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A comparative study on stability of different types of coconut (*Cocos nucifera*) oil against autoxidation and photo-oxidation

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This study aimed to compare the stability of five types of coconut (*Cocos nucifera*) oil available in Sri Lanka. The types of coconut oil studied were coconut pairing oil (CPO), white coconut oil (WCO), refined bleached and deodorized coconut oil (RBDCO), virgin coconut oil (VCO) and coconut oil (CO). Oils were exposed to elevated temperature (60±5°C) in the presence of excess air or oxygen to induce autoxidation and florescent light (2650 lux) to induce photo-oxidation in 28 days of storage. Samples were taken on 0, 1, 3, 5, 7, 14, 21 and 28 days to assess the level of oxidation by peroxide value (PV), conjugated dienes (CD), conjugated trienes (CT) and thiobarbituric acid reactive substances (TBARS) with time. WCO and VCO possessed the highest oxidative stability against autoxidation followed by CO and CPO, while, RBDCO showed the lowest stability. WCO and VCO possessed highest stability against photo-oxidation. CO was more susceptible to photo-oxidation among the oils examined followed by CPO and RBDCO. WCO and VCO possessed similar stability against both autoxidation and photo-oxidation. VCO has highest monounsaturated fatty acid: polyunsaturated fatty acid ratio (4.28) and suitable n6/n3 ratio (5:1); hence applicable as whole purpose oil which is resistant to processing and storage conditions with good nutritive value.

Key words: Autoxidation, coconut oil, fluorescent, oxidative stability, photo-oxidation.

INTRODUCTION

Coconut (*Cocos nucifera*) is one of the major plantation crops cultivated in Sri Lanka over many decades and coconut oil is the widely used edible oil in the country. Coconut fat accounts for 80% of fat intake among Sri Lankans (Amarasiri and Dissanayake, 2006). In 2015, the total area under coconut cultivation was approximately 455,000 ha which produced almost 3,056 million nuts, while coconut oil production was approximately 52,790 MT (Central Bank of Sri Lanka, 2017). Coconut oil is the major dietary source of medium chain triacylglycerols (MCTs). The MCTs are quickly absorbed and utilized when as consumed compared to long chain triacylglycerols

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(Krishna et al., 2010). MCTs may reduce the incorporation and storage of dietary fats and oil in adipose tissue, thus, reducing body fat (Adhikari et al., 2010). In addition, coconut oil is considered to be the most stable for deep frying because of high level of saturation as compared to other edible oils. Therefore, recently, there is a trend to encourage the use of coconut oil for deep frying to reduce the health risks arising from *trans* fats (Manchanda and Passi, 2016).

Oxidative stability is an important indicator to determine quality and shelf life of edible oils (Choe and Min, 2006). Most plant oils in their natural forms do not possess sufficient oxidative stability, which limits their application for food processing and long term storage (Comandini et al., 2009). Lipid oxidation causes deterioration of the oils leading to loss of essential fatty acids and alteration of sensory qualities such as flavour, aroma and colour; thus reducing palatability. Moreover, oxidation of lipids produces primary and secondary oxidative products such as peroxides, hydroperoxides, aldehydes, ketones, acids and alcohols which adversely affect the sensory quality, nutritive value and safety of the oil (Choe and Min, 2006; Vidrih et al., 2010).

Chemical mechanisms such as autoxidation and photo-oxidation are responsible for the oxidation of lipids during processing and storage (Choe and Min, 2006). Autoxidation and photo-oxidation of lipids are influenced by heat, light, composition of fatty acids, type of oxygen and minor compounds such as metals, pigments, phospholipids, free fatty acids, mono- and di-acylglycerols, thermally oxidized compounds and antioxidants. In contrast, phenolic compounds, especially carotenoids, decrease autoxidation in oil, while tocopherols, chlorophylls and phospholipids exhibit both antioxidant and pro-oxidant activity depending on the oil system and storage conditions (Shahidi and Zhong, 2005; Choe and Min, 2006).

Triplet ($^3O_2$) and singlet ($^1O_2$) oxygen act as the initiators of oxidation of the lipids (Ahmed et al., 2016). Unsaturated fatty acids undergo autoxidation, which is initiated by triplet oxygen via the free radical chain reaction through attack on alpha methylene of the carbon double bonds leading to the formation of hydroperoxides (primary oxidation product), which are further decomposed to form secondary oxidation products (Rukmini and Raharjo, 2010). Lipid autoxidation involves three steps: initiation, propagation and termination stages. In the initiation step, when lipid is exposed to an initiator (heat, light or metal ions), hydrogen atom of double bond is abstracted and alkyl radical is formed. This free radical abstracts hydrogen from other lipid molecules and reacts with the hydrogen to form hydroperoxide, which is a tasteless and odourless compound and another alkyl radical. Alkyl radical also reacts with triplet oxygen to form peroxy radical, which abstracts hydrogen from other lipid molecules and reacts with hydrogen to form hydroperoxide and another alkyl radical. Radicals react with each other to form non-radical species and the reaction is terminated (Choe and Min, 2006; Lee et al., 2004). Hydroperoxide further break down via several steps into secondary oxidation products such as carbonyl compounds responsible for objectionable odour and taste such as aldehyde, ketones, acids, alcohols, acids, esters and short-chain hydrocarbons via monomolecular and bimolecular reactions (Choe and Min, 2006).

Singlet oxygen initiates photo-oxidation via the direct attack of the extremely electrophilic singlet oxygen on the unsaturated fatty acids. In the presence of light, photosensitizers such as chlorophyll and porphyrin convert triplet oxygen into singlet oxygen, which is a highly reactive non-radical molecule. If wavelength of solar light is less than 220 nm, unsaturated fatty acids cannot absorb light; however, photosensitizers can absorb light energy and convert triplet state sensitizer to singlet state sensitizer (Ahmed et al., 2016). Two types of photo-oxidation mechanisms have been proposed; an electron or a hydrogen atom transfers between an excited triplet sensitizer and a substrate, producing free radicals or radical ions; and triplet oxygen can be excited by light to singlet oxygen, which reacts with the double bond of unsaturated fatty acids, producing an allylic hydroperoxide (Gordon, 2001; W¹sowicz et al., 2004; Song et al., 2007; Galano et al., 2015; Ahmed et al., 2016). The autoxidation is comparatively a slower process than photo-oxidation (Rukmini and Raharjo, 2010).

Although, the oxidative stability of some oils is higher compared to other edible oils due to lower amounts of polyunsaturated fatty acids and presence of natural antioxidants, natural photosensitizers in oils such as chlorophyll may initiate photo-oxidation (Kim et al., 2000). Coconut oil is an example for saturated oils. Thus, greater concern should be given to photo-oxidation than auotoxidation of coconut oil. Generally, the oils are packaged in transparent plastic or glass bottles during storage display of retail markets. The opacity of the packaging material to light is of fundamental importance to minimize the deterioration of edible oils by photo-oxidation (Méndez and Falqué, 2007). Therefore, prevention of light exposure during storage of vegetable oils is necessary to preserve the quality and extend its shelf-life (Gargouri et al., 2015).

