Antifungal Potential of Three Natural Oils and Their Effects on the Thermogravimetric and Chromatic Behaviors When Applied to Historical Paper and Various Commercial Paper Sheets

Maisa M. A. Mansour,[a] Mohamed Z. M. Salem,[b] Rushdya Rabee Ali Hassan,[a] Hayssam M. Ali,[c,d,*] Dunia A. Al Farraj,[c] and Mohamed S. Elshikh [c]

Three natural extracted oils from *Citrus reticulata* peels, *C. aurantifolia* leaves, and *Linum usitatissimum* (linseeds) were used as antifungal agents against the growth of *Aspergillus flavus* and *Penicillium chrysogenum*. The following main compounds (determined *via* gas chromatography–mass spectrometry) were found. The essential oil (EO) from *C. aurantiifolia* leaves contained limonene (22.96%), geranyl acetal (13.53%), and geraniol acetate (13.33%); the *n*-hexane oil from *C. reticulata* peels contained methyl-13-cyclopentyltridecanoate (16.74%), and *D*-limonene (16.06%); and linseed oil contained linoleic acid (27.36%), and oleic acid (19.01%). The inhibition of fungal growth significantly was reached 100% against *A. flavus* at all tested *C. aurantifolia* leaf EO concentrations and at a concentration of 2000 μL/mL for linseeds oil. The growth inhibition reached 100% against *P. chrysogenum* with *C. aurantifolia* leaf EO concentrations of 125-2000 μL/mL. *Citrus reticulata* peel EO had 100% growth inhibition of *P. chrysogenum* at concentrations of 2000 μL/mL and 1000 μL/mL, while linseeds oil had 100% growth inhibition at 2000 μL/mL. Thermogravimetric analysis showed that *C. aurantifolia* EO yielded the greatest thermal stability and color change protection to cotton pulp, while linseed oil was found to protect wood pulp-based and historical papers.

Keywords: Antifungal potential; Chromatic behaviors; Natural oils; Paper; pH, TGA; FTIR

*Contact information: a: Conservation Department, Faculty of Archaeology, Cairo University, Giza 12613 Egypt; b: Forestry and Wood Technology Department, Faculty of Agriculture (EL-Shatby), Alexandria University, Alexandria 21545 Egypt; c: Botany and Microbiology Department, College of Science, King Saud University, P.O. Box 2455, Riyadh 11451 Saudi Arabia; d: Timber Trees Research Department, Sabahia Horticulture Research Station, Horticulture Research Institute, Agriculture Research Center, Alexandria 21526 Egypt; *Corresponding author: hayhassan@ksu.edu.sa*

INTRODUCTION

Fungi are the main deterioration agents for wood, wood products, paper, historical manuscripts, leathers, and other heritage artifacts. They degrade the polysaccharide components and cause cell wall degradation or discoloration of organic materials (da Silva *et al.* 2006; Zyani *et al.* 2009; Sequeira *et al.* 2014; Hassan and Mansour 2018; Abo Elgat *et al.* 2020a; Hassan *et al.* 2020a; Mansour *et al.* 2020a; Mansour *et al.* 2020b; Salem *et al.* 2020a). Most of these fungi, including *Aspergillus flavus* and *Penicillium chrysogenum*, cause damage to historical papers in practice (Ljaljević-Grbić *et al.* 2013; Pinheiro *et al.* 2019).

Medicinal/aromatic plants and edible seeds are good sources for natural oils and

extracts with potential biological activities against different groups of pathogens, *e.g.*, antibacterial (Abbassy *et al.* 2020; Ashmawy *et al.* 2020a, b), antifungal (El-Hefny *et al.* 2019; Salem *et al.* 2019a,b; Behiry *et al.* 2020; Mansour *et al.* 2020a; Mohamed *et al.* 2020a, b; Salem *et al.* 2020b), and insecticidal (Hussein *et al.* 2017; Hamada *et al.* 2018; El-Sabrout *et al.* 2019; Hamad *et al.* 2019; Salem *et al.* 2020b), as well as providing antioxidant properties (Salem *et al.* 2016a; Elansary *et al.* 2017; El-Hefny *et al.* 2018; Al-Huqail *et al.* 2019; Okla *et al.* 2019a). In the present work, extracted oils from *Citrus aurantifolia, C. reticulata*, and *Linum usitatissimum* were used.

The peel oils of many species of *Citrus* showed the presence of limonene as the primary compound (Moufida and Marzouk 2003; Golmohammadi *et al.* 2018; Okla *et al.* 2019b; Abo Elgat *et al.* 2020b), which provides strong antifungal activity against *A. flavus* (Velázquez-Nuñez *et al.* 2013). The essential oil from *C. aurantifolia* showed strong inhibitory effects towards *Aspergillus parasiticus* and aflatoxins production (Rammanee and Hongpattarakere 2011). The primary compound in *C. aurantifolia* leaves essential oil, *D*-limonene, showed promising antibacterial activity against *Staphylococcus aureus* and *Escherichia coli* strains with excellent *in vitro* antioxidant activity (Al-Aamri *et al.* 2018). Essential oils and extracts from *C. aurantifolia* are known to exhibit important biological activities against several pathogens, *e.g.*, *Staphylococcus aureus*, *Escherichia coli*, *Klebsiella pneumonia*, *Aspergillus niger*, and *Candida albicans*, such as antiaflatoxigenic and anticancer properties (Aibinu *et al.* 2007; Razzaghi-Abyaneh *et al.* 2009; Pathan *et al.* 2012; Narang and Jiraungkoorskul 2016). Limonene (46.7%), as well as other compounds such as geranial, neral, and geranyl, were identified in the oil extracted from *Citrus reticulata* Blanco, which all show promising antifungal activity (Chutia *et al.* 2009). The peel oil from *C. reticulata* proved to be more toxic to *Sitophylus zeamais* adults than *C. aurantiifolia* oil (Fouad and da Camara 2017).

Flaxseed (*Linum usitatissimum* L., Linaceae family) is a good source of α-linolenic acid (an omega-3 fatty acid), as well as phenolic compounds, peptides, cyanogenic glycosides, alkaloids, polysaccharides, proteins, and fixed oil, which promises several health benefits (Hall *et al.* 2006; Krajčová *et al.* 2009; Bayrak *et al.* 2010; Goyal *et al.* 2014; Shim *et al*. 2014; Chauhan *et al.* 2015). The unsaturated fatty acids and lignans are the two primary groups of metabolites in flaxseed that exhibit antimicrobial activities (Paiva *et al.* 2010; Fadzir *et al.* 2018). The extracts derived from flaxseeds have been suggested to be effective in prohibiting the growth of *Escherichia coli, Salmonella paratyphii, Lactobacillus*, *Staphylococcus aureus*, *Proteus vulgaris, Klebsiella pneumoniae,* and *Saccharomyces cerevisiae* (Narender *et al.* 2016). Flaxseed oil was applied as a constrictive bioantifungal and exhibited average insecticidal properties (Kaithwas and Majumdar 2013). The *n*-hexane extract showed promising antimicrobial activity against *S. aureus*, *S. epidermis, Enterococcus faecalis, Escherichia coli*, and *K. pneumoniae* (Al-Mathkhury *et al*. 2016). The extracted oligosaccharides from flaxseed were found to be able to control the growth of *Alternia alternata* and *Alternia solani* (Guilloux *et al.* 2009).

This study was carried out in order to investigate *in vitro* the antifungal potency of extracted oils from *Citrus aurantifolia* leaves, *Citrus reticulata* peels, and *Linum usitatissimum* seeds against two fungal strains, *Aspergillus flavus* (acc#MH355958) and *Penicillium chrysogenum* (acc#MH352451). In this context, this is the first time these extracts have been evaluated for their effects on the thermogravimetric and chromatic properties of historical papers in comparison to paper made from the pulp of softwood and cotton.

EXPERIMENTAL

Preparation of the Natural Oils

The oil from *Citrus aurantifolia* leaves was extracted *via* the hydrodistillation method, where approximately 150 g of small pieces of leaves were put in 2 L flask containing 1500 mL of distilled water then connected to a Clevenger unit and heated for 3 h under refluxing (Abdelsalam *et al.* 2019). The obtained essential oil was kept dry in an Eppendorf tube.

Peels from *C. reticulata* were collected as the byproduct from fruits and linseeds (*Linum usitatissimum* L.) were purchased from an herbarium store located in Alexandria City, Egypt. Approximately 250 g of ripened linseeds and *C. reticulata* peels, in form of small pieces, were soaked (separately) in 200 mL of n-hexane solvent for 24 h. After the extraction process, the materials were filtered through a cotton plug using filter paper (Whatman No. 1), to removal any solid residues and to obtain the dissolved oils in n-hexane solvent (Ashmawy *et al.* 2020b). The solvent was evaporated, and the oils were obtained and preserved at 4 °C in a refrigerator until needed.

Chemical Analysis of the Oils

The oil extracts collected from ripened flax seeds and *Citrus reticulata* peels *via* n-hexane solvent extraction were analyzed for their chemical constituents with a Trace GC Ultra-ISQ mass spectrometer (Thermo Scientific, Austin, TX) with a direct capillary column TG–5MS (30 m × 0.5 mm × 0.25 µm film thickness) apparatus at the Atomic and Molecular Physics Unit, Experimental Nuclear Physics Department, Nuclear Research Centre, Egyptian Atomic Energy Authority (Inshas, Cairo, Egypt). The column oven temperatures and the chemical separation and identification conditions can be found in the study by Salem *et al.* (2019a). The conditions used to separate and identify the chemical compounds in the essential oil from the *Citrus aurantifolia* leaves can be found in the study by Okla *et al.* (2019b). Xcalibur 3.0 data system in the GC-MS with its values of threshold were used to confirm that all the mass spectra of the identified compounds were attached to the library. Furthermore, the measurement indices of Standard Index (SI) and Reverse Standard Index (RSI) with values ≥ 650 were used to confirm the identified compounds (Abdelsalam *et al.* 2019; Salem *et al.* 2019a,b; Ashmawy *et al.* 2020a,b; Mohamed *et al.* 2020a,b; Behiry *et al.* 2020).

