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Potential of Fourier-transform infrared spectroscopy in adulteration detection and quality assessment in buffalo and goat milks

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ABSTRACT

Adulteration of higher priced milks with cheaper ones to obtain extra profit can be the cause of adverse health effects as well as economic loss. In this study, it was aimed to differentiate goat-cow and buffalo-cow milk mixtures and also to estimate the critical quality parameters of these milks by the evaluation of Fourier-transform infrared (FTIR) spectroscopic data with chemometric methods. Raw goat and buffalo milks were mixed with cow milk at 1–50% (v/v) concentrations and FTIR spectra of the pure and mixed samples were obtained at 4000–650 cm^{-1}. Orthogonal partial least square discriminant analysis (OPLS-DA) resulted in differentiation of goat-cow and buffalo-cow milk mixtures with 93% and 91% correct classification rates, respectively. Detection level for mixing is determined as higher than 5% for both milks. Total fat, protein, lactose and non-fat solid contents were predicted from FTIR spectral data of the combination of three types of milks by partial least square models with R^2 values of 0.99. As a result, FTIR spectroscopy provides rapid and simultaneous detection of adulteration and prediction of quality parameters regardless of the milk type.

1. Introduction

As it is very well known, composition of milk varies depending on factors such as type and diet of animal. Various types of milk have higher market values owing mostly to their compositions. It was reported that goat milk is very rich in terms of calcium, phosphorus, magnesium, copper and conjugated linoleic, omega 3 and 6 fatty acids [1]. Short and medium chain fatty acid contents of goat milk are higher compared to cow's milk and higher amounts of these fatty acids enable the lipase enzyme to work more effectively; therefore, goat milk is easier to digest [2]. Goat milk is also suitable for the consumption by people who have allergies because it has lower share of αs_1-casein fraction compared to cow milk causing reduced sensitivity to β-lactoglobulin, a protein with high allergy potential [3]. Along with these positive health effects, goat milk has been reported to have other beneficial properties such as antioxidant, anti-inflammatory, anticarcinogenic, antidiabetic, antihypertensive and antiobesity activities [4].

Buffalo milk contains higher percentages of fat and protein compared to cow milk; moreover, lower cholesterol levels were reported for this type of milk [5]. Buffalo milk has high quality proteins and it also contains antibacterial peptides with essential and branched amino acids [6]. The levels of oligosaccharides in buffalo milk are significantly higher than cow and goat milks. These constituents activate the immune system by providing defense against bacterial and viral infections on the intestinal epithelial surfaces, and they also support beneficial flora in the colon [7].

Goat and buffalo milks are the preferred milk type for some cheeses and yogurts due to the differences in their compositions and sensory attributes [8]. Market prices of goat and buffalo milks are generally higher in most parts of the world compared to cow milk because of the factors such as availability and consumer demand. Therefore, one way for fraudsters to obtain extra profit is to mix these milks with cow milk [9]. Depending on the nature of the adulterant, these mixtures can cause disturbances in people who have allergies and/or sensitive digestive systems [10]. In addition to its economic and health aspects, mixed milks can cause religious, ethical and cultural problems [9]. Another problem related with milk mixtures could be encountered in the production of dairy products. Because various properties of buffalo, goat and cow milks such as hydrolysis time, colloidal stability, dispersion state and creaming rate that affect the processing conditions along with the final quality of the product, including textural and sensory characteristics, differ from each other. These properties are very much related

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with compositional and physical characteristics of raw milk [3]. Therefore, it is important to determine the quality characteristics and authenticity of the milk considering all aspects of milk usage.

Immunological, electrophoresis and chromatographic techniques along with DNA-based methods have been investigated to determine adulteration of milk with various adulterants [11]. Some of these techniques are time consuming, require qualified personnel and produce considerable amount of chemical waste. Rapid spectroscopic techniques have been also used to determine various adulterants in milk [12]. These techniques, in general, do not have pre-sample preparation steps and they require none/minimum amounts of chemicals. Raman spectroscopy was able to differentiate buffalo and cow milks based on the presence of β-carotene [13]. Synchronous fluorescence and Raman spectroscopy techniques were used in separating buffalo and cow milk mixtures with a detection limit of 6% [14]. Another spectroscopic method that provides successful results in adulteration detection is Fourier-transform infrared (FTIR) spectroscopy. It is a well-established analytical tool for rapid, high-yield, non-destructive analysis of a wide variety of samples and it provides fingerprint capability for biochemical substances in the sample. This spectroscopic method has been used in detection of water, starch, sodium citrate, formaldehyde, sucrose, detergents, melamine, urea and sodium bicarbonate addition into milk [15–19]. Determination of mixtures of various types of milk as soy and cow-buffalo [20], and goat, sheep, and cow milks [21] was also investigated with FTIR spectroscopy. However, there are not any investigations of buffalo and cow mixtures with FTIR spectroscopy in literature.

