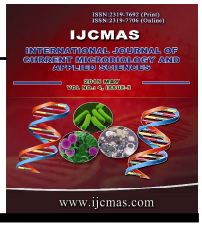
Int.J.Curr.Microbiol.App.Sci (2015) 4(5): 537-554

International Journal of Current Microbiology and Applied Sciences
ISSN: 2319-7706 **Volume 4 Number 5 (2015) pp. 537-554**
http://www.ijcmas.com



Original Research Article

Citrus Seeds Oils as Sources of Quality Edible Oils

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A B S T R A C T

Keywords

Nigeria, citrus seeds, lipids composition

The study is aimed to isolate and analyse the lipids of citrus seeds. The fat (%), total fatty acid (%) and energy (kJ/100 g) had range values of 21.6-26.2, 17.3-20.9 and 639-775 respectively. Saturated fatty acids ranged from 33.3 -36.7 %. Total unsaturated acids values ranged from 63.4-67.0 %. Phospholipids level was high at total value of 491-590 mg/100 g. Among the phytosterols, sitosterol was highly concentrated (283-381 mg/100 g) within a total of 360-485 mg/100 g.

Introduction

Fruits are natural staple food of man containing essential nutrients in adequate proportion. Fruits are excellent sources of minerals, vitamins and enzymes. They are easily digested and bring about a cleansing effect on the blood and the digestive tract. Hence, the ailments usually caused by the consumption of unnatural foods can easily be treated with fruits. Apart from being a very good source of food, fruits are also good medicine (Raiyemo, 2014). Citrus is one of the most important fruit crops grown all over the world. Citrus are rich in vitamin C (ascorbic acid) and folic acid, as well as a good source of fiber. They are fat free, sodium free and cholesterol free. In addition they contain K, Ca, foliate, thiamin, niacin, vitamin B_6 (pyridoxine), P, Mg and Cu. Citrus species are grown for the juice of their fruits. The commonly grown citrus species belong to the family Rutaceae.

The most important citrus species grown in Nigeria are shown in table 1 (Raiyemo, 2014).

In Nigeria, about 930,000 tons of citrus fruits are produced annually from an estimated hectarage of 3 million hectares of land (FAO, 2008). Citrus is grown in the rainforest and guinea savannah, most of these farmlands is in the remote part of the country with poor roads. About 30-50 % of these citrus fruits get spoilt on the way before getting to the final consumers in the urban centres.

Most citrus production is accounted for by oranges, but significant quantities of grape fruits, lemons and limes are also grown. Current annual worldwide citrus production is estimated at over 105 million tons, with

more than half of these being oranges (FAO, 2008).

Citrus fruits are mainly used for dessert, juice and jam production. The food and agro-food processing industry yields considerable amount of waste or by-products such as peels, seeds and pulps which represents 50 % of the raw processed fruit (Vasudeva and Sharma, 2012).

Citrus seeds have long been recognized as a source of edible oil with characteristics suitable for human consumption and food use. At the turn of the last century, German researchers identified palmitic, stearic, oleic, linolenic acids as constituents of citrus and glyerides (Diedrichs, 1914). The same researchers also determined many of the physical and chemical characteristics of the seed oils from sour oranges and lemons (Peters and Frerichs, 1902). The earliest references described the oil as having an intense bitter taste due to the presence of limonin (Peters and Frerichs, 1902). Other early research describes the appearance and proximate analyses of four varieties of orange and grape fruit seed oils (Anonymous, 1920). The chemical and physical characteristics of grapefruit seed oil have been reported (Dunn et al., 1948, Hendrickson and Kesterson, 1961).

Analyses have been performed on orange seeds and seed oils to determine component fatty acids of the glycerides (Dhingra et al., 1957, French, 1962) and other physicochemical characteristics and processing methods (Hendrickson and Kesterson, 1951, Hendrickson and Kesterson, 1965). Mandarin orange seed oils have been separately characterized (Hendrickson and Kesterson, 1964), as have seed oils from tangerines (Swift, 1949), Sri Lanka sweet oranges (Weerakoon, 1960), bitter oranges (Nomura, 1950), and several types of Indian citrus, including shaddock and calamondins (Agrawal et al., 1959). Descriptions of lemon seed oils have been published (Franguelli amd Mariani, 1959) and lime seed oil has been described by Oilar as light-coloured oil, desirable as a salad oil (Oilar, 1954).

Other more recent works on citrus seeds oils were the determination of chemical composition and antimicrobial activity of essential oil of *Citrus limettioides* Tanaka commonly known as sweet lime (Vasudeva and Sharma, 2012); determination of physicochemical characteristics of citrus seeds from Kerman, Iran (Reazai et al., 2014) and the preliminary studies on the characterization of orange seed and pawpaw seed oils (Okoye et al., 2011). There is paucity of information on the study of lipids composition in Nigeria citrus seeds where they are generally discarded as waste. Even in the literature from the study of chemical composition of citrus seeds from international sources, the information on the phytosterols and phospholipids evaluation is very scanty. Therefore, in order to make more efficient use of citrus seeds, it is worth investigating the lipids composition (crude fat, fatty acids, phospholipids, sterols) of citrus seed in Nigeria particularly seeds of grape, sweet orange and tangerine fruits. Such information may extend the zone of available dietary oil source.

Experimental

Collection and treatment of samples: The various citrus seeds were collected from homes within the Ekiti State University, in 2011, weighed, sun dried, deshelled and ground into fine flour.

Oil extraction: The fine flour of the three different seeds was subjected individually to solvent extraction for 5 h with petroleum spirit between 40–60^{o}C boiling ranges using Soxhlet extraction method (AOAC, 2005).

Production and analyses of FAME: The crude extraction was converted to the methyl ester using the boron trifluoride method (AOAC, 2005). The gas chromatographic conditions for the analyses of FAME (fatty acid methyl esters) were as follows: The GC was the HP 5890 powered with HP ChemStation rev A09.01 [1206] software [GMI, Inc, Minnesota, USA] fitted with a flame ionization detector (FID).

A split injection with split ratio of 20:1 was used. GC inlet temperature was 250^{o}C with an oven programme of initial temperature at 60^{o}C, first ramping at 10^{o}C/min for 20 min (maintained for 4 min), second ramping at 15 ^{o}C/min for 4 min (maintained for 10 min) and detector temperature at 320^{o}C. A capillary column (30 m x 0.25 mm) packed with a polar compound (HP INNOWAX) with a diameter (0.25 µm) was used to separate the esters. The peaks were identified by comparison with standard fatty acid methyl esters. Carrier gas used was nitrogen.

