali* (Fitch.)  $\alpha$ -Pinene was the most toxic fumigant compound found in thyme essential oil (LD<sub>50</sub>=9.85µl L<sup>-1</sup> air) followed by  $\beta$ -pinene (LD<sub>50</sub>=11.85µl L<sup>-1</sup> air) and linalool (LD<sub>50</sub>=21.15µl L<sup>-1</sup> air). The mixture of  $\alpha$ - and  $\beta$ -pinene exhibited stronger fumigant toxicity than  $\alpha$ - or  $\beta$ -pinene itself against the mushroom fly adults.

Negahban et al. (2007)<sup>[37]</sup> determined the content of essential oil extracted from *Artemisia sieberi* (Besser). The oil contained camphor (54.7%), camphene (11.7%), 1,8-cineol (9.9%),  $\beta$ -thujone (5.6%) and  $\alpha$ - pinene (2.5%).The mortality of 7 days old adults of *C. maculatus*, *S. oryzae*, and *T. castaneum* increased with concentration from 37 to 926 µl L<sup>-1</sup> and with exposure time from 3 to 24 h. A concentration of 37 µl L<sup>-1</sup> and an exposure time of 24 h were

sufficient to obtain 100% kill of the insects. *C. maculatus* was significantly more susceptible than *S. oryzae* and *T. castaneum*.

Rozman et al. (2007)<sup>[22]</sup> investigated the toxicity of 1,8-cineole, camphor, eugenol, linalool, carvacrol, thymol, borneol, bornyl acetate and linalyl acetate against adults of *S. oryzae*, *R. dominica* and *T. castaneum*. The most sensitive species was *S. oryzae*, followed by *R. dominica*. *T. castaneum* was highly tolerant of the tested compounds. 1,8-Cineole, borneol and thymol were highly effective against *S. oryzae* when applied for 24 h at the lowest dose (0.14  $\mu\text{L L}^{-1}$ ). For *R. dominica* camphor and linalool were highly effective and produced 100% mortality in the same conditions. Against *T. castaneum* no oil compounds achieved more than 20% mortality after exposure for 24 h, even with the highest dose (139  $\mu\text{L L}^{-1}$ ). However, after 7 days exposure, 1,8-cineole produced 92.5% mortality, followed by camphor (77.5%) and linalool (70.0%).

Stamopoulos et al. (2007)<sup>[38]</sup> were tested vapor form of monoterpenoids terpinen-4-ol, 1,8-cineole, linalool, *R*-(+)-limonene and geraniol against different stages of *T. confusum*. The  $\text{LC}_{50}$  values ranging between 1.1 and 109.4  $\mu\text{L L}^{-1}$  air for terpinen-4-ol, from 4 and 278  $\mu\text{L L}^{-1}$  air for (*R*)-(+)-limonene (with  $\text{LC}_{50}$  and from 1,8-cineole 3.5 and 466  $\mu\text{L L}^{-1}$  air were the most toxic to all stages tested, followed by linalool (with  $\text{LC}_{50}$  values ranging between 8.6 and 183.5  $\mu\text{L L}^{-1}$  air) while the least toxic monoterpenoid tested was geraniol with  $\text{LC}_{50}$  values ranging between 607 and 1627  $\mu\text{L L}^{-1}$  air.

Korunic and Rozman (2008)<sup>[39]</sup> carried out three different experiments with 1,8-cineole. The authors conducted experiment in order to determine the efficacy of 509  $\text{g m}^{-3}$  of cineole against different developmental stages of *S. oryzae*, *R. dominica* and *Cryptolestes ferrugineus* (Steph.) in wheat grain in the space 50% filled up with grain. Apparently, applied dose of 50  $\text{g m}^{-3}$  was not sufficient for effective control of younger developmental stages of *S. oryzae*, *R. dominica* and even *C. ferrugineus*, the most sensitive species among tested. In the second experiment authors tested the effective concentration of cineole against adults of *S. oryzae*, *R. dominica*, *T. castaneum* and *C. ferrugineus* in space 50% filled up with wheat applying cineole in the concentration range of 50, 100, 150, 200, 250  $\text{g m}^{-3}$ . The 100% mortality of *C. ferrugineus* was obtained with 50  $\text{g m}^{-3}$  (lowest applied concentration). However, 100% mortality of *R. dominica* was obtained with concentration of 150  $\text{g m}^{-3}$  and 100% mortality of *S. oryzae* and *T. castaneum* at concentration of 250  $\text{g m}^{-3}$ . In the third experiment the concentration of 50  $\text{g m}^{-3}$  cineole in spaces differently filled up with wheat (empty space, 50% and 95% filled up) was assessed against the same four species. This concentration in empty space induced nearly 100% mortality in all four tested insect species. However, fumigation in a space 50% filled up with wheat, cineole was absolutely effective against *C. ferrugineus* only, with 50% to 60% efficacy against rice weevil and lesser grain borer, and only 11% against red flour beetle. In space 95% filled up with wheat mortality of rusty grain beetle was 88%, rice weevil 34%, lesser grain borer 64% and red flour beetle only 4.5%.