Another advantage of spectroscopic methods is that they allow prediction of compositional parameters of an analyzed sample with a single run with the help of multivariate statistical analysis of the data. There are several reports of the estimation of various chemical constituents such as fatty acid and protein compositions of different types of milks with FTIR spectroscopy [22]. However, these prediction studies were mostly performed with milk samples from single animal species. However, in the current study, it is intended to obtain several quality parameters of milk samples by combining data from three types of milk (cow, goat and buffalo), from mid-IR spectral data.

It was aimed to identify mixtures of buffalo-cow and goat-cow milks with mid-IR spectroscopy and to determine some quality characteristics (%fat, %protein, % lactose, %solid non-fat) of pure milk samples regardless of the milk type with the use of multivariate statistical analysis in this study.

2. Materials and methods

2.1. Milk samples and mixtures

A total of 72 milk samples of about 2 L each were obtained directly from nine commercial dairy farms and taken to the laboratory in chilled conditions. Each farm supplied one species of milk in raw form and the samples were obtained throughout a 12-month period. Farms are located in Aegean Region of Turkey. At least one sample from each animal type was obtained monthly and the pure samples were the mixtures of daily production of each farm.

Number of pure raw buffalo, goat and cow milk samples used in the study were 16, 19 and 37, respectively (Fig. 1 and Table S1, Supplementary Material), and all of these samples were used in prediction of milk quality parameters. Sixteen blends (at least one blend for each month) were prepared by adding cow milk to goat and buffalo milks separately for authentication purpose. As a result, 16 blends having 1, 5, 10, 20, 30, 40 and 50% (v/v) mixture ratio for buffalo-cow and goat-cow milk mixtures were obtained. For both goat-cow and buffalo-cow milk mixtures, 112 blends (16 blends × 7 concentrations) were separately prepared and used in adulteration part of the study.

The detailed schematic representation of the experimental plan for both prediction and adulteration parts is given as Fig. 1. Sampling and analysis were done in the same day and the samples were used in their raw form without any pretreatment.

2.2. Determination of selected quality parameters of milk samples

Fat, protein, lactose, and solid non-fat contents of pure milk samples were determined with Lactostar Dairy Analyser (Funke Gerber, Berlin, Germany) with two repetitions. In each run, the equipment was cleaned with cleaning reagents to avoid any residual.

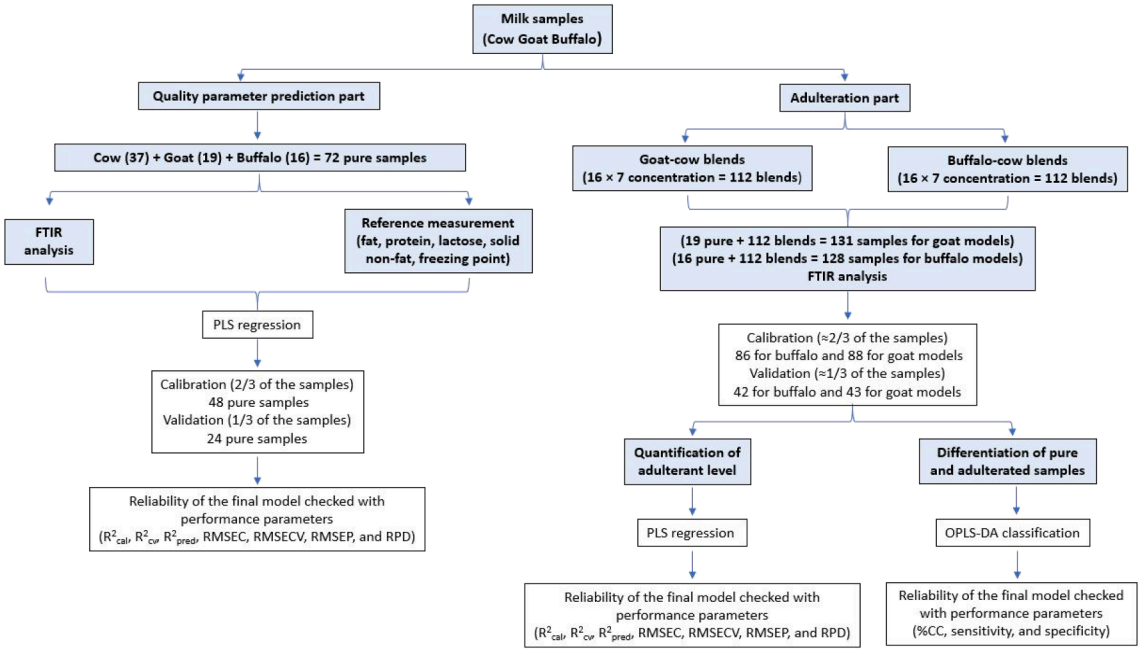

Fig. 1. Flow diagram of the sample preparation and experimental design for prediction and adulteration part.

