Short note

Fatty acids profile, atherogenic and thrombogenic health lipid indices of refined vegetable edible oils and their thermal stability

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In Algeria, olive oil is not available in large quantities, which explains its high price and consequently the use of refined vegetable oils by households for frying. It is well known that, unlike oils extracted from seeds, olive oil has a beneficial effect on health due to its resistance to oxidation. The focus of this paper is two-fold; the first objective is to present the fatty acids (FAs) composition and pro-oxidant heavy metals content of five brands of the most popular Algerian vegetable edible oils, namely: LaBelle, oleor, elio, afia and fleurial. The second objective is to assess their degree of oxidation during heating. The atherogenic and thrombogenic indices were calculated from the FAs composition of the same five Algerian brands of vegetable edible oils. In order to evaluate their oxidative stability, these oils were subject to a Rancimat test and thermogravimetric analysis to calculate activation energy. Our results showed that these oils recorded high polyunsaturated fatty acids (PUFAs) content (mean 60%) and contain pro-oxidant metals (iron and copper). "Feurial" sunflower oil is more sensitive to oxidation than "LaBelle" and "oleor" soybean oils. In fact, sunflower oil, which had the highest PUFAs content (mean 65%) and the highest pro-oxidant copper content (0.058 mg/kg) displayed the lowest induction period (8.27 h) and the lowest activation energy (438.16 kJ/mol) compared to soybean oils (55% PUFAs, 0.004 mg Cu/kg, 12 h and 566 kJ/mol) respectively. The presence of PUFAs at high levels and transition metals even in small amounts enhance the sensitivity of these oil to oxidation during heat processing. Sunflower oil, which is more susceptible to oxidation, should only be used for seasonings.

Keywords: Refined oils; fatty acid composition; heavy metals; oxidative stability; atherosclerosis.

1. INTRODUCTION

Vegetable oils are the most consumed food in the world. In our country, refined vegetable oils are mainly used for frying, cooking and baking because of their lower cost compared to olive oil. These oils have a high nutritional value due to the high content of polyunsaturated fatty acids (PUFAs), especially linoleic acid (18:2, ω6) [1]. It should be noted that PUFAs found in refined vegetable oils are endowed with nutritional and therapeutic properties. However, these oils are very sensitive to oxidation.
In addition, several factors influence the oxidative instability of edible oils. These include the composition of oil in fatty acids (FA) and the presence of pro-oxidant heavy metals like copper (Cu) and iron (Fe). The rate of oxidation reactions increases with the degree of unsaturation of fats [2]. α-linolenic acid (18:3, ω3) oxidizes the fastest, followed by linoleic (18:2, ω6) and oleic acids (18:1, ω9) [3]. Furthermore, heavy metals in vegetable oils can accelerate oil oxidation even by a very tiny amount by promoting the formation of free radicals from FAs or hydroperoxides [4, 5, 6]. Free radicals in fried foods cause ischaemic heart disease and aging.

Until now, nothing or little has been known about the content of heavy metals in Algerian edible vegetable oils. This study aims to determine the physical and chemical parameters of the five most consumed brands of vegetable oils in Algeria, for example: FAs profile, free fatty acids (FFA) content, atherogenic, thrombogenic and peroxide indices and iodine value (IV). Moreover, this research was carried out in order to shed light on the oxidative stability of the same five brands of Algerian vegetable edible oils, such as: the Rancimat test and calculating the energy of activation using thermogravemetric analysis.

2. MATERIALS AND METHODS

2.1. REFINED OILS STUDIED

One litre bottles of each of the five most consumed brands of Algerian vegetable edible oils were puchased in a local supermarket, namely: fleurial, LaBelle, oleor, elio and afia with a shelf life of two years. The five bottles were kept in the fridge at 4°C until subsequent analyses.
It should be noted that fleurial is a pure sunflower oil, wherea LaBelle and oleor are pure soybean oils. Conversely, elio and afia are blended oils and are prepared by blending soybean and sunflower oils at a ratio of 80:20 for the first oil and the blending of soybean with corn oils at a ratio of 95:5 for the second oil. Both the physical and chemical characteristics of these oils were determined; each oil was analyzed in triplicate.

