Assessing the effects of Sunlight on the Photooxidation of Tropical Oils

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Abstract

This study aims to investigate the influence of sunlight on the photooxidation of tropical oils (TOs). Coconut oil (CNO), palm oil (PO), and palm kernel oil (PKO) were chosen for determining the indicators of photooxidation when exposed to and in the absence of sunlight for seven weeks. The results showed a significant ($p < 0.05$) increase in free fatty acid (FFA) levels and peroxide value (PV) when the TOs were exposed to sunlight. The iodine value (IV) and colour content decreased significantly ($p < 0.05$) due to the decomposition of unsaturated FFAs owing to the breaking-down of the π-bonds and the degradation of colour pigments during photooxidation. FTIR analysis showed strong vibrational absorptions at 1721 and 3505 cm^{-3}, 1720 and 3560 cm^{-3}, and 1721 and 3554 cm^{-3} for the CNO, PO, and PKO samples exposed to sunlight, respectively. These bands can be attributed to the presence of secondary oxidation products, which were absent in the TOs that were not exposed to sunlight. A simulation was performed to support the FTIR results, which also indicated peaks from the secondary oxidation products at 1744 and 3660 cm^{-3}. The study also revealed that the rate of photooxidation was different for each TOs. The rate of oxidation followed the order PO > PKO > CNO. In contrast, no notable changes were observed in the TOs kept away from the sunlight. These results suggest that exposing TOs to sunlight influences their oxidation stability and quality.

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ABSTRACT

This study aims to investigate the influence of sunlight on the photooxidation of tropical oils (TOs). Coconut oil (CNO), palm oil (PO), and palm kernel oil (PKO) were chosen for determining the indicators of photooxidation when exposed to and in the absence of sunlight for seven weeks. The results showed a significant ($p < 0.05$) increase in free fatty acid (FFA) levels and peroxide value (PV) when the TOs were exposed to sunlight. The iodine value (IV) and colour content decreased significantly ($p < 0.05$) due to the decomposition of unsaturated FFAs owing to the breaking-down of the π-bonds and the degradation of colour pigments during photooxidation. FTIR analysis showed strong vibrational absorptions at 1721 and 3505 cm^{-3}, 1720 and 3560 cm^{-3}, and 1721 and 3554 cm^{-3} for the CNO, PO, and PKO samples exposed to sunlight, respectively. These bands can be attributed to the presence of secondary oxidation products, which were absent in the TOs that were not exposed to sunlight. A simulation was performed to support the FTIR results, which also indicated peaks from the secondary oxidation products at 1744 and 3660 cm^{-3}. The study also revealed that the rate of photooxidation was different for each TOs. The rate of oxidation followed the order PO > PKO > CNO. In contrast, no notable changes were observed in the TOs kept away from the sunlight. These results suggest that exposing TOs to sunlight influences their oxidation stability and quality.

KEYWORDS: Tropical oils, Photooxidation, Sunlight, Saturation, Unsaturation, Oxidation

INTRODUCTION

With the increasing awareness about health and safety in modern societies, the usage of tropical oils (TOs) is declining. Typical TOs include coconut oil (CNO), palm oil (PO), and palm kernel oil (PKO), which predominantly comprise saturated fatty acids (Ramadan, 2019). The high degree of saturation in TOs (CNO, [?] 93%; PKO, [?] 82%; PO, [?] 50%) (Dubois *et al.* , 2007) has raised several health concerns and debates on the safety and health risks of saturated fatty acid consumption (Cassiday, 2017)(Noordwijk, 2020). However, TOs are essential for food and other uses and are potential solutions for achieving food security in the future (Puah and Kochhar, 2019). To meet the requirements of the ever-increasing world population, improvement and reassessment of the existing food resources is critical.

TOs differ majorly in their chemical compositions and applications. CNO and PKO predominantly comprise lauric (C12) and myristic acids (C14) which contain a high proportion of medium-chain fatty acids (MCFAs) (Gunstone, 2011). PO also comprises predominant levels of long-chain fatty acids (LCFAs), such as palmitic acid (C16) and oleic acid (C18:1) (Kostik, Memeti and Bauer, 2013). In addition to the differences in their chemical compositions, some novel potential applications of TOs have been recently discovered. PO and PKOs are currently considered as substitutes for green fossil fuels despite inadequate clarity on the sustainability of such usage. Ana et al. reported that PO-based biofuels possess the potential for increasing greenhouse gas savings in the near future (Hu and Cheng, 2013). CNO is also emerging as a solution to various health problems and could play a critical role in the health sector (Woolley et al., 2020).

Notwithstanding the emerging applications and advantages, the quality of TOs has always been a concern. The poor handling and storage of TOs adversely affects their stability and quality, which depreciates their market, economic, and nutritional values. These issues can be widely attributed to the unconventional storage of TOs in the presence of sunlight. In Africa, one of the traditional refining practices by many TOs producers is the exposure of oils to sunlight (Tonfack Djikeng*et al.* , 2019). This approach is cost-effective, convenient, and economical for evaporating the residual water in oils. A related approach adopted by the distributors and vendors of TOs involves exposing the oils to sunlight for marketing purposes (Mwanza and Ingari, 2015). This is due to the high viscosity, high melting points, and the saturated fatty acids present in TOs, owing to which, these oils solidify readily under ambient conditions within a short time. Marketers or vendors address this challenge by exposing the oils to sunlight for liquefying the solidified oils and maintaining the liquid or semi-solid forms (Ngono Ngane Annie, 2014), which increases the marketability and consumer acceptance. While these practices are suitable from the marketing and economics outlook, from a scientific perspective, they pose risks to the stability of the oils by oxidation or the related decomposition and are inappropriate (Maszewska et al., 2018 & Oh et al., 2014).

