

Characterization of fatty acids and triacylglycerols in vegetable oils by gas chromatography and statistical analysis

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Abstract

Fatty acids and triacylglycerols from *Argania* tree seeds (argania oil) were identified. Common fatty acids and triacylglycerols were quantified using gas chromatography on a capillary column. A frequent adulteration of some kinds of argania oil was determined using statistical methods. Hierarchical cluster analysis clearly distinguished between oil directly isolated from seeds and bought in a pharmacy, and oil bought in the grocer's shop. ©1999 Elsevier Science B.V. All rights reserved.

Keywords: *Argania spinosa* oil; Fatty acids and triacylglycerols; Cluster analysis by SPSS; Capillary gas chromatography

1. Introduction

Argan (*Argania spinosa*, also known as *A. sideroxylon*, Sapotaceae) is a medium-sized thorny evergreen tree. It originates from southwestern Morocco, from a narrow strip of land along the Atlantic coast. Several criss-crossed stems sometimes occur on its knotty trunk; the leaves are small, lanceolate, evergreen, but pale on the underside. The tree is in flower from May to June and its hermaphrodite flowers have a greenish-yellow color. The fruit of argan tree is a green ovoid drupe containing an almond with fleshy oily albumen. The fruits have a bitter pericarp surrounding a stone-like structure and contain 1–3 kernels with a high (over 50%) oil content. In the country of its ori-

gin, the argan tree serves as a source of feed for wild animals, a source of oil and a source of firewood [1].

The original process for argan oil isolation is the following. The berries are harvested and dried in the sun. The pulp is removed from the dried fruit and the kernels are hand broken. Then the almonds are sorted, roasted slowly and crushed. The mixture is then treated in hot water and the floating oil is collected. The oil has a brown color and an unpleasant taste but it brightens when decanted. The pulp is collected and used as a feed for cattle. The resulting output is rather small – 100 kg of fresh fruit giving approximately 2 kg of oil. It is interesting to note the typical and original kernel harvesting method: the goats are very fond of argan fruit and to get at it, even climb up the trees. They digest the pericarp but excrete the kernels that are collected by the peasants and used for oil preparation [2,3].

The dietetic value of the oil is high as the total unsaturated fatty acids/total saturated fatty acids ratio

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is approximately 4.5, which is similar to that of olive oil [2]. The oil has a unique aroma and the Moroccans consider it to be the best culinary oil. The price of argan oil is US\$ 43 per litre in comparison with US\$ 4 per litre of the olive oil. The oil yield is around 1 kg per tree annually for the best specimens [3].

Modern analytical methods including capillary column chromatography enabled us to disclose the adulteration of vegetable oils including argan oil. Unfortunately, vegetable oils do not contain a wide fatty acid spectrum and thus, due an excellent knowledge of the single fatty acid content, the oils are easily falsified. The majority of references concern olive oil and cocoa butter adulteration [4–6]. All the references agree that in order to disclose oil adulteration it is necessary to determine not only the fatty acid percentage range but also the qualitative and quantitative triacylglycerol content [7–12]. As mentioned above, argania oil is unique because of its occurrence, and thus also suitable for adulteration. Based on our former works [13–15] we compared four kinds of argania oil: the oil isolated in our lab directly from the seeds, oil bought from a pharmacy, another one bought from a grocer's shop and finally oil falsified (artificially setup) in our laboratory. Capillary gas chromatography was used to assay the samples for fatty acids and triacylglycerols and compare them with other vegetable oils. The methods of statistical analysis were used to obtain the overall image of the fatty acid and triacylglycerol content in vegetable oils.

2. Experimental

The fruits were gathered in March 1998 near the road from Agadir to Essaouira, the oil was bought from a shop in Agadir and from a pharmacy in Marrakesh (Morocco). The other oils were already present in the lab as they were formerly used for experiments [13–15].

The capillary gas chromatography analysis was done according to the method, which was worked out soon. The method comprises total triacylglycerols analysis on a fused capillary column 25 m $\times$ 0.25 mm ID $\times$ 0.1 mm phase TAP (Chrompack, NL). Hydrogen was used as a carrier gas (100 cm/s), the temperature was set from 320 to 350°C and the rate was 1.5°C. The injector OCI-3 (SGE, Kensington, Australia)

was adapted to the 'movable on-column' injection technique [16].

Detector FID (375°C) and chromatographic apparatus PU 4900 (Pye Unicam, Cambridge, UK) were used for analysis. The samples were dissolved in isooctane and 0.2 ml of a 0.1% solution was injected (57 : 0 was the internal standard).

