TITLE: Detection of adulteration in edible oil using FT-IR spectroscopy and machine learning

ABSTRACT

Aims: To detect the adulterant in edible oil rapidly

Study Design: Authenticity and adulteration detection in edible oils are the increasing challenges for researchers, consumers, industries and regulatory agencies. Traditional approaches may not be the most effective option to combat against adulteration in edible oils as that's are complex, laborious, expensive, require a high degree of technical knowledge when interpreting data and produce hazardous chemical. Consequently, a cost effective, rapid and reliable method is required.

Place and Duration of the Study: The experiment was conducted jointly in the laboratory of the Department of Food Technology and Rural Industries, Bangladesh Agricultural University, Mymensingh and the Institute of Food Science and Technology, BCSIR, Dhaka.

Method: In this study, Fourier Transform Infrared spectroscopy coupled with multivariate analysis was used for adulteration detection in sunflower and rice bran oil. Sunflower oil was adulterated with soybean oil in the range of 10-50% (v/v) and rice bran oil was adulterated with palm oil in the range of 4-40% (v/v) at approximately 10% and 5% increments respectively. FTIR spectra were recorded in the wavenumber range of 4000-650cm^{-1}.

Results: FTIR spectra data in the whole spectral range and reduced spectral range were used to develop a partial least square regression (PLSR) model to predict the level of adulteration in sunflower and palm oils. Good prediction model was obtained for all PLSR models with a coefficient of determination (R^2) of >= 0.985 and root mean square errors of calibration (RMSEC) in the range of 0-1.7325%.

Conclusion: The result suggested that FTIR spectroscopy associated with multivariate analysis has the great potential for a rapid and non-destructive detection of adulteration in edible oils laborious conventional analytical techniques.

Keywords: *FT-IR, Spectroscopy, Adulteration, Edible oil, Authenticity*

1. INTRODUCTION

History across the world reminds that our food is under continuous threat from adulteration. It has existed if food has been made and sold. Since prehistoric times humans have altered the state of food to extend its longevity or improve its taste. However, the eighteenth and nineteenth centuries saw an explosion of food adulteration when foodstuffs were often adulterated with inedible and even poisonous/toxic substances, as all stakeholders' (i.e. farmers, suppliers, and grocers) tried to maximize their profits [1] and now a global issue addressed by several researchers, involving economic, quality, safety, ethical and socio-religious issues [2]. Major food adulteration events appear to regularly occur, for instance adulteration of spices with Sudan Red dye in 2003, milk powder with melamine in 2008, dioxins in pork in 2008, milk with detergent, fat, and even urea in 2012 and processed beef products with horsemeat in 2013 [3-5]. In some cases, there might be no issue related to safety (i.e. horsemeat scandal), however, such adulteration is always a concern with individuals allergic to species, or those with religious taboos or ethical aversions [5, 6]. Adulteration varies widely among the thousands of food products, range from tragic, as in the toxic oil syndrome or simply toxic syndrome disaster in Spain in 1981 where thousands were hospitalized and an approximately 600 people died due to rapeseed oil intended for industrial use being sold as olive oil [7, 8] to authentication of the species [9, 10], variety [11, 12], purity [13-15] and geographical origin of a product [6, 16-18]. Although the extent and dangers of food adulteration have received huge public attention, the prevalence of fraud is not easy to assess [19]. Basically, detecting adulteration is difficult without resorting to highly sophisticated analytical techniques because the adulterant components are usually very similar to the authentic product [2, 20]. On the other side, fraudsters are always one step ahead of the regulatory agencies and their techniques are increasingly becoming more and more sophisticated with time. Once a specific test has been developed

by the scientists to identify an adulterant, fraudsters can become aware of this and then add or remove that component from the adulterated foodstuff [3]. Many methods are available for the detection of qualitative and/ or quantitative adulteration in food. Currently the methods often used for food authentication and detection of fraud include polymerase chain reaction (PCR), chromatography, electrophoretic separation of proteins, immunological procedure and DNA based techniques, and enzymatic assays [19], all of which are well documented. However, these techniques are invasive, time consuming, laborious, demand highly skilled personnel, and thus they are not suitable for online application and routine analysis. Consequently, a cost effective, efficient, rapid, and reliable method is required. There is a great interest in developing non-destructive optical technologies that have the capability of monitoring in real-time assessment.

Among optical sensing technologies, FT-IR spectroscopy is more sensitive and perhaps more suited to detect and quantify the presence of adulterants within complex food matrices. The high spectral signal-to-noise ratio obtained from FT-IR analysis allows the detection of constituents present in very low concentrations as well as subtle compositional differences between and among multi constituent specimens. Basically, the IR spectrum is formed because of the absorption of electromagnetic radiation at characteristics bands/wavenumbers that correlate to the vibration of specific sets of chemical bonds/functional groups from within a molecule. Different functional groups absorb characteristic frequencies of IR radiation. Analysis of these absorption characteristics reveals details about the molecular structure of the sample. However, this is complex task due to the overlap of frequencies characteristic and also because of overtone and combinations bands [8]. There are several alternatives for quantifying a compound/functional group in a multicomponent mixture based on IR spectra, ranging from the univariate calibration method, which correlates the intensity (i.e. absorbance) of an isolated, intense band in the spectrum to the concentration of the compound/functional group, through the peak fitting, which enables quantification of the compound/functional group from the absorption by scaling fixed peak shapes to the spectrum, or fitting parameterized line shapes (such as Gaussian) to particular regions of the spectrum, up to the more sophisticated multivariate approach in which nearly entire spectra are utilized by carrying out correlation between spectral data and the concentrations or other measurable properties obtained from the ordinary laboratory measurements. Univariate analysis or peak fittings can become complex when absorbance peaks overlap [21, 22] or if there are multiple absorption peaks for the same functional group. On the other side, multivariate calibration approach is especially useful when data are highly collinear [23] as in the case for FT-IR spectra. Several chemometric algorithms/multivariate analyses are available to appropriately extract meaningful information in an efficient way from the spectra. By applying chemometric tools, one can perform reliable quantitative analysis of a multicomponent system even in the case of very complex spectra. In its ample applications, FT-IR spectroscopic technique in tandem with multivariate analysis has proved its potential for detecting adulteration and authenticity in edible oils.

Edible oils are routinely used as cooking oils, salad oils, shortenings, spreads and ingredients in several food products formulation and a large variety of edible oils treated and marketed in Bangladesh. Some edible oils are expensive compared to others as tempting to adulterate with the lower price edible oils. Adulteration of high value edible oils can occur either by mislabeling of less expensive oil or by adding less expensive oils to increase the volume and therefore profits. Different physical parameters such as refractive index, viscosity, melting point, saponification and iodine value can be measured to detect adulteration in edible oils. However, these parameters are not anymore practical to detect adulteration as these properties are easy to manage in adulterated oils to mask the adulteration. On the other side, it is also possible to use both major (triacylglycerols) and minor components (sterols, carotenoids, tocopherols, chlorophylls etc.) as detection tool. Among different edible oils, some have particular component at a known level which is absent in other one. Therefore, the presence and amounts of this particular component can be considered as a detection tool [9]. Many analytical techniques can be used to detect adulteration in edible oils. Most of these techniques are based upon the chromatographic methods, which rely on the determination of fatty acids [24]. However, these methods are time consuming, complex, laborious, expensive and destructive, require a skilled operator and produce hazardous chemical waste. In this study, an attempt has been made to develop a rapid and accurate analytical protocol based on FT-IR spectroscopy for the determination of adulteration (palm oil in rice bran oil and soybean oil in sunflower oil) in edible oils and apply machine learning technique such as PLSR to develop calibration model for detecting adulteration in edible oils.