Antifungal Activity of the Oils

The three oils were prepared at concentrations of 2000, 1000, 500, 250, 125, and 62 µL/mL by dissolving them 10% dimethyl sulfoxide (DMSO) followed by 0.5 mL of tween 80, which was used to emulsify the carrier oils in the solvent (Salem *et al.* 2016b, 2019b). Potato dextrose agar (PDA) medium was used to grow the two tested fungi, *Aspergillus flavus* (acc#MH355958) and *Penicillium chrysogenum* (acc#MH352451), at 26 °C at a relative humidity of 65±5%. The PDA medium was sterilized, and then the concentrated oils were added to the PDA medium and poured into sterilized Petri dishes. For each fungus type, fungal mycelial (7-day-old culture) discs, with a diameter of 0.5 cm, were put directly on the surface of the treated medium at the center of the Petri dishes. All the inoculated plates were incubated at 26 °C, and after the control treatment had finished growing (inoculated plates did not contain plant oils), the fungal diameter growth was measured in triplicate (Salem *et al.* 2017). The percentage of growth inhibition (GI) was calculated according to Eq. 1,

$$GI\% = [(G_1 - G_2)/G_1] \times 100 \quad (1)$$

where the GI is the mycelial growth inhibition (%), and G_1 and G_2 are the average diameters (mm) of the fungal colonies of the control (10% DMSO) and treatment, respectively. The minimum inhibitory concentrations (MICs) of the studied oils were measured as they were prepared at concentrations of 2 μL/mL to 62 μL/mL, using the broth dilution method according to CLSI (2008).

TGA Measurements

Source of papers

Paper samples with approximate dimensions of 7 cm × 15 cm with a 0.05 mm thickness were selected for the study and were not aged any further. The paper samples used were purified cotton linter cellulose (40 g/m²), paper sheets made from mechanical softwood pulp (40 g/m²) with a SR° of 40 in a Jokro (Rakta paper mill- Alexandria) (Hassan and Mohamed 2017), and a historical paper sample from the manuscript of "Tafsir Al Khazen", which is a book completely made of paper. The historical paper sample was received by the venerable Prince Louaa Ayoub, formerly Dafter Dar of Egypt, Mohamed Abu El Dahab in 1779 AD. The paper sheets were prepared according to the previous works (Hassan 2016; Hassan and Mohamed 2017; Hassan and Mansour 2018). All the paper samples were treated with the highest MICs values reported from the antifungal activity test. To study the effect of oil on paper, the samples were placed in Petri dishes contains cotton saturated with these oils, without contact between the oil and the paper, while the process was carried out through the sublimation of the oil (Massoud *et al.* 2012). Therefore, there is not impregnation with the oils. Furthermore, text-free samples were used.

TGA methodology

Thermogravimetric analysis was carried out with a Shimadzu TGA-50 device (Kyoto, Japan) at a temperature range of 22 °C to 760 °C in a static nitrogen atmosphere with a heating rate of 10 °C/min. The temperature ranges of the specimens in this study were evaluated *via* differential mass loss curves.

Measuring the Color Change

The color change parameters *L*, *a*, and *b* were measured with a HunterLab Labscan 600 spectrocolorimeter (version 3.0; Hunter Associates Laboratory Inc., Reston, VA), where *L* refers to the black-to-white color, *a* refers to the green-to-red color, and *b* refers to the blue-to-yellow color. The total color change of all oil treated paper types was expressed as ΔE, according to Eq, 2,

$$\Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2} \quad (2)$$

where $(\Delta L)^2$, $(\Delta a)^2$, and $(\Delta b)^2$ are the differences between the values of the color indices before and after oil treatment (Ali *et al.* 2018; Salem *et al.* 2020c; Salim *et al*. 2020).

Statistical Analysis

The fungi inhibition percentages were statistically analyzed using ANOVA in a completely randomized design with two factors (oil source and oil concentration) using Statistical Analysis System software (version 8.2, SAS, Cary, NC), and compared with the values of the control. The means were compared with a least significant difference (LSD) test at a significance level of a *p*-value less than 0.05.

RESULTS AND DISCUSSION

Chemical Composition and the Antifungal Activity of the Oils

The chemical compounds of the essential oil (EO) from *Citrus aurantiifolia* leaves are shown in Table 1. The primary compounds in the EO were limonene (22.96%), geranyl acetal (13.53%), geraniol acetate (13.33%), *γ*-dodecalactone (7.51%), caryophyllene oxide (7.4%), *β*-caryophyllene (7.36%), spathulenol (4.95%), neryl acetal (4.38%), citronellal (3.13%), *β*-citral (2.52%), and (*E*)-citral (2.10%).

Table 1. Phytochemicals of the Essential Oil from *Citrus aurantiifolia* (leaves) Analysed *via* GC-MS

Compound	Percentage in the Oil (%)	SI[1]	RSI[2]
D-Limonene	22.96	932	932
trans-β-Ocimene	0.57	828	853
Linalool	0.76	939	951
Citronellal	3.13	949	951
Citronellol	0.41	911	924
Nerol (*cis*-Geraniol)	0.61	891	906
Geraniol	0.54	932	934
β-Citral	2.52	903	905
γ-Dodecalactone	7.51	864	880
(*E*)-Citral	2.10	916	964
Geranyl acetal	13.53	800	905
Citronellyl formate	0.85	865	903
Neryl acetal	4.38	939	945
Geraniol acetate	13.33	967	973
Linalyl acetate	0.36	831	836
β-Caryophyllene	7.36	938	940
α-Caryophyllene	0.61	901	921
α-Farnesene	0.77	869	904
β-Bisabolene	0.71	881	909
γ-Elemene	0.48	833	887
Caryophyllene oxide	7.4	921	928
Spathulenol	4.95	852	884
2-Methylene-5*α*-cholestan-3*β*-ol	1.19	820	842
Arachidonic acid methyl ester	0.36	802	832
Oleic acid	0.23	794	807
1-Heptatriacotanol	0.18	788	826
Methyl hexadecadienoate	1.19	774	805
Note: SI = Standard Index; and RSI = Reverse Standard index			

Table 2 shows the chemical composition of the *n*-hexane oil (fixed oil) from *Citrus reticulata* peels as analyzed via gas chromatography–mass spectrometry (GC/MS). The primary compounds were methyl-13-cyclopentyltridecanoate (16.74%), *D*-limonene (16.06%), diethyl phthalate (13.37%), oleic acid (8.02%), methyl (16*E*)-16-octadecenoate (5.68%), 14-pentadecynoic acid methyl ester (4.39%), 1,3-diolein (4.33%), and *cis*-7-hexadecenoic acid methyl ester (4.22%). Table 3 presents the chemical constituents of linseed fixed oil; the primary compounds were linoleic acid (27.36%), oleic acid (19.01%), palmitic acid (18.28%), and methyl hexadecadienoate (16.26%).

Table 2. Phytochemicals of the Fixed Oil from *Citrus reticulata* Peels Extracted Using *n*-Hexane

Compound	Percentage in the Oil (%)	SI[1]	RSI[2]
D-Limonene	16.06	906	917
Methyl dihydromalvalate	1.00	797	840
14-Pentadecynoic acid methyl ester	4.39	819	819
cis-7-Hexadecenoic acid methyl ester	4.22	779	790
2-Methylene-5*α*-cholestan-3*β*-ol	1.20	813	836
Diethyl phthalate	13.37	714	848
Methyl-13-cyclopentyltridecanoate	16.74	777	778
Methyl hexadecadienoate	2.02	819	829
cis-9,10-Epoxy-octadecanoic acid	2.33	808	818
Oleic acid	8.02	823	831
Ethyl iso-allocholate	0.88	804	806
Methyl 14-Methylpentadecanoate	14.02	850	852
1,3-Diolein	4.33	812	819
Methyl (16E)-16-octadecenoate	5.68	838	863
Note: SI = Standard Index; and RSI = Reverse Standard index			

Table 3. Phytochemicals of the Fixed Oil from Flaxseed (*Linum usitatissimum*) Analysed *via* GC-MS

Compound	Percentage in the Oil (%)	SI[1]	RSI[2]
Linoleoyl chloride	5.32	752	783
α-Linoleic acid	27.36	798	815
7-Methyl-*Z*-tetradecen-1-ol acetate	6.91	763	783
Palmitic acid	18.28	788	797
Methyl hexadecadienoate	16.26	789	794
Stearic acid	3.54	794	808
Oleic acid	19.01	829	830
Note: SI = Standard Index; and RSI = Reverse Standard index			

As indicated in Table 4, the oils from *Linum usitatissimum*, *Citrus reticulata*, and *C. aurantifolia* showed different antifungal activity levels against the studied fungi (*Aspergillus flavus* and *Penicillium chrysogenum*). Generally, the inhibitory effect of the oils increased in proportion with an increase in concentration, and maximum inhibition was reached at the final concentration of 2000 µL/mL. Table 4 presents the growth inhibition (GI) percentage of the *A. flavus* and *P. chrysogenum* fungal mycelial, and how the GI values were affected by the three oils. At all the studied concentrations, the GI% significantly reached 100% as the essential oil from *C. aurantifolia* leaves was tested as antifungal agent against the growth of *A. flavus*, while it reached 100% as seed oil from *L. usitatissimum* was tested at the concentration of 2000 µL/mL in comparison with the control (*p*-value less than 0.05 *via* ANOVA). While *C. reticulata* peel EO yielded GI% values of 57.41% in terms of *A. flavus* growth, *C. aurantifolia* oil yielded the highest GI% values (100%) with significant antifungal activity (*p*-value less than 0.05) against *P. chrysogenum* at concentrations of 125 µL/mL, 250 µL/mL, 500 µL/mL, 1000 µL/mL, and 2000 µL/mL and reached a GI value of 81.48% at 62 µL/mL, when compared to the control treatment. *Citrus reticulata* peel oil yielded significant (*p*-value less than 0.05) GI values, with 100% of the fungal growth inhibited at the concentrations of 2000 µL/mL and 1000 µL/mL against *P. chrysogenum*. In addition, *L. usitatissimum* seed oil applied at 2000 µL/mL yielded a GI value of 100%, when compared to the control treatment.

