Potential of Fourier Transform Infrared Spectroscopy for the Authentication of Vegetable Oils

Yoke W. Lai,† E. Katherine Kemsley, and Reginald H. Wilson*

AFRC Institute of Food Research, Norwich Laboratory, Norwich Research Park, Colney, Norwich NR4 7UA, U.K.

Fourier transform infrared (FTIR) spectroscopy was used in conjunction with principal component analysis (PCA) and discriminant analysis to investigate the potential of the technique for determining the authenticity of vegetable oils. PCA applied to spectra from a range of seed oils revealed clustering according to plant species. When extra virgin and refined olive oils were subjected to discriminant analysis, 93% of samples in a calibration set and 100% of samples in an independent validation set were correctly classified, despite these two types of oil being chemically and spectroscopically very similar. The method, therefore, has potential as a rapid method for the determination of product authenticity.

1. INTRODUCTION

Adulteration of "added value" food products, involving the replacement of high-cost ingredients with lower grade, cheaper substitutes, could potentially be very lucrative for a manufacturer or raw material supplier. Composition, declared on the label by the manufacturer, often legally determines what a product can be called and at what price it can be sold. Chemical or sensory analysis of the product may be the only means of verifying the product composition. Substitution or adulteration of a product with a cheap ingredient is not only a major economic fraud but may also have severe health implications to consumers. In Spain, some 400 deaths and 20 000 casualties have occurred since May 1981 from the disease now known as "Spanish toxic syndrome", which is caused by the consumption of adulterated cooking oil (Jimeno, 1982; Kochlar and Rossell, 1984). Therefore, the detection of food adulteration is of vital importance.

Authentication of food constituents is a major challenge in food analysis, and it is often a time-consuming and laborious process. Very often, complex chemical treatment of the sample and the use of sophisticated instruments are required. In this paper, the potential of Fourier transform infrared (FTIR) spectroscopy for the authentication and detection of adulteration in an important class of food materials, edible oils, is investigated. Particular attention is paid to the discrimination of "extra virgin" and refined olive oils.

Infrared spectroscopy has been widely used as an analytical technique in industrial, forensic, food, and agricultural laboratories. The use of the near-infrared (NIR) region of the electromagnetic spectrum (15 000–4000 cm^{-1}, 0.72–2.5 μm) for rapid quantitative analysis of many constituents, such as moisture, fat, protein, and fiber, in food and agricultural products is well documented (Osborne and Fearn, 1986; Williams and Norris, 1987). However, the use of the mid-infrared (MIR) region of the spectrum (4000–400 cm^{-1}, 2.5–25 μm) has been largely overlooked by the food analyst, mainly due to instrumental and sampling problems (Wilson, 1990). Many food samples are opaque and highly scattering and contain water, which is a very strong infrared absorber. In addition, these materials are often difficult to prepare as pellets or mulls for traditional mid-IR analysis, and dispersive mid-IR measurements are relatively slow. Nevertheless, there are significant advantages in using the mid-IR region for analysis: absorption bands are well-resolved and can be assigned to specific chemical groups; quantitative analyses are more or less constituent specific. Advances in infrared instrumentation, in particular the incorporation of Fourier transform techniques (Hirschfeld, 1983, 1984), and new sample presentation techniques such as attenuated total reflectance (ATR), photoacoustic detection (PAS), and diffuse reflectance (DRIFT) (Coates et al., 1987a,b), have led to a resurgence of interest in the application of mid-IR to the analysis of food samples. FTIR spectroscopy offers significant advantages over dispersive infrared methods (Chenery and Sheppard, 1987), among which are higher signal-to-noise ratios and rapid spectral acquisition. A combination of FTIR with the new sample presentation techniques has created a valuable tool for food analysis (Wilson, 1990).

The products studied in this paper are vegetable oils, including extra virgin and refined olive oils. Olive oil is extracted from the fruit of the olive tree, *Oleo europea* L. Good quality olive oil is characterized by a fragrant and delicate flavor, which is highly prized. There is a large price gap between olive and other vegetable oils and also between the various grades of olive oil: extra virgin may cost nearly twice as much as refined oils. Consequently, adulteration of olive with cheaper oils is a temptation. International concern about the adulteration of these products has led to the development of draft standards for olive oil [International Olive Oil Council (IOOC), 1984; World Health Organization (WHO), 1984]. Three grades are commonly available in the supermarket: extra virgin, virgin, and (refined) olive oil.

Adulteration of olive oil with seed oils is a problem that has existed for centuries. More than 170 years ago (Poutet, 1819), a procedure was reported for identifying adulteration of olive with poppyseed and rapeseed oils. More recently, a monitoring program conducted by the U.S. Food and Drug Administration in 1983–1984 (Firestone et al., 1985) found that among the 28 samples of olive oil analyzed, the content of 20 samples did not correspond to the declaration on the label. Most of the samples analyzed were found to have been substituted in whole or in part with lower grade olive (residue and esterified) and/or other seed oils (soya and corn). A follow-up survey by the same

* Author to whom correspondence should be addressed.
† Present address: Department of Chemistry Malaysia, Jalan Sultan, 46661 Petaling Jaya, Malaysia.

