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Some Biophysical and Biochemical Properties of Edible Oils Exposed To Environmental Conditions

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Abstract:

In daily life most oils and fats in local markets are exposed to environmental conditions such as storage on shelves, exposure to UV radiation and day light. The present study deals with the influence of these environmental conditions for a duration period of one year on some physicochemical properties of a number of edible oils used in daily life. These oils include olive oil, sesame oil, sunflower oil, corn oil, palm oil and cotton oils. The results revealed that significant changes occurred in some biophysical and biochemical properties of these oils mainly in flash point, viscosity, specific gravity, iodine number, peroxide value, acid value, and saponification value. The exposure to daylight was more effective rather than the other two environmental conditions. These changes resulted in variation in the oils structure and the formation of new fatty acids fragments which were confirmed using GC-MS and NMR techniques.

Keywords: Physico-chemical properties of edible oils environmental conditions, fatty acids composition.

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1. Introduction

In normal human diet, about 25% to 50% of the caloric intake consists of fats and oils. These substances are the most concentrated form of food energy in our diet. When metabolized, fats produce about 9.5 kcal of energy per gram. Carbohydrates and proteins produce less than half this amount. For this reason, animals tend to build up fat deposits as a reserve source of energy. They do this, only when their food intake exceeds their energy requirements. In times of starvation, the body metabolizes these stored fats. Even so, some fats are required by animals for bodily insulation and as a protective sheath around some vital organs. (1)
Many vegetable oils, such as olive oil, sesame (sim, sim) oil, sunflower oil, maize oil, coconut oil and others are consumed directly or used as ingredients in food, a role that they share with some animal fats, including butter and ghee. (2)The beneficial properties of olive oil are located in its chemical structure. This oil is rich in monounsaturated fatty acids, low in saturated and polyunsaturated fats, and high in antioxidants. (5)

Physically, oils are liquid and fats aresolid at room temperature. Chemically, both oils and fats are composed of triglycerides, as contrasted with waxes which lack glycerol in their structure. Although many different parts of plants may yield oil, in commercial practice, oil is extracted primarily from seeds. (1-3)

Oils can be heated and used to cook other foods. Oils that are suitable for this purpose must have a high flash point. Such oils include the major cooking oils; canola, sunflower, peanut and others. (4, 5).Oil quality is concerned with the present state of oil acceptability while oil stability is related to its resistance to future and environmental changes, e.g., oxidation, temperature, and humidity. (4)

Different chemical mechanisms, autoxidation and photosensitized oxidation are responsible for the oxidation of edible oils during processing and storage depending upon the types of oxygen. Two types of oxygen can react with edible oils. One is the atmospheric triplet oxygen, $^3O2$, and the other is singlet oxygen, $^1O2$. $^3O2$ reacts with lipid radicals and causes autoxidation, which is a free radical chain reaction. The chemical properties of $^3O2$ are to react with lipid radicals which can be easily explained by the molecular orbital of the oxygen. The $^3O2$ in the ground state with 2 unpaired electrons in the $2p\pi$ ant bonding orbital has a permanent magnetic moment with 3 closely grouped energy states under a magnetic field and is called triplet oxygen. $^3O2$ is a radical with 2 unpaired orbital in the molecule. It reacts with radical food compounds in normal reaction conditions according to spin conservation. Photosensitized oxidation of edible oils occurs in the presence of light, sensitizers, and atmospheric oxygen, in which $^1O2$ is produced. (6)

In this connection, and in general, it is stressed that positive health benefits would be achieved by deriving 30-40% of calories from dietary fats that have a 1:1 ratio of polyunsaturated to

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saturated fatty acids. [7] Lack of information on the composition and utilization of the many and varied oil seeds indigenous to the topic are more of problem than is the real shortage of oils that take place in most worldwide. [8]

This work aims to study the effect of some environmental factors such as storage, exposure to daylight and UV radiation on some biophysical and biochemical properties of some local edible oils used in daily life.

2. Materials and Methods:

Edible oil samples:
Six edible oils namely; olive oil, sesame oil, sunflower oil, corn oil, palm oil and cotton oil were used. The mass of each oil sample was 2 Kg. The oils were collected from the groceries in the local market, as fresh oil samples.

a) Samples Preparations:
Each sample was divided into two main parts as follows:

Part I: Was used as a control part for the estimation of the physical and chemical properties at the start point of the work.