Different types of coconut oil are available in Sri Lanka. Coconut pairing oil (CPO) is extracted from fresh pairings resulting from the desiccated coconut, coconut cream and coconut milk industry and contains free fatty acid content of maximum 0.8%. Virgin coconut oil (VCO) is manufactured from fresh kernel either through dry or wet process without high heat treatment. Coconut oil (CO) is prepared from traditional copra (dried coconut kernel). Refined and bleached and deodorized coconut oil (RBDCO) is obtained from crude coconut oil by refining process. White coconut oil (WCO) is obtained from best

quality copra and contains free fatty acid percentage of less than 0.8%. These oils may differ in their susceptibility to oxidation because of the processing methods which may lead to differences in the presence of minor components. For example, chemical refining may remove major proportion of natural antioxidants (Ayyildiz et al., 2015). Thus, the knowledge on the stability of different types of coconut oil for autoxidation and photo-oxidation will be useful to suit the oil type for different cooking or processing techniques and to select suitable packaging materials to extend their shelf life. In this backdrop, the aim of this study was to examine and compare the stability of five types of oils from coconut available in Sri Lankan market against autoxidation and photo-oxidation in order to understand the impact of storage conditions such as temperature and florescent light on the oxidation of selected oils and to select the oil which is most resistant to oxidation during processing and storage.

MATERIALS AND METHODS

The study was carried out at the Department of Food Science and Technology, Faculty of Agriculture, University of Peradeniya, Sri Lanka.

All oil samples were collected from a local oil mill within three days after manufacture. It was ensured that the oils do not contain any additive. Oil samples in glass bottles wrapped with aluminum foil were stored at -20°C after flushed with nitrogen. After an oil sample was drawn for a particular analysis, the bottle was flushed with nitrogen again, covered with the original cap and sealed with parafilm. All the chemicals used in this study were of analytical grade with highest purity available (>99.5%) and obtained from Sigma Chemicals Company (MO, USA).

Determination of fatty acid composition of oils

Fatty acids profile of selected oils was determined by gas-liquid chromatography (GLC). Fatty acid methyl esters (FAMEs) were prepared according to the method explained by Christie (1992). To prepare FAMEs, 3 mL of 0.5 M sodium methoxide and 0.3 mL of dichloromethane were added to the 0.2 g of oil sample. The mixture was kept at 50°C for 30 min. The mixture was allowed to cool to room temperature and 5 mL of deionized water was added. Then, 0.1 mL of acetic acid and 0.5 mL of hexane was added and centrifuged at 1500 rpm for 10 min at 5°C. The distinct upper layer of hexane containing FAMEs was separated for analysis. Analysis of FAMEs was carried out on GLC (GC-14B, Shimadzu, Japan) equipped with Flame Ionization Detector (FID) and capillary column (SP^{TM} 2560, 100 m × 0.25 mm ID, 0.20 µm film). The initial column oven temperature was maintained at 140°C for 5 min and increased to 220°C at the rate of 4°C/min, then maintained at that temperature for 10 min. Both injector and detector temperatures were maintained at 260°C. Helium was used as the carrier gas at flow rate of 30 mL/min. The injection volume was 1 µL at split ratio of 100:1. Fatty acids were identified by comparison of their retention time with authentic standards (Supelco^{TM} 37 component FAME mix).

Extraction of phenolic fraction

Phenolic fraction present in 50 g of oil was extracted into methanol by passing oil through a gravity column packed with silica (60-Å pore diameter) according to the method described by Steel et al. (2005). Hexane and methanol (1:1) mixture was used for conditioning the column and hexane and ethyl acetate (9:1) mixture was used for washing the column. The oil sample thoroughly mixed with hexane was introduced to the column and the phenoilc fraction bound to silica was subsequently extracted into methanol. The extract was recovered after desolventizing *in vacuo* at 40°C. The prepared extracts were stored under frozen condition (-20°C) after flushing with nitrogen. The frozen extracts were thawed and appropriately diluted before chemical analysis.

Determination of total phenolic content

The total phenolic content (TPC) of the extracts was determined colorimetrically using Folin-Ciocalteu's reagent as described by Thaipong et al. (2006) with minor modifications. Twenty microliters of oil extract and 1.58 mL of deinozed water were mixed with 100 µL of Folin-Ciocalteu's reagent, left for 3 min and subsequently 300 µL of sodium carbonate (0.7 M) was added and vortexed. The absorbance of the resulting mixture was read at 725 nm using a UV visible spectrophotometer (UV 1601, Shimadzu, Japan) after leaving for 30 min at room temperature (27°C). The results were expressed as mg of gallic acid equivalents (GAE) per liter of extract using a gallic acid (50 to 500 mg/L) standard curve.

Determination of oxidative stability of oils

Oxidative stability of selected oils was examined by accelerated oxidative stability tests. The rate of oxidation was monitored by the measurement of peroxide value (PV), conjugated dienes (CD), conjugated trienes (CT) and thiobarbituric acid reactive substances (TBARS) with the time.

Preparation of samples for *Schaal* oven test method

The oxidative stability of the investigated oil samples was studied using the *Schaal* oven accelerated oxidation test (Shahidi et al., 1997). The test was performed in an oven at constant temperature. Sixteen samples (5 mL) of each oil type were placed in glass vials (2 cm ID × 4.5 cm) and kept in the oven at the temperature of 60±5°C for up to 28 days without lids to facilitate the oxidation. Two samples from each oil type were drawn on days 0, 1, 3, 5, 7, 14, 21 and 28, and the level of oxidation of oil samples was analyzed.

Preparation of samples for accelerated photo-oxidation method

The stability against light induced oxidation of selected oils was evaluated under fluorescent light (2650 lux) (Khan and Shahidi, 1999) up to 28 days of storage period. Sixteen samples (5 mL) of each oil type were placed in glass vials (2 cm ID × 4.5 cm) and glass vials were placed in a polypropylene box (70 cm length x 35 cm width x 25 cm height) equipped with 80 Watt cool white fluorescent lights fixed on the lid above the surface of the oil containers. The remaining open space of the box was covered with aluminum foil. The fluorescent radiation was at a level of 2650 Lux and the temperature inside the container was maintained at 27±1°C. Two samples from each oil type were drawn from the box on days 0, 1, 3, 5, 7, 14, 21 and 28 to assess the level of oxidation.