0021-8561/94/1442-1154$04.50/0

agency in 1985–1986 (Firestone et al., 1988) showed that the problem still existed. Therefore, continuous vigilance is required to control adulteration of olive oil products and to protect the interest of the consumers as well as the industry.

Various physical and chemical tests have been proposed to establish the authenticity of olive oil and to detect the level of adulterants in it. UV spectroscopy based on absorptivity in the regions of 208–210and 310–320 nm has been widely used to detect adulteration of virgin with refined olive oil (Passaloglou-Emmanouillidou, 1990). A second derivative spectrophotometry method was reported to be able to detect adulteration at a level of 6% (Kapoulas and Andrikopoulos, 1987). Analysis of the fatty acid profile of an oil after methylation by gas chromatography is a well-established method for the quantitative analysis of mixtures of olive and other seed oils (Kapoulas and Passaloglou-Emmanouillidou, 1981; Morchio et al., 1989; Lanzou et al., 1989). HPLC analysis of the triglycerides and fatty acid composition is also widely used to detect adulteration in olive oil (Sanchis Rodriguez and Rodriguez Serrano, 1991). Nuclear magnetic resonance analysis (Sacchi et al., 1970) and a spectrophotofluorometric method (Marini et al., 1990) have also been reported.

Infrared spectroscopy has been used as an analytical tool in the oil and fat industries for many years (Kochlar and Rossell, 1987; Chapman, 1965). However, the mid-IR region has not been used for the detection of adulteration in olive oils. The most likely reason for this is that most vegetable oils contain the same fatty acids (mainly with carbon number 16 or 18), and their triglyceride contents are also quite similar (carbon number 50, 52, or 54). As a result, the mid-IR spectra of most seed oils are superficially similar and dominated by the C–H and C–O vibrations of the polymethylene chains. For oils as chemically alike as the various grades of olive, the spectral differences can be very small indeed. Nevertheless, subtle dissimilarities between spectra do exist, due to differences in the structure of the polymethylene chains, so that the spectrum of an oil actually contains a great wealth and variety of information about the composition of the sample. The analytical problem is to extract the information from the spectra in a way that can be used to differentiate the oil types.

The recent introduction of multivariate statistical methods such as partial least square (PLS), principal component analysis (PCA), and discriminant analysis into FTIR has opened up new frontiers in analysis. Multivariate statistics have been used extensively in the characterization of oils in recent years (Forina et al., 1983; Peipponen et al., 1983; Consiglieri and Bruschi, 1992). PCA has been used in the authentication of olive oil, and the detection of adulteration with seed oils, based on the triglyceride and fatty acid content obtained by high-performance liquid chromatography (HPLC) analysis (Tsimidon et al., 1987a,b). Therefore, it may be possible to determine the authenticity and the adulteration of olive oil by using multivariate techniques in conjunction with FTIR. The first stage in this work, reported here, is to ensure that oils of different botanical origin can be differentiated.

The data processing reported in this paper covers two areas: exploratory analysis on a data set comprising spectra from eight different plant oils, with the aim of investigating groupings according to oil type; and discriminant analysis on a data set from extra virgin and refined olive oils, with the aim of assigning spectra to either of the two classes. For both analyses, PCA has been used as a first step, since it is an efficient "data reduction" method. A typical spectrum contains several hundred data points (or "variates"); a typical data set comprises perhaps several tens of such spectra (or "observations"). Hence, the number of variates far exceeds the number of observations. This is not a propitious situation for the application of multivariate statistical procedures, which require that the number of observations exceeds the number of variates. PCA offers a means of overcoming this difficulty. The variates in the original data set will normally be correlated with one another, to a greater or lesser extent. PCA removes this redundancy by transforming the original data into a set of new, uncorrelated variates, termed the principal component (PC) scores. In doing so, a rearrangement takes place, such that only the first few PC scores are required to describe the information contained in the many original variates. The number of significant PC scores is less than the number of observations, and multivariate methods (such as the calculation of Mahalanobis distances required in discriminant analysis) can proceed.

A further advantage of the reduction in variates is the resultant simplification of the data set, enabling easier visualization of relationships within the data. For example, in the PCAs performed in this paper, the first 2 PC scores accounted for 65–75% of the information content of the original data. Hence, important relationships between observations may be revealed in plots of these PC scores against one another. (In contrast, pairs of original variates account for much smaller percentages of the total information present in the data set and are usually correlated to some extent; plots of these are unlikely to yield any interesting findings.) Plots of PC scores have been used to explore data clustering in both the eight- and two-oil data sets.

Table 1. Oil Samples Used in the Experiment

type of oil	no. of samples	type of oil	no. of samples
extra virgin olive	10	groundnut	4
refined olive	8	grapeseed	4
sunflower	5	corn	3
rapeseed	4	walnut	3

Table 2. Composition of the Two-Oil Calibration and Validation Sets

	calibration set		validation set	
	no. of spectra	no. of samples	no. of spectra	no. of samples
extra virgin olive oil	32	7	13	3
refined olive oil	29	6	7	2
total	61	13	20	5

2. PROCEDURE

2.1. Samples. All oils used in this experiment were purchased from local retailers. The number of samples and their types are given in Table 1. All samples were stored at 1 °C and were allowed to warm to room temperature before thorough mixing and sampling.