2. MATERIALS AND METHODS

2.1 MATERIALS

Four different edible oils such as sunflower oil, soybean oil, rice bran oil and palm oil were purchased from the local super market.

2.2 PREPARATION OF ADULTERATED SAMPLES

The first study was carried out for detection of palm oil in rice bran oil (palm oil is cheaper than rice bran oil) and the second study was carried out for detection of soybean oil in sunflower oil (sunflower oil is expensive compared to soybean oil). For the first study, rice bran oil was adulterated with palm oil in the range of 5-40% (v/v) at approximately 5% increments. For the second study, sunflower oil was adulterated with soybean oil in the range of 10-50% (v/v) at approximately 10% increments. A total of 8 samples were prepared for palm oil adulteration in rice bran oil and only 5 samples were prepared for soybean oil adulteration in sunflower oil. Additionally, a spectrum of each pure oil was also extracted to compare the IR spectra of different oils.

2.3 FTIR SPECTRA ACQUISITION

Fourier transform infrared spectroscopy (FT-IR) was performed using a Perkin Elmer Universal ATR spectrophotometer (UATR-FT-IR, USA) equipped with a Zn Se crystal for the FT-IR spectroscopy. Transmittance was measured as the function of the wave number between 4000 and 650 cm−1 with their solution of 4 cm−1 and the number of scans equal to 12.

2.4 SPECTRA PRE-PROCESSING

In spectral instruments, sample physical properties and discrepancies in instrument response can cause undesired effects such as light scattering and random noise. These effects can induce spectral variations that are not associated with the studied responses and affect the reliability of multivariate calibration models. These effects can be eliminated from the spectral data by applying some mathematical treatments. However, there is still no single recipe available to select the best pre-treatment technique that needs to be applied. In this study, various pre-processing techniques such as first derivative, second derivative, multiplicative scatter correction (MSC), and standard normal variate (SNV) were separately applied to the spectral data prior to the development of multivariate model.

2.5 ANALYSIS OF SPECTRAL DATA

Partial least squares regression (PLSR) was developed to correlate the FT-IR spectra of different oil samples and their corresponding level of adulteration. It was developed to calibrate the FT-IR spectra of laboratory standards to their corresponding moles of functional group. A detailed description of the PLS can be found in our previous studies [25-27]. In recent years, PLSR has become the *de facto* standard in multivariate spectral analysis in different fields [28-31]. PLSR is emerging as the most robust and reliable chemometric method for constructing multivariate models when the measured variables are many and highly collinear (correlated) as in the case of FTIR spectra [32]. PLS regression is used to find the fundamental relations between the predictors (X) and the responses (y), thus reducing the original predictors to a new variable set called latent variables (LVs), which have the best predictive power. These LVs are statistically independent i.e. uncorrelated and ideally carry all relevant information to be correlated with reference values (i.e., true measured values) leading to more stable predictions [23, 33]. Generally, the PLS regression builds a linear model by decomposing both X (*n, m)* and y (n, 1) and constructs the following relations [34]:

$$X = TP^T + E \quad (1)$$

$$y = Tq^T + f \quad (2)$$

where, P (m, k) is the matrix of X-loadings, T(n, k) is the matrix of X-scores, q (k, 1) is the loading vector of y, E (n, m) and f (n, 1) are error terms which are not explained by the model, and k is the number of LVs used in the PLS model.

The X-scores are predictors of y and model X, i.e., both y and X are assumed to be, at least partly, modeled by the same LVs. The scores can be computed by a linear combination of the variables in X with the weights W^* (m, k) as $T=XW^*$. These weights are computed so that each of them maximizes the covariance between responses and the corresponding scores.

The regression coefficients b (m, 1) of y on X can then be calculated through the weights W* as $b=W^*q^T$, where $W^* = W(P^TW)^{-1}$.
Finally, the PLS latent variable model can be re-organized as a simple prediction equation similarly as for multiple linear regression:

$$\hat{y} = Xb + f \quad (3)$$

To avoid either over- or under-fit problem of the model, cross validation using leave-one-out method was used during the calibration step to select the optimum number of LVs for PLSR model. It was determined at the minimum value of the root mean square error of cross-validation (RMSECV). To estimate the actual predictive capability of the calibration model, the performance of the developed model was validated using an independent test set with samples not included in the original calibration samples.

2.5 EVALUATION OF THE CALIBRATION MODELS

The predictive capabilities of the calibration model were evaluated by examining the coefficient of determination (R^2), and the root mean square errors (RMSE). The R^2 and RMSE are defined as follows:

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(\hat{y}_i - y_i)^2}{\sum_{i=1}^{N}(\hat{y}_i - \bar{y}_i)^2} \quad (4)$$

$$RMSEC \text{ or } RMSECV = \sqrt{\frac{\sum_{i=1}^{N}(\hat{y}_i - y_i)^2}{N}} \quad (5)$$

where $\hat{y}_i$ = predicted value of the i^{th} sample, y_i= measured value of the i^{th} sample, N = number of samples, N_c = number of samples in the calibration set and N_p = number of samples in the validation (testing) set.
Generally, the accuracy of multivariate calibration model is considered as excellent when the R^2 is 0.90 or higher [35, 36]. However, it is always expected to obtain R^2 as close as 1 with errors as close as 0.

2.6 SOFTWARE

All spectral transformations were carried out in Unscrambler (CAMO AS, Trondheim, Norway). The PLSR analysis was carried out in MATLAB.

3. RESULTS AND DISCUSSION

3.1 FT-IR SPECTRA OF THE DIFFERENT PURE AND ADULTERATED EDIBLE OILS

FT-IR spectra of four different pure oils (rice bran, palm, sunflower and soybean) in the spectral range of 4000-650 cm^{-1} are shown in Fig 1 and individual spectra shown in Fig 2. The spectra of the tested samples of different oils showed similar trends throughout the whole spectral range. Despite the similarity, the studied original spectra were different in absorbance values. Although, the difference in the spectral profile is not clear by naked eye in the whole spectral range, it is clearly seen if the spectra are zoomed at some selected spectral range Fig 3. In general, objects present similar spectral patterns indicate their similarity in chemical composition. However, different concentrations of the major chemical compositions in the tested object make the difference in absorbance values. It is seen that the differences between different oils is small and the spectra in the FTIR region have well-resolved bands that can be assigned to different functional groups present in the oils. Basically, the IR spectrum is formed as a consequence of the absorption of electromagnetic radiation at characteristics bands/wavenumbers that correlate to the vibration of specific sets of chemical bonds/functional groups from within a molecule. Different functional groups absorb characteristic frequencies of IR radiation. Analysis of these absorption characteristics reveals details about the molecular structure of the sample [37]. Chemically, fats and oils are glycerol esterified with fatty acids. Some of the fats and oils might have quite similar composition; consequently, it is often difficult to detect adulteration of fats and oils physically [13, 38]. However, because of its

capability as a fingerprint technique, IR spectroscopy allows one to differentiate authentic oils and those adulterated with others by observing the spectra changes due to the adulteration [7, 39].

