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Fatty Acid Composition and Physicochemical Properties of Four Varieties of *Citrullus lanatus* Seed Oils Cultivated in Côte d'Ivoire

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Authors' contributions

This work was carried out in collaboration between all authors. Author EAD designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors AMNN carried out all laboratoire work. Authors SB and YDD performed the statistical analysis, managed the analyses of the study. Authors SD and JPENK managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

In this study, the physicochemical properties and fatty acids composition of four varieties of *Citrullus lanatus seed* oils cultivated in Côte d'Ivoire were investigated. There was one variety (Bebu cultivar) with oval flat seeds and thick edges (COS), and three varieties (wlêwlê cultivars) with flat seeds disentangled and sharp extremity of various size (big seed (CBS), average seed (CAS) and small seed (CSS)). The pH at 30ºC was ranged from 5.85±0.003-6.03±0.001. The results for density (g/ml), refractive index, unsaponifiable matter (% of oil) and impurity (%) were 0.876±0.005-0.924±0.001; 1.4595±0.004-1.4739±0.001; 0.89±0.450-2.07±0.160; 0.05±0.002-

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0.1±0.001; respectively. The free fatty acid (%), acid value (mgKOH/g oil), peroxide index (meq O2/kg oil), saponification value (mgKOH/g oil), iodine value (mg of g I_2/100 g oil), Total polyphénolics content (mg gallic acid/kg oil) values of the oils were obtained in the range; 0.80±0.050-2.03±0.010; 6.03±0.350-7.93±0.100; 10.93±0.470-13.80±0.100; 228.43±0.040-244.76±0.010; 84.07±11.250-93.38±1.090; 0.125±0.050-0.291±0.010; respectively. The most abundant fatty acid detected in the four oils was oleic acid ranged from 46.56-59.97. This acid constituted with the aldaic acid, the monounsaturated fatty acids in these oils and accounted for 64.82-73.07. This study showed the presence of others main saturated fatty acids as stearic acid and palmitic acid. The properties of these oils were quite comparable to those of previously reported cucurbitaceaes seeds oils, suggesting their potential use as good table and cooking oils. With a high yield of oil and physicochemical characteristics similar to those of the other commercial edible oils, the *Citrullus lanatus* seed oil can be considered as a new and valuable source of edible oil. This study also showed potential for *Citrullus lanatus* seeds oils to ave relatively high oxidative stability that would be suitable for food and industrial applications.

Keywords: Citrullus lanatus; varieties; seed oil; physicochemical properties; fatty acids composition.

1. INTRODUCTION

Citrillus lanatus belongs to the family of Cucurbitaceae which has a tremendous genetic diversity, extending to vegetative and reproductive characteristics [1]. They thrive in tropical, subtropical, arid deserts and temperate locations. This specie is an annual, herbaceous, monoecious plant with a non-climbing creeping habit [1]. This flowering plant produces a special type of fruit known by botanists as a pepo, which has a thick rind (exocarp) and fleshy centre (mesocarp and endocarp); pepos are derived from an inferior ovary and are characteristic of the Cucurbitaceae. The watermelon fruit, loosely considered a type of melon has a smooth exterior rind (green and yellow) and a juicy, sweet, usually red, but sometimes orange, yellow, or pink interor flesh [2]. *Citrillus lanatus* seeds have both nutritional and cosmetic importance, the seed contain vitamins (B1, minerals, riboflavin, fat, carbohydrates and proteins [3].

In west Africa, a region where soups are integral to life, *Citrillus lanatus* seeds are a major soup ingredient and a common component of daily meals. Coarsely ground up, they thicken stews and contribute to widely enjoyed steam dumplings. Some are soaked, fermented, boiled and wrapped in leaves to form a favourite food seasoning. Despite being a significant foodstuff even by global standard, pistache is hardly known to nutritionists outside a few west African nations. Little nutrional detail on *Citrullus lanatus* seed oil is readly available to an international readership. Research studies have shown that these seeds contained about 50% oil [4], 42-57% oil [5], 44-53% oil [6] for seeds cultivated in different bioclimatic regions of cameroon and 56,9% oil for *Citrullus lanatus* from Nigeria [7]. These studies showed that *Citrullus lanatus* seeds contained good amounts of oil that can be exploited. The aim of the present study therefore is to determine some physical and chemical properties as well as the fatty acid composition from four varieties of *Citrullus lanatus* seeds oils cultivated in Côte d'Ivoire.