2.2. Instrumentation. All spectra were collected on a Spectra-Tech (Applied Systems Inc.) MONITIR FTIR spectrometer system, equipped with room temperature infrared source, sealed and desiccated interferometer, and deuterated triglycine detector. An overhead attenuated total reflectance (ATR) accessory was built into one of two dedicated sampling stations. The accessory comprised transfer optics within a desiccated chamber sealed from the atmosphere by two potassium bromide windows. Through these windows the infrared radiation could be directed into the detachable ATR element. The element was a 10-reflection zinc selenide crystal mounted into a plate with a shallow trough for sample containment. The crystal geometry was a 45° parallelogram with mirrored angled faces. Due to the presence of the potassium bromide windows, the ATR

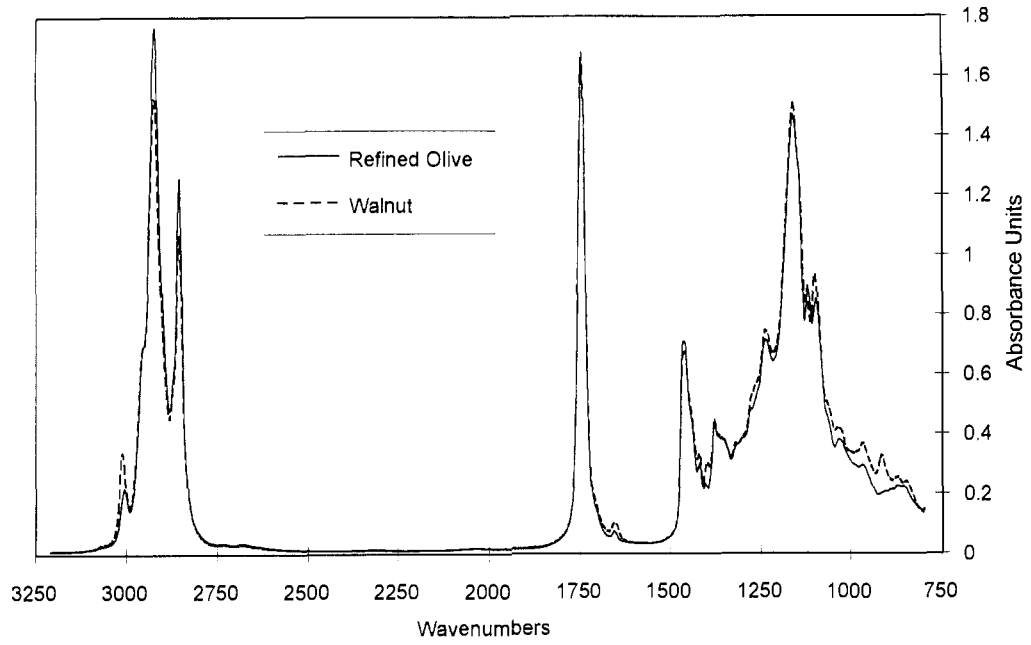

Figure 1. Example FTIR spectra of vegetable oils.