Table 4. Antifungal Activity of the Tested Oils Against *Aspergillus flavus* and *Penicillium chrysogenum*

Oil	Conc (µL/mL)	*Aspergillus flavus*	*Penicillium chrysogenum*
Citrus reticulata (peels)	0 (10% DMSO, control)	0.00	0.00
	62	16.2 ± 0.37	38.5±0.37
	125	20.3± 0.37	71.4±0.37
	250	27 ± 0.37	75.9 ± 0.37
	500	41.8 ± 0.37	80.3 ± 0.37
	1000	57.4 ± 0.37	100
	2000	100	100
Citrus aurantifolia (leaves)	0 (10% DMSO, control)	0.00	0.00
	62	100	81.4 ± 0.37
	125	100	100
	250	100	100
	500	100	100
	1000	100	100
	2000	100	100
Linum usitatissimum (seed)	0 (10% DMSO, control)	0.00	0.00
	62	4.8 ± 0.37	72.9 ± 0.37
	125	17 ± 0.37	74.8 ± 0.37
	250	36.2 ± 0.37	80.3 ± 0.37
	500	42.9 ± 0.37	82.5 ± 0.37
	1000	47 ± 0.37	84.8 ± 0.37
	2000	100	100
p-value		< 0.0001	< 0.0001

Table 5 shows the MIC results of the studied oils, where the lowest MIC values were less than 2 µL/mL. This level of *C. aurantifolia* leaf essential oil was applied to inhibit the growth of *A. flavus* and *P. chrysogenum*, respectively. Therefore, the highest MIC values were 6 µL/mL, 2 µL/mL, and 32 µL/mL for *C. reticulata* (peels), *C. aurantifolia* (leaves), and *Linum usitatissimum* (seed), respectively. These concentrations were used to treat the paper made with mechanical softwood pulp, cotton paper, and historical paper.

Table 5. Minimum Inhibitory Concentrations (MICs) of the Oil Treatments

Source of Oil	MIC (µL/mL)	
	Aspergillus flavus	*Penicillium chrysogenum*
Citrus reticulata (peels)	4	6
Citrus aurantifolia (leaves)	< 2	2
Linum usitatissimum (seed)	32	16

In a study by Razzaghi-Abyaneh (2018), D-limonene was found to make up 22.96% of the compounds in the essential oil of *C. aurantifolia* leaves, but it reached 85.5% in the plants grown in Iran. In a study by Al-Aamri *et al.* (2018), D-limonene (63.35%) formed the major constituent of *C. aurantifolia* essential oil; however, other compounds, including 3,7-dimethyl-2,6-octadien-1-ol, geraniol, *E*-citral, *Z*-citral, and β-ocimene (7.07%, 6.23%, 4.35%, 3.29%, and 2.25%, respectively), were found. In a study by Ibrahim *et al.* (2019), D-limonene (57.84%) was the primary compound in *C. aurantifolia* leaf essential oil, with

notable compounds, including neral, linalool, sulcatone, and isogeraniol (7.81%, 4.75%, 3.48%, and 3.48%, respectively), were identified. A study by Lemes *et al.* (2018) found limonene, linalool, citronellal, and citronellol as the main constituents (77.5%, 20.1%, 14.5%, and 14.2%, respectively), in the essential oils from *C. aurantifolia* leaves and fruit peels, which showed promising activity against *Streptococcus mutans* and *Lactobacillus casei*.

Samples of *C. aurantifolia* from Italy contained limonene, β-myrcene, citral, γ-terpinene, β-pinene, and β-bisabolene as the primary compounds (Tundis *et al.* 2012; Spadaro *et al.* 2012). In addition, limonene and β-pinene were the major components in the essential oil extracted from *C. aurantifolia* collected in South Korea (Hong *et al.* 2017). *Citrus aurantifolia* leaf essential oil showed an inhibition value against *A. parasiticus* (47.8%) and therefore was considered to possess the ability to suppress this fungus (Rammanee and Hongpattarakere 2011). According to a study by Abo Elgat *et al.* (2020b), *Citrus sinensis* peel essential oil showed potential antifungal activity against *A. flavus* with a GI of 86.66% when applied at a concentration of 50 µL/mL. Dongmo *et al.* (2009) observed that *C. aurantifolia* essential oil had a fungicidal inhibiting action on the radial growth of *Phaeoramularia angolensis*.

Limonene (46.7%), followed by geranial, neral, geranyl acetate, geraniol, β-caryophyllene, nerol, neryl acetate (19%, 14.5%, 3.9%, 3.5%, 2.3%, 2.6%, and 1.1%, respectively) were found in the oil extracted from *C. reticulata* Blanco grown in India (Chutia *et al.* 2009), which possessed good antifungal activity against plant pathogenic fungi *Alternaria alternata, Rhizoctonia solani*, *Curvularia lunata*, *Fusarium oxysporum*, and *Helminthosporium oryzae*. *Citrus reticulata* essential oil at a concentration of 0.94% showed a 100% reduction of the growth of *A. flavus* and *P. chrysogenum* (Viuda-Martos *et al.* 2008).

The antifungal activity of the extracted oils is associated with the phytochemical components, *e.g.*, monoterpenes (Matasyoh *et al.* 2007), which are able to diffuse into cell membrane structures and damage them. Sokovic and Griensven (2006) observed that limonene and α-pinene possessed antifungal activity (a MIC of 4.0 µL/mL to 9.0 µL/mL) against *Verticillium fungicola* and *Trichoderma harzianum*, which are found at different amount in different plant essential oils (limonene and α-pinene). The essential oils and their related substances made the cell membrane of the fungus permeable, causing leakage (Piper *et al*. 2001).

Fatty acids, *i.e.*, linoleic, oleic, and palmitic, are the primary identified compounds in linseed essential oil. It was reported by Coşkuner and Karababa (2007) that the primary oil constituents were linoleic acid, oleic acid, and α-linolenic acid, with values ranging from 8% to 29%, 12% to 30%, and 35% to 67%, respectively. In addition, α-linolenic acid, linoleic acid, palmitic acid, oleic acid, and stearic acid were found in the ranges of 39.9% to 60.42%, 12.25% to 17.44%, 4.9% to 8%, 13.44% to 19.39%, and 2.24% to 4.59%, respectively, in a study by Goyal *et al.* (2014); additional studies found these compounds comprised 53%, 17%, 5%, 19%, and 3% of the primary compounds, respectively (Simopoulos 2002; Bernacchia *et al.* 2014). Linseed oil from a Romanian plant contained high levels of linolenic acid (53.21%) followed by oleic acid, linoleic acid, palmitic acid, and stearic acid (18.51%, 17.25%, 6.58%, and 4.43%, respectively) (Popa *et al.* 2012). Fatty acids α-linolenic (51.37%), oleic (20.59%), linoleic (15.8%), palmitic (5.86%), and stearic (5.57%) were reported as the primary compounds in the linseed oil analyzed in a study by Danish and Nizami (2019).

For linseed oil, the biological activity action of fatty acids is attributed to its

unsaturated long-chain fatty acids, *i.e.*, linoleic, linolenic, and oleic (Xu *et al.* 2008; Mueller *et al.* 2010; Chandrasekaran *et al.* 2011), while its saturated long-chain fatty acids, *i.e.*, stearic and palmitic, are less active (Seidel and Taylor 2004). The potential antifungal activity of linseed oil against *Aspergillus ochraceus* and *A. flavus* could be due to its rich α-linolenic acid and linoleic acid content (Abdelillah *et al*. 2013). Petroleum ether extract showed good antifungal activity against *Candida albicans* (Guilloux *et al.* 2009; Kaithwas *et al*. 2011), Flaxseed flour showed promising fungistatic activity against *Fusarium graminearum, A. flavus*, and *Penicillium chrysogenum* (Xu *et al.* 2008), while defatted flaxseed powder exhibited bioactivity against *A. flavus* and *A. niger* (Barbary *et al*. 2010). Linseed powder at a 6% concentration completely inhibited the development of *A. flavus* (Xu *et al.* 2008).

Thermogravimetric Properties of the Treated Paper Samples

Thermogravimetric analysis (TGA) is a simple and accurate method for studying the decomposition pattern and thermal stability of paper after treatment. Figures 1, 2, and 3 show the primary thermo-grams and derivato-grams for the reference paper sources, softwood pulp, cotton, and historical paper, respectively, as well as the samples treated with oil.

The references paper samples (wood-based, cotton, and historical paper) had an initial weight loss of 3.6%, 3.3%, and 2.86%, respectively, at approximately 105 °C, which is primarily due to the evaporation of any absorbed moisture (Madera-Santana *et al.* 2002; Hassan 2020). The primary decomposition proceeds in one step (Nasr and Ismail 2010) for each type of paper, *i.e.,* in the cotton sample the weight loss of 3.3% occurs at a decomposition temperature (T_d) of 107 °C and the weight loss of 3.6% for the wood-based sample occurred at a T_d of 105.6 °C. Furthermore, the authors were able to detect similar behavior between the untreated samples (Table 6).

The data in Table 6 show the difference in the results of the modern paper samples and the historical sample; the historical sample started the initial weight loss at high temperatures than the modern samples (the T_d of the historical sample was approximately 201 °C). Thermal gravimetric analysis allowed a conclusion to be drawn that the thermo oxidation destruction of historical paper, before and after treatment, was a multistage process, which involved at least three stages.

The maximum rate of the first stage of thermal oxidation can be determined by the weight loss rate, which is considered a major determinate of the degree of paper destruction. The data from Table 6 shows that the historical paper samples with the highest degree of destruction exhibited the highest rate of destruction during the first stage. For the historical paper samples, the lowest mass loss that occurs during stage II of the paper destruction process is related to the partial splitting of cellulose macromolecules, and therefore, increases the heterogeneousness of its structures (Kamel *et al.* 2004).

To examine the mass loss brought about by high temperatures, dM/dT curves (calculated by deriving weight loss *vs*. temperature data) are given for references and treated paper with oils, as shown in Figs. 1 through 3. Degradation of untreated paper started at a temperature lower than 105 ºC and degraded with much higher speed than treated samples. Initial degradation temperature of treated samples were much lower than references ones, and also the speed was much slower. In the heat *versus* weight loss curve, the mass loss peaks of untreated and treated took place at different temperatures. For wood-based paper, the mass loss peak occurred at a much lower temperature than cotton samples. Treating paper by oils made paper more thermally stable and increased the ash content, as

shown in Figs. 1 and 2. Although the improvement was (as it was not detected very clearly), a small improvement in thermal stability was considered important. It should be noted that the samples treated with *Linum usitatissimum* gave the highest thermal stability, especially with cotton samples. In accordance with the literature (Le Moigne 2008; Youssef *et al.* 2012), there was no degradation before 50 °C. Above this temperature, thermal stability gradually decreased, and decomposition of the fibers occurred in treated paper with two different steps: one peak below 40 °C, and another one at 105 °C. The first peak was assigned to the decomposition of oils, and it shifted to higher temperature than in untreated samples. The second inflection with the sharp peak was at the same T_d of original paper. The data show the effect of treatment on total initial weight loss; it is obvious that the treatment decreased total initial weight loss, especially for the samples treated with *C. reticulata* oil. Treated cotton with *Linum usitatissimum* and wood-based paper treated with *Citrus reticulata* had the highest and lowest stability, respectively.

