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Seed Oil from Ten Algerian Peanut Landraces for Edible Use and Biodiesel Production

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Abstract: As a result of a recent *ad hoc* prospection of the Algerian territory, a collection of peanut (groundnut; *Arachis hypogaea* L.) landraces was established, covering a remarkable array of diversity in terms of morphological and physiological features, as well as of adaptation to local bioclimatic conditions. In the present work, the oils extracted from the seeds of these landraces were evaluated in terms of edible properties and suitability for biodiesel production. As for edible use, a low free acidity (ranging from 0.62 to 1.21%) and a high oleic acid content (44.61-50.94%) were common features, although a poor stability to oxidation [high peroxide values, high spectrophotometric indices, and low % of inhibition in the 2,2-diphenyl-1-picrylhydrazyl radical (DPPH)· test] was observed in a few cases. As for biodiesel production, low values of acidity [1.23-2.40 mg KOH (g oil)$^{-1}$], low iodine values [90.70-101.54 g I_2 (g oil)$^{-1}$], high cetane numbers (56.95–58.88) and high calorific values (higher heating value 37.34–39.27 MJ kg^{-1}) were measured. Edible properties and suitability for biodiesel production were discussed with respect to the German standard DIN 51605 for rapeseed oil and to the EN 14214 standard, respectively. One way ANOVA and Hierarchical Cluster Analysis showed significant differences among the oils from the Algerian peanut landraces.

Key words: *Arachis hypogaea* L., biodiesel, DIN 51605, EN 14214, FAME, groundnut, landrace, peanut, vegetable edible oil.

1 INTRODUCTION

Peanut or groundnut (*Arachis hypogaea* L.) is an annual herbaceous plant belonging to the Fabaceae family. The world peanut production in the 2011-2012 was 35,367,000 metric tons, to which the most important contributing Countries were China (45%), India (16%), USA (5%), Nigeria (4%), Argentina (3%), and Indonesia (3%)[1)]. Today, peanut is consumed as an edible seed and is used for its edible oil, from which butter is also prepared.

A. hypogaea is well adapted to grow under high temperature, drought and low soil fertility, owing to their nitrogen-fixing ability and their potential to form effective symbiotic association. Therefore, peanut can play an important role in sustainable agricultural development, particularly in the Maghreb and the sub-Saharan regions, where drought and salinity frequently limit crop production. Water availability is fundamental in crop production and was proved to affect the seed oil content and composition of safflower[2)], sunflower[3)], and *Jatropha curcas* L.[4)].

Vegetable oil can also be a source for biodiesel production and many studies have been conducted in such context. The raw material (vegetable oils) useful for an efficient biodiesel conventional production process must be of high quality, in terms of low content of free fatty acids, water and unsaturated triglycerides[5)]. In the last decades, energy demand on a global scale is steadily increasing, whereas traditional energetic resources, such as fossil fuels, are mainly concentrated in specific geographic areas. For this reason, alternative sources of energy, such as biofuels, are becoming increasingly important, especially for those Countries devoid of geological reservoirs, in order to reduce their energy dependence.

The present work lies in the vein of ongoing projects launched in recent years aiming at defining a dynamic conservation strategy and a reasoned exploitation of the genetic diversity embedded in food legume landraces collected across the Algerian territory. The main aim was to study the composition of oils obtained from the seeds of

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ten Algerian peanut landraces in relation to their edible use and as a source for biodiesel production. An additional aim was to evaluate the influence of the landraces geographical origin on the oil composition and properties.

2 EXPERIMENTAL

2.1 Plant material

Seeds from a total of 10 Algerian landraces of peanut (*Arachis hypogaea* L.) were used in the present work as the starting plant material. These landraces, all showing a sustained degree of tolerance to water stress, were selected in different geographical areas of Algeria (Table 1). The seeds were from the collections obtained from an *ad hoc* prospection of the Algerian territory brought about within the framework of the project "Amélioration des légumineuses alimentaires pour la tolérance au stress hydrique", run by the Ecole Nationale Supérieure Agronomique of Algiers (ENSA) in 2004 and 2005, and then maintained by multiplication. Plants were cultivated on a substrate mixture composed by sandy soil (50% w/v), garden soil (20% w/v) and river sand (30% w/v). During their growth, plants did not receive either pesticides or fertilizers, to not interfere with symbiotic nodulation. Each plant was irrigated with 200 mL of a 50% mixture (vol/vol) of deionized + tap water each four days. Each landrace was harvested in two consecutive years (2011 and 2012). Seeds of each landrace were randomly and manually collected from 20 plants and then air dried in the dark and stored in paper bags at 4 ℃. Further details concerning plants' growth conditions are reported elsewhere[6].

2.2 Oil content and oil recovery

Three aliquots of seeds (100 g each) for each landrace were randomly selected. Each aliquot was separately grinded with a home grinder. The powder obtained from each seed aliquot was placed in a glass beaker, in which three volumes of petroleum ether were then added. The mixture was submitted to three consecutive room temperature stirring extractions (RTSE, each for 30 min) of the oil. After each RTSE, the liquid fraction (petroleum ether and oil) was separated by filtration from the solids and oil content was quantified after solvent evaporation under vacuum in a Rotary evaporator apparatus at room temperature. At the end, the three oil fractions were weighted and summed.

The oil content was expressed as the oil quantity (g) for 100 g of grinded dried seeds. The oil recovery was calculated as the quantity of oil (g) recovered in each extraction on the total oil recovered. Recoveries from RTSE and from Soxhlet extraction (SE) were compared, being the latter conducted on the peanut powder using a previously described method[7].

After the RTSE, the oil was immediately filtered through a paper filter and stored into a 50 mL brown glass bottle. The physicochemical analyses were conducted on the oil from RTSE within 5 days from extraction.

2.3 Nitrogen and crude proteins

Nitrogen and crude protein (CP) were determined as follows: 1) digestion of defatted seeds in boiling H_2SO_4 at 370 ℃; 2) neutralization of the mineralized solution with NaOH; 3) distillation of the produced ammonia gas into a trapping solution containing H_3BO_3; 4) titration with a 0.1 N HCl solu-

Table 1 Geographic origin and coordinates of the Algerian peanut (*Arachis hypogaea*, L.) landraces used in the present study.