The results of Shaaya et al. (1997)<sup>[25]</sup>, Lee et al. (2004)<sup>[21]</sup> and Korunic and Rozman (2008)<sup>[39]</sup> demonstrated the significant effect of different quantity of wheat grain in the same volume on the effectiveness of EO against stored grain insect pests. In a space filled with grains for the successful control several times higher concentrations has to be applied in the comparison with concentrations applied in an empty space. This may be one of a very important limited factor for wider use of EO in grain fumigation.

### **The current price of essential oils on the market and the cost of the fumigation**

Currently, EO are sold in different packages containing 5 ml, 14.75 g (1/2 oz) up to 907.2 g (32 oz) and 3780 ml (US gallon). The prices depend on the type of the essential oil,

technology of the extraction, the size of the package and on producers, as well. The prices of EO sold by different producers, generally speaking, may be significantly different (Table 1). The size of the package greatly affects the cost of EO. One gram of Citronella EO in the package of 14.175 g (1/2 oz) costs US \$ 0.49 but in a gallon (3789 ml) 1 ml costs US \$0.065; 1g of Lavandin organic EO in the package of 14.175g costs US \$ 0.69 costs but in a gallon 1 ml costs US \$0.16; 1 g of Lavender Provence-Organic EO in a package of 14.175 g costs US \$1.28 but in a gallon 1 ml costs US \$0.46, etc. Also, the prices of various EO are significantly different. For example, in the package of 14.175 g (1/2 oz) 1 g of different oils costs from US \$ 0.49 (Citronella) to US \$1.3 (Juniperus Berry). In the package of 907.2 g (32 oz) 1 g costs from US \$0.32 (myrtle) to US \$5.54 (Jasmine Absolute). In the package of 3789 ml (US gallon) 1 ml of different essential oils costs from US \$0.064 (Citronella) to US \$0.47 (Oregano) (the producer Dreaming Earth Botanicals, LLC, Ashenwill, NC, USA).

**Table 1.** The approximate prices of essential oils (EO)

Essential oil	Producer	Size of package*	Cost of package (US \$)	Cost of 1 g or 1 ml (US \$)
Lavender Provence Organic (France)	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3780 ml (US galloon)	173.00	0.045
Lavender Spike France	Snowdrift Farm, Inc.	2268g (80 oz)	169.95	0.074
Bulgarian Lavender	Snowdrift Farm, Inc.	2268g (80 oz)	285.00	0.125
Lavandin FCC France	Snowdrift Farm, Inc.	2268g (80 oz)	159.95	0.070
Lavandin Organic	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3780 ml (US galloon)	623.00	0.164
Geranium	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3780 ml (US gallon)	1055.00	0.278
Geranium Burbon	Snowdrift Farm, Inc.	2268g (80 oz)	639.20	0.277
Geranium Egyptian Rose	Snowdrift Farm, Inc.	2268g (80 oz)	356.25	0.157
Rosemary (Maroccan)	Snowdrift Farm, Inc.	2268g (80 oz)	154.95	0.068
Rosemary (Spanish)	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3780 ml (US gallon)	528.00	0.139
Juniperus (Italy)	Snowdrift Farm, Inc.	2268g (80 oz)	525.00	0.231
Juniperus Berry Organic (Croatia)	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3789 ml (US gallon)	2360.00	0.622
Thyme linalool	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	907.2g (32 oz)	408.00	0.449
Bay (Laurus nobilis)	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3789 ml (US gallon)	1214.00	0.320
Tagetes	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3789 ml (US gallon)	1598.00	0.421
Clove bud	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	3789 ml (US gallon)	998.00	0.263
Myrtle	Dreaming earth botanicals, LLC, Ashenwill, NC, USA	907.2g (32 oz)	321.00	0.353