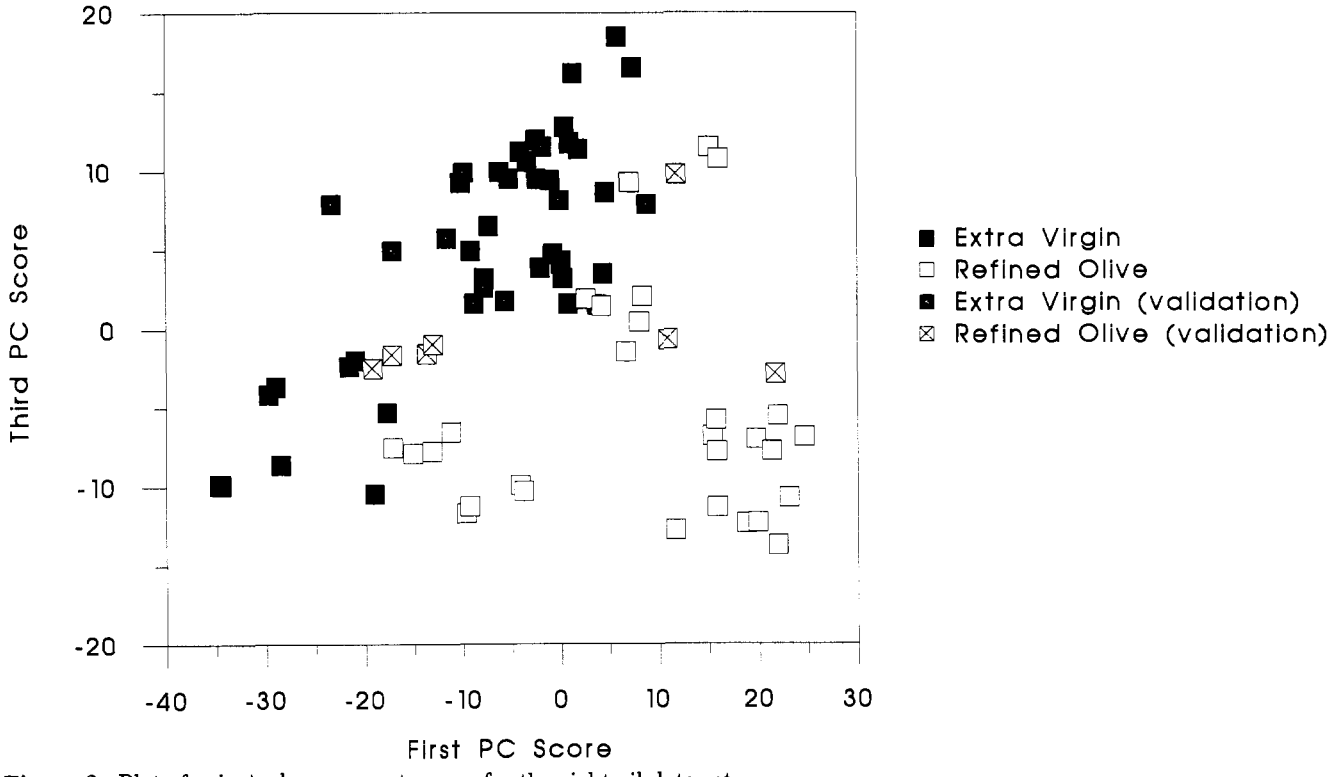

Figure 2. Plot of principal component scores for the eight-oil data set.

plate could be removed for cleaning without allowing the ingress of water vapor into the spectrometer.

2.3. Spectral Acquisition. Each sample (approximately 2 mL) was applied to the ATR plate by direct transfer from the bottle. Multiple spectra (between three and five) were collected of each sample, at intervals over a period of weeks. All spectra were recorded from 4800 to 800 cm^{-1} at a resolution of 4 cm^{-1}. For each spectrum, 256 interferograms were co-added before Fourier transformation and zero-filled to give a data point spacing of approximately 2 cm^{-1} in the frequency domain. Triangular apodization was employed, and each sample single-beam spectrum was ratioed to a single-beam spectrum of the clean ATR plate collected under identical conditions, before conversion into absorbance units. The ATR plate was thoroughly cleaned between each sample by removing the previous sample with tissue and cleaning with hexane, 2% Triton X-100, distilled water, and finally acetone. This procedure was found to efficiently remove all traces of oil from the ATR crystal.

2.4. Exploratory and Discriminant Analysis. All data analysis was carried out using SPIDA (Statistical Package for Interactive Data Analysis) software (Statistical Computing Laboratory, NSW, Australia); 560 data points were extracted from all spectra in two regions: 3100–2800 and 1800–1000 cm^{-1}. Two data matrices were constructed, the first comprising spectral data from all eight seed oils and the second from the extra virgin olive and refined olive oils only. The second data matrix was further divided into "calibration" and "validation" matrices, each containing spectra from randomly selected samples of each type of olive oil. The composition of the calibration and validation sets is given in Table 2. PCAs were carried out on the eight-oil data matrix and on the two-oil calibration matrix. Plots of the PC scores were constructed to investigate data clustering.

Discriminant analysis was applied to the PC scores from the two-oil data set only. The analysis comprised the following procedure:

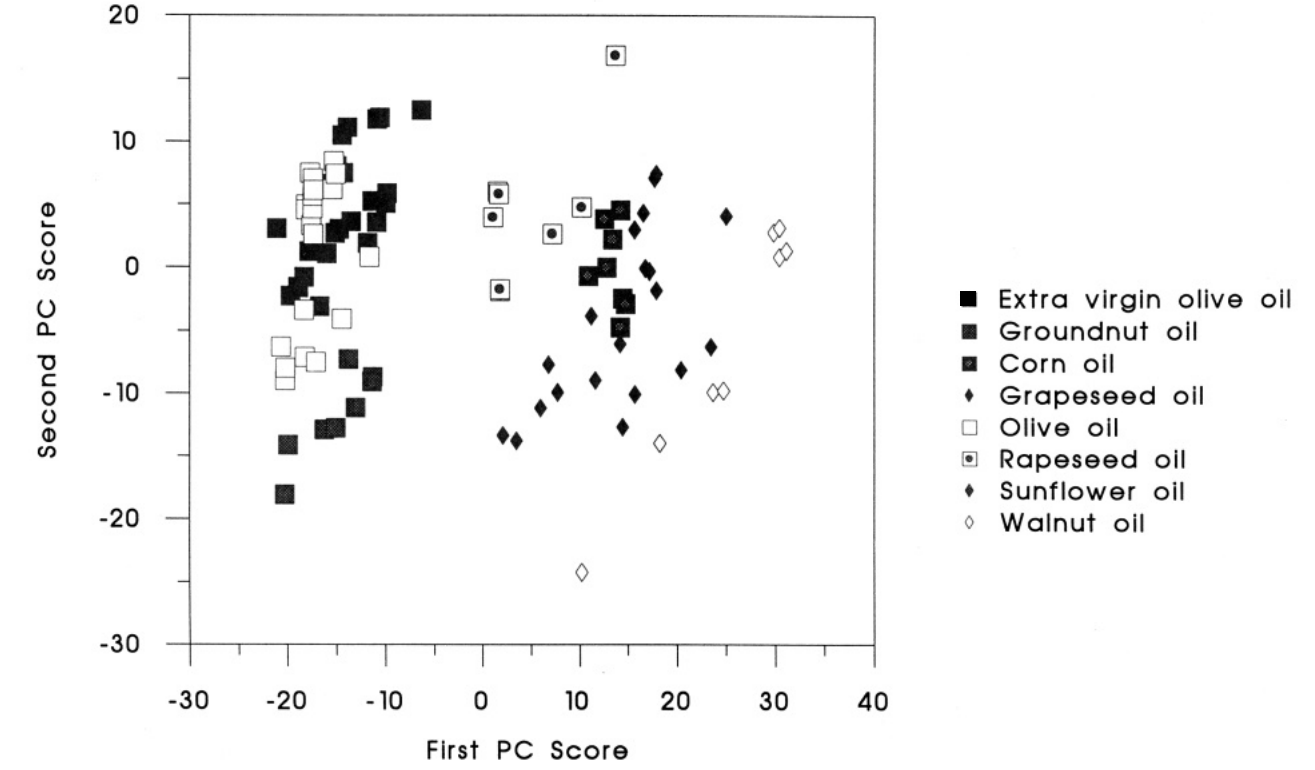

Figure 3. Plot of principal component scores for the two-oil data set.

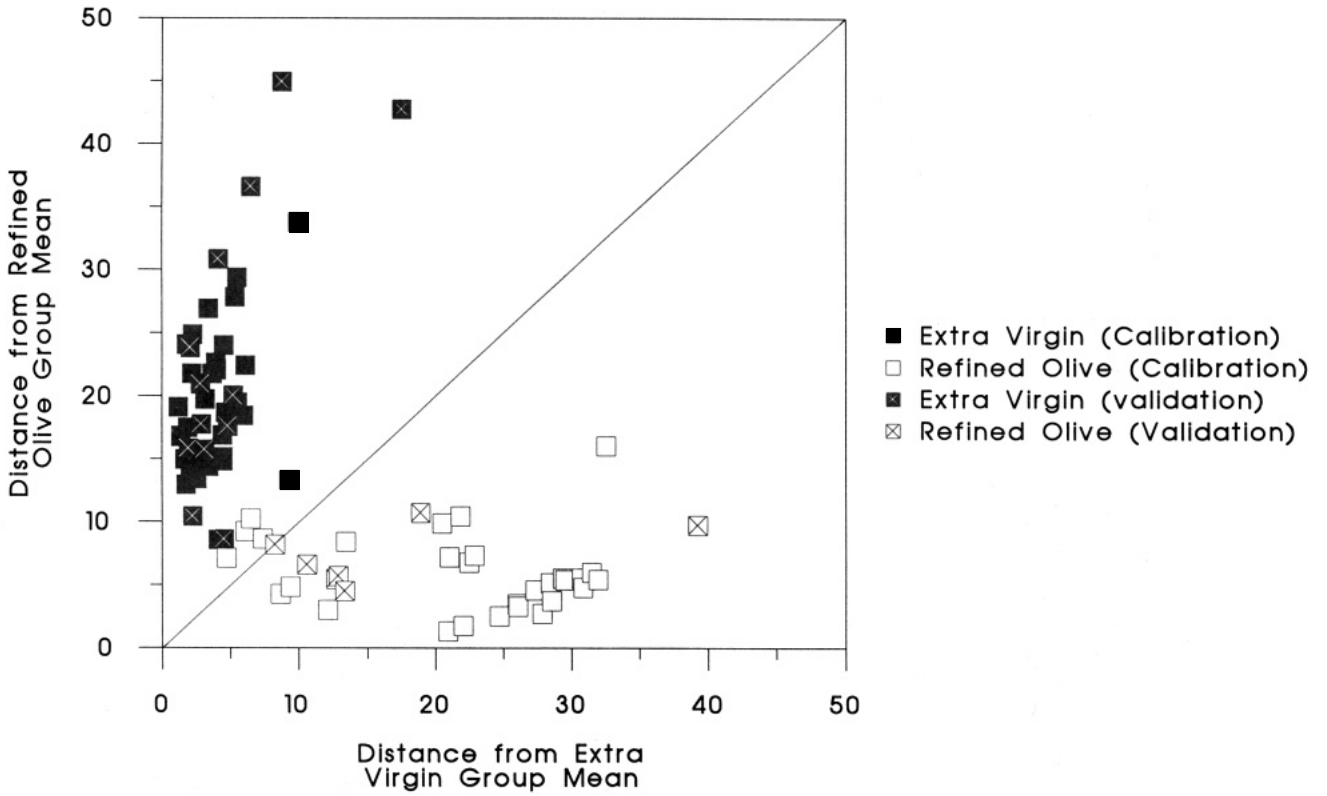

Figure 4. Plot showing results of discriminant analysis, as plots of squared Mahalanobis distances from each group mean.