2.2. PHYSICAL PARAMETERS ANALYSIS

2.2.1 Fatty acid analysis

The FA composition of the five brands of Algerian vegetable edible oils was determined by gas chromatography (GC) in order to evaluate the content of PUFA that are sensitive to the oxidation process. The presence of two double bonds in the FA structure causes faster oxidation than with a single double bond.
The FAs composition of the samples was determined as fatty acid methyl esters (FAMEs). FAMEs were obtained by the transesterification of edible oils with BF3-MeOH after alkaline hydrolysis [7]. 1 mg of the vegetable oil sample and 500 µL of C17 were mixed with 1 mL of 0.5N methanolic NaOH, and the mixture was saponified at 70°C for 15 minutes. After cooling to room temperature, 1 mL 14% BF3-MeOH solution was added and the mixture was heated at 70°C for 10 minutes to promote the formation of the methyl ester. Again, the mixture was cooled to room temperature followed by the addition of 6 mL of distilled water and 2 mL of pentane. The solution was allowed to settle until two layers were formed, and the supernatant was collected and transferred into GC vials and frozen at -20°C prior to analysis. FAMEs were analyzed GC-MS using an Agilent 7890A gas chromatograph system equipped with a flame ionization detector. Separation was performed on a capillary fused silica column (30 m × 0.25 mm). The stationary phase consisted of 80% biscyanopropyl and 20% cyanopropylphenyl while hydrogen was used as a carrier gas. The temperature was set at 45, 195, 220 and 240°C for 2, 7, 2 and 2 minutes respectively for a total duration analysis 21.9 minutes. The injector and temperature detector were maintained at 220 and 280°C, respectively. The results were expressed as percentage of identified FAs.

2.2.2 Atherogenic and thrombogenic index

The polyunsaturated/saturated (P/S) ratio considerably influences the occurrence of coronary heart disease. However, this parameter has been replaced by indices of atherogenicity and thrombogenicity [8].
The atherogenic and thrombogenic indices (A.I. and T.I.) were calculated from the FAs composition of the five brands of Algerian edible oils according to the following equations, described in a previous study [9].

$$AI = \frac{C12:0 + 4 * C14:0 + C16:0}{\sum AGMI + \sum AGPI,\omega 6 + \sum AGPI,\omega 3}$$

$$AI = \frac{C14:0 + C16:0 + C18:0}{0{,}5 * \sum AGMI + 0{,}5 * \sum AGPI,\omega 6 + 3 * \sum AGPI,\omega 3}$$

where C12:0, C14:0, C16:0, C18:0, ω3 and ω6 are lauric, myristic, palmitic, stearic, linoleic and linolenic acids, respectively.

2.2.3 Pro-oxidant heavy metals content

Trace metals in refined vegetable oils may originate from the soil and fertilizers, or from contamination during processing and storage. The presence of small amounts of trace metals in oils and fats is known to negatively affect oil quality. The strongest and most notable pro-oxidants are copper and iron, which produce a significant oxidative effect at concentrations as low as 0.005 and 0.03 ppm respectively [10].
In our study, the measurement of heavy metals in fresh refined vegetable edible oils was carried out using a Perkin Elmer PINAACLE 900F graphite flame atomic absorption spectrometer. Results obtained are expressed in mg per kg of oil. Previously, a calibration curve was drawn using increasing known concentrations of heavy metals.
To establish the calibration curve for the determination of heavy metals (iron and copper) in oil samples by Atomic Absorption Spectrocopy (A.A.S.), 1 mL of reference solution (1000 ppm) was transferred to a 100 mL volumetric flask and filled to the mark with distilled water to form the stock solution (100 ppb). From the latter, 1mL was transferred to a 100mL volumetric flask and filled to the mark with

double-distilled water. Lastly, 40 µL of each solution underwent spectrophotometry, which automatically established the calibration curve. For the determination of heavy metal in oils studied by A.A.S., 3 g of the oil studied was placed in a graphite oven (MAB-FM74) at a gradually increasing temperature of 25-500°C (2°C/min) in order to allow the slow charring of the sample until all the carbon had been removed. To the cooled ash, a 5 mL aliquot of HNO_3 (60%) was added to promote dissolution. A nitric acid solution was filled to 10 mL and diluted 10 times. Finally, 50 µL were injected in a graphite tube for analysis; the results were displayed automatically. All reagents used were of analytical-reagent grade.

2.3. CHEMICAL PARAMETERS ANALYSIS

The six brands of Algerian edible oils were chemically characterized according to the AFNOR method [11].

2.3.1 Free fatty acid content

The FFAs are one of the chemical parameters that impact considerably on the quality of edible oils. These molecules turn oil rancid. Rancidity is the unpleasant odour and that develops spontaneously in oils. There are two type of rancidity: hydrolytic and oxidative. Acid value (AV) is a measurement of the FFAs content in the oil. The higher the AV, the higher the level of FFAs, which translates into lower oil quality. During refining, FFAs are very carefully removed because they promote oxidation, thus fully refined edible oils become less prone to oxidation. The percentage of FFAs is the amount of potassium hydroxide expressed in milligrams required to neutralize the FFAs present in one gram of fat or oil. FFAs content was determined according to the method of [11].