Geographic zone	Bioclimatic floor	Region of the collection	Landraces	Geographic origin	Longitude	Latitude	Altitude (asl)
Coastal	Subhumid	El Kala	Oum Tboul (OT)	North Eastern Lake of Tonga	08° 31E	36° 53N	2.2
Coastal	Subhumid	El Kala	El Frin (FR)	North-Eastern Lake of Oubaira	08° 42E	36° 83N	28
Coastal	Subhumid	El Kala	Boumalek (BMK)	Mellah West	08° 33E	36° 87N	3
Coastal	Subhumid	El Kala	Tonga (TO)	South Eastern Lake of Tonga	8° 32' E	36° 49N	2.2
Coastal	Subhumid	El Kala	Berrihane (BER)	West Eastern Lake of Oiseaux, Berrihane	08° 07E	36° 42N	10
North Sahara	Saharian	Ghardaia	Metlili (MET), SebSeb (SEB)	Metlili, SebSeb	02° 87E 03° 55E	30° 57N 32° 95N	380 503
North Sahara	Saharian	Oued Souf	Oued souf (OS)	Guémar, Oued Souf	06° 52E	33° 22N	77
Central Sahara	Saharian	Adrar	Adrar (AD)	Adrar, Tsabit	00° 25W	28° 36N	256
Central Sahara	Saharian	Timimoun (TIM)	Timimoun (TIM)	Adrar, Timimoun	00° 27E	29° 30N	335

tion and determination of the amount of N by calculation; 5) calculation of CP by multiplying the N content by 6.25 as the conversion factor (AOAC method No 988.05)[8].

2.4 Refractive Index (RI)

RI was determined by an Abbe refractometer according to AOAC No 921.08 method[9]. Each measurement was conducted at 20 ℃.

2.5 Free Acidity (FA)

An exactly weighted aliquot of 2 g of each oil was dissolved in a diethyl ether/ethanol neutralized mixture (1:1) and titration was conducted with a 0.1 N NaOH solution, after adding 3-4 drops of phenolphthalein [1% (v/v) solution in ethanol][10].

2.6 Acid Value (AV)

Each oil was titrated in alcoholic medium and analysis was carried out following the AOAC standard 969.17[11].

2.7 Peroxide Value (PV)

Peroxide value was determined by the method proposed by the Consolidated text for olive oil analysis, Annex III[10].

2.8 Iodine Value (IV)

One g of each oil, 20 mL of a cyclohexane/acetic acid solution (1:1, v/v), and 20 mL of Wijs reagent were put into a glass flask. Then, the stopper was inserted and the flask was placed in the dark for 1 hour. Then, 20 mL of a 100 g L^{-1} KI aqueous solution and 150 mL of deionized water were added. A blank was prepared under identical conditions but omitting the oil. Both sample and blank solutions were titrated with a 0.1 N thiosulfate solution, to which five drops of a 3% aqueous starch solution had been previously added (Consolidated text for olive oil analysis, Annex XVI)[10].

2.9 Saponification Value (SV)

Each oil was saponified by refluxing with a known excess of alcoholic KOH solution. The alkali required for saponification was determined by titrating the excess KOH with standard HCl solution, adding phenolphtalein as the indicator (AOAC, 920.160)[12]. The results were expressed as mg KOH (g of oil)$^{-1}$.

2.10 Spectrophotometric indices

These were measured in the ultraviolet range following the method described in the Consolidated text for olive oil analysis, Annex IX[10].

2.11 2,2-diphenyl-1-picrylhydrazyl radical test (DPPH$^{\cdot}$ test)

The test of antioxidant capacity by using the DPPH$^{\cdot}$ test was conducted spectrophotometrically by following the method of Kalantzakis *et al.*[13] modified as follows. The oil was diluted with ethyl acetate (1:10, v/v) and a 10^{-4} M solution of DPPH in ethyl acetate was prepared. Ethyl acetate was used as a blank solution. A 2 mL aliquot of DPPH solution, or of ethyl acetate alone as the blank, was put into a rectangular quartz cuvette, with cover and having an optical length of 1 cm, after which a first A_{515} value was recorded (t0; Agilent mod 8453 spectrophotometer, Santa Clara, CA, USA). Then, 0.5 mL of oil solution was added to both the sample cuvette, containing the DPPH solution, and to the reference cuvette, containing ethyl acetate alone. Continuous stirring was maintained for 30 min in both cuvettes, after which A_{515} was measured again (t30). The result was expressed as % of DPPH reduction = $100 \times [1 - (t30 - t0)]$.

2.12 Higher Heating Value (HHV)

A bomb calorimeter (Mahler calorimeter) was used. Because the direct combustion of the oils was difficult, each of them was mixed with cellulose powder before combustion, by using 25 kg cm^{-2} as the O_2 pressure. The analysis was firstly conducted on 1 g of cellulose powder pressed as a pellet (3 mm diameter and 2 g weight), and its HHV was quantified. Then, another 1 g pellet of cellulose powder was pressed, over which 1 g of oil was slowly and accurately dropped before HHV quantification. Finally, the HHV of the oil was calculated as the difference among oil plus cellulose powder and the cellulose powder alone[14].

2.13 Cetane number (CN)

The CN is dependent on the FAME composition[15, 16]. Consequently, the CN of the oils can be obtained as the predicted cetane number. Viola *et al.*[17] calculated the experimental CN for FAMEs whose maximal chain length was 22 carbon atoms. Because the oil from peanut seeds contains C24:0 as the FAME with the maximal chain length, the experimental C24:0 CN was calculated here also on the basis of the equation proposed by Freedman and Bagby[15] and the CN of each peanut oil was predicted as the sum of the mass fraction of all the detected FAME.

2.14 Fatty acids methyl esters (FAME)

Transesterification of each oil with a cold methanolic solution of 2 N KOH was conducted following the Consolidated text for olive oil analysis, Annex X, method A[10]. One mL of heptane and 0.1 g of oil were put into a glass vial and shaken. Then, 0.2 mL of KOH were added and the vial shaken again for 30 seconds. After phase separation, the upper heptane phase was injected onto a gas-chromatograph under the conditions previously described[18].

2.15 Calcium and magnesium

Calcium and magnesium in the oils were quantified by atomic emission spectroscopy as described by Giuffrè *et al.*[14].

2.16 Chemicals

Fatty acid methyl ester (FAME) standards were sourced from Sigma-Aldrich (St. Louis, MO, USA); hexane was from WWR International (Milan, Italy); Wijs' reagent and methanol were from Panreac (Barcelona, Spain); all other reagents were from Carlo Erba (Milan, Italy).

2.17 Statistical analyses

The determination of mean values and standard deviation was conducted by the Microsoft Excel software (2007 version). Each value is the mean of three replicates/year per two harvest years ± SD.