The statistical analysis was done using the statistical package SPSS 8.0.1 (SPSS Inc. Chicago, USA).

3. Results and discussion

The experimental results are summarized in Tables 1 and 2. Table 1 shows the percentage content of individual fatty acids. The ordinary fatty acids identified, included especially palmitic acid and a mixture of fatty acids with 18 carbon atoms (especially oleic and linoleic acids, but stearic and linolenic acids also in smaller amounts). The content of fatty acids with 20 or more carbon atoms in the chain was negligible. Unfortunately, using any method including GC-MS of methyl esters or oxazolines, we did not succeed in determining at least trace amounts (more than 0.005% of total fatty acids) of docosahexaenoic acid, which was formerly identified in argania oil [17]. The percentage content of triacylglycerols in argania oil and in other vegetable oils is shown in Table 2. The most frequent triacylglycerols are: OLL, OOL, POL, OOO and POO. These results are in good agreement with literature data [18]. It is necessary to remark that argania oil is noted for the presence of some specific species of triacylglycerols, e.g. PSO and SSL. These triacylglycerols are mostly unique to argania oil (see Table 2). It is clear from Table 1 that the fatty acid content differs in argania oil bought from the shop, but almost no differences were found between argania oil isolated from seeds, that bought from a pharmacy and the oil prepared in our lab.

Completely different results were obtained from the cluster analysis of triacylglycerols. At first sight, it is clear that it was not possible to prepare artificially argania oil with triacylglycerol content equivalent to that isolated directly from the argania seeds. This test fully documented the merchants' dishonesty and is in good correspondence with already published data.

We tried to present the similarity of oils by means of statistical analysis. Oils are the objects under study

Table 1
Fatty acid composition from different plant oils separated by CGC

FAME ^a	Corn	Cotton	Grape	Olive	Peanut	Soy	Palm	Cocoa	Sunflower	Argania	Pharmacy	Shop	Falsified
14:0	0.2	0.8	0.0	0.0	0.0	0.0	0.9	0.1	0.1	0.1	0.1	0.1	0.1
16:0	13.0	27.3	7.0	10.2	12.5	11.6	43.7	0.1	5.5	10.3	10.5	6.8	10.8
16:1	0.0	0.8	0.1	0.7	0.0	0.3	0.1	0.3	0.1	0.1	0.1	0.1	0.1
17:0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0
18:0	2.5	2.0	3.0	2.5	2.5	4.2	4.5	44.6	4.7	4.2	4.1	3.1	4.0
18:1	30.5	18.3	22.1	78.1	37.9	21.6	39.8	48.1	19.5	43.8	43.1	62.8	43.6
18:2	52.1	50.5	67.2	7.1	41.3	53.7	10.5	4.9	68.5	36.9	37.5	24.3	36.8
18:3	1.0	0.0	0.5	0.6	0.3	7.5	0.3	0.1	0.1	3.8	3.4	2.4	3.9
20:0	0.5	0.3	0.1	0.5	0.5	0.8	0.2	1.5	0.3	0.4	0.3	0.2	0.3
20:1	0.2	0.0	0.0	0.3	0.7	0.3	0.0	0.1	0.1	0.3	0.2	0.1	0.3
22:0	0.0	0.0	0.0	0.0	2.5	0.0	0.0	0.1	0.9	0.1	0.1	0.1	0.1
22:1	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.4	0.0	0.0
24:0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	0.0	0.2	0.0	0.2	0.0	0.0

^a First no.: no. of carbon atoms in the chain; second no.: no. of double bonds.

and percentage contents of fatty acids or triacylglycerols are the variables. Cluster analysis is usually used for assessing the similarity of objects. Some other methods exist for the solution of our task, e.g. multidimensional scaling. If we investigate a transposed data matrix (oils are considered as variables), factor analysis could be used. The results of methods mentioned above can be presented in charts. The disadvantage of factor analysis and multidimensional scaling consists of the possibility to graphically represent the similarity of objects (variables) on the basis only of two or three factors (dimensions). The charts obtained on the basis of any of these methods can sometimes be more intelligible, but sometimes the points and their labels can be overlapping. In our opinion, the best way of representing similarity objects is a dendrogram, which gives the possibility to present the succession of finding of similarity.

