The Influence of the Essential Oils on the Pests in the Old Book Collections

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Abstract:
The use of essential oils for disinfection and pest control of old book collections can be a non-residual, ecological and economical alternative to chemical treatments, being safe both for the objects and for the staff. The repellant and biocidal activity of some volatile oils has been proven effective against a variety of microorganisms and insects.

Keywords: *essential oils, biodeterioration, old book*

Introduction

In the process of degradation and deterioration of the materials that make up libraries and archives, the biological type are among the most common; in certain microclimate conditions or in exceptional cases they can attain to catastrophic proportions and impact.

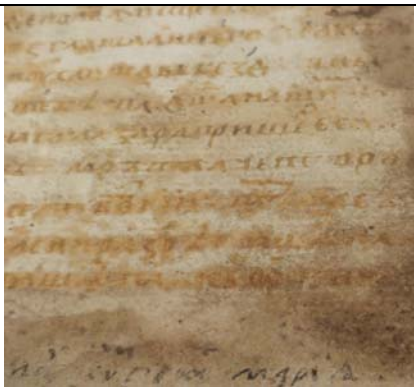
The most important biological agents involved in deteriorating documents with paper binding are: microorganisms (*i.e.* bacteria, fungi), insects and rodents.

The most frequent types of degradation caused by the microbiological agents include: enzymatic degradation of cellulose, degradation of protein materials (such as: leather, parchment, adhesives), chromatic alterations, degradation of the gallotannins that cause discoloration of inks, lowering of the pH in paper caused by acid products of bacterial metabolism (fumaric acid, citric acid, lactic acid). Many species of Aspergillus and Penicillium make up large amounts of oxalic acid, especially if the nutrient environment promotes an alkaline reaction in which the acid is transformed into the oxalate.

The irreversible damage caused by the bibliophages consisted of: loss of integrity of the piece (holes, galleries, abrasions, punctures) (Figure 1a), decreased resistance of the paper caused by the consumed gluing agents (Figure 1b), lowered pH level of the paper and its weakening as a result of the action caused by the acidic by-products of the metabolism (Figure 1c), and chromatic alterations caused by the droppings (Figure 1d).

a

b



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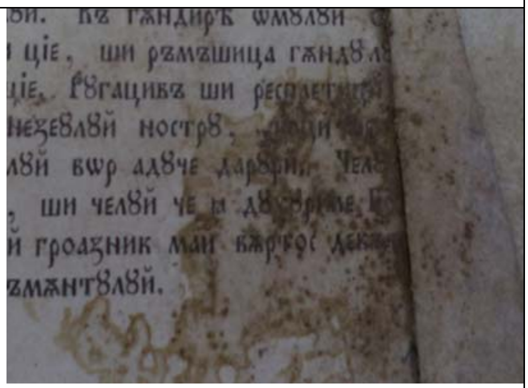

d

Figure 1. Effects caused by biological agents on the materials in the archives and book deposits

The negative impact on the staff working in the archives should be mentioned in addition to the damage caused by the biological agents on the documents. The main micro-fungi from the collections of rare books and documents that precipitate human infections are species of Aspergillus, Penicillium, and Fusarium. In time, various harmful effects on the chemicals (synthesized biocides) were proven to have happened, not only on paper but also on the staff, warehouses personnel, etc. The presence of biological agents in the archive can cause occupational diseases such as asthma, allergic rhinitis, pneumonia, endocarditis, aspergillosis, dermatitis, etc. The mycotoxins produced by the fungi cause mycotoxicoses – "poisoning" of the body by means of toxic substances released by the fungi on the substrate, the symptoms being similar to poisoning by pesticides or heavy metals.

Aflatoxins (produced by *Aspergillus flavus, A. parasiticus, A. zonatus, A. clavatus, A. sojae, A. toxicarius, A. nidulans* etc.), citrinin, patulin, the cyclopiazonic acid, the penicillic acid, ochratoxin, rubratoxin, cyclochlorotine, islanditoxin and luteoskirin produced by species of Penicillium are among the by-products produced by the metabolism of microorganisms. These mycotoxins affect the thyroid gland, the muscles, the circulatory system, and the heart causing localized tumors (Nisipeanu et al. 2009).