Determination of PV

PV of the oil samples was measured according to the modified IDF

method (Hornero-Méndez et al., 2001). The sample (0.01 to 0.05 g) was dissolved in 1 mL of chloroform/acetic acid (2:3), with addition of 100 µL Fe (II) solution, mixed for 15 s in a vortex mixer and left in the dark for 10 min. Deionized water (2 mL) was added and 4 mL of diethyl ether was added. Organic phase was discarded and remaining ether in the aqueous phase was removed under N_2 current for a few seconds. Aqueous phase (1 mL) was transferred into another tube and 100 µL of saturated ammonium thiocyanate solution was added. After 10 min, absorbance at 470 nm was read against water blank using a UV visible spectrophotometer (UV 1601, Shimadzu, Japan). A reaction blank also prepared. PV was calculated and the results were expressed as meq/kg of oil using a Fe (III) solution (0 to 40 µg/mL) calibration curve.

Determination of TBARS

The sample of oil (0.05 to 0.10 g) was thoroughly mixed with 5 mL 1-butanol solution and 5 mL of 0.2% (w/v) thiobarbituric acid (TBA) in 1-butanol solution was added. Prepared solution was incubated for 2 h in a water bath maintained at 95°C and cooled immediately. Absorbance of the solution was taken at 532 nm using a UV visible spectrophotometer (UV 1601, Shimadzu, Japan). The results were expressed as mg of malonaldehyde equivalent/g of oil using TMP (1×10^{-6} - 1×10^{-5} M) as standard (Yi et al., 2011).

Determination of CD and CT

CD and CT values were determined by IUPAC II.D.23 analytical method (Paquot, 1979). Oil sample (0.01 to 0.03 g) was thoroughly mixed with 25 mL of iso-octane using vortex mixer for 15 s. Then, the absorbances were measured separately at 233 nm for CD and 268 nm for CT using a UV visible spectrophotometer (UV 1601, Shimadzu, Japan). The results were calculated as extinction values ($E_{1\%}$) using following equation.

$$E_{1\%} = \frac{A_\lambda}{(C\times l)}$$

Where, A_λ is the absorbance measured at either 233 or 268 nm; C represent the concentration of oil solution (g/100 mL); l is the path length of the cuvette (cm).

Statistical analysis

The data were analyzed using Minitab 16 (Minitab Inc., UK) and Microsoft (Excel) procedures. All measurements were performed in triplicate and results are expressed as mean ± SD. The ANOVA tables were constructed using GLM procedure. Duncan's new multiple range test was used to determine significant differences at 0.05 significant levels.

RESULTS AND DISCUSSION

PV of all oils ranged between 0.21 and 0.42 meq/Kg. As per the Codex Standard (Codex Alimentarius, 1999), the maximum limit of peroxide values for any refined oil and virgin oils are 10 and 20 meq/kg, respectively. Thus, initial quality of the oil sample used in this study complied with this standard.

Fatty acid composition of oils

The fatty acid composition of oils is presented in Table 1. All oil samples were characterized by five to seven different types of saturated fatty acids of chain length C6 to C22 and three to six different types of unsaturated fatty acids of chain length C18 to C22. Palmitic acid (C16:0) and stearic acid (C18:0) were common in all saturates.

The amount of saturated fatty acid of the selected oils ranged from 87 to 93%. Lauric acid (43-52%) was most predominant followed by myristic acid (16-18%). Moreover, oils contained considerable amounts of short chain fatty acids such as caproic, caprylic and capric. The average content of oleic acid and lauric acid were significantly higher in WCO and VCO as compared to that of other coconut oils tested. Results of this study appeared identical with those of earlier published findings in the context of major fatty acids of the respective oils (Vidrih et al., 2010). It is recommended that the *n*-6/*n*-3 fatty acid ratio in the diet should be between 4:1-5:1 and 10:1 (Candela et al., 2011). The most suitable ratios were found in all types except CO (Table 1).

In addition to fatty acid composition, the presence of minor components in edible oils contributes to their oxidative stability (Bendini et al., 2009; Shinagawa et al., 2017). Due to this reason, even though all types of coconut oils selected for this study contained more than 87% of saturated fatty acid, oxidative stability of these oils may differ. Table 2 shows the total phenolic content of the oils. WCO contained highest amount of total phenolic content followed by VCO, whereas, CPO and RBDCO had the lowest. Lowest level of total phenolic content of RBDCO indicates that refining process reduced the phenolic content. VCO, WCO and CO did not undergo refining chemical refining process. However, the lower total phenolic content of the CO than VCO and WCO could be due to the heat applied during the processing. Szydłowska-Czerniak and Łaszewska (2015) reported that the refining process of rapeseed oils reduced the antioxidant capacity by about 60% and total phenolic content by above 80%.

Oxidative stability of oils against autoxidation

Accelerated storage tests have been used extensively in researches to evaluate the stability of edible oils against oxidation. In this test, the oxidation of oils is accelerated by keeping the oil at 60 to 65°C. By increasing the temperature of storage, the oxidation rate is increased (accelerated). One day of storage at this condition is equal to one month of storage at ambient temperature (Evans et al., 1973). Therefore, the present study has evaluated the stability of oils for 28 months of storage at the ambient temperature.

Table 3 summarized the rate of increment of PV, CD, CT and TBARS per day of the oils tested to study their

Table 1. Percentage of saturated (SFA), monounsaturated (MUFA) and polyunsaturated (PUFA) fatty acids of each type of oil.

Fatty acid	CPO	WCO	RBDCO	VCO	CO
SFA	92.29[a]	90.72[a]	92.86[a]	87.88[b]	91.36[a]
MUFA	4.19[d]	6.81[b]	5.18[c]	9.76[a]	5.82[c]
PUFA	2.81[a]	2.45[b]	1.37[c]	2.28[b]	2.83[a]
MUFA/PUFA	1.75[d]	2.78[c]	3.78[b]	4.28[a]	2.06[c]
n-6/*n*-3	9:1	4:1	4:1	5:1	24:1

Values (means) with different superscript letters in the same row indicate significant differences ($p < 0.05$). CPO, coconut pairing oil; WCO, white coconut oil; RBDCO, refined bleached and deodorized coconut oil; VCO, virgin coconut oil; CO, coconut oil; SFA, saturated fatty acids, MUFA, monounsaturated fatty acids; PUFA, polyunsaturated fatty acids.

Table 2. Total phenolic content of oils.

Oil	TPC (mg GAE/ g extract)
WCO	28.37 ± 0.46[a]
VCO	24.52 ± 0.34[b]
CO	20.40 ± 0.41[c]
CPO	12.78 ± 0.03[d]
RBDCO	12.64 ± 0.28[d]

Values (means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

Table 3. Rate of value increment (per day) of PV, CD, CT and TBARS of different oils stored at 60±5°C during storage period.