The assignment of functional groups, shown in Fig 1, are dominated by some peaks at 3013, 2924, 2855, 1745, 1650, 1462, 1416, 1376, 1242, 1160, 1114, 1099, 1033, 968 and 723 cm^{-1}. The observed absorption bands were consistent with reported peak assignments based on published literature [40]. Absorbance between 3008 and 2852 cm^{-1} are due to bands arising from CH_2 stretching vibrations, asymmetric and symmetric, respectively. The major peak at 1745 cm^{-1} arises from C=O stretching vibrations of aldehydes and ketons. The stretching C=C was observed at 1650 cm^{-1}. The bands at 1462, 1416 and 1376 cm^{-1} arise from CH_2 and CH_3 scissoring vibration of ethers, while those at 1242, 1160, 1114 1099 and 1033 cm^{-1} are associated with the C=O stretching vibration. The C=C component in oil samples was observed at 968 cm^{-1}. A small peak at 723 cm^{-1} corresponds to CH_2 rocking mode. These observations are in agreement with the results of other studies performed with oils [41, 42], which have a composition and spectra very similar to the ones obtained in this work.

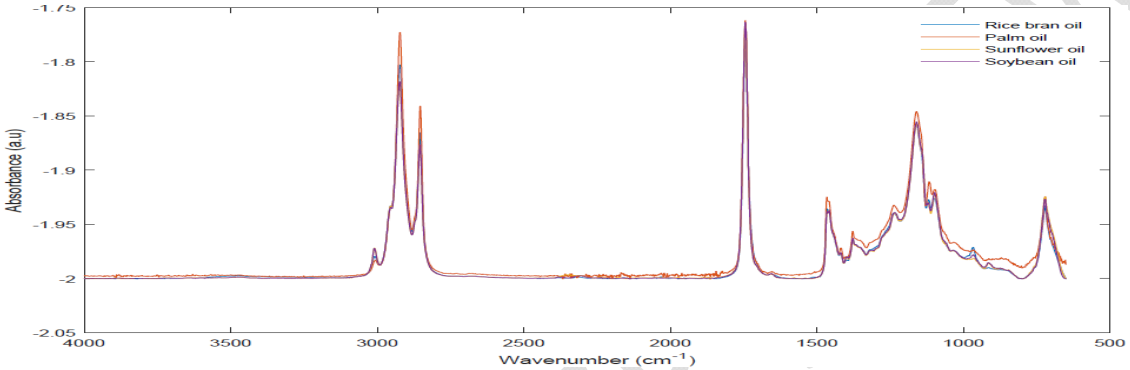


Fig1. FTIR spectra of rice bran, palm, sunflower, and soybean oils in the spectral range of 4000-650 cm^{-1}.

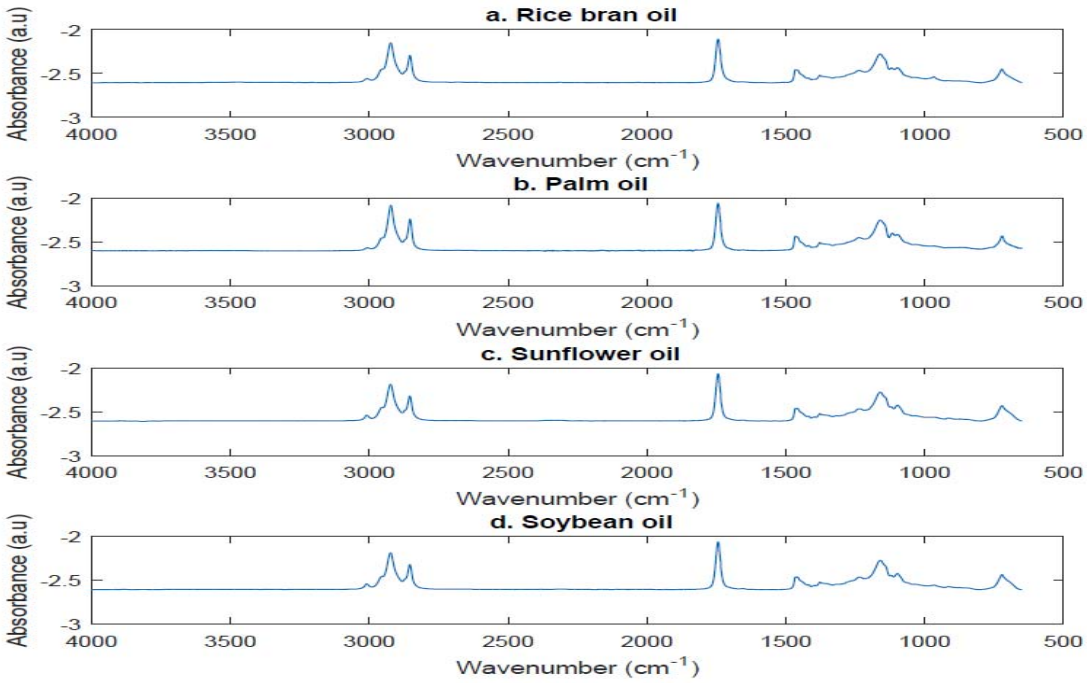


Fig 2: Spectra of rice bran, palm, soybean and sunflower oil in the spectral range of 4000-650 cm^{-1}.