2. MATERIALS AND METHODS

2.1 Sample Collection

This study was carried out on four varieties of *Citrullus lanatus* grown at the experimental station of Nangui Abrogoua University, at Abidjan Côte d'Ivoire. These varieties were authenticated by a taxonomist in the Department of crop Science of this University. At maturity (3-4 months), the fruits were harvested and transported to laboratory. After the berries fermentation (5 days at 25ºC), seeds of the four obtained varieties were washed, dried (oven drying (60ºC) during 24 hours) and peeled. For this work, one variety (Bebu cultivar) with oval flat seeds and thick edges (COS) and three varieties (wlêwlê cultivars) with flat seeds disentangled and sharp extremity of various size (big seed (CBS), average seed CAS) and small seed (CSS)) were used (Fig. 1).

2.2 Oil Extraction

The dried samples (200g) were ground into powder with the laboratory pestle and mortar and kept or stored in air tight containers until required for subsequent analysis. Oil from seeds was extracted in soxhlet using hexane as solvent and

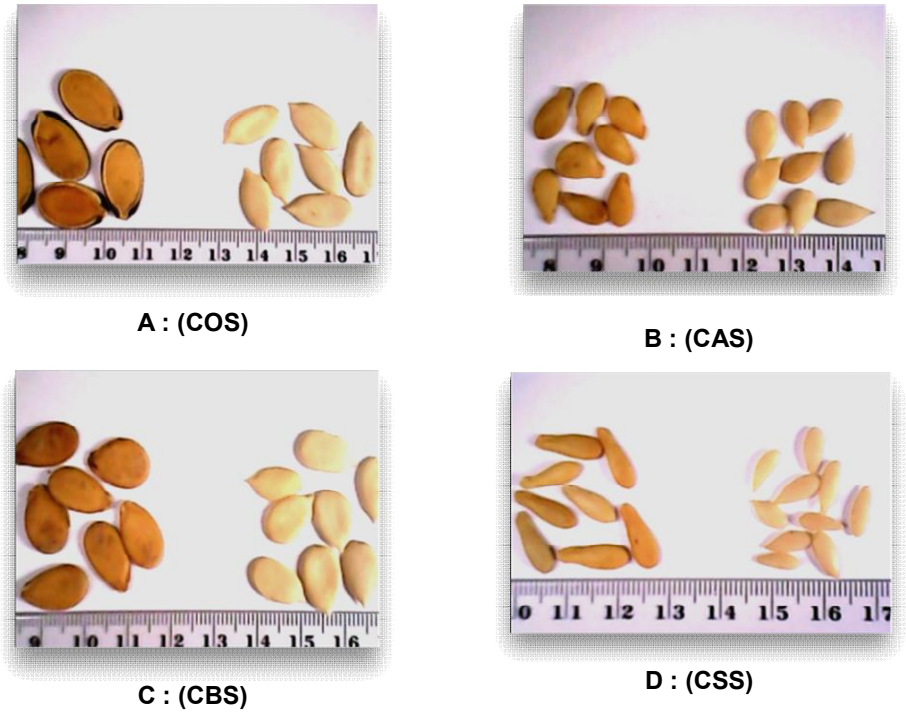
the lipid value (crude fat) was evaluated. The oil extracted after drying was transferred into a translucent glass bottle and kept in the refrigerator (4ºC) until required for analysis. The peeled seeds are dried in the steam room at 40ºC during 24 hours. 200g of almonds were weighed and crushed by means of a mixer. The obtained powder (the taking), was used to extract the oil. Extraction was done for heating during 12 hours. The organic phase containing the fat is evaporated until elimination of the solvent. Then, balloon containing oil is placed in the steam room at 70ºC during 30 min for the evaporation of solvent.

2.3 Physicochemical Properties Analysis

The physicochemical parameters (refractive index, acid, iodine, peroxide and saponification values, density) were carried out according to the methods described [8]. Colour was measured with a Lovibond tintometer ; unsaponifiable matters was carried out by the procedure of Lozano et al. [9]. Determination of the total polyphenolics content was done spectrophotometrically using Folin-Ciocalteau's reagent as described by Capannesi et al. [10]. The determination of the pH value is carried out by measurement of the potential difference between electrodes immersed in standard and test solutions. The standard solutions used are assigned a definite pH value by convention.