However, there was a change of the destruction mechanism from hydrolytic to hemolytic, which might be due to stearic acid (in the chemical structure of the oils), which it is believed to enhance the hydrophobicity of the negative active material. It also reduces the extension of oxidation in the open atmosphere (Wang *et al.* 2007). Moreover, the loss of free water in paper results in a loss of flexibility, while the loss of bound water results in its deterioration, due to changes that occur in its chemical structure and physical properties (Hassan 2015).

Paper samples treated with linseed oil showed the highest mass loss values, but it should be taken into consideration that the end temperature was dramatically increased in comparison to the standard sample, which confirms the impact of linseed oil on the thermal characteristics of treated paper at high temperatures. The authors were able to identify two mechanisms of oxidation in linseed oil: (i) the poor oxidative stability of linseed oil at low temperatures can attributed to a high α-linolenic acid content (Rudnik *et al.* 2001); and (ii) the good oxidative stability of linseed oil at high temperatures can increase the rate of protection from oxidation at high temperatures, which was consistent with various studies. Khattab *et al.* (1999) found that as the linseed oil concentration in the cotton paper sample increases, the apparent activation energies of pyrolysis and oxidation decrease, due to the hypothesized formation of free radicals *via* the oxidation of linseed oil, which then catalyze the pyrolysis reactions of cotton. Linseed oil oxidation, which incorporates cross-linking reactions, involves oxygen consumption, and thus induces increased sample mass. Therefore, the mass reading of a TGA instrument corresponds to a combined effect of mass gaining reactions, *i.e.*, oxygen consumption, and mass losing reactions, *i.e.*, emission of carbon oxides and water. For this reason, gravimetric techniques alone are insufficient to investigate the spontaneous ignition of linseed oil impregnated into cellulose materials.

The results show the effectiveness of the oils at improving the thermal stability properties of the treated paper at different temperatures, especially *C. aurantifolia* oil, which improved the thermal properties of both cotton paper and wood-based paper dramatically, as the end temperature of the primary decomposition temperature was higher than the decomposition temperature of the standard sample. However, it must be noted that the improvement mechanism was linked to a closed link with the type of paper pulp, *i.e.*, *C. aurantifolia* oil yielded the best results with cotton pulp, while *L. usitatissimum* oil yielded promising results with softwood mechanical pulp-based and historical papers.

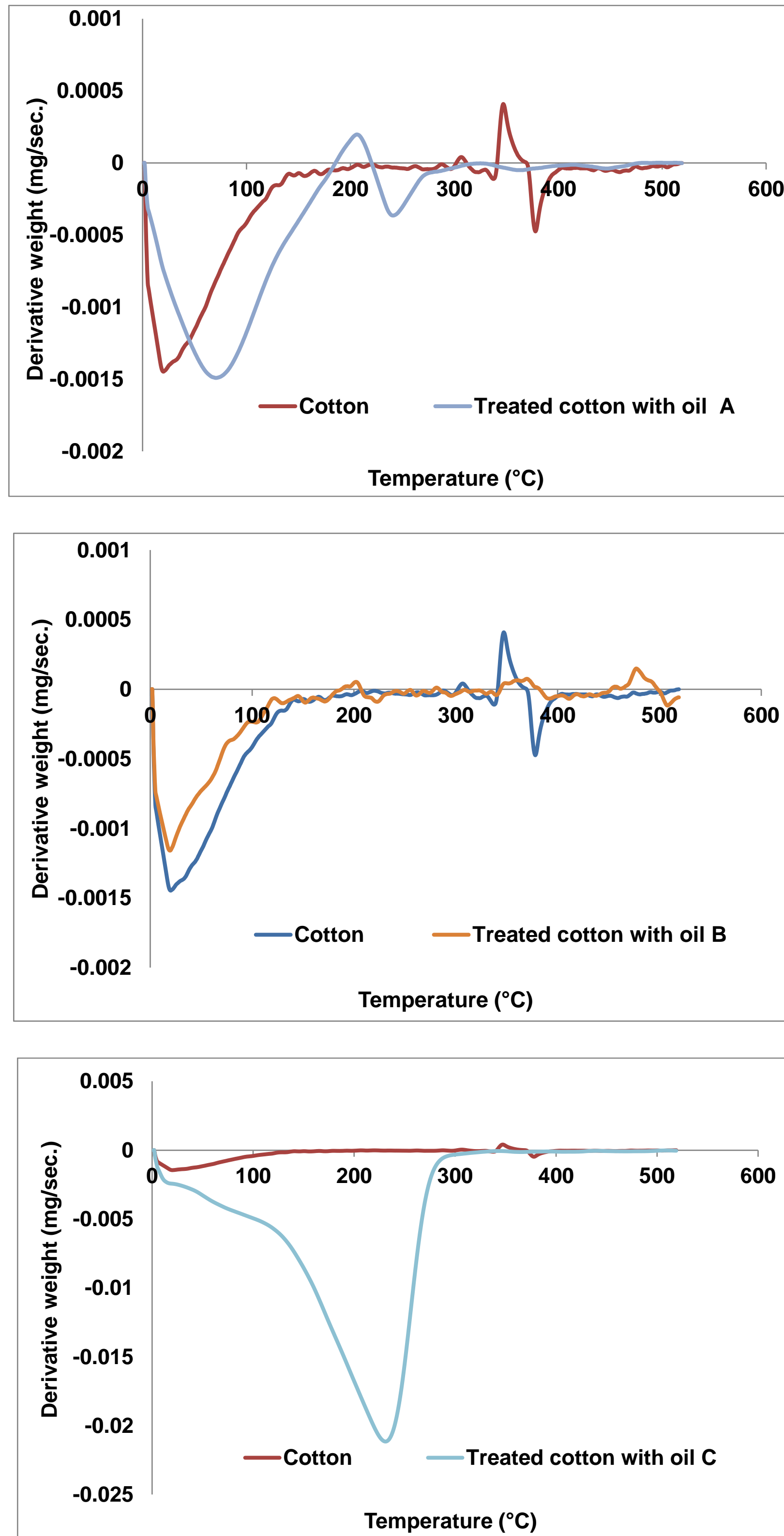

Fig. 1. Thermogravimetric analysis (TGA) for the weight change (mg/sec.) for the cotton paper samples before and after treatment with the three oils (A) *Citrus reticulata*; (B) *Citrus aurantifolia*; (C) *Linum usitatissimum*

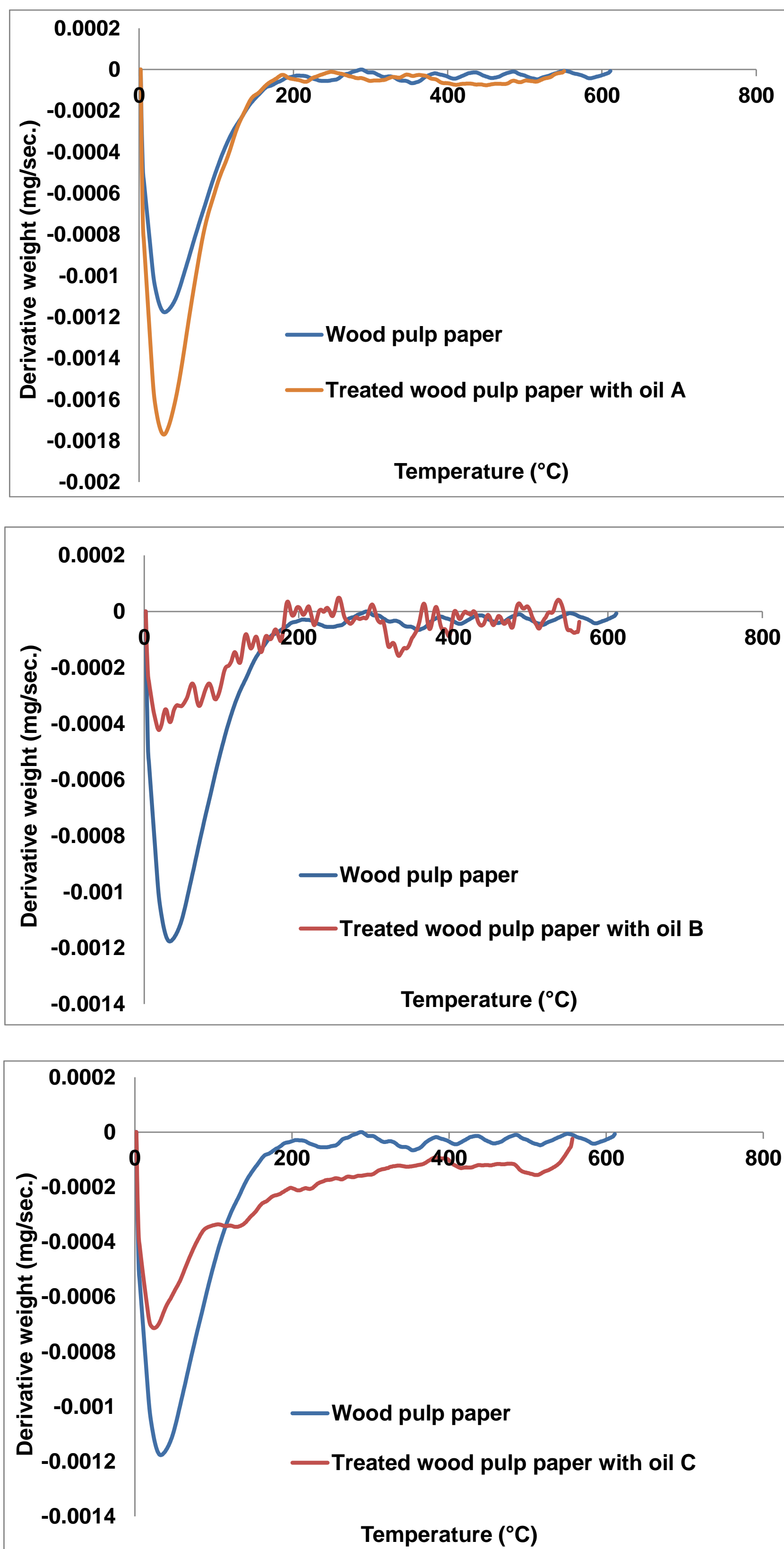

Fig. 2. Thermogravimetric analysis (TGA) for the weight change (mg/sec.) for the wood-based paper samples before and after treatment with the three oils A) *Citrus reticulata*; (B) *Citrus aurantifolia*; (C) *Linum usitatissimum*

