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A Comparative Study of the Properties of Selected Melon Seed Oils as Potential Candidates for Development into Commercial Edible Vegetable Oils

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Abstract A comprehensive compositional and characterization study was carried out on five seed oils from varieties of the melons *Citrullus lanatus* and *C. colocynth* in order to evaluate their suitability for large-scale exploitation as edible vegetable oils. The oils were extracted by Soxhlet with a 3:1 mixture of *n*-hexane/2-propanol with yields that ranged from 24.8 to 30.0% (wt/wt). The refractive indices and relative densities of the oils fell within the narrow ranges of 1.465–1.469 and 0.874–0.954 g/cm^3, respectively. Saponification values ranged between 182.1 and 193.8 mg KOH/g, whilst iodine values (IV) ranged from 95.8 to 124.0 (Wijs). The ranges of the values for free fatty acid (AV), 1.2–4.0 mg KOH/g, peroxide (PV), 1.1–10.9 meq/kg and *p*-anisidine (*p*-AV), 0.2–9.0, indicated that secondary oxidation products were barely present. GC analysis gave total unsaturation contents of 67.93–82.36%, with linoleic acid (18:2) being the dominant fatty acid (55.21–66.85%). The GC results agreed closely with those from proton NMR analysis of the fatty acid classes. The physicochemical and compositional properties determined in this study show that the qualities of the test Cucurbitacea seed oils are highly comparable to those of soybean, sunflower and groundnut seed oils. Therefore, the test melon seed oils could be developed into commercial products to serve as alternate vegetable oils in Southern and West Africa, the regions where these melons grow.

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Introduction

Species of the genus *Citrullus*, belonging to the Cucurbitacea family, usually consist of a large number of varieties that are generally known as melons. Melons are a major food crop in many tropical and subtropical parts of the world, especially in sub-Saharan Africa. In West, East and Southern Africa, several varieties of *Citrullus lanatus* and *C. colocynth* serve as a major food source. Some varieties like the watermelons are eaten as fruits, whilst others (like the tsama melon grown in the Kalahari Desert) serve as a good source of water during the dry seasons.

Throughout sub-Saharan Africa, the seeds from several melon varieties are greatly relished and are used in a variety of dishes. The use of melon seeds as a food source appears to be justified by their reported nutritional value. The dry seed of *C. lanatus* has been reported to contain on average about 22 g of protein, 30 g of fat and 11 g of carbohydrate as well as good amounts of micronutrients per 100 g sample [1]. Thus the nutritional value of melon seed compares very favorably with those of soybean, sunflower seed and groundnut [2].

Melon seeds are generally a rich source of oil. However, this resource is only used on a limited scale. For example, sesoswane seeds (from Botswana) and wrewre seeds (from Ghana) are extracted in a few rural areas for use as cooking oil. The world's supply of vegetable oil is currently in excess of 100 million metric tones (MMT), and demand is increasing at a rapid pace due to increasing demand for non–food uses of vegetable oil, for example in biodiesel, oleochemicals, lubricants, pharmaceuticals and cosmetics. However, only about 12 of the ~500,000 known plant species are currently commercially exploited—by big companies with vast resources for promotion and marketing—to produce vegetable oils in order to meet the world's increasing demand. Thus, the widespread availability of vegetable oils around the world has resulted in the development of other oil-bearing plant species being neglected, especially in developing countries, which have become net importers of vegetable oils even though they have rich resources for producing their own vegetable oil products.

This study investigates the physicochemical and compositional properties as well as the stabilities of oils obtained from the seeds of wrewre (from Ghana), sesoswane, tsama and desert melons (from Botswana), which are all varieties of *C. lanatus*, and agusi seed oil, *C. colocynth* (from Ghana). All of these melon seeds are edible and their oils are extracted on a very limited scale in the growing areas. There are a few reports in the literature about the fatty acid compositions of seed oils from some varieties of melons (*C. colocynth)* and some acknowledgement of the good quality of these seed oils [3, 4]. However, because of the current limited scale of use of the seed oils from *Citrullus* species, detailed information about their physicochemical properties, their stabilities and their potential as sources of vegetable oils for the world market is still scarce.

In this study we compare the properties of seed oils from four varieties of *C. lanatus* and one variety of *C. colocynth* with those of three vegetable oils marketed worldwide—soybean, sunflower and groundnut oils—in some detail, to find out whether these properties justify the development of these *Cucurbitacea* seed oils into commercial products. Development of these seed oils will not only add great value to these agricultural crops, but will also increase the number of sources of vegetable oil available, at least in the growing regions of the world.