2.3. Fourier-transform infrared spectroscopy

Spectra of all milk samples were collected in the mid-IR region (4000–650 cm^{-1}) with Perkin Elmer Spectrum 100 FTIR spectrometer (PerkinElmer Inc., USA) having a deuterated tri-glycine sulphate (DTGS) detector. FTIR device equipped with a horizontal attenuated total reflectance (HATR) sampling accessory with ZnSe crystal was used in obtaining the FTIR spectra of pure and adulterated milk samples. For each spectrum, the resolution was set at 4 cm^{-1} and the number of scans was 64. After each analysis, the ZnSe crystal was cleaned with hexane, ethanol and deionized water. Then, the crystal was dried under nitrogen flow prior to each run. All measurements were repeated at least twice.

2.4. Multivariate statistical analysis

SIMCA 14.1 software (Umetrics, Umeå, Sweden) was used for the statistical evaluation of the FTIR spectral information. First, replicated spectral data were averaged then, appropriate pre-treatment techniques as second order derivative (SD), third order derivative (TD) and orthogonal signal correction (OSC) were applied by trial and error approach. The strategy on trial and error approach was based on first application of each pre-treatment to each model and then the most successful model was determined based on performance parameters. Pre-treated data set was randomly divided into calibration (2/3 of total samples) and external validation (1/3 of total samples) sets for both classification and prediction (Fig. 1).

In classification part, orthogonal partial least square-discriminant analysis (OPLS-DA) was used to visualize the separation of adulterated (112 samples) and raw goat (19 samples) and buffalo (16 samples) milk samples by using the pre-treated data. In OPLS-DA analysis, a dummy Y matrix (variable vector) which has two classes (adulterated and non-adulterated samples) was correlated with X matrix (spectral data) [23]. The results of OPLS-DA analysis are shown as a misclassification table and a score plot. Performance parameters such as number of latent variables (LVs), coefficient of determination for calibration (R^2_{cal}) and cross validation (R^2_{cv}) were determined to test the validity of the models. Correct classification rate (%CC), sensitivity and specificity were calculated according to definitions given in literature [24]. The accuracy of the discrimination models is related with the higher %CC along with sensitivity and specificity [25].

In addition, PLS regression was used to determine the levels of adulteration (1–50% v/v) as an authentication approach. Linear calibration models that enable the prediction of chemical parameters (fat, protein, lactose and solid non-fat contents) were also constructed with the same regression technique. PLS regression algorithm is based on the ability to correlate mathematically any quantitative data to a matrix of the property of interest. In this case, Y block represents the measured chemical data and the level of adulteration and X block represents FTIR spectroscopic data. One of the advanced signal correction algorithms, OSC, was used as a pre-treatment method for both PLS regression models. Performance parameters, LVs, R^2_{cal}, R^2_{cv} and external validation (R^2_{pred}) were determined for each chemical parameter and adulteration level models. Root mean square error (RMSE) of calibration (RMSEC), root mean square value of cross-validation (RMSECV), root mean square value of prediction (RMSEP), residual predictive deviation (RPD) and the slope were also used in the performance evaluation. A robust quantification model should possess RPD value greater than 3.0 and slope value varies between 0.9 and 1.1 [26]. R^2 values should be close to one, error values should be small and close to each other in order to obtain a robust prediction model by keeping the balance between the error value ranges created and to reduce the error as much as possible [27].