(*i*) Each spectrum in the calibration matrix was assigned to a group according to its grade, "extra virgin" or "refined" olive oil.

(*ii*) The multivariate mean of the PC scores for each group was calculated. Only the first five PC scores were used; this ensured that the group sizes sufficiently exceeded the number of dimensions of the multivariate means.

(*iii*) The squared Mahalanobis distances of each observation in the calibration matrix from the two group mean observations were calculated.

(*iv*) Each observation was reassigned to the nearest group, on the basis of the calculated square Mahalanobis distance.

(*v*) The percentage of correct reclassification was compared with that obtained from randomly generated data (where no class structure exists) to determine whether the result was significant. This is a method of determining the successfulness of the discriminant model.

A second method was also used to evaluate the discriminant analysis: the model was applied to the independent spectral data contained in the validation matrix. The data in this matrix were transformed to PC space using the scaling constants and PC loadings determined from the PC analysis on the calibration set. Squared Mahalanobis distances from the established group means

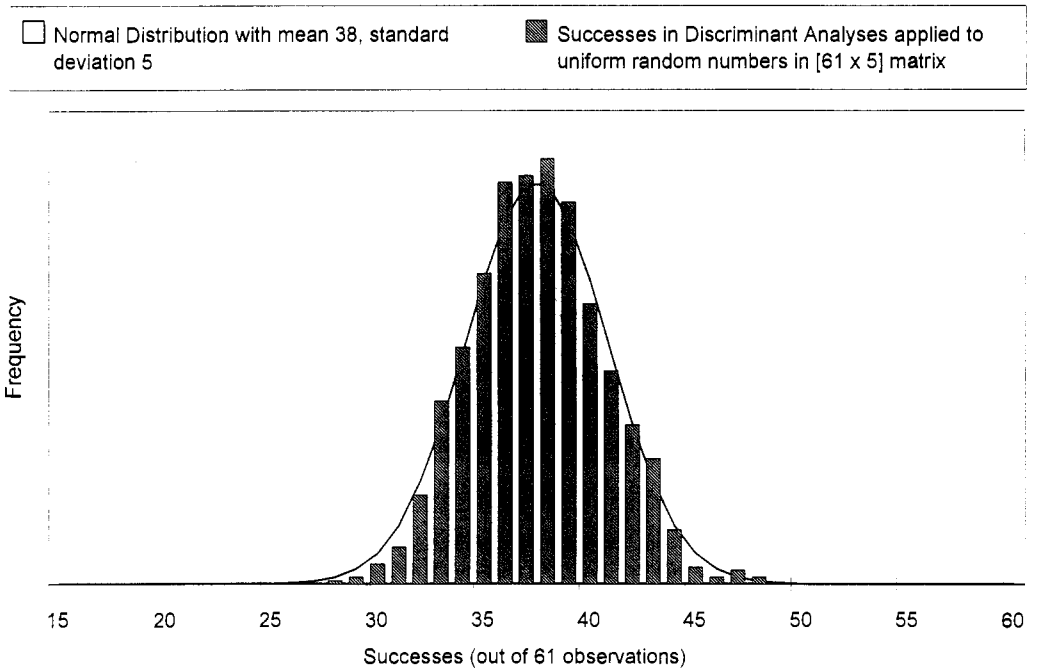

Figure 5. Histogram showing distribution of success rate of discriminant analyses applied to data with no underlying class structure.

were calculated for each validation spectrum's PC scores, and each validation observation was assigned to the nearest group. These predicted assignments were compared with the known classes to evaluate the efficacy of the discrimination.

3. RESULTS AND DISCUSSION

3.1. Spectra. Typical spectra are shown in Figure 1. The superimposed spectra of walnut and refined olive oil illustrate the high quality of the data obtained. Special regard should be paid to the high signal-to-noise ratio obtained, especially in the region around 1600 cm^{-1}. It is in this region that water vapor absorption will be seen in poorly purged, nondesiccated instruments. Early experiments with such equipment showed that spectral contamination from water vapor severely affected the ability to carry out discriminant analysis. It should be noted that the oils exhibit only minor spectral differences to the eye. Furthermore, most of the spectral information is contained in the regions 3100–2800 and 1800–1000 cm^{-1}, so that the use of data points extracted from these regions only in the PCA and discriminant analysis does not reject important data.

3.2. Exploratory and Discriminant Analysis. PCA of the complete oils data set showed that the first 12 PCs accounted for approximately 99% of the information in the original data matrix, with a good proportion (75%) accounted for by the first 2 PCs alone. A plot of the first two PC scores is shown in Figure 2. Grouping of the spectra according to oil type is already apparent. The olive oils are clustered closely together and are well separated from the other seed oils. There is a degree of overlap of some of the oils in this plot. However, a two dimensional plot of this kind may not reflect the true separation of the different oil types in PC space, as the dimensions representing the third, fourth, fifth, etc. PC scores may enhance the distances between oil types.