Experimental Procedures

Materials

Agusi seeds, *C. colocynth*, and wrewre seeds, *C. lanatus*, were purchased from Makola Market, Accra, Ghana. The Botswana Forestry Association supplied sesoswane, tsama and desert melon seeds, varieties of *C. lanatus.*

Extraction

Solvents (Fischer Scientific, Loughborough, UK) used were of analytical grade and were not further purified. The seed samples were macerated in a Waring blender and the powders were extracted with a mixture of *n*-hexane/2-propanol (3:1 v/v) in a Soxhlet apparatus for 6 h. The resulting oils were further dried by azeotropic distillation with benzene.

Physicochemical Properties

The bulk physical and chemical properties (Table 1) were determined according to standard IUPAC methods for the analysis of oils and fats [5]. All experiments were conducted in triplicate.

Lipid Class Composition

Analysis of the lipid classes (Table 2) in each oil sample was carried out by adsorption column chromatography, using florisil (7% water w/w, Saarchem Pvt Ltd., Muldersdrift, Republic of South Africa) and gradient elution using *n*-hexane, mixtures of *n*-hexane/diethyl ether, diethyl ether, methanol and acetone [6].

Separation of Acylglycerols

Triacylglycerols (TAG), diacylglycerols with free fatty acids (DAG + FFA) and monoacylglycerols (MAG) in the oil samples were further separated by gradient elution on silica gel (Saarchem Pvt Ltd.) using benzene (100%), benzene:diethyl ether (9:1) and diethyl ether (100%), respectively [6].

Fatty Acid Composition: Sample Preparation

The oil samples (2 g each) were transesterified by refluxing in dry methanol that contained ethanoyl chloride to yield fatty acid methyl esters (FAME). These were then used for chromatographic and ^{1}H NMR analyses [7].

Instrumentation and Separation Conditions

Fatty acid compositions were determined using a Perkin Elmer Autosystem Gas Chromatograph (Norwalk, CT, USA) with on-column injection and flame ionization detection interfaced with a PE Nelson computer. The column was an Omegawax TM 320 capillary column (30 m × 0.32 mm × 0.25 mm i.d.; Supelco, Bellefonte, PA, USA). The carrier gas was nitrogen at a pressure of 15.0 psi. The oven, injection and detection temperatures

Table 1 Physicochemical properties of the *Citrullus* seed oils compared with Codex standards for soybean, sunflower and groundnut oils

Chemical parameter	Sesoswane	Agusi	Wrewre	Tsama melon	Desert melon	Codex standards[a]		
						Soybean	Sunflower	Groundnut
Refractive index (40 ± 1°C)	1.465 ± 0.001	1.467 ± 0.002	1.466 ± 0.001	1.469 ± 0.001	1.468 ± 0.001	1.466–1.467	1.467–1.469	1.460–1.465
Relative density (30 ± 1°C)	0.874 ± 0.001	0.874 ± 0.001	0.878 ± 0.001	0.889 ± 0.001	0.954 ± 0.001	0.919–0.925	0.918–0.923	.912–.914
Saponification value (mg KOH/g)	193.8 ± 0.1	189.5 ± 0.2	190.8 ± 0.1	184.4 ± 0.1	182.1 ± 0.1	189–195	188–194	187–196
Iodine value (Wijs)	107.8 ± 0.1	104.4 ± 0.2	112.2 ± 0.1	95.8 ± 0.1	124.0 ± 0.2	120–143	110–143	80–106
Acid value (mg KOH/g)	1.9 ± 0.1	4.0 ± 0.3	1.2 ± 0.1	1.8 ± 0.1	2.5 ± 0.1	4.0	4.0	4.0
Peroxide value (meq/Kg)	3.5 ± 0.1	1.1 ± 0.1	1.1 ± 0.2	9.8 ± 0.1	10.9 ± 0.1	10	10	10
p-Anisidine value (mmol/kg)	1.3 ± 0.1	0.6 ± 0.1	0.2 ± 0.1	2.2 ± 0.1	9.0 ± 0.1	–	–	–
Oxidation value[b]	8.3 ± 0.3	2.8 ± 0.3	2.5 ± 0.3	21.8 ± 0.1	30.8 ± 0.1	–	–	–
Flavor score[c]	4.8 ± 0.1	6.7 ± 0.1	6.8 ± 0.1	0.1 ± 0.0	–3.1 ± 0.0	–	–	–
Unsaponifiable matter (%, w/w)	1.2 ± 0.1	0.6 ± 0.1	1.1 ± 0.1	2.6 ± 0.1	1.8 ± 0.1	1.5	1.5	1.5
Yield (% w/w)	24.8 ± 0.2	30.0 ± 0.9	27.5 ± 0.5	24.8 ± 0.2	28.0 ± 0.4	20–22	40–50	40–45