3. Results and discussion

3.1. Spectral and quality parameter evaluation of the milks

Quality characteristics of raw milks used in this study are presented in Table S1 (Supplementary Material), and Fig. 2 shows raw FTIR spectral profiles of buffalo, goat and cow milks along with their second and third derivative spectra. As it can be seen from spectra, it is hard to detect the differences among milks visually (Fig. 2a). Absorption band at 3650–3000 cm^{-1} can be associated with strong O–H stretching vibrations [28]. The other major peak in the spectra between 1700 and 1500 cm^{-1} is the characteristic of amide I (C-O) and amide II (N–H) absorptions and is correlated with milk proteins [29]. Protein peaks are also located in 1550, 1560–1520, 1300–1230, 1300–1230, and 1100–1060 cm^{-1} regions [29]. In the fingerprint region (near the 1200–1000 cm^{-1}),

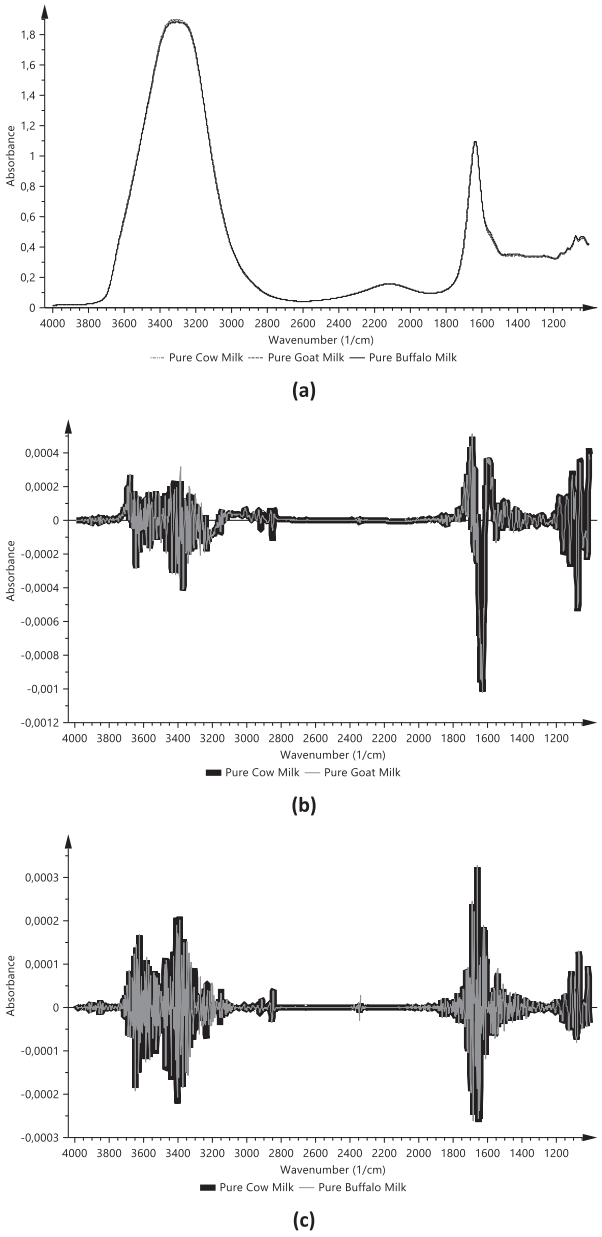

Fig. 2. a) Average raw spectra of pure buffalo, goat and cow milks b) second derivative spectra of pure goat and cow milks c) third derivative spectra of pure buffalo and cow milks recorded between 4000 and 1000 cm^{-1} (Profiles belong to average of 37 cow, 19 goat, and 16 buffalo milk samples).

several emerged peaks could be attributed to C–H stretching, ketone C-O stretching and bending, C-O–H in-plane bending and C-O stretching vibrations of lipids, organic acids, amino acids, and carbohydrate derivatives [28,29]. 1200–900 and 1045 cm^{-1} region are associated with lactose while absorption at 3000–2800 cm^{-1} and 1400–800 cm^{-1} are related with carbohydrates, mainly lactose with smaller amounts of monosaccharides and oligosaccharides in milk composition [29]. Fat related regions are 3000–2800, 1745–1725 and 970 cm^{-1} [29]. Second derivative of pure goat and cow milk samples indicates that 3000–2800 cm^{-1} and 16000–1800 cm^{-1} along with 1000–1200 cm^{-1} regions have different absorption profiles from pure cow milk samples due to compositional differences explained in the following part (Fig. 2b). For pure buffalo and cow milk samples, the same wavelengths but a wider range of 2800–3500 cm^{-1} are responsible for differentiation (Fig. 2c). These findings are also supported with variable importance in projection (VIP) scores (Fig. S1, Supplementary Material). Although spectra were collected in 4000–650 cm^{-1} frequency range, elimination of 1000–650 cm^{-1} part of the spectra due to noise improved the performances of all statistical models.