Pepper, black	Dreaming earth botanicals, LLC, Ashenville, NC, USA	3789 ml (US gallon)	752.00	0.198
Bergamot	Dreaming earth botanicals, LLC, Ashenville, NC, USA	907.2g (32 oz)	338.00	0.372
Lemon Organic	Dreaming earth botanicals, LLC, Ashenville, NC, USA	3789 ml (US gallon)	623.00	0.164
Patchouli	Dreaming earth botanicals, LLC, Ashenville, NC, USA	3789 ml (US gallon)	731.00	0.192
Patchouli (India)	Snowdrift Farm, Inc.	2268g (80 oz)	475.00	0.209
Jasmin Absolute	Dreaming earth botanicals, LLC, Ashenville, NC, USA	850.5g (30 oz)	4717.00	5.546
Nutmeg FCC (East Indian)	Snowdrift Farm, Inc.	2268g (80 oz)	299.00	0.131
Basil	Dreaming earth botanicals, LLC, Ashenville, NC, USA	3789 ml (US gallon)	647.00	0.170
Oregano	Dreaming earth botanicals, LLC, Ashenville, NC, USA	3789 ml (US gallon)	1806.00	0.476

\*the largest package available; smaller packages are significantly more expensive.

**Table 2.** The approximate prices of essential oils (EO) and approximate cost of fumigation of cubic meter

Essential oil	Producer	Size of package*	Cost of package (US \$)	Cost of 1 g or 1 ml (US \$)	Approximative concentration; reference	Approximate cost (US \$) of EO to fumigate 1 cubic meter**
Eucalyptus globulus (the usual eucalyptus)	Dreaming earth botanicals, LLC, Asheville, NC, USA	3780 ml (US gallon)	557.00	0.147	<b>LD50</b> = 28.9 $\mu\text{l L}^{-1}$ against <i>S. oryzae</i> <sup>[29]</sup>	Much more than 4.2
					<b>LD50</b> =23.5 $\mu\text{l L}^{-1}$ against <i>S. oryzae</i> <sup>[29]</sup>	Much more than 5.
					<b>LC100</b> =50 $\mu\text{l L}^{-1}$ against <i>S. oryzae</i> , <i>T. castaneum</i> , <i>O. surinamensis</i> , <i>Musca domestica</i> , <i>Blatella germanica</i> <sup>[20]</sup>	11.8
					LD95 for <i>S. oryzae</i> was 47.9 $\mu\text{l L}^{-1}$ , for <i>R. dominica</i> was 30.4 and for <i>T. castaneum</i> 21 $\mu\text{l L}^{-1}$ , in an empty space <sup>[21]</sup>	4.95 to 11.3
1,8-cineole	Acros organic	100 g	23.60	0.236	<b>LD50</b> =from 3.5 to 3.5 to 466g $\mu\text{l L}^{-1}$ against <i>T. confusum</i> all stages <sup>[38]</sup>	Much more than 0.82 to 109.9
					<b>92.5%</b> mortality of <i>T. castaneum</i> after 7 days of exposure to 138.8 $\mu\text{l L}^{-1}$ <sup>[22]</sup>	32.7
					In an empty space <b>LC100</b> =50g $\text{m}^{-3}$ against <i>S. oryzae</i> , <i>R. dominica</i> and <i>T. castaneum</i> <sup>[39]</sup>	11.8
						11.8 (C.

					In a space 50% filled up with grain, <b>LC100</b> =50g m <sup>-3</sup> for <i>C. ferrugineus</i> , 150g m <sup>-3</sup> for <i>R. dominica</i> , and 250g m <sup>-3</sup> for <i>S. oryzae</i> and <i>T. castaneum</i> <sup>[39]</sup>	<i>ferrugineus</i> ) 35.5 ( <i>R. dominica</i> ) 59.0 ( <i>S. oryzae</i> , <i>T. castaneum</i> )
					In a space 95% fill up with grain 50 g m <sup>-3</sup> caused mortality of <b>88%</b> ( <i>C. ferrugineus</i> ), <b>64%</b> ( <i>R. dominica</i> ) and <b>4.5%</b> ( <i>T. castaneum</i> ) <sup>[39]</sup>	Much more than 11.8
Camphor	Aldrich	100g	74.45	0.744	<b>77%</b> mortality of <i>T. castaneum</i> after 7 days of exposure to 139 µl L <sup>-1</sup> <sup>[22]</sup>	Much more than 74.4
Linalool	Aldrich	100 g	25.93	0.259	<b>70%</b> mortality of <i>T. castaneum</i> after 7 days of exposure to 139 µl L <sup>-1</sup> <sup>[22]</sup>	Much more than 25.9 to 44.9
					<b>LD50</b> =from 8.6 to 183.5 µl L <sup>-1</sup> against <i>T. confusum</i> all stages <sup>[38]</sup>	Much more than 2.2 to 47.5
					<b>LD90</b> =14 µl L <sup>-1</sup> against <i>Tyrophagus putrescentiae</i> <sup>[28]</sup>	More than 3.6
Thyme linalool	Dreaming earth botanicals, LLC, Asheville, NC, USA	907.2g (32 oz)	408.00	0.449	<b>LD50</b> =21.5 µl L <sup>-1</sup> against <i>Lycoriella mali</i> <sup>[36]</sup>	Much more than 5.5 to 9.6
Aniseed (Anise seed)	Dreaming earth botanicals, LLC, Asheville, NC, USA	3789 ml (US gallon)	375.00	0.098	Anise essential oil <b>LC99</b> =98.5 µl L <sup>-1</sup> against <i>E. kuehniella</i> and <i>T. castaneum</i> <sup>[27]</sup>	9.65