Parameter	CPO	WCO	RBDCO	VCO	CO
PV	0.471	0.044	0.044	0.029	0.030
CD	0.025	0.034	0.044	0.022	0.021
CT	0.013	0.017	0.025	0.022	0.035
TBARS	0.163	0.064	0.199	0.112	0.127

oxidative stability against autoxidation. PV is an indicator of the extent of primary oxidative products formed in oils (Yi et al., 2011). The effect of storage condition at 60±5°C on the formation of primary oxidative products is expressed as PV. The PV of the oils with storage time is shown in the Figure 1 and the fold increment of PV with duration of storage is presented in Table 4. Development of PV was observed in all samples except VCO during the experimental period. Level of oxidation gradually increased with the storage time in all oil samples. However, rate of formation of hydroperoxides decreased with extent of duration. Li et al. (2014) reported that the higher rate of increment during early storage, decrease the rate during the latter period during the storage of oil blends under accelerated condition for 24 days. Instability of hydroperoxides under higher degree of oxidation could be the reason for lower detection values. In the later stages of lipid oxidation, the hydroperoxides are broken down into secondary oxidative products (Ullah et al., 2003). Autoxidation is affected by temperature. Since these oils were stored at 60±5°C, which might have accelerated autoxidation and rapid production of secondary oxidative products. The PV varied among oils due to different oxidative stabilities of the oils. During the first three days of storage, no significant decrease ($p>0.05$) in level of the primary oxidative products was observed in WCO and VCO. The induction period of the oils that contained considerable value of MUFA: PUFA ratio and high antioxidant activity is high. The rapid oxidation that occurred in CPO and RBDCO indicated that the oxidative stability deteriorated quickly during autoxidation. Based on the PV, highest oxidative stability

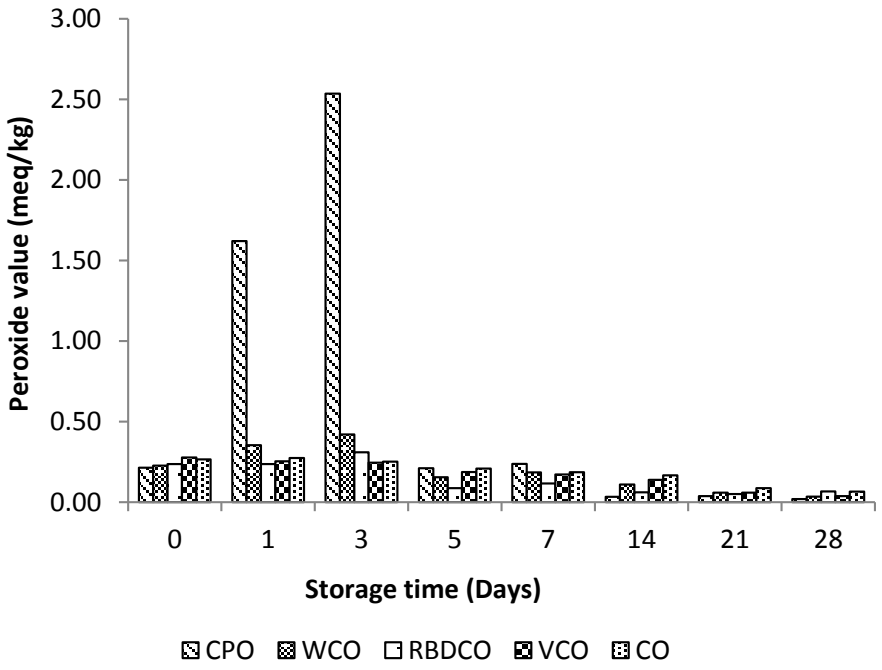

Figure 1. Peroxide value of oils stored under *Schaal* oven conditions at 60±5°C.

Table 4. Fold increment of PV of different oils stored at 60±5°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	7.58±0.20^{b}	1.56±0.01^{a}	1.00±0.02^{b}	0.91±0.01^{a}	1.03±0.03^{a}
3	11.85±0.37^{a}	1.19±0.44^{ab}	1.31±0.01^{a}	0.88±0.01^{a}	0.94±0.01^{b}
5	0.98±0.05^{c}	0.37±0.02^{c}	0.37±0.01^{d}	0.67±0.01^{b}	0.78±0.01^{bc}
7	1.11±0.07^{c}	1.20±0.02^{bc}	0.49±0.01^{c}	0.62±0.01^{c}	0.70±0.01^{b}
14	0.16±0.01^{d}	0.59±0.02^{c}	0.26±0.02^{e}	0.50±0.01^{d}	0.62±0.02^{b}
21	0.17±0.01^{d}	0.54±0.01^{c}	0.21±0.02^{e}	0.21±0.01^{e}	0.33±0.03^{d}
28	0.09±0.01^{d}	0.58±0.03^{c}	0.28±0.01^{e}	0.13±0.01^{f}	0.24±0.02^{c}

Values (Means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

was observed in VCO. Highest oxidation rate was executed in CPO, RBDCO and WCO.

CD value is also useful for monitoring the early stages of lipid oxidation that formed almost immediately after peroxides are produced (Xia and Budge, 2017). The oxidation of the tested oils expressed as CD value is presented in Figure 2 and fold increment of the values are shown Table 5.

All oil samples revealed the increase of CD value during the experiment. Therefore, oxidative stability of oils decreased with storage conditions. The CD values of all oils were slowly decreased after third or fifth day of storage since they would have completed conjugation of unsaturated fatty acids available. Based on the CD values, the highest oxidative stability was observed in VCO followed by CPO and CO. The rate of autoxidation of RBDCO was the highest under accelerated storage conditions.

The oxidation of the tested oils expressed as CT value is presented in Figure 3 and the fold increment of CT value of oils obtained during experimental period is presented in the Table 6. The CT values of all oils tested increased with the storage time. According to the data shown in Table 3, the rate of formation of CT value per day was lowest in WCO followed by CPO, VCO and RBDCO.