One way ANOVA was applied to the ten peanut landraces using SPSS version 15.0 for Windows (SPSS Inc., Chicago IL, U.S.A.); the Tukey test was applied to determine any significant difference among all landraces at $p < 0.05$. The same statistical software also allowed to run a Hierarchical Cluster Analysis (HCA) (furthest neighbour and Euclidean distance) to identify different groups among the studied set of peanut landraces.

3 RESULTS AND DISCUSSION

3.1 Oil content and oil recovery

The highest oil quantities were extracted from the seeds of BER (44.03%), AD (42.35%) and MET (42.12%) peanut landraces (Table 2). In all the remaining landraces the oil quantity was lower than 40%, being the lowest in BMK, which yielded 30.96%. Özcan *et al.*[19] found 31.52% and 44.09% oil contents in two Turkish peanut cultivars after a Soxhlet/diethyl ether extraction. By using Soxhlet/petroleum ether extraction, Grosso *et al.*[20] found oil contents ranging from 45.7% to 51.8% in wild peanuts species collected in Argentina. Wang *et al.*[21] by using an Ankom apparatus and petroleum ether as a solvent, found the range 31.52% - 44.09% in fifty peanut accessions from the US germplasm collection. Russo and Webber[22] extracted oil quantities ranging from 5.6% to 23.8% in wet peanut seeds from plants grown under varying cropping conditions.

3.2 Nitrogen and crude protein

The N content of the seeds was in the range 3.54% - 8.14% (Table 3). The fifty percent of the landraces showed a N content in the seeds ranging from 4% to 6%.

The two lowest CP content in defatted peanuts were found in MET (22.15%) and in SEB (27.07%), both from the Ghardaia Region (Tables 1 and 3). The highest CP content was found in TO (50.89%), being twice those of MET and SEB, and more than twice respect to those employed by Scerra *et al.*[23] for preparing the meals given to lambs in a study aimed at evaluating leguminous protein sources, such as soybean (CP 19.50%), broad bean (CP 19.92%) and pea (CP 19.56%). Consistently with the present results, Asibuo *et al.*[24] found a CP content ranging from 18.92% to 30.53% in twenty groundnut varieties from Ghana.

3.3 Refractive index

The RI is a dimensionless parameter indicating the speed of light passing from oil to air, whose value is characteristic for the oil of each species. Here, RI values ranged from 1.4612 in AD to 1.4688 in MET (Table 3), supporting the findings of Davis *et al.*[25] in normal oleic peanut oil.

The Codex Standard for named vegetable oils indicates 1.460 and 1.465 as the minimum and the maximum RI values, respectively, for peanut oil measured at 40 ℃[26]. In

Table 2 The effects of three extractions, the total oil contents and the oil recoveries from the ten peanut landraces of Table 1. Means in the same column with different lowercase letters are statistically different. Significance level: n.s. = not significant; *** $p < 0.001$.

	Oil content on dried seeds (%)				Oil recovery (%)		
	1st extraction	2nd extraction	3rd extraction	Total	1st extraction	2nd extraction	3rd extraction
OT	27.31 ± 1.02bc	7.68 ± 0.63a	2.24 ± 0.11a	37.23 ± 0.70cd	73.33 ± 1.77a	20.65 ± 1.48a	6.02 ± 0.31a
FR	24.84 ± 1.07c	7.59 ± 1.64a	2.37 ± 0.11a	34.81 ± 0.01d	71.37 ± 3.13a	21.81 ± 3.36a	6.82 ± 0.23a
SEB	28.64 ± 0.82bc	6.88 ± 1.00a	1.82 ± 1.94a	37.34 ± 2.02cd	76.90 ± 5.13a	18.38 ± 1.20a	4.72 ± 3.97a
BMK	24.87 ± 4.14c	4.96 ± 3.31a	1.13 ± 0.17a	30.96 ± 2.18e	80.01 ± 9.51a	16.32 ± 8.84a	3.67 ± 0.69a
TO	27.80 ± 4.65bc	8.01 ± 4.42a	1.29 ± 1.72a	37.10 ± 0.42cd	74.88 ± 11.94a	21.64 ± 8.59a	3.49 ± 3.44a
BER	37.48 ± 2.83a	5.72 ± 1.69a	0.83 ± 0.52a	44.03 ± 1.76a	85.05 ± 3.95a	13.04 ± 3.13a	1.91 ± 0.93a
MET	36.42 ± 0.69a	4.44 ± 1.44a	1.26 ± 0.38a	42.12 ± 0.09ab	86.47 ± 1.84a	10.53 ± 2.62a	3.00 ± 0.82a
AD	33.93 ± 3.26ab	7.10 ± 1.27a	1.32 ± 0.53a	42.35 ± 2.70ab	79.99 ± 3.86a	16.86 ± 2.85a	3.15 ± 1.01a
OS	31.41 ± 1.19abc	6.34 ± 1.48a	1.26 ± 0.78a	39.01 ± 0.84bc	80.54 ± 3.95a	16.24 ± 2.55a	3.22 ± 1.40a
TIM	32.91 ± 1.66ab	6.05 ± 0.45a	0.74 ± 0.64a	39.70 ± 1.17bc	82.86 ± 2.30a	15.26 ± 1.13a	1.88 ± 1.17a
Sign.	***	n.s.	n.s.	***	n.s.	n.s.	n.s.

Table 3 Nitrogen, crude seed proteins in seeds and physico-chemical properties of the peanut oils for edible use. Means in the same column with different lowercase letters are statistically different. Significance level: *** $p < 0.001$.