2.4 Fatty Acid Composition Analysis

The fatty acid composition of oils was determined by gas chromatography (GC) as fatty acid methyl esters (FAMEs). FAMEs were prepared by saponification/methylation with sodium methylate according to European Regulations (EEC 2568/91). A chromatographic analysis was performed in a Hewlett-Packard model 4890D gas chromatograph equipped with a 30m x 0.25 mm x 0.25µm film thickness fused silica capillary column (Innowax) coupled to a flame ionization detector (column temperature 210ºC). Both the injector and the detector were maintained at 230ºC and 250ºC, respectively. Nitrogen was used as the carrier gas at 1ml/min with Split injector system (Split ratio 1 :100). Fatty acids were identified by comparing their retention times with those of standard compounds (butyric; caprylic; capric; lauric; mirystoleic; myristic; palmitoleic; palmitic; linolenic; linoleic; oleic; aldaric; stearic; arachibonic and arachidique acids from Sigma-Aldrich).



A : (COS) **B : (CAS)**

C : (CBS) **D : (CSS)**

Fig. 1. Seeds of cucurbit *Citrullus lanatus* varieties: A: Oval flat seeds and thick edges (COS); B: average seeds (CAS); C: big seeds (CBS); D: small seeds (CSS)

3. RESULTS AND DISCUSSION

3.1 Physicochemical Characterization of Four Varieties of *Citrillus lanatus* Seed Oils

The total lipid content of *Citrullus lanatus* seeds (Table 1) were 56.08±2.16; 55.62±0.49; 51.43±1.05; 49.24±0.77 respectively for CSS; CAS; CBS and COS. From the results, these varieties of *Citrullus lanatus* could be considered as good lipid sources. These values were similar to those reported by Essien and Eduok [11] for southern Nigeria *Citrullus lanatus* seed oil (57.26 %), Fokou et al. [12], for Cucurbitaceae oils from four different regions in Cameroon, (49.01-52.15g/100g) but higher in value than L. breviflora seed oil (22.9%) [13] or *Lagenaria siceraria* (Mol.) cultivars from Nigeria (38.10-43.65%) [14].

Physicochemical properties of ivorian *Citrullus lanatus* seeds oils are shown in Table 1.

The refractive indices measured in the present work (ranging from 1.4595-1.4739), were similar to those reported by Lazos [3] for pumpkin (1.4616), Oluba et al. [7] for egusi melon seed oil (1.45) or Ardabili et al. [15] for pumpkin seeds (1.4662).

This shows that these oils are less thick compared with most drying oils whose refractive indices were between 1.48 and 1.49 [16].

The refractive index is used by most processors to measure the range in unsaturation as the fat or oil is hydrogenated. The refractive index of oils depend on their molecular weight, fatty acid chain length, degree of unsaturation [17].

Saponification index is an indicator of the average molecular weight and hence, chain length. It depends on the neutralization of the free fatty acids in oil and complete saponification of the fatty material [18]. The saponification indices of Ivorian *citrullus lanatus* (CSS; CAS; CBS and COS) were respectively 244.76; 228.43; 239.61 and 239.34 mg KOH/g of oil. These values agree with those of saturated fatty acids rich oils such as *Cocos nucifera* coconut (248-265 mg KOH/g of oil) and *Elais guineensis*, palm kernel oils (230-254 mg KOH/g of oil) [19]. As reported by Pearson [20], oil with higher saponification values contain high proportion of lower fatty acids. Therefore, the values obtained for *citrullus lanatus* oils in this study show that they contain high amounts of higher fatty acids.

These values were also slightly higher than those of non-conventional oils such as Dacryodes elulis, the African pear (201.4 mg KOH/g of oil) [21], Coula edulis (180-185 mg KOH/g of oil) [22], C. schweinfurthii (177-197.79 mg KOH/g of oil) [23].

The iodine indices obtained in this study for the studied oils were 93.38; 92.97 83.25 84.07±11.25, g I_2/100g respectively for CSS; CAS; CBS and COS. The iodine index is a characteristic of the unsaturation of a fatty acids or its esters. Lipids with unsaturated fatty acids (containing one or more double bonds) are easily assimilated and broken down to produce calorific energy than saturated fatty acids. The higher the iodine value, the more unsaturated the oil. However, when the iodine value becomes too high, the stability of the oil reduces because it is more likely to undergo oxidation. These iodine values of CSS, CAS and CBS oils, ranged (80 to 100 g I_2/100g), were similar to those of unsaturated fatty acid rich oils such as peanut (86.06-107.0 g I_2/100g), cotton seed (100.0-123.0 g I_2/100g), but lower than that of sunflower (118.0-141.0 g I_2/100g), of soybean oil (124.0-139.0 g I_2/100g) [24]. However, *C. lanatus* has higher iodine value than those of saturated fatty acid-rich oils such as Theobroma cacao, cocoa butter (32.0-42.0 g I_2/100g) [25], coconut (6.0-10.0 g I_2/100g), palm oil (50.0-55.0 g I_2/100g), palm kernel (14.0-1.0 g I_2/100g) [24].