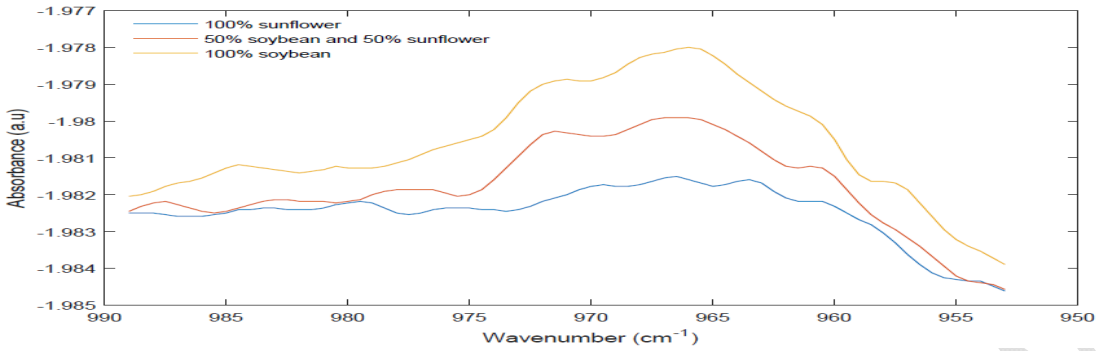
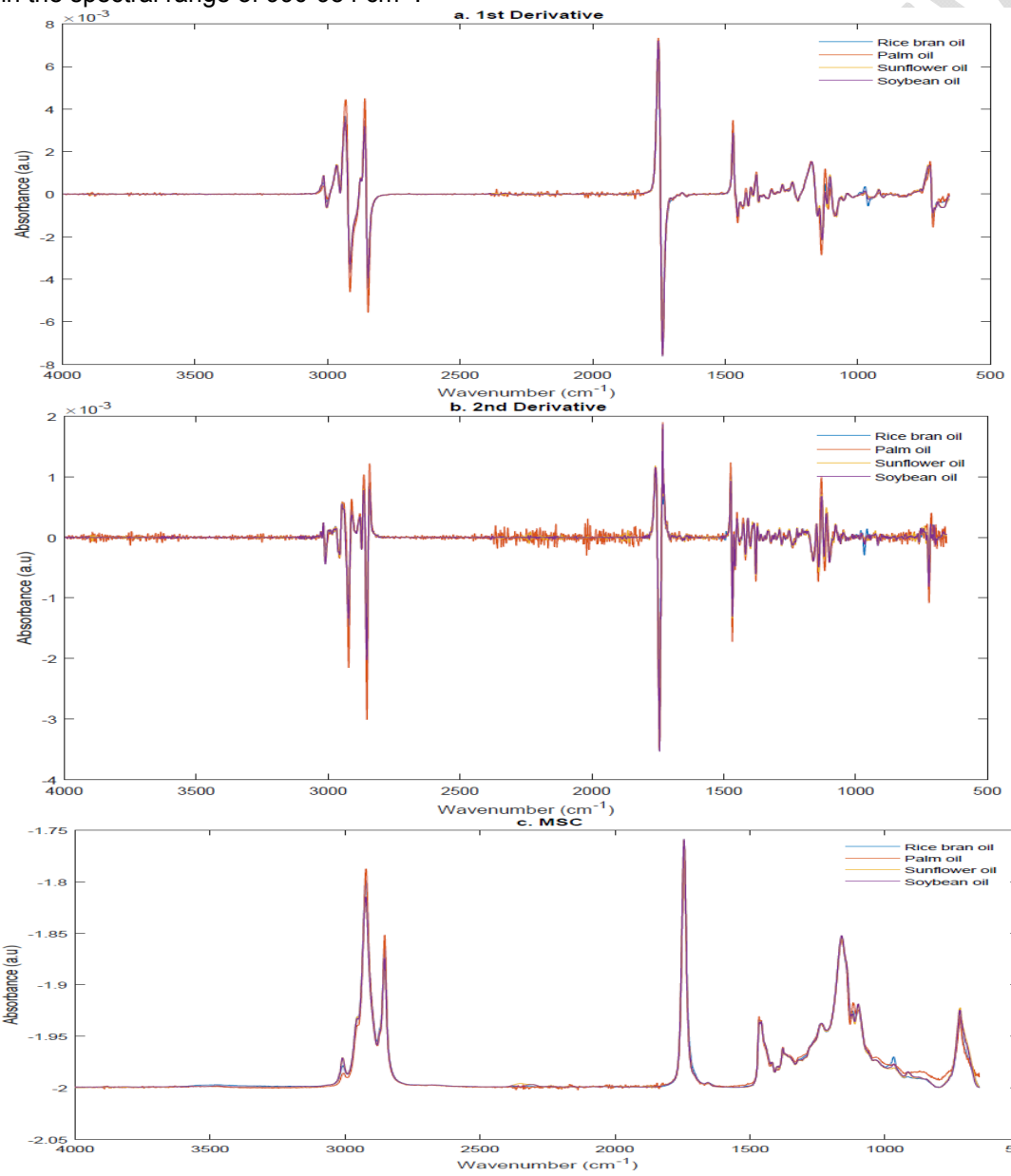


Fig 3: FTIR spectra (zoomed) of pure sunflower oil, soybean oil and 1:1 mixed of sunflower and soybean in the spectral range of 990-954 cm^{-1}.



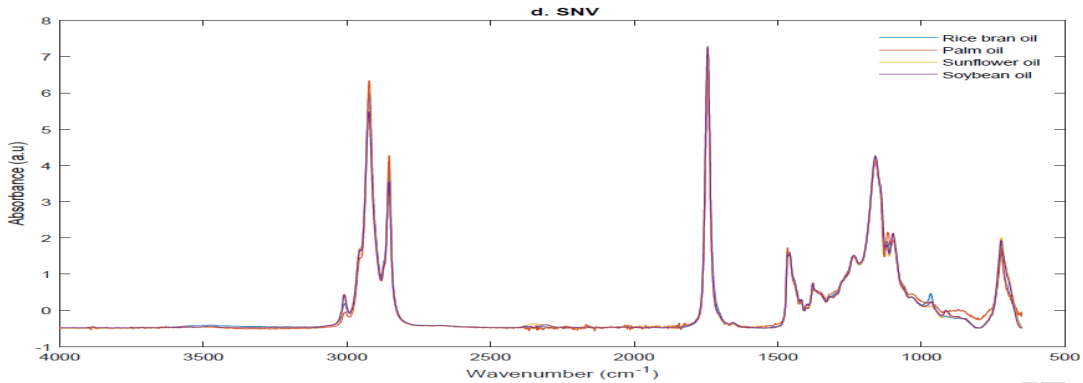


Figure 4: Spectral features with various pre-treatment procedures in the spectral range of 4000-650 cm^{-1}: (a) first derivative, (b) second derivative, (c) MSC and (d) SNV

To correct the scatter effect, different spectral pre-treatment techniques such as derivatives (both first and second derivative) MSC and SNV were applied and the resulting spectra are shown in Fig 4. It is apparent that all the pre-treatments effectively suppressed the scatter effect. SNV and MSC worked similarly in data preprocessing and provided equivalent results as shown in Fig 4 (c, d) and this agreed well with some previous investigations [43, 44]. As expected, several new absorption spectral bands are apparent in the derivative spectra as illustrated in Fig 4 (a,b); those were difficult to understand in the original reflectance spectra as shown in Fig 1.

3.2 SELECTION OF SPECTRAL RANGE

It is not always possible to distinguish infrared spectra of adulterated samples from pure samples with visual inspection. Therefore, it is necessary to analyze the data by multivariate methods to develop predictive models and to achieve an accurate study due to the fact that some regions could be statistically different. Similar changes in the absorbance on some regions are proportional to the degree of adulteration. In this study, the most sensitive region from whole spectra was selected for multivariate analysis based on visual inspection [40]. The region 3040-2995 cm^{-1} and 1000-960 cm^{-1} were selected for detecting palm oil adulteration in rice bran oil, while the regions 3040-2995 cm^{-1} and 990-954 cm^{-1} were selected for detecting soybean oil adulteration in sunflower oil. The FTIR spectra depicted Fig 5 and 6 revealed the differences in absorbance in these range due to the change in concentration of functional groups for the addition of adulterants (i.e. palm oil in rice bran and soybean oil in sunflower) in different level. These spectral ranges, in addition of whole spectral range, were used to develop multivariate calibration for detecting adulteration in edible oil. Therefore, a total of six calibration models was developed for adulteration detection in rice bran oil and sunflower oil.