Sterol analyses: The sterol analysis was as described by AOAC (2005). The aliquots of the processed fat were added to the screw-capped test tubes. The samples were saponified at 95^{o}C for 30 min, using 3 ml of 10 % KOH in ethanol, to which 0.20 ml of benzene had been added to ensure miscibility.

Deionised water (3 ml) was added and 2 ml of hexane was added in extracting the non-saponifiable materials. The extractions, each with 2 ml of hexane, were carried out for 1 h, 30 min and 30 min respectively. The hexane was concentrated to 1 ml in the vial for gas chromatography analysis and 1 µl was injected into the injection pot of GC. The GC conditions of analyses were similar to the GC conditions for methyl esters analyses. The peaks were identified by comparison with standard sterols.

Phospholipids analyses: The method of Raheja et al. (1973) was adopted in the analyses of the phospholipids content determination. The GC conditions for analyses of phospholipids were similar to FAME analyses except in the following: Column type was HP5, oven programme initial temperature at 50 ^{o}C, second ramping at 15 ^{o} C/min for 4 min, maintained for 5 min and the detector was pulse flame photometric detector (PFPD).

Quality assurance: For the purpose of ensuring the accuracy of the results obtained, the followings were prepared for sterols, phospholipids and fatty acid methyl esters which were then compared with respective analytical results; calibration curves were prepared for all the standard mixtures and correlation coefficient determined for fatty acids, sterols and phospholipids. Correlation is a statistical index that shows the quality assurance of the calibration curve performed. It was prepared with the Hewlett Packard Chemistry (HPCHEM) software (GMI, Inc 6511 Bunker Lake Blvd Ramsey, Minnesota, 55303, USA).

Calculation of fatty acids per 100 g in sample as food: At the data source and reference database levels, values for individual fatty acids (FAs) are usually expressed as percentages of total FAs. At the user database levels, values per 100 g of food are required.

A conversion factor derived from the proportion of the total lipid present as FAs is required for converting percentages of total FAs to FAs per 100 g of food. Total lipid level was multiplied by a conversion factor of 0.80 to convert it to total fatty acids (Anderson, 1976). (0.80 is a conversion factor to convert total lipid to total fatty acids.) For fatty acids, precision is best limited to 0.1 g/100 g of fatty acids

(Greenfield and Southgate, 2003), with trace being set at < 0.06 g/100 g total fatty acids.

Statistical analyses: Statistical analyses were carried out to determine mean, standard deviation, coefficient of variation in per cent. Further statistical analysis was carried out using the Chi-square (X^2) method as appropriate, the α value for the X^2 was 0.05.

Results and Discussion

In Table 2 are shown the crude fat levels of the samples (dry weight). The range was 21.6–26.2 % with grape seed oil being the lowest (21.6 %) and tangerine being the highest (26.2 %). The variation was low between the three samples with the coefficient of variation (CV %) being 9.53. The Chi-square (X^2) values showed no significant differences. The calculated total fatty acids (crude fat x 0.80) and calculated energy levels followed the trend as in the crude fat results in CV % but whilst total fatty acid was not significantly different, the calculated energy was significantly different at $\alpha_{=0.05}$. The significant difference would have been due to the highest value of energy contribution due to tangerine seeds oil (775 kJ/100 g or 36.2 %).

The present crude fat results in Table 2 were better than the values in the lipid composition of the seeds of three types of chillies consumed in Nigeria whose values ranged as 10.8-12.1 % (Adeyeye et al., 2013), making about one half of the present result; they are also much better than the values in sorghum (1.83 %) and in other cereals: millet (1.10 %), maize (1.72 %) and rice (0.63 %) (Adeyeye and Ajewole, 1992). The crude fat levels were reasonable enough and so the seeds of the citrus fruits could be said to be average sources of dietary fat. The total fatty acid (TFA) profiles showed that grape seeds had lowest level of TFA (17.3 %) and highest in tangerine with a value of 20.9 % dry weight. The citrus seeds from Iran (Reazai et al., 2014) had the following oil content: Qaleh Ganj [lemon (41.5 %), citrus (34.1 %)], Jiroft [lemon (41.9 %), citrus (37.2 %)], Anbarabad [lemon (40.3 %), citrus (33.4 %)].

In Table 3, fatty acid profiles of the samples in % total fatty acids are shown. The following fatty acids (FAs) recorded 0.00 % value: C6:0-C14:0, C14:1 (*cis*-9) and C18:1 (*trans*-11) whereas C2:0-C5:0 recorded not detected in all the three samples. The highest saturated fatty acid (SFA) came from C16:0 having a range of 28.5-32.0 % with variation (CV %) of 5.85 whereas total SFA range was 33.3-36.7 % and CV % of 5.10. This meant that the SFA values were generally close as shown by the low CV %. The monounsaturated fatty acids *cis* (MUFA *cis*) levels were close to the values of the SFA with values of 21.6-29.4 and CV % of 18.0 showing the values to be less homogenous than in the SFA. The MUFA (*trans*) levels were of insignificant values with 0.017-0.034 % and CV % of 32.5.

The C18:2 (*cis*-9, 12) had significant high levels in all the samples having a range of 32.5 -40.4 % and low CV % of 10.8. The SFA and C18:2 (*cis*-9, 12) values are close but with reversed concentration thus: in SFA it is grape (36.7 %) > tangerine (36.1 %) > sweet orange (33.3 %) whereas in C18:2 (*cis*-9, 12) it is tangerine (40.4 %) > grape (36.8 %) > sweet orange (32.5 %). Other PUFA members having values greater than 1 % were C18:3 (*cis*-6, 9, 12) and C18:3 (*cis*-9, 12, 15) with respective range values of 1.39-1.91 % (CV % = 16.6) and 1.56-2.13 % (CV % = 17.0). All other PUFA values were very low in all the samples with none contribution up to 1.00 % to the total PUFA; actually total PUFA ranged as 37.3-45.3 % and CV % of 9.68.