In cluster analysis and in multidimensional scaling, similarity of objects is determined on the basis of the measure of distance (similarity or dissimilarity). This measure must correspond to the type of data. The default distance measure of statistical packages may be used only when the absolute differences of the values related to certain objects (oils) measured for two variables, correspond to the dissimilarity of the oils in terms of these variables. However, this is not our case. If the two oils exhibit similar absolute differences between the values measured for a major acid and a minor fatty acid, then in terms of the minor fatty acid, the oils are more different. Even very small difference in the percentage content of a minor fatty acid

can completely change the characteristics of the oil. The standardisation of the values (an average of values of the observed variable is subtracted from the measured value and the result is divided by the standard deviation) cannot eliminate this discrepancy. It could be partially eliminated by transforming the values to the interval from 0 to 1 (each value is divided by the maximum of observed values).

Statistical packages provide a possibility to use different measures and different ways of value transformations. For interval data, SPSS offers Square Euclidean (SEUCLID) distance, Euclidean (EUCLID) distance, correlation coefficient as a similarity measure, Chebychev distance metric, city block or Manhattan distance and distance in an absolute power metric, for frequency count data there are chi-square and phi-square measures of dissimilarity. For frequencies chi-square distance is recommended because exact tests based on hypergeometric distribution are too complicated. In relation to this type of data we consider two nominal variables – ‘oils’ and ‘fatty acids’ (‘triacylglycerols’). The categories of variable ‘oils’ are single kinds of oils. So, our data are percentage counts in the contingency table. Other measures are not adequate because the variability of variables is different.

We compared the graphical representations of similarity of objects by cluster analysis and by multidimensional scaling (with the same measures). The distances of objects in charts are very similar. We illustrate the results on the basis of SEUCLID distance for original data, EUCLID distance for the data transformed

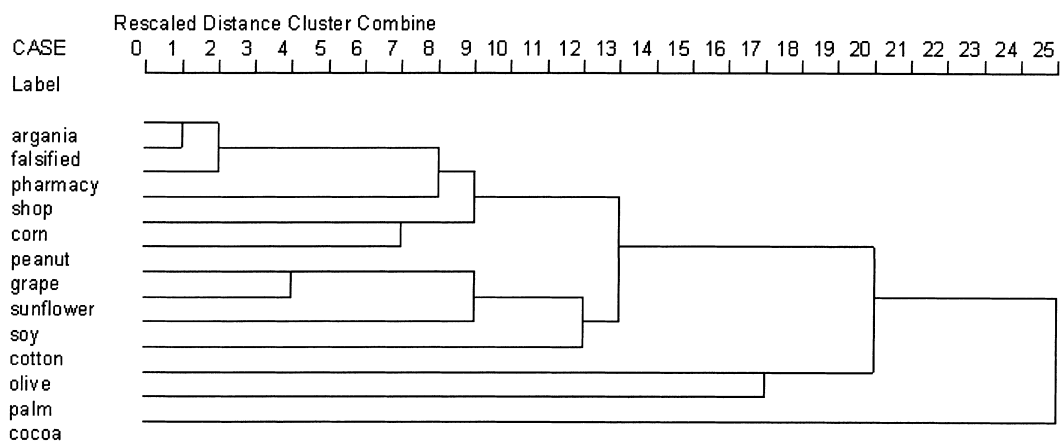


Fig. 1. Similarity of oils based on the percentage content of fatty acids using the chi-square dissimilarity measure.

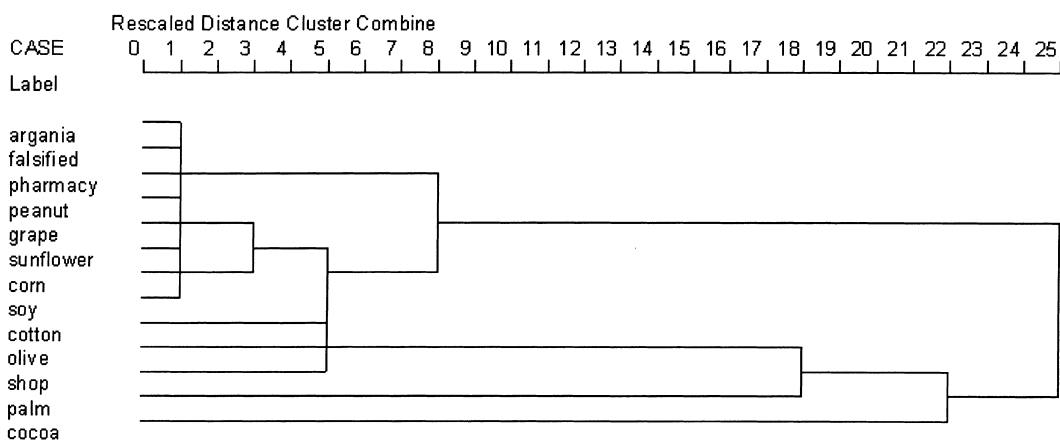


Fig. 2. Similarity of oils based on the percentage content of fatty acids using SEUCLID distance measure.