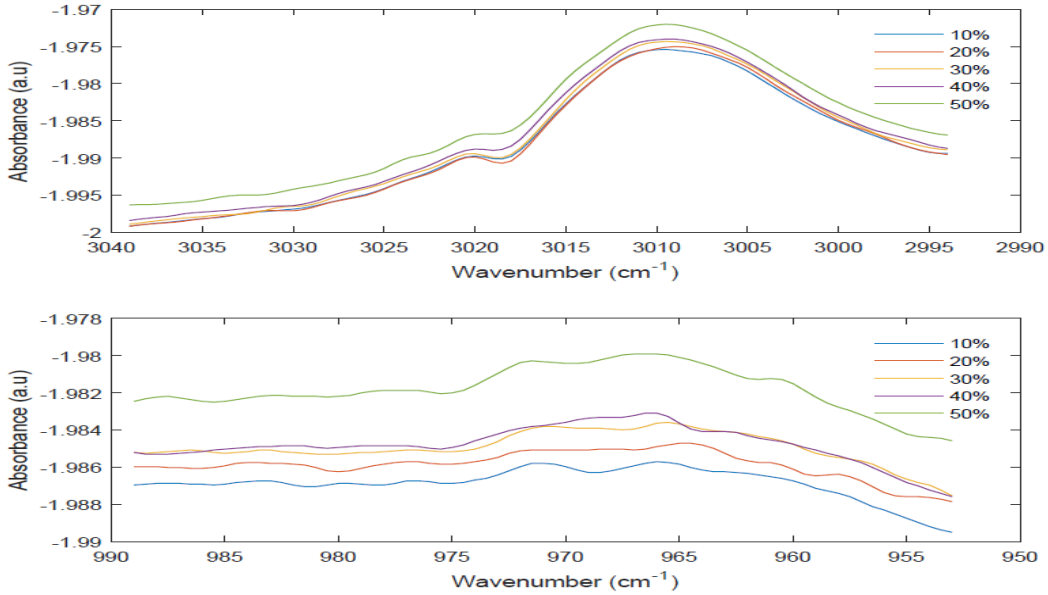


Fig 5: FTIR spectra in the spectral range of 3040-2995 cm^{-1} (top) and 990-954 cm^{-1} (bottom) for soybean oil (10-50%) adulteration in pure sunflower oil.

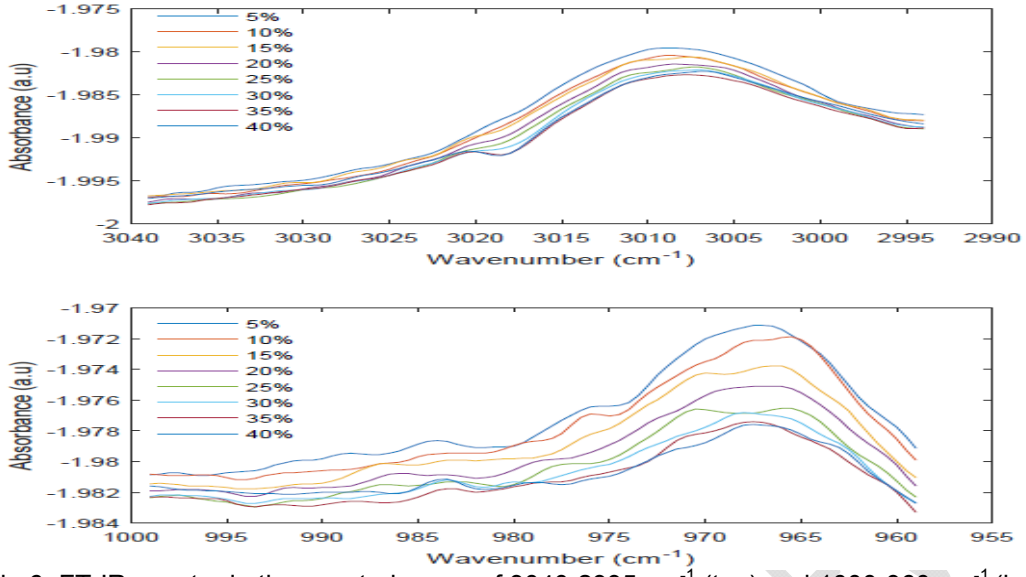


Fig 6: FT-IR spectra in the spectral range of 3040-2995 cm^{-1} (top) and 1000-960 cm^{-1} (bottom) for palm oil adulteration (5-40%) in pure rice bran oil.

3.3 DEVELOPMENT OF CALIBRATION MODEL BASED ON FT-IR SPECTRA

Application of IR spectroscopy combined with chemometric methods is a relatively new approach to determine authenticity and adulteration detection in edible oil industry. Use of chemometric technique as an analytical procedure is fast and very cheap, not very accurate but enough accurate for many actual problems. In this study, a chemometric algorithm called partial least squares regression (PLSR) was used. Three different PLSR models were developed using full spectra of 4000-650 cm^{-1} and reduced spectra of 3040-2995 cm^{-1} and 1000-960 cm^{-1} for palm oil adulteration in rice bran oil. On the other hand, another three models were developed in the spectral range of 4000-650 cm^{-1}, 3040-2995 cm^{-1} and 990-954 cm^{-1} for soybean oil adulteration detection in sunflower oil. The selection of the optimum number of LVs is the key step in building a robust PLSR model to obtain efficient and reliable prediction [30]. Selecting many or few LVs may lead to over- or under-fitting of the model. There are numerous ways to select optimum number of LVs. In this study, the optimum number of LVs was selected at the minimum value of RMSECV (Fig 7a and Fig 8a). The calibration statistics of different PLSR models are summarized in Table 1. To visualize the PLSR calibration models, the actual percent level of adulteration and its predicted percent level of adulteration obtained from the PLSR models are plotted and displayed in Fig 7 (b, c, d) and Fig 8 (b, c, d). The PLSR models developed using the raw spectra were very accurate with R_c^2 >0.985 for all three spectral ranges for both adulterants. The results found in this study were similar to those mentioned by [40, 45] for predicting adulteration in edible oils using FT-IR spectroscopy. Quiñones-Islas et al. [40] reported R^2 of >0.99 using PLSR for predicting sunflower, canola and soybean adulteration in avocado oil [15, 45], whereas [45] obtained R^2 of 0.999 for predicting sunflower and corn oil in extra virgin coconut oil.

Table1: Performance of PLS model for detecting soybean oil and palm adulteration in sunflower oil and rice bran oil, respectively.