2.3.2 Peroxide index

The peroxide value (PV) of oil samples was measured according to the method of [11]. This technique is based on iodometric titration that quantifies iodine produced from potassium iodide by peroxides present in oil. The results were expressed as milliequivalents of active oxygen per kilogram of edible oil. PV highlights the extent of primary oxidation and is a key indicator of oil quality and potential rancidity.

2.3.3 Iodine value

The I.V. indicates the degree of unsaturation of edible oils. It is defined as the number in grams of iodine absorbed by 100 g of oil. The I.V. value was determined using method [11].

2.4. OXIDATIVE STABILITY

In this investigation, the Rancimat test and calculation of activation energy were used in order to compare the thermal oxidation stabilities of the five brands of refined oils.

2.4.1 Rancimat measurement

The Rancimat analyses served to evaluate the oxidative stability of oils and the latter was expressed as the oxidative induction period expressed in hours and defined as the duration of oil's natural resistance to oxidation, due to the presence of naturally occurring antioxidants which delay oxidation during frying [12]. Rancimat tests of all the samples were performed according to the method of [13] with 743 Rancimat apparatus (Methrom AG, Herissau, Switzerland). It should be noted that 3 g of oil was weighed in the reaction vessel and heated at 98°C under a constant air flow (10 L/h). The induction times were printed automatically by the apparatus software with an accuracy of 0.005. All analyses were carried out in triplicate and the mean value was recorded.

2.4.2 Calculating activation energy

Thermal deterioration of the oils due to lipid oxidation can be also examined by thermogravimetric analysis (T.G.A.). The oxidation process in oil was assessed by monitoring mass change (gain or loss) as a function of temperature or time [14, 15]. In this study, the thermogravemetric analyses of oils were carried out using «NETZSCH STA 449C» apparatus. TGA involves heating oil samples of 2-5 g from 20°C to 500°C, under a constant air flow (25 mL/min) and constant heating rates. Activation energies (Ea) were calculated by means of Arrhenius equations. Activation energy is the minimum energy required to start oxidation reaction. Therefore, it is an indicator of thermal stability and degradation behaviour of edible oils.

2.5. STATISTICAL ANALYSIS

Statistical analyses were performed with The Stat. Box software. The data were analyzed by one-way analysis of variance (ANOVA), and the means were compared by the least significant difference (LSD) test at a significance level of 0.05.

3. RESULTS AND DISCUSSION

3.1. REFINED VEGETABLE OILS CHARACTERISTICS

Refined vegetable oils are cheaper than olive oil, which makes them a popular choice among Maghreb households with low purchasing power. In Algeria, olive oil is expensive, costing 10 Euros/L, so the majority of the population cannot afford it. Conversely, refined sunflower oil is prized by a large segment of the Algerian population in salad dressings, not only because it is cost-effective, but it also contains many added synthetic vitamins as A, D and E. Moreover, sunflower oil does not impart any new flavour when used for seasoning salads, thereby maintaining the unique flavour profile of leafy vegetables.

3.2. LEVELS OF PHYSICAL PARAMETERS

The results regarding the physical parameters of the five most consumed brands of Algerian vegetable edible oils are shown in Table I. The physical parameters are within the official ranges for these oils as specified in the Codex Alimentarius [16].

3.2.1 Fatty acids composition

Table I shows that there were significant differences between the five brands of edible oils in terms of FAs composition. The content in saturated fatty acids (S.F.As.) ranged from 10.959% in "fleurial" (pure sunflower oil) to 15.331% in "LaBelle" (pure soybean oil). In addition, palmitic acid (C16:0) was the major SFA in all our oils, followed by stearic acid (C18:0). The percentage of palmitic acid was the lowest in "fleurial" oil (6.472%) and the highest in "LaBelle" oil (10.624%).

Our results regarding total SFAs are in accordance with those reported by Zambiazi et al [17]; Tasan et al. [18]; Kostik et al. [19]; Kaur et al. [20]. As they pointed out, soybean oils are more saturated compared to sunflower oils; the values obtained were respectively 15.10 *v.s* 12.36%, 14.24% *v.s* 9.45%, 13.5% *v.s* 8.8% and 16.34% *v.s* 10.62%. Consequently, they suggest that soybean oil may be more resistant to thermal oxidation than sunflower oil. Furthermore, our oil samples did not contain lauric acid. These results are in line with Gregorio [21] and Gopala et al. [22], who revealed that only coconut oil is a major source of lauric acid C12:0. However, these values were lower than those found by Asgary et al. [23] in Iranian edible oils (18.9%).