Like many other edible oils, TOs can undergo lipid oxidation upon prolonged exposure to sunlight. Light is a photochemical initiator capable of inducing photochemical reactions when exposed to food (Zeb*et al.* , 2008). The mechanism of the photochemical oxidation of vegetable oils has been extensively studied (Choe & Min, 2006; Koutchma, 2019), and the oxidation of lipids under light has been termed photooxidation. Irradiation of edible oils reduces their oxidative stability and renders them prone to rancidity (Min and Boff, 2002). TOs are susceptible to oxidation due to their low unsaturated fatty acid contents and the presence of colour pigments (photosensitisers) in minor quantities. Moreover, investigations have revealed that antioxidants that prevent oxidation in oils can be destroyed when exposed to prolonged sunlight (Oh, Lee and Choe, 2014). Photooxidation leads to the formation of peroxides and other volatile and harmful products in oils. This renders the oil less stable, reduces its value (economic, nutrition, and market) and safety. Importantly, the oils also lose their flavour and sensory quality and become unattractive and unacceptable to consumers, which results in economic losses to both the food and non-food industries (Redondo-Cuevas *et al.* , 2018).

Therefore, the impact of sunlight on TOs is not well understood, and comprehensive studies that compare and evaluate the effects of sunlight on the photooxidation of TOs are sparse. Hence, this study aims to monitor the changes in three TOs after exposure to sunlight for seven consecutive weeks. Iodine value (IV), free fatty acid (FFA), colour content and peroxide value (PV) were measured as the indicators of the degree of photooxidation in the TOs. Fourier transform infrared spectroscopy (FTIR) was performed to study the photooxidation in TOs further. A simulation was performed to support the FTIR results and study the oxidation of TOs. Statistical analysis was performed to verify the validity and significance of these findings.

EXPERIMENTAL PROCEDURE

Sampling of TOs

Freshly prepared CNO, PO, and PKO were purchased from selected locations of Kotokuraba (Cape Coast, Ghana) and Bantama (Kumasi, Ghana) local markets. All TOs were immediately stored in 1500 mL dark polypropylene bottles, away from light, to avoid premature photochemical reactions. The caps on each bottle containing the oils were well sealed and locked to limit oxygen penetration. Each set of TOs stored in the dark polypropylene bottle was divided into two subsets: one bottle was labelled as the control sample, and the other labelled as the test sample. The control sample was stored away from light, whereas the test samples were exposed to sunlight for seven consecutive weeks.

Irradiation Procedure

Seven samples of each oil test sample were exposed to sunlight for seven consecutive weeks (8 h of exposure per day) at Adum (Kumasi, Ghana), between May and June on sunny days. The mean maximum and minimum temperatures from May to June were 33–29 °C. After each day of exposure, the sample was analysed for the FFA, PV, IV, and colour content. All analyses and characterisations were performed at the Ghana Nut Company limited, Techiman (Ghana), and Kwame Nkrumah University of Science and Technology (KNUST), Kumasi, Ghana.

Free Fatty Acids

The percentage (%) of FFA in the oil was evaluated according to the AOCS (Ca 5a-40) (AOCS, 2017) protocol. The oil samples were shaken thoroughly to ensure even mixing of the oil. The oil (2.0 g) was weighed with a Sartorius analytical balance in a conical flask. Neutralised ethanol (50 mL) in a flask was heated to 70 °C on a heating mantle, which dissolved all of the oils in the flask. Phenolphthalein indicator (2 to 3 drops) was added to the heated mixture, and the mixture was titrated against 0.1 N NaOH solution with constant swirling until the first appearance of the pink colour, which persisted for 15–20 s. The procedure was performed in triplicate, and the FFA level was calculated from equation 2.

$$\% \, FFA = \frac{\text{T x N}}{\text{W}} \times 0.02 \, mg, \tag{1}$$

where T is the titer volume (mL), N is the normality of NaOH, and W is the weight of samples taken (g).

Peroxide Value

According to the AOCS (Cd 8b-90) protocol (AOCS Official Method Cd 8b-90, 2003), PV was evaluated by the titrimetric method, and the results were expressed in Meq O_2 / kg. The control and test samples were shaken thoroughly to ensure homogeneity of the TOs. The TOs (5.0 g) were weighed with a Pasteur pipette into a 250 ml dark Erlenmeyer flask, and 50 mL of acetic acid-isooctane solution (3:2, v/v) was added to the oil. The mixture was swirled to ensure dissolution. Potassium iodide (0.5 mL) was pipetted into the flask, and the flask was stored away from light before the titration. The resulting solution was shaken for 1 min, after adding 30 mL of distilled water. SDS solution (0.5 mL, 10%) and approximately 10 mL of the starch indicator was added, and titration was performed. The solution was titrated with 0.1 N standardised sodium thiosulfate with constant and vigorous agitation to liberate the iodine from the solvent layer completely. Thiosulfate was added dropwise until the solution turned from blue-black to colourless. The procedure was performed in triplicate, and the PV was calculated using equation 2.

$$\text{PV} \left(\frac{\text{meq}}{\text{kg}}\right) = \frac{(S - B) \text{ x N}}{W} \times 1000 \tag{2}$$

where S is the volume of Na_2SO_3required by the samples (mL), B represents the volume of Na_2SO_3 for the blank sample (mL),N is the normality of sodium thiosulfate (0.1 N), and W is the weight of the sample (g).