Part II: Was divided into three sub-parts for studying the effects of storage, exposure to day light, and exposure to UV radiation on the biophysical and biochemical properties of these edible oils. All samples under the research all time at Rome temperature and tested measurements at 25 ºC**.**

b) Storage:
To study the effect of storage on the biochemical and biophysical properties of the considered oils, the oil bottles were wrapped in black paper and kept in suitable places far from daylight or UV radiation exposure, for a total period of one year.

c) Exposure to daylight:
To study the effect of exposure to daylight, bottles of the considered oil samples were exposed daily today light, to simulate the exposure of oil bottles on shelves in Moles and local markets. The exposure period extended up to one year.

d) Exposure to UV radiation:
To study the effect of exposure to UV radiation on the biochemical and biophysical properties of the considered edible oils, the samples were exposed daily for 12 hrs, up to one year. The UV radiation equipment was Truman GAI-16 with UV lamp, a mosquito killer apparatus. That is used in Moles and local markets**.**

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Methods:

a) The physicochemical properties:
For each oil sample, whether fresh or stored and/or exposed to environmental day light or UV radiation, the following parameters were measured, namely; saponification value[9], Iodine Number[10], Peroxide value (PV) [11], the Acid Value[12], Flash point [13], refractive index [14] by Abbe refractometer (Bausch and Lomb Abbe), viscosity[15], and the specific gravity.

b) Determination of fatty acid profile for edible oil: for this purpose, GC-MS, was used:
A flame ionization detector, A DB-225, 30 m x 0.25 mm ID and 0.15µm Restek Rtx-PCB column was used. The injector and detector temperatures were set at 250ºC.

c) Nuclear Magnetic Resonance (NMR):
The 1H NMR spectra were recorded on a JEOL JNM ECA 500 Plus spectrometer operating at 500 MHz, Each oil sample, weighing 20 mg was mixed with 400 µl of deuterated chloroform; this mixture was introduced into a 5 mm diameter tube. The acquisition parameters of 1H NMR were: spectral width 5000 Hz, relaxation delay 3 s, number of scans 32, acquisition time 3.744 s and pulse width 90, with a total acquisition time of 3.37 min. The experiment was carried out at 25ºC. Spectra were acquired periodically throughout the oxidation process.

3. Results and Discussion

a) The Biophysical Parameters:

Refractive index:
The effect of exposure to UV radiation or exposure to day light and even oils wrapped in black paper at the end of one year storage resulted in an increasing in the refractive index .The order of change was as follows: oils exposed to day light [1.460±0.008 to 1.467±0.008] > oils exposed to UV radiation [1.460±0.008 to 1.466±0.008] > oils wrapped in black paper [1.460±0.008 to 1.462±0.007]. It's clear , that the increase in refractive index mean value is more elevation as a result of exposure to day light than both the effect of storage or exposure to UV radiation. The variations of refractive index with time for the different types of tested edible oils, in case of exposure to daylight are illustrated in Fig. 1.

Viscosity at 25˚C:
The results of the present work concerning the viscosity of the oils at 25 ºC showed that exposure to day light or UV radiation and even in oils wrapped in black paper resulted in an increase in the viscosity of all the six considered edible oils. The results of the increase in viscosity at the end of one year storage time was in the order: oils wrapped in black paper [81.5±7 to 82.6 ±7 cp] < oils exposed to UV radiation [81.5±7 to 85.8 ±7 cp] < oils exposed

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to day light [81.5±7 to 86.1±7 cp]. It's clear, that, there is more increase in mean viscosity value at 25˚C as a result of exposure to day light than both the effect of storage or exposure to UV radiation. The variation of viscosity of the different oils as a result of exposure to daylight is illustrated in Fig. 2. The reason of why exposed samples to day light are more affected than that exposed to UV and control samples? That's because the day light contains IR radiation, visible light and UV radiation. And the reason of increasing viscosity, that's because the viscosity must be closely correlated with the structural parameters of the fluid particles. On the basis of published data concerning flow properties of oils, the oil viscosity has a direct relationship with some chemical characteristics of the lipids, such as the degree of unsaturation and the chain length of the fatty acids that constitute the triacylglycerol. The viscosity slightly decreases with increased degree of unsaturation [16] and according to our results the exposure to either day light or UV the double bond decreases and the linear configuration of the oils become closer to each other leading to decrease oil fluidity resulting in increased viscosity.

The present values represent slight elevating in viscosity owing to summation of three active changes which are the decomposition of few parts of oils, to short chain fatty acids, decreases degree of unsaturation and changes amount of functional groups in some parts of oil back bone chain. The first one leads to decrease in viscosity but the main change and the dominant one is the change in the functional groups which leads to elevation of viscosity. This is in agreement with another study, on various oil, which indicate that the oils contain more functional groups are the more viscous. [17]

Specific gravity:

The specific gravity of all the edible oils considered in this work is less than one at 25 ^{o}C. As a result of exposure to day light or UV radiation and even storage oils bottles in black paper, slight increase in specific gravity occurred. The elevation followed the order: oils wrapped in black paper [0.910±0.011 to 0.911±0.110] < oils exposed to UV radiation [0.910±0.011 to 0.916±0.112] < oils exposed to day light [0.910±0.011 to 0.917±0.112]. It's clear, that, there is increase in mean specific gravity value is. More elevation occurs as a result of exposure to day light than both the effect of storage or exposure to UV radiation. The variation of specific gravity of the different oils as a result of exposure to daylight is illustrated in Fig. 3.