"Fleurial" oil exhibited the lowest content in monounsaturated fatty acids (M.U.F.A.) acids (24.117%), whereas "LaBelle" oil displayed the highest content in MUFAs (29.763%). "LaBelle" oil had the highest oleic acid (28.055%) content, while "Afia" (blended oil) registered the lowest content in oleic acid (20.977%). Our results regarding total content in MUFAs are not in accordance with those obtained by Kaur et al. [20]. Our pure "fleurial" sunflower oil (24.117%) was not as rich in MUFA as sunflower oil analyzed by Kaur et al. [20], who reported (37.40%). However, our soybean oils were richer in MUFAs in comparison with Indian soybean oils [20].

"LaBelle" oil showed the lowest content in polyunsaturated fatty acids (P.U.F.A) at (54.565%), whereas "fleurial" oil displayed the largest PUFA content (64.769%). Linoleic acid (C18:2, ω6) was the major PUFA present in all analyzed oils : "fleurial" oil (64.442%), "LaBelle" (49.366%). Unlike linoleic acid, an average amount of α-linolenic acid was found in "Afia" oil (6.376%) *vs.* (0.327%) in "fleurial" oil. ''Fleurial" oil contains a low proportion of SFAs and a considerable quantity of MUFAs (84.378%) as well as a maximum level PUFAs (88.886%), which gave it a higher unsaturated FAs (HUFAs) content than the other analyzed pure soybean oils.

Our results regarding the MUFA and PUFA content of our edible oils are not in accordance with those of Kaur et al. [20]. Pure "fleurial" sunflower oil was richer in PUFAs than Indian sunflower oil, which contained

Table I - Fatty acids composition of studied refined vegetable oils

Fattyacids (% w/w)	Marks of oils				
	'LaBelle'	'Oleor'	'Elio'	'Afia'	'Fleurial'
C14:0	0.080±0,000[a]	0.080±0,000[b]	0.070±0,000[d]	0.070±0,000[c]	0.070±0,000[e]
C16:0	10.611±0,016[d]	10.637±0,003[c]	8.050±0,001[e]	10.749±0,012[b]	6.472±0,008[f]
C18:0	3.984±0,0016[c]	3.919±0,000[d]	3.517±0,000[f]	4.493±0,003[b]	3.857±0,004[e]
C20:0	0.423±0,003[b]	0.432±0,002[a]	0.288±0,002[e]	0.358±0,000[d]	0.273±0,001[f]
C24	0.201±0,001[c]	0.202±0,003[c]	0.213±0,002[b]	0.124±0,002[e]	0.245±0,005[a]
C18:1, ω9	28.055±0,030[a]	27.184±0,012[b]	23.537±0,002[c]	20.977±0,003[e]	23.266±0,004[d]
C18:1, ω7	1.458±0,002[a]	1.441±0,006[b]	1.059±0,003[e]	1.335±0,001[c]	0.699±0,002[f]
C18:2, ω6	49.366±0,020[f]	50.240±0,027[e]	60.475±0,002[b]	55.063±0,007[c]	64.442±0,043[a]
C18:3, ω3	5.199±0,003[d]	5.297±0,001[c]	2.320±0,000[e]	6.376±0,000[b]	0.327±0,001[f]
C20:1, ω9	0.250±0,001[b]	0.266±0,006[a]	0.173±0,003[e]	0.183±0,001[d]	0.152±0,001[f]
∑ SFA	15.331±0,023[c]	15.281±0,013[d]	12.188±0,000[e]	15.797±0,010[b]	10.959±0,016[f]
∑ MUFA	29.763±0,033[a]	28.891±0,012[b]	24.769±0,002[c]	22.495±0,003[e]	24.117±0,005[d]
∑ PUFA	54.565±0,023[f]	55.537±0,028[e]	62.795±0,003[b]	61.439±0,007[c]	64.769±0,042[a]
P / S	3.560±0,006[f]	3.633±0,004[e]	5.150±0,001[b]	3.890±0,004[c]	5.909±0,013[a]
ω6 / ω3	9.494±0,004[c]	9.474±0,005[c]	26.060±0,000[b]	8.630±0,000[d]	195.271±0,121[a]
A.I.	0.1296	0.1297	0.0951	0.1314	0.0759
T.I.	0.2660	0.2639	0.2347	0.2644	0.2297

SFA: saturated fatty acids; MUFA: monounsaturated fatty acids; PUFA: polyunsaturated fatty acids; P / S: PUFA / SFA; AI: atherogenic index; TI: thrombogenic index
Means ± SD (standard deviation) within a row with the same lower case letters are not significantly different at $p < 0.05$

only 51.23% of linoleic acid. However, our soybean oils contained less PUFAs than Indian soybean oils (59.27%) and (53.58%) of linoleic acid [20]. Finally, all soybean oils had similar α-linolenic content.