Adulterants	Spectral range (cm^{-1})	LVs	RMSEC	R^2
Soybean	4000-650	4	0.00	1.00
	3040-2995	4	0.00	1.00
	990-954	2	1.7325	0.9850
Palm oil	4000-650	5	0.031	1.00
	3040-2995	2	0.728	0.996
	1000-960	2	0.801	0.995

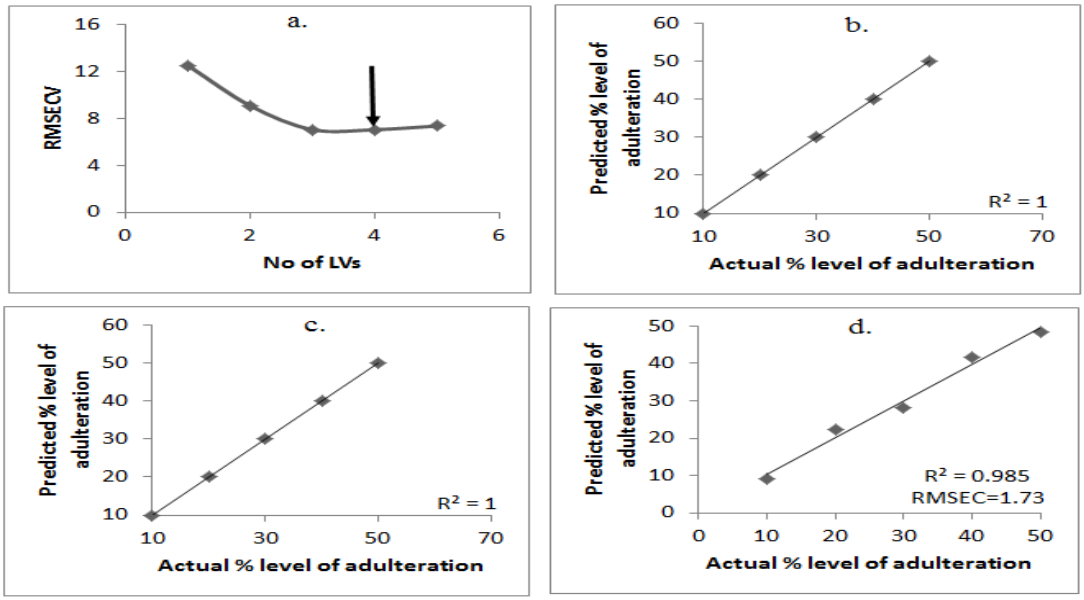


Fig. 7: Prediction of soybean oil adulteration in sunflower oil using PLSR models. a. An example of determination of LVs at the minimum value of RMSECV. a. Measured vs. PLSR prediction of % level of adulteration in the range of b. 4000-650 cm^{-1}, c. 3040-2995 cm^{-1} and d. 990-954 cm^{-1}.

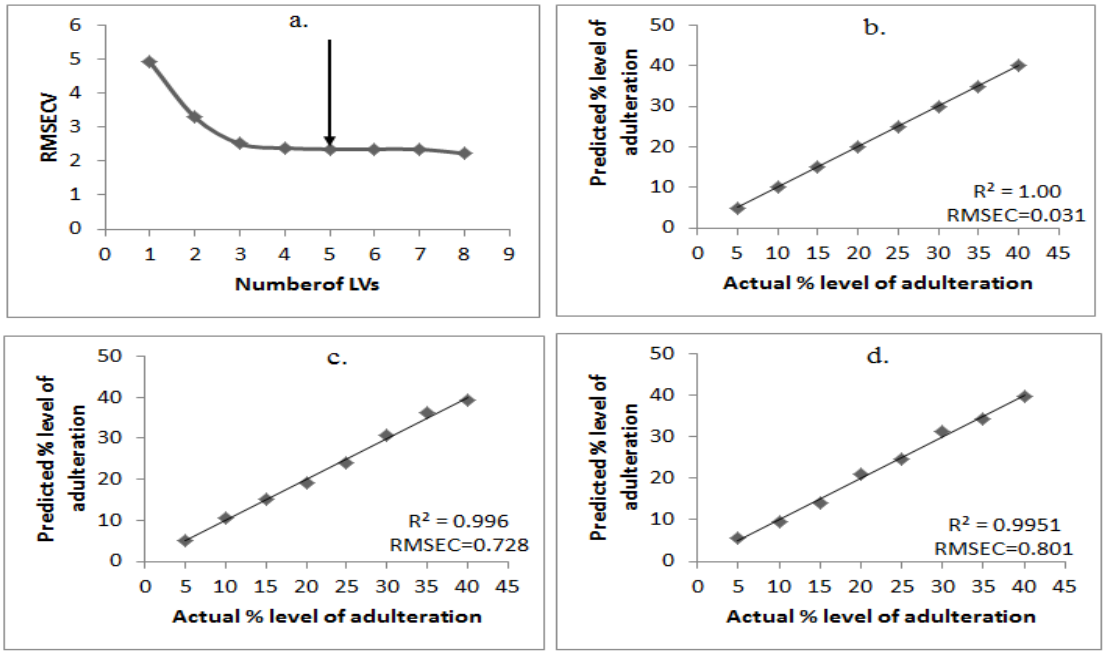


Fig. 8: Prediction of palm oil adulteration in rice bran oil using PLSR models. a. An example of determination of LVs at the minimum value of RMSECV. Measured vs. PLSR prediction of % level of adulteration in the range of b. 4000-650 cm^{-1}, c. 3040-2995 cm^{-1} and d. 1000-960 cm^{-1}.

Table 2. Measured and PLSR predicted values of palm oil adulteration in rice bran oil.

% Measured	% Predicted (4000-650 cm^{-1})	% Predicted (3040-2995 cm^{-1})	% Predicted (1000-960 cm^{-1})
5.00	4.98	4.99	5.53
10.00	10.00	10.67	9.45
15.00	15.00	15.01	14.09
20.00	20.06	19.16	21.01
25.00	24.97	24.12	24.75
30.00	30.00	30.64	31.36
35.00	34.95	36.12	34.17
40.00	40.02	39.20	39.63

Table 3. Measured and PLSR predicted values of soybean oil adulteration in sunflower oil.

% Measured	% Predicted (4000-650 cm^{-1})	% Predicted (3040-2995 cm^{-1})	% Predicted (990-954 cm^{-1})
10.00	10.00	10.00	9.02
20.00	20.00	20.00	22.36
30.00	30.00	30.00	28.25
40.00	40.00	40.00	41.81
50.00	50.00	50.00	48.55

4. CONCLUSION

The samples were collected from the local market and the preparation was done in the laboratory. This study mainly concerned with the development of optical sensing technique based on FTIR spectroscopy for adulteration detection in edible oils. Two types of oils such as palm oil and soybean oil were selected as adulterants. Rice bran oil was adulterated with palm oil at 5% increments upto 40% (v/v), while sunflower oil was adulterated with soybean oil at 10% increments upto 50% (v/v). FTIR spectra in the wave number interval of 4000–650 cm^{-1} were then collected at room temperature using a Perkin Elmer Universal ATR spectrophotometer (UATR-FT-IR, USA). Various pre-processing techniques such as first derivation, second derivative, multiplicative scatter correction (MSC), and standard normal variate (SNV) were separately applied to the spectral data prior to the development of multivariate model. Partial least squares regression (PLSR) was developed to correlate the FT-IR spectra of different oil samples and their corresponding level of adulteration. Cross validation using leave-one-out method was used during the calibration step to select the optimum number of LVs for PLSR model. It was determined at the minimum value of the root mean square error of cross-validation (RMSECV). Six different PLSR models were developed based on whole spectral range as well as selected spectral range. For all models, the level of adulteration in pure oil was predicted with determination coefficients (R^2) of > 0.985. This study revealed that FT-IR spectroscopy coupled with PLSR can be successfully utilized as a rapid screening technique to detect and quantify the level of adulteration in edible oil.