Average fat contents of buffalo, goat and cow milks in the sample set were 6.43 ± 0.92%, 3.81 ± 0.60% and 5.22 ± 2.73%, respectively (Table S1, Supplementary Material). Average protein contents of the same milks were close to each other and buffalo milks had the highest average protein content of 2.93 ± 0.17%, goat and cow milk followed it with 2.55 ± 0.23% and 2.48 ± 0.20%, respectively. Average lactose contents were determined as 4.14 ± 0.25% for buffalo milk, 3.67 ± 0.34% for goat milk and 3.48 ± 0.28% for cow milk. Buffalo milk had the highest solid non-fat content of 7.69 ± 0.45% followed by goat (6.77 ± 0.61%) and cow (6.48 ± 0.50%) milks. Quality parameters are in the range of the values that could be found in literature although variations in the values and the trends exist in literature studies due to many factors such as animal diet, season and lactation stage [3,30]. Therefore, the values belonging to milks obtained from similar conditions could provide a more accurate comparison.

3.2. Prediction of selected quality parameters from FTIR spectral data

Fat, protein, lactose, and solid non-fat contents as measured parameters were predicted from FTIR spectra of pure milk samples by constructing PLS regression models. A total of 72 samples by combining pure buffalo, goat and cow milks were used in model building and 48 of these samples were used in calibration model and the rest in validation model (Fig. 1). Table 1 lists the prediction performances of the models evaluated by internal and external as well as cross validation parameters. Several pre-treatments were applied to the data and the best performances were obtained with OSC for all models. For fat content prediction, constructed model with five LVs has R^2_{cal}, R^2_{cv} and R^2_{pred} of 0.99, 0.98 and 0.99, respectively. RMSE values (0.15, 0.42, 0.26) of this model are small compared to measurement range and close to each other. Slope of 0.99 and RPD value of 8.9 are the indications of a robust model. All other prediction models have also five LVs (Table 1). Their R^2 values are in the range of 0.98–0.99 and slopes are 0.99 for all of them. Quite high RPD values are obtained for each prediction model. Therefore, it could be concluded that all variables (fat, protein, lactose, and solid non-fat) could be quite accurately predicted from FTIR spectral data.

Several studies that aimed to estimate the properties of especially cow milk such as fatty acid composition [31,32], protein profile [33–35] and mineral content [36] with FTIR spectroscopy are available in the literature and these properties were quite successfully calculated using various chemometric approaches. In the current study, on the other hand, data from three different raw milk types were combined. It was shown that prediction of chemical parameters (fat, protein, lactose, solid non-fat) with FTIR could be achieved quite successfully regardless of the milk source with robust models. Results of the current study are quite comparable with those in the literature although milk types having different compositions and properties were used as the data set. Resulting models have wider ranges of prediction for quality characteristics due to the compositional variability of different milks compared to models developed using single milk type. In addition, statistical models provided good prediction of chemical constituents. Therefore, simultaneous rapid measurement of these parameters for different types of milks could also be helpful in quality monitoring of dairy processes since they affect the final product attributes such as textural and organoleptic properties. Technological suitability characteristics of different milks for various processes along with their nutritional quality are also defined by milk composition [3]. Furthermore, these parameters could provide information regarding the authenticity of the raw milk.

3.3. Prediction of adulteration of buffalo and goat milks with cow milk

OPLS-DA and PLS regression were used to determine the mixing of buffalo and goat milks with cow milk in the concentration range of 1–50% and calibration and validation models of each analysis were built for this purpose (Fig. 1). Table 2 and Fig. 3 show the OPLS-DA statistical parameters of constructed models and score plots for both milk mixtures, respectively. In addition, Table 3 and Fig. 4 provide the PLS regression outputs for the same mixtures.

OPLS-DA score plot (Fig. 3a), which was obtained from TD pretreated spectroscopic profile, indicates a very good separation between pure buffalo and mixtures of buffalo-cow milks. Most of the mixtures are placed on the right half of the ellipse while pure buffalo

Table 2
Statistical parameters of OPLS-DA calibration and validation models of pure and adulterated milk samples obtained from FTIR spectroscopic data.

Milk type	Specifications[1]	Model[2]	% CC[3]	Sensitivity	Specificity
Buffalo	Pre-treatment: TD, LVs:1 + 2, R^2_{cal}:0.90, R^2_{cv}:0.30	Cal	100	100	100
		Val	93	75	95
Goat	Pre-treatment: SD, LVs:1 + 4, R^2_{cal}:0.96, R^2_{cv}:0.32	Cal	100	100	100
		Val	93	80	95

[1] SD: second derivative, TD: third derivative, [2]Models for buffalo milk and goat milk consist of 86 and 88 samples for calibration (cal.) and 42 and 43 samples for external validation (val.), respectively, [3]correct classification rate (accuracy).