The relationship between SFAs and PUFAs content is expressed as the P/S index. Table I reveals that all analyzed vegetable oils displayed higher total PUFAs content than SFAs content. These oils had a P/S ratio that varied from 3.560 for "LaBelle" oil to 5.909 for "fleurial" oil. Moreover, all refined vegetable oils marketed in Algeria are highly unsaturated. "Fleurial" oil had high PUFAs (C18:2, ω6 + C18:3, ω3) content (64.769%) with linoleic acid being the major FA (64.442%) and α-linolenic acid the minor one. It is clear that oils with high content are more prone to oxidation. Oil source, plant genotype, environmental conditions - particularly temperature- and water supply considerably influenceFAs composition.

3.2.2 Atherogenic and thrombogenic index

The atherogenicity Index (A. I.) explains the relationship between the sum of the main SFAs and the main classes of unsaturated fatty acids (UFAs). The thrombogenicity Index (T.I.) explains the tendency of blood cells to form clots in the blood vessels due to the adhesion of lipids with blood cells. SFAs are pro-atherogenic : they promote their adhesion to cells in the circulatory system and are pro-thrombogenetic. Conversely, UFAs are anti-atherogenic : they inhibit the clotting of blood cells and they are anti-thrombogenetic : they prevent the appearance of coronary diseases [24, 25].

Pareira et al [9] developed two very useful equations, one for calculating the index of thrombogenicity (T.I.) and another for calculating the index of atherogenicity (A.I.). Indices of atherogenicity and thrombogenicity of the five brands of Algerian edible refined oils are summarized in Table I. Note that the T.I. was 0.2297 for "fleurial" (pure sunflower oil), and 0.2660 for "LaBelle" (pure soybean oil). In contrast, for A. I. the values were 0.0759 for "fleurial", and 0.1296 for "LaBelle". Our results regarding atherogenic and thrombogenic indices of Algerian refined edible oils are in agreement with those reported by Kaur et al. [20], who worked on refined Indian soybean and sunflower oils. However, the values were very low, when compared to those of olive oil ; 0.14 and 0.32 respectively for A.I. and T.I. [8].

A.I. and T.I. values were the lowest in sunflower oil (0.07 and 0.21) and the highest in soybean oil (0.13 and 0.27). However, "elio" oil (blended soybean and sunflower oils at a ratio of 80:20) showed a significant decrease in both indices, thereby revealing a lower risk of developing coronary diseases [24, 25]. Indeed, Mennoti et al. [26] pointed out that there is a positive and highly significant correlation between A.I. and T.I. and the mortality rate from coronary heart disease. They also observed a direct link with all-cause mortality at 25 and 50 years of age.

In addition, A.I. and T.I. provide very useful knowledge about the impacts of SFAs, MUFAs and PUFAs in the development of coronary heart diseases [27,9]. Some FAs have a greater role in atherosclerosis, while others play a key role in thrombogenesis. Palmitic acid has a cholesterol-raising effect and is therefore considered as atherogenic SFAs, while stearic acid influences clot formation in blood vessels and is known as thromogenic SFAs [27]. C14:0, C16:0 and C18:0 contents in our oils were similar to those obtained by Kaur et al. [20]. The myristic acid is considered to be four times more atherogenic than palmitic acid.

MUFAs and PUFAs in edible oils considerably reduce the atherogenic and thrombogenic effects. PUFAs are more prominent in preventing atherosclerosis and thrombogenesis than MUFAs [8]. There were significant differences between our sunflower oils and Indian sunflower oils in terms of MUFAs and PUFAs fractions. Pure Algerian "fleurial" sunflower oil showed the lowest MUFAs content compared to Indian sunflower edible oils, and ''fleurial" oil had a higher PUFAs content than its Indian counterpart. MUFAs and PUFAs are represented respectively by oleic and linoleic acids. However, Ng et al. [28] pointed out that edible oils heated repeatedly many times have potentially adverse health effects and are responsible for the occurrence of atherosclerosis. In contrast, phenolic compounds, which occur naturally in small quantities in fresh edible oils, exert an antioxidant effect ; heating oils over and over again destroys and eliminates the antioxidant properties of these substances. Consumption of deep-fried foodstuffs alters the physiology and vasorelaxation of the endothelium [29].

3.2.3 Heavy pro-oxidant metals content

As is shown in Table II, there were significant differences between Algerian edible oils in terms of heavy metals content. Our results were in agreement with the Codex Alimentarius [16], 1.5 mg/kg oil for iron and 0.1 mg/kg oil for copper. The iron levels in the analyzed oils were 0.047 in "elio", 0.049 in "fleurial", 0.066 in "oleor", 0.070 in "Afia" and 0.082 mg/kg in "LaBelle" oils. The copper concentrations were 0.0010 mg/kg in "oleor", 0.007 in "LaBelle", 0.023 in "Afia", 0.031 in "elio" and 0.058 in "fleurial" oils. Soybean oil displayed the lowest copper levels, while sunflower oils exhibited the highest concentrations of copper.

