A COMPREHENSIVE OVERVIEW OF NEAR INFRARED AND INFRARED SPECTROSCOPY FOR DETECTING THE ADULTERATION ON FOOD AND AGRO-PRODUCTS—A CRITICAL ASSESSMENT

TINJAUAN KOMPREHENSIF SPEKTROSKOPI INFRAMERAH DEKAT DAN INFRAMERAH UNTUK MENDETEKSI PEMALSUAN PADA MAKANAN DAN PRODUK PERTANIAN—PENILAIAN KRITIS

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ABSTRACT

In the past decade, fast and non-destructive methods based on spectroscopy technology have been studied to detect and discriminate against food adulteration and agro-products. Numerous linear and nonlinear chemometric approaches have been developed for spectroscopy analysis. Recently, various approaches have been developed for spectroscopic calibration modelling to detect and discriminate adulteration food and agroproducts. This article discusses the application of spectroscopy technology, including near infrared and infrared, in detecting and discriminating the adulteration of food and agro-products based on recent research and delivered a critical assessment on this topic to serve as lessons from current studies and future outlooks. The current state-of-the-art techniques, including detection and classification of various adulteration in food and agro-products, have been addressed in this paper. Key findings from this study, near infrared and infrared spectroscopy is a non-destructive, rapid, simple-preparation, analytical rapidity, and straightforward method for classification and determination of adulteration in the food and agro-products so it is suitable for large-scale screening and on-site detection. Although there are still some unsatisfactory research results, especially in detecting tiny adductors, these technologies can potentially detect any adulteration in the various food and agro-products at an economically viable level, at least for the initial screening process. In that respect, near infrared and infrared spectroscopy should be expanded to cover all food and agro-products sold in the market. Only then will there be an acceptable deterrent in place to stop adulteration activity in widely consumed food and agro-products ingredients.

ABSTRAK

Dalam satu dekade terakhir, metode cepat dan non-destruktif berdasarkan teknologi spektroskopi telah banyak dipelajari untuk mendeteksi dan membedakan pemalsuan produk makanan dan pertanian. Banyak pendekatan kemometrik linier dan nonlinier telah dikembangkan untuk analisis spektroskopi. Baru-baru ini, berbagai pendekatan telah dikembangkan juga untuk pemodelan kalibrasi spektroskopi dalam mendeteksi dan membedakan pemalsuan produk makanan dan pertanian. Artikel ini membahas penerapan teknologi spektroskopi, termasuk inframerah dekat dan inframerah, dalam mendeteksi dan membedakan pemalsuan produk makanan dan pertanian berdasarkan penelitian terbaru dan menyampaikan penilaian kritis tentang topik ini untuk dijadikan pelajaran dari studi saat ini dan pandangan dimasa depan. Teknik mutakhir saat ini, termasuk deteksi dan klasifikasi berbagai pemalsuan dalam produk makanan dan pertanian, telah dibahas dalam makalah ini. Temuan utama dari penelitian ini, spektroskopi inframerah dekat dan inframerah adalah metode non-destruktif, cepat, sederhana, kecepatan analitis, dan metode yang mudah untuk klasifikasi dan penentuan pemalsuan dalam produk makanan dan pertanian sehingga cocok untuk skala besar, penyaringan dan deteksi di tempat. Meskipun masih ada beberapa hasil penelitian yang tidak memuaskan, terutama dalam mendeteksi adduktor kecil, teknologi ini berpotensi mendeteksi pemalsuan dalam berbagai produk makanan dan pertanian pada tingkat yang layak secara ekonomi, setidaknya untuk proses penyaringan awal. Dalam hal ini, spektroskopi inframerah dekat dan inframerah harus diperluas untuk mencakup semua produk makanan dan pertanian yang dijual di pasar. Hanya dengan demikian akan ada pencegah yang dapat diterima untuk menghentikan aktivitas pemalsuan bahan makanan dan produk pertanian yang dikonsumsi secara luas.

INTRODUCTION

In today's worldwide economy, concerns about food authenticity are a top priority. Customers' primary focus has changed to the originality of food and agro-products commodities, due to the growing desire for local products (*Amirvaresi et al.*, 2021; *Wongsaipun et al.*, 2021; *Tao et al.*, 2021). As a result, indigenous food and agro-products are frequently chosen over imported ones. Consumers consider freshness and geographical origin when selecting high-quality food products to consume daily, such as meat, flour, flavouring, herbs, and spices.