ATR-FTIR Analysis

A Bruker Alpha FTIR spectrometer equipped with platinum attenuated total reflectance (ATR-FTIR, Bruker, Karlsruhe, Germany) was used for the FTIR measurement. All measurements were performed at room temperature (25 °C) at the KNUST central laboratory (Kumasi, Ghana). The ATR-FTIR employed a diamond crystal, which was cleaned with isopropanol before any background scan was acquired. The oil sample

was placed directly on the crystal, and a pressure gauge was applied to ensure maximum contact. The background scan and the oil samples were sequentially measured from 4000 to 400 cm^{-1}. The spectra were obtained with 24 scans at 4 cm resolution using the OPUS software (Bruker, Karlsruhe, Germany).

Iodine Value

IV of oils was analysed according to the AOCS (Cd 1-25) protocol (AOCS, 2009), and the results were expressed in grams of iodine / 100 g of oil. The oils were shaken thoroughly to ensure even mixing, and 0.50 g of the oil was weighed with a Sartorius analytical balance in a 250 ml Erlenmeyer flask. Cyclohexane-acetic acid (1:1) solution (20 mL) was added to the flask, followed by 25 ml of Wijs solution; the mixture was swirled to ensure homogeneity. The flask was stored in a dark enclosed area for 1 h. Potassium iodide solution (20 mL) and distilled water (100 mL) were immediately added to the solution. The resulting solution was titrated with 0.1 N standardised sodium thiosulfate. At the endpoint, the yellow colour of the solution completely disappeared, at which point, 1–2 mL of the starch indicator was added to the solution. The titration was continued until the solution turned from blue to colourless. The procedure was performed in triplicate, and IV was calculated from equation 3.

$$\text{IV}\left(\frac{\text{g of iodine}}{100g\ of\ Oil}\right) = \frac{(B-S)\ \text{x N}\ x\ 12.69}{W} \tag{3}$$

where S is the volume of $Na_2SO_3$for all samples (mL), B is the volume of Na_2SO_3 for the blank sample (mL),N is the normality of sodium thiosulfate (0.1 N), and Wrepresents the weight of the sample (g).

Colour Content

According to the AOCS (Cc 13b-45) protocol (Associaton of Offical Analytical Chemists, 2000), the oil colour content was determined analytically. The instrument used was the Lovibond Tintometer PFX-i series. The results were expressed in the AOCS tintometer red (R) and yellow (Y) units.

FTIR Analysis

Computational Modelling of FTIR Spectra

The Gaussian 16 software (Frisch *et al.* , 2016) was used for all computational modelling and calculations. Oleic acid, palmitic acid, and lauric acids were used as the fatty acids present in TOs required to simulate the photooxidation of TOs (Cheng et al., 2018). The initial geometries of the fatty acids were optimised, and the vibrational frequencies were obtained. The geometry optimisation and vibrational frequency calculations were performed with the density functional theory (DFT) in the gas phase (Pereira *et al.* , 2017). The basis set used was Becke's three parameters, and the Lee-Yang-Parr nonlocal correlation functional (B3LYP) at the 6-311++G (d, p) basis set. The final geometries of the fatty acids after the vibrational frequencies were visualised with GaussView (version 6.0.16) (Dennington, Keith and Millam, 2016). A scaling procedure (Palafox, 2019) was implemented to improve the wavenumbers and transmittance of the simulated spectra to match those of the experimental data.

Statistical Analysis

All data were obtained from at least three measurements, and each replicate was reported as the mean standard± deviation. OriginPro (version 2020, Northampton, MA, USA) and SPSS (IBM Corp., 2020, version 27, Armonk, N.Y., USA) were used for the one-way analysis of variance (ANOVA). The significant difference was evaluated by the P-values ($P < 0.05$).

