JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

J. Agric. Food Chem. **2003,** *51,* 5871−5876 **5871**

Dietary Supplement Oil Classification and Detection of Adulteration Using Fourier Transform Infrared Spectroscopy

BANU F. OZEN, ILAN WEISS, AND LISA J. MAUER*

Food Science Department, Purdue University, 745 Agriculture Mall Drive, West Lafayette, Indiana 47907

Fourier transform infrared spectroscopy (FT-IR) methods and common chemometric techniques [including discriminant analysis (DA), Mahalanobis distances, and Cooman plots] were used to classify various types of dietary supplement oils (DSO) and less expensive, common food oils. Rapid FT-IR methods were then developed to detect adulteration of DSO with select common food oils. Spectra of 14 types of DSO and 5 types of common food oils were collected with an FT-IR equipped with a ZnSe attenuated total reflectance cell and a mercury cadmium telluride A detector. Classification of DSO and some common food oils was achieved successfully using FT-IR and chemometrics. Select DSO were adulterated (2−20% v/v) with the common food oils that had the closest Mahalanobis distance to them in a Cooman plot based on the DA analysis, and data were also analyzed using a partial least-squares (PLS) method. The detection limit for the adulteration of DSO was 2% v/v. Standard curves to determine the adulterant concentration in DSO were also obtained using PLS with correlation coefficients of >0.9. The approach of using FT-IR in combination with chemometric analyses was successful in classifying oils and detecting adulteration of DSO.

KEYWORDS: Dietary supplement oils; Fourier transform infrared spectroscopy; FT-IR; adulteration

INTRODUCTION

Prior to 1994, the U.S. Food and Drug Administration regulated most dietary supplements to ensure they were safe, wholesome, and truthfully labeled. However, with the passage of the Dietary Supplements Health and Education Act (DSHEA) of 1994, dietary ingredients used in dietary supplements are no longer subject to safety evaluations required of other new food ingredients or new uses of old food ingredients. The intent of the DSHEA was to ensure that safe and appropriately labeled dietary supplements remain available to the millions of consumers who believe they provide health benefits, and the dietary supplement market has grown steadily since 1994. Currently, the dietary supplement industry has multibillion dollar annual sales.

Dietary supplement oils (DSO) are a significant part of the growing dietary supplement industry. Recent patents on several of these oils have been filed (*1*−*3*). Health claims, both scientifically based and folklore, related to dietary supplement grapeseed, flaxseed, borageseed, and evening primrose oils indicate that they are essential for maintaining good health (*4*); necessary for energy metabolism, cardiovascular, and immune health (*5*); have anti-inflammatory properties, prevent the formation of blood clots, and help to keep cell membranes flexible (*6*); are a remedy for high cholesterol (*7*); abd prevent coronary heart disease and stroke, autoimmune disorders (lupus and nephropathy), cancers of the breast, colon, and prostate, mild hypertension, and rheumatoid hypertension (*8*−*12*).

According to The Hartman Group, a consulting firm specializing in natural products, the total retail sales of flaxseed oil in 1999 was $58 million (*13*). The estimated U.S. market in 1996 for grapeseed and evening primrose oils alone was over $17 million dollars (*14*). Additionally, in 1998 one of the top two therapeutic categories in terms of growth was “other therapeutic goods”, which included evening primrose oil (*15*). With the market for these specialty oils increasing on an annual basis, the regulation and detection of adulterated products as well as the ability to monitor and ensure quality are necessary to sustain consumer confidence.

Authenticity is a quality criterion for foods and food ingredients that is attracting increased attention as consumers agree to pay higher prices for products labeled as organic or 100% natural, as well as for those with perceived health benefits. Methods of food adulteration have become more sophisticated in recent years, and traditional methods used in the detection of adulterants are getting more expensive and time-consuming as adulterators find ways to avoid detection. Not only is detection of adulteration needed to protect consumer bank-balances and uphold truth-in-labeling laws, but also it is needed to protect consumer health and ensure product quality.

Conventional methods for detecting oil adulteration involve hydrolysis and methylation of the resulting fatty acids prior to analysis and are therefore time-consuming and destructive. Also, information associated with the location of the fatty acids on

* Author to whom correspondence should be addressed [e-mail mauer@foodsci.purdue.edu; telephone (765) 494-9111; fax (765) 494-7953].