TBARS value is an indicator for the extent of secondary lipid oxidative products, aldehydes (Ross and Smith, 2006). Data illustrated in Table 7 and Figure 4 indicated that the TBARS value of the oils fluctuated with experimental time. During the first few days of the storage period, WCO, CPO and CO showed the highest level of

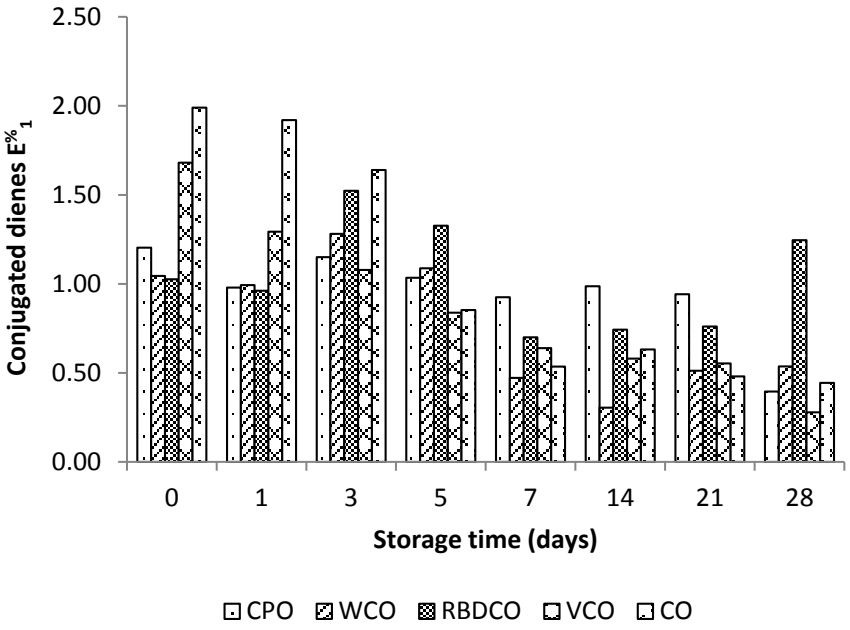

Figure 2. CD of different oils stored under *Schaal* oven conditions at 60±5°C.

Table 5. Fold increment of CD of different oils stored at 60±5°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	0.81±0.03^{ab}	0.95±0.10^{a}	0.94±0.14^{bc}	0.77±0.01^{a}	0.96±0.04^{a}
3	0.96±0.01^{a}	1.23±0.24^{a}	1.48±0.14^{a}	0.64±0.06^{ab}	0.82±0.04^{a}
5	0.86±0.11^{a}	1.04±0.19^{a}	1.29±0.04^{a}	0.50±0.10^{bc}	0.43±0.07^{b}
7	0.77±0.09^{ab}	0.45±0.13^{b}	0.68±0.01^{c}	0.38±0.01^{c}	0.27±0.06^{bc}
14	0.82±0.01^{ab}	0.29±0.02^{b}	0.72±0.14^{c}	0.35±0.02^{cd}	0.32±0.01^{bc}
21	0.78±0.11^{ab}	0.49±0.06^{b}	0.74±0.01^{c}	0.33±0.02^{cd}	0.24±0.02^{bc}
28	0.33±0.05^{b}	0.51±0.01^{b}	1.21±0.02^{ab}	0.17±0.01^{d}	0.22±0.04^{c}

Values (Means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

TBARS value and then reduction was observed. This could be attributed to the rapid production of tertiary oxidative product by these oils. Therefore, they did not respond positively for tests which evaluate secondary oxidative product. Thus, the increasing temperature may quicken the breakdown of fatty acids in the above mentioned oils. VCO and RBDCO positively responded to the test with the accelerated heating. TBARS values of these oils increased with time. As data shown in Table 3, based on the rate of development of TBARS value per day, WCO and VCO possessed higher oxidative stability as compared to other oils examined. RBDCO showed higher rate of oxidation.

Based on results obtained during the oxidation in the oven, WCO and VCO possessed the highest oxidative stability. Despite higher oxidation rate as measured by PV and CD value, WCO showed the lowest rate of formation of TBARS and CT (Table 3). Therefore, WCO can be categorized as oil which has the strongest oxidative stability against autoxidation. Oxidation rate of both primary and secondary oxidative parameters was lower for VCO. Therefore, oxidative stability of VCO was more or less similar to that of WCO. Oxidative stability of the oil tested can be arranged in descending order as WCO, VCO, CO, CPO and RBDCO.

High oxidative stability of VCO and WCO is mainly due to its higher phenolic content (Table 2) than other types of oils examined as well as fatty acid composition, in particular, to the high MUFA to PUFA ratio (Table 1). Even though the RBDCO contain higher MUFA: PUFA ratio than WCO, RBDCO showed poor oxidative stability than WCO, which could be attributed to the loss of minor components in RBDCO during refining. Several studies reported that the refined oils have less stability against

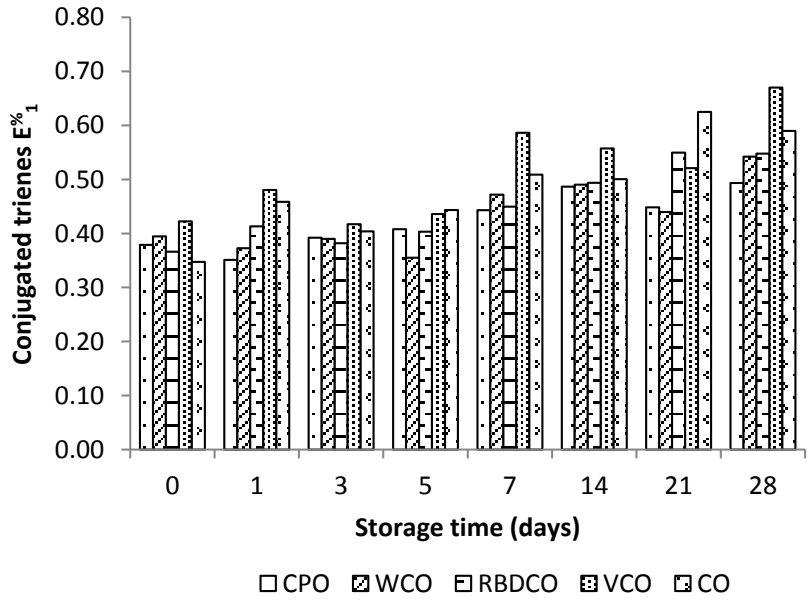

Figure 3. CT of different oils stored under *Schaal* oven conditions at 60±5°C.

Table 6. Fold increment of CT of different oils stored at 60±5°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	0.93±0.02^{c}	0.94±0.04^{bc}	1.13±0.37^{c}	1.14±0.10^{ab}	1.32±0.03^{cd}
3	1.03±0.09^{bc}	0.99±0.01^{bc}	1.04±0.50^{c}	0.99±0.02^{b}	1.16±0.01^{d}
5	1.08±0.05^{abc}	0.90±0.06^{c}	1.10±0.37^{c}	1.03±0.13^{b}	1.28±0.08^{cd}
7	1.17±0.04^{ab}	1.20±0.06^{abc}	1.23±0.70^{b}	1.39±0.07^{ab}	1.46±0.03^{bc}
14	1.28 ± 0.05^{a}	1.24±0.11^{ab}	1.35±0.38^{ab}	1.32±0.33^{ab}	1.44±0.16^{bcd}
21	1.18±0.09^{a}	1.11±0.08^{abc}	1.50±0.61^{a}	1.23 ±0.06^{ab}	1.80±0.15^{a}
28	1.30±0.04^{a}	1.37±0.12^{a}	1.50±0.38^{a}	1.59±0.02^{a}	1.70±0.03^{ab}

Values (means (n=3) ± SD) with different letters in the same column imply significant differences (p <0.05).