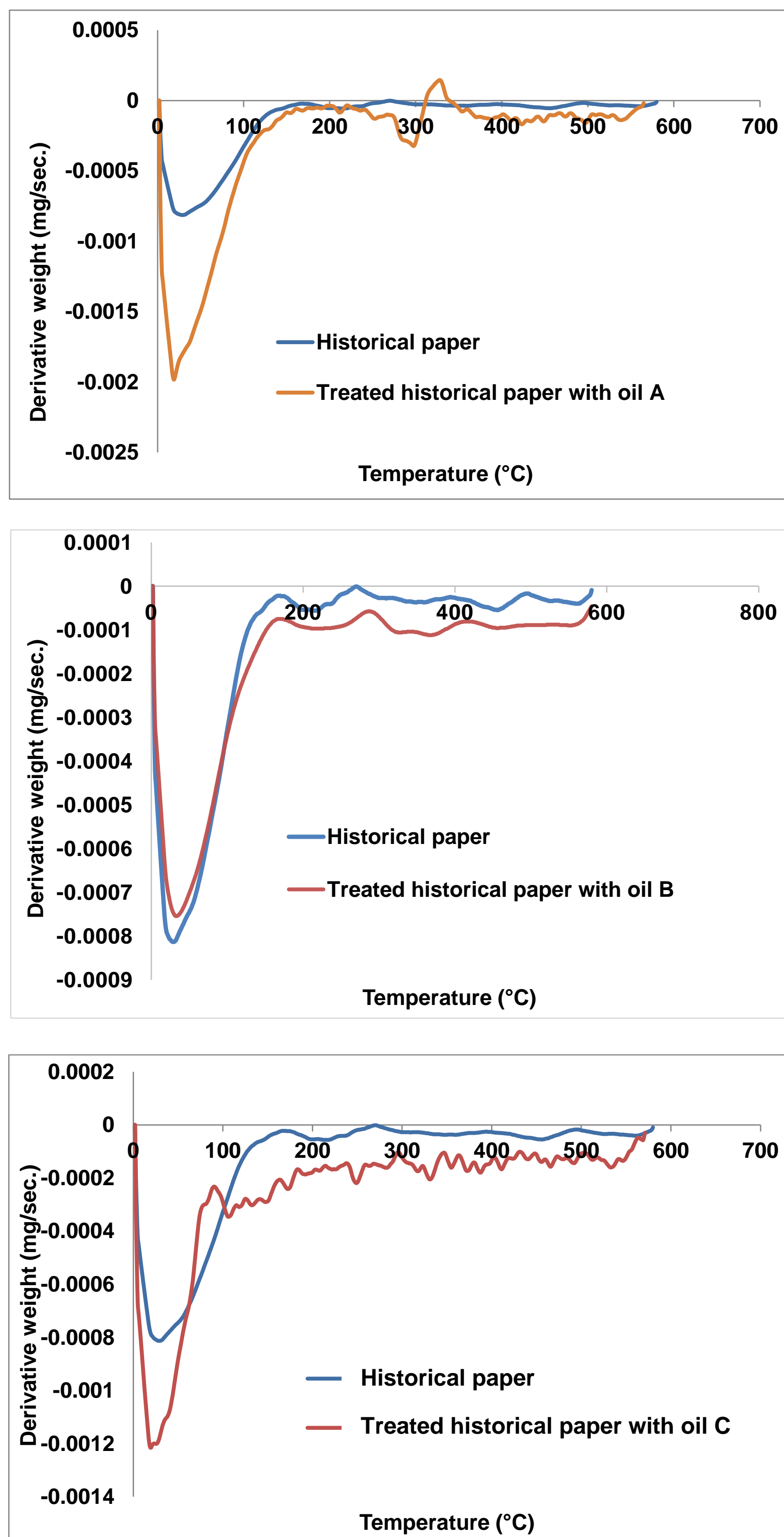

Fig. 3. Thermogravimetric analysis (TGA) for the weight change (mg/sec.) for the historical paper samples before and after treatment with the three oils. A) *Citrus reticulata*; (B) *Citrus aurantifolia*; (C) *Linum usitatissimum*

Table 6. Parameters of the Thermo-Oxidation Destruction for Paper Samples Before and After Oil Treatment

Source of Paper	Treated with Tested Oil	Start (°C)	End (°C)	Weight Loss (mg)
Cotton	Control	37.94	107	-0.102 (3.3%)
	Citrus reticulata	39.45	95.1	-0.06 (2.9%)
	Citrus aurantifolia	18.61	201.8	-0.17 (4.22%)
	Linum usitatissimum	35.7	148.8	-2.45 (6.7%)
Wood-based	Control	43.88	105.6	-1.01 (3.6%)
	Citrus reticulata	36.3	104.6	-0.13 (3.6%)
	Citrus aurantifolia	38.5	107.9	-0.01 (2.0%)
	Linum usitatissimum	37.55	201.3	-0.13 (4.4%)
Historical	Control	44.25	201.7	-0.09 (2.86%)
	Citrus reticulata	37.98	100.59	-0.130 (3.16%)
	Citrus aurantifolia	41.97	98.25	-0.063 (2.03%)
	Linum usitatissimum	36.54	201.06	-0.148 (4.9%)

Total Color Differences (ΔE)

The results of the total color change values (ΔE) for the paper samples before and after treatment with the tested three oils are shown in Table 7. The results confirmed that after the oil treatments, the ΔE values of the treated wood-based paper samples decreased significantly; the ΔE of the wood-based paper sample treated with *L. usitatissimum* oil was 2.11, while the ΔE was even higher when treated with *C. aurantifolia* oil (ΔE 9.33) and *C. reticulata* (6.65 ΔE).

Table 7. Total Color Differences of Paper Samples Before and After Treatment with Oils

Source of samples	Oil source	*L*	*a*	*b*	ΔE
Paper from softwood pulp	Blank	91.77	0.64	8.84	
	C. reticulata	87.27	1.41	13.68	6.65
	C. aurantifolia	86.31	2.69	16.13	9.33
	L. usitatissimum	90.92	0.78	7.43	2.11
Cotton paper	Blank	91.10	0.69	9.03	
	C. reticulata	85.47	3.08	8.95	6.11
	C. aurantifolia	89.97	0.50	10.84	2.14
	L. usitatissimum	84.64	1.79	10.93	6.82
Historical paper	Blank	86.72	1.79	10.30	
	C. reticulata	90.49	1.48	3.74	7.57
	C. aurantifolia	80.45	2.42	11.08	6.34
	L. usitatissimum	84.94	1.96	9.10	2.15

For treated the cotton paper samples, significant color change was found in the samples treated with *C. aurantifolia* and *L. usitatissimum* oils, with ΔE values of 6.11 and 6.82, respectively, however the ΔE decreased to 2.14 when treated with *C. aurantifolia* oil. The treated historical paper samples yielded ΔE values of 7.57, 6.34, and 2.15 when the paper was treated with the oils from *C. aurantifolia*, *C. aurantifolia* and *L. usitatissimum*, respectively. These results confirmed that the oils from *L. usitatissimum*, *C. aurantifolia* and *L. usitatissimum* played a vital role in reducing the color change values in wood-based paper, cotton paper, and historical paper, respectively. Table 6 shows that *L. usitatissimum* oil provided the best color change protection, as yielded the lowest ΔE values for the treated samples, which were classified as not noticeable to the naked eye, since their value was less than 5 (Hassan 2019, 2020b).

CONCLUSIONS

1. The effect of using three natural oils as antifungal agents, as well as their effects on the thermogravimetric, chromatic behaviors of a historical paper sample, and paper sheets made from softwood mechanical pulp and cotton were studied. The results indicated that the three oils assayed possessed antifungal properties against *Aspergillus flavus* and *Penicillium chrysogenum*, ranked in the following order: *C. aurantifolia* was greater than *C. reticulata*, which was greater than *L. usitatissimum*. These three essential oils could be used to control fungal infestations on various types of paper.
2. The thermogravimetric and chromatic alternations analyses demonstrated a positive effect of *C. aurantifolia* EO and color change protection to cotton pulp, while linseeds oil showed positive effects with wood pulp-based and historical papers.
3. The positive effect of the oils on the thermal properties of the papers provided a significant increase in the initial decomposition temperatures after treatment, but this improvement was linked to the type of paper.

ACKNOWLEDGMENTS

The authors would like to extend their sincere appreciation to the Deanship of Scientific Research at the King Saud University for funding this work through research group No. RG 1435-011.

REFERENCES CITED

Abbassy, M. M. S., Salem, M. Z. M., Rashad, N. M., Afify, S. M., and Salem, A. Z. M. (2020). "Nutritive and biocidal properties of agroforestry trees of *Moringa oleifera* Lam., *Cassia fistula* L., and *Ceratonia siliqua* L. as non-conventional edible vegetable oils," *Agroforestry System* 94, 1567-1579, DOI: 10.1007/s10457-018-0325-4

Abdelillah, A., Houcine, B., Halima, D., Meriem, C. S., Imane, Z., Eddine, S. D., Abdallah, M., and Daoudi C. S. (2013). "Evaluation of antifungal activity of free fatty acids methyl esters fraction isolated from Algerian *Linum usitatissimum* L. seeds against toxigenic *Aspergillus*," *Asian Pacific Journal of Tropical Biomedicine* 3(6), 443-448. DOI: 10.1016/S2221-1691(13)60094-5

Abdelsalam, N. R., Salem, M. Z. M., Ali, H. M., Mackled, M. I., EL-Hefny, M., Elshikh, M. S., and Hatamleh, A. A. (2019). "Morphological, biochemical, molecular, and oil toxicity properties of *Taxodium* trees from different locations," *Industrial Crops and Products* 139, DOI: 10.1016/j.indcrop.2019.111515

Abo Elgat, W. A. A., Kordy, A. M., Böhm, M., Černý, R., Abdel-Megeed, A., and Salem, M. Z. M. (2020b). "*Eucalyptus camaldulensis*, *Citrus aurantium* and *Citrus sinensis* essential oils as antifungal activity against *Aspergillus flavus*, *Aspergillus niger*, *Aspergillus terreus*, and *Fusarium culmorum*," *Processes* 8(8), 1-16. DOI: 10.3390/pr8081003