Table 1
Statistical parameters of PLS regression models generated from FTIR spectroscopic data for prediction of various chemical measurements of milk samples.

Parameters (%)	Min-Max	Pre-treatment	LVs	R^2_{cal}	R^2_{cv}	R^2_{pred}	RMSEC	RMSECV	RMSEP	RPD	Slope
Fat	1.41–10.88	OSC	5	0.99	0.98	0.99	0.15	0.42	0.26	8.9	0.99
Protein	1.58–3.25	OSC	5	0.99	0.98	0.99	0.02	0.08	0.02	10.4	0.99
Lactose	2.25–4.60	OSC	5	0.99	0.99	0.99	0.03	0.13	0.03	11.3	0.99
Solid non-fat	4.18–8.54	OSC	5	0.99	0.99	0.99	0.06	0.22	0.06	11.6	0.99

OSC: orthogonal signal correction, LVs: latent variables, R^2_{cal}: coefficient of determination for calibration, R^2_{cv}: coefficient of determination for cross validation, R^2_{pred}: coefficient of determination for external validation, RMSEC: root mean square error of calibration, RMSECV: root mean square error of cross-validation, RMSEP: root mean square error of prediction, and RPD: residual predictive deviation.

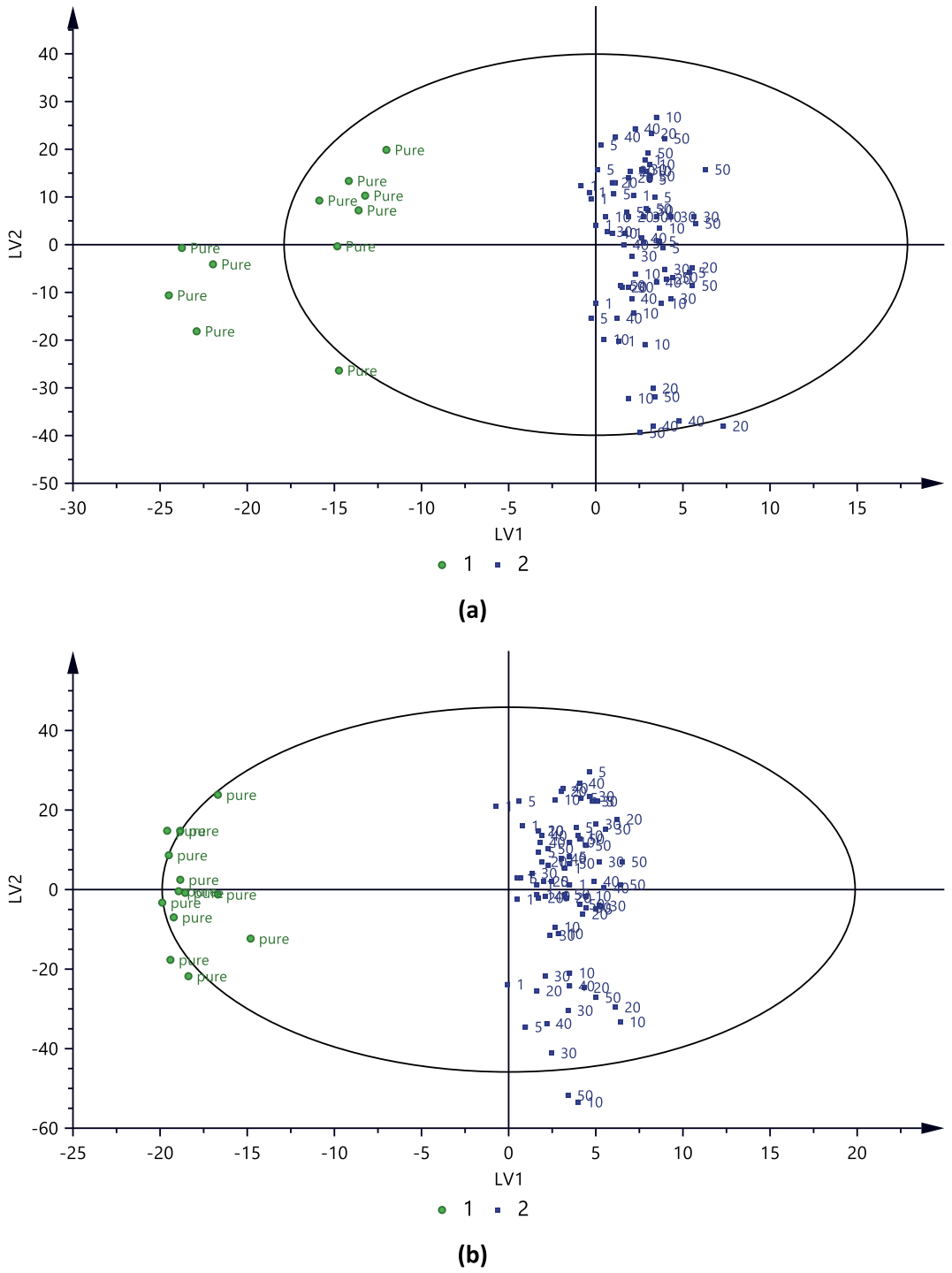