The percentage of impurity (acid/saponification index) ranged from 0.01 (COS) to 0.05 (CSS; CAS and CBS). The percentage of impurities (high amount of free fatty acids) may be due to processing methods, storage conditions and extraction method used in the laboratory [25].

Unsaponifiable matters in the vegetale oils are a variety of nonglyceridic bioactive substances containing variable mixture of hydrocarbons, aldehydes, ketones, alcohols, sterols, pigments, and fat-soluble vitamins that may occur naturally or may be formed during processing or degradation of oils [26]. The content of unsaponifiable matters (2.07±0.16; 0.89±0.45; 1.31±0.32; 1.31±0.31) for CSS; CAS; CBS and COS respectively were much lower than the values reported by Answar et al. [27] for rice bran oil (3-7%), Ardabili et al. [15], for the pumpkin seed oil (5.7±0.8%). But *citrullus lanatus* unsaponifiable matters from CAS; CBS and COS were in a close agreement with the result of Milovanovic and Jovanovic [28], for *Citrullus colocynthis* L. seed oil (1.02%).

The acid value represents free fatty acid content due to enzymatic activity. The acid values were 7.93±0.1; 6.92±0.17; 6.03±0.35 and 7.23±0.13 mg KOH/g for CSS; CAS; CBS and COS respectively. While the free fatty acid values were 1.84±0.02; 2.03±0.01; 1.53±0.03; 0.80±0.05% respectively which were below 5.00% free fatty acids content recommended as the maximum for non-rancid oil [29,30]. This implies that the studied oils were not rancid.

The phenolic compounds obtained in this study were 0.125±0.05; 0.291±0.01; 0.251±0.013 and 0.167±0.03 g/l respectively for *citrullus lanatus* CSS; CAS; CBS and COS. These compounds represent potentially health-promoting substances and have industrial applications [31]. These naturally occuring compounds have proven to play important role in the stability and sensory and nutritional characteristics of the product and may prevent deterioration through quenching of radical reactions responsible for lipid oxidation [32]. Phenolic compounds values of this work were in agreement with those usually obtained from seed oils (0.1 to 0.3 mg/kg) [15]. Cultivar, extraction system, and the conditions of processing and storage are critical factors for the content of phenolic compounds [33].

3.2 Oil Fatty Acid Composition

Table 2 shows that the most abundant fatty acid detected in the four varieties of *Citrullus lanatus* seeds oils from Côte d'Ivoire was oleic acid (46.56-59.97).

This study showed the presence of others main fatty acids as elaidic acid (13.10-21.23), palmitic acid (12.03-13.83) and stearic acid (09.49-11.84) in the oils.

The monounsaturated fatty acids (MUFA) of cultivars oils we studied (64.82; 69.90 ; 73.07, 67.79, respectively for CBS, COS, CAS, CSS) were similar to those reported by Essien et *al*. [14] for *lagenaria siceraria* seeds oils (66.68-71.89%) but higher than those reported by Oluba et al. [7], for Nigerian *Citrullus lanatus* seeds oils (57.4%). As can be seen in Table 2, monounsaturated fatty acid have great importance because of their nutritional implication and effect on the oxidative stability oils. These oils high in MUFA have been well documented to provide numerous health benefits [34,35]. Our results with oleic acid as the predominant level in these *Citrullus lanatus* seeds oils were so similar to those of Idouraine et al. [36] and Zdunczyk et al. [37] who showed that C. pepo seeds contain oleic acid as the most abundant fatty acid and C. pepo subsp. These results revealed that ivorian's *Citrullus lanatus* seeds oils were in agreement with all other studies of Cucurbitaceae seeds oils which were low in linolenic acid (<1%) ([38-40]). Unsaturated fatty acids values of *Citrillus lanatus* seeds oils from Cote d'Ivoire suggest that they could be some goods sources of edibles oils (for cooking) and a potential antidote for fight against cardiovascular disease.