Flash Point:

The changes in flash point was in the order: oils wrapped in black paper [326.5±1.2 ^{o}C to 324.6±1.5 ^{o}C] < oils exposed to UV radiation [326.5±1.2 ^{o}C to 319.5±1.7 ^{o}C< oils exposed to day light [326.5±1.2 ^{o}C to 318.8±2.9 ^{o}C]. This indicates the liberation of more fatty acids , i.e., the formation of short chain free fatty acids owing to decomposition of long chain fatty acids which are more volatile and hence more flammable, as a result of exposure to day light or UV radiation and even storage with wrapping oils in black paper. The long chain fatty acid under effects of exposure to the radiation lead to more saturated (elevation of iodine number)

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and so more viscous which agree with 0this linear "zigzag" organization enables the chains to be lined up close to each other and intermolecular -interactions such as Van der Waals interactions can take place. This system inhibits flow of the fluid, resulting in the relatively high viscosity of the oils.
Moreover, the least changes or reduction in flash point occurred during storage in black paper. The depression in mean value of flash point was more declined as a result of exposure to day light than both the effect of storage or exposure to UV radiation, which is clear in Fig.

b) Biochemical changes

Iodine number:
Our study denoted that the depression in iodine number mean value is more declined as a result of exposure to day light than both the effect of storage or exposure to UV radiation at the end of one year. The reduction in the mean of all oils iodine number followed the order: oils bottles wrapped in black paper [98±26 to 91.6±26] < oils exposed to UV radiation [98±26 to 96±25] < oils exposed to day light [98±26 to 89±24]. The depression in the iodine number indicates depression in the number of double bonds. This also indicates that the edible oils considered in this work resulted in the formation of saturated fatty acids due to the Decrease in the number of double bonds. This leads to affect the quality of the oil. The variation of iodine value with time in case of exposure to day light is illustrated in Fig. 5.

Peroxide Value:
The study denoted that the there is more increase in peroxide mean value as a result of exposure to day light than both the effect of storage or exposure to UV radiation. Exposure to day light or UV radiation and even storage of oils wrapped in black paper resulted in elevation of the peroxide levels in all the edible oils used in this work. The elevation of the mean value changes in the peroxide value followed the order: oils wrapped in black paper [3±1 to 5±2 mg eq. of O_2/kg $_{sample}$] < oils exposed to UV radiation [3±1 to 11±2 mg eq. of O_2/kg $_{sample}$] < oils exposed to day light [3±1 to 12±1 mg eq. of O_2/kg $_{sample}$]. The pattern of increasing is similar for all the oils used, as described in case of exposure to daylight, Fig. 6.

Saponification Value:
From our study, it was found that the saponification value of all edible oils used in this work increased after both exposures to day light or UV radiation and even in oils wrapped in black paper.
The increase in the mean saponification number was in the order: oils wrapped in black paper [191±6.6 to 193±6.4 mg KOH/1g_{sample}] < oils exposed to UV radiation [191±6.6 to 202±7.84 mg KOH/1g_{sample}] < oils exposed to day light [191±6.6 to 203±8.34 mg KOH/1g_{sample}].The variation of saponification value is more elevated as a result of exposure to day light than both

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the effect of storage or exposure to UV radiation. The pattern of increasing is similar for all the oils tested, as described in case of exposure to daylight, Fig. 7.

The acid value:
There was more elevation in the mean acid value as a result of exposure to day light than both of storage or exposure to UV radiation**.** The order of change was as follows: oils wrapped in black paper [0.12±0.05 to 0.20±0.07 mg of KOH/1g_{sample}] < oils exposed to UV radiation [0.12±0.05 to 0.59±0.02 mg of KOH/1g_{sample}] < oils exposed to day light [0.12±0.05 to 0.60±0.02 mg of KOH/1 g_{sample}]. Where the increasing in case of the both exposure to UV radiation and exposure to daylight are more but the pattern of increasing is similar for all the oils tested, as described in case of exposure to daylight, Fig. 8.

c) Determination of fatty acid profile for edible oil by using:

I. Gas Chromatographic / Mass Spectrometry

The six number of oils were analyzed by GC-Mass Spectrometry for estimating the composition of fatty acid Triacylglycerols (TAGs) and other constituents during storage and exposure to either UV or daylight radiation. The fatty acids composition percent for the oil in GC-MS peaks were calculated by dividing the single peak area over the total peaks area.
GC-MS is used to detect the difference in fatty acid between fresh oil samples and the tested samples exposed to UV radiation. After 6 months the wrapped sample in the black paper, is used as a control sample.

The results of using GC –MS, revealed that the main components of the control sample are at retention time 44.2 minute and 38 minute are 70 % 9-Octadecenoic acid methyl ester and 18% Hexadecanoic acid methyl ester, respectively. Exposure of olive oil to UV radiation the components of 9-Octadecenoic acid methyl ester reduced to 65.6% (~ 6 % decrease) and to 60 % (~14%) after six and eight month, respectively, while Hexadecanoic acid methyl ester did not suffer any changes (60%) after exposure to UV radiation.