Abo Elgat, W. A. A., Taha, A. S., Böhm, M., Vejmelková, E., Mohamed, W. S., Fares, Y. G. D., and Salem, M. Z. M. (2020a). "Evaluation of the mechanical, physical, and anti-fungal properties of flax laboratory papersheets with the nanoparticles

treatment," *Materials* 13(2), 1-23. DOI: 10.3390/ma13020363
Aibinu, I., Adenipekun, T., Adelowotan, T., Ogunsanya, T., and Odugbemi, T. (2007). "Evaluation of the antimicrobial properties of different parts of *Citrus aurantifolia* (lime fruit) as used locally," *African Journal of Traditional, Complementary and Alternative Medicines* 4(2), 185-190.
Al-Aamri, M. S., Al-Abousi, N. M., Al-Jabri, S. S., Alam, T., and Khan, S. A. (2018). "Chemical composition and in-vitro antioxidant and antimicrobial activity of the essential oil of *Citrus aurantifolia* L. leaves grown in Eastern Oman," *Journal of Taibah University Medical Sciences,* 13(2), 108-112. DOI: 10.1016/j.jtumed.2017.12.002
Al-Huqail, A. A., Behiry, S. I., Salem, M. Z. M., Ali, H. M., Siddiqui, M. H., and Salem, A. Z. M. (2019). "Antifungal, antibacterial, and antioxidant activities of *Acacia saligna* (Labill.) H. L. Wendl. flower extract: HPLC analysis of phenolic and flavonoid compounds," *Molecules* 24(4), 1-14. DOI: 10.3390/molecules24040700
Ali, M. F., Mansour, M. M. A., Badr, N. M., and Salem, M. Z. M. (2018). "A study of biodeterioration and chromatic alterations of painted and gilded mummy cartonnage at the Saqqara museum storeroom, Egypt," *Archaeometry* 60(4), 845-858. DOI: 10.1111/arcm.12340
Al-Mathkhury, H. J. F., Al-Dhamin, A. S., and Al-Taie, K. L. (2016). "Antibacterial and antibiofilm activity of flaxseed oil," *Iraqi Journal of Science* 57(2B), 1086-1095.
Ashmawy, N. A., Al Farraj, D. A., Salem, M. Z. M., Elshikh, M. S., Al-Kufaidy, R., Alshammari, M. K., and Salem, A. Z. M. (2020a). "Potential impacts of *Pinus halepensis* Miller trees as a source of phytochemical compounds: Antibacterial activity of the cones essential oil and *n*-butanol extract*," Agroforestry System* 94, 1403-1413, DOI: 10.1007/s10457-018-0324-5
Ashmawy, N. A., Behiry, S. I., Al-Huqail, A. A., Ali, H. M., and Salem, M. Z. M. (2020b). "Bioactivity of selected phenolic acids and hexane extracts from *Bougainvilla spectabilis* and *Citharexylum spinosum* on the growth of *Pectobacterium carotovorum* and *Dickeya solani* bacteria: An opportunity to save the environment," *Processes* 8(4), 1-16. DOI: 10.3390/pr8040482
Barbary, O. M., El-Sohaimy, S. A., El-Saadani, M. A., and Zeitoun, A. M. A. (2010). "Antioxidant, antimicrobial and anti-HCV activities of lignan extracted from flaxseed," *Research Journal of Agriculture and Biological Sciences* 6(3), 247-256.
Bayrak, A., Kiralan, M., Ipek, A., Arslan, N., Cosge, B. and Khawar, K. M. (2010). "Fatty acid compositions of linseed (*Linum usitatissimum* L.) genotypes of different origin cultivated in Turkey," *Biotechnology & Biotechnological Equipment* 24(2), 1836-1842. DOI: 10.2478/V10133-010-0034-2
Behiry, S. I., Nasser, R. A., Abd El-Kareem, M. S. M., Ali, H. M., and Salem, M. Z. M. (2020). "Mass spectroscopic analysis, MNDO quantum chemical studies and antifungal activity of essential and recovered oil constituents of lemon-scented gum against three common molds," *Processes* 8(3), 1-24. DOI: 10.3390/pr8030275
Bernacchia, R., Preti, R., and Vinci, G. (2014). "Chemical composition and health benefits of flaxseed," *Austin Journal of Nutrition and Food Sciences*, 2(8), 1-9.
Chandrasekaran, M., Senthilkumar, A., and Venkatesalu, V. (2011). "Antibacterial and antifungal efficacy of fatty acid methyl esters from the leaves of *Sesuvium portulacastrum* L," *European Review for Medical and Pharmacological Sciences* 15(7), 775-780.
Chauhan, R., Chester, K., Khan, Y., Tamboli, E. T., and Ahmad, S. (2015).

"Characterization of *Linum usitatissimum L.* oil obtained from different extraction technique and in vitro antioxidant potential of supercritical fluid extract." *Journal of Pharmacy & BioAllied Science* 7(4), 284-288. DOI: 10.4103/0975-7406.168027

Chutia, M., Bhuyan, P. D., Pathak, M. G., Sarma, T. C., and Boruah, P. (2009). "Antifungal activity and chemical composition of *Citrus reticulata* Blanco essential oil against phytopathogens from North East India," *LWT - Food Science and Technology* 42(3), 777-780. DOI: 10.1016/j.lwt.2008.09.015

Clinical and Laboratory Standards Institute (CLSI) (2008). *Reference Method for Broth Dilution Antifungal Susceptibility Testing of Filamentous Fungi; Approved Standard-Second Edition* (CLSI document M38-A2), Clinical and Laboratory Standards Institute, Wayne, PA.

Coşkuner, Y., and Karababa, E. (2007). "Some physical properties of flaxseed (*Linum usitatissimum* L.)," *Journal of Food Engineering* 78(3), 1067-1073. DOI: 10.1016/j.jfoodeng.2005.12.017

da Silva, M., Moraes, A. M. L., Nishikawa, M. M., Gatti, M. J. A., de Alencar, M. A. V., Brandão, L. E., and Nóbrega, A. (2006). "Inactivation of fungi from deteriorated paper materials by radiation," *International Biodeterioration & Biodegradation*, 57(3), 163-167. DOI: 10.1016/j.ibiod.2006.02.003

Danish, M., and Nizami, M. (2019). "Complete fatty acid analysis data of flaxseed oil using GC-FID method," *Data in Brief* 23, 103845. DOI: 10.1016/j.dib.2019.103845

Dongmo, P. M. J., Tatsadjieu, L. N., Sonwa, E. T., Kuate, J., Zollo, P. H. A., and Menut, C. (2009). "Essential oils of *Citrus aurantifolia* from Cameroon and their antifungal activity against *Phaeoramularia angolensis*," *African Journal of Agricultural Research* 4(4), 354-358.

Elansary, H. O., Salem, M. Z. M., Ashmawy, N. A., Yessoufou, K., and El-Settawy, A. A. A. (2017). "*In vitro* antibacterial, antifungal, and antioxidant activities of *Eucalyptus* spp. leaf extracts related to phenolic composition," *Natural Product Research* 31(24), 2927-2930. DOI: 10.1080/14786419.2017.1303698

El-Hefny, M., Abo Elgat, W. A. A., Al-Huqail, A. A., and Ali, H. M. (2019). "Essential and recovery oils from *Matricaria chamomilla* flowers as environmentally friendly fungicides against four fungi isolated from cultural heritage objects," *Processes* 7(11), 1-12. DOI: 10.3390/pr7110809

El-Hefny, M., Mohamed, A. A., Salem, M. Z. M., Abd El-Kareem, M. S. M., and Ali, H. M. (2018). "Chemical composition, antioxidant capacity and antibacterial activity against some potato bacterial pathogens of fruit extracts from *Phytolacca dioica* and *Ziziphus spina-christi* grown in Egypt," *Scientia Horticulturae* 233, 225-232. DOI: 10.1016/j.scienta.2018.01.046

El-Sabrout, A. M., Salem, M. Z. M., Bin-Jumah, M., and Allam, A. A. (2019). "Toxicological activity of some plant essential oils against *Tribolium castaneum* and *Culex pipiens* larvae," *Processes* 7(12), 1-24. DOI: 10.3390/pr7120933

Fadzir, U. A., Darnis, D. S., Mustafa, B. E., and Mokhtar, K. I. (2018). "*Linum usitatissimum* as an antimicrobial agent and a potential natural healer: A review," *Archives of Orofacial Science* 13(2), 55-62.

Fouad, H. A., and da Camara, C. A. G. (2017). "Chemical composition and bioactivity of peel oils from *Citrus aurantiifolia* and *Citrus reticulata* and enantiomers of their major constituent against *Sitophilus zeamais* (Coleoptera: Curculionidae),' *Journal of Stored Products Research* 73, 30-36. DOI: 10.1016/j.jspr.2017.06.001

Golmohammadi, M., Borghei, A., Zenouzi, A., Ashrafi, N., and Taherzadeh, M. J.

(2018). "Optimization of essential oil extraction from orange peels using steam explosion," *Heliyon* 4(11), 1-18. DOI: 10.1016/j.heliyon.2018.e00893

Goyal, A., Sharma, V., Upadhyay, N., Gill, S., and Sihag, M. (2014). "Flax and flaxseed oil: An ancient medicine & modern functional food," *Journal of Food Science Technology* 51(9), 1633-1653. DOI: 10.1007/s13197-013-1247-9

Guilloux, K., Gaillard, I., Courtois, J., Courtois, B., and Petit, E. (2009). "Production of arabinoxylan-oligosaccharides from flaxseed (*Linum usitatissimum*)," *Journal of Agricultural and Food Chemistry* 57(23), 11308-11313. DOI: 10.1021/jf902212z

Hall III, C., Tulbek, M. C., and Xu, Y. (2006). "Flaxseed," in: *Advances in Food and Nutrition Research*, S. L. Taylor (ed.), Elsevier, Amsterdam, Netherlands, pp. 1-97. DOI: 10.1016/S1043-4526(06)51001-0

Hamad, Y. K., Abobakr, Y., Salem, M. Z. M., Ali, H. M., Al-Sarar, A. S., and Al-Zabib, A. A. (2019). "Activity of plant extracts/essential oils against three plant pathogenic fungi and mosquito larvae: GC/MS analysis of bioactive compounds," *BioResources* 14(2), 4489-4511. DOI: 10.15376/biores.14.2.4489-4511

Hamada, H. M., Awad, M., El-Hefny, M., and Moustafa, M. A. M. (2018). "Insecticidal activity of garlic (*Allium sativum*) and ginger (*Zingiber officinale*) oils on the cotton leafworm, *Spodoptera littoralis* (Boisd.)(Lepidoptera: Noctuidae)," *African Entomology* 26(1), 84-94. DOI: 10.4001/003.026.0084

Hassan, R. R. A. (2016). "Thermal degradation of paper: the structure changes of fibres," *Egyptian Journal of Archaeological and Restoration Studies* 6(2), 71-84. DOI: 10.21608/ejars.2016.23543