Copper is an essential nutrient for the human body. Together with iron, it enables the formation of red blood cells. It helps maintain healthy bones, blood vessels, nerves, and the immune system, and it contributes to iron absorption. Copper and iron are required in our diet. Biologically, these two heavy minerals exhibit a wide range of functions. Iron is involved in numerous processes necessary for normal bodily functions: oxygen transport, oxidative phoshorylation, metabolism of neurotransmitters, and DNA synthesis all require iron. Iron deficiency is prevalent in most of the developing world; the main cause is a diet low in bioavailable iron [30]. Copper is responsible for structural and catalytic properties of multiple enzymes necessary for normal bodily functions [31]. This metal is required for infant growth, host defence mechanisms, bone strength, red and white cell maturation, iron transport and brain development [32]. Anaemia, neutropenia, and bone abnormalities (osteoporosis, fractures, etc.) are the main symptoms of copper deficiency. Other effects described include hypopigmentation of the hair and skin, hypotonia, impaired growth, increased incidence of infections and altered immunity [32, 33]. However, a high content of PUFAs, copper and iron in edible oils triggers oxidation [34, 35].

The presence of heavy metals in our refined vegetable oils might be due to crude oil imported by our country and/or introduced during the refining carried out locally. The values were higher than those reported by Pehlivan et al., [36] who worked on Turkish sunflower oils (mean 0.00918 mg/kg oil). Finally, Acar [37] found 1.52 and 1.51 mg/kg of copper and iron in Turkish soybean and sunflower oils respectively.

Algerian oils had lower concentrations of iron compared to their Nigerian, Iranian, Spanish and Chinese counterparts : Nnorom and Ewuzie [38] in Nigeria recorded 50-425 mg/Kg, while Farzin and Moassesi [39] in Iran measured 17.66 µg/g and Garrido et al. [40] in Spain found 0.27-1.31 µ/g sunflower oil and 0.38-0.76 µg/g soybean oil. Finally, Zhu et al. [41] in China obtained 27.3-29.8 and 21.6-23.1 µ/g of sunflower and soybean oils respectively.

The values of iron content in Algerian edible oils were higher than those reported by Pehlivan et al. [36] for Turkish sunflower oils (0.01652 mg/kg oil), while Acar [37], who worked on the same Turkish brands, obtained 0.07 mg/kg sunflower oil and 0.09 mg/kg soybean.

Furthermore, Algerian edible oils contain less copper than their Nigerian, Iranian, Spanish and Chinese counterparts : Nnorom and Ewuzie [38] in obtained 0.22-4.99 mg/kg, while, Farzin et al. [39] in Iran found 0.25 and 0.18 µg/g for sunflower and soybean oils respectively, whereas Garrido et al. [40] in Spain recorded 0.03-0.19 µg/g and 0.04-0.12 µg/g for sunflower and soybean oils respectively, while Zhu et al [41] in China reported 0.052-0.068 and 0.048-0.056 µg/g of sunflower and soybean oils respectively. Finally, Dugo et al. [42] in Italy found 0.1068 and 0.67458 mg/kg for sunflower and soybean oils respectively.

3.3. CHEMICAL PARAMETERS LEVELS

The results regarding the chemical parameters of the five most consumed brands of Algerian vegetable edible oils are shown in Table II. The FFAs content was lowest in the blended oil ”elio” (0.044) and the highest in the percentage of FFAs was the pure sunflower oil “LaBelle” (0.054). The minimum PV was observed in pure “fleurial” sunflower oil (1.61 meq/kg), whereas the maximum PV was found in pure “LaBelle” soybean oil (2.00 meq/kg). There were no significant differences ($p \geq 0.05$) between the five brands of Algerian vegetable edible oils in terms of IV.

On one hand, AV measures FFAs in oil. Normally, FAs occur in form of triglycerides (TGs), however, during processing the FAs may get hydrolyzed into FFAs. The higher the AV found, the higher the level of FFAs, which translates as decreased oil quality. Acceptable levels for all oil samples must be below 0.6 mg KOH/g (measured in potassium hydroxide per gram) and 0.3% (as oleic acid) [16]. Therefore, according to edible oil specifications, all five brands of Algerian edible oils fell within the acceptance range.

On the other hand, IV measures of the degree of unsaturation in oil TG. The greater the IV, the more unsaturation and the higher the susceptibility to oxidation. The Codex Alimentarius [16] established IV at 118-141 for sunflower oil and 124-139 for soybean oil. Therefore, according to these specifications, all the five brands of oils were within the set ranges. IV is an indicator oil unsaturation, which is the most important analytical characteristic of oil [43].