In case of fresh sunflower oil (wrapped in black paper) the main constituents appeared at retention time 44.25 minute and 42.5 minute were 62.98% of 9-12-Octadecadienoic acid methyl ester and 17% 9-Octadecenoic acid methyl ester respectively. The exposure to UV radiation for six months resulted in decrease in the content of fatty acids to 58.28% (~7.5% reduction), 54.68% (~13% reduction) in 9-12-Octadecadienoic acid methyl ester respectively while the exposure for 8 months UV radiation resulted in lower values of 15% (~12% reduction) and 12.4% (reduction of ~ 27%) in 9-Octadecenoic acid methyl ester, receptively.

In case of fresh corn oil, the main components at retention time 44.2 minute and 42.5 minute were 54%9-12-Octadecadienoic acid methyl ester and 30.5 % 9-Octadecenoic acid methyl ester respectively. The exposure to UV radiation for six months resulted in the decrease in fatty acid contents to of these acid 48.8% (~10% reduction) and to 27.4% (~10%reduclion),

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respectively. The exposure to 8 months of UV radiation on the other hand, resulted in more reduced contents to 46.8 % (~13%reduclion) and to 26.4% (~13%reduclion) in 9-12-Octadecadienoic acid methyl ester and 9-Octadecenoic acid methyl ester respectively.

In case of fresh palm oil, the main components at retention time 39 minute and 44. 5 minute the main components were 43.5 % Hexadecanoic acid methyl ester and 39.8 % C- 9-Octadecenoic acid methyl ester, respectively. The Exposure to UV radiation for six and eight months resulted in no changes in Hexadecanoic acid methyl ester. However, the same exposure periods resulted in reduced values to 35.5% (~11% reduction) and reduced 31.2% (~22% reduction) in 9-Octadecenoic acid methyl ester, respectively.

In case of sesame, at retention time 44.2 minute and 46.9 minute, the main components were 42% 9-Octadecenoic acid methyl ester, 41% 9-12-Octadecadienoic acid methyl ester, respectively. Exposure to UV radiation reduced 9-Octadecenoic acid methyl ester To 40%(~10%reduction),and to 37% (~12% reduction) in 9-Octadecenoic acid methyl ester after exposure to six months and to 38.1% (~7%reduction) and to 35.1% (~12% reduction) in 9-12-Octadecadienoic acid methyl ester ,respectively, after 8 months exposure to UV radiation.

In case of fresh cotton oil, the main components at 44.9 minute and 37.9 minute retention time were 54.7% 9-12-Octadecadienoic acid methyl ester and 21.5% Hexadecanoic acid methyl ester, respectively. The exposure to UV radiation resulted in lower level to 49.1% (~10% reduction) and to 45 % (~18%reduction) in 9-12-Octadecadienoic acid methyl ester after exposure for six and 8 months, receptivity.

The changes in the 9-Octadecenoic acid methyl ester occurred after exposure to UV radiation may be due mainly to the reduction in C=C as a result of exposure to UV radiation. On the other hand, the stability of C- 9-Octadecenoic acid methyl ester before and after exposure to UV radiation is due to mainly the absence of C=C, as it is a saturated fatty acid.

The results of the analysis by gas chromatography indicated very interesting results. The exposure to UV radiation resulted in the liberation of glycerol which is a good indication of the decomposition of the triglycerides. Hexadecanoic acid (C16:0) and Octadecanoic acid (C18:0) did not suffer any changes because they are saturated fatty acids. However, 9-Octadecenoic acid (C18:1), 9-12-Octadecadienoic acid (C18:2) and 9-12-15-Octadecanoic acid (C18:3) decreased in their percentages after exposure to UV radiation. The reduction in the percentages of C=C, C=C-C=C and C=C-C=C-C=C in these acids, respectively. The exposure to UV radiation resulted in the breakdown of the double bonds. This depression of double bond lead to the decrease of the iodine number [18], and increases of the free fatty acids and the formation of new fragments which are in good agreement with the decrease of flash point.[19,20] and the formation of new functional groups that tend to increase of both the peroxide value and the oil viscosity.[21,22]

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II. Nuclear Magnetic Resonance

Studying the nuclear magnetic resonance spectrum for oils, as a biological material, describe ,in general, a number of regions which can be considered as a base line of comparison to differentiate the properties of fresh oils from that exposed to any physical or chemical factors. So, the aim was to use this technique to introduce an evaluation of the biochemical and/or the biophysical changes occurred as a result of exposure of the oils to either day light or UV radiation and storage wrapped in black paper.

The base line regions are as follows:

- **Region 1** is known as the methyl group region. It ranges between 0.8 – 0.9 ppm.
- **Region 2** is known as the allyl region. It ranges between 1.9 – 2.1 ppm.
- **Region 3** is known as the bisallyl CH2. It ranges between 2.7 – 2.8 ppm.
- **Region 4** is known as the olefeinic,.i.e, CH=CH. It ranges between 5.2 – 5.4 ppm.
- **Region 5** is known as the hydroperoxides. It ranges between 5.9 – 7.2 ppm.
- **Region 6** is known as the aldehyde group, CHO. It ranges between 9.5 – 9.7 ppm.