	N (%)	Crude Protein (%)	Refractive Index (20 ℃)	Free acidity (%)	Peroxide Value [meq O_2 (kg of oil)$^{-1}$]	DPPH (% Inhibition)	Saponification Value [mg KOH (g of oil)$^{-1}$]
OT	5.49 ± 0.10f	34.31 ± 0.65f	1.4668 ± 0.00d	0.78 ± 0.06cd	19.26 ± 0.21e	25.56 ± 0.50d	185 ± 1.03c
FR	6.92 ± 0.11c	43.27 ± 0.67c	1.4657 ± 0.00e	0.62 ± 0.05d	25.31 ± 0.28c	22.24 ± 0.39f	192 ± 0.60b
SEB	4.33 ± 0.04h	27.07 ± 0.26h	1.4683 ± 0.00ab	1.21 ± 0.08a	13.49 ± 0.19g	37.10 ± 0.29a	198 ± 0.61a
BMK	6.56 ± 0.08d	41.03 ± 0.48d	1.4678 ± 0.00bc	0.81 ± 0.02c	23.40 ± 0.23d	30.03 ± 0.23b	191 ± 0.56b
TO	8.14 ± 0.04a	50.89 ± 0.25a	1.4658 ± 0.00e	0.79 ± 0.04cd	11.25 ± 0.27h	27.14 ± 0.37c	174 ± 0.49e
BER	5.20 ± 0.01g	32.48 ± 0.07g	1.4672 ± 0.00cd	0.62 ± 0.05d	16.69 ± 0.27f	23.27 ± 0.36e	186 ± 0.53c
MET	3.54 ± 0.04i	22.15 ± 0.22i	1.4688 ± 0.00a	1.01 ± 0.05b	62.88 ± 0.45a	6.58 ± 0.28i	176 ± 0.63d
AD	5.28 ± 0.02fg	32.99 ± 0.11fg	1.4612 ± 0.00f	0.78 ± 0.07cd	31.66 ± 0.20b	11.43 ± 0.34h	171 ± 0.66f
OS	5.99 ± 0.14e	37.44 ± 0.88e	1.4613 ± 0.00f	1.19 ± 0.06a	19.14 ± 0.79e	11.71 ± 0.27h	199 ± 0.53a
TIM	7.34 ± 0.09b	45.86 ± 0.59b	1.4687 ± 0.00ab	1.01 ± 0.08b	23.05 ± 0.23d	12.90 ± 0.24g	199 ± 0.57a
Sign.	***	***	***	***	***	***	***

the present work, the RI for MET oil might have exceeded the above maximum because of the differences in the measuring temperatures (20 ℃ instead of 40 ℃), because an inverse relationship is expected among RI and temperature (the lower the temperature, the denser the oil, with light having to pass more molecules). In addition, RI tends to increase in oxidized vegetable oils, and in fact the MET oil showed the highest PV and K_{274} indices among the tested peanut landraces (see below).

3.4 Free acidity

A low FA most commonly testifies a high chemical and nutritional quality of an edible vegetable oil. Here, the lowest FA (0.62%) was found in the oils from FR and BER peanut landraces, both from the Coastal East of Algeria (Table 3). Instead, the highest FA values were found in oils extracted from the Sahara landraces, such as OS (1.19%), SEB (1.21%), MET (1.01%) and TIM (1.01%). When the European Regulation[27] or the trade standard of the International Olive Council[28] are considered, all the peanut oils studied here were well below the threshold value for a virgin olive oil (2.00%) and five of them even below the threshold for an extra virgin olive oil (0.80%), with BMK (0.81%) standing at the borderline (Table 3).

3.5 Peroxide value

Lipids oxidation is one of the major deterioration process affecting the chemical and sensorial quality of food, as well as its nutritional value. By quantifying the peroxides (hydroperoxides) formed during oxidative processes, the PV reflects the actual state of oil oxidation. In the studied peanut oils, the best PV [i.e. the lowest one, 11.25 meq O_2 (kg of oil)$^{-1}$] was found for Tonga, from Central-Eastern Algeria, whereas the worst PV [(i.e. the highest one, 62.88 meq O_2 (kg of oil)$^{-1}$], more than five times the lowest, was found for Metlili, again from the oases area (Table 3). Zhang *et al.*[29] found a PV of 10.65 in a peanut oil obtained from enzymatic extraction. The Codex Stan 210[26] establishes a PV of up to 10 and up to 15, respectively, for a refined peanut oil and for a cold-pressed, virgin peanut oil. The EU Regulation[27] and the trade standard of the IOC[28] state a PV threshold of 20 for an edible olive oil, so that five out of ten Algerian peanut oils studied here were within the above limit (Table 3). The MET oil was the only one largely above the aforementioned threshold, thus needing rectification.

3.6 DPPH$^{\cdot}$ test

The DPPH$^{\cdot}$ is a very stable radical species whose color changes from red-purple to yellow upon being reduced by an antioxidant molecule. It is therefore possible to quantify the overall antioxidant capacity of a given substance, i.e. its radical scavenging ability, by measuring spectrophotometrically how much it inhibits the absorbance of DPPH$^{\cdot}$, showing a maximum at 515 nm: the stronger the decrease in A_{515}, the higher the antioxidant power of the substance. In the present work, the strongest radical scavenging activity was shown by the SEB oil (37.10% inhibition of A_{515}; Table 3), which also showed the lowest K_{232} value (Table 3) and the second lowest PV and K_{274} ones. On the contrary, the lowest DPPH$^{\cdot}$ scavenging activity was shown by the MET oil (6.58%), which also showed the highest PV, IV, and K_{270} values, as well as the second highest K_{232} value among all the studied oils (Tables 3 and 4).

3.7 Saponification value

This gives an estimation of the molecular mass of a fat or an oil. The lower the SV, the longer the chain and the

larger the molecular weight of fatty acids in the triglycerides, and vice-versa. From the point of view of the use for biodiesel production, a low saponification value is related to a reduction of the corrosion problems of the diesel engine.

The Codex Stan 210[26] indicates for peanut oil the SV range 187-196, expressed as mg of KOH (g of oil)$^{-1}$. In the present work, the oils from TO (SV = 174), MET (SV = 176), and AD (SV = 171) were those showing the most exceeding values with respect to the above range (Table 3), being OT at the borderline (SV = 185). All the remaining Algerian peanut landraces gave oils within the range of the Codex Standard. Anyasor *et al.*[30] reported SV values ranging from 112.20 to 140.25 for oils obtained from Nigerian peanut varieties, thus largely below those exhibited by the Algerian oils.

3.8 Spectrophotometric indices

According to EU regulation[27], K_{232} values have to be 2.50 and 2.60 for an extra virgin and a virgin olive oil, respectively, being the corresponding K_{270} values 0.22 and 0.25, respectively. The K_{232} values in the present peanut oils ranged from 2.77 for SEB to 3.21 for AD, whereas the K_{270} ranged from 0.27 for OT to 0.67 for MET, thus in all cases exceeding the aforementioned limits for an edible olive oil (Table 4). Although olive oil is extracted through physico-mechanical processes from a fruit, whereas the present peanut oils were extracted by RTSE, the spectrophotometric characteristics of the latter are not remarkably different from the ones of a virgin olive oil.

3.9 Fatty acid methyl esters

In the present peanut oils, eleven FAME, ranging from 14 to 24 carbon atoms, were detected by gas-chromatography, six of them being saturated (myristic, palmitic, stearic arachidic, behenic and lignoceric) and five unsaturated (palmitoleic, oleic, linoleic, linolenic and eicosenoic) (Tables 5 and 6).