Staff contamination occurs both by inhaling polluted air, carrying 2-3 mm spores, and by direct contact with infected objects.

In this context, the use of unconventional treatments for disinfecting and pest control on the rare book collections is recommended by specialists as an ecological and economical alternative to chemical treatments typically used in biological pest control today. Special attention should be given to essential oils of all the unconventional treatments for both the treatment of the infested books and air decontamination in these areas.

The use of essential oils

The history of essential oils is one and the same with the history of medicinal plants. The Egyptians have used herbs for medicinal and cosmetic purposes and for the embalming process since 3000 BC. In ancient Greece, Hippocrates mentions in his writings two-hundred and thirty-six herbs. Some knowledge about plants and their use for health benefits were taken from Arab and Byzantine medicine, having been passed along to scholars of the Middle Ages. In the 16^{th} century, there were many books that contained recipes for obtaining oils, fragrant essences as well as herbal combinations.

There are treatises by Avicenna, the Arabian doctor dating from the 10^{th} century in which he mentions over eight hundred herbs and their beneficial effects on the body. During the Renaissance, Theophrastus Paracelsius, a scholar and professor at the University of Basel, achieved a qualitative leap in the use of medicinal plants. By using chemistry in phyto-therapy, he proved that not the whole plant, but the active substance it contains can heal. He called that active part "Arcanum" or "quinta esentia", outlining the concept of "active substances" (active ingredients) in the modern era (Radu and Andronescu 1981). After that, the extraction, isolation and purification of a growing number of herbal substances took place, subsequently being synthesized in laboratories.

In recent decades, plants have been used more and more in therapy, leading phyto-therapy to a spectacular rebirth. In current conditions, when microorganisms have developed a high resistance to conventional antibiotics, essential oils began to be used both for treating various diseases, and for purifying the air in hospitals, in order to limit the spread of microbial germs (De Billerbeck 2007).

Essential oils: generalities

Essential oils are generally liquids with an oily texture, being insoluble in water but soluble in alcohol and organic solvents. They have the distinct smell of the powerful substances they contain, which give away the characteristic scent of plants, flowers, fruit, seeds, or tree bark.

Chemically speaking, essential oils are complex mixtures of aliphatic and aromatic hydrocarbons, aldehydes, alcohols, esters and other components, terpenoid compounds being the most frequently encountered.

Although they are called oils, these substances do not contain fats: a drop of essential oil placed on a sheet of paper will not leave any traces, unlike vegetable oil.

Volatile oils can be extracted from various parts of the plant: flowers (chamomile, rose, lavender), seeds (cardamom), leaves (mint, basil), bark (cinnamon), roots (iris), floral buds (cloves), wood (sandalwood), fruits (anise), etc.

The content of essential oils is very small; for example, in order to obtain 1 liter of essential oil we need about 10 kg of cloves or 150 kg of lavender flowers.

There are three ways in which essential oils can be obtained:

- steam distillation, a process invented in the 11th century and frequently used now;
- cold pressing;
- solvent extraction.

Each species of plant has specific components in varying proportions, depending on the conditions of its growth. The place where the plant is grown, the climate, altitude, soil composition influence the chemical composition of the extracted oils. Also, depending on the distillated part of the plants (fruits, leaves, flowers, bark), the same plant can provide essential oils with different biochemical characteristics and properties. The composition of essential oils is very complex. Essential oils may contain hundreds of different aromatic molecules, each molecule having specific properties. These molecules act synergistically, which explains the efficiency and versatility of these essential oils.