Hassan R. R. A., and Mansour M. M. A. (2018). "A microscopic study of paper decayed by *Trichoderma harzianum* and *Paecilomyces variotii*," *Journal of Polymers and the Environment* 26(7), 2698-2707. DOI: 10.1007/s10924-017-1147-6

Hassan, R. R. A. (2015). "Behavior of archeological paper after cleaning by organic solvents under heat accelerated ageing," *Mediterranean Archaeology and Archaeometry* 15(3), 141-150. DOI: 10.5281/zenodo.18365

Hassan, R. R. A. (2019). "The restoration of two historic leather bindings according to a new strategy," *Journal of the Institute of Conservation* 42(3), 210-225. DOI: 10.1080/19455224.2019.1654532

Hassan, R. R. A. (2020). "In vitro study for use of cactus gel in enhancing the mechanical strengths of vegetable tanned leathers under accelerated aging," *International Journal of Precision Engineering and Manufacturing* 21, 145-155. DOI: 10.1007/s12541-019-00178-x

Hassan, R. R. A. (2020). "Using Polaroid- zinc oxide nanocomposites in strengthening a historical printed paper: Application to "*Annales Agricoles*"- 1829 AD," *Pigment & Resin Technology*, 49(5), 369-375. DOI: 10.1108/PRT-02-2020-0012

Hassan, R. R. A., Ali, M. F., Fahmy, A.-G. A., Ali, H. M., and Salem, M. Z. M. (2020). "Documentation and evaluation of an ancient paper manuscript with leather binding using spectrometric methods," *Journal of Chemistry* 2020, 1-10. DOI: 10.1155/2020/6847910

Hassan, R. R. A., and Mohamed, W. S. (2017). "Effect of methyl methacrylate/ hydroxyethyl methacrylate copolymer on optical and mechanical properties and long-term durability of paper under accelerated ageing," *International Journal of Conservation Science* 8(2), 237-250. DOI: 10.1007/s00339-018-1989-3

Hong, J. H., Khan, N., Jamila, N., Hong, Y. S., Nho, E. Y., Choi, J. Y., Lee, C. M., and Kim, K. S. (2017). "Determination of volatile flavour profiles of *Citrus* spp. fruits by

SDE-GC-MS and enantiomeric composition of chiral compounds by MDGC-MS," *Phytochemical Analysis* 28(5), 392-403. DOI: 10.1002/pca.2686

Hussein, H. S., Salem, M. Z. M., and Soliman, A. M. (2017). "Repellent, attractive, and insecticidal effects of essential oils from *Schinus terebinthifolius* fruits and *Corymbia citriodora* leaves on two whitefly species, *Bemisia tabaci*, and *Trialeurodes ricini*," *Scientia Horticulturae* 216, 111-119. DOI: 10.1016/j.scienta.2017.01.004

Ibrahim, F. A., Usman, L. A., Akolade, J. O., Idowu, O. A., Abdulazeez, A. T., and Amuzat, A. O. (2019). "Antidiabetic potentials of *Citrus aurantifolia* leaf essential oil," *Drug Research* 69(4), 201-206. DOI: 10.1055/a-0662-5607

Kaithwas, G., and Majumdar, D. K. (2013). "Effect of *L. usitatissimum* (flaxseed/linseed) fixed oil against distinct phases of inflammation," *International Scholarly Research Notices*, 735158. DOI: 10.1155/2013/735158

Kaithwas, G., Mukerjee, A., Kumar, P., and Majumdar, D. K. (2011). "*Linum usitatis-simum* (linseed/ flaxseed) fixed oil: Antimicrobial activity and efficacy in bovine mastitis," *Inflammopharmacology* 19(1), 45-52. DOI: 10.1007/s10787-010-0047-3

Kamel, S., El-Sakhawy, M., and Nada, A. M. A. (2004). "Mechanical properties of the paper sheets treated with different polymers," *Thermochimica Acta* 421(1-2), 81-85. DOI: 10.1016/j.tca.2004.03.005

Khattab, M. A., El-Ashael, A. A., and Kandil, S. H. (1999). "Effect of contamination of cotton fabric with linseed oil on the activation energies of pyrolysis and oxidation of the fabric," *Fire Materials* 23(3), 131-137. DOI: 10.1002/(SICI)1099-1018(199905/06)23:3<131::AID-FAM678>3.0.CO;2-4

Krajčová, A., Schulzová, V., Hajšlová, J., and Bjelková, M. (2009). "Lignans in flaxseed," *Czech Journal of Food Sciences* 27(S1), 252-255. DOI: 10.17221/1062-CJFS

Ljaljević-Grbić, M., Stupar, M., Vukojević, J., Maričić, I., and Bungur, N. (2013). "Molds in museum environments: biodeterioration of art photographs and wooden sculptures," *Archives of Biological Sciences*, 65(3), 955-962. DOI: 10.2298/abs1303955g

Le Moigne, N. (2008). *Swelling and Dissolution Mechanisms of Cellulose Fibres*, Ph.D. Dissertation, École Nationale Supérieure des Mines de Paris, Paris, France.

Lemes, R. S., Alves, C. C. F., Estevam, E. B. B., Santiago, M. B., Martins, C. H. G., Santos, T. C. D., Crotti, A. E. M., and Miranda, M. L. D. (2018). "Chemical composition and antibacterial activity of essential oils from *Citrus aurantifolia* leaves and fruit peel against oral pathogenic bacteria," *Anais da Academia Brasileira de Ciências* 90(2), 1285-1292. DOI: 10.1590/0001-3765201820170847

Madera-Santana, T. J., Aguilar-Vega, M. J., Márquez-Lucero, A., and Vázquez-Moreno, F. (2002). "Production of leather-like composites using chemically modified short leather fibers. I: Chemical modification by emulsion polymerization," *Polymer Composites* 23(1), 49-60. DOI: 10.1002/pc.10411

Mansour, M. M. A., EL-Hefny, M., Salem, M. Z. M., and Ali, H. M. (2020a). "The biofungicide activity of some plant essential oils for the cleaner production of model linen fibers similar to those used in ancient Egyptian mummification," *Processes* 8(1), 1-19. DOI: 10.3390/pr8010079

Mansour, M. M. A., Hamed, S. A. E.-K. M., Salem, M. Z. M., and Ali, H. M. (2020b). "Illustration of the effects of five fungi on *Acacia saligna* wood organic acids and ultrastructure alterations in wood cell walls by HPLC and TEM examinations," *Applied Sciences* 10(8), 1-14. DOI: 10.3390/app10082886

Massoud, M. A., Saad, A. S. A., Soliman, E. A., and El-Moghazy, A. Y. (2012). "Antifungal activity of some essential oils applied as fumigants against two stored grains fungi," *J. Adv. Agric. Res. (Fac Agric Saba Basha)*, 17(2), 296-306.

Matasyoh, J. C., Kiplimo, J. J., Karubiu, N. M., and Hailstorks, T. P. (2007). "Chemical composition and antimicrobial activity of essential oil of *Tarchonanthus camphorates*," *Food Chemistry* 101(3), 1183-1187. DOI: 10.1016/j.foodchem.2006.03.021

Mohamed, A. A., Behiry, S. I., Ali, H. M., EL-Hefny, M., Salem, M. Z. M., and Ashmawy, N. A. (2020a). "Phytochemical compounds of branches from *P. halepensis* oily liquid extract and *S. terebinthifolius* essential oil and their potential antifungal activity," *Processes* 8(3), 1-18. DOI: 10.3390/pr8030330

Mohamed, A. A., El-Hefny, M., El-Shanhorey, N. A., and Ali, H. M. (2020b). "Foliar application of bio-stimulants enhancing the production and the toxicity of *Origanum majorana* essential oils against four rice seed-borne fungi," *Molecules* 25(10), 1-19. DOI: 10.3390/molecules25102363

Moufida, S., and Marzouk, B. (2003). "Biochemical characterization of blood orange, sweet orange, lemon, bergamot and bitter orange," *Phytochemicals* 62(8), 1283-1289. DOI: 10.1016/S0031-9422(02)00631-3

Mueller, K., Eisner, P., Yoshie-Starc, Y., Nakada, R., and Kirchhoff, E. (2010). "Functional properties and chemical composition of fractionated brown and yellow linseed meal (*Linum usitatissimum* L.)," *Journal of Food Engineering* 98(4), 453-460. DOI: 10.1016/j.jfoodeng.2010.01.028

Narang, N., and Jiraungkoorskul, W. (2016). "Anticancer activity of key lime, *Citrus aurantifolia*," *Pharmacognosy Reviews* 10(20), 118-122. DOI: 10.4103/0973-7847.194043

Narender, B. R., Tejaswini, S., Sarika, M., Karuna, N., Shirisha, R., and Priyanka, S. (2016). "Antibacterial and antifungal activities of *Linum usitatissimum* (flax seeds)," *International Journal of Pharmacy Education and Research* 3(2), 4-8.

Nasr, H. E., and Ismail, A. (2010). "Improving the leather performance via treatment with different adducts and grafting with 1-vinyl-2-pyrrolidinone," *New York Science Journal* 3(9), 112-119.

Okla, M. K., Alamri, S. A., Alatar, A. A., Hegazy, A. K., Al-Ghamdi, A. A., Ajarem, J. S., Faisal, M., Abdel-Salam, E. M., Ali, H. M., Salem, M. Z. M., *et al.* (2019a). "Antioxidant, hypoglycemic, and neurobehavioral effects of a leaf extract of *Avicennia marina* on autoimmune diabetic mice," *Evidence-Based Complementary and Alternative Medicine* 2019, 1-8. DOI: 10.1155/2019/1263260

Okla, M. K., Alamri, S. A., Salem, M. Z. M., Ali, H. M., Behiry, S. I., Nasser, R. A., Alaraidh, I. A., Al-Ghtani, S. M., and Soufan, W. (2019b). "Yield, phytochemical constituents, and antibacterial activity of essential oils from the leaves/twigs," branches, branch wood, and branch bark of Sour Orange (*Citrus aurantium* L.). *Processes* 7(6), 1-15. DOI: 10.3390/pr7060363

Paiva, P. M. G., Gomes, F. S., Napoleão, T. H., Sá, R. A., Correia, M. T. S., and Coelho, L. C. B. B. (2010). "Antimicrobial activity of secondary metabolites and lectins from plants," in: *Current Research, Technology and Education Topics in Applied Microbiology and Microbial Biotechnology,* A. Méndez-Vilas (ed.), Formatex Research Center, Badajoz, Spain, pp. 396-406.