Values represent the average of three replicate analyses ±SD

[a] From [9]

[b] Values calculated from Holm's equation [10]

[c] Values calculated from List's equation [10]

Table 2 Percentage composition of lipid classes and composition of acylglycerols in *Citrullus* seed oils, as estimated from adsorption column chromatography

Citrullus seed oil	Neutral lipids				Polar lipids			
	HC	TAG	STE + FFA	FST	DAG	MAG	GL	PL
Lipid classes								
Sesoswane	0.41	73.90	3.71	13.71	1.60	1.40	0.65	4.71
Agusi	0.32	74.31	3.68	13.79	1.78	1.81	0.39	3.92
Wrewre	0.29	75.62	3.67	14.00	1.42	1.70	0.79	2.51
Acylglycerols								
Sesoswane		95.6			6.73[a]	2.91		
Agusi		96.7			6.32[a]	1.85		
Wrewre		94.5			8.31[a]	0.88		

Values represent the average of two replicate analyses

HC hydrocarbons, *STE* sterol esters, *FFA* free fatty acid, *FST* free sterols, *GL* glycolipids, *PL* phospholipids

[a] DAG + FFA

were fixed at 200, 250 and 260°C, respectively. Reference compounds were standard FAME mixtures from Supelco and Sigma (Sigma Chemical Co., St Louis, MO, USA).

Nuclear Magnetic Resonance (NMR) Analysis

Proton NMR spectra of the FAME were acquired at 300 MHz using a Bruker (Billerica, MA, USA) Avance DPX300 Spectrometer.

Statistical Analysis

All experiments were carried out in triplicate unless otherwise stated; results are expressed as means ± SD. Statistical analysis was carried out using a one-way ANOVA with a significance level of $P < 0.05$. The software used for the statistical analysis was the SPSS for Windows statistical package (v.10.0.6; SPSS, Chicago, IL, USA).

Results and Discussion

The yields of oils obtained from Soxhlet extractions of the *Citrullus* seeds are shown in Table 1, which also shows literature yields for soybean, sunflower and groundnut oils [8]. The yields of sunflower and groundnut oils are appreciably higher than those of the test *Citrullus* seeds. However, it is quite clear from Table 1 that the oil contents in the melon seeds are higher than the oil contents of soybeans. Thus, this good oil yield coupled with the ease of cultivating the melon plants make these melon plants good candidates for commercial exploitation.

The physicochemical properties of the test *Citrullus* seed oils are compared with Codex standards for soybean, sunflower and groundnut oils in Table 1 [9]. It is instructive to note that the refractive indices (RIs) for the melon seed oils and those of soybean, sunflower and groundnut oils fall within the same range of 1.465–1.469. This similarity in RI suggests that the two sets of oils have similar degrees of unsaturation. The relative densities of the melon seed oils are comparable with those of soybean, sunflower and groundnut oils, again suggesting similarities in the textures of the two sets of oils. This observation is supported by the rather similar saponification values (SVs) for all of the seed oils shown in Table 1. The range of SVs observed in Table 1 clearly suggests that the melon seed oils consist mainly of medium-chain fatty acids (i.e., C16 and C18), as indeed is the case for soybean, sunflower and groundnut oils. The range of iodine values (IVs) for the melon seeds, 95.8–124.0 (Wijs), is slightly higher than the range of IVs for groundnut oil (80–106), but lower than the IV range for soybean and sunflower oils, 110–143. However, the differences in the IVs are rather small and this again corroborates the inference drawn from the RI values regarding the similarity of unsaturation in the two sets of oils. The range of acid values (AVs) for the melon seed oils, 1.2–4.0 mg KOH/g, falls well within the Codex standards for virgin vegetable oils, whilst the range of peroxide values (PVs) for the melon seed oils, 1.1–10.9 meq/kg, falls within the Codex standards for refined vegetable oils. Again except for desert melon seed oil, which has a *p*-anisidine value (*p*-AV) of 9.0 mmol/kg, the range of *p*-AVs observed for the rest of the melon seed oils, 0.2–2.2 mmol/kg, indicates the presence of only very small amounts of secondary oxidation products in these unrefined seed oils, especially for sesoswane, wrewre and agusi. These favorable stability parameters indicate that formal refinement of the melon seed oils will further improve their general quality and hence their competitiveness tremendously.