The results concerning the use of nuclear magnetic resonance, and using the considered six types of edible oils wrapped in black paper or submitted to exposure to UV radiation for six or eight months exposure, showed no changes in the methyl region in the range from 0.8 – 0.9 ppm, which is attributed to the fact that this region, i.e., the methyl group represents a non-active group.

However, exposure of oils to UV radiation for six or eight months resulted in changes in the five remaining regions. Table 1: describes the changes in the number of protons, as an example, as a result of exposure to eight months UV radiation. It is clear from this table that the change in the proton number described in the allyl region, the bisallyl region, and the olefeinic region these oils after exposure to UV radiation tend to shift towards increase of saturation, i.e., reduced number of double bonds. This is going side by side with the results of the iodine number which decreased with increasing the exposure to either day light or UV radiation, Fig.9.

On the other hand, in the hydroperoxides region, and in the peroxide group and aldehyde group region, the changes in the proton number in these two groups are more interested; because of the formation of either the peroxide or the aldehyde group occurred in these types of edible oils considered in this work. These groups produce free radicals and short chain fatty acids which add flavor to the oil which is known as rancidity.

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Table 1: Percentage changes in the proton number after exposure to eight months UV radiation.

Region / Oil	1.9 – 2.1ppm	2.7 – 2.8ppm	5.2 – 5.4ppm	5.9- 7.2 ppm	9.5- 9.7ppm
Sunflower	40	98	60	100	100
Corn	40	75	75	100	100
Cotton	0	33.3	40	100	0
Sesame	22.2	50	20	100	100
Olive	20	40	30	33	50
Palm	2	40	28	15	44

In fact, 1H NMR spectroscopy has proved to be very useful in evaluating the oxidative status of oils and fats, determining their oxidative stability, monitoring oil degradation processes, as well as in providing information on the nature and proportions of the aldehydes generated in these processes which are present in the oil liquid phase. The chemical properties results were confirmatory to those obtained by the physical properties results which are agreed by the results obtained by NMR spectrum. For all oils under investigation, the evaluation of the changes due to degradation conditions by exposure to day light or UV radiation.

4. Conclusion

The exposure of some edible oils, namely; olive oil, corn oil, sunflower oil, palm oil, sesame oil and cotton oil to UV or day light and even storage oil bottles wrapped in black paper induced some biophysical changes in refractive index, viscosity, flash temperature, and some biochemical changes in saponification number, iodine number, acid value and peroxide value that must be taken into consideration. The effect of exposure to day light proved to be higher than that produced as a result of exposure to UV radiation or storage wrapped in black paper. The storage of oils wrapped in black paper proved to be of least effect with respect to the two other exposure procedures used, for period less than four months.

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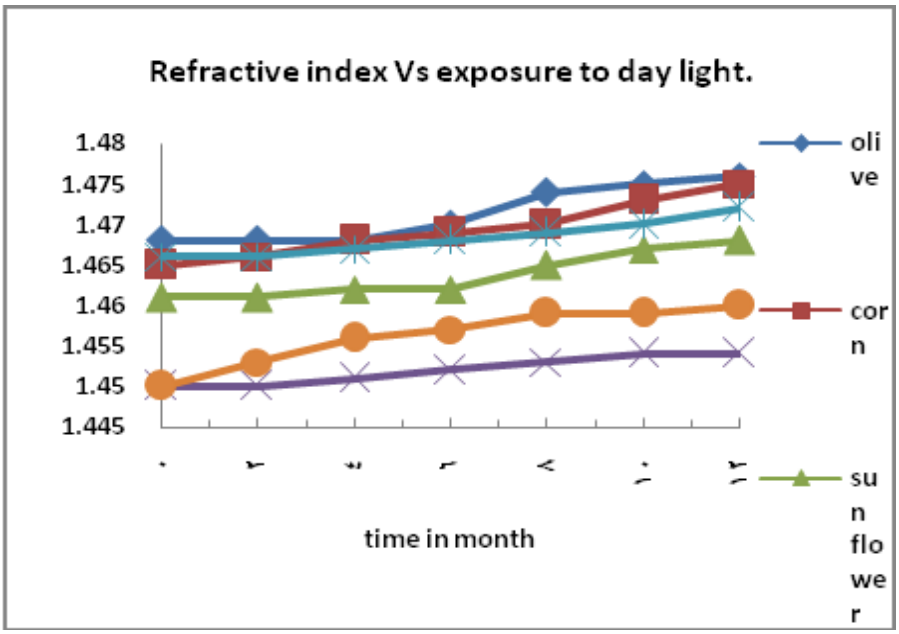

Fig. 1: Variation of refractive index with time for the different types of tested edible oils.