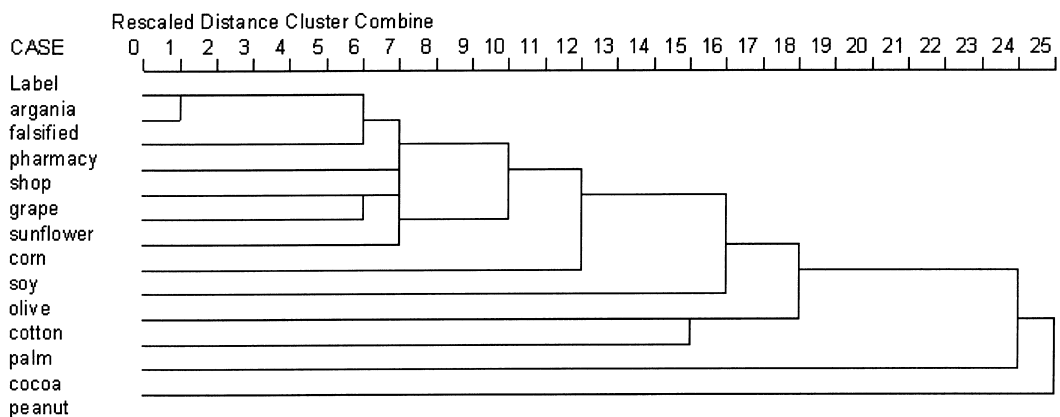


Fig. 3. Similarity of oils based on the percentage content of fatty acids using EUCLID distance measure for the data transformed to the interval from 0 to 1.

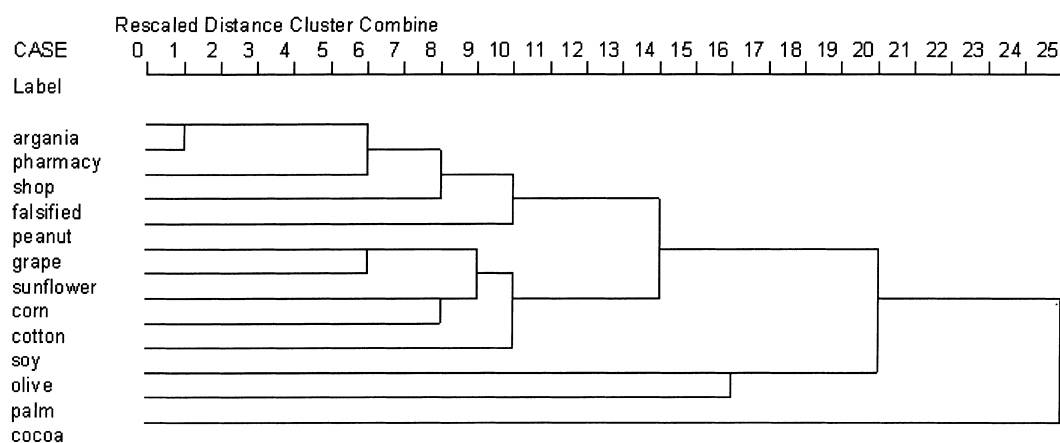


Fig. 4. Similarity of oils based on the percentage content of triacylglycerols using the chi-square dissimilarity measure.