\*the largest package available; smaller packages are significantly more expensive.

\*\* µl/L is equal to ml in cubic meter; close to g in cubic meter depending on the density of EO (for example density of cineole is 0.9225 g cm<sup>-3</sup>, linalool 0.858-0.868 g cm<sup>-3</sup>).

## The potential of the introduction and the use of essential oil to fumigate stored grain

Analyzing the prices of EO produced by numerous producers by searching data available on internet, by direct contact with the producers and by analyzing the results of the effectiveness of EO published by numerous authors, it is obvious that the prices may be the limited factor for the adoption and its wider use (Table 2). It is a great difference in approximate concentrations of phosphine, methyl bromide and EO 1,8-cineole to give 95% and higher mortality of *S. oryzae* with 24 h exposure. According to Champ and Dyte (1976)<sup>[40]</sup> and re-calculated from Ct based on 20 h exposure, the approximate concentration of phosphine is 0.03 g m<sup>-3</sup>; the approximate concentration of methyl bromide, from Ct based on 5 h exposure, is 1 g m<sup>-3</sup>. Lee et al. (2004)<sup>[21]</sup> determined the concentration of 42 g m<sup>-3</sup> of 1,8-cineole to give 95% mortality of *S. oryzae*. Korunic and Rozman (2008)<sup>[39]</sup> determined that 50 g m<sup>-3</sup> of cineole in an empty space and 48 h exposure caused 100% mortality of *S. oryzae*, in a space 50% filled with wheat grain the mortality was 57% and in a space filled up 95% with wheat grain the mortality was 34% only. Shaaya et al. (1997)<sup>[18]</sup> found out that the highly active *Labiatae* sp. oil ZP51, in a concentration of 1.4–4.5 µl L<sup>-1</sup> air and exposure time of 24 h caused 90% killed *T. castaneum*, *S. oryzae*, *R. dominica* and *O. surinamensis*. However, in columns 70% filled up with wheat, a concentration of 50 µl L<sup>-1</sup> and 7 d exposure were needed to obtain 94–100% kill of the insects. Lee et al. (2004)<sup>[21]</sup> found out that EO extracted from *Eucalyptus nicholii*, *E. codonocarpa*, *E. blakelyi*, *Callistemon sieberi*, *Malaleuca fulgens* and *M. armillata* were 3 to 5 times less toxic to *S. oryzae*, *R. dominica* and *T. castaneum* in a space 50% filled up with wheat in comparison with the toxicity in an empty space.

One (1) kg of phosphine pellets costs about US \$41.00 US, whilst 1 kg of cineole in packages of 100g reaches about US \$236.00. When the highest dosage of phosphine pellets is applied (30 pellets t<sup>-1</sup>) with 1 kg of phosphine it is possible to fumigate approximately 55 tons of grain. It means the cost of phosphine to fumigate one tone of grain is about US \$0.74. With 1 kg of 1,8-cineole it is possible to fumigate 4 tons (Korunic and Rozman (2008)<sup>[39]</sup>; 95% space filled up with wheat) to about 23 tons of grain (Lee et al. 2004<sup>[21]</sup>; 50% space filled up with wheat). It means the cost of 1,8-cineole to fumigate one ton of grain is from approximately US \$10.00 to US \$59.00. The great effect of grain on the reduction of the effectiveness of EO may greatly increase the cost of the grain fumigation with EO and make them too expensive to be adopted for wider grain fumigation use.

### Conclusion

Besides of different barriers under the process of the registration, we find such a high price for cineole, and for other essential oils as well, considering other characteristics (scent, sorption, penetration, aeration etc.), as a serious limiting factor for the application of natural essential oils in practice. We believe that there are two solutions to overcome the mentioned barriers; significant reduction of the prices of natural EO, or if possible, to produce the active components of natural EO synthetically.

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