Adulteration of food products involves the replacement of high cost ingredients with inferior quality substitutes. Expensive edible oil is sometimes adulterated with cheaper oil as a means of illegally

increasing profit. Therefore, the challenge is to develop a cost effective and efficient analytical method to detect adulteration and confirm authenticity. Using FTIR spectroscopy coupled with multivariate analysis could be used as an alternative analytical tool to detect adulteration in sunflower oil and rice bran oil. Therefore, the laborious and time-consuming conventional analytical techniques could be replaced by spectral data to provide a rapid and nondestructive testing technique. More results compare with other analytical techniques need to be addressing to validate the models.

COMPETING INTEREST

Authors have declared that no competing interests exist.

REFERENCES

1. Filby FA. (1976). A history of food adulteration and analysis. Allen & Unwin, London.
2. Alamprese, C., Casale, M., Sinelli, N., Lanteri, S., & Casiraghi, E. (2013). Detection of minced beef adulteration with turkey meat by UV–vis, NIR and MIR spectroscopy. *LWT - Food Science and Technology, 53*(1), 225-232. doi:10.1016/j.lwt.2013.01.027.
3. Ellis, D. I., Brewster, V. L., Dunn, W. B., Allwood, J. W., Golovanov, A. P., & Goodacre, R. (2012). Fingerprinting food: current technologies for the detection of food adulteration and contamination. *Chem Soc Rev, 41*(17), 5706-5727. doi:10.1039/c2cs35138b.
4. Kamruzzaman, M., Makino, Y., Oshita, S., & Liu, S. (2015). Assessment of Visible Near-Infrared Hyperspectral Imaging as a Tool for Detection of Horsemeat Adulteration in Minced Beef. *Food and Bioprocess Technology, 8*(5), 1054-1062. doi:10.1007/s11947-015-1470-7.
5. Zhao, M., Downey, G., & O'Donnell, C. P. (2014). Detection of adulteration in fresh and frozen beefburger products by beef offal using mid-infrared ATR spectroscopy and multivariate data analysis. *Meat Sci, 96*(2 Pt A), 1003-1011. doi:10.1016/j.meatsci.2013.10.015.
6. Urickova, V., & Sadecka, J. (2015). Determination of geographical origin of alcoholic beverages using ultraviolet, visible and infrared spectroscopy: A review. *Spectrochim Acta A Mol Biomol Spectrosc, 148*, 131-137. doi:10.1016/j.saa.2015.03.111.
7. Beaten V, Fernández Pierna J A, Dardenne P, Meurens M, García-González D L and Aparicio-Ruiz R (2005). Detection of the presence of hazelnut oil in olive oil by FT-Raman and FT-MIR spectroscopy. *Journal of Agriculture and Food Chemistry, 53*, 6201–6206.
8. García-González, D. L., Aparicio-Ruiz, R., & Aparicio, R. (2008). Virgin olive oil - Chemical implications on quality and health. *European Journal of Lipid Science and Technology, 110*(7), 602-607. doi:10.1002/ejlt.200700262.
9. Espiñeira, M., Vieites, J. M., & Santaclara, F. J. (2010). Species authentication of octopus, cuttlefish, bobtail and bottle squids (families Octopodidae, Sepiidae and Sepiolidae) by FINS methodology in seafoods. *Food Chemistry, 121*(2), 527-532. doi:10.1016/j.foodchem.2009.12.042.
10. Kamruzzaman, M., Barbin, D., ElMasry, G., Sun, D.-W., & Allen, P. (2012a). Potential of hyperspectral imaging and pattern recognition for categorization and authentication of red meat. *Innovative Food Science and Emerging Technologies, 16*, 316-325. doi:http://dx.doi.org/10.1016/j.ifset.2012.07.007.
11. Manley M, De Bruyn N and Downey G (2003). Classification of three-year old, unblended South African brandy with near-infrared spectroscopy. *NIR News, 14*, 8-9.
12. Ruoff K, Luginbuhl W and Kunzli R (2006). Authentication of the botanical and geographical origin of honey by mid-infrared spectroscopy. *Journal of Food Agricultural Chemistry, 54*, 6873-6880.
13. Li, B., Wang, H., Zhao, Q., Ouyang, J., & Wu, Y. (2015). Rapid detection of authenticity and adulteration of walnut oil by FTIR and fluorescence spectroscopy: a comparative study. *Food Chem, 181*, 25-30. doi:10.1016/j.foodchem.2015.02.079.
14. Mamani-Linares, L. W., Gallo, C., & Alomar, D. (2012). Identification of cattle, llama and horse meat by near infrared reflectance or transflectance spectroscopy. *Meat Sci, 90*(2), 378-385. doi:10.1016/j.meatsci.2011.08.002.
15. Sun, X., Zhang, L., Li, P., Xu, B., Ma, F., Zhang, Q., & Zhang, W. (2015). Fatty acid profiles based adulteration detection for flaxseed oil by gas chromatography mass spectrometry. *LWT - Food Science and Technology, 63*(1), 430-436. doi:10.1016/j.lwt.2015.02.023.