The citrus seed oils all have virtually similar compositions. Distinguishing features are the generally higher palmitic acid (C16:0) content of the grape fruit, higher oleic acid (C18:1 *cis*-9) in orange fruit and higher linoleic acid (C18:2 *cis*-9. 12) in tangerine seed. The properties and characteristics of the crude and refined oil are similar to cottonseed oil, except that the citrus seed oil may be more subject to oxidative rancidity because of the presence of linolenic acid in the glycerides; this fatty acid is present only in traces in cottonseed oil (Braddock and Kesterson, 1973). Our present results can favourably compare with some literature information from range of fatty acid composition in Florida citrus and other seed oils. Orange (Hendrickson and Kesterson, 1963, Nordby and Nagy, 1969): palmitic (26-31 %), palmitoleic (0.1 %), stearic (3-5 %), oleic (24-28 %), linoleic (35-37 %) and linolenic (2-4 %); Grapefruit (Hendrickson and Kesterson, 1961, Teles et al., 1972): palmitic (26-36 %), palmitoleic (0.1-0.3 %), stearic (1-4 %), oleic (18-25 %), linoleic (32-40 %) and linolenic (3-6 %); Mandarins (Hendrickson and Kesterson, 1964): palmitic (22-30 %), palmitoleic (0.1-1.0 %), stearic (2-5 %), oleic (20-25 %), linoleic (37-45 %), linolenic (3-5 %); Lemons (Nordby and Nagy, 1969, Hendrickson and Kesterson, 1961, Hendrickson and Kesterson, 1963): palmitic (20-24 %), palmitoleic (0.1-0.3 %), stearic (2-4 %), oleic (26-31 %), linoleic (31-38 %) and linolenic (8-12 %); Limes (Dunn et al., 1948, Nordby and Nagy, 1969, Teles et al., 1972): palmitic (24-29 %), palmitoleic (0.1-0.5 %), stearic (3-5 %), oleic (20-22 %), linoleic (37-40 %), linolenic (6-11 %); Cottonseed (Barroso et al., 1972): palmitic (20-23 %), palmitoleic (-), stearic (1.3 %), oleic (23-35 %), linoleic (42-54 %), linolenic (-); Soybean (Bailey's Industrial Oil and Fat Products, 1964): palmitic (7-11 %), palmitoleic (-), stearic (2-6 %), oleic (15-33 %), linoleic (43-56 %), linolenic (5-11 %). The citrus seeds from Iran (Reazai et al., 2014) had the following fatty acid compositions of the oils extracted from different citrus seed species (%). Qaleh Ganj: palmitic [lemon (29.4), citrus (27.6)], palmitoleic [lemon (0.7), citrus (0.6)], stearic [lemon (4.7), citrus (6.5)], oleic [lemon (26.4), citrus (27.1)], linoleic [lemon (34.1), citrus (34.0)], linolenic [lemon (6.2), citrus (3.2)], other fatty acids [lemon (0.5), citrus (1.0)]; Jiroft: palmitic [lemon (27.8), citrus (27.3)], palmitoleic [lemon (0.9), citrus (0.4)], stearic [lemon (4.1), citrus (4.8)], oleic [lemon (24.8), citrus (29.3)], linoleic [lemon (35.7), citrus (36.3)], linolenic [lemon (7.0), citrus (3.3)], other fatty acids [lemon (0.6), citrus (0.9)]; Anbarabad: palmitic [lemon (23.5), citrus (26.5)], palmitoleic [lemon (0.6), citrus (0.6)], stearic [lemon (4.2), citrus (6.5)], oleic [lemon (28.5, citrus (28.6)], linoleic [lemon (33.7), citrus (32.2)], linolenic [lemon (7.8), citrus 4.1], other fatty acids [lemon (1.4), citrus (1.5)].

Researches that have further been conducted on properties of citrus seed oils are as follows. Anwar et al. (2008) found that linoleic acid is the main acid in citrus seed oil (36.1-39.8 %) and the other key fatty acids were palmitic acid (25.8-32.2 %), oleic acid (21.9-24.1 %), linolenic acid (3.4-4.4 %), and stearic acid (2.8-4.4 %). In a study by Matthaus and Ozcan, the oil content of seeds was reported somewhat between 32.1 g/100 g and 58.8 g/100 g. In descending order, the main fatty acids in the oil samples were oleic (12.8-70.1 %), linoleic (19.5-58.8 %), and palmitic (5.1-28.3 %). Furthermore, content of stearic, vaccenic, linolenic and arachidic acids was negligible (Matthaus and Ozcan, 2012). Mahmud et al. (2009) reported good quality of unsaturated acids found in citrus fruit oil (49.92 %). In our present results, good qualities of unsaturated

acids were in the range of 63.3-66.9 %. Such high content of unsaturated acid is expected in quality edible oil. In another study, Saloua et al. (2009) indicated that linoleic (76.19 %), oleic (13.87 %), stearic (6.76 %) and palmitic (2.40 %) acids are the key fatty acids found in crude oil. Waheed et al. (2009) showed that, in sum, content of lipids C18:3 were 4.66 % in *C. aurantium* and 3.58 % in *C. paradise* (Waheed et al., 2009). Our results are mostly within the range of fatty acid composition when compared to the values in the literature. As the results discussed herein showed, orange, grape and tangerine seeds oil is a rich source of unsaturated fatty acids so that citrus seed oil can be of the best oils for man.

Some calculated parameters from Table 3 are shown in Table 4. These calculated parameters predicted the nutritional qualities of the citrus seeds in their lipid compositions. The X^2 analyses showed that only three parameter: *n*-6/*n*-3, MUFA/SFA and PUFA/SFA had Table (critical) values less than the sample values thereby making them to be significantly different at $\alpha = 0.05$. The original American Heart Association (AHA) Step I fat recommendation was perceptive because it recognized the significance of the fatty acid balance at approximately 1:1:1 for SFA: MUFA: PUFA. Careful review of numerous reports in the literature has revealed the importance of this balance for generating the LDL/HDL ratio. Furthermore, it would appear that the balance is critical at any level of fat intake if one wishes to avoid adversely affecting the lipoprotein profile (Hayes, 2002). The best dietary fat would contain an ideal balance (7:1) of *n*-6 linoleic to *n*-3 linolenic acids. This balance is not available in partially hydrogenated margarines, in which most of the *n*-3 linolenic acid has been destroyed by processing, and is also unlike most vegetable oils that contain only a small amount of this important fatty acid (Hayes, 2002). Our LA/ALA results showed a range of 17.6-26.5 which highly deviated from 7:1 as recommended above; the reason had been due to the high level of LA (37.2-45.2 %) and very low level of ALA (0.085-0.135 %). The ratio of PUFA/SFA (P/S ratio) is important in determining the detrimental effects of dietary fats. The higher the P/S ratio the more nutritionally useful is the oil. This is because the severity of atherosclerosis is closely associated with the proportion of the total energy supplied by SFA and PUFA fats (Honatra, 1974). These proportions are positive towards PUFA far more than SFA. On the overall *n*-6/*n*-3 ratios, the value of 334-485 showed that the PUFA composition was skewed towards *n*-6 much more than the *n*-3. Whilst it would be easy for the body to synthesis AA [C20:4 (*n*-6)], it may be difficult to synthesise the *n*-3 PUFA series especially eicosapentaenoic acid [C20:5 (*n*-3) or EPA] and so the seed might need enhancement in this PUFA. The MUFA/SFA levels in the samples ranged from 0.600-0.883 which was less than in the P/S levels. The relative proportion of MUFA/SFA is an important aspect of phospholipids compositions and changes to this ratio have been claimed to have effects on such disease states as cardiovascular disease, obesity, diabetes, cancer and neurophatological conditions.