10.1021/jf034245h CCC: $25.00
Published on Web 08/23/2003

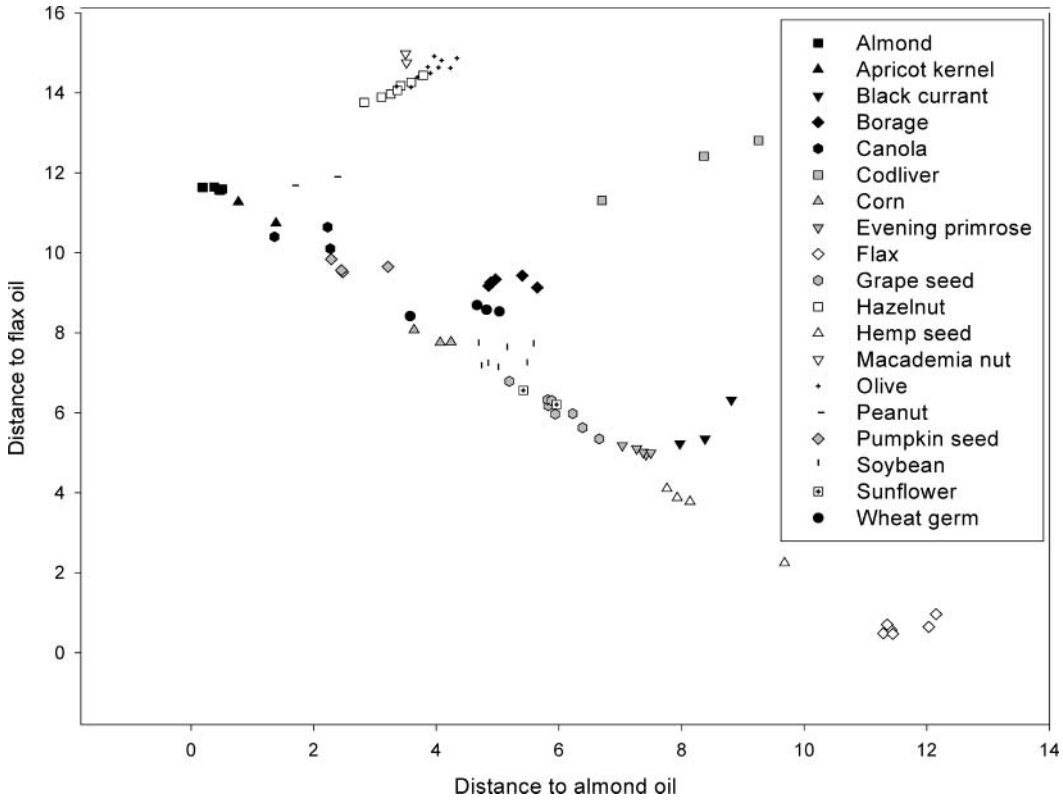

Figure 1. Cooman plot for the classification of DSO and common food oils with 3050–2775, 1780–1630, and 1500–650 cm^{-1} regions used for DA.

the original glycerol backbone is lost. The most common chromatographic methods for detecting oil adulterations are gas chromatography (*16*−*19*), high-performance liquid chromatography (*20*), and gas−liquid chromatography (*21*). Other analytical methods include nuclear magnetic resonance (*22*, *23*), a spectrofluorometric method (*24*), UV spectroscopy (*25*), and a second-derivative spectrometry method (*26*). Gas chromatography methods were also used in the detection of pumpkinseed oil adulteration (*27*, *28*). However, either most of these methods require time-consuming sample preparation steps prior to analysis or the analysis takes more time than analysis using Fourier transform infrared spectroscopy (FT-IR).

Recent studies (*29*, *30*) have shown that FT-IR, a type of mid-infrared (MIR) spectroscopy, is a rapid, nondestructive, authentication tool capable of detecting the adulteration of various oils using common chemometric techniques such as discriminant analysis (DA). Adulteration detection levels can be as low as 2% (*30*, *31*). Other examples of adulteration screening using MIR spectroscopic analysis include authentication of coffee, fruit purees, honey, and meats (*32*). MIR is a well-established analytical tool in the chemical industry and forensic sciences, and its use for rapid food analysis techniques has great promise. An infrared spectrum contains features arising from vibrations of molecular bonds, and the MIR region (4000−400 cm^{-1}) is highly sensitive to the precise composition of the sample being analyzed (*33*). Data obtained from FT-IR absorption spectra will provide information on numerous compounds, including quantitative, qualitative, physical, and chemical information related to individual components. Recent FT-IR instrumentation and multivariate statistical analysis techniques (chemometrics) allow for the detection of constituents present in very low concentrations (as low as 0.0003%) as well as subtle compositional differences between and among multiconstituent specimens (*34*).

The objectives of this study were to use FT-IR spectroscopy and multivariate statistical procedures to (1) classify different types of DSO and less expensive common food oils and (2) develop a rapid method and determine detection limits for the detection of adulterated DSOs with the less expensive food oils.