RESULTS AND DISCUSSION

Effects on Free Fatty Acids

FFA is considered as a weight parameter that is linked to the oxidative stability of vegetable oils. Table 1 shows the various changes in %FFA for all TOs at 95% confidence level (CL). The %FFA of the exposed and unexposed CNO, PKO, and PO were initially 1.32±0.01, 8.42±0.01, and 3.81±0.01, respectively. After seven weeks of sunlight exposure, the %FFA in the CNO, PKO, and PO substantially increased to 3.75±0.03, 15.04±0.03, and 6.59±0.03respectively. This shows that the effect of sunlight on FFA is significantly different ($p < 0.05$) for the control and sunlight-exposed TOs. The results presented in Figure 1 indicate that the increase in %FFA is faster in PO than in CNO and PKO. This can be attributed to the differences in the amounts of the saturated FFA present in the TOs. CNO and PKO are characterised by the most representative portion of saturated FFA consisting primarily of lauric acid (54% CNO and 50% PKO). PO is characterised by a substantial level of saturated and monounsaturated FFA, with the prominent constituents being palmitic (52%) and oleic acids (43%)(Kostik, Memeti and Bauer, 2013). Hence, PO is more prone to photooxidation due to its high level of unsaturation compared to CNO and PKO (Maszewska *et al.* , 2018). Second, the TOs stored away from exposure to the sun recorded the lowest %FFA. After the seven weeks of protection from light, the %FFA increased slightly from 1.30±0.01 to 1.40±0.01 (CNO), 3.81±0.01 to 3.96±0.03 (PKO), and 8.42±0.01 to 8.68±0.02 (PO), among the oils. This increase in %FFA can be attributed to enzymatic or hydrolytic oxidation, which is initiated by the presence of moisture or enzymes remaining in the TOs after processing.

Similar results have already been reported by several researchers and agree well with those of the present study. For instance, Fekarurhobo et al. (2009) investigated the short-term exposure of sunlight on PO and PKO. After eight months of sunlight exposure, they reported an increase in %FFA from 5.0 to 6.0 and 2.5 to 4.1 in PKO and PO, respectively. Henry (2011) also reported variations in the FFA of crude PO when exposed to various light radiations and found that the %FFA in the PO rose from 0.738 to 1.084 after 20 days of sunlight exposure. These results showed an appropriate correlation with the results of Rukmini et al. (2011), who monitored the effects of fluorescent light on virgin coconut oil (VCO). They reported a similar increase in %FFA in various commercial VCOs after exposure to fluorescent light for 5 hrs. The %FFA in several commercial VCOs increased from 0.18 to 0.84, 0.29 to 1.24, 0.48 to 3.11, 0.16 to 1.14, and 0.08 to 0.25 after exposure to light. Figure 1 shows that the higher the degree of the unsaturation in the TO, the faster is the oxidation reaction. Hence, the present study can conclude that the oxidative stability of the TOs decreases in the order: PO < PKO < CNO, due to different degrees of unsaturation. This results in an increase in the %FFA, which decreases the oxidative stability and shelf life of the TOs. Figure 1 shows that the PO with the highest level of unsaturated fats is more prone to photooxidation than CNO and PKO. Thus, the type of FFA present in TOs plays a crucial role in their stability against oxidation. The one-way ANOVA of the mean for the protected and unprotected TOs showed a statistically significant ($p > 0.05$) difference in %FFA.

Effects on Peroxide Value

PV is an important indicator that provides the initial evidence of primary oxidation in TOs. As shown in Table 2, the evolution of PV was monitored for seven consecutive weeks at 95% CL. The TOs that were not exposed to sunlight showed the lowest increase in PV as a function of time. The PV ($meqO_2$ / kg) in the TOs increased slightly from 2.03±0.01 to 2.14±0.02 (CNO), 6.13±0.01 to 6.23±0.04 (PKO), and 3.09±0.01 to 3.22±0.04 (PO) for the oils. The increase in PV ($meqO_2$ / kg) despite the protection from light can be attributed to the presence of dissolved oxygen in the TOs (Johnson and Decker, 2015). Dissolved oxygen can induce oxidation of unsaturated FFAs in the absence of light (Johnson and Decker, 2015). In addition, the results in Figure 1 show that the increase in PV was more pronounced in the sunlight-exposed TOs. The TOs exposed to light showed a significant increase of the PV from 2.03±0.01 to 4.8±0.03 (CNO), 6.13±0.01 to 9.33±0.03 (PKO), and 3.09±0.01 to 8.43±0.05 (PO). This shows that the effect of sunlight on PV is significantly different ($p < 0.05$) for the control and exposed TOs. The rapid increase in PV can be ascribed

to the chemical changes that occur in the TOs when exposed to sunlight. Therefore, prolonged exposure of TOs to relatively high light intensities caused significant initiation of photooxidation in the oils. These findings are in agreement with the work of Rukmini and Raharjo (2010), who observed a similar pattern. They reported an increase in the PV of VCO upon exposure to fluorescent light at higher storage times. Such an increase in PV has also been studied by Almeida et al. (2019), who stored refined PO (RPO) in an enclosed dark area (20–25 °C) and at ambient temperature with exposure to natural light (26–32 °C). After 12 months of storage with exposure to light, the PV (meq O2/ kg) for the RPO increased drastically from 00.52 to 85.29. The RPO stored away from light increased slightly from 00.59 to 14.64 meq O_2/kg.