For example, MUFA/SFA have been shown to have cytoprotective actions in pancreatic β-cells. *Cis*-Monoenoic acids have desirable physical properties for membrane lipids in that they are liquid at body temperature, yet are relatively resistant to oxidation. They are now recognized by nutritionists as being beneficial in the human diet. The essential PUFA status index (EPSI), (ratio of sum of all *n*-3 and *n*-6 FAs and the sum of all *n*-7 and *n*-9 FAs) is an indicator of essential PUFA status. The higher the EPSI the better

the essential PUFA status. The EPSI values in the samples ranged from 1.27-2.09. A high ratio between AA and DGLA is an indicator of Δ-5 desaturase activity which can be related to good insulin sensitivity; values here ranged from low to high (0.150-1.95). The synthesis of mead acid is promoted if there are insufficient concentrations of LA and ALA to meet the need for the synthesis of long-chain PUFA. EPA and DHA inhibit mead acid synthesis; the presence of mead acid indicates a general shortage of all essential PUFA. The present results had ratios of 1.0 -1.0 for EPA/DHA and no mead acid was produced.

In Table 5, the fatty acids calculated as food lipids sources are shown. Categories of fatty acids of significance in lipid food composition were: C16:0 (5.53-6.26 g/100 g); total SFA (6.34-7.55); MUFA (*cis*) (3.84-5.76 g); C18:2 (*cis*-9, 12) (6.36-8.46 g/100 g) and total PUFA (7.09-9.48 g/100 g).

This type of information is required to be able to calculate the energy contribution by each type of fatty acid. As expected, the concentration of the fatty acids as food went as (g/100 g): SFA (6.34-7.55 g/100 g) > MUFA (*cis*) (3.84-5.76 g/100 g) < PUFA (7.09-9.48 g/100 g).

The energy contribution of the fatty acids in the citrus seed samples is shown in Table 6. The energy contributions were as varied as the fatty acids distribution. The contributions were total SFA (235-279 kJ/100 g or 36.8-36.0 %); MUFA (*cis*) (142-213 kJ/100 g or 22.2- 29.3 %); MUFA (*trans*) (0.110-0.245 g/100 g or 0.017-0.034 %); C18:2 (*cis*-9, 12) (235-313 kJ/100 g or 36.8-40.4 %); C18:3 (*cis*-9, 12, 15) (9.97-16.5 g/100 g or 1.56-2.13 %); PUFA (total) (262-351 kJ/100 g or 41.0-45.3 %) whereas contributions in PUFA due to LA was 87.1-89.7 % and in ALA it was 3.81-5.72 %. Some notable energy contributions were (kJ/100 g): C16:0 (205-232 or 32.1-29.9 %); C18:0 (29.3-46.8 or 4.59-6.04 %). On the other hand, the percentage contribution of energy to total energy in SFA due to C16:0 was 83.2-87.2 % and C18:0 was 12.5 -16.8 %; in MUFA (*cis*), energy contribution due to C18:1 (*cis*-6) was 36.8-53.1 % and C18:1 (*cis*-9) was 44.6 -59.5 %; in PUFA, energy contribution due to C18:2 (*cis*-9, 12) was 87.1-89.7 % and C18:3 (*cis* -9, 12, 15) was 3.81-5.72 % . In the samples total energy intake as contributed from SFA was greater than 10 % E but the recommended range ADMR (acceptable macronutrient distribution range) for PUFA is 6.11 % E (WHO, 2008) was less than our values; our results could lead to the replacement of SFA with PUFA (*n*-3 and *n*-6) in the diet.

From Table 4, the SFA: MUFA: PUFA in the samples were: grape was 1:1.65:0.893, orange was 1:1.33:0.893 and tangerine was 1:1.48:0.799. The original AHA Step 1 fat recommendation (≤ 30 % fat, 8-10 % SFA and < 300 mg of cholesterol per day) (Kris-Etherton, 1999) was perceptive because it recognized the significance for SFA: MUFA: PUFA. Careful review of numerous reports in the literature has revealed the importance of this balance for generating the best LDL/HDL ratio. Furthermore, it would appear that the balance is critical at any level of fat intake if one wishes to avoid adversely affecting the lipoprotein profile (Hayes, 2002); this appears to be the situation with the present results. From literature, this type of scenario was seen in the following samples for SFA: MUFA: PUFA in red bell pepper (1:1.23:0.20), scotch bonnet pepper (1:1.80:0.28) and chili pepper (1:1.55:024) (Adeyeye et al., 2013).

The concentrations of C18:1 (*cis*-6) and C18:1 (*cis*-9) need further discussion. From Table 3, the respective fatty acid levels

were: C18:1 (*cis*-6) (7.99-15.5 %) and C18:1 (*cis*-9) (10.3-13.1 %); in Table 5, their contributions as food source were: C18:1 (*cis*-6) (1.67-3.04 g/100 g) and C18:1 (*cis*-9) (1.78-2.70 g/100 g); in Table 6, their energy contributions were: C18:1 (*cis*-6) (61.9-113 kJ/100 g or 7.99-15.6 %) and C18:1 (*cis*-9) (65.9-99.9 kJ/100 g or 10.3-12.9 %) both corresponding values being very close to each other. Oleic acid [9c-18:1 or 18:1 (*n*-9)] is by far the most abundant monoenoic fatty acid in plant and animal tissues, both in structural lipids and in depot fats. It is the highest concentrated MUFA only in tangerine being slightly higher than C18:1 (*cis* -6) by 38.1 %. Olive oil contains up to 78 % oleic acid, and it is believed to have especially valuable nutritional properties as part of the Mediterranian diet. It has a number of important biological properties, both in the free and esterified form. Oleic acid is the biosynthetic precursor of a family of fatty acid with the (*n*-9) terminal structure and with chain-lengths of 20-24 or more. Petroselinic acid (6c-18:1) occurs up to a level of 50 % or more in seed oils of Umbelliferae family, including carrot, parsely and coriander. In the present report, petroselinic acid occupied the highest position in the *cis*-18:1 FA in both grape (11.7 %) and orange (15.5 %) but second highest in tangerine (7.99 %).