MATERIALS AND METHODS

Samples. At least two varieties of each DSO (almond, apricot kernel, black currant, borage, cod liver, evening primrose, flaxseed, grapeseed, hazelnut, hempseed, macadamia nut, olive, pumpkinseed, and wheat germ) and common food oil (canola, corn, peanut, soybean, and sunflower) were obtained from local grocery and health food stores and Internet suppliers. Blends of each selected DSO (borage, evening primrose, flaxseed, grapeseed, and pumpkinseed) from different sources and different brands were adulterated with blends of selected common food oils at levels of 2, 5, 10, 15, and 20% (v/v). This range was selected on the basis of industry reports identifying 2−20% as the target for detection (*35*) and results from previous studies (*30*, *31*). Infrared spectra of pure oil samples and adulterated samples were obtained at least in duplicate, and each point in **Figure 1** represents the averages of these repetitions for each source of oil (e.g., the averages of duplicate spectra for each type of six sources of flax oil are shown by the six diamond points in the lower right-hand corner of **Figure 1**).

Instrumentation. All infrared spectra were acquired using a ThermoNicolet Nexus 670 FT-IR spectrometer (ThermoNicolet Analytical Instruments, Madison, WI) equipped with a mercury cadmium telluride A (MCTA) detector and KBr optics. Measurements were performed by using 128 scans at 4 cm^{-1} resolution with a ZnSe single-bounce attenuated total reflectance (ATR) accessory.

Statistical Analysis. Analysis of the data was performed by common classical multivariate procedures, including discriminant analysis (DA) and partial least-squares (PLS) analysis, using TQ Analyst software (ThermoNicolet). DA was used for the classification of the samples. The DA enables the separation of the oil spectra based on principal component (PC) analysis or total urea under the spectra in the spectral regions defined by the researcher (*36*). The TQ Analyst program selects the principal components that explain the spectral variability in the defined spectral regions. Then a Cooman plot can be constructed using the TQ Analyst software by plotting the Mahalanobis distance between the oils using either principal components or different oil categories

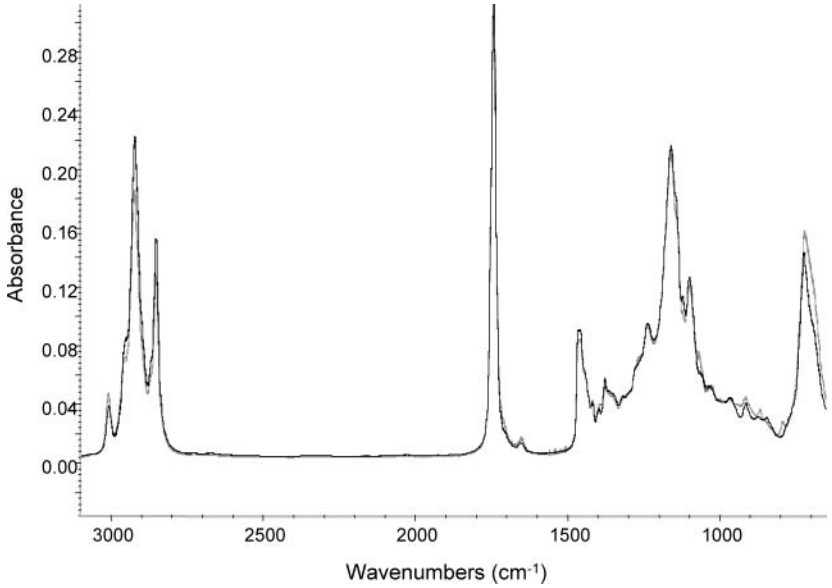

Figure 2. FT-IR spectra of flaxseed (gray line) and sunflower (black line) oils in the 3100–650 cm^{-1} region.

Table 1. Number of Samples Used To Construct PLS Standard Curves and Calibration and Validation Standard Errors of Prediction

sample	no. of samples	no. of validation samples	no. of PLS factors	R^2	RMSEC[a]	RMSEP[b]
sunflower oil in flaxseed oil	15	4	1	0.96	1.79	1.36
sunflower oil in borage oil	15	4	3	0.97	1.51	2.12
sunflower oil in evening primrose oil	15	4	4	0.97	1.83	2.57
sunflower oil in grapeseed oil	16	4	5	0.98	1.28	1.79
canola oil in pumpkinseed oil	14	5	8	0.99	0.155	6.39

[a] Root-mean-square error of calibration. [b] Root-mean-square error of prediction.

(as done in this project) on the axes. The Mahalanobis distance is a useful description of the similarity between samples and represents standard deviations from the mean of one set of samples to another sample. Samples that are further apart on a Cooman plot have greater differences than samples that are close together on the plot (as shown in **Figure 1**). The DA method classifies the calibration and validation standards, which are specified by the researcher. The TQ Analyst calculates boundaries for each class based on the number of standards in the class. The software reports a given standard as misclassified only if there are fewer differences between the standard and the incorrect class than there are between the standard and its correct class (i.e. there was a smaller Mahalanobis distance to the incorrect class than to the correct class).