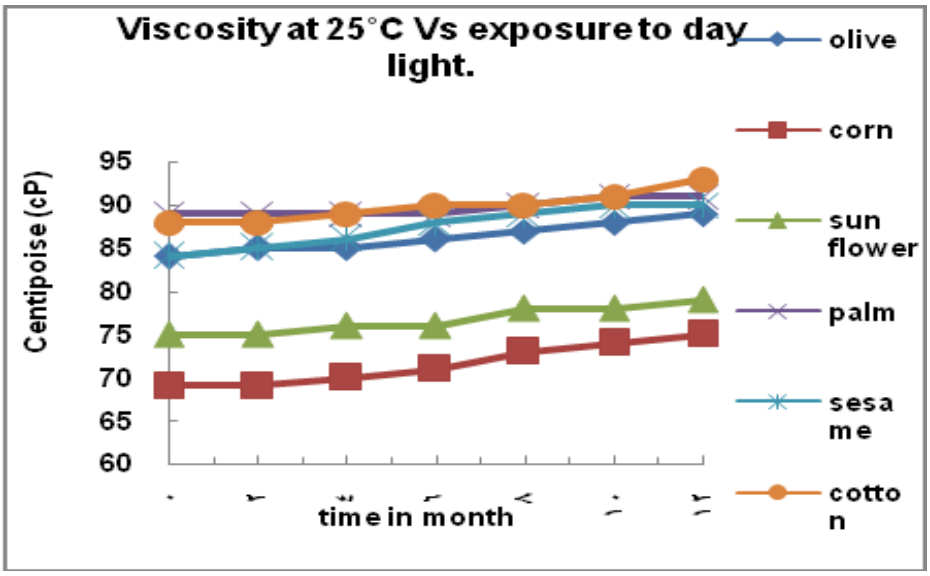

Fig. 2: Variation of viscosity with time for the different types of tested edible oils.

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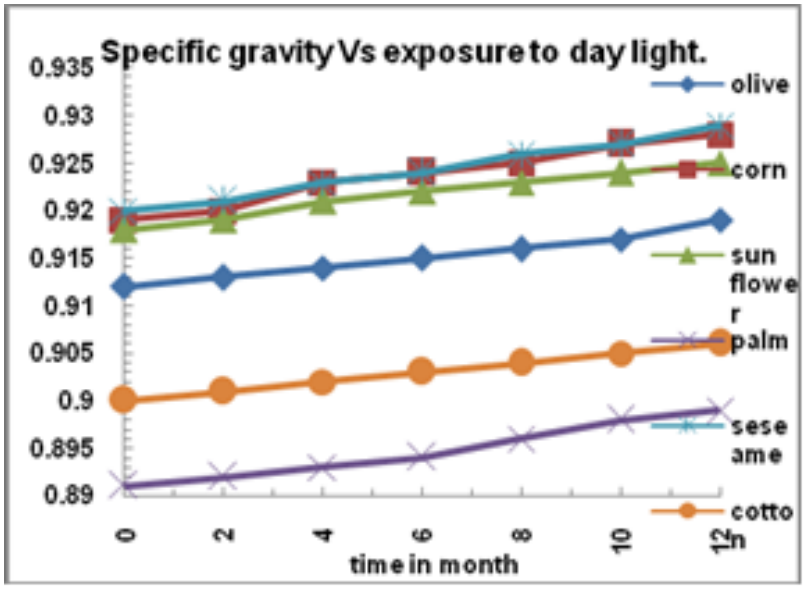

Fig. 3: Variation of the specific gravity with time for the different types of tested edible oils.

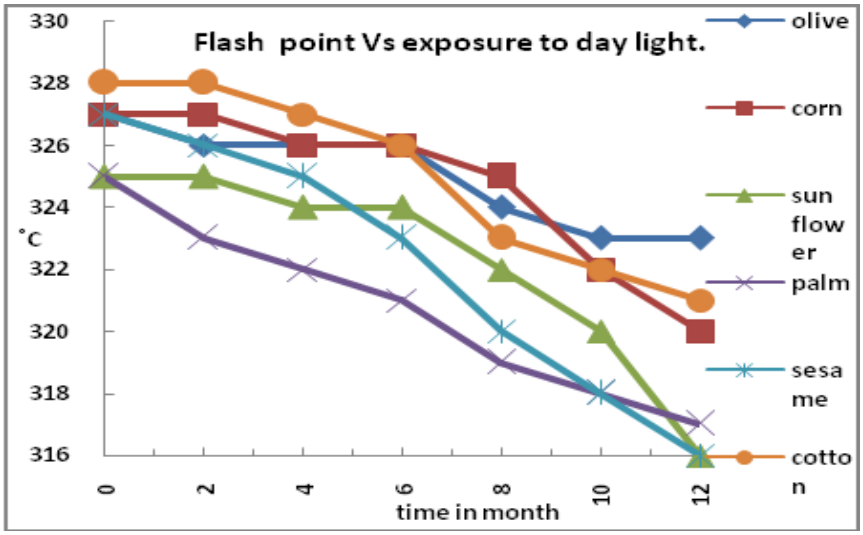

Fig. 4: Variation of the flash point with time for the different types of tested edible oils.