However, different types of oils can either undergo the same or different changes during the initial stages of oxidation. This is evidenced by the variations in the levels of PV for each TOs shown in figure 2. The maximum PV was recorded for PO on the 7^{th} day (Figure 2), followed by PKO and CNO. At all stages, the increase in the PV of PO was more rapid than in CNO and PKO, which can be attributed to the increase in PV with the increase the degree of unsaturation in the TOs. PO consists of higher levels of unsaturated FFAs (C18:1) compared to CNO (C18:1) and PKO (C18:1), owing to which, PO showed a rapid increase in PV when exposed to sunlight than the PKO and CNO. Therefore, the observed increase in PV agrees well with the degree of unsaturation in the TOs. Suryani et al. (2020) also reported that CNO should theoretically exhibit a low rate of oxidation due to its low unsaturated FFA content. Thus, unsaturated FFAs readily form peroxides (or hydroperoxides) compared to the saturated congeners, which causes the high PV. A higher PV reflects lower chemical stability of the oil and renders the oil less safe for food and non-food uses (Frankel and Huang, 1994). Therefore, the chemical stability of TOs needs to be enhanced by protecting the oils from direct exposure to sunlight. This can be achieved by the appropriate revision and improvement of the packaging and storage conditions of these oils. Attention to such handling of TOs will improve their acceptability.

Effects on Iodine Value

IV is a vital characteristic of oils and represents the degree of unsaturated fatty acids (double bonds) present in the oil (Kumar*et al.* , 2012). This amount represents the unsaturated fatty acids before and after the oxidation of the oil. Thus, IV indicates the oxidative stability of oil and is a useful parameter for studying the changes in the unsaturated fatty acid contents in TOs exposed to sunlight. Table 3 presents the decrease in IVs of TOs stored under different conditions at 95% CL. The TOs stored away from light exhibited the lowest reduction of IV per week, unlike the oils exposed to light. The IV (g of iodine/100 g of oil) in the unexposed TOs decreased from 6.10±0.01 to 6.03±0.02 for CNO, 18.25±0.01 to 18.12±0.02 for PKO, and 49.35±0.01 to 49.19±0.03 for PO. The exposed TOs showed a significant decrease in IV per week from 6.10±0.01 to 4.94±0.04 for CNO, 18.25±0.01 to 16.6±0.01 for PKO, and 49.35±0.01 to 44.12±0.05 for PO. This shows that the effect of sunlight on IV is significantly different ($p < 0.05$) for the control and exposed TOs. IV decreases in TOs because it is proportional to the amount of iodine required to saturate the FFAs present in the TOs. The different types of FFA in the TOs account for the varied IV values for each oil. Approximately 51% of unsaturated fatty acids are found in PO (oleic acid), 35% in PKO (myristic acid), and 15% in CNO (myristic acid) (Gunstone, 2011). In addition, Figure 3 provides a comparison between the decreasing rates of IVs of protected and unprotected TOs using a bar chart, which shows that the decrease in IV is more pronounced when the TOs are exposed to sunlight. Thus, when the TOs are exposed to sunlight, a higher number of unsaturated FFA (C=C) in TOs are likely decomposed into radicals, thereby reducing the degree of unsaturation (C=C) in TOs. Dawodu et al. (2015) reported a similar trend when PKO was exposed to different temperatures. The IV in the PKO, initially at 5.00, decreased to 7.00 when the temperature was increased to 300 °C. Fekarurhobo et al. (2009) also reported a decrease in IV from 20.0 to 18.5 and 50.0 to 40.1 in PKO and PO, respectively. In their research, PO and PKO were exposed to sunlight for a shorter time (25 days) than that in the present study (49 days). However, PKO and PO showed a decrease in IV after 25 days of sunlight exposure. They further noted that the reduction of IV could be ascribed to the release of unsaturated FFAs as the π-bonds broke down during photooxidation. In addition, the highest significant decrease of IV was found in PO, followed closely by PKO and CNO. As previously mentioned,

the degree of unsaturation of oils accounts for the rate at which the IV decreases. PO with the highest number of unsaturated FFA are likely to have more rapid reduction of IV than PKO and CNO. PKO has a higher number of unsaturated FFA, and therefore, its IV decreases faster than that of CNO. In summary, IV is a useful diagnostic of TOs photooxidation and provides useful information on the oxidative stability of TOs. In addition, it is a straightforward and convenient method for evaluation of TOs in typical laboratory settings and can be used by TOs producers to monitor the changes in TOs exposed to sunlight.

Effects on Colour

Colour is an indirect measure of product quality or product condition. The colours of the TOs were measured using a Lovibond Tintometer Pfx 880 series. The Lovibond colour scale was used to express the colour in the red and yellow AOCS tintometer units. Table 4 depicts the colour changes in the TOs samples during the seven day test period. The initial colours of the TOs were 1.11R±3.40Y, 2.07R±18.2Y, and 3.63R±9.40Y for CNO, PKO, and, PO, respectively, before exposure to sunlight. After seven days of maintaining the samples in the absence of sunlight, the colours of the TOs reduced to 1.04R±3.33Y, 2.05R±9.36Y, and 3.57R±9.36Y for CNO, PKO, and PO, respectively. These results indicate an insignificant change in the colour of TOs stored in the absence of sunlight. However, the TOs exposed to light showed a notable decrease in colour to 0.44R±2.51Y, 1.13R±16.8Y, and 1.80R±9.08Y for CNO, PKO, and PO, respectively. This shows that the effect of sunlight on the colour content is significantly different ($p < 0.05$) for the control and exposed TOs. These results are supported by the findings of Tonfack et al. (2019) (Tonfack Djikeng *et al.* , 2019) who reported the decrease in colour from 1.00R±6.00 to 0.20R±0.80Y after 90 days of sunlight exposure. Almeida et al. (2019) and Taluri et al. (2019) also reported that high-temperature storage conditions could reduce the colour content of oils. They reported that the degradation of colour pigments in oil during photooxidation affects the colour of the oil. These changes are pronounced in oils with a higher degree of unsaturation during prolonged exposure to light. The colour changes of the exposed TOs can be attributed to the damage of the colour pigments present in the oil. Thus, prolonged exposure to sunlight can deteriorate the colour pigments in oils. Choe et al. (2014) reported that colour changes could also be affected by the type of packaging material used for the oils. Transparent PET bottles used in the packaging of oils allow the transmission of light through oils, thereby destroying the pigments in oils. Hence, the colour content is reduced, changing the appearance of the oil; this change could indicate a problem during storage or the exposure of the oil to adverse conditions. The bar chart (Figure 4) shows that the decrease in colour was faster in PO than in PKO and CNO. As previously discussed, the degree of unsaturation is closely related to the variation of colour in oils. The higher the degree of unsaturation, the more likely are the colour pigments degraded during photooxidation. Therefore, the colour of TOs is a significant indicator of oil stability.