It is important to remember that the scatter of points on the plot represents variations not only between samples but also between replicate spectral acquisitions of individual samples at different times. Points representing replicate spectra tend to group together but are far from coincident. Moreover, the degree of the scatter within these groups of replicates is of the same order as that between samples. This implies that variations in the spectral data due to differences in oil type are so subtle that sampling of replicates (that is, sampling reproducibility) introduces variations of a comparable magnitude. It must be concluded that for a robust analytical method the calibration set should contain replicate spectra of individual samples obtained over a period of time, thereby building into the model variations due to factors such as instrumental drift and alignment, ambient temperature fluctuations, and aging of the sample.

The PCA on the calibration set of extra virgin and refined olive oils supported the evidence for clustering according to oil type. Plots of the first five PCs against one another revealed the most grouping, and these five scores were used for the subsequent discriminant analysis. An example of such a plot is shown in Figure 3. The data in the validation matrix were also transformed to PC space and are included in the plot.

Discriminant analysis was carried out on the two-oil calibration set, using the first five PC scores. Of the 61 spectra (from 13 samples), 57 were correctly classified by the discriminant model. These results are summarized pictorially in Figure 4. In a discriminant analysis of this kind, which uses the squared Mahalanobis distance metric, the number of observations must sufficiently exceed the number of variates (dimensions) of the observations or the condition of "over-fitting" can occur, in which a model calibrates well (in the extreme case, 100% correct classification occurs) but has no predictive ability. It can therefore be difficult to judge whether a discriminant model is effective on the basis of a calibration set alone. To assess the significance of the result, the same discriminant analysis procedure was applied repeatedly to [61 × 5] matrices of randomly generated data with no underlying class structure. The "success rate" was found to be approximately normally distributed, with a mean of 38 "correctly" classified and a standard deviation of 5 (Figure 5). Hence, the probability of 57 correct classifications occurring when there is no class structure present can be calculated: it was found to be 0.007%, that is, extremely unlikely. Therefore, the discriminant analysis results are strong evidence of spectral grouping according to oil type.

The four incorrectly classified spectra in the calibration set were from the replicate measurements on a single refined olive oil sample. This is not surprising, since oils sold in supermarkets as olive oil are often mixtures of refined and virgin oils. Consequently, some of the samples assigned to the refined olive set may have a degree of extra virgin character and may contain chemical constituents associated with both of the defined group types. Discrimination analysis should not then be expected to be as successful as if pure, well-characterized samples were available.

Further evidence of the ability to discriminate between the two oil types is gained from analysis of the independent validation set. The results of applying the discrimination model to the 20 spectra in the validation matrix are also shown in Figure 4.

Conclusions. Ninety-three percent of spectra in the two-oil training set and 100% in the validation set were correctly classified according to type. It is interesting to note that these oils, which are chemically the most similar and are the most closely clustered in the exploratory PC analysis, can be almost completely discriminated using this technique. As these two types represent the most challenging set for discriminant analysis, the implication must be that discrimination between these oils and other seed oils should be even more robust. Successful discriminant analysis is the first step toward producing a method that not only correctly classifies but also reliably detects adulterated samples. To produce a reliable robust method, the database of samples and spectra must be increased considerably. Equally importantly, the database must be constructed from samples whose origin and composition are determined absolutely. (The samples used in this experiment were commercially obtained.) This work will continue using such samples and will be expanded to detect and, if possible, quantify adulteration.

In conclusion, the FTIR method clearly has the potential of becoming an easily employed, rapid method for the detection of adulteration of oils. Once the database is established and the model developed, each individual analysis can be achieved in a matter of minutes, offering considerable savings in time and costs compared to alternative methods.

ACKNOWLEDGMENT

This work was, in part, conceived as part of the European Community FLAIR QUEST concerted action. Y.W.L. thanks the government of Malaysia for financial support.

LITERATURE CITED

Chapman, D. Infrared spectroscopy of lipids. *J. Am. Oil Chem. Soc.* **1965**, *42*, 353–371.

Chenery, D. H.; Sheppard, N. An evaluation of the practical performance of a Digilab FTS-14 Fourier transform infra-red interferometer working in the region of 4000 to 400 cm^{-1}. *Appl. Spectrosc.* **1978**, *32*, 79–89.

Coates, J P.; D'Agostino, J. M.; Friedman, C. R. Quality control analysis by infrared spectroscopy. Part I: Sampling. *Int. Lab.* **1987a**, May, 70–77.

Coates, J. P.; D'Agostino, J. M.; Friedman, C. R. Quality control analysis by infrared spectroscopy. Part II: Practical Applications. *Int. Lab.* **1987b**, June, 58–65.

Consiglieri, C.; Bruschi, R. Origin and provenance of commercial extra virgin olive oil. Chemometric applications. *Ind. Aliment. (Pinerolo, Italy)* **1992**, *31* (306), 643–648.

Firestone, D.; Summers, J. L.; Reina, R. J.; Adams, W. S. Detection of adulterated and misbranded olive oil product. *J. Am. Oil Chem. Soc.* **1985**, *62*, 1558–1562.

Firestone, D.; Carson, K. L.; Reina, R. J. Update on control of olive oil adulteration and misbranding in the United States. *J. Am. Oil Chem. Soc.* **1988**, *65*, 788–792.

Forina, M.; Armanino, C.; Lanteri, S.; Tiscornia, E. Classification of olive oils from the fatty acid composition. In *Food Research and Data Analysis*; Martens, H., Russuwurm, H., Eds; Applied Science: London, 1983; pp 189–214.

Hirschfeld T., FTIR now and in the future. Part I. *Eur. Spectrosc. News* **1983**, *51*, 13–18.

Hirschfeld, T. FTIR now and in the future Part II. *Eur. Spectrosc. News* **1984**, *55*, 15–21.

International Olive Oil Council (IOOC). "International trade standards applying to olive oils and olive-residue oils"; COI/T.15/NC NO I; IOOC, Madrid, 1984.

Jimeno, S. A. The Spanish toxic symptoms. *Trends Anal. Chem.* **1982**, *1*, 4–6.

Kapoulas, V. M.; Andrikopoulos, N. K. Detection of virgin olive oil adulteration with refined oils by second derivative spectrophotometry. *Food Chem.* **1987**, *23*, 183–192.

Kapoulas, V. M.; Passaloglou-Emmanouilldou, S. Detection of adulteration of olive oil with seed oils by a combination of column and gas chromatography. *J. Am. Oil Chem. Soc.* **1981**, *58*, 694–697.

Kochlar, S. P.; Rossell, J. B. The Spanish toxic oil syndrome I. *Nutr. Food Sci.* **1984**, *90*, 14–15.

Kochlar, S. P.; Rossell, J. B. Analysis of oils and fats using infrared spectroscopy. *Int. Anal.* **1987**, *1*, 23–26.

Lanzon, A.; Cert, A.; Albi, T. Detection of refined olive oil in virgin olive. *Grasas Aceites* **1989**, *40* (6), 385–388.

Marini, D., Grassi, L., Balestrieri, F.; Pascucci, E. Spectrophotofluorometric analysis of olive oil. *Riv. Ital. Sostanze Grasse* **1990**, *67* (2), 95–99.

Morchio, G., Di Bello, A., Mariani, C.; Fedeli, E. Detection of refined oils in virgin olive oil. *Riv. Ital. Sostanze Grasse* **1989**, *66* (5), 251–257.

Osborne, B. G.; Fearn, T. *Near Infrared Spectroscopy in Food Analysis;* Longman: Harlow, U.K., 1986.

Passaloglou-Emmanouillidou, S. A comparative study of UV spectrophotometric methods for the detection of olive oil adulteration by refine oils. *Z. Lebensm. Unters. Forsch.* **1990**, *191* (2), 132–134.

Peipponen, S., Vainionpaa, A.; Roos A. Classification of some food oils. In *Food Research and Data Analysis*; Martens, H., Russuwurm, H., Eds.; Applied Science: London, 1983; p 426.

Poutet, M. Procedure for detecting the falsification of olive oils with that of other seeds. *Ann. Chem. Phys.* **1819**, *12* (2), 58.

Sacchi, P.; Addeo, F.; Giudicianni, I.; Paolillo, L. Application of carbon-13 NMR to the determination of mono- and diglycerides and free fatty acids in virgin and refined olive oil. *Riv. Ital. Sostanze Grasse* **1990**, *67* (5), 245-252.

Sanchis Rodriguez, J.; Rodriguez Serrano, J. Rapid HPLC procedure for the detection of adulteration of olive oil by seed oils. *Alimentaria (Madrid)* **1991**, *28* (224), 27–29.

Tsimidon, M., Macrae, R.; Wilson, I. Authentication of virgin olive oil using principal component analysis of triglycerides and fatty acid profiles. Part I: Classification of Greek Olive Oil. *Food Chem.* **1987a**, *25*, 227–239.

Tsimidon, M.; Macrae, R.; Wilson, I. Authentication of virgin olive oil using principal component analysis of triglycerides and fatty acid profiles. Part II: Detection of adulteration with other vegetable oils. *Food Chem.* **1987b**, *25*, 251–258.

Williams, P. C.; Norris, K. H. In *Near Infrared Technology in the Agricultural and Food Industries*; American Association of Cereal Chemists: St. Paul, MN, 1987; p 330.

Wilson, R. H. Fourier transform mid-infrared spectroscopy for food analysis. *Trends Anal. Chem.* **1990**, *9*, 127–131.

World Health Organization. "Recommended International Standard for Olive Oil, Virgin and Refined, and for Refined Olive-Residue Oil"; Codex Alimentarius Commission CAC/RS 33; Rome, Italy, 1970.

Received for review November 2, 1993. Accepted January 28, 1994.[⊗]

[⊗] Abstract published in *Advance ACS Abstracts,* April 1, 1994.