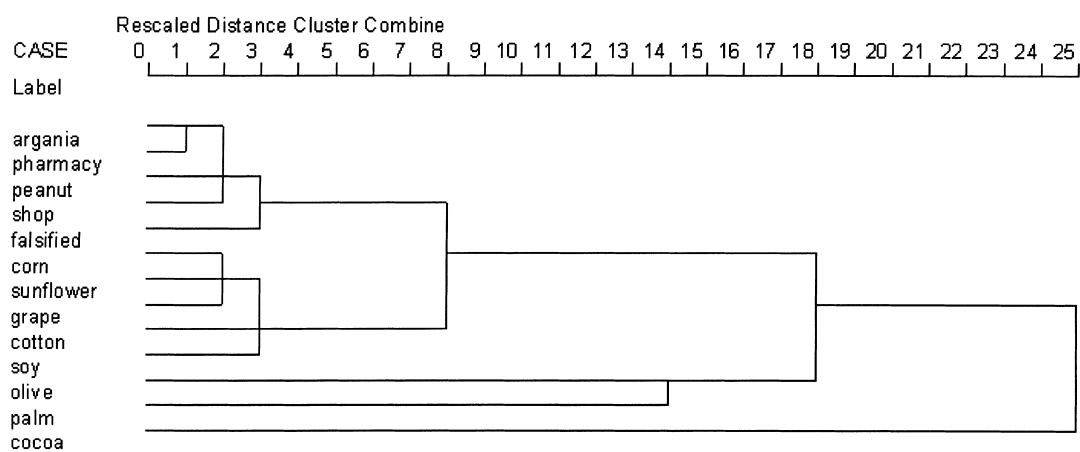


Fig. 5. Similarity of oils based on the percentage content of triacylglycerols using SEUCLID distance measure.

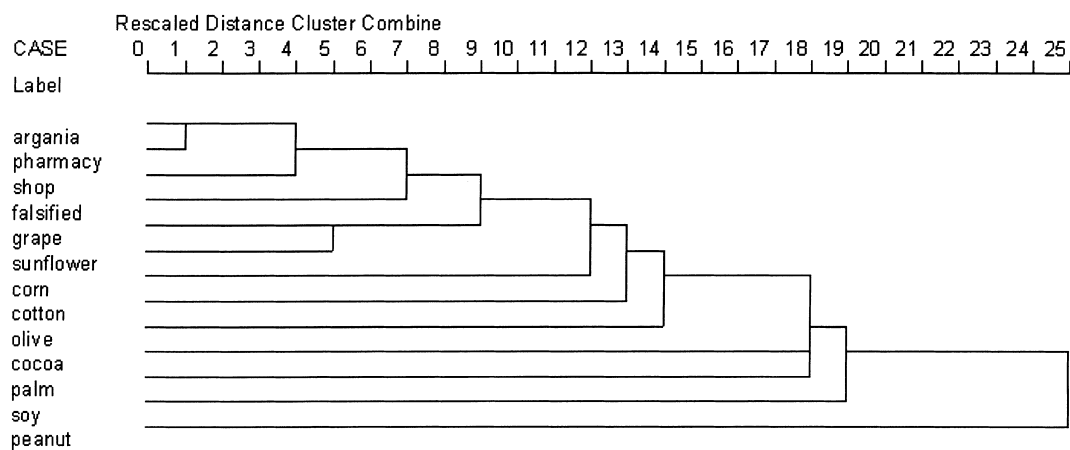


Fig. 6. Similarity of oils based on the percentage content of triacylglycerols using EUCLID distance measure for the data transformed to the interval from 0 to 1.