This pattern was also observed in three types of chillies consumed in Nigeria where *cis*-18:1 occupied the highest position in both scotch bonnet (5.35 %) and chili (5.53 %) but second highest in red bell (5.88 %) (Adeyeye et al., 2013). Studies in vitro by Weber et al. (1995) revealed that triacylglycerols containing petroselinoyl [18:1(*n*-12)] moieties are hydrolysed by pancreatic lipase at much lower rates than other triacylglycerols. Consumption of coriander (*Coriandrum sativum*) oil compared with the other oils led to significant greater weights. No significant differences were observed among the groups fed various levels of oleic acid in body weight, the weights of heart, liver, kidneys, spleen or testes, lipid content of heart or total cholesterol, HDL cholesterol and triacylglycerol concentrations of blood plasma. Ingestion of coriander oil led to incorporation of 18:1 (*n*-12) into heart, liver and blood lipids and to a significant reduction in the concentration of arachidonic acid in the lipids of hearts, liver and blood with a concomitant increase in the concentration of linoleic acid compared with results for the other groups. The data showed that petroselinic acid from dietary triacyglycerols is absorbed by rats as readily as oleic acid but the former reduces the concentration of arachidonic acid in tissue lipids suggesting (in view of earlier studies (Mohrhauer et al., 1967)) petroselinic acid-mediated inhibitor of arachidonic acid synthesis.

Table 7 shows the phospholipids levels in the samples. The samples were low to high in the concentration of the various phospholipids. On the low side were phosphatidylserine with 15.3-26.4 mg/100 g and CV % of 28 % and lysophosphatidylcholine with 3.69-7.02 mg/100 g having highest CV % of 31.4; the medium range concentration was observed in phosphatidylethanolamine having a range of 86.3-101 mg/100 g having lowest CV % of 8.88; the high range group were phosphatidylinositol (171-216 mg/100 g, CV % of 17.9). Significant differences were observed in the phosphatidylcholine, phosphatidylinositol and the total groups with sweet orange (295 mg/100 g) being responsible for the significant difference in phosphatidylcholine, tangerine (216 mg/100 g) being responsible for the significant difference in phosphatidylinositol and sweet orange (590 mg/100 g) again being responsible for the significant difference in the total group. Other parameters were not

significantly different among themselves. The various concentration values were generally close with CV % ranging between 8.88 and 31.4. The results of phospholipids in the citrus seed oils were very different from our earlier observation in three types of chillies seeds consumed in Nigeria where total phospholipids range was 1.98 e-4 to 5.37 e-4 mg/100 g (Adeyeye et al., 2013).

The phospholipid fraction of the seed lipids consists primarily of phsphatidylcholine (lecithin) and phosphatidylinositol. These two phospholipids comprise approximately 77.7 to 80.0 % of the total phospholipids; the remainder being phosphatidylethanolamine (14.8-17.7 %), phosphatidylserine (3.12-4.47 %) and lysophosphatidylcholine (0.63-1.23 %). These results were different from those obtained from Florida citrus where lecithin and phosphatidylethanolamine (cephalin) comprise approximately 85 to 90 % of the total phospholipids, the remainder being phosphatidylinositol (10 to 15 %) and phosphatidylserine (1 to 5 %) (Braddock and Kesterson, 1973); these ranges represent analyses performed for one season only. Lecithin is usually the most abundant phospholipid in animals and plants, often amounting to almost 50 % of the total, and as such it is the key building block of membrane bilayers. This observation is true for lecithin in the samples having concentration range of 39.8 % to 50.0 %. This is within the range of dika nut [128 mg/100 g, 46.4 %] and much better than in conophor nut [302 mg/100 g, 22.1 %] (Adeyeye, 2012). Phosphatidylcholine (PC) is a class of phospholipids that incorporate choline as a headgroup. They are a major component of biological membranes. They are also a member of the lecithin group of yellow-brownish fatty substances occurring in animal and plant tissues. Cephalin is found in all living cells, although in human physiology it is found particularly in nerves, such as the white matter of brain, nerves, neural tissue and in spinal cord.

The cholesterol level was in trace in each sample as shown in Table 8 (2.19 e-3 to 6.63 e-3 mg/100 g). Other animal sterols present at trace levels in the samples were cholestanol (2.24 e-3 to 0.021 mg/100 g) and ergosterol (0.023-0.045 mg/100 g). Three sterols were moderately concentrated in the samples: campesterol (54.4-68.1 mg/100 g and CV % of 11.4), stigmasterol (12.7-17.0 mg/100 g and CV% of 2.48) and 5-avenasterol (9.93-18.5 mg/100 g and CV % of 30.4). The plant sterol of major concentration in the samples was sitosterol with values range of 283-381 mg/100 g and CV % of 15.0. Only the sitosterol and the total phytosterol were significantly different among themselves; with sweet orange being responsible for the significant difference in the two groups. For the phytosterols with average concentrations, the following percentages were contributed: campesterol (14.0-15.1 %; dominated by orange contribution), stigmasterol (3.51-4.13 %; dominated by tangerine), 5-avenasterol (2.76-3.81 %, dominated by orange) and sitosterol (77.9-78.6, dominated by grape). Our present values in campesterol, stigmasterol and sitosterol were lower than the values in chillies with corresponding levels of 87.7-96.2 mg/100 g or 13.7-15.2 %, 162-164 mg/100 g or 15.3-25.4 % and 341-389 mg/100 g or 57.1-61.0 % (Adeyeye et al., 2013).

The cholesterol-lowering effect of dietary plant sterols (phytosterols) has been studied since the 1950s and is well known (Lees et al., 1977). Grundy and Mok (1976) demonstrated that 3 g/d of sitosterol was sufficient to lower serum cholesterol levels. They suggested that plant sterol could be considered a form of dietary treatment rather

than a drug to lower cholesterol because plant sterols are naturally present in plant based foods. Plant sterols interfere with the uptake of both dietary and biliary cholesterol from the intestinal tract in humans (Heinemann et al., 1991). The reason for this is not fully understood; however, plant serols appear to decrease the solubility of cholesterol in the oil and micellar phases, thus displacing cholesterol from bile salt micelles and interfering with its absorption (Ikeda and Sugano, 1998).

The present study showed that the phytosterols in the citrus seed samples compared very favourably with most literature values such as: campesterol (mg/kg): corn oil (2691), cottonseed (170), olive (28), palm (358), rapeseed (1530), safflower (452), soybean (720) and sunflower (313); stigmasterol (mg/kg): corn oil (702), cottonseed (42), olive (14), palm (204), rapeseed (-), safflower (313), soybean (720), sunflower (313); β-sitosterol (mg/kg): corn oil (7722), cottonseed (3691), olive (1310), palm (1894), rapeseed (3594), safflower (1809), soybean (1908) and sunflower (2353) (Gunstone et al., 1994). Our results, using similar unit, we had campesterol (544-681 mg/kg), stigmasterol (127-170 mg/kg) and β-sitosterol (2830-3810 mg/kg).