Oleic acid is one of the most important fatty acid for its capacity to decrease the low density lipoprotein content in the blood, while increasing the high density lipoprotein. In the peanut oils studied here, oleic acid was the most represented fatty acid, ranging from 44.61% for TO to 50.94% for OT (Table 5). Such figure is remarkably higher with respect to the oleic acid previously found in the oils extracted from the seeds of comparable peanut germplasm collections: indeed, Grosso *et al.*[20] found 30.6-46.8% oleic acid in oils from peanut wild accessions belonging to a germplasm bank in Argentina; Anaysor *et al.*[30] reported 41.67-44.20% oleic acid in Soxhlet-extracted oils from six groundnut varieties from eastern, western and northern Nigeria, and Berry[31] found 37.94- 41.90% oleic acid in the oils form 16 peanut varieties cultivated in Malaysia. Instead, higher oleic acid percentages were found in the oils extracted from two Turkish peanut cultivar, namely NC-7 (43.13%) and ÇOM (55.07%)[19]. In a wider context, the oleic acid contents of the Algerian peanut landraces were similar to those found on average in USA cultivars, and higher than in Spanish or Argentinian cultivars, as well as in wild peanut relatives[32].

Linoleic acid, an essential fatty acid (EFA), ranged from 29.92% in OT to 35.17% in BMK (Table 5). Similar results were obtained for peanut oil in Malaysia (34.59- 37.51%)[32], whereas a lower content in linoleic acid (19.58 - 20.89%) was found in Nigerian peanut oil[30]. Wang *et al.*[21] studied 50 accessions of *Arachis hypogea* L. and found a linoleic acid content ranging from 25.18% to 39.74%.

In the oils from the Algerian landraces, the sum of oleic and linoleic acid was around the 80% of the total FAME content, similarly to olive oil produced in this same

Table 4 Spectrophotometric indices in the ten peanut landraces. Means in the same column with different lowercase letters are statistically different. Significance level: n.s. = not significant; *** $p < 0.001$.

	K_{232} nm	K_{266} nm	K_{270} nm	K_{274} nm	ΔK	K_{232}/K_{270}
OT	2.8518 ± 0.03de	0.2657 ± 0.00h	0.2679 ± 0.00h	0.2575 ± 0.00g	0.01 ± 0.00	10.65 ± 0.02
FR	2.9307 ± 0.03cd	0.3490 ± 0.02ef	0.3521 ± 0.02ef	0.3384 ± 0.02de	0.01 ± 0.00	8.35 ± 0.53
SEB	2.7734 ± 0.01e	0.2892 ± 0.01gh	0.2952 ± 0.01gh	0.2821 ± 0.01fg	0.01 ± 0.00	9.40 ± 0.29
BMK	2.9484 ± 0.02bc	0.3328 ± 0.02ef	0.3373 ± 0.02ef	0.3228 ± 0.02de	0.01 ± 0.00	8.75 ± 0.38
TO	2.9230 ± 0.02cd	0.3129 ± 0.00fg	0.3182 ± 0.00fg	0.3054 ± 0.00ef	0.01 ± 0.00	9.19 ± 0.04
BER	3.1416 ± 0.02a	0.5069 ± 0.02c	0.5105 ± 0.02c	0.4913 ± 0.02b	0.01 ± 0.00	6.16 ± 0.32
MET	3.1858 ± 0.03a	0.6696 ± 0.00a	0.6725 ± 0.00a	0.6356 ± 0.00a	0.02 ± 0.00	4.74 ± 0.03
AD	3.2106 ± 0.02a	0.5474 ± 0.00b	0.5501 ± 0.00b	0.5186 ± 0.00b	0.02 ± 0.00	5.84 ± 0.08
OS	2.9993 ± 0.04bc	0.3692 ± 0.01e	0.3727 ± 0.01e	0.3529 ± 0.01d	0.01 ± 0.00	8.05 ± 0.13
TIM	3.0314 ± 0.06b	0.4296 ± 0.01d	0.4324 ± 0.01d	0.4125 ± 0.01c	0.01 ± 0.00	7.01 ± 0.05
Sign.	***	***	***	***	n.s.	***

Table 5 Fatty acid methyl ester composition of the ten peanut landrace oils. Means in the same column with different lowercase letters are statistically different. Significance level: n.s. = not significant; *** $p < 0.001$.

	Myristic (%)	Palmitic (%)	Palmitoleic (%)	Stearic (%)	Oleic (%)	Linoleic (%)	Linolenic (%)	Arachidic (%)	Eicosenoic (%)	Behenic (%)	Lignoceric (%)
OT	0.02 ± 0.01a	9.45 ± 0.07b	0.02 ± 0.01b	3.36 ± 0.02b	50.94 ± 0.13a	29.92 ± 0.24f	0.09 ± 0.01ef	1.57 ± 0.01a	0.91 ± 0.01g	2.39 ± 0.02f	1.34 ± 0.01i
FR	0.03 ± 0.01a	9.09 ± 0.02cd	0.02 ± 0.01b	2.27 ± 0.02g	46.73 ± 0.10e	34.79 ± 0.14a	0.12 ± 0.01cde	1.33 ± 0.01e	1.35 ± 0.01d	2.61 ± 0.02d	1.65 ± 0.01c
SEB	0.01 ± 0.01a	8.53 ± 0.08f	0.04 ± 0.01ab	2.65 ± 0.03e	49.76 ± 0.11b	32.35 ± 0.10d	0.11 ± 0.01de	1.39 ± 0.01d	1.19 ± 0.01e	2.42 ± 0.04ef	1.55 ± 0.01f
BMK	0.01 ± 0.02a	8.92 ± 0.07de	0.02 ± 0.01b	2.11 ± 0.01h	46.21 ± 0.17f	35.17 ± 0.06a	0.19 ± 0.02a	1.26 ± 0.01g	1.50 ± 0.02b	2.84 ± 0.05c	1.76 ± 0.01b
TO	0.01 ± 0.01a	10.90 ± 0.14a	0.04 ± 0.02ab	3.45 ± 0.04a	44.61 ± 0.11h	33.55 ± 0.07c	0.08 ± 0.01f	1.57 ± 0.02a	0.88 ± 0.01g	3.54 ± 0.05a	1.36 ± 0.01h
BER	0.02 ± 0.00a	9.21 ± 0.01c	0.04 ± 0.01a	3.02 ± 0.02d	48.65 ± 0.07d	32.37 ± 0.12d	0.12 ± 0.02bcd	1.48 ± 0.01c	1.13 ± 0.01g	2.49 ± 0.02e	1.46 ± 0.01g
MET	0.01 ± 0.01a	8.78 ± 0.05e	0.02 ± 0.01b	2.12 ± 0.01h	45.83 ± 0.09g	35.07 ± 0.14a	0.15 ± 0.01ab	1.32 ± 0.01e	1.74 ± 0.01a	3.13 ± 0.04b	1.82 ± 0.02a
AD	0.02 ± 0.02a	8.42 ± 0.05f	0.03 ± 0.02ab	3.21 ± 0.01c	49.37 ± 0.16c	31.59 ± 0.09e	0.05 ± 0.01g	1.53 ± 0.01b	1.40 ± 0.01c	2.77 ± 0.05c	1.61 ± 0.02d
OS	0.02 ± 0.01a	8.78 ± 0.06e	0.03 ± 0.01ab	2.12 ± 0.01h	47.05 ± 0.16e	34.42 ± 0.09b	0.15 ± 0.01bc	1.27 ± 0.01g	1.51 ± 0.02b	2.87 ± 0.03c	1.78 ± 0.01b
TIM	0.01 ± 0.01a	8.77 ± 0.06e	0.02 ± 0.01ab	2.38 ± 0.02f	48.54 ± 0.11d	33.26 ± 0.17c	0.14 ± 0.01bcd	1.29 ± 0.02f	1.37 ± 0.02cd	2.62 ± 0.02d	1.58 ± 0.01e
Sign.	n.s.	***	***	***	***	***	***	***	***	***	***