Fig. 3. OPLS-DA score plots for pure and adulterated buffalo milk (a) and for pure and adulterated goat milk (b) samples obtained from FTIR spectroscopic data (1: pure milk samples 2: adulterated milk samples (1–50% (v/v)).

Table 3
Statistical parameters of PLS regression models generated from FTIR spectroscopic data for prediction of adulterant level of milk samples.

Milk type	Pre-treatment	LVs	R^2_{cal}	R^2_{cv}	R^2_{pred}	RMSEC	RMSECV	RMSEP	RPD	Slope
Buffalo	OSC	5	0.98	0.96	0.97	2.31	4.14	3.02	5.8	0.98
Goat	OSC	3	0.99	0.96	0.97	2.14	3.55	2.84	6.2	0.99

OSC: orthogonal signal correction, LVs: latent variables, R^2_{cal}: coefficient of determination for calibration, R^2_{cv}: coefficient of determination for cross validation, R^2_{pred}: coefficient of determination for external validation, RMSEC: root mean square error of calibration, RMSECV: root mean square error of cross-validation, RMSEP: root mean square error of prediction, and RPD: residual predictive deviation.

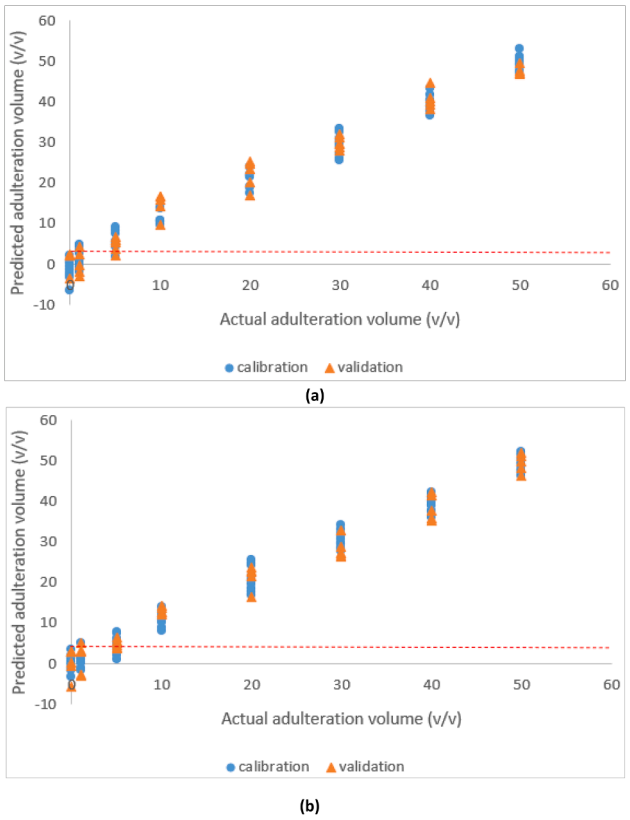

Fig. 4. PLS regression plots of actual versus predicted adulteration volume of cow milk in buffalo milk (a) and cow milk in goat milk (b) generated from FTIR spectroscopic data.