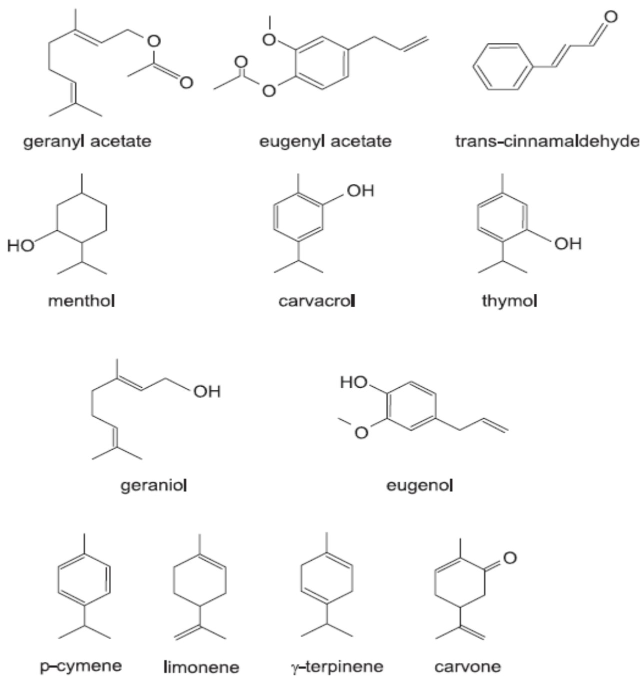

Figure 2. Essential oils components (Burt, 2004)

Using essential oils in the protection of cultural heritage

Since ancient times, certain flowers or leaves of plants have been inserted between the pages of books to protect them from pest attack (Kathpalia 1978). The essential oils from herbs and medicinal plants are known for their antibacterial, antifungal, acaricide (Araujo et al. 2012; Hussein et al. 2006), insecticide qualities (the orders of Lepidoptera, Coleoptera, Diptera, Isoptera, Hemiptera) (Tripathi et al. 2009) and nematicide qualities (Bai, Liu and Liu 2011). These properties of the essential oils have led to the idea of their use to decontaminate infected documents and the premises that house artifacts or documents with paper bindings.

The use of essential oils extracted from plants in preventing and fighting bio-deterioration of works of art is both a method that is complementary to other interventions to protect the collections and an alternative treatment to effectively combat pests. As a method of treatment, these powerful oils can be applied by means of direct application or fumigation.

The effects of essential oils on microorganisms

The antimicrobial activity of some essential oils and their components has been demonstrated to be effective against a variety of microorganisms, bacteria (Gram + and Gram-) and fungi. Apparently Gram- bacteria are more resistant to biocide activity obtained from plants, due to lipopolysaccharide present in the outer membrane, but there are also some exceptions (González-Lamothe et al. 2009).

Essential oils exhibit a differentiated antimicrobial activity, closely related to the chemical structures of the components that are present in the oil, being influenced by the specific mechanisms of synergism / antagonism between the components or by environmental factors or specific microorganisms against which they are used.

Studies on the antimicrobial effect of essential oils have proved their effectiveness both in vitro and in vivo (Saviuc et al. 2011; Soković 2012).

Essential oils act on bacteria in three stages:

- The essential oil attacks the cell wall of the bacteria, which results in an increase in permeability and then loss of cellular components.
- There is a lowering of the pH level in the bacteria, an acidification that will cause changes in the existing enzymes in the bacteria, thus blocking cellular energy production and the synthesis of the substances that are necessary to the bacterial cell.
- The last step is the destruction of genetic material leading to cell death (Popa 2014).

Although the mechanism of action on fungi is not known in detail, essential oils were found to influence both their growth rate and their morphological structure. Studies have proven the action of essential oils on the plasma membrane, whose structure and function are altered and the transport of nutrients is disrupted (Di Pasqua et al. 2006; Ferdes and Ungureanu 2012).

Methods for testing antimicrobial activity bring together classical microbiological methods (the antibiogram – an adapted diffusion method in several methodological versions, the kinetic method and the microdilution method) as well as microscopic methods (for proving morphological changes in the studied microbial strains).