FTIR Studies on Photooxidation

ATR-FTIR analysis was performed to study the photooxidation of the TOs. Computational simulation was used to support the findings of the ATR-FTIR. Figures 5–7 show the experimental and simulated FTIR spectra for the exposed and unexposed (control) TOs. The principal absorption bands relevant for studying photooxidation were located at 3505 cm^{-3}, 3560 cm^{-3}, and 3554 cm^{-3}, corresponding to CNO, PO, and PKO, respectively. The absorption bands were ascribed to the presence of secondary oxidation products, such as hydroperoxides or alcohol-related compounds(Navarra *et al.* , 2011). The bands indicated the stretching vibration of the O-H bonds during oxidation. The stretching vibration was observed to be weak in the unexposed TOs due to the low degree of oxidation. A weak vibration was also observed for the exposed CNO due to the high degree of saturation, which renders it more stable against oxidation. These results comply with the those from the simulated spectra demonstrated in Figure 7, producing a strong absorption band at 3640 cm^{-3} for oxidised fatty acids and a less intense band for the unoxidised fatty acids (Figure 6). A similar result was reported by Poiana et al. (2015) (Poiana *et al.* , 2015), who used FTIR spectroscopy to evaluate the oxidation of edible oils after heating and frying. The band located at 3008 cm^{-3} in the spectrum of PO was assigned to the symmetric stretching vibration of the C-H bonds in the cis-double bonds (Araújo

et al. , 2011). This absorption was absent in the spectra of CNO and PKO because of the low amounts of unsaturated double bonds present in the oils. The simulated FTIR spectra from figure 6 &7 also showed located band spectra for the absorption band at 3009-3006 cm^{-3}. The absorption band at 2850-2920 cm^{-3} in the spectra of all the TOs can be assigned to C-H bonds for the symmetric and asymmetric stretching vibrations of the aliphatic CH_2 and CH_3 groups (Karabacak*et al.* , 2012). Another significant absorption band relevant for studying photooxidation was located near 1746 cm^{-3} in the spectra of the unexposed TOs. This band was assigned to the ester carbonyl functional group, which was more dominant in the less oxidised oils, as seen in the unexposed TOs (Poiana *et al.* , 2015). However, the vibrational frequencies of the absorption band decreased for the exposed TOs. Absorption bands were detected in the spectra of the oils at 1721 and 3505 cm^{-3} for the exposed CNO, 1720 and 3560 cm^{-3} for the exposed PO, and 1721 and 3554 cm^{-3} for the exposed PKO, and the reduction in these frequencies is attributed to the transformation of secondary oxidation products, such as aldehydes and other carbonyl group-containing compounds of a portion of the esters, during the oxidation process (Poiana *et al.* , 2015). These transformations caused the lowering of the vibrational frequencies of the ester groups with increasing absorbance. The simulated FTIR spectra also indicated a decrease in the absorption bands of the oxidised fatty acids. The simulated unoxidised lauric, palmitic, and oleic fatty acids exhibited a strong band at 1749 cm^{-3}, which decreased negligibly to 1741 cm^{-3} after oxidation. Another vital absorption band observed at 888–990 cm^{-3} was the bending vibration of the trans group of disubstituted olefins (Poiana *et al.* , 2015). This band provides related oxidation information about the bands at 3009 and 3550 cm^{-3}. The absorption band is extremely weak, with a low intensity compared with the other bands. Additional absorption bands and shifts in wavenumbers and their comparison to the simulated FTIR are summarised in Table 5. The results of the FTIR spectra from this study were comparable to those from the reported FTIR studies on edible oils (Guillen and Goicoechea, 2007). The results also agreed well with the simulated FTIR spectra shown in Figure 6 & 7.