Table 6 Fatty acid methyl ester elaboration of the ten peanut landrace oils. Means in the same column with different lowercase letters are statistically different. Significance level: *** $p < 0.001$.

	SFA(%)	UFA(%)	MUFA(%)	PUFA (EFA) (%)	UFA/SFA	MUFA/ PUFA	SFA/PUFA	C18:1/C18:2	C18:1/C16:0	18:2ω6/18:3ω3
OT	18.12 ± 0.11b	81.88 ± 0.11f	51.87 ± 0.14a	30.01 ± 0.24g	4.52 ± 0.03f	1.73 ± 0.58a	0.49 ± 0.00	1.70 ± 0.53a	5.39 ± 1.80c	332.44 ± 31.88bc
FR	16.98 ± 0.05de	83.02 ± 0.05cd	48.10 ± 0.10e	34.91 ± 0.13bc	4.89 ± 0.02c	1.38 ± 0.82g	0.54 ± 0.00	1.34 ± 0.74ef	5.14 ± 6.21e	289.92 ± 20.81cd
SEB	16.56 ± 0.03g	83.44 ± 0.03a	50.98 ± 0.12b	32.46 ± 0.10e	5.04 ± 0.01a	1.57 ± 1.16c	0.49 ± 0.00	1.54 ± 1.10b	5.83 ± 1.33a	294.04 ± 10.79cd
BMK	16.91 ± 0.12e	83.09 ± 0.12c	47.74 ± 0.15f	35.35 ± 0.08a	4.91 ± 0.04c	1.35 ± 1.93g	0.60 ± 0.00	1.31 ± 2.63g	5.18 ± 2.41de	185.11 ± 12.72d
TO	20.84 ± 0.05a	79.16 ± 0.05g	45.53 ± 0.11g	33.63 ± 0.07d	3.80 ± 0.01g	1.35 ± 1.56g	0.50 ± 0.00	1.33 ± 1.62fg	4.09 ± 0.82f	419.38 ± 43.75b
BER	17.68 ± 0.04c	82.32 ± 0.04e	49.82 ± 0.07c	32.49 ± 0.10e	4.66 ± 0.01e	1.53 ± 0.65d	0.51 ± 0.00	1.50 ± 0.59c	5.29 ± 13.22cd	269.75 ± 26.54cd
MET	17.18 ± 0.05d	82.82 ± 0.05d	47.59 ± 0.09f	35.22 ± 0.14ab	4.82 ± 0.02d	1.35 ± 0.66g	0.48 ± 0.00	1.31 ± 0.67g	5.22 ± 2.07de	233.80 ± 6.66cd
AD	17.57 ± 0.09c	82.43 ± 0.09e	50.79 ± 0.18b	31.64 ± 0.09f	4.69 ± 0.03e	1.61 ± 1.92b	0.49 ± 0.00	1.56 ± 1.88b	5.86 ± 3.19a	631.80 ± 72.93a
OS	16.84 ± 0.05ef	83.16 ± 0.05bc	48.60 ± 0.14d	34.57 ± 0.09c	4.94 ± 0.02bc	1.41 ± 1.51f	0.62 ± 0.00	1.37 ± 1.78e	5.36 ± 2.60c	229.47 ± 14.56cd
TIM	16.65 ± 0.08fg	83.35 ± 0.08ab	49.94 ± 0.10c	33.41 ± 0.17d	5.01 ± 0.03ab	1.50 ± 0.59e	0.56 ± 0.00	1.46 ± 0.64d	5.54 ± 1.91b	237.57 ± 13.78cd
Sign.	***	***	***	***	***	***	***	***	***	***

Country[18].

The contents of linolenic acid, the second EFA, was along the same line with linoleic acid and was always lower than 0.20% (Table 5). Similar results were reported by other Authors[19, 21, 30].

The fatty acids in Table 5 were grouped as saturated (SFA), unsaturated (UFA), poly-unsaturated (PUFA), and mono-unsaturated (MUFA). As linoleic and linolenic acids are known to have beneficial health effects, being essential fatty acids, they were grouped as EFA. Certain ratios among individual fatty acids or among groups of them were also calculated. UFA were largely the most represented in the oils from the Algerian landraces. In the oil from Tonga (TO) the UFA were below 80%, whereas in all the remaining oils the UFA content was among 81.88 (OT) and 83.44% (SEB). Among the studied landraces, the lowest MUFA was found in TO (45.53%), mainly as a consequence of its lowest oleic acid content.

Biodiesel (fatty acid methyl esters) is derived from the transesterification of vegetable oils catalyzed by acids, bases or enzymatic catalysts, in the presence of methanol[33]. The methyl esters mixture (or biodiesel fuel) is similar to fossil diesel fuel in terms of cetane number and kinematic viscosity, but does not contain petroleum products and sulfur compounds[34]. By considering the Algerian peanut oils as a possible raw material for biodiesel production, under the specifications of the European standard EN 14214:2014[35], the contents of linolenic acid methyl ester found in all the tested oils were largely below 12%, and the PUFA (4 double bonds) methyl esters were in all cases lower than 1% of the total FAME (Tables 5 and 6).