Spectral regions where variations were observed (3050−2775, 1780−1630, and 1500−650 cm^{-1} regions in **Figure 2**) were chosen for developing DA and PLS models to classify oils and for quantifying levels of adulteration, respectively. For PLS analysis, the number of calibration samples used was at least 20% of the number of samples used for model building (**Table 1**). Several diagnostics including eigen analysis, cross-validation, and predicted residual error sum of squares (PRESS) were run to check the validity of the developed models.

RESULTS AND DISCUSSION

Classification of various DSO and regular oils was performed by DA using 3050−2775, 1780−1630, and 1500−650 cm^{-1} regions. These regions include the portions of the IR spectra where peaks were observed, and regions that were not included did not have peaks (refer to **Figure 2**). **Figure 1** shows the classification of oils using a Cooman plot. A total of 14 different classes of DSO and 5 common food oils are represented in this figure. The oils shown in **Figure 1** have different fatty acid compositions (**Table 2**), which produce different FT-IR spectra. Chemometric analysis of these spectra (using TQ Analyst software) generated a Cooman plot by plotting the Mahalanobis distance between the spectra of the oils. The Cooman plot (**Figure 1**) shows that nut oils with olive and canola oils, which are very rich in oleic acid (18:1) (**Table 2**), are clustered in the upper left corner of the plot. Olive and hazelnut oils are close to each other in the plot, in some cases overlapping. According to another study, olive and hazelnut oils have very similar compositions and spectra, and detection of adulteration of olive oil with hazelnut oil is possible only at concentrations >20% using FT-IR techniques (*31*). The lower right corner of the Cooman plot (**Figure 1**), on the other hand, contains oils (flaxseed, hempseed, and black currant oils) with high α-linolenic acid (18:3ω3) concentrations. Flaxseed oil is also separated from hempseed and black currant oils. Hempseed, black currant, and evening primrose oils contain significant amounts of γ-linolenic acid and form a cluster near the middle of the plot. The middle part of the Cooman plot (**Figure 1**) contains oils such as wheat germ, corn, and soybean oils that have high concentrations of linoleic and oleic acids in their structures. In addition to linoleic and α-linolenic acids, cod liver oil contains polyunsaturated fatty acids such as 20:5, 22:5, and 22:6, which the vegetable, seed, and nut oils used in this study do not contain. Because of this compositional difference, cod liver oil is separated from the rest of the oils in the upper right corner of the Cooman plot (**Figure 1**).

In addition to differences between types of oils, slight differences between different samples of the same type of oil are apparent on the Cooman plot (e.g., there are relatively small distances between the six types of flax oil shown in the lower right-hand corner of **Figure 1**). These differences can be attributed to natural variations in fatty acid composition between

Table 2. Fatty Acid Composition of Oils Used in the Current Study (Data Are Compiled from References *37–40*)

oil	palmitic 16:0	stearic 18:0	oleic 18:1	linoleic 18:2	linolenic 18:3ω3	γ-linolenic 18:3ω6
almond	4–13	0–10	43–60	20–34		
apricot kernel	5–7	1–3	72–84	6–22	<1	
black currant	6–7	1–2	9–11	45–60	12–15	15–19
borage	11	4	16.5	37	<1	23
canola	4	2	64	19	9	
codliver	10.6	2.8	20.7	0.94	0.94	
corn	12	2	27	57		
evening primrose	4–12	1–7.5	4–12	65–72	0	3–15
flaxseed	4.9	5.2	23.7	15.2	50.1	0
grapeseed	5–11	3–6	12–28	58–78	0.4	
hazelnut	5–7	1–3	72–84	6–22	<1	
hempseed	6–9	2–3	10–16	50–70	15–25	1–6
macademia nut		12	71	10	10	
olive	8–20	1–5	55–83	4–21	<1	
peanut	8–14	2–4	36–67	14–43	trace	
pumpkinseed	16	5	24	54	0.5	
soybean	10–13	3–5	18–25	50–57	5–10	
sunflower	5–8	2–7	13–40	40–74	<1	
wheat germ	12–19	<3	14–23	50–56	3–7	

the samples due to the origin of the oil, plant variety, and processing conditions, as well as differences in sample handling, changing environmental conditions, and spectrometer drift during analysis. In some cases, these variations will result in misclassification of oils, such as the misclassification problems encountered for wheat germ and apricot kernel oils used in this study. One of the wheat germ oil samples was misclassified by the TQ Analyst program as corn oil, and one of the canola oils was misclassified as apricot kernel oil. This misclassification problem should be expected due to compositional similarities between these oils and variations based on the origin of the oil (species, soil, and area of cultivation); however, misclassifications could be reduced by increasing the number of samples used for each type of oil.