The effectiveness of essential oils is assessed by measuring 3 concentrations:

• MIC, Minimum Inhibitory Concentration, is the minimum concentration of an antimicrobial agent that inhibits growth of the bacteria after incubation under standard conditions compared with the control/blank sample. Without an antimicrobial agent, the microorganisms remain viable.

• MBC, Minimum Bacterial Concentration, is the minimum concentration of the antimicrobial agent required to kill initial inoculum after incubation under standard conditions; the microorganisms no longer remain viable. MLC (Minimal Lethal Concentration) is also used. MBC and MLC can be used in the study of antifungal activity.

• MFC, Minimum Fungicidal Concentration, the minimum concentration of an antimicrobial agent that totally inhibits fungal growth after incubation under standard conditions; the microorganisms no longer remain viable (Klaric, Mastelic and Pieskova 2006).

The three concentrations are expressed in µl/ml or % (vol/vol).

Previous research presented the effects of essential oils on some species of fungi developed in old books' storage facilities: *Aspergillus niger*, *Aspergillus flavus*, *Trichoderma viride*, *Chaetomium globosum*, *Penicillium funiculosum*, *Penicillium chrysogenum*, *Fusarium solani*, etc. Studies were done on three stages of microorganism development: spores, germination system and mycelium, each of these stages being distinct in terms of structure, metabolism and providing a proper response to the action of the disinfectants.

The essential oils of myrtle, wormwood, thyme, sage, chenopodium, eucalyptus, lavender, rosemary, ragweed, etc., were applied by direct contact or fumigation and were analyzed in terms of effects, of the minimum concentration levels required and in terms of their effectiveness compared with the effects of synthetic products: cineol, linalool, eugenol, citronellal, etc.

Tests have shown the following:

• The minimum required concentration of essential oils is higher by contact than by fumigation.

• The synthetic compounds corresponding to natural oils are not more effective than the natural ones, but there are phenomena of synergy or antagonism between certain natural compounds.

• Sensitivity to these treatments depends on the fungal species, and stage of development; thus the *Penicillium crysogenum* is the most sensitive and the *Aspergillus flavus* is the most resistant of all the spores. At the level of the germination system there is great sensitivity toward the *Aspergilllus versicolor* and high resistance by the *Penicillium variotii*.

• It is obvious that essential oils cause morphological changes in the structure of the fungi: swelling, shrinkage and/or distortion.

Taking into account all the results, the effectiveness of the studied products can be classed/sorted into the following classes: for the synthetic products: linalool, citronellal, eugenol, and for the essential oils: chenopodium, myrtle, thyme, eucalyptus, sage, lavender and rosemary.

Fungicidal power is about 400-500 ml/m3, and the fungistatic action is obtained in less than 300 ml/m3. In light of these observations, the specialists' conclusion is that research should continue in order to reduce the amount of product used (*Désinfection des documents par des procédés physiques, CRCDG).*

Effects of the essential oils on insects

Studies present *Stegobium paniceum* (Coleoptera: Anobiidae) (Moşneagu 2012), *Lasioderma serricorn*e (Coleoptera: Anobiidae), *Tribolium castaneum* (Coleoptera, Tenebrionidae), *Liposcelis bostrycophila* (Psocoptera, Liposcelididae), *Dermestes maculatus* (Coleoptera: Dermestidae), the genus Attagenus (Coleoptera: Dermestidae) with its species: *Attagenus fasciatus, Attagenus piceus* among the species of pests that damage collections of old books and affected by essential oils (Ali et al 2011) (Table 1).