3.10 Iodine value

The IV, expressed as g I_2 (100 g of oil)$^{-1}$, defines how much iodine monochloride reacts with the unsaturated bonds to produce a di-halogenated single bond. The IV measures the drying property of an oil: if its value is below 100, between 100 and 140, or above 140, the oil is labeled as non-siccative, semi-siccative or siccative, respectively. An high IV denotes an oil not suitable as a food.

The Codex-standard[26] indicates an IV range between 86 and 107 for peanut oil and Table 7 shows that the present oils were all within the above range. Although within the Codex-Standard range, the oil from the MET landrace (Northern Sahara) showed the highest IV (101.54) and the sole exceeding the threshold of 100, above which a vegetable oil is reputed to behave as a semi-siccative one. Such feature confirms that the oil from the MET landrace exhibited the worst physicochemical quality among the studied Algerian landraces. As far as the IV is concerned, the oil from AD (Central Sahara) was the best among the studied group of peanut landraces, with an IV as low as 90.70. All the peanut oils studied in the present work showed an IV well below the 123.22 figure found in Turkish peanut oil[36].

The IV value is related to the number of double bonds of the FAME (biodiesel); the higher the double bond number, the higher the attitude of the biodiesel to undergo oxidation. As for food purpose, also for biodiesel production the best attitudes, among the studied peanut landraces, were exhibited by AD and OT, whose IV were 90.70 and 91.85, respectively (Table 7). These values were low if compared with other vegetable oils, e.g. tomato seed oil, for which a 108.6 – 118.5 IV range was reported[14].

3.11 Acid Value

For biodiesel production from a vegetable oil, a low acid value, i.e. a low content of free fatty acids (FFA), is required. Ideally, the FFA level in an oil should be below 0.3-0.5%, for alkaline transesterification to take place[37].

In the presence of FFA, in fact, chemical transesterifica-

Table 7 Properties of the ten peanut landrace oils for biodiesel use. Means in the same column with different lowercase letters are statistically different. Significance level: *** $p < 0.001$.

	Acid value (mg KOH/g)	Iodine Value (g I_2/100g of oil)	Ca (mg kg^{-1})	Mg (mg kg^{-1})	Cetane number	HHV (Mj kg^{-1})
OT	1.55 ± 0.12cd	91.85 ± 0.77d	315.80 ± 0.31c	46.62 ± 0.20d	58.64 ± 0.08a	37.34 ± 0.22c
FR	1.23 ± 0.10d	95.38 ± 0.49c	180.38 ± 0.37h	32.37 ± 0.11g	57.07 ± 0.03g	39.27 ± 0.16a
SEB	2.40 ± 0.16a	98.24 ± 0.71b	279.85 ± 0.26d	47.20 ± 0.13c	57.54 ± 0.01e	37.46 ± 0.12c
BMK	1.61 ± 0.04c	98.76 ± 0.55b	180.39 ± 0.20h	30.65 ± 0.14h	56.88 ± 0.03i	38.21 ± 0.79abc
TO	1.56 ± 0.08cd	94.88 ± 0.86c	364.07 ± 0.18b	48.62 ± 0.18b	58.50 ± 0.01b	38.84 ± 0.54ab
BER	1.24 ± 0.10d	94.50 ± 0.36c	186.82 ± 0.15g	32.52 ± 0.13g	57.91 ± 0.03d	37.46 ± 0.09c
MET	2.00 ± 0.10b	101.54 ± 0.50a	235.38 ± 0.17f	37.96 ± 0.12e	56.95 ± 0.04hi	37.72 ± 0.16bc
AD	1.55 ± 0.14cd	90.70 ± 0.97d	259.64 ± 0.17e	33.24 ± 0.15f	58.03 ± 0.01c	37.85 ± 0.30bc
OS	2.36 ± 0.12a	97.74 ± 0.64b	364.84 ± 0.14a	62.83 ± 0.14a	57.03 ± 0.00gh	37.92 ± 0.18bc
TIM	2.01 ± 0.16b	98.40 ± 0.56b	160.83 ± 0.18i	29.34 ± 0.20i	57.29 ± 0.06f	37.76 ± 0.73bc
	***	***	***	***	***	***

tion brought about by alkaline catalyst (such as NaOH, KOH, CH_3ONa or CH_3OK), reduces biodiesel yield due to collateral and undesired saponification reaction[33]. A high FFA content in an oil deactivates the catalyst, and the addition of excess catalyst as compensation makes things worse, giving rise to the formation of emulsions which increases oil viscosity. This leads to the formation of gels and the associated problems of glycerol separation and loss in ester yields[38].

The standard DIN V 51605[39] for rapeseed oil establishes 2.0 mg KOH (g of oil)$^{-1}$ as the maximum admissible acid value. Among the tested peanut landraces, only SEB [2.40 mg KOH (g of oil)$^{-1}$] and OS [2.36 mg KOH (g of oil)$^{-1}$], both of Saharian origin, slightly exceeded the above limit (Table 7); MET [2.00 mg KOH (g of oil)$^{-1}$] and TIM [2.01 mg KOH (g of oil)$^{-1}$] were borderline oils, whereas all the remaining landraces produced oils well below the maximum of the German Standard[39]. Zhang *et al.*[29] found an acid value ranging from 0.38 to 0.61 mg KOH g^{-1} in peanut oils from roasted and unroasted seeds after aqueous enzymatic extraction.

3.12 Calcium and magnesium

Calcium and magnesium, as well as other metals, are regarded as undesirable components of fuels because of their attitude to lower the performances of the engine (corrosion). The highest level of Ca were found in the OS and TO oils (364.84 and 364.07 mg kg^{-1}, respectively), twice or more than twice higher than in FR (180.38 mg kg^{-1}), BMK (180.39 mg kg^{-1}), or TIM (160.83 mg kg^{-1}; Table 7). The highest Mg content (62.83 mg kg^{-1}) was found in the OS oil, whereas all the remaining landraces showed a Mg content in their oils lower than 50 mg kg^{-1}. Interestingly, a relationship was found among the levels of the two metals in the peanut oils ($R^2 = 0.829$): the higher the Ca content, the higher the Mg content. Lamas *et al.*[40] found 95.34 mg kg^{-1} Ca and 105.04 mg kg^{-1} Mg in crude sunflower oil, whereas the Ca + Mg content was lowered after oil degumming.

3.13 Cetane number

The CN is an important parameter when evaluating the ignition properties of a diesel/biodiesel, or the suitability of a vegetable oil as a source for biodiesel production. A fuel with an high cetane number is faster to ignite and improves the combustion quality, whereas a low CN implies ignition delay and incomplete combustion. The calculated CN for the Algerian peanut oils ranged from 56.88 ± 0.03, found in BMK, to 58.64 ± 0.08 found in OT (Table 7), i.e. largely above the 39 minimum reported in the DIN 51605:2010[39] for rapeseed oil and slightly higher than the value range found in tomato seed oil (52.81 – 54.47)[14].