Pathan, R. K., Gali, P. R., Pathan, P., Gowtham, T., and Pasupuleti, S. (2012). "*In vitro* antimicrobial activity of *Citrus aurantifolia* and its phytochemical screening," *Asian*

Pacific Journal of Tropical Disease 2(1), 328-331. DOI: 10.1016/S2222-1808(12)60176-5
Pinheiro, A. C., Sequeira, S. O., and Macedo, M. F. (2019). "Fungi in archives, libraries, and museums: a review on paper conservation and human health," *Critical Reviews in Microbiology*, 45(5-6), 686-700. DOI: 10.1080/1040841X.2019.1690420
Piper, P., Calderon, C. O., Hatzixanthis, K., and Mollapour, M. (2001). "Weak acid adaptation: The stress response that confers resistance to organic acid food presser-vatives," *Microbiology* 147(10), 2635-2642. DOI: 10.1099/00221287-147-10-2635
Popa, V.-M. , Gruia, A., Raba, D.-N., Dumbrava, D., Moldovan, C., Bordean, D., and Mateescu, C. (2012). "Fatty acids composition and oil characteristics of linseed (*Linum usitatissimum* L.) from Romania," *Journal of Agroalimentary Processes and Technologies* 18(2), 136-140.
Rammanee, K., and Hongpattarakere, T. (2011). "Effects of tropical citrus essential oils on growth, aflatoxin production, and ultrastructure alterations of *Aspergillus flavus* and *Aspergillus parasiticus*," *Food and Bioprocess Technology* 4(6), 1050-1059. DOI: 10.1007/s11947-010-0507-1
Razzaghi-Abyaneh, M., Shams-Ghahfarokhi, M., Rezaee, M.-B., Jaimand, K., Alinezhad, S., Saberi, R., and Yoshinari, T. (2009). "Chemical composition and antiaflatoxigenic activity of *Carum carvi* L., *Thymus vulgaris* and *Citrus aurantifolia* essential oils," *Food Control* 20(11), 1018-1024. DOI: 10.1016/j.foodcont.2008.12.007
Rudnik, E., Szczucinska, A., Gwardiak, H., Szulc, A., and Winiarska, A. (2001) "Comparative studies of oxidative stability of linseed oil," *Thermochimica Acta* 370(1-2), 135-140. DOI: 10.1016/S0040-6031(00)00781-4
Salem, M. Z. M., Zayed, M. Z., Ali, H. M., and Abd El-Kareem, M. S. M. (2016a). "Chemical composition, antioxidant and antibacterial activities of extracts from *Schinus molle* L. wood branch growing in Egypt," *Journal of Wood Science* 62(6), 548-561. DOI: 10.1007/s10086-016-1583-2
Salem, M. Z. M., Zidan, Y. E., Mansour, M. M. A., El Hadidi, N. M. N., and Abo Elgat, W. A. A. (2016b). "Antifungal activities of two essential oils used in the treatment of three commercial woods deteriorated by five common mold fungi," *International Biodeterioration & Biodegradation* 106(1), 88-96. DOI: 10.1016/j.ibiod.2015.10.010
Salem, M. Z. M., Mansour, M. M. A., Mohamed, W. S., Ali, H. M., and Hatamleh, A. A. (2017). "Evaluation of the antifungal activity of treated *Acacia saligna* wood with Paraloid B-72/TiO_2 nanocomposites against the growth of *Alternaria tenuissima*, *Trichoderma harzianum*, and *Fusarium culmorum*," *BioResources* 12(4), 7615-7627. DOI: 10.15376/biores.12.4.7615-7627
Salem, M. Z. M., Behiry, S. I., and EL-Hefny, M. (2019a). "Inhibition of *Fusarium culmorum*, *Penicillium chrysogenum* and *Rhizoctonia solani* by n-hexane characterized extracts of three plant species as a wood-treated oil-fungicide model," *Journal of Applied Microbiology* 126(6), 1683-1699. DOI: 10.1111/jam.14256
Salem, M. Z. M., Hamed, S. A. E.-K. M, and Mansour, M. M. A. (2019b). "Assessment of efficacy and effectiveness of some extracted bio-chemicals as bio-fungicides on wood," *Drvna Industrija* 70(4), 337-350. DOI: 10.5552/drvind.2019.1837
Salem, M. Z. M., Abo Elgat, W. A. A., Taha, A. S., Fares, Y. G. D., and Ali, H. M. (2020a). "Impact of three natural oily extracts as pulp additives on the mechanical, optical, and antifungal properties of paper sheets made from *Eucalyptus camaldulensis* and *Meryta sinclairii* wood branches," *Materials* 13(6), 1-27. DOI: 10.3390/ma13061292

Salem, M. Z. M., Ali, M. F., Mansour, M. M. A., Ali, H. M., Abdel Moneim, E. M., and Abdel-Megeed A. (2020b). "Anti-termitic activity of three essential oils, chlorpyrifos, and a bioagent compound (Protecto) against termite *Microcerotermes eugnathus* Silvestri (Isoptera: Termitidae) in Egypt," *Insects* 11(11), 756. DOI: 10.3390/insects11110756

Salem, M. Z. M., Ibrahim, I. H., Ali, H. M., and Helmy, H. M. (2020c). "Assessment of the use of natural extracted dyes and pancreatin enzyme for dyeing of four natural textiles: HPLC analysis of phytochemicals," *Processes* 8, 59. DOI:10.3390/pr8010059

Salim, E., Abdel-Hamied, M., Salim, S., Gamal, S., Mohamed, S., Galal, F. E.-Z., Tarek, F., Hassan, R. R. A., Ali, H. M., and Salem, M. Z. M. (2020). "Reduction of borax / agar-based gel residues used to neutralize acidity of a historical manuscript with use of different paper barriers: Artificial ageing results," *BioResources* 15(3), 6576-6599. DOI: 10.15376/biores.15.3.6576-6599

SAS (2001). *User Guide: Statistics* (Release 8.02), SAS Institute, Cary, NC.

Seidel, V., and Taylor, P. W. (2004). "In vitro activity of extracts and constituents of *Pelagonium* against rapidly growing mycobacteria," *International Journal of Antimicrobial Agents* 23(6), 613-619. DOI: 10.1016/j.ijantimicag.2003.11.008

Sequeira, S. O., Cabrita, E. J., and Macedo, M. F. (2014). "Fungal biodeterioration of paper: How are paper and book conservators dealing with it?," *Restaurator. International Journal for the Preservation of Library and Archival Material* 35(2), 181-199. DOI: 10.1515/rest-2014-0005

Shim, Y. Y., Gui, B., Arnison, P. G., Wang, Y., and Reaney, M. J. T. (2014). "Flaxseed (*Linum usitatissimum* L.) bioactive compounds and peptide nomenclature: A review," *Trends in Food Science & Technology* 38(1), 5-20. DOI: 10.1016/j.tifs.2014.03.011

Simopoulos, A. P. (2002). "The importance of the ratio of omega-6/omega-3 essential fatty acids," *Biomedicine & Pharmacotherapy* 56(8), 365-379. DOI: 10.1016/S0753-3322(02)00253-6

Soković, M., and van Griensven, L. J. L. D. (2006). "Antimicrobial activity of essential oils and their components against the three major pathogens of cultivated button mushroom *Agaricus bisporus*," *European Journal of Plant Pathology* 116, 211-224. DOI: 10.1007/s10658-006-9053-0

Spadaro, F., Costa, R., Circosta, C., and Occhiuto F. (2012). "Volatile composition and biological activity of Key lime *Citrus aurantifolia* essential oil," N*atural Product Communications* 7(11), 1523-1526. DOI: 10.1177/1934578X1200701128

Tundis, R., Loizzo, M. R., Bonesi, M., Menichini, F., Mastellone, V., Colica, C., and Menichini, F. (2012). "Comparative study on the antioxidant capacity and cholinesterase inhibitory activity of *Citrus aurantifolia* Swingle, *C. aurantium* L., and *C. bergamia* Risso and Poit. peel essential oils," *Journal of Food Science* 77(1), 40-46. DOI: 10.1111/j.1750-3841.2011.02511.x

Velázquez-Nuñez, M. J., Avila-Sosa, R., Palou, E., and López-Malo, A. (2013). "Antifungal activity of orange (*Citrus sinensis* var. Valencia) peel essential oil applied by direct addition or vapor contact," *Food Control* 31(1), 1-4. DOI: 10.1016/j.foodcont.2012.09.029

Viuda-Martos, M., Ruiz-Navajas, Y., Fernández-López, J. and Pérez-Álvarez, J. (2008). "Antifungal activity of lemon (*Citrus lemon* L.), mandarin (*Citrus reticulata* L.), grapefruit (*Citrus paradisi* L.) and orange (*Citrus sinensis* L.) essential oils," *Food Control* 19(12), 1130-1138. DOI: 10.1016/j.foodcont.2007.12.003

Wang, Z.-J., Liang, C.-L., Li, G.-M., Yu, C.-Y., and Yin, M. (2007). "Stearic acid protects primary cultured cortical neurons against oxidative stress 4," *Acta Pharmacologica Sinica* 28(3), 315-326. DOI: 10.1111/j.1745-7254.2007.00512.x
Xu, Y., Hall III, C., and Wolf-Hall, C. (2008). "Antifungal activity stability of flaxseed protein extract using response surface methodology," *Journal of Food Science* 73(1), 9-14. DOI: 10.1111/j.1750-3841.2007.00576.x
Xu, Y., Hall III, C., Wolf-Hall, C., and Manthey, F. (2008). "Fungistatic activity of flaxseed in potato dextrose agar and a fresh noodle system," *International Journal of Food Microbiology* 121(3), 262-267. DOI: 10.1016/j.ijfoodmicro.2007.11.005
Youssef, A. M., Kamel, S., El-Sakhawy, M., and El Samahy, M. A. (2012). "Structural and electrical properties of paper–polyaniline composite," *Carbohydrate Polymers* 90(2), 1003-1007. DOI: 10.1016/j.carbpol.2012.06.034
Zyani, M., Mortabit, D., Mostakim, M., Iraqui, M., Haggoud, A., Ettayebi, M., and Koraichi, S. I. (2009). "Cellulolytic potential of fungi in wood degradation from an old house at the Medina of Fez," *Annals of Microbiology* 59(4), 699-704. DOI: 10.1007/BF03179211

Article submitted: Sept. 18, 2020; Peer review completed: Nov. 15, 2020; Revised version received and accepted: November 22, 2020; Published: November 24, 2020.
DOI: 10.15376/biores.16.1.492-514