The next step in this study was to identify the common food oils with the closest Mahalanobis distance (i.e., those most structurally similar and exhibiting similar spectra) to select DSO, as shown in **Figure 1**. The logic for this was that common food oils which are the most similar to the DSO could potentially be used for the adulteration of these oils, a problem associated with hazelnut oil adulteration of olive oil (*35*). Successful detection of adulteration with these structurally similar common food oils will most likely mean that adulteration with other types of oils with greater structural differences could also be detected using FT-IR and chemometric techniques. Selected DSO samples for adulteration were borage, evening primrose, flaxseed, grapeseed, and pumpkinseed oils. As shown in **Figure 1**, canola oil had the least distance to pumpkinseed oil, whereas the other DSO were closest to sunflower oil. Therefore, pumpkinseed oil was adulterated with canola oil, whereas sunflower oil was used in the adulteration of the other DSO. The selected adulterant oil was added to the DSO at concentrations of 2−20% v/v, FT-IR spectra were collected, and chemometric methods were used to analyze the data.

For data analysis of adulterated flaxseed oil spectra, the same regions used for the classification of all the oils (**Figure 1**) were used again because these encompassed the differences in the spectra of flaxseed and sunflower oils (3050−2775, 1780−1630, and 1500−650 cm^{-1} regions). **Figure 2** shows the spectra of pure flaxseed and sunflower oils in the 3100−650 cm^{-1} region. The peaks in the 3050−2800 cm^{-1} region result from C−H stretching vibrations, whereas the large peak around 1740 cm^{-1} is due to C═O vibration. Different forms of C−H and C−O vibrations cause the appearance of peaks in the 1500−650 cm^{-1} region for oils. Differences between the spectra of flaxseed and sunflower oils in the 3050−2800 cm^{-1} (**Figure 3**) and 1780−1630 cm^{-1} (**Figure 4**) regions were due to the differences in the intensities of the peaks in these regions. In the 1500−650 cm^{-1} region, on the other hand, there are shifts in the peaks, changes in peak intensities, and new peaks (**Figure 4**). For example, the peak at 1070 cm^{-1}, which corresponds to C−O stretching, is very clear for flaxseed oil, whereas it is just a shoulder for sunflower oil (**Figure 4**). Shifting of the peaks in

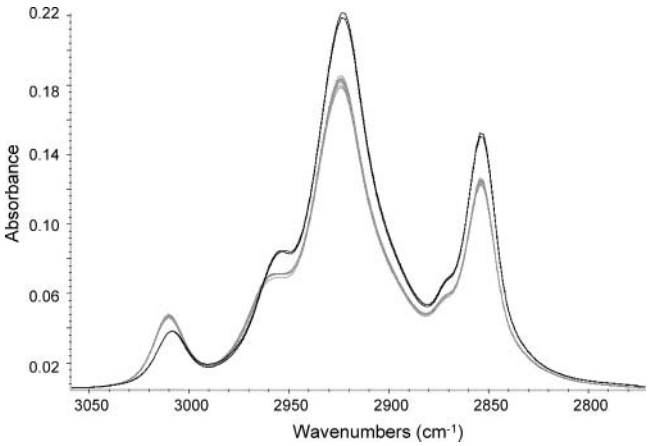

Figure 3. FT-IR spectra of flaxseed (gray line) and sunflower (black line) oils in the 3050–2800 cm^{-1} region.

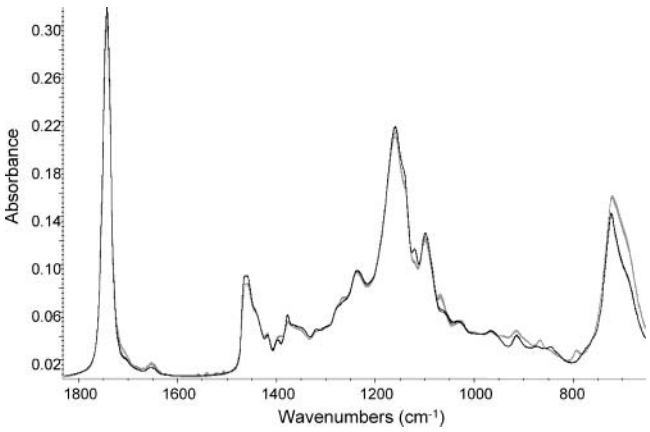

Figure 4. FT-IR spectra of flaxseed (gray line) and sunflower (black line) oils in the 1800–650 cm^{-1} region.