Table 1. Physicochemical characteristics of four varieties of *Citrullus lanatus* seeds oils from Côte d'Ivoire

Cultivars / Parameters	CSS	CAS	CBS	COS
Total lipid content	56.08±2.16^{a}	55.62±0.49^{a}	51.43±1.05^{a}	49.24±0.77^{a}
pH at 30ºC	5.93±0.001^{b}	5.90±0.002^{b}	5.85±0.003^{c}	6.03±0.001^{a}
Density (g/ml)	0.891±0.003^{b}	0.924±0.001^{a}	0.916±0.001^{a}	0.876±0.005^{c}
Refractive Index	1.4732±0.001^{a}	1.4686±0.003^{a}	1.4739±0.001^{a}	1.4595±0.004^{a}
Unsaponifiable matter (% of oil)	2.07±0.16^{a}	0.89±0.45^{c}	1.31±0.32^{b}	1.31±0.31^{b}
Impurity (%)	0.05±0.002^{b}	0.05±0.002^{b}	0.05±0.002^{b}	0.1±0.001^{a}
Free fatty acid, (% of oleic acid)	1.84±0.02^{b}	2.03±0.01^{a}	1.53±0.03^{c}	0.80±0.05^{d}
Acid value (mgKOH/g of oil)	7.93±0.1^{a}	6,92±0.17^{b}	6.03±0.35^{c}	7.23±0.13^{b}
Peroxide index meq O2/kg of oil	12.34±0.23^{b}	10.93±0.47^{c}	12.38±0.3^{b}	13.80±0.1^{a}
Saponification value mgKOH/g oil	244.76±0.01^{a}	228.43±0.04^{c}	239.61±0.02^{b}	239.34±0.02^{b}
Iodine value (g I_2/100 g of oil)	93.38±1.09^{a}	92.97±0.98^{a}	83.25±1.34^{b}	84.07±11.25^{b}
Total polyphénolics content (mg gallic acid/kg oil)	0.125±0.05^{c}	0.291±0.01^{a}	0.251±0.013^{b}	0.167±0.03^{c}
Color	Yellow orange	yellow	Light yellow	Light yellow

Mean± standard deviation of three determinations. In the same row, mean values followed by the same letter (superscript) are not significantly different (p>0.05)

Table 2. Fatty acids composition of four varieties of *Citrullus lanatus* seeds oils from Côte d'Ivoire

samples / Fatty acid	Carbons and bonds	CBS	COS	CAS	CSS
Lauric acid	C12 :0	1.67	1.07	1.03	1.34
Myristic acid	C14 :0	1.10	_	_	1.24
Palmitic acid	C16 :0	13.82	13.44	12.03	12.71
Stearic acid	C18 :0	11.84	11.62	09.49	09.71
Oleic acid	C18 :1	50.27	52.68	59.97	46.56
Elaidic acid	C18 :1 Trans-9	14.55	17.22	13.10	21.23
Saturated fatty acids (SFA)		28.43	26.13	22.55	25
Monounsaturated fatty acids (MUFA)		64.82	69.90	73.07	67.79

The four cucurbits oils studied contents low amounts of saturated fatty acids (22.55-28.43) and high contents of unsaturated fatty acids (64.82-73.07). It is now widely accepted that diet with low saturated fatty acids and high in unsaturated fatty acids is beneficial for health.

The SFA such as lauric; myristic and to a lesser extent, palmitic acids, elevate serum cholesterol and LDL levels. Stearic acid does not elevate serum cholesterol or LDL levels, but its other health effects are yet undefined [41].

The colour of the oil is used preliminarily in judging the quality and in determining the degree of bleaching of the oil. For Powe [42], the darker the colour, the poorer the quality. Therefore the pale yellow colour of *Citrullus lanatus* seeds oils shows that the quality of the oil is good and confirms to Encyclopedia of Chemical Technology [42,43].

4. CONCLUSION

The results of the present study showed that ivorian's *Citrullus lanatus* seeds oils contained appreciable physicochemical characteristics which suggest that they could be good sources of edibles oils for cooking. The oils extracted from four varieties of *Citrullus lanatus* revealed that the most abundant fatty acid detected was oleic acid (46.56-59.97). Theses oils contents make them suitable as food supplement. They may also enjoy applications as industrial ingredients in soap production, cosmetics and food complements. Their chemical properties and fatty acids was quite comparable to those of previously reported cucurbitaceae seeds oils, suggesting their potential use as good table and cooking oils. With a high yield of oil and physicochemical characteristics similar to those of the other commercial edible oils, the *Citrullus lanatus* seeds oils can be considered as a new and valuable source of edible oil. This study also showed potential for *Citrullus lanatus* seeds oils to have relatively high !oxidative stability that would be suitable for food and industrial applications.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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