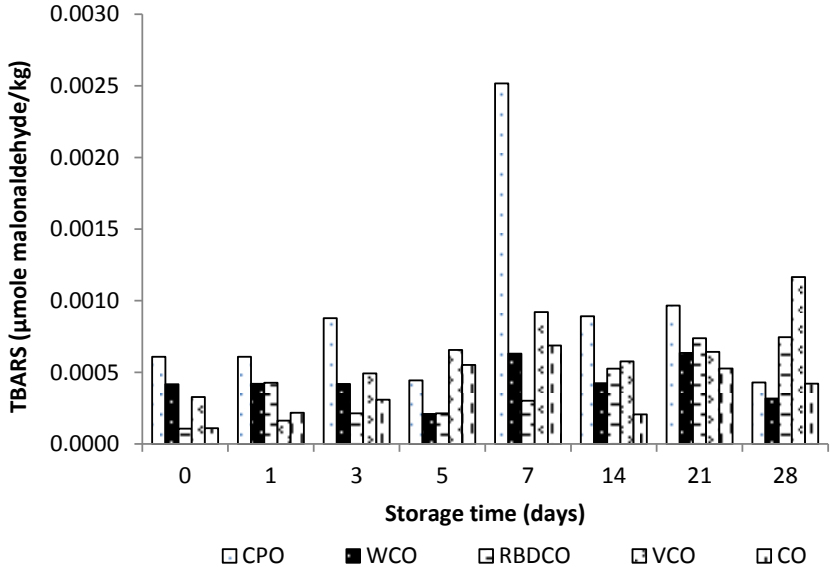

Figure 4. TBARS values of oils stored under *Schaal* oven conditions at 60±5°C.

Table 7. Fold increment of TBARS of different oils stored at 60±5°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	1.00±0.01^{cd}	1.01±0.01^{b}	4.00±0.02^{c}	0.50±0.23^{c}	1.99±0.01^{e}
3	1.44±0.18^{bc}	1.01±0.03^{b}	2.00±0.02^{e}	1.50±0.21^{bc}	2.82±0.32^{d}
5	0.73±0.09^{d}	0.50±0.01^{d}	2.00±0.05^{e}	2.00±0.46^{b}	5.03±0.02^{b}
7	4.14±0.17^{a}	1.51±0.04^{a}	2.83±0.23^{d}	2.81±0.61^{a}	6.27±0.33^{a}
14	1.47±0.34^{b}	1.02 ± 0.03^{b}	4.93 ± 0.18^{b}	1.76 ± 0.69^{bc}	1.88 ± 0.02^{e}
21	1.59±0.25^{b}	1.53±0.02^{a}	6.92±0.50^{a}	1.96±0.03^{b}	4.80±0.06^{b}
28	0.71±0.10^{d}	0.76±0.01^{c}	6.99±0.31^{a}	3.55±0.52^{a}	3.84±0.03^{c}

Values (means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

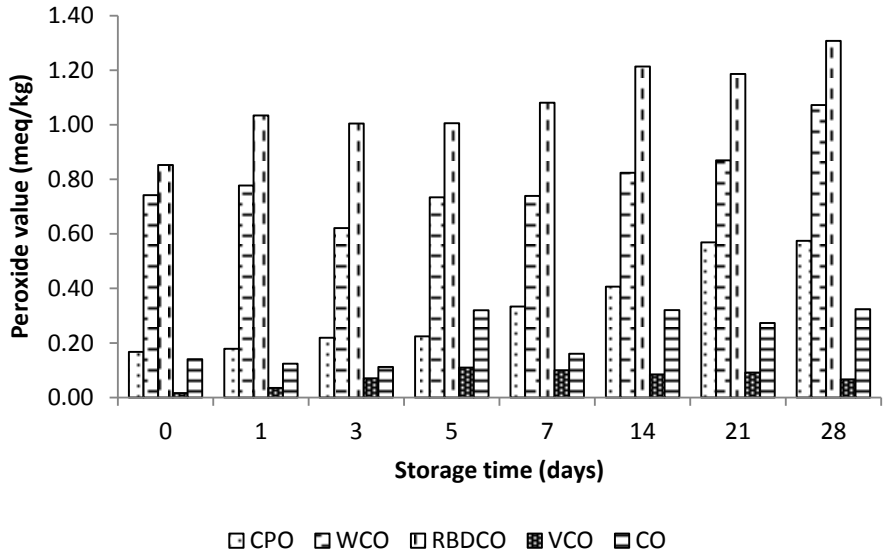

Figure 5. Peroxide value of different oils stored under fluorescent light at 27±1°C.

Table 8. Rate of increment (per day) of PV, CD, TBARS and CT of different oils stored under fluorescent light at 27±1°C during storage.

Parameter	CPO	WCO	RBDCO	VCO	CO
PV	0.087	0.025	0.014	0.115	0.061
CD	0.011	0.014	0.014	0.006	0.018
TBARS	0.340	0.066	0.205	0.120	0.525
CT	0.040	0.013	0.023	0.019	0.058

oxidation than their unrefined counterparts attributed to the loss of natural antioxidants during chemical refining process (Velasco and Dobarganes, 2002; Medina-Juárez and Gámez-Meza, 2011; Szydłowska-Czerniak and Łaszewska, 2015). Same reason is applicable in the present study.

Oxidative stability of oils under fluorescent light

The rate of increment of PV, CD, CT and TBARS per day of tested oils is summarized in Table 8. The oxidation of edible oils exposed to fluorescent light as measured by PV is shown in Figure 5 and Table 9. According to the results, the peroxide value of oils increased up to 28 days of storage under fluorescent light. The oxidative stability of oils decreased with the storage period. At the end of the storage period, PV of WCO and RBDCO exceeded the acceptable level which is less than 1 meq/kg (Khan and Shahidi, 1999). The concentration of peroxides and hydroperoxides was low in all oils tested during 28 days of storage under the fluorescent light. Despite the fact

Table 9. Fold increment of PV of different oils stored under fluorescent light at 27±1 °C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	1.07±0.09^{e}	1.05±0.02^{cd}	1.21±0.03 ^{cd}	2.14±0.32^{b}	0.88±0.01^{d}
3	1.31±0.09^{d}	0.84 ± 0.01^{e}	1.18±0.02 ^{d}	4.32±0.88^{ab}	0.80±0.05^{d}
5	1.34±0.11^{d}	0.99±0.02^{d}	1.18±0 .02^{d}	6.73±0.80^{a}	2.28±0.04^{a}
7	1.99±0.10^{c}	1.00±0.03^{d}	1.27±0.04^{c}	6.14±0.60^{a}	1.15±0.01^{c}
14	2.43±0.08^{b}	1.11±0.01^{bc}	1.42±0.01^{b}	5.20±0.35^{a}	2.28±0.02^{a}
21	3.40±0.19^{a}	1.17±0.01^{b}	1.39±0.03^{b}	5.60±0.41^{a}	1.95±0.02^{b}
28	3.43±0.19^{a}	1.45±0.01^{a}	1.53±0.04^{a}	4.08±0.43^{ab}	2.31±0.02^{a}

Values (means (n=3) ± SD) with different letters in the same column imply significant differences (p <0.05).