PV reflects the freshness of the oil. As with FFAs, all five brands of Algerian edible oils were in compliance with the specifications in terms of PV. The latter provides useful knowledge regarding the level of lipid oxidation, and must not exceed the upper limit (10

Table II - Physical and chemical properties of studied refined oils

	Marks of oils				
	‘LaBelle’	‘Oleor’	‘Elio’	‘Afia’	‘Fleurial’
Free fatty acids (%)	0.054 ± 0.003^{a}	0.051 ± 0.003^{a}	0.044 ± 0.003^{b}	0.050 ± 0.001^{a}	0.051 ± 0.005^{a}
Peroxide value (meq kg^{-1})	2.00 ± 0.20^{bc}	1.97 ± 0.12^{bc}	1.70 ± 0.20^{c}	1.85 ± 0.10^{b}	1.61 ± 0.21^{a}
Iodine value(g $100g^{-1}$)	127.05 ± 2.07^{a}	$124.07 \pm 4,10^{a}$	125.14 ± 4.82^{a}	126.25 ± 4.03^{a}	129.91 ± 2.78^{a}
Copper content (mg/kg)	0.007±0.001	0.0010±0.000	0.031±0.001	0.023±0.001	0.058±0.001
Iron content (mg/kg)	0.082±0.001	0.066±0.001	0.047±0.001	0.070±0.001	0.049±0.001

meqO2/kg) established by [16]. So, the greater the PV, the higher the rate of oxidation of the oil [44].
Malondialdehyde is an organic compound formed during lipid peroxidation. It is a biomarker of oxidative stress: a high level of this by-product indicates greater fat degradation and causes oxidative damage in cells and tissues. The urinary excretion of malondialdehyde and lipophile carbonyl coupounds has been observed in animals and humans [45].
The peroxydes impair the physiological function of the blood vessels endothelium and consequently negatively affect the amount of nitrogen monoxide (NO) ; this molecule plays a key role in maintaining cardiovascular homeostasy, thanks to its impact on blood vessels dilatation [46, 29]. In addition, studies have shown that consumption of oxidized fats causes carcinogenesis [47]. It should be noted that fat peroxydes, once absorbed, cause the formation of other fat peroxydes by chain reaction. Furthermore, induced oxydative stress stimulates the production of pro-inflammatory cytokines [48]. However, fresh food intake provides protection of the blood vessels structure. In fact, many researchers demonstrated that polyphenols ingested simultaneously with oxydable foods prevent fat oxidation in the stomach. These molecules break down the oxidation chains of membrane phospholipids [49].
Vitamins and minerals are contained mainly in plant foods like fruits, vegetables and cereals and are endowed with antioxidant properties, considerably reducing the peroxydation and inflammation that occur during digestion [49, 50, 51]. The amount of oxydated lipoproteins diffused in the blood circulation plummets substantially, providing the best state of health to consumers.
Note that fried foods should be accompanied with green vegetables since the latter are rich in antioxidant molecules which, on one hand, reduce the number of free radicals in the digestive tract and, on the other hand, at low levels in the body, reactive oxygen molecules are useful and play an essential role in the mechanism of cell signalling, thereby contributing to maintaining cell homeostasis [52].
It is almost impossible to prevent oxidation in edible oils. However, it is possible to influence the kinetics of the reaction, slow down the speed at which this reaction occurs and delay this phenomenon considerably. In the past, our grandparents filtered soybean oil after frying and incorporated slices of green apples into the cooled oil before storing it in sealed containers in dark cupboards. When frying, they also introduced a raw peeled carrot into the oil in order to regulate its temperature and keep it clean. Current research has shown that apple polyphenols neutralize and reduce the concentration of polar compounds that occur during frying. Finally, current research emphasises the effect of high temperatures on advanced oil degradation.
Antioxidants found in fresh fruits and vegetables increase the physiological capacity of the human organism to release free radicals outside the body, and thereby sustain the function of endothelium. Introducing a slice of raw apple in used cooking oil enables its reuse, thanks to the phenolic compounds that sequestrate the free radicals that are formed during frying. A large amount of lipoperoxydes is harmful and a risk factor in cardiovascular diseases [49].
In Algeria, edible oil is packaged in non-inert plastic bottles, yet plastic packaging may contain chemicals that can dissolve in edible oil and contaminate it. However, the frequency of fried food consumption exceeds the recommendation of only twice a week so the new trend means that the risk of oil contamination by chemicals from plastic packaging is eliminated since the oil is consumed before chemicals can build up in the oil.

3.4. THERMAL STABILITY EVALUATION

The results from the Rancimat test and the calculation of activation energy of the five brands of Algerian vegetable edible oils are summerized in Table III.