The oxidation states of the oils were further examined by combining the PVs and *p*-AVs in Holm's equation, $OV = p\text{-AV} + 2(PV)$, which is used to describe the degree of oxidation of oils [10]. Their corresponding flavor scores were also calculated from List's equation, $F = 7.7 - 0.35\ (OV)$, for predicting theoretical flavor scores [10]. The results are given in Table 1, which shows good oxidation values for wrewre and agusi seed oils, and their corresponding flavour scores are also quite acceptable. However, the oxidation value and flavor score for sesoswane are rather average, whilst the OVs for tsama and desert melon seed oils and their corresponding flavor scores are not that good. The implication here is that significant oxidation activity has taken place in these seed oils, especially for tsama and desert melon. The rather poor oxidation properties of these oils, as revealed by Holm's and List's equations, could in part be attributed to the age of the seeds, as these were bought from the open market. However, the results indicate that formal processing and refining of these oils will be required to enhance their oxidation properties and hence their general quality and acceptability.

The percent unsaponifiable matter (UM), shown in Table 1 for sesoswane (1.2%), agusi (0.6%) and wrewre (1.1%) seed oils, are within the Codex recommended maximum values for refined sunflower, soybean and groundnut oils [9], whilst the UM values for tsama and desert melon seed oils, 2.6 and 1.8%, respectively, are comparable with the Codex recommended maximum values for maize and grapeseed oils [9]. The physicochemical properties of the test melon seed oils shown in Table 1 generally indicate that these *Citrullus* seed oils will only require a simple refining treatment to become universally acceptable.

Compositional studies carried out on the *Citrullus* seed oils included determination of the lipid classes and separation of the acylglycerols by adsorption column chromatography and capillary GC determination of the fatty acid compositions. The results from the adsorption column chromatography given in Table 2 shows that seed oils from sesoswane, agusi and wrewre are composed mainly of neutral lipid classes dominated by triacylglycerols. The percent compositions of sterol esters and free sterols given in Table 2 are rather exaggerated, as TLC of these fractions showed the presence of significant amounts of TAG. The contents of polar lipids, glycolipids and phospholipids in the test oils is quite low, again suggesting that these melon seed oils would only require simple refinement procedures. Adsorption column separation of the acylglycerols gave the usual pattern of composition, i.e., the triacylglycerols constituted well over 90% of each sample, as shown in Table 2.

The fatty acid compositions of the melon seed oils are given in Table 3, which shows the principal fatty acid components in the melon seed oils to be linoleic (18:2n-6), oleic (18:1n-9), palmitic (16:0) and stearic (18:0) acids. From Table 3, the close resemblance between the melon seed oils and the oils from soybean, sunflower and groundnut is very apparent. Stearic acid composition in the melon seed oils ranges from 7.81 to 11.41%, which is higher than it is in the commercial oils. However, palmitic acid content in the melon seed oils (9.91–20.80%) is comparable with those in soybean, sunflower and groundnut oils. Thus the level of total saturation (17.72–32.12%) in the *Citrullus* seed oils is only slightly higher than that in the commercial oils (10–30.5), Table 3. However, this level of saturation is much lower than the saturation levels in palm nut oil (35–55%) [11]. The unsaturation profiles of the *Citrullus* seed oils are remarkably similar to those of soybean and sunflower oils, where both sets of seed oils have linoleic acid as the most abundant fatty acid, followed by oleic acid. In general, polyunsaturation in soybean oil is higher than it is in the melon seed oils because of its significant content of α-linolenic acid, 18:3n-3 (5.5–10%). The only melon seed oil with a trace of α-linolenic acid observed in this study was wrewre (0.11%), which contains α-linolenic acid at levels similar to those in sunflower and groundnut oils. Groundnut oil, however, differs from all of the other seed oils in Table 3 because it has oleic acid as the predominant fatty acid.

The fatty acid profiles of the *Citrullus* seed oils as determined by capillary GC, Table 3, are in close agreement with the percent compositions of the fatty acid classes estimated from proton NMR analysis of the oils [7]. Both GC and proton NMR techniques give linoleic acid as the dominant fatty acid in the melon seed oils, and both techniques also give similar ratios of total unsaturation to total saturation in all of the melon seeds.

The major results from this study, as shown in Tables 1 and 3, demonstrate the close similarities between the melon seed oils and the oils from soybean, sunflower and groundnut. The test melon seeds are all edible and they all have reasonably high oil contents (average 27% wt/wt) with high unsaturation (~70%) and good physicochemical properties that make them attractive candidates for commercial exploitation. The potential value of these melon seeds, especially sesoswane, agusi and wrewre seeds, in terms of their multiple use in oil and food products, is comparable to those of soybean, sunflower and groundnut. Development of these melon seed oils into commercial products will add tremendous value to these melon crops, and will expand the number of sources of vegetable oil available, at least in the growing regions of the world.