Table 10. Fold increment of CD of different oils stored under fluorescent light at 27±1°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	0.98±0.01^{b}	0.97±0.10^{b}	1.25±0.10^{a}	0.83±0.05^{a}	1.26±0.16^{a}
3	1.00±0.12^{b}	1.10±0.10^{ab}	1.29±0.11^{a}	0.91±0.20^{a}	1.24±0.33^{a}
5	1.04±0.07^{b}	1.01±0.01^{b}	1.22±0.02^{a}	0.66±0.26^{a}	1.15±0.09^{a}
7	1.15±0.10^{ab}	1.12±0.02^{ab}	1.18±0.09^{a}	0.83±0.06^{a}	1.35±0.44^{a}
14	1.17±0.06^{ab}	1.19±0.06^{ab}	1.32±0.16^{a}	0.75±0.06^{a}	1.29±0.18^{a}
21	1.28±0.09^{a}	1.23±0.04^{ab}	1.36±0.17^{a}	0.88±0.07^{a}	1.40±0.09^{a}
28	1.29±0.01^{a}	1.36±0.04^{a}	1.48±0.02^{a}	0.96±0.10^{a}	1.55±0.31^{a}

Values (means (n=3) ± SD) with different letters in the same column imply significant differences (p <0.05).

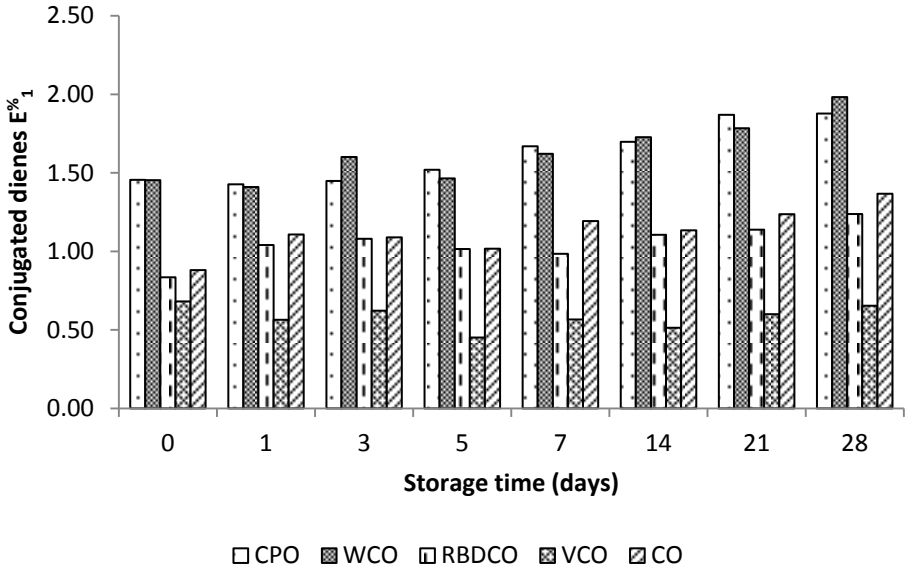

Figure 6. Conjugated dienes of different oils stored under fluorescent light at 27±1°C.

that initial stages of photo-oxidation progressed rapidly, the level of oxidation is less as compared to that of autoxidation. In this experiment, RBDCO and WCO showed higher oxidative stability against the fluorescent light oxidation as measured by increment of PV per day VCO was more susceptible to photo-oxidation (Table 8).

The CD values and fold increments of CD values of tested oils under fluorescent light are presented in Figure 6 and Table 10, respectively. The variation of CD values during experiment period followed a similar trend as PV,

Table 11. Fold increment of CT of different oils stored under fluorescent light at 27±1°C up to 28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	0.88±0.36[a]	0.95±0.18[a]	0.99±0.24[a]	0.65±0.63[a]	1.07±0.73[a]
3	1.02±0.50[a]	0.95±0.06[a]	1.10±0.34[a]	0.46±0.60[a]	1.40±0.41[a]
5	0.80±0.04[a]	0.90±0.04[a]	0.93±0.05[a]	0.52±0.50[a]	1.52±0.02[a]
7	0.88±0.19[a]	1.05±0.15[a]	1.17±0.09[a]	0.78±0.55[a]	1.16±0.08[a]
14	1.16±0.21[a]	0.94±0.22[a]	1.16±0.44[a]	0.61±0.04[a]	1.78±0.30[a]
21	1.03±0.38[a]	0.86±0.18[a]	0.84±0.05[a]	0.59±0.18[a]	2.24±0.34[a]
28	0.98±0.35[a]	0.91±0.11[a]	1.03±0.05[a]	0.95±0.46[a]	2.17±0.81[a]

Values (means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

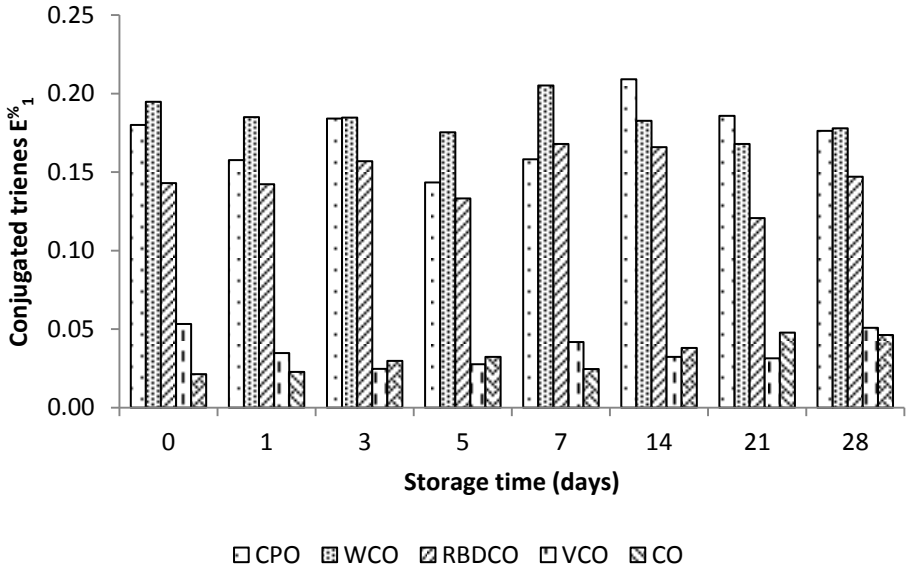

Figure 7. Conjugated trienes of different oils stored under fluorescent light at 27±1°C.