milks are on the far left. Several mixtures with low cow milk concentration (1%) are located on the line separating negative and positive sides of the 1st LV. Statistical parameters indicate that correct classification rates (%CC) of OPLS-DA model (1 + 2 LV) are 100% and 93% (two samples out of 42 samples were falsely identified as adulterated and one sample was misclassified as pure) for calibration and validation, respectively (Table 2). While sensitivity and specificity values of calibration model were calculated as 100%, same values are 75% and 95% for validation model, in order. Frequency ranges of 3800–2800 cm^{-1} and 1800–1000 cm^{-1} having VIP values higher than one are significant variables in developed classification models (Fig. 2 and Fig. S1a, Supplementary Material). These ranges are correlated mainly with water, carbohydrate, protein and fat composition of milks. Therefore, it could be concluded that all major fractions of milks have a role in discrimination as expected and this can be also supported by the differences in chemical compositions of milk samples (Table S1, Supplementary Material) which correlated with differences in FTIR spectra (Fig. 2). PLS regression models were also built to obtain a relation between actual mixture ratios (Table 3) and ratios calculated from spectroscopic data. PLS plot for buffalo-cow mixture is shown in Fig. 4a. Five LVs resulted a PLS model for buffalo-cow mixture with R^2 values in the range of 0.96–0.98, RPD of 5.8 and slope of 0.98. RMSE values are small compared to 1–50% blend ratio. Therefore, this model could accurately predict cow milk amount in goat milk. It is also determined that detection of buffalo-cow milk mixtures at concentrations higher than 5% from PLS regression model is possible (Fig. 4a). There is no comparable study in the literature performed with FTIR spectroscopy for determination of buffalo-cow milk mixtures. However, another spectroscopic method, Raman spectroscopy was successful in differentiation between buffalo milk and buffalo-cow milk with chemometric analysis [13]. In another study, cow α-lactalbumin was used as a marker to detect cow milk in buffalo milk by capillary electrophoresis with minimum 1% limit [8].

Same statistical analyses were also used in evaluating goat-cow milk mixtures (Table 2). SD of FTIR spectroscopic data was used in model building. As can be seen from the score plot (Fig. 3b), goat and cow milk mixtures are very well separated from pure goat milks. This observation from score plot was also confirmed by OPLS-DA model parameters listed in Table 2. OPLS-DA calibration model built with 1 + 4 LV correctly classified 100% of the samples (out of 88 samples, none of the samples were misclassified) while correct classification rate of validation model was 93% (two samples out of 43 samples were falsely identified as adulterated and one sample was misclassified as pure). Sensitivity and specificity calculations yielded values of 100% for calibration and 80% and 95% for validation models, respectively. As in buffalo-cow mixtures, VIP values (Fig. S1b, Supplementary Material) indicated that frequency ranges of major peaks at 3000–2800 cm^{-1} and 1800–1000 cm^{-1}, which are correlated with major constituents as water, carbohydrate, protein and fat, are significant for goat-cow milk OPLS-DA model. PLS regression model was also constructed for goat-cow milk mixtures (Table 3). PLS regression analysis of OSC pre-treated data provided the best results and this model has three LVs and R^2 values of 0.96–0.99. Slope and RPD values are, in order, 0.99 and 6.2 showing that the model is quite accurate and robust. According to Fig. 4b, presence of cow milk in goat milk at ratios higher than 5% could be accurately predicted with FTIR spectroscopy. In a previous study, FTIR spectroscopy was also used to quantify binary and tertiary mixtures of cow, goat and sheep milks with PLS and non-linear kernel PLS (KPLS) [21]. Good predictive value with an error level of 4–6% was obtained for binary mixtures and error level for tertiary mixtures were 3.4–4.9%. Various approaches such as NIR spectroscopy [37], duplex PCR [38], polar metabolite pool measurement with GC–MS [39], primary protein profile determination with 2D-gel electrophoresis [40] and laser induced breakdown spectroscopy [41] were used in detection of cow milk adulteration of goat milk with varying detection levels as low as 0.5% in the literature. Listed literature studies indicate detection limits as low as 0.5% to 6%. Therefore, 5% limit of this study falls in the range of the previous methods, and current method has also the advantages of being rapid and environmentally friendly as other spectroscopic techniques.

4. Conclusion

Evaluation of FTIR spectroscopic data from three types of milks (buffalo, goat and cow) with PLS regression resulted in reliable predictions of several quality parameters of these milks including fat, protein, lactose and solid non-fat contents regardless of milk type. Quick determination of these parameters could be helpful in monitoring the quality of raw milk before processing as well as in their authentication.

This technique was also used in determination of binary mixtures of goat-cow and buffalo-cow milk mixtures and these mixtures were identified at ratios higher than 5% (v/v) level. When it is considered that profitable mixtures prepared by fraudsters are generally at higher levels than the threshold level determined in the present study, use of FTIR spectroscopy technique will help to solve one of the adulteration detection problems for milk.

As a result, this spectroscopic technique could be used for rapid analysis of different types of milks and not only quality parameters but also adulteration at levels higher than 5% could be determined with a single run.

CRediT authorship contribution statement

Sevval Sen: Investigation, Methodology, Writing - original draft, Funding acquisition. **Zahide Dundar:** Investigation, Methodology, Writing - original draft, Funding acquisition. **Oguz Uncu:** Methodology, Formal analysis, Writing - original draft, Supervision. **Banu Ozen:** Conceptualization, Methodology, Writing - original draft, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.microc.2021.106207.

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