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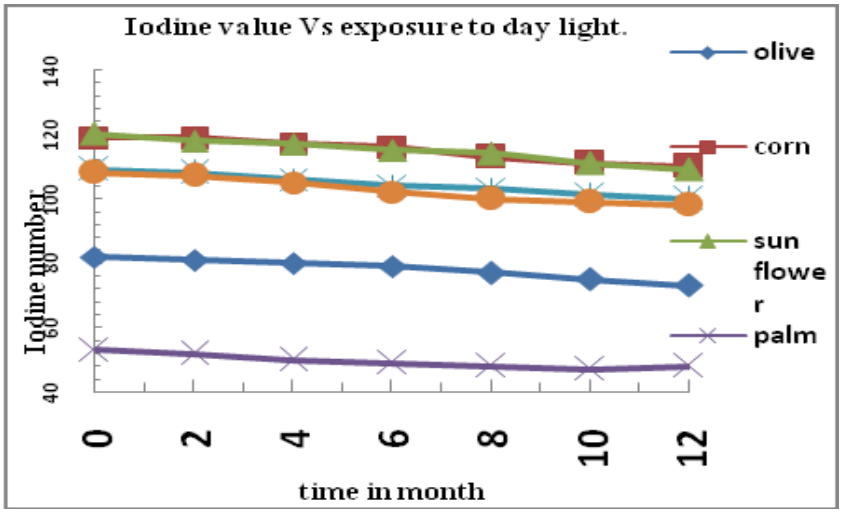

Fig. 5: Variation of the iodine value with time for the different types of tested edible oils.

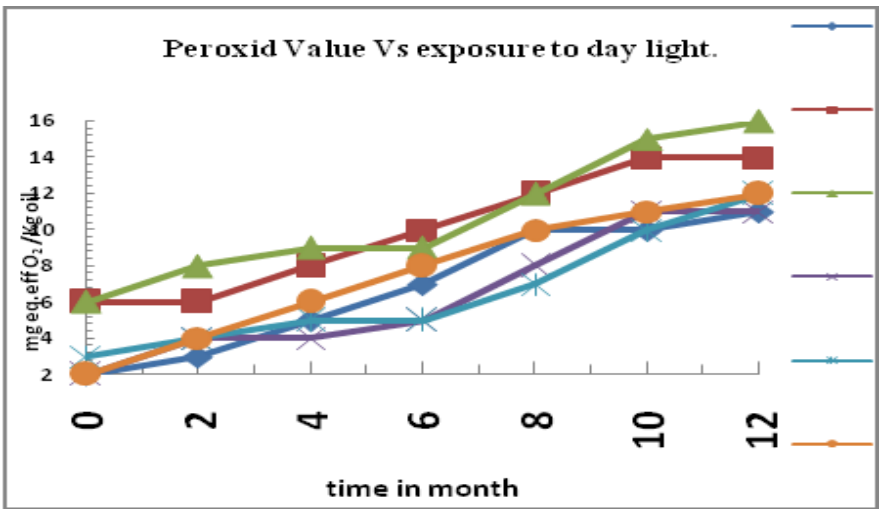

Fig. 6: Variation of the peroxide value with time for the different types of tested edible oils.

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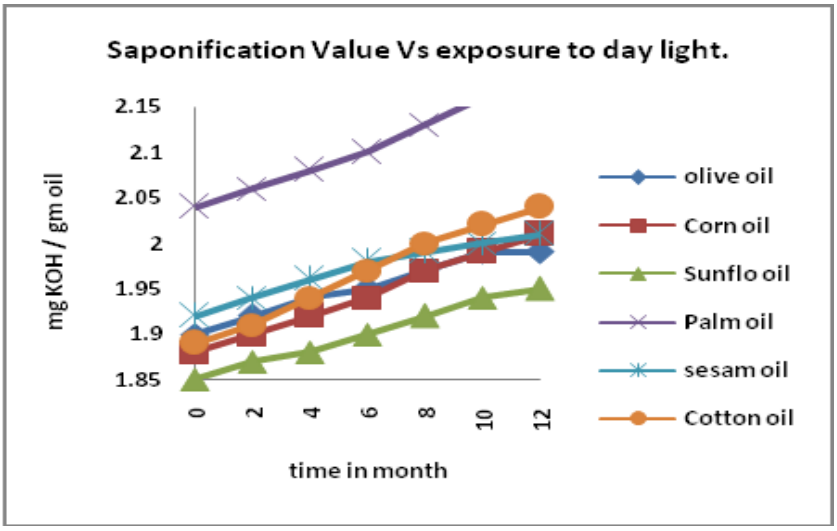

Fig. 7: Variation of saponification value with time for the different types of tested edible oils.