3.14 Higher heating value

The calorific value of a fuel decreases, and the same does its HHV, as far as its oxygen content increases. The calorific value of FAME is lower than that of diesel because of the oxygen content of the former. For this reason, the calorific value of biodiesel is found to be slightly lower than that of diesel[41].

The standard DIN 51605 for rapeseed oil sets 36 MJ kg^{-1} as the minimum value for a biodiesel. The HHV in peanut oils from the Algerian landraces ranged from 37.34 in OT to 39.27 in FR (Table 7), thus all of them were well above the afore mentioned limit. The oil produced from FR seeds showed therefore the best characteristics. Ramadhas *et al.*[42] listed the Lower Heating Value of certain vegetable oils and found: 37.5 in rubber seeds oil, 39.5 in sunflower seed oil, 37.6 in rapeseed oil, 39.6 in cotton seed oil and 39.6 in soybean oil, by and large in the same range of the Algerian peanut oils.

3.15 One way ANOVA

Oil content and oil recovery of the ten peanut landraces showed highly significant differences among each other ($p < 0.001$) in the 1st oil extraction and in the total oil content calculated on dry seeds (Table 2). No significant inter-sample variability was instead observed in the 2nd and 3rd extractions and in the oil recovery calculation (Table 2). The geographical origin of the peanut landraces high significantly ($p < 0.001$) influenced oil free acidity and saponification number. The oils from SEB and OS landraces (both of Saharian origin) showed at the same time significantly higher FA and SV respect to the other peanut landraces (Table 2).

Remarkably, the oil extracted from the MET landrace showed significantly much higher PV, RI, IV, K_{232}, K_{266}, K_{270}, K_{274} values than the other landraces, together with a significantly lower DPPH value (Table 3). This clearly indicates that, among the tested peanut oils, the MET one exhibited not only the worse oxidative status already upon extraction, but also the higher proneness to further worsen thereafter, which is confirmed by its significantly higher PUFA content (Table 6).

Difference among the ten peanut landraces in terms of Ca and Mg contents were highly significant ($p < 0.001$); OS and TIM were the two landraces showing the significantly highest and lowest metals contents, respectively, among the landraces group (Table 7).

3.16 Hierarchical Cluster Analysis

HCA was built on the basis of the following parameters: N, CP, total oil content, FA, SV, PV, RI, DPPH$^{\cdot}$ test, spectrophotometric indices, FAME, AV, IV, Ca, Mg, CN and HHV (Fig. 1). The obtained dendrogram suggests the existence of three clusters, grouped at a linkage level of 4. The peanut landraces belonging to the first cluster are FR, BER, BMK, TIM, and MET and are characterized by comparatively high values of PV, N, CP, K_{232}, K_{266}, K_{270}, K_{274},

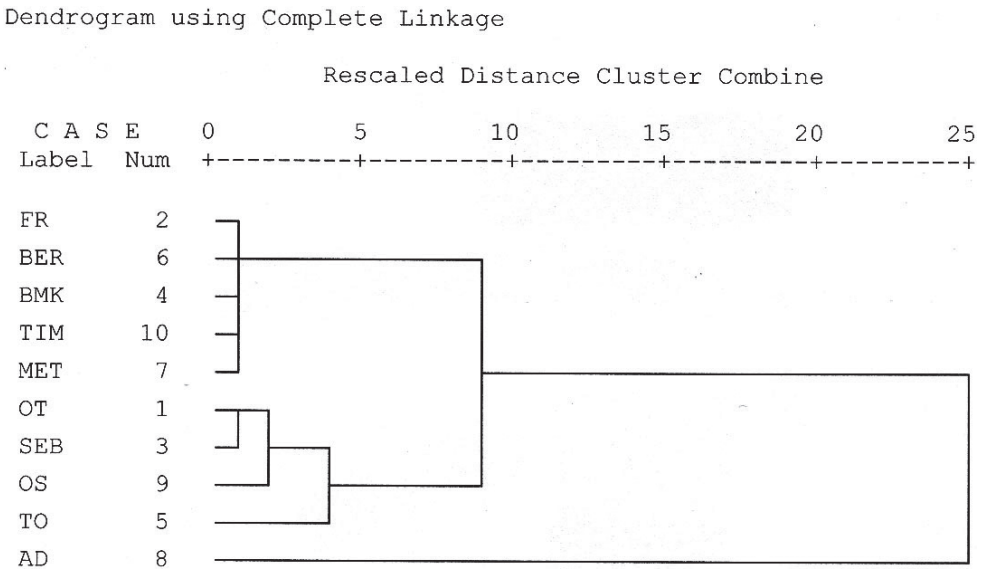

Fig. 1. Dendrogram resulting from a cluster analysis selecting the Euclidean distance as similarity measurement and Furthest neighbour method as amalgamation rule. Geographical origin: OT (Oum Tboul), FR (El Frin), BMK (Boumalek), TO (Tonga), BER (Berrihane), MET (Metlili), SEB (Seb Seb), OS (Oued souf), AD (Adrar), TIM (Timimoun).

C18:1, C18:2 and low C16:0, Ca and Mg values. The second cluster includes OT, SEB, OS, and TO and show high Ca and Mg contents, as well as an oil content in the range 37-39%.

The third cluster contains the AD landrace alone, the most southern one, with the highest oil content, K_{232} index (3.2106), linoleic/linolenic ratio (631.80), and oleic/palmitic ratio (5.86), and the lowest SV, RI, and C16:0 (8.42%)

4 CONCLUSION

The oils extracted from the seeds of ten Algerian peanut landraces were evaluated here for their edible use as well as a feedstock for biodiesel production. As for edibility, and globally speaking, positive physicochemical properties were apparent in terms of oil content, free acidity, oleic acid, and essential fatty acids, whereas a criticism has to be raised in relation to oil stability to oxidation. The defatted seeds also showed a good crude protein content. As for biodiesel production, the peanut oils showed, on average, low acid values and iodine values. In general, calorific values were slightly above the minimum requested by the German standard for rapeseed oil, whereas the cetane numbers were largely above the minimum imposed by the same standard, suggesting that the peanut oil from the 10 Algerian landraces can be used for biodiesel production. One way ANOVA and hierarchical cluster analysis demonstrated that ample and significant differences exist among the oils of the ten studied peanut landraces.

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