Table 1. The effectiveness of essential oils on pests in museums

The species that damages books	The source of the essential oil	Work conditions	Stage	Results	Author
Stegobium paniceum	*Dendranthema indicum* L. Des Moul.	Contact	adults	LD_{50}: 5,82 µg/adult	Zhang W.J. et *al.*, 2015

	Chamazulene - extracted from *Dendranthema indicum*	contact	adults	LD_{50}: 4,3 µg/adult	
	Zanthoxylum bungeanum M.	96 ul/l fumigation	larvae	LD_{50}:292,13 ppm/48h	Can L., 2011
			adults	LD_{50}:6784,18 ppm/48h	
		contact: 1000 ppm, time:144 h.	larvae	Mortality100%	
			adults		
	Ocimum basilicum	fumigation for a week	Larvae	Mortality 100%	Moşneagu M., 2012
	Cyperus rotundus			LD_{50}: 0,784% v/v	Kokate et *al*., 1980
Lasioderma serricorne	*Zanthoxylum dissitum* Hemsl	Contact	adults	LD_{50}: 13,8 µg/adult	Wang C.F. et al., 2015
	Mentha piperita (Labiatae) peppermint	fumigation : 1 ml/80 cm^3	larvae	LD_{95}:4,75	Bakr et *al*. (2010)
			adults	LD_{95}:9,75	
	Ocimum basilicum (Lamiaceae) basil		larvae	LD_{95}:7,73	
			adults	LD_{95}:10,41	
	Citrus lemon (Rutaceae) lemon		larvae	LD_{95}:9,58	
			adults	LD_{95}:11,59	
	Citrus sinensis (Rutaceae) orange		larvae	LD_{95}:10,04	
			adults	LD_{95}:10,56	
	Foeniculum vulgare	contact, time: one day, dosage:0,105 mg cm^{-2}	adulţi	Mortality100%	Kim & Ahn, 2001
Attagenus fasciatus	*Mentha piperita* (Labiatae) peppermint	fumigation : 1 ml/80 cm^3, time: 24h	larvae	LD_{95}:4,33	Bakr et *al*. , 2010
			adults	LD_{95}:6,32	
	Ocimum basilicum (Lamiaceae) basil		larvae	LD_{95}:8,19	
			adults	LD_{95}:9,92	
	Citrus lemon (Rutaceae) lemon		larvae	LD_{95}:8,53	
			adults	LD_{95}:11,10	
	Citrus sinensis (Rutaceae) orange		larvae	LD_{95}:10,30	
			adulţi	LD_{95}:11,22	
Attagenus piceus	*Zanthoxylum dissitum* Hemsl	Contact	Larvae	LD_{50}: 96,8 µg/larvae	Wang C.F. et al., 2015
Dermestes maculatus	*Tea Tree*	contact, time: 3days	larvae	LC_{50}: 71,18 mg/g	Maksoud G.A. et *al*., 2013
	Illicium verum	fumigaţie, timp: 3 zile	Larve	LC_{50}:o,86	Zhang B., 2012
			adults	LC_{50}: 2,07	

Tribolium castaneum	*Zanthoxylum dissitum* Hemsl	Contact	adults	LD_{50}: 43,7 µg/adult	Wang C.F. et al., 2015
	Dendranthema indicum L. Des Moul.	Contact	adults	No contact toxicity	Zhang W.J. et *al*., 2015
	Chamazulene – extracted from *Dendranthema indicum*	Contact	adults	LD_{50}: 29,52µg/adult	
	Mintha piperita L.	fumigation : 15µl/litre, time: 3 h	adults	Mortality 75%	Shaaya et *al*., 1991 (in Thorayia *et al*., 2012)
	Ocimum gratissimum	fumigation	adults	Little effect	Ogendo J.O. et *al*., 2008
	Cupressus sempervirens	Contact	adults	LD_{50}:0,74 µl/ cm^2	Tapondjou et *al*., 2005
	Eucalyptus oil			LD_{50}:0,48 µl/ cm^2	
	cymol			LD_{50}:0,96 µl/ cm^2	
Liposcelis bostrycophila	*Acorus calamus* (Acoraceae) Calamus	Contact	adults	LD_{50}:100,21 µg/cm^2	Xin Chao Liu et *al*., 2013
		fumigation	adults	LC_{50}: 392,13µg/L aer	

LD – lethal dose
LC – lethal concentration

The oils' routes of entry into the insects' bodies are: the respiratory system (by inhalation), through the skin (by absorption), through the digestive system (by ingestion) (Tripathi et al. 2009), through the sense organs on the antennae (Ahmed, Yasui and Ichikawa 2009; Thorayia et al. 2012). Essential oils have a higher effect on the insects by fumigation than by direct contact, therefore by penetrating the respiratory system (Bakr et al. 2010).