3.4.1 Rancimat test

The Rancimat test is an easy method for determining the induction time of lipid oxidation. Several parameters impact the Rancimat test such as sample quantity, air flow rate and temperature [53]. Induction time is the time that the hydroperoxides produced by oil oxidation take to decompose [54]. The Rancimat induction time at 98°C for the five brands of edible oils varied from 8.27 h to 14.05 h; pure "fleurial" sunflower oil displayed minimum oxidative stability, while pure soybean oil "oleor" had maximum thermal stability. "LaBelle" required a relatively short induction time (10.15 h), while the blended oils ("elio": 80% soybean + 20% sunflower oil and "afia": 95% soybean + 5% corn oil) were characterized by a longer induction time (14.05 h and 13.46 h) respectively.

Table III - Stability index of studied refined vegetable oils

	Marks of oils				
	'LaBelle'	'Oleor'	'elio'	'Afia'	'Fleurial'
Induction time (h)	10.15	14.05	13.46	13.25	8.27
Activation energy (kJ/mol)	550.09	581.52	570.50	570.03	438.16

Furthermore, pure soybean oil ‘’oleor”, characterized by a high induction time, was more stable than pure “fleurial” sunflower oil with a low induction time. “Fleurial” oil recorded a shorter induction period, higher IV and higher copper content than ‘’oleor”, confirming that ‘’fleurial” was highly sensitive to oxidation. There were significant differences between ‘’LaBelle” and ‘’oleor” oils in terms of induction times. These differences were due to the variations in copper content. “LaBelle” oil had a higher copper concentration than “oleor” oil. Therefore, ‘’oleor” oil was thermally more stable and more resistant to oxidation. The sensitivity of “fleurial” oil to oxidation is due to its high PUFA content, large copper concentration and short induction time.

3.4.2 Activation energy

Thermogravimetric analysis evaluates the oxidative and thermal stability of edible oils by detecting the mass change of the related oil sample through thermal degradation [15]. Activation energy represents the minimum energy required to start the first step of thermal decomposition and the beginning of FAs decomposition; the higher the activation energy, the harder it is for a reaction to occur and vice versa. This is expressed in kilojoules per mole (kJ/mol). Our results with regard to the activation energy of the five oils are summerized in Table III. The results regarding activation energy of the five brands of Algerian oils were in line with the results of the Rancimat test. Note that activation energy values ranged from 438.16 kJ/mol for pure “fleurial” sunflower oil to 581.52 kJ/mol for pure “oleor” soybean oil.

The content of FAs and the presence and concentration of heavy metals (copper and iron) considerably influence oil activation energy. Our data established the order in terms of the thermal stabilities of the five oils as follows: oleor > elio > Afia > LaBelle > fleurial. On one hand, ‘’fleurial” oil exhibited the lowest activation energy among the five oils due to having the highest content in PUFAs and pro-oxidants (copper). On the other hand, refined pure ‘’oleor” soybean oil displayed the highest activation energy and therefore the slowest oxidation due to its lower copper content. In fact, copper accelerates hydrogen peroxide decomposition to a speed 50 times faster than ferrous ion (Fe^{2+}), and ferrous ion speeds up this thermal degradation reaction to 100 times faster than ferric ion (Fe^{3+}) [55]. In addition, copper increases the rate of oil oxidation due to the reduction of the activation energy of the initiation step in auto-oxidation down to 63~104 kJ/mol [56]. Indeed, the rate of the formation of the primary oxidation products increases as the amount of PUFAs and pro-oxidant metals in oils rises. Finally, these transition metals catalyze the decomposition of hydroperoxides and lead to more rapid formation of undesirable substances [36].

4. CONCLUSION

In this study, physical, chemical and oxidative stability analyses of the five top most consumed Algerian vegetable edible oils were conducted in order to assess their thermal stability. Our results showed that these five edible oils are within the acceptance range of the physical and chemical parameters described in the Codex Alimentarius. The five brands of oils behaved differently in terms of thermal degradation and oxidative stability characteristics. Based on the Rancimat test and thermogravimetric analysis, our data showed that pure ‘’fleurial” sunflower oil is the most prone to thermal degradation and oxidation because of its high content in PUFAs and heavy metals (copper and iron). Conversely, pure ‘’oleor” soybean oil and ‘’LaBelle” are the most thermally stable due to their low concentrations in copper and iron. The blended oils “elio” and “Afia” displayed intermediate values of thermal and oxidative stabilities because they are produced by mixing sunflower and soybean oils at different ratios. Finally, in order to improve the thermal and oxidative stability of pure ‘’fleurial” sunflower oil, the full-refining of this oil is recommended in order to get rid of all FFAs, and any heavy metals (copper and iron) while adding stabilizers like olive oil polyphenols.

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