Table 2

Triacylglycerols composition from different plant oils separated by CGC

TG ^a	Corn	Cotton	Grape	Olive	Soy	Cocoa	Palm	Sunflower	Peanut	Argania	Pharmacy	Shop	Falsified
LLLn	1.2	1.5	0.0	0.0	8.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OLnLn	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
LLL	25.6	23.2	32.6	1.2	16.8	0.0	0.0	28.0	3.6	6.9	6.2	15.0	14.6
PoLL	0.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OLLn	0.6	0.0	0.0	3.3	4.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6
PLLn	0.0	0.0	0.0	0.0	2.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
OLL	20.5	15.4	19.1	4.3	15.4	0.0	1.0	23.1	18.9	14.2	12.8	17.7	13.7
OOLn	0.3	0.0	0.0	0.0	1.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
PLL	16.4	24.5	17.6	2.8	13.8	0.0	4.1	16.8	6.1	6.0	5.9	10.5	9.8
POLn	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MPL	0.0	0.0	0.0	0.0	0.0	0.0	0.6	0.0	0.0	0.0	0.0	0.0	0.0
PPLn	0.1	1.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
SLL	1.9	1.1	6.9	0.0	4.0	0.0	0.6	10.2	1.2	1.7	2.0	5.3	5.1
OOL	10.9	5.1	3.4	2.4	10.1	0.0	2.6	7.5	18.1	17.3	18.1	13.5	5.0
POL	10.4	10.9	15.5	9.5	10.7	1.0	11.0	6.0	11.6	11.9	12.0	9.5	7.8
PPL	0.0	7.7	0.3	3.0	1.4	3.3	10.7	1.2	2.0	1.3	2.0	1.7	2.1
PMgO	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
MPO	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0
OLGa	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.6	0.0	0.0	0.1	0.0
OOO	3.1	1.6	0.0	36.7	2.3	1.7	4.7	1.4	8.1	14.7	14.5	8.6	19.1
PLGa	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.1	0.0	0.0	0.1	0.0
SOL	1.2	0.9	1.9	0.0	2.9	1.3	0.5	2.4	2.1	3.0	2.8	2.6	1.2
POO	2.5	2.0	1.4	23.4	1.9	6.3	22.5	0.6	7.6	12.2	13.4	7.6	12.0
PSL	2.8	1.1	0.1	0.0	1.0	5.1	2.4	1.0	0.8	1.7	1.1	1.0	0.5
PPO	1.5	1.5	0.0	4.9	0.2	18.0	24.8	0.0	1.2	3.1	3.3	1.8	2.4
LLBe	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.5	0.0	0.0	0.1	0.0
PPP	0.0	0.0	0.0	0.5	0.0	0.0	7.2	0.0	0.0	0.0	0.0	0.0	0.2
OOGa	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0
POGa	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.2	0.0	0.0	0.1	0.0
SOO	0.3	0.7	0.2	5.7	0.5	7.9	1.7	0.8	3.4	2.8	2.4	1.8	3.2
SSL	0.0	0.6	0.7	0.0	0.2	1.6	0.0	0.4	0.9	1.1	1.0	0.7	0.2
PSO	0.1	0.5	0.3	1.5	0.4	30.8	4.9	0.6	0.9	1.8	1.5	1.1	1.1
PPS	0.4	0.0	0.0	0.0	0.2	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0
OLBe	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.6	0.0	0.0	0.1	0.0
OOA	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	2.5	0.0	0.0	0.2	0.0
SLA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	0.1	0.0
SSO	0.0	0.0	0.0	0.8	0.2	22.0	0.0	0.0	0.8	0.3	1.0	0.6	0.4
OOBe	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.2	0.0	0.0	0.1	0.0
SLBe	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0
SOA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.7	0.0	0.0	0.1	0.0
OAA	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.5	0.0	0.0	0.0	0.0

^a M: myristic acid; P: palmitic acid; Po: palmitoleic acid; Mg: margaric acid; S: stearic acid; O: oleic acid; L: linoleic acid; Ln: linolenic acid; A: arachidic acid; Ga: gadoleic acid and Be: behenic acid.

to the interval from 0 to 1 and chi-square measure for original data. If the two objects are marked x and y , then SEUCLID distance is counted according to the following formula:

$$\text{SEUCLID}(x, y) = \sum_{i=1}^m (x_i - y_i)^2,$$

where x_i is the value of i th variable for object x , y_i is the value of i th variable for object y and m is the number of variables. EUCLID distance is the square root of SEUCLID:

$$\text{EUCLID}(x, y) = \sqrt{\sum_{i=1}^m (x_i - y_i)^2}.$$

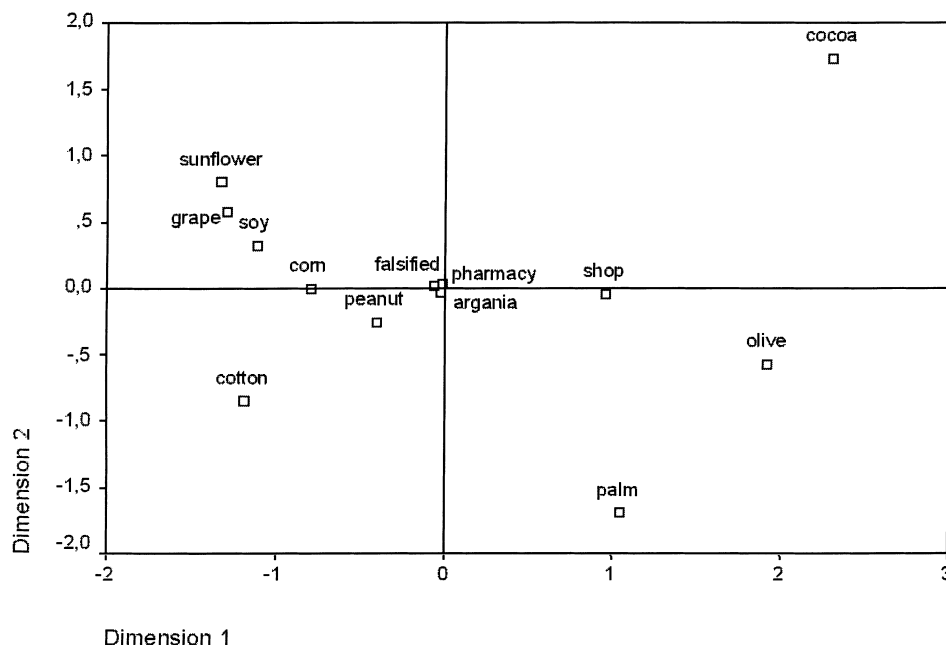


Fig. 7. Similarity of oils based on the percentage content of fatty acids using the chi-square dissimilarity measure – two dimensions.