CONCLUSIONS

The exposure of food to sunlight has substantial effects on its safety and quality. Therefore, this study focuses on the influence of sunlight exposure on tropical oils. The study showed a significant ($p< 0.05$) increase in FFA levels and PV for the exposed TOs. The IV and colour content also decreased significantly ($p <0.05$). FTIR analysis showed intense vibrational absorptions at 1721 and 3505 cm^{-3}, 1720 and 3560 cm^{-3}, and 1721 and 3554 cm^{-3} for the exposed CNO, PO, and PKO, respectively. These bands correspond to the secondary oxidation products, which were absent in the unexposed TOs. Simulation was performed to support the FTIR results, which also indicated the absorptions from the secondary oxidation products at 1744 and 3660 cm^{-1}. However, the rate of oxidation of different types of TOs differed significantly, and the trend followed the order: PO > PKO > CNO. This study, therefore, suggests several ways of minimising photooxidation, which should be confirmed by further studies: (1) reduce the exposure of TOs during processing, trading, and prolonged exposure to sunlight; (2) improve the packaging materials to reduce the transmittance of light through oils; and (3) increase the awareness on the consequences of exposing TOs to sunlight. Hence, the findings of this work are essential and need to be addressed to ensure appropriate handling of tropical oils during distribution and storage.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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Table 1: Changes in FFA profiles of TOs exposed and not exposed to sunlight for 7 weeks.[msd]

[a]**Time [weeks]**	**CNO**	**CNO**	**PO**	**PO**	**PKO**	**PKO**
	[b]Control	[c]Test	[b]Control	[c]Test	[b]Control	[c]Test
0	1.32±0.01	1.32±0.01	8.42±0.01	8.42±0.01	3.81±0.01	3.81±0.01
1	1.32±0.01	1.83±0.02	8.42±0.03	8.76±0.03	3.83±0.02	3.98±0.01
2	1.33±0.02	2.11±0.04	8.45±0.01	9.35±0.01	3.84±0.01	4.22±0.02
3	1.34±0.02	2.67±0.02	8.48±0.01	10.27±0.02	3.86±0.02	4.64±0.02
4	1.35±0.01	3.17±0.01	8.50±0.01	11.34±0.01	3.86±0.03	5.04±0.03
5	1.37±0.03	3.39±0.01	8.53±0.02	13.05±0.02	3.90±0.01	5.67±0.01
6	1.40±0.01	3.65±0.01	8.57±0.01	14.71±0.01	3.91±0.01	6.14±0.01
7	1.40±0.01	3.75±0.03	8.68±0.02	15.04±0.03	3.96±0.03	6.59±0.03

[msd] ± FFA (%) values indicate the 95% confidence interval for the respective mean and standard deviation.[a] Time (weeks) represents 8 h of exposure per day for seven weeks. [b] Control represents all TOs stored away from sunlight for 7 consecutive weeks.[c] Test represents the TOs exposed to sunlight for seven consecutive weeks.

Table 2: Changes in PV profile of TOs exposed and unexposed to sunlight for 7 weeks.[msd]

[a]**Time [weeks]**	**CNO**	**CNO**	**PO**	**PO**	**PKO**	**PKO**
	[b]Control	[c]Test	[b]Control	[c]Test	[b]Control	[c]Test
0	2.03±0.01	2.03±0.01	3.09±0.01	3.09±0.01	6.13±0.01	6.13±0.01
1	2.04±0.01	2.28±0.02	3.10±0.03	3.74±0.02	6.13±0.01	6.35±0.03
2	2.04±0.03	2.93±0.04	3.12±0.01	5.07±0.03	6.15±0.02	7.04±0.01
3	2.07±0.01	3.13±0.02	3.12±0.01	6.16±0.05	6.16±0.02	7.77±0.03
4	2.09±0.02	3.84±0.03	3.15±0.02	7.43±0.03	6.18±0.01	8.39±0.02
5	2.10±0.01	4.06±0.01	3.17±0.02	8.00±0.01	6.20±0.02	8.69±0.01
6	2.13±0.01	4.32±0.02	3.19±0.01	8.28±0.02	6.23±0.02	9.25±0.01
7	2.14±0.02	4.85±0.03	3.22±0.04	8.43±0.05	6.23±0.04	9.33±0.03

[msd] ± PV (meq/kg) values indicate the 95% confidence interval for the respective mean and standard deviation.[a] Time (weeks) represents 8 hours of exposure per day for 7 weeks. [b] Control represents all TOs

stored away from sunlight for seven consecutive weeks.[c] Test represents the TOs exposed to sunlight for seven consecutive weeks.

Table 3: Changes in IV profile of TOs exposed and unexposed to sunlight for 7 weeks.[msd]

[a]Time [weeks]	[a]Time [weeks]	[a]Time [weeks]	CNO	CNO	CNO	PO	PO	P
			[b]Control	[c]Test	[c]Test	[b]Control	[b]Control	[c]T
0	6.10±0.01	6.10±0.01	6.10±0.01	6.10±0.01	6.10±0.01	49.35±0.01	49.35±0.01	49
1	6.10±0.01	6.10±0.01	6.10±0.01	5.93±0.02	5.93±0.02	49.33±0.02	49.33±0.02	49
2	6.09±0.02	6.09±0.02	6.09±0.02	5.76±0.01	5.76±0.01	49.33±0.02	49.33±0.02	48
3	6.08±0.01	6.08±0.01	6.08±0.01	5.62±0.02	5.62±0.02	49.30±0.01	49.30±0.01	47
4	6.07±0.01	6.07±0.01	6.07±0.01	5.38±0.03	5.38±0.03	49.27±0.01	49.27±0.01	46
5	6.07±0.02	6.07±0.02	6.07±0.02	5.17±0.02	5.17±0.02	49.22±0.02	49.22±0.02	45
6	6.04±0.02	6.04±0.02	6.04±0.02	4.94±0.04	4.94±0.04	49.20±0.01	49.20±0.01	44
7	6.03±0.02	6.03±0.02	6.03±0.02	4.94±0.04	4.94±0.04	49.19±0.03	49.19±0.03	44

[msd] ± IV (g of iodine / 100 g of oil) values indicate the 95% confidence interval for the respective mean and standard deviation. [a] Time (weeks) represents 8 h of exposure per day for 7 weeks. [b] Control represents all TOs stored away from sunlight for seven consecutive days.[c] Test represents the TOs exposed to sunlight for seven consecutive weeks.