Depending on the type of essential oil, concentration, exposure time, the development stage of the insect, essential oils can cause birth defects which prevent the growth of the individual to adulthood, the resulting adults being unfit for breeding or their offspring being malformed. Some essential oils have a repellant or biocidal action on larval stages and adults.

Most monoterpenes are cytotoxic to plants and animals, causing decrease in the number of mitochondria, affecting breathing, lowering the permeability of the cell membrane (Tripathi et al. 2009).

Essential oils may disrupt communication among insects in the mating season by blocking the operation of the antennae sensilla (recipient organs), thereby reducing the populations of pests by lowering fertility, the number of eggs deposited (Ahmed, Yasui and Ichikawa 2001; Bakr et al. 2010). Ahmed et al. (2001) studied the decreased fertility of *Callosobrucus chinensis* species treated with Neem oil vapors.

The antennae are considered to be the most important channel of perception of external stimuli by insects. Capturing volatile substances is achieved through several types of sensilla, of different shapes and sizes. Essential oil substances that can be perceived by the insects can be sex pheromones, kairomones substances indicative of the host plant. The pheromones diffuse through the pore wall of the sensilla in its lymph, are solubilized, encapsulated by the specific proteins and transported to specific olfactory receptors (Leal 2005). The adults *Lasioderma serricorne* and *Attagenus fasciatus*, resulting from stage 3 larvae exposed to vapors of essential oil of peppermint for 24 hours, have suffered malformations and defects of the antenna. The malformations consisted of the widening of the membranes between antennomeres, and damage in the trichoid sensilla, especially in males, these sensilla being considered to have a role in receiving the female sex pheromones. Peppermint oil may cause suffocation and prevents some biosynthetic processes in the development of the insect to the stage of imago (Bakr et al. 2010).

Peppermint essential oil (*Mintha piperita*) causes adverse effects to the development of *Callosobruchus maculatus* species (Bruchidae): the higher the quantity of the oil is, the less adults resulting from the treated there are; the number of Bohm sensilla decreases; there are malformations in the sensilla, changes of direction/guidance of the sensilla, the merger of the trichoid sensilla; malformations such as swelling in the joints of the antennomeres, and the broadening of the membranes between segments; malformations of the mouthparts (Thorayia et al. 2012).

Mesbah et al. (2006) consider that essential oils act mainly as insect growth inhibitors, causing disruption of their development and resulting in abnormality ultimately leading to lower pest populations. Insect growth regulators inhibit chitin deposition causing abnormal endocuticular deposits, affecting the moulting process as well.

Conclusions

The first area that used bioactive principles from natural sources was alternative medicine that uses plants as sources of antioxidant and antimicrobial agents, in alternative treatments of various diseases.

The composition of essential oils is influenced by genotype, the extraction solvent, the geographical origin of the plants from which they are obtained and, last but not least, the agricultural and environmental conditions in which the plants grow.

The use of essential oils in plants has several advantages: some oils can be used to prevent pest attacks by acting as repellent substances, some oils can be used to control pests that damage cultural goods, acting as an insecticide; some oils are specific, pest control being based on the relationship of plant to insect (thus acting only on the harmful species, biocoenosis being less disrupted as compared to broad-spectrum insecticides).

Essential oils are environmentally friendly, economical and easy to obtain.

There are some limitations in using essential oils: increased volatility, absence of residual effect, possible allergic reactions of the staff. However, in this context, further research on the use of essential oils for decontamination treatments of both infested items and air in the spaces that house collections of rare books, is a very appealing alternative.

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