The chi-square measure is expressed by the following formula:

$$\text{CHISQ}(x, y) = \sqrt{\sum_{i=1}^m \frac{(x_i - E(x_i))^2}{E(x_i)} + \sum_{i=1}^m \frac{(y_i - E(y_i))^2}{E(y_i)}}$$

where E is the expected value from the model of independence of variables. It is computed as:

$$E(x_i) = \frac{(x_i + y_i) \sum_{i=1}^m x_i}{\sum_{i=1}^m x_i + \sum_{i=1}^m y_i} \quad \text{and}$$

$$E(y_i) = \frac{(x_i + y_i) \sum_{i=1}^m y_i}{\sum_{i=1}^m x_i + \sum_{i=1}^m y_i}.$$

Besides the choice of the distance measure, the choice of the cluster method is done in cluster analysis. Unweighted average linkage between groups, which is set up in SPSS as default, was used as a cluster method for our case. In our task, the cluster method influences only the levels of grouping of objects, not the order of objects for grouping. In multidimensional scaling we considered two and three dimensions.

The results of investigation of oils similarity on the basis of fatty acid composition are presented in Figs.

1–3 and 7–8. In the case of multidimensional scaling, we only show the charts based on the chi-square measure of dissimilarity (Fig. 7 shows graphical representation for two dimensions, Fig. 8 for three dimensions). For other measures the results obtained by both methods are also comparable.

When using SEUCLID distance measure (see Fig. 2), some objects are difficult to distinguish. By using

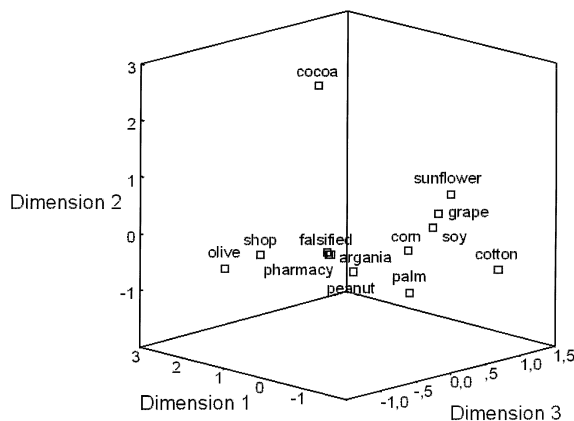


Fig. 8. Similarity of oils based on the percentage content of fatty acids using the chi-square dissimilarity measure – three dimensions.