Table 4: Changes in colour profile of TOs exposed and unexposed to sunlight for 7 weeks.[msd]

[a]Time [weeks]	[a]Time [weeks]	[a]Time [weeks]	CNO	CNO	CNO	PO
			[b]Control	[c]Test	[c]Test	[b]Control
0	1.11R + 3.40Y	1.11R + 3.40Y	1.11R + 3.40Y	1.11R + 3.40Y	1.11R + 3.40Y	3.63R +9.40Y
1	1.10R + 3.40Y	1.10R + 3.40Y	1.10R + 3.40Y	0.92R + 2.98Y	0.92R + 2.98Y	3.63R +9.40Y
2	1.09R + 3.38Y	1.09R + 3.38Y	1.09R + 3.38Y	0.82R + 2.91Y	0.82R + 2.91Y	3.61R +9.40Y
3	1.09R + 3.38Y	1.09R + 3.38Y	1.09R + 3.38Y	0.70R + 2.81Y	0.70R + 2.81Y	3.60R +9.38Y
4	1.08R + 3.38Y	1.08R + 3.38Y	1.08R + 3.38Y	0.66R + 2.78Y	0.66R + 2.78Y	3.60R +9.38Y
5	1.06R + 3.36Y	1.06R + 3.36Y	1.06R + 3.36Y	0.62R + 2.73Y	0.62R + 2.73Y	3.58R +9.37Y
6	1.06R + 3.36Y	1.06R + 3.36Y	1.06R + 3.36Y	0.53R + 2.68Y	0.53R + 2.68Y	3.58R +9.36Y
7	1.04R + 3.33Y	1.04R + 3.33Y	1.04R + 3.33Y	0.44R + 2.51Y	0.44R + 2.51Y	3.57R +9.36Y

[msd] R+Y values indicate the 95% confidence interval for the respective Lovibond Red and Yellow scales.[a] Time (weeks) represents 8 h of exposure per day for 7 weeks. [b] Control represents all TOs stored away from sunlight for seven consecutive days.[c] Test represents TOs exposed to sunlight for seven consecutive weeks.

Table 5: Summary of dominant functional groups in the TOs studied according to FTIR spectroscopy and computation modelling (Poiana*et al.* , 2015)(Vlachos *et al.* , 2006)(Alshuiael and Al-Ghouti, 2020).

Wavenumber (cm^{-1})	Wavenumber (cm^{-1})	Wavenumber (cm^{-1})	Ma
Experiment	**Simulation**	**Functional Group**	
3345-3350	3600-3700	Secondary oxidation products such as hydroperoxides and alcohols	Str
3000-3010	3000-3006	*Cis*-double bonds	Sy
2920-2925	2910-2970	Aliphatic CH_3 group	Sy
2850-2855	2850-2890	Aliphatic CH_2 group	As
1720-1744	1730-1750	Carbonyl compounds such as esters, aldehydes, and others	Str
1650-1660	1660-1670	*Cis*-olefins	C=

Wavenumber (cm^{-1})	Wavenumber (cm^{-1})	Wavenumber (cm^{-1})	Ma
1460-1470	1450-1470	Aliphatic CH_2 and CH_3 groups	C-I
1415-1420	1410-1430	*Cis*-disubstituted olefins	Ro
1375-1780	1360-1780	Aliphatic CH_2 group	C-I
1100-1120	1160-11700	C-O in ester groups	C-(
870-890	840-880	Trans group of disubstituted olefins	Be
720-725	720-735	Aliphatic CH_2	Ro

Figure Captions

Fig.1 Initial and final %FFA values for the exposed and unexposed TOs. Final values indicate the values obtained on the 7^{th} week. Values are reported as mean±SD at 95% confidence interval (N=4)

Fig.2 Initial and final peroxide values for the exposed and unexposed TOs. Final values indicate the values obtained on the 7^{th} week. Values are reported as mean±SD at 95% confidence interval (N=4)

Fig.3 Initial and final iodine value values for the exposed and unexposed TOs. Final values indicate the values obtained on the 7^{th} week. Values are reported as mean±SD at 95% confidence interval (N=4)

Fig.4 Initial and final colour content for the exposed and unexposed TOs. Final values indicate the values obtained on the 7^{th} week. Values are reported as mean±SD at 95% confidence interval (N=4)

Fig.5 ATR-FTIR spectra of the TOs exposed and unexposed to sunlight

Fig.6 Simulated IR spectra of the dominant fatty acids in TOs before oxidation

Fig.7 Simulated IR spectra of the dominant fatty acids in TOs after oxidation

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