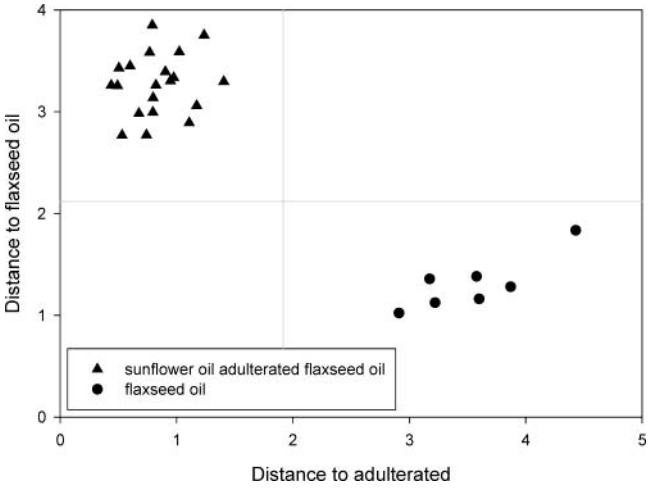

Figure 5. Cooman plot for the classification of pure flaxseed oil and sunflower oil adulterated flaxseed oil (vertical and horizontal lines show 95% confidence interval).

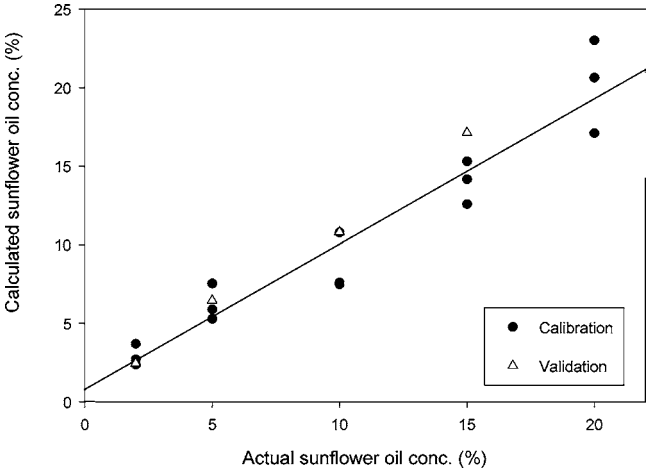

Figure 6. PLS calibration curve for sunflower oil adulterated flaxseed oil.

the 980−780 cm^{-1} region was also observed for flaxseed and sunflower oil spectra, and an additional peak at 793 cm^{-1} (C−H vibration, phenyl ring substitution) formed for flaxseed oil, which does not exist in sunflower oil spectra (**Figure 4**). These differences in spectra result from compositional differences between the oils.

Results of DA using the specified regions showed that detection of adulteration of flaxseed oil with sunflower oil could be achieved successfully even at a 2% level (**Figure 5**). Eigen analysis defines how much variation principal components explain in the spectra. For this case, eigen analysis revealed that it is possible to obtain 99% of the desired information with six principal components. The data were also analyzed by PLS to construct a calibration curve. **Figure 6** shows the concentration values obtained from the PLS model versus the actual concentration of sunflower oil in flaxseed oil. Differences between the actual adulteration concentration and the calculated adulteration concentration obtained from the model were very small, and the correlation coefficient, R^2, was calculated as 0.96 (**Table 1**). Several diagnostics (cross-validation, PRESS, and eigen analysis) were run to validate the developed model. Cross-validation was performed by removing one standard at a time, and a final R^2 value of 0.89 was obtained. Also, PRESS values were calculated for different principal component factors. This diagnostic shows how the PRESS value changes as the number of factors used to calibrate each component in the active PLS method is increased. PRESS values and the eigen analysis suggested that one of the principal components was enough to extract 99% of the desired information used for detecting the adulteration. These analyses indicated that the developed FT-IR method and chemometric analysis were very useful for quantifying sunflower oil added to flaxseed oil at 2−20% v/v.

For the adulteration of borage, evening primrose, and grapeseed oils with sunflower oil study, the same spectral regions (3050−2775, 1780−1630, and 1500−650 cm^{-1}) were used for analyzing spectra using the chemometric methods. However, only the 1500−650 cm^{-1} region was used for the analysis of canola oil adulterated pumpkinseed oil spectra because the differences in the canola oil and pumpkinseed oil spectra were observed only in this region. Canola and pumpkinseed spectra are very similar to each other, and this similarity also could be seen by the small distances between these oils in **Figure 1**. The major differences between the spectra of canola and pumpkinseed oils were in the intensities of the peaks at 872, 912, and 965 cm^{-1}, which correspond to out-of-plane bending in combination with C═C bonds. For the adulteration of borage, evening primrose, and grapeseed oils with sunflower oil as well as the canola oil adulteration of pumpkinseed oil, DA classified 100% of all samples accurately either as the pure DSO or as the adulterated oil even at a 2% adulteration level (figures are not shown but are similar to **Figure 5**). PLS was also used to construct the standard curves (similar to **Figure 6**), and good fits with R^2 values >0.9 were obtained for the models developed with PLS (**Table 1**). Again, these analyses indicated that the developed FT-IR method and chemometric analysis were very useful for quantifying the adulterant oil added to the DSO at 2−20% v/v, and future work in this area could investigate lower detection limits at <2% v/v.