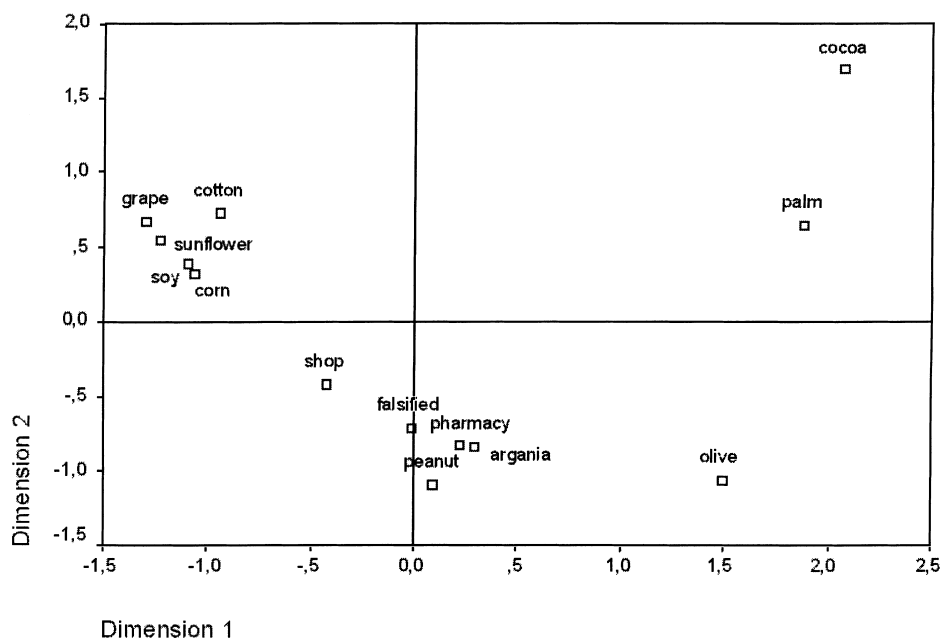


Fig. 9. Similarity of oils based on the percentage content of triacylglycerols using the chi-square dissimilarity measure – two dimensions.

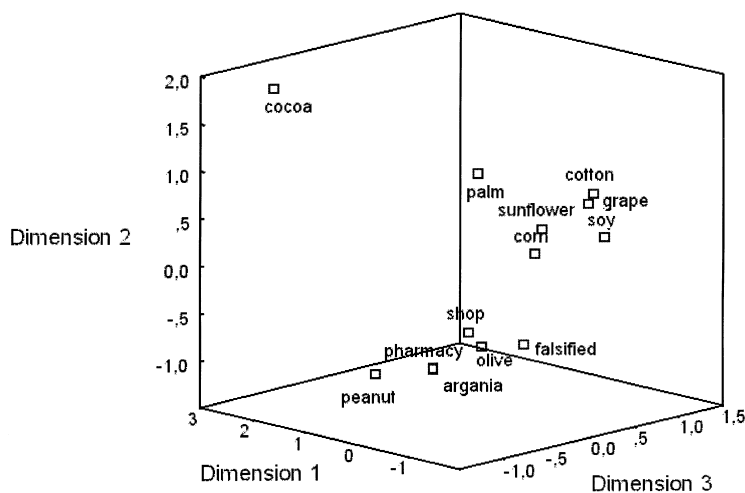


Fig. 10. Similarity of oils based on the percentage content of triacylglycerols using the chi-square dissimilarity measure – three dimensions.

EUCLID distance measure for the data transformed to the interval from 0 to 1 (see Fig. 3) we could distinguish between peanut and other oils. Only relationships between objects obtained by chi-square measure (see Fig. 1) correspond with the real variance between oils. The dendrogram in Fig. 1 depicts the close relationship of three samples: oil isolated from the seeds, oil bought from the shop and oil prepared in our lab.

The similarity of the oils also depends on botanical relations of the plants, from which the oils have been isolated. There is a close relationship between grape and sunflower oils, while the cocoa, palm and olive oils differ considerably. It must be mentioned here that the similarity was assessed, based on the percentage content of only 10 fatty acids. More than half of them were minor acids, which is not sufficient for clear dif-

ferentiation as was shown in the case of argania oil.

The results of investigation of oils similarity on the basis of triacylglycerol composition are presented in Figs. 4–6 and 9–10. In the case of multidimensional scaling, we again show only the charts based on chi-square measure of dissimilarity (Fig. 9 shows graphical representation for two dimensions, Fig. 10 for three dimensions). The results obtained by both methods are also comparable with other measures. The dendrogram in Fig. 4, obtained by hierarchical cluster analysis, and the charts in Fig. 10, obtained by multidimensional scaling (both with the chi-square dissimilarity measure), pose many questions for discussion.

A final note about statistical analysis concerns the principal component analysis, which is used in a paper [7]. This method measures distances between variables on the basis of the correlation coefficient, which is not convenient for comparing oils on the basis of percentage contents. The explanation of this fact is given above.

We consider cluster analysis and multidimensional scaling with chi-square measure of dissimilarity the best methods for determination of similarity of oils on the basis of percentage contents (fatty acids or triacylglycerols). In terms of graphical representation, we consider dendrogram the best chart.

Finally, we can state that after 15 years of experience in the field of analytical chemistry we did not succeed in falsifying the oil so perfectly that the adulteration could not be disclosed using capillary gas chromatography of triacylglycerols. It is of course possible to falsify oils by mixing appropriate amounts of individual molecular types of triacylglycerols. However, this method cannot be industrially used, as it is too expensive. Analytical detection of possible oil adulteration is much easier and shorter when multivariate

statistical methods are used. With modern analytical methods including that capillary gas chromatography, the commercially produced software it makes possible to determine very easily any oil adulteration. This ease of analysis is of great economical importance (see the introduction).

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