16. Mohammed Kamruzzaman, Y. M., Seiichi Oshita. (2014). An appraisal of hyperspectral imaging for non-invasive authentication of geographical origin of beef and pork *Proceedings International Conference of Agricultural Engineering, Zurich*.
17. Longobardi F,Casiello G and Cortese M (2015). Discrimination of geographical origin of lentils (Lens culinaris Medik.) using isotope ratio mass spectroscopy combined with chemometrics. *Food Chemistry*, 05, 020.
18. Picque D, Cattenoz T and Corrieu G (2005). Discrimination of red wines according to their geographical origin and vintage year by the use of mid-infrared spectroscopy. *Science Aliments, 25,* 207- 220.
19. Ballin, N. Z. (2010). Authentication of meat and meat products. *Meat Sci, 86*(3), 577-587. doi:10.1016/j.meatsci.2010.06.001.
20. Rasmus Bro, Van Den Berg F., Anette Thybo, Charlotte M. Anderseny, Bo M. Jørgensenx and Henrik Andersen. (2002). Multivariate data analysis as a tool in advanced quality monitoring in the food production chain. *Trends in Food Science & Technology, 13*, 235-244.
21. Russell, L. M., Bahadur, R., Hawkins, L. N., Allan, J., Baumgardner, D., Quinn, P. K., & Bates, T. S. (2009). Organic aerosol characterization by complementary measurements of chemical bonds and molecular fragments. *Atmospheric Environment, 43*(38), 6100-6105.
22. Takahama, S., Johnson, A., & Russell, L. M. (2013). Quantification of carboxylic and carbonyl functional groups in organic aerosol infrared absorbance spectra. *Aerosol Science and Technology, 47*(3), 310-325.
23. Svante Wold, M. S. a. L. E. (2001). PLS-regression: a basic tool of chemometrics *Chemometrics and Intelligent Laboratory Systems, 58*, 109-130.
24. Safwan M. Obeidat, M. S. K. a. W. M. O. (2009). Classification of Edible Oils and Uncovering Adulteration of Virgin Olive Oil Using FTIR with the Aid of Chemometrics *Australian Journal of Basic and Applied Sciences, 3*(3), 2048-2053.
25. Kamruzzaman, M., Makino, Y., & Oshita, S. (2016). Rapid and non-destructive detection of chicken adulteration in minced beef using visible near-infrared hyperspectral imaging and machine learning. *Journal of Food Engineering, 170*, 8-15. doi:10.1016/j.jfoodeng.2015.08.023.
26. Maggio, R. M., Cerretani, L., Chiavaro, E., Kaufman, T. S., & Bendini, A. (2010). A novel chemometric strategy for the estimation of extra virgin olive oil adulteration with edible oils. *Food Control, 21*(6), 890-895. doi:10.1016/j.foodcont.2009.12.006.
27. Oussama, A., Elabadi, F., Platikanov, S., Kzaiber, F., & Tauler, R. (2012). Detection of Olive Oil Adulteration Using FT-IR Spectroscopy and PLS with Variable Importance of Projection (VIP) Scores. *Journal of the American Oil Chemists' Society, 89*(10), 1807-1812. doi:10.1007/s11746-012-2091-1.
28. Abdel-Rahman, E. M., Mutanga, O., Adam, E., & Ismail, R. (2014). Detecting Sirex noctilio grey-attacked and lightning-struck pine trees using airborne hyperspectral data, random forest and support vector machines classifiers. *ISPRS Journal of Photogrammetry and Remote Sensing, 88*, 48-59. doi:10.1016/j.isprsjprs.2013.11.013.
29. Kamruzzaman, M., ElMasry, G., Sun, D. W., & Allen, P. (2012). Prediction of some quality attributes of lamb meat using near-infrared hyperspectral imaging and multivariate analysis. *Anal Chim Acta, 714*, 57-67. doi:10.1016/j.aca.2011.11.037.
30. Perez-Rodriguez, M., Horak-Terra, I., Rodriguez-Lado, L., & Martinez Cortizas, A. (2016). Modelling mercury accumulation in minerogenic peat combining FTIR-ATR spectroscopy and partial least squares (PLS). *Spectrochim Acta A Mol Biomol Spectrosc, 168*, 65-72. doi:10.1016/j.saa.2016.05.052.
31. Ruthenburg, T. C., Perlin, P. C., Liu, V., McDade, C. E., & Dillner, A. M. (2014). Determination of organic matter and organic matter to organic carbon ratios by infrared spectroscopy with application to selected sites in the IMPROVE network. *Atmospheric Environment, 86*, 47-57. doi:10.1016/j.atmosenv.2013.12.034.
32. Gurdeniz, G., & Ozen, B. (2009). Detection of adulteration of extra-virgin olive oil by chemometric analysis of mid-infrared spectral data. *Food Chemistry, 116*(2), 519-525. doi:10.1016/j.foodchem.2009.02.068.
33. Hirri A, Gammouh M, Gorfti A, Kzaiber F, Bassbasi M, Souhassou S, Balouki A and Oussama A (2015). The use of Fourier transform mid infrared (FT-MIR) spectroscopy for detection and

estimation of extra virgin olive oil adulteration with old olive oil, *Sky Journal of Food Science, 4*, 060 -066.
34. Kowalski, P. G. B. R. (1986). Partial Least-Squares Regression: A Tutorial. *Analytica Chimica Acta, 186*, 1-17.
35. Rinnan, Å., Berg, F. v. d., & Engelsen, S. B. (2009). Review of the most common pre-processing techniques for near-infrared spectra. *TrAC Trends in Analytical Chemistry, 28*(10), 1201-1222. doi:10.1016/j.trac.2009.07.007.
36. Urbano Cuadrado, M., Luque de Castro, M. D., Perez Juan, P. M., & Gomez-Nieto, M. A. (2005). Comparison and joint use of near infrared spectroscopy and Fourier transform mid infrared spectroscopy for the determination of wine parameters. *Talanta, 66*(1), 218-224. doi:10.1016/j.talanta.2004.11.011.
37. Zhang, Q., Liu, C., Sun, Z., Hu, X., Shen, Q., & Wu, J. (2012). Authentication of edible vegetable oils adulterated with used frying oil by Fourier Transform Infrared Spectroscopy. *Food Chem, 132*(3), 1607-1613. doi:10.1016/j.foodchem.2011.11.129.
38. Christy AA, K. S., Du Y and Ozaki K. (2004). The detection and quantification of adulteration in olive oil by near-infrared spectroscopy and chemometrics. *Analytical Sciences, 20*, 935-940.
39. Yap, K. Y.-L., Chan, S. Y., & Lim, C. S. (2007). Infrared-based protocol for the identification and categorization of ginseng and its products. *Food Research International, 40*(5), 643-652. doi:10.1016/j.foodres.2006.11.009.
40. Quiñones-Islas, N., Meza-Márquez, O. G., Osorio-Revilla, G., & Gallardo-Velazquez, T. (2013). Detection of adulterants in avocado oil by Mid-FTIR spectroscopy and multivariate analysis. *Food Research International, 51*(1), 148-154. doi:10.1016/j.foodres.2012.11.037.
41. Mauer, B. F. O. A. L. J. (2002). Detection of Hazelnut Oil Adulteration Using FT-IR Spectroscopy. *Agricultural and Food Chemistry, 50*, 3898-3901. doi:doi:10.1021/jf0201834.
42. Tay A, Singh RK, Krishnan SS and Gore JP (2002). Authentication of olive oil adulterated with vegetable oils using Fourier transform infrared spectroscopy. *LWT-Food Science and Technology.35,* 99-103.
43. Elzey, B., Pollard, D., & Fakayode, S. O. (2016). Determination of adulterated neem and flaxseed oil compositions by FTIR spectroscopy and multivariate regression analysis. *Food Control, 68*, 303-309. doi:10.1016/j.foodcont.2016.04.008.
44. Rohman, A., & Man, Y. B. C. (2011). The use of Fourier transform mid infrared (FT-MIR) spectroscopy for detection and quantification of adulteration in virgin coconut oil. *Food Chemistry, 129*(2), 583-588. doi:doi:10.1016/j.foodchem.2011.04.070.
45. Rohman, A., & Man, Y. C. (2010). Fourier transform infrared (FTIR) spectroscopy for analysis of extra virgin olive oil adulterated with palm oil. *Food Research International, 43*(3), 886-892. doi:doi:10.1016/j.foodres.2009.12.006.