VCO, WCO, CPO, RBDCO and CO showed more or less constant fold increment throughout the time of illumination.

Fold increment of CT value and development of CT value of the oils obtained during experimental period is presented in the Table 11 and Figure 7. The oxidation of all oils tested increased with the storage time. Therefore, the effect of fluorescent light on oxidation was obvious. CPO, WCO, RBDCO, VCO and CO showed an ability to maintain a statistically more or less constant fold of increment throughout the experiment. This may be due to the presence of higher amount of antiphoto-oxidative compounds of theses oils. According to the results obtained as rate of formation of CT value per day (Table 8), CO showed lowest stability followed by CPO, RBDCO, VCO and WCO.

According to the data illustrated in the Figure 8 and Table 12, level of oxidation has increased with the extended storage life in all the oil samples. The results suggested that the oxidation of these oils accelerated by the fluorescent light effect. First few days of the storage period of CO and VCO showed the highest level of TBARS value and then reduction was observed. In the later stages of lipid oxidation, the secondary oxidative products are broken down into tertiary oxidative products (Dąbrowska et al., 2015). The TBARS values of other oils excluding the aforementioned oils were well correlated with the storage time. Based on the data shown in Table 8, WCO showed higher photo-oxidative stability. CO showed the highest oxidative rate followed by CPO and RBDCO.

The role of light in the oxidation process is clearly shown by the rate of oxidation. Based on the results presented as PV, CD, TBARS and CT values, CO is more susceptible to photo-oxidation among the oils examined. The second lowest photo-oxidative stability was exhibited by CPO followed by RBDCO. Based on the results, the susceptibility of these oils for photo-oxidation

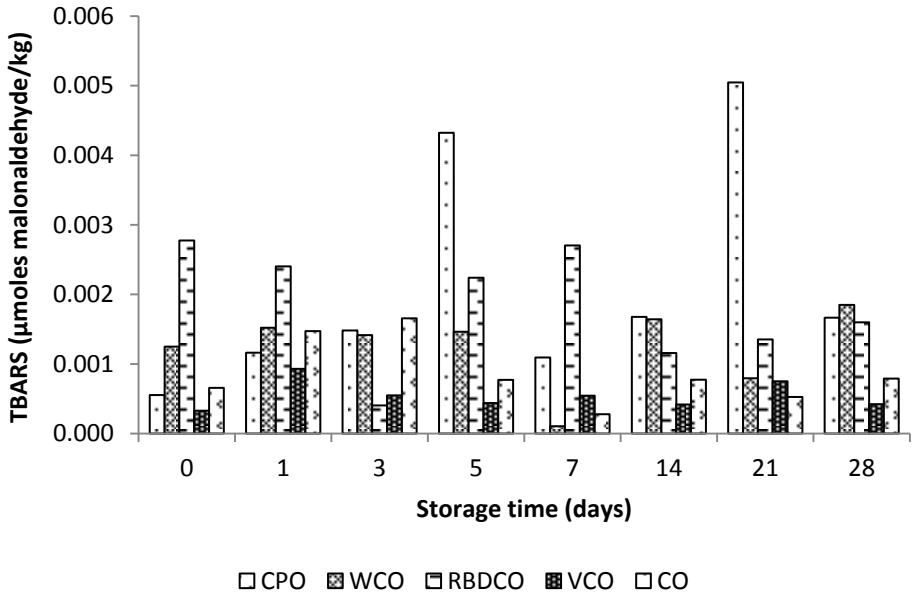

Figure 8. TBARS value of different oils stored under fluorescent light at 27±1°C.

Table 12. Fold increment of TBARS of different oils stored under fluorescent light at 27±1°C up to28 days.

Storage time (days)	CPO	WCO	RBDCO	VCO	CO
1	2.10±0.41^{c}	1.22 ± 0.21^{a}	0.87 ± 0.03^{ab}	2.84±0.26^{a}	2.24±0.11^{a}
3	2.68±0.12^{c}	1.13±0.23^{a}	0.15±0.01^{d}	1.67±0.03^{c}	2.52±0.03^{a}
5	7.82±0.22^{b}	1.17±0.15^{a}	0.81±0.02^{b}	1.33±0.01^{c}	1.17±0.01^{b}
7	1.98±0.05^{c}	0.08±0.01^{c}	0.97±0.02^{a}	1.66±0.05^{c}	0.42±0.12^{c}
14	3.03±0.21^{c}	1.31±0.25^{a}	0.42±0.02^{c}	1.27±0.01^{c}	1.18±0.12^{b}
21	9.13±0.53^{a}	0.64±0.14^{b}	0.49±0.08^{c}	2.29±0.04^{b}	0.80±0.01^{bc}
28	3.01±0.11^{c}	1.48±0.25^{a}	0.58±0.03^{c}	1.29±0.02^{c}	1.20±0.12^{b}

Values (means (n=3) ± SD) with different letters in the same column imply significant differences ($p < 0.05$).

can be arranged as WCO<VCO<RBDCO<CPO<CO. High content of total phenolics could be the reason for higher stability of WCO and VCO against photo-oxidation than other types of oils. Furthermore, pigments such as riboflavin and porphyrins (chlorophyll) can act as initiators of lipid photo-oxidation (W¹sowicz et al., 2004). Therefore, the presence of pigments in addition to the less total phenolics could have led to less stability of CPO and CO.

Conclusion

In conclusion, WCO and VCO showed higher stability against both autoxidation and photo-oxidation than RBDCO, CPO and CO. This may be attributed to the high phenolic and other minor natural compounds as well as high MUFA to PUFA ratio. While the unrefined oils contain high amount of phenolic and other natural antioxidants including vitamins, which act to increase the oxidative stability, presence of prooxidants such as free fatty acids, trace metals and others may act to reduce the stability. Thus, it is necessary to quantify all minor compounds present in these oils to make a clear conclusion regarding the stability of these oils against oxidation. Thus, further studies are needed to identify and quantify the minor compounds (pro-and antioxidants) in order to identify the exact reason for the variation in the oxidative stabilities among these various types of coconut oils. In addition, further research is necessary to examine the effect of packaging materials on the susceptibility of these oils to oxidation, which will help to minimize the oxidative deterioration and thus improve the organoleptic, nutritional and economic value of the coconut oils.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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