Table 3 Fatty acid compositions (%, w/w) of *Citrullus* seed oils (obtained from capillary GC and proton NMR integrals) compared with the fatty acid compositions of soybean, sunflower and groundnut oils

Fatty acid class	Sesoswane	Agusi	Wrewre	Tsama melon	Desert melon	Soybean[a]	Sunflower[a]	Groundnut[a]
16:0	20.80 ± 0.09	14.41 ± 0.01	15.43 ± 0.02	9.91 ± 0.07	10.10 ± 0.11	7–14	3–10	6–16
16:1	ND	ND	ND	ND	ND	0.5	<1.0	<1.0
18:0	11.32 ± 0.03	11.41 ± 0.01	8.90 ± 0.02	7.81 ± 0.04	9.22 ± 0.02	3–5.5	1–10	1.3–6.5
18:1	12.72 ± 0.02	12.24 ± 0.02	14.22 ± 0.03	15.51 ± 0.03	18.20 ± 0.04	18–26	14–35	35–72
18:2n-6	55.21 ± 0.03	62.00 ± 0.06	61.50 ± 0.02	66.85 ± 0.08	62.46 ± 0.06	50–57	55–75	13–45
18:3n-3	–	–	0.11 ± 0.02	–	–	5.5–10	<0.3	<0.3
20:0	–	–	–	–	–	<0.6	<1.5	1–3
22:0	–	–	–	–	–	<0.5	<1.0	1–5
Total saturated	32.12	25.82	24.33	17.72	19.32	10–20.6	4–22.5	9.3–30.5
Total unsaturated	67.93	74.24	75.72	82.36	80.66	73.5–93.5	69– >95	48– >95
Fatty acid classes	**Estimated from proton NMR integrals (% mol)[b]**							
α-Linolenic	–	–	–	–	–	–	–	–
Diunsaturated	51.4	65.5	51.8	60.7	–	–	–	–
Monounsaturated	17.3	14.3	21.8	15.9	–	–	–	–
Saturated	31.3	20.2	26.4	23.4	–	–	–	–
Average carbon number	17.0	18.0	17.0	18.0	–	–	–	–

Capillary GC values represent the average of three replicate analyses ±SD

[a] From [9]

[b] Compositions of FA classes were estimated from one proton NMR experiment for each seed oil

References

1. Wickens GE, Goodin JR, Field DV (1984) Plants for arid lands. George Allen and Unwin, London, pp 231–241
2. Holland B, Welch AA, Unwin ID, Buss DH, Paul AA (1995) Southgate DAT, McCance and Widdowson's—the composition of foods, 5th edn. The Royal Society of Chemistry and Ministry of Agriculture, Fisheries and Food, UK, pp 223–321
3. Yaniv Z, Shabelsky E, Schafferman D (1999) Colocynth: potential arid land oilseed from an ancient cucurbit. In: Janick J (eds) Perspectives on new crops and new uses. ASHS, Alexandria, VA, pp 257–261
4. Akoh CC, Nwosu CV (1992) Fatty acid composition of melon seed oil lipids and phospholipids. J Am Oil Chem Soc 69:314–316
5. Dieffenbacher A, Pocklington WD (1987) Standard methods for the analysis of oils, fats, and derivatives. International Union of Pure and Applied Chemistry/Blackwell, Oxford
6. Sempore BG, Berzard A (1996) Analysis of neutral lipids: fatty acids. In: Nollet LML (ed) Handbook of food analysis, vol. 1. Marcel Dekker, New York, pp 331–394
7. Yeboah SO, Holmback J, Ketshajwang KK (1998) Quality and compositional studies of some edible leguminosae seed oils in Botswana. J Am Oil Chem Soc 75(6):741–743
8. Bockisch M (1998) Fats and oils handbook. AOCS, Champaign, IL, pp 174–326
9. Kirk SR, Sawyer R (1991) Pearson's composition and analysis of foods, 9th edn. Longman, UK, pp 617–620
10. List GR, Evans CD, Kwolek WF, Warner K, Boundy BK, Cowan JC (1974) Oxidation and quality of soybean oil: a preliminary study of the anisidine test. J Am Oil Chem Soc 51:17–21
11. Bockisch M (1998) Fats and oils handbook. AOCS, Champaign, IL, pp 208–213