Utilization of citrus seed oils has been evaluated (Hendrickson and Kesterson, 1961), and some unusual applications have been proposed. Kaplan (1941) patented the use of grape fruit seed oil as a lubricant and preservative to treat textile fibers such as silk, wool and rayon; Stambovsky (1942) discusses utilization of citrus seed oil as a food, in paints, and for soaps. Our results will find accommodation in Stambovsky discussion.

Table.1 Important citrus species grown in Nigeria

Common name	Botanical names	Local names in Nigeria
Sweet orange	*Citrus sinensis*	Osan mimu, Orombo didun
Grape fruit	*Citrus paradise*	Osan gerepu, Osan paya
Lime	*Citrus aurantifolia*	Osan-wewe, Afotanta, Epe nkirisi
Lemon	*Citrus limon*	Osan-laimu, Oroma-nkiri, Babban leemul
Tangerine	*Citrus reticulate*	
Sour orange	*Citrus aurantium*	Osan, Orombo-igun, Olomaoyibo, gangan

Table.2 Crude fat and total fatty acids (%) composition of grape (O1), orange (O2) and tangerine(O3) seeds

Parameters	O1	O2	O3	Mean	SD	CV %	X^2	TV	Remark
Crude fat	21.6	24.5	26.2	24.1	2.30	9.53	0.438	5.99	NS
Total fatty acid*	17.3	19.6	20.9	19.3	1.85	9.61	0.356	5.99	NS
Energy kJ/100g	639	726	775	713	68.6	9.61	13.2	5.99	S

*Crude fat x 0.80

Table.3 Fatty acids composition of grape (O1), orange (O2) and tangerine (O3) seed (%)

Fatty acids	O1	O2	O3	Mean	SD	CV %
C2:0	-	-	-	-	-	-
C3:0	-	-	-	-	-	-
C4:0	-	-	-	-	-	-
C5:0	-	-	-	-	-	-
C6:0	0.00	0.00	0.00	0.00	0.00	0.00
C8:0	0.00	0.00	0.00	0.00	0.00	0.00
C10:0	0.00	0.00	0.00	0.00	0.00	0.00
C12:0	0.00	0.00	0.00	0.00	0.00	0.00
C14:0	0.00	0.00	0.00	0.00	0.00	0.00
C16:0	0.049	0.170	0.469	0.229	0.216	94.3
C18:0	0.016	0.031	0.023	0.023	0.008	32.2
C20:0	0.044	0.086	0.065	0.065	0.021	32.3
C22:0	0.041	0.080	0.060	0.060	0.020	32.3
C24:0	0.005	0.010	0.007	0.007	0.003	34.3
TOTAL SFA	0.155	0.377	0.624	0.385	0.235	60.9
C14:1(*cis*-9)	32.0	28.5	29.9	30.1	1.76	5.85
C16:1(*cis*-9)	4.59	4.62	6.04	5.08	0.829	16.3
C18:1(*cis*-6)	11.7	15.5	7.99	11.7	3.76	32.0
C18:1(*cis*-9)	10.3	13.1	12.9	12.1	1.56	12.9
C20:1 (*cis*-11)	0.166	0.326	0.246	0.246	0.08	32.5
C22:1(*cis*-13)	0.014	0.027	0.021	0.021	0.007	32.5
C24:1(*cis*-15)	0.005	0.248	0.007	0.087	0.140	161
TOTAL MUFA *cis*	22.2	29.4	21.6	24.4	4.31	18.0
C18:1 (*trans*-6)	0.016	0.031	0.023	0.023	0.008	32.5
C18:1 (*trans*-9)	0.001	0.003	0.002	0.002	0.001	33.3
C18:1 (*trans*-11)	0.00	0.00	0.00	0.00	0.00	0.00
TOTAL MUFA *trans*	0.017	0.034	0.026	0.026	0.008	32.5
MUFA total	22.3	29.4	21.7	24.4	4.31	17.6
C20:2 (*cis*-11,14)	0.006	0.012	0.009	0.009	0.003	32.6
C20:3 (*cis*-11,14,17)	0.027	0.053	0.040	0.040	0.013	32.5
C20:5 (*cis*-5,8,11,14,17)	0.005	0.010	0.007	0.007	0.002	32.4
C22:6 (*cis*-4,7,10,13,16,19)	0.046	0.010	0.079	0.045	0.035	76.7
TOTAL (n-3)	0.085	0.085	0.135	0.101	0.029	28.9
C18:2 (*cis*-9,12)	36.8	32.5	40.4	36.6	3.96	10.8
C18: 2 (*trans*-9,11)	0.019	0.036	0.027	0.027	0.009	32.5
C18:3 (*cis*-6,9,12)	1.39	1.85	1.91	1.72	0.284	16.6
C18:3 (*cis*-9,12, 15)	1.56	2.13	2.13	1.94	0.329	17.0
C20:3 (*cis*-8,11,14)	0.406	0.609	0.587	0.534	0.111	20.9
C20:3 (*cis*-11,14,17)	0.269	0.053	0.040	0.121	0.129	107
C20:4 (*cis*-5,8,11,14)	0.792	0.0916	0.0914	0.325	0.404	124
C22:2 (*cis*-13,16)	0.005	0.010	0.007	0.007	0.002	32.4
TOTAL (n-6)	41.2	37.3	45.2	41.2	3.96	9.60
PUFA Total	41.3	37.4	45.3	41.3	3.98	9.63

Table.4 Calculated parameters from fatty acid composition of seeds of grape (O1), orange (O2) and tangerine (O3)