In summary, 100% success was obtained in the classification of adulterated and pure oil samples, and the detection limit for adulteration was 2% v/v for the oils used in this study. On the basis of this success, future work could investigate lower detection limits for adulteration scenarios of interest. Considering that data collection using the FT-IR methods took <5 min, FT-IR methods could be a helpful tool for the rapid and accurate detection of the adulteration of dietary supplement oils by common, less expensive oils. **Figures 1** and **5** also demonstrate how FT-IR and chemometric techniques can be used to classify pure oils and differentiate between types of oils on the basis of fatty acid and other structural differences.

ACKNOWLEDGMENT

We thank Yashodar Burgula for editorial advice.

LITERATURE CITED

(1) Gibson, R.; Regtop, H.; Bode, O.; Drummond, C.; Newman, M.; Drummond, G. Supplement for baby infant formula and a method of delivering that supplement. Int. Patent Appl. AU 95-1378, 1996.

(2) Plummer, N. Nutritional supplement. U.K. Patent Appl. GB2324457A, 1998.

(3) Veret, A. A new food supplement formulation comprising four vegetable oils (borage, evening primrose, wheat germ and grapeseed oils. Fr. Patent Appl. FR2794941A1, 2000.

(4) Omega Nutrition. *Borage and Flax Oil Products*; 2001; www.omegaflo.com/products/.

(5) Allman, M. A.; Pena, M. M.; Pang, D. Supplementation with flaxseed oil versus sunflowerseed oil in healthy young men consuming a low fat diet: effects on platelet composition and function. *Eur. J. Clin. Nutr.* **1995**, *49*, 169−178.