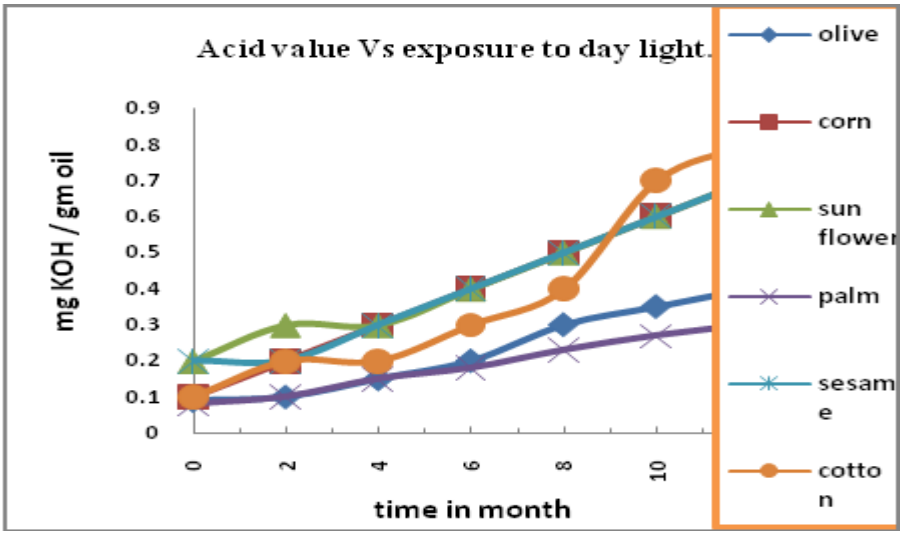

Fig. 8: Variation of acid value with time for the different types of tested edible oils.

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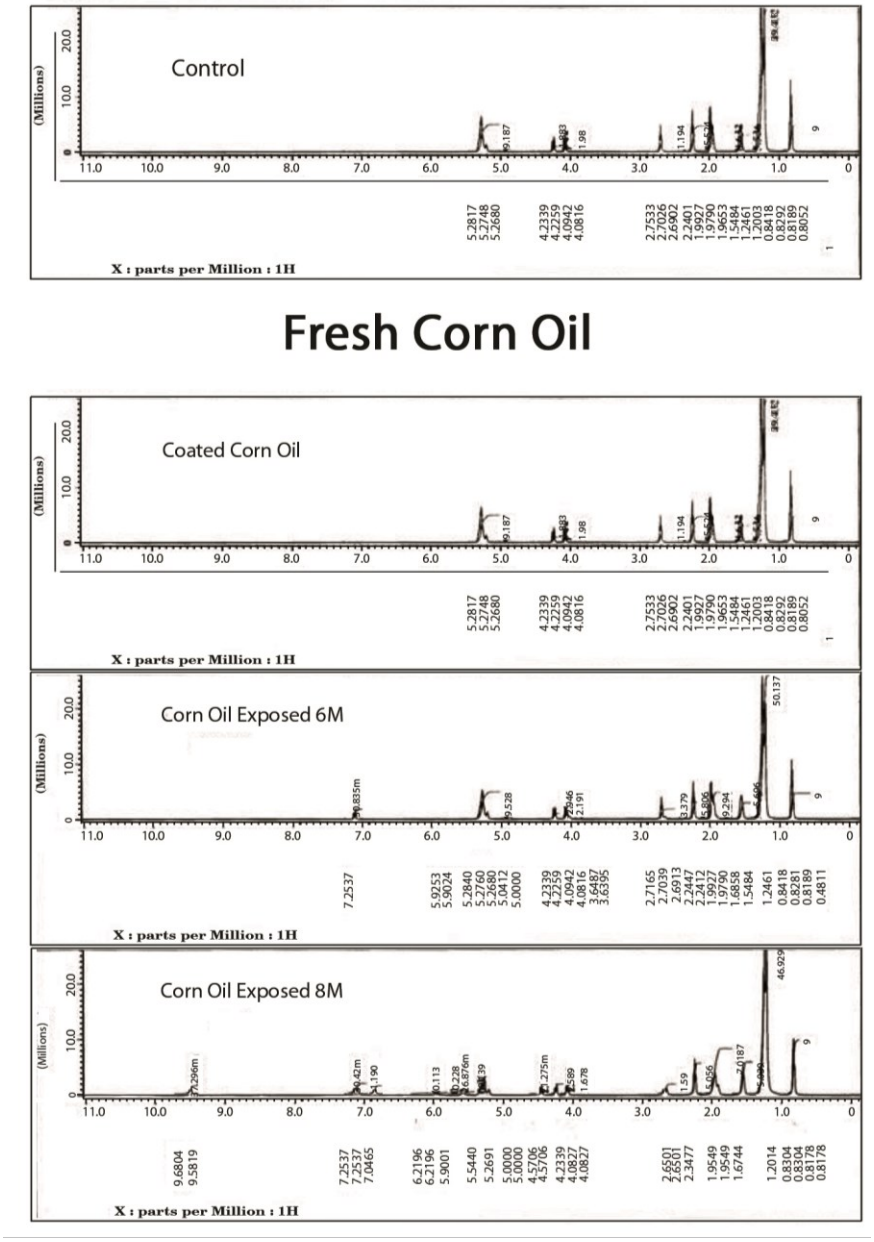

Fig.9:1H-NMR spectrum of corn oil in contrast to the corn oil exposed to UV radiation.