Parameters	O1	O2	O3	Mean	SD	CV %	X^2	TV	Remark
SFA	0.155	0.377	0.624	0.385	0.235	60.9	0.286	5.99	NS
MUFA *cis*	58.8	62.3	57.1	59.4	2.66	4.5	0.239	5.99	NS
MUFA *trans*	0.017	0.034	0.026	0.026	0.008	32.5	0.005	5.99	NS
MUFA total	58.8	62.4	57.1	59.4	2.67	4.5	0.240	5.99	NS
n-3 PUFA	0.085	0.085	0.135	0.101	0.029	28.9	0.017	5.99	NS
n-6 PUFA	41.2	37.3	45.2	41.2	3.96	9.6	0.759	5.99	NS
Total PUFA	41.3	37.4	45.3	41.3	3.98	9.6	0.767	5.99	NS
DUFA *cis*	36.8	32.5	40.4	36.6	3.95	10.8	0.854	5.99	NS
DUFA *trans*	0.019	0.036	0.027	0.027	0.009	32.5	0.006	5.99	NS
DUFA total	36.8	32.6	40.4	36.6	3.95	10.8	0.851	5.99	NS
TUFA *cis*	3.65	4.69	4.71	4.35	0.606	13.9	0.169	5.99	NS
TUFA *trans*	-	-	-	-	-	-	-	-	-
TUFA total	3.65	4.69	4.71	4.35	0.606	13.9	0.169	5.99	NS
MUFA/SFA	379	165	91.6	212	149	70.5	211	5.99	S
PUFA/SFA	267	99.1	72.6	146	105	72.0	151	5.99	S
n-6/*n*-3	488	441	334	421	78.9	18.7	29.6	5.99	S
EPSI	1.85	1.27	2.09	1.74	0.422	24.3	0.205	5.99	NS
LA/ALA	26.5	17.6	21.2	21.7	4.48	20.6	1.85	5.99	NS
EPA/DHA	1.00	1.00	1.00	1.00	0.00	0.00	0.00	5.99	NS
AA/DGLA	1.95	0.150	0.156	0.752	1.04	138	2.86	5.99	NS

X^2= Chi-Square, TV= table value at α=0.05, n-1=2, S= signifant, NS= not significant

Table.5 Fatty acid composition (%) of the seeds of grape (O1), orange (O2) and tangerine (O3) as food

Fatty acids	O1	O2	O3	Mean	SD	CV %
C2:0	-	-	-	-	-	-
C3:0	-	-	-	-	-	-
C4:0	-	-	-	-	-	-
C5:0	-	-	-	-	-	-
C6:0	0.00	0.00	0.00	0.00	0.00	0.00
C8:0	0.00	0.00	0.00	0.00	0.00	0.00
C10:0	0.00	0.00	0.00	0.00	0.00	0.00
C12:0	0.00	0.00	0.00	0.00	0.00	0.00
C14:0	0.00	0.00	0.00	0.00	0.00	0.00
C16:0	0.008	0.033	0.098	0.047	0.046	99.3
C18:0	0.003	0.006	0.005	0.005	0.002	36.8
C20:0	0.008	0.017	0.014	0.013	0.005	37.0

C22:0	0.007	0.016	0.013	0.012	0.004	37.0
C24:0	0.001	0.002	0.001	0.001	0.001	38.4
TOTAL SFA	0.027	0.074	0.131	0.077	0.052	67.4
C14:1(*cis*-9)	5.53	5.59	6.26	5.79	0.406	7.00
C16:1(*cis*-9)	0.79	0.91	1.26	0.99	0.25	24.9
C18:1(*cis*-6)	2.02	3.04	1.67	2.25	0.71	31.7
C18:1(*cis*-9)	1.78	2.57	2.70	2.35	0.50	21.2
C20:1 (*cis*-11)	0.03	0.06	0.05	0.05	0.02	37.2
C22:1(*cis*-13)	0.00	0.01	0.00	0.00	0.00	37.2
C24:1(*cis*-15)	0.00	0.05	0.00	0.02	0.03	161
TOTAL MUFA *cis*	10.2	12.2	12.0	11.4	1.13	9.8
C18:1 (*trans*-6)	0.0027	0.0061	0.0049	0.0046	0.0017	37.2
C18:1 (*trans*-9)	0.0002	0.0005	0.0004	0.0004	0.0002	38.0
C18:1 (*trans*-11)	0.00	0.00	0.00	0.00	0.00	0.00
TOTAL MUFA *trans*	0.003	0.007	0.005	0.005	0.002	37.3
MUFA total	10.2	12.2	12.0	11.5	1.13	9.85
C20:2 (*cis*-11,14)	0.001	0.002	0.002	0.002	0.001	37.3
C20:3 (*cis*-11,14,17)	0.005	0.010	0.008	0.008	0.003	37.2
C20:5 (*cis*-5,8,11,14,17)	0.001	0.002	0.002	0.001	0.001	37.2
C22:6 (*cis*-4,7,10,13,16,19)	0.008	0.002	0.017	0.009	0.007	83.1
TOTAL (n-3)	0.015	0.017	0.028	0.020	0.007	37.4
C18:2 (*cis*-9,12)	6.36	6.38	8.46	7.07	1.21	17.1
C18: 2 (*trans*-9,11)	0.003	0.007	0.006	0.005	0.002	37.2
C18:3 (*cis*-6,9,12)	0.240	0.363	0.400	0.334	0.084	25.0
C18:3 (*cis*-9,12, 15)	0.270	0.418	0.446	0.378	0.095	25.1
C20:3 (*cis*-8,11,14)	0.070	0.119	0.123	0.104	0.030	28.3
C20:3 (*cis*-11,14,17)	0.046	0.010	0.008	0.022	0.021	98.7
C20:4 (*cis*-5,8,11,14)	0.137	0.018	0.019	0.058	0.068	118
C22:2 (*cis*-13,16)	0.001	0.002	0.002	0.001	0.001	37.2
TOTAL (*n*-6)	7.08	7.30	9.46	7.95	1.31	16.5
PUFA total	7.09	7.32	9.48	7.97	1.32	16.6

Table.6 Energy contributions (kJ/100g) from the various fatty acids composition of seeds of grape (O1), orange (O2) and tangerine (O3)

Fatty acids	O1	O2	O3	Mean	SD	CV %
C2:0	-	-	-	-	-	-
C3:0	-	-	-	-	-	-
C4:0	-	-	-	-	-	-
C5:0	-	-	-	-	-	-
C6:0	0.00	0.00	0.00	0.00	0.00	0.00
C8:0	0.00	0.00	0.00	0.00	0.00	0.00
C10:0	0.00	0.00	0.00	0.00	0.00	0.00
C12:0	0.00	0.00	0.00	0.00	0.00	0.00
C14:0	0.00	0.00	0.00	0.00	0.00	0.00
C16:0	205	207	232	214	15.0	7.00
C18:0	29.3	33.5	46.8	36.6	9.11	24.9