The increasing population and high cost of produced food and agro-products have created opportunities to use adulteration in postharvest processing. The quality control of these products still relies on laboratory testing based on chemical analysis. Regrettably, these methods seem expensive, complicated to use, usually time-consuming and require a sample preparation step before analysis, in turn, they need many kinds of chemical solvent. In that respect, the option of spectroscopy technology, including near infrared and infrared, offers a valid key to overcoming some of the abovementioned disadvantages since they allow performing a non-destructive evaluation, rapid, easy, eco-friendly, and directly in situ (*Galvin-King et al.*, 2021a; *Silva et al.*, 2020; *Ndlovu et al.*, 2019). This is why researchers have worked over the years to find another application as standard analysis in various fields, especially food science (*Ozaki et al.*, 2021).

According to the recent literature, many studies have been using spectroscopy technology, including near infrared and infrared, to detect and classify the adulteration of food and agro-products. Yet, to date, no comprehensive study has reported on it or provided a critical assessment on this topic. Therefore, the article presents an overview of the application of near infrared and infrared spectroscopy in detecting and discriminating the adulteration of food and agro-products based on recent research.

METHODS

Applications of spectroscopy technology, including near infrared and infrared, to assess fraud, particularly in food and agro-products, have increased each year (Fig. 1). Research papers were searched in February 2022 via the electronic database Scopus (www.scopus.com). The keyword for finding the research papers using "NIR" or "near-infrared" and "adulteration". From the first search, research papers can be categorized into an article (447), conference article (56), review (41), book chapter (15), conference review (5) and short survey (1). Most of the articles published come from China (33.6%), followed by Brazil (11.7%), the United States (8.3%), Spain (6.2%), the UK (4.8%), India (4.4%), Italy (4.2%), Ireland (4.1%), Malaysia (3.2%), and France (3.0%). The most popular keywords were infrared device (50.4%), near infrared spectroscopy (50.4%), adulteration (29.9%), least squares approximations (23.7%), chemometrics (20.4%), principal component analysis (19.6%), and spectroscopy, near infrared (18.8%).

Subsequently, the abstracts of the paper were investigated to include or exclude them in this article. From there, 447 documents were further examined, and inappropriate documents were excluded. Excluded research papers were carried out because they did not use near infrared or infrared spectroscopy to detect adulteration, papers that did not use food and agro-products as the main object of the study, conference papers, book chapters, conference reviews, short survey, and review articles. A total of 126 documents were used in the further study. An overview of the research papers is shown in Table 1 to Table 3.



Fig. 1 – Metadata Scopus record of research paper per annum and cumulative total of articles until 2021

NEAR INFRARED AND INFRARED SPECTROSCOPY FOR FOOD AND AGRO-PRODUCT

Infrared (IR) spectroscopy uses the spectral range between 800 and 500000 nm, which can be further subdivided into the far IR (FIR: 25000 to 500000 nm), the mid IR (MIR: 2500 to 25000 nm), the near IR (NIR: 800 to 2500 nm), and ultraviolet-visible (UV-VIS: 200 to 780) (*Reich*, 2016; *Ozaki et al.*, 2021). The application of near infrared and infrared spectroscopy for food and agro-products has long been known in the industrial world and continues to expand today (*Wesley et al.*, 1995). In general, this technology is utilized to evaluate food and agro-products in the form of quantitative and qualitative analysis. The wavelengths used vary widely from near infrared spectroscopy (780–2500 nm) to MIR spectroscopy (2500–25000 nm) (*Santos et al.*, 2021; *Alamar et al.*, 2020; *Pereira et al.*, 2019). Meanwhile, some researchers combine the wavelength of the near infrared spectroscopy (*Pandiselvam et al.*, 2022; *Valinger et al.*, 2021b; *Ndlovu et al.*, 2021b).

Likewise, several wavelength ranges in near infrared and infrared spectroscopy for food and agroproducts that have been studied are shown in Fig. 2. Unfortunately, although it has limitations in the spectral range, visible near infrared technology (340–780 nm) is still used to detect and discriminate adulteration in food and agro-products. However, full-wavelength near infrared (780–2500 nm) and infrared (2500–16000 nm) spectroscopy with wider wavelengths are more commonly used for detecting adulterations of food and agroproducts. On the other hand, some studies also combine ultraviolet, visible, and near infrared wavelength ranges known as UV-VIS-NIR (325–2500 nm).



Fig. 2 - Wavelength range of near infrared and infrared spectroscopy technology

Near infrared spectroscopy technology (780-2500 nm)

The spectral band represents the interaction of molecules with the near infrared wavelength. The chemical content on the samples tends to absorb specific frequencies of light when a sample is irradiated with near infrared spectroscopy. Thus, near infrared spectroscopy can provide a fingerprint of the content in a sample, especially in food and agro-products. Near infrared spectroscopy has been used in a wide range of investigations to find adulteration in foods and agro-products such as livestock (*dos Santos Pereira et al.*, 2021a; *Teixeira et al.*, 2021a; *Mabood et al.*, 2020), flour (*Ndlovu et al.*, 2021a; *Ayvaz et al.*, 2021b; *Tao et al.*, 2021), liquid agro-product (*Tan et al.*, 2021; *Valinger et al.*, 2021b; *Du et al.*, 2021b), and herbs and spices (*Castro et al.*, 2021; *Cantarelli et al.*, 2020; *Rukundo and Danao*, 2020).

Near infrared spectroscopy offers a fast, effective, and low-cost alternative procedure that can provide clues about the chemical content and physical properties of the samples. The more affordable near infrared spectroscopy technology is due to the fact that more and more mechatronic industries are developing spectrometer packages that are simpler, more portable, and smaller in size than the benchtop types available in the laboratory.

Several studies have reported that it detects adulteration in food and agro-products using portable near infrared spectroscopy in the wavelength range of 908–1676 nm, 950–1650 nm, 1351-2551 nm and 1600–2400 nm (*dos Santos Pereira et al.*, 2021b; *Oliveira et al.*, 2020; *Aykas and Menevseoglu*, 2021; *Correia et al.*, 2018; *Silva et al.*, 2020; *Torres et al.*, 2021; *Santos et al.*, 2013). Although many industries have developed near

infrared spectroscopy technology packages, unfortunately, they will still be relatively expensive over the next few years. On the other hand, near infrared spectroscopy instruments generate a large amount of data that require an adequate method to build useful analytical information. Combining chemometric and near infrared spectroscopy techniques is required to collect as much associated information from the spectral data as possible (*Genis et al.*, 2021). In this case, chemometrics is the science of extracting information from a chemical system through data-driven methods.

The use of a wider spectral region allowed them to obtain more information related to the stretching and deformation vibrations of the C–H, O–H, and N–H groups that are abundant in a sample. For example, from a honey sample, wavelengths in the visible region up to near infrared (400–2500 nm) are related to those compounds in the honey that absorb in the blue-violet range, giving the characteristic orange-amber color of the honey (*Yang et al.*, 2020). In the near infrared region, the wavelength at 1451 nm is related to the first overtone of the vibrational mode of the O–H stretch from water (*Huang et al.*, 2020a). Therefore, signal regions of near-infrared and infrared spectra are needed to understand the compound in the samples with greater precision. With that in mind, the next step is to focus only on the few wavelength regions that can provide the information that correlates with the compounds in our sample. In addition, portable near infrared spectroscopy with a narrow wavelength region can be utilized, while providing high accuracy.

Infrared spectroscopy technology (2500–16000 nm)

Infrared spectroscopy data cover the 2500 to 16000 nm range used to represent fundamental vibrations, molecular overtones, and combination vibrations. The absorption areas are predominantly composed of hydrogen-containing groups related to the acid, oil content, protein, sugar, and water of food and agroproducts. Consequently, the spectral contains chemical information by reflecting the molecular structures from the samples.

Several recent studies have been carried out using infrared spectroscopy technology to detect and discriminate adulteration of food and agricultural products for livestock products, including milk and eggs (*Hosseini et al.*, 2021; *Botelho et al.*, 2015; *Uysal and Boyaci*, 2020). In addition, flour products have been investigated for products including pistachios and peppers (*Aykas and Menevseoglu*, 2021; *Galvin-King et al.*, 2020a). Liquid products have also been studied for products including yogurt, guava pulp, durum wheat pasta, and butter oil (*Temizkan et al.*, 2020b; *Alamar et al.*, 2020; *De Girolamo et al.*, 2020b; *Pereira et al.*, 2019). For herbs and spices, products have been studied, including those of black pepper, garlic, and saffron (*Wilde et al.*, 2019; *Galvin-King et al.*, 2021a; *Amirvaresi et al.*, 2021). Nevertheless, the most challenging thing for researchers in adulteration studies in this range spectral is to explain the connection between absorption in the spectral region with the chemical content of food and agro-products. Occasionally, the various intrinsic properties to be determined usually lead to non-linear patterns. Finally, many linear and non-linear chemometric approaches have been developed for quantitative and qualitative analyses to tackle this problem.

ANALYSIS DATA

Spectral data analysis is the most important part of obtaining the information contained therein. In general, the procedure that must be followed in extracting the information in the near infrared and infrared spectra, especially related to the purity of food and agro-products, is presented in Fig. 3. Food and agro-products that have been adulterated with an adulterating agent will create different infrared spectra data as a result of the various functional groups in the material. However, this will not necessarily produce information without developing a calibration model, which is followed by testing to build a predictive model. Furthermore, the predictive model performance should also be tested with several unknown datasets to create a proven model.

In many cases of adulteration of food and agro-products, the processing and pre-treatment steps are very important to reduce noise spectra data. Furthermore, many linear and nonlinear chemometric approaches, including Partial Least Squares Regression (PLSR), Principal Component Regression (PCR), Support Vector Machine (SVM), and Artificial Neural Network (ANN), have been developed to quantify the physical and chemical properties of food and agricultural products to acquire information from spectral data. The last two algorithms are the newest, along with the k-nearest neighbour (k-NN), the Convolutional Neural Network (CNN), and the Radial Basis Function Neural Networks (RBFNN) based on machine learning, which are reported to produce the best predictive models compared to PLSR and PCR (*Xie et al.*, 2008; *Alamar et al.*, 2020; *Liu et al.*, 2021).



Fig. 3 – Procedure of model construction and performance evaluation

Pre-processing data

The difficulty of using spectral data for food and agro-products quality assessment stems from the need for a strong and accurate model with low sensitivity and low-intensity spectral data. Almost all studies involving near infrared and infrared spectroscopy use pre-processing data to avoid noise from light scattering, instrumental drift, particle size variation, and also high overlaps between combination bands and overtones to address this problem. Pre-processing is a method used to go from raw data to clean data ready for analysis including removing baseline artifacts, peak selection, or alignment. Pre-treatment is to transform the pre-processed data to make them suitable for analysis, including normalization, scaling, transformations, and removing any outliers in the data.

The application of pre-processing does not always provide the best results. For example, *Valinger et al.* (2021b) did not apply pre-processing or pre-treatment to its spectral data. However, they could provide an RPD value greater than 3 using the PLSR algorithm to detect fructose corn in honey. However, *Santos et al.* (2021) reported that pre-processing of SNV to detect adulteration of cocoa solids gave better results than without the application of pre-processing. Therefore, we conclude that applying pre-processing to near infrared and infrared spectroscopy data is a procedure that must be tested regardless of the results obtained.

Linear approach

A linear approach in near infrared and infrared spectroscopy data analysis will be successful if a linear association exists between the absorbance spectra and predicted content, more commonly referred to as the Beer-Lambert law. It is capable of conducting qualitative and quantitative analyses of adulteration in food and agro-products. The linear chemometric methods that were used most frequently to formulate a qualitative and quantitative analysis of adulteration in food and agro-products were PLSR, PCR, Partial Least Squares Discriminant Analysis (PLS-DA) and Principal Component Analysis-Linear Discriminant (PCA-DA) (*Kazazić et al.*, 2001; *Paradkar et al.*, 2002a; *Gayo et al.*, 2006).

In general, linear chemometric methods from IR spectroscopic data can be evaluated with several parameters. The parameters most used, including calibration and cross-validation (CV), are the determination coefficients (R²), the coefficients correlation (r), the Root Mean Square Error (RMSE) and the Standard Error (SE). In addition, some use difference average value between predicted and measured values (Bias), Range Error Ratio (RER), and Predicted Deviation Ratio (RPD). Each parameter has its own purpose in evaluating the model. Coefficient determination indicates how well a model performs in terms of the proportion of variance in the dependent variable predicted by the independent variables. The RPD shows the robustness of the model. SE and RMSE indicate the level of precision and accuracy of the developed model.

Non-linear approach

Another method to analyse near infrared and infrared spectroscopy data of adulteration in food and agro-products associated with chemometrics is a non-linear approach. This approach is required when the

Table 1

connection between the spectral absorption region of the IR spectroscopy is non-linear. The origin of these non-linear relationships is diverse and challenging to identify, but according to *Ramírez-Morales et al.* (2016), in some cases, due to the disparities in viscosity, temperature, pH, particle dimensions, and chemical content. Calibration is generally achieved utilizing non-linear methods and multivariate analysis for this reason. A reasonable variable selection aimed at collecting a small sub-group with lower sensitivity to non-linear or excluding the most wavelengths is usually effective in enhancing the model's performance (*Kaufmann et al.*, 2022; *Pandiselvam et al.*, 2022).

The research that applies a non-linear approach in chemometrics for detecting and authenticating adulteration on food and agro-product is currently in constant expansion. As mentioned before, a non-linear approach to analysing near infrared and infrared spectroscopy data can also perform qualitative analysis and quantitative prediction of adulteration in food and agro-products. Machine learning-based chemometric research is rapidly expanding at the moment. ANN, CNN, k-NN, RBFNN, RF, SVM are also more reported to analyze IR spectroscopy data of adulteration in food and agro-product as these techniques are based on pattern recognition (*Weng et al.*, 2020; *Ding and Xu*, 2000; *Le Nguyen Doan et al.*, 2021).

SOME CASE ADULTERATION ON FOOD AND AGRO-PRODUCTS

Near infrared and infrared spectroscopy analysis has been applied to both detecting and discriminating adulteration of food and agro-products. Qualitative evaluation can be the detecting of adulteration in livestock products, flour products, liquid agro-product, and herbs and spices (Table 1). In contrast, the quantitative study concentrates on predicting multiple contents adulteration of food and agro-products has been reported quite a lot recently (Table 2). In the present studies, various IR spectroscopy ranges are utilized for the quantitative and qualitative analysis of food and agro-products, including near infrared and infrared spectroscopy data (Table 3).

	Some qualitative study of food and agro-product adulteration								
#	Source	Objective	Adulterant	Range of	The be	st of	Prediction		
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results		
1	(de Araújo et al., 2021)	Gourmet ground roasted coffees (90)	Traditional and superior coffees	1205 – 2128	Offset correction	SIMCA	Specificity = 100%		
2	(Srinuttrakul et al., 2021)	Hom Mali ríce (170)	Rice from northern and north-eastern regions of Thailand	740 – 1070 2500 – 22222	MSC+ 1 st dev	PLS-DA	Accuracy =84.85 – 86.96% Accuracy = 96.97 –100%		
3	(Tan et al., 2021)	Stingless bee honey (30)	High fructose corn syrup	900 – 1700	Cutting + Gaussian smoothing	LR	Accuracy = 98.2%		
4	(dos Santos Pereira et al., 2021a)	Goat milk (146)	Cow milk	900 – 1650	Moving mean + Baseline offset	iSPA-PLS- DA	Accuracy = 98.3%		
5	(Shannon et al., 2021)	Basmati rice (1399)	Other varieties basmati rice	740 – 1070	Raw	PLS-DA	F1_score = 0.93		
6	(<i>Tao et al.</i> , 2021)	Wheat flour (48)	Eight varieties of cassava flour	1150 – 2150	Raw	PLS-DA	Accuracy = 97.53%		
7	(Galvin-King et al., 2021b)	Garlic (117)	12 types of white powder	833 – 2500 2500 – 18182	SNV + 1 st dev SG	OPLS-DA	Youden index = 0.98 Youden index = 1		
8	(<i>Teixeira et al.</i> , 2021b)	Yogurt and Cheese from goat milk (576)	Cow milk	1000 – 2500	Smoothing + 2 nd dev SG	PLS-DA	Sensitivity = 99.2 – 100% Specificity = 99.2 – 100%		
9	(<i>Torres et al.</i> , 2021)	Sweet almonds (216)	Bitter almonds	950 – 1650	SNV + 1 st dev SG	PLS-DA	Non-error rate = 86 – 100%		
10	(Le Nguyen Doan et al., 2021)	High-quality rice (200)	Low-quality rice	740 – 1070	1 st dev SG + mean centered	PLS-DA	Accuracy = 82.6%		
11	(Cantarelli et al., 2020)	Cinnamon verum (120)	Cinnamon cassia	940 – 1640	Raw	PNN	Accuracy = 99.25%		
12	(<i>Huang et al.</i> , 2020b)	Honey (224)	Syrup	1000 – 2500 2222 – 12500	2 nd dev SG	SVMC	Accuracy = 100%		

#	Source	Objective	Adulterant	Range of	Range of The best of		Prediction
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results
13	(Galvin-King et	Powdered	Varying seed/pod	833 –	SNV + 1 st + 2 nd	OPLS-DA	R2 = 0.85
	<i>al.</i> , 2020b)	paprika (159)		2500	dev SG		
				2500 –	SNV + 1 st dev		R2 = 0.94
				18182	SG		
14	(Alamar et al.,	Guava pulp	Sugar and water	1000 -	MSC	k-NN	Accuracy =
	2020)	(240)		2500			100%
				2500 -			Accuracy =
15	(Do Cirolomo at	Durwent	Durum wheet peets from	25000	Maan baadina		100%
15		Durum wheat	Argontino	1000 -	Mean Daseline	PLS-DA	Accuracy = $07 100\%$
	al., 2020a)	pasta 110111 Italy (280)	Argentina	2500 -			97 - 100%
		naly (200)		25000	detrending		96 - 97%
16	(Teixeira et al	Goat milk	Water urea bovine	1000 -	1 st dev SG +	PLS-DA	Precision -
10	2020)	(600)	whey, and cow's milk	2500	SNV	I LO DA	100%
17	(Visconti et al	Grated	Microcrystalline	1000 -	1 st dev SG	PLS-DA	Precision =
	2020)	cheese (196)	cellulose, silicon dioxide.	2500			100%
	,	()	wheat-flour, wheat-				
			semolina, sawdust				
18	(Jahani et al.,	Lime juices	Water and citric acid	900 -	MSC	k-NN	Precision =
	2020)	(56)		1700			100%
19	(Wilde et al.,	Black pepper	papaya seeds, chili and	833 –	SNV + 1 st dev	OPLS-DA	Precision =
	2019)	(126)	non-functional black	2500	SG		90 – 100%
			pepper material	2500 –			Precision =
				25000			92 – 100%
20	(Karunathilaka et	Milk powder	11 potential adulterants	800 -	SNV + 1 st dev	SIMCA	Accuracy =
04	al., 2018)	(383)	Mala as is a	2500	SG		100%
21	(Chen et al.,	MIIKS (102)	Melamine	1000 -	SNV	OC-PLS	Accuracy =
22	2017) (Shop of ol	Southoon	Six types of per protein	2500	1 st day SC		89% Sopoitivity –
22	(Shen et al., 2016)	Soybean mool (88)	six types of non-protein	1202 -		FLS-DA	300%
23	(Ziealer et al	Wheat	Bread wheat shelt	1200 -	1 st dev SG		
20	2016)	kernels and	durum emmer and	2400 650		I LO DA	80 - 100%
	2010)	flours (1225)	einkorn	- 2500			00 10070
24	(<i>Xu et al.</i> , 2015)	Tea (100)	Exogenous amino acids	833 -	SNV	PLS-DA	Accuracy =
	(2500			0.936
25	(Schmutzler et	Pork meat	Pork fat	833 –	2 nd dev SG	SVMC	Accuracy =
	al., 2015)	(84)		2500			83.3%
26	(Botelho et al.,	Raw cow milk	Water, starch, sodium	2500 -	1 st dev SG +	PLS-DA	Sensitivity =
	2015)	(155)	citrate, formaldehyde,	16667	Smoothing		88.5 – 100%
			and sucrose				
27	(Ding et al.,	Sweet potato	purple and white sweet	700 –	Selection	LDA	Accuracy =
	2015)	powder (116)	potato	2500	wavelength		100%
~~				1000	using GA-PLS	011404	•
28	(Lopez et al.,	Hazelnut	Almond paste and	1000 -	Offset	SIMCA	Accuracy =
00	2014) (7 /	paste (135)	Chickpea flour	2740	correction	0)/11/0	96.3%
29	$(\angle nang et al., 2014)$	Raw cow milk	pseudo proteins (urea,	1000 -	SNV	SVMC	Precision = 06.62%
	2014)	(800)	animonium nitrate,	2500			90.02%
			thickeners (devtrin and				
			Starch)				
30	(Xu et al. 2013a)	Chinese	Extraneous adulterants	1000 -	2 nd dev SG	OC-PLS	Specificity -
00	(700 01 01., 20100)	alutinous rice	unwanted variations	2500	2 00/00	00120	0.92
		flour (215)		2000			0.02
31	(<i>Xu et al.</i> , 2013b)	Chinese	Edible gelatine.	833 –	SNV	OC-PLS	Specificity =
-	(,	yogurt (257)	industrial gelatine, soy	2500	-		0.95
		,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	protein powder				
32	(<i>Xu et al.</i> , 2013c)	Lotus root	Four cheaper starches	833 –	SNV	SIMCA	Specificity =
		powder (85)		2500			0.94
33	(Chen et al.,	Honey (144)	High fructose corn syrup	1000 –	1 st dev SG +	PLS-DA	Accuracy =
	2011)			2500	smoothing +		96.88%
			A	101-	mean centering	0.4	
34	(<i>Zhu et al.</i> , 2010)	Honey (135)	Sweeteners materials	1000 -	SNV +	SVM	Accuracy =
~-	(Min at -1, 0000)	Dune hard	Mater	2500	Smoothing SG		95.1%
35	(Xie et al., 2008)	Pure bayberry	vvater	800 -	SINV	KRENN	Accuracy =
36	(Downey of al	Juice (129) Honey (200)	Fructose and ducose	2400 400 -	2nd day SC		97.02 Accuracy -
50	(2003)	(300)	i nuclose and giucose	2498	2 464 30	1 LO-DA	96%
				2700			00/0

1st dev SG = First derivatives Savitzky-Golay; 2nd dev SG = Second derivatives Savitzky-Golay; iSPA-PLS-DA = Intervals SPA – Partial least squares – algorithm discriminant analysis; k-NN = k-nearest neighbour; LDA = Linear discriminant analysis; LR = Logistic Regression; MSC = Multiplicative scatter correction; OC-PLS = One class – partial least squares; OPLS-DA = Orthogonal partial least squares – discriminant analysis; PLS-DA = Partial least squares – discriminant analysis; NV = Partial least squares – discriminant analysis; SNV = Partial least squares – discriminant analysis; SNV = Standard normal variate; SIMCA = Soft independent modelling of class analogy; SVMC

= Support vector machines classification.

Table 2

	Some quantitative study of food and agro-products adulteration							
#	Source	Objective	Adulterant	Range of	The best	of	Prediction	
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results	
1	(<i>Ndlovu et al.</i> , 2021a)	Green banana flour (72)	Wheat flour	400 – 2500	SNV + Baseline	PLSR	RPD = 3.9	
2	(<i>Ndlovu et al.</i> , 2021b)	Green banana flour	Wheat flour	400 – 2500	2 nd dev + Detrend	PLSR	RPD = 6.24	
3	(<i>Ayvaz et al.</i> , 2021b)	Einkorn flour	Wheat flour	1000 – 2500	MN + MSC + 1 st dev	PLSR	RPD=19.3	
4	(Santos et al., 2021)	Cocoa solids (110)	Cocoa solids content	1100 – 2500	SNV	PLSR	RPD = 31.09	
	,	· · ·		2500 – 16667			RPD = 17.28	
5	(<i>Valinger et al.</i> , 2021b)	Acacia honey (135)	Fructose corn syrup	325 – 900; 904 – 1699	Raw	PLSR	RPD = 3.32	
6	