(6) Hanrahan, C. *Borage Oil. Gale Encyclopedia of Alternative Medicine*; 2001; www.findarticles.com/cf_dls/g2603/0002/2603000221/print/jhtml.
(7) Cunnane, S. C.; Ganguli, S.; Menard, C.; Liede, A. C.; Hamadeh, M. J.; Chen, Z.; Wolever, T. M. S.; Jenkins, D. J. A. High α-linolenic acid flaxseed (*Linum usitatissimum*): some nutritional properties in humans. *Br. J. Nutr.* **1993**, *69*, 443−453.
(8) Fan, Y.; Chapkin, R. S. Importance of dietary γ-linolenic acid in human health and nutrition. *J. Nutr.* **1998**, *128*, 411−1414.
(9) Rose, D. P.; Connolly, J. M. Omega-3 fatty acids as cancer chemopreventive agents. *Pharm. Ther.* **1999**, *83*, 217−244.
(10) Connor, W. E. Importance of n-3 fatty acids in health and diseases. *Am. J. Clin Nutr.* **2000**, *71*, 171S−175S.
(11) Uauy, R.; Valenzuela, A. Marine oils: the health benefits of n-3 fatty acids. *Nutr.* **2000**, *16*, 680−684.
(12) Trautwein, E. n-3 Fatty acids-physiological and technical aspects for their use in food. *Eur. J. Lipid Sci. Technol.* **2001**, *103*, 45−55.
(13) Challener, C. Specialty dietary supplement ingredients are the hot spot. *Chem. Market Rep.* **2000**, Sept 25.
(14) Pang, T. *China: Traditional Medicine—the Road to International Recognition*; World Markets Research Centre; 2001; http://www.worldmarketsanalysis.com/InFocus2002/articles/asia_china_health.html.
(15) U.S. Department of Commerce. *Best Sales Prospects. Industry Canada*; 1999; http://strategis.ic.gc.ca/SSG/dd70746e.html.
(16) Kapoulas, V. M.; Passaloglou-Emmanouilidou, 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.
(17) Morchio, G.; DiBello, A.; Mariani, C.; Fedeli, E.; Kapoulas, V. M.; Andrikopulos, N. K. Detection of virgin olive oil adulteration with refined oils by second derivative spectrophotometry. *Food Chem.* **1987**, *23*, 183−192.
(18) Lanzou, A.; Cert, A.; Albi, T. Detection of refined olive oil in virgin olive. *Grasas Aceites* **1989**, *40*, 6.
(19) Grob, K.; Giuffre, A. M.; Leuzzi, U.; Mincione, B. Recognition of adulterated oils by direct analysis of the minor components. *Fat Sci. Technol.* **1994**, *96*, 286−290.
(20) Sanchis Rodriguez, J.; Rodriguez Serrano, J. Rapid HPLC procedure for the detection of adulteration of olive oils by seed oils. *Alimentaria (Madrid)* **1991**, *28*, 27−29.
(21) Manku, M. A comparison of GLC and HPLC methods for determining fatty acid composition of evening primrose and soybean oil. *J. Chromatogr. Sci.* **1983**, *21*, 367−369.
(22) Mavromoustakos, T.; Zervou, M.; Thedoropoulou, E.; Panagiotopoulos, D.; Bonas, G.; Day, M.; Helmis, A. ^{13}C-NMR analysis of the triacylglycerol composition of Greek virgin olive oils. *Magn. Reson. Chem.* **1997**, *35*, S3−S7.
(23) Sachhi, R.; Addeo, F.; Paolillo, L. ^{1}H and ^{13}C-NMR of virgin olive oil. An overview. *Magn. Reson Chem.* **1997**, *35*, S133−S145.
(24) Marini, D.; Grassi, L.; Balesteri, F.; Pascucci, E. Spectrophotofluorometric analysis of olive oil. *Riv. Ital. Sostanze Grasse* **1990**, *67*, 95−99.
(25) Passaloglou-Emmanouilidou, S. A comparative study of UV spectrophotometric methods for the detection of olive oil adulteration by refined oils. *Z. Lebensm. Unters. Forsch.* **1990**, *191*, 132−134.
(26) Kapoulas, V. M.; Andrikopulos, N. K. Detection of virgin olive oil adulteration with refined oils by second derivative spectrophotometry. *Food Chem.* **1987**, *23*, 183−192.
(27) Mandl, A.; Reich, G.; Lindner, W. Detection of adulteration of pumpkinseed oil by analysis of content and composition of specific Δ7-phytosterols. *Eur. Food Res. Technol.* **1999**, *209*, 400−406.
(28) Spangenberg, J. E.; Ogrinc, N. Authentication of vegetable oils by bulk and molecular carbon isotope analyses with emphasis on olive oil and pumpkinseed oil. *J. Agric. Food Chem.* **2001**, *49*, 1534−1540.
(29) Kupper, L.; Heise, H. M.; Lampen, P.; Davies, A. N.; McIntyre, P. Authentication and quantification of extra virgin olive oils by attenuated total reflectance infrared spectroscopy using silver halide fiber probes and PLS. *Appl. Spectrosc.* **2001**, *55*, 563−570.
(30) Tay, A.; Singh, R. K.; Krishnan, S.; Gore, J. Authentication of vegetable oils using FTIR spectroscopy. *Lebens. Wiss. Technol.* **2002**, *35* (1), 99−103.
(31) Ozen, B. F.; Mauer, L. J. Detection of hazelnut oil adulteration using FT-IR spectroscopy. *J Agric. Food Chem.* **2002**, *50*, 3898−3901.
(32) Downey, G. Food and food ingredient authentication by mid-infrared spectroscopy and chemometrics. *Trends Anal. Chem.* **1998**, *17*, 418−424.
(33) van de Voort, F. R.; Ismail, A. A. Proximate analysis of foods by mid-FTIR spectroscopy. *Trends Food Sci. Technol.* **1991**, *2*, 13−17.
(34) Sivakesa, S.; Irudayaraj, J. A rapid spectroscopic technique for determining honey adulteration with corn syrup. *J. Food Sci.* **2001**, *66*, 787−792.
(35) European Union Research Committee. Development and assessment of methods for the detection of adulteration of olive oil with hazelnut oil; press release, Brussels, Belgium, Oct 10, 2001.
(36) Kemsley, E. K. *Discriminant Analysis and Class Modelling of Spectroscopic Data*; Wiley: West Sussex, U.K., 1998.
(37) Deferne, J.; Pate, D. W. Hemp seed oil: a source of valuable essential fatty acids. *J. Int. Hemp Assoc.* **1996**, *3* (1), 4−7.
(38) Chu, Y.; Hwang, L. S. Food lipids. In *Chemical and Functional Properties of Food Components*; Sikorski, Z. E., Ed.; CRC Press: Boca Raton, FL, 2002.
(39) Pokorny, J. Phospholipids. In *Chemical and Functional Properties of Food Lipids*; Sikorski, Z. E., Kolakowska, A., Eds.; CRC Press: Boca Raton, FL, 2003.
(40) U.S. Department of Agriculture. *National Nutrient Database*; 2003; http://www.nal.usda.gov/fnic/cgi-bin/nut_search.pl.

Received for review March 13, 2003. Revised manuscript received July 24, 2003. Accepted July 28, 2003. This research was supported by the Purdue University Agricultural Research Programs. This is Journal Article 17142 of the Purdue University Agricultural Research Programs.

JF034245H