C20:0	0.281	0.624	0.504	0.470	0.174	37.0
C22:0	0.262	0.581	0.465	0.436	0.161	37.0
C24:0	0.032	0.073	0.054	0.053	0.020	38.4
TOTAL SFA	235	242	279	252	24.2	9.59
C14:1(*cis*-9)	0.00	0.00	0.00	0.00	0.00	0.00
C16:1(*cis*-9)	0.313	1.23	3.63	1.73	1.71	99.3
C18:1(*cis*-6)	74.8	113	61.9	83.1	26.3	31.7
C18:1(*cis*-9)	65.9	95.1	99.9	87.0	18.4	21.2
C20:1 (*cis*-11)	1.06	2.37	1.91	1.78	0.662	37.2
C22:1(*cis*-13)	0.089	0.198	0.160	0.149	0.056	37.2
C24:1(*cis*-15)	0.032	1.80	0.054	0.629	1.01	161
TOTAL MUFA *cis*	142	213	168	175	36.1	20.7
C18:1 (*trans*-6)	0.101	0.225	0.181	0.169	0.063	37.2
C18:1 (*trans*-9)	0.009	0.020	0.016	0.015	0.006	38.0
C18:1 (*trans*-11)	0.00	0.00	0.00	0.00	0.00	0.00
TOTAL MUFA *trans*	0.110	0.245	0.198	0.184	0.069	37.3
MUFA Total	142	213	168	175	36.1	20.7
C20:2 (*cis*-11,14)	0.040	0.089	0.071	0.066	0.025	37.3
C20:3 (*cis*-11,14,17)	0.172	0.383	0.309	0.288	0.107	37.2
C20:5 (*cis*-5,8,11,14,17)	0.032	0.071	0.057	0.053	0.020	37.2
C22:6 (*cis* 4,7,10,13,16,19)	0.297	0.071	0.611	0.326	0.271	83.1
TOTAL (*n*-3)	0.540	0.614	1.05	0.734	0.274	37.4
C18:2 (*cis*-9,12)	235	236	313	261	44.7	17.1
C18: 2 (*trans*-9,11)	0.118	0.264	0.212	0.198	0.074	37.2
C18:3 (*cis*-6,9,12)	8.89	13.4	14.8	12.4	3.09	25.0
C18:3 (*cis*-9,12, 15)	9.97	15.5	16.5	14.0	3.51	25.1
C20:3 (*cis*-8,11,14)	2.60	4.42	4.55	3.85	1.09	28.3
C20:3 (*cis*-11,14,17)	1.72	0.383	0.309	0.804	0.794	98.7
C20:4 (*cis*-5,8,11,14)	5.06	0.665	0.708	2.15	2.53	118
C22:2 (*cis*-13,16)	0.032	0.071	0.057	0.053	0.020	37.2
TOTAL (*n*-6)	264	271	350	295	48.0	16.3
PUFA Total	264	271	351	296	48.3	16.3

Table.7 Phospholipids level (mg/100g) in grape (O1), orange (O2) and tangerine (O3) seeds

Phospholipids	O1	O2	O3	Mean	SD	CV %	X^2	TV	Remark
Phosphatidylethanolamine	86.3	87.6	101	91.6	8.14	8.88	1.45	5.99	NS
Phosphatidylcholine	213	295	227	245	43.9	17.9	15.7	5.99	S
Phosphatidylserine	15.3	26.4	18.5	20.1	5.71	28.5	3.25	5.99	NS
Lysophosphatidylcholine	5.20	3.69	7.02	5.30	1.67	31.4	1.05	5.99	NS
Phosphatidylinositol	171	177	216	188	24.4	13.0	6.35	5.99	S
Total	491	590	570	550	52.3	9.50	9.93	5.99	S

CV= coefficient of variation, X^2=Chi-square test, TV= table value at α=0.05, n-1=2, NS= not significant, S= significant

Table.8 Phytosterols level (mg/100g) in grape (O1), orange (O2) and tangerine (O3) seeds

Phytosterol	O1	O2	O3	Mean	SD	CV %	X^2	TV	Remark
Cholesterol	2.19e-3	6.63e-3	5.35e-3	0.005	0.002	48.4	0.002	5.99	NS
Cholestanol	2.24e-3	2.61e-3	0.0208	0.009	0.011	124	0.026	5.99	NS
Ergosterol	0.0450	0.0454	0.0226	0.038	0.013	34.6	0.009	5.99	NS
Campesterol	54.4	68.1	59.7	60.7	6.91	11.4	1.57	5.99	NS
Stigmasterol	12.7	17.0	17.0	15.6	2.48	15.9	0.792	5.99	NS
5- Avenasterol	9.93	18.5	13.9	14.1	4.29	30.4	2.61	5.99	NS
Sitosterol	283	381	321	328	49.4	15.0	14.9	5.99	S
Total	360	485	412	419	62.6	14.9	18.7	5.99	S

CV= coefficient of variation, X2=Chi-square test, TV= table value at α=0.05, n-2, NS= not significant, S= significant

From the standpoint of human nutrition, a major benefit to be derived from the consumption of citrus seed oils (and many other seed oils) in preference to animal fats is their high content of the essential fatty acids particularly linoleic acid whose content of citrus seed oils is from 37.2-45.2 % of the fatty acids present, and certainly makes this oil one of those high in polyunsaturated fatty acids. Other nutritional attributes were high levels of monounsaturated fatty acids of 21.6-29.4 %; phospholipids levels of 491-590 mg/100 g and sitosterol level of 283-381 mg/100 g. On the overall summary of parameter distribution and representing grape as O1, orange as O2 and tangerine as O3, we have:

Total fatty acid (and crude fat): O3 > O2 >O1
SFA: O1 > O3 > O2
MUFA (*cis*): O2 > O1 >O3
PUFA : 03 > O1 > O2
PUFA/SFA: O3 >O2 = O1
MUFA/SFA: O2 >O1 >O3
Phytosterols: O2 > O3 >O1
Phospholipids: O2 > O3 >O1

That is: O1 was overall highest in: SFA (1/8 or 12.5 %), overall second in PUFA, MUFA (*cis*) and MUFA/SFA (3/8 or 37.5 %) and overall third in Total fat (crude fat), PUFA/SFA, Phytosterols and Phospholipids (4/8 or 50.0 %). For O2, it was overall highest in: MUFA (*cis*), MUFA/SFA, Phytosterols and Phospholipids (4/8 or 50.0 %), overall second in Total fat and PUFA/SFA (2/8 or 25.0 %) and overall third in SFA and PUFA (2/8 or 25.0 %). For O3, it was overall highest in: PUFA, PUFA/SFA and Total fat (3/8 or 37.5 %), overall second in SFA, Phytosterol and Phospholipids (3/8 or 37.5 %) and overall third in MUFA (*cis*) and MUFA/SFA (2/8 or 25.0 %). Based on the above information we can categorise the samples on their nutritional quality characteristics as O2 (sweet orange) > O3 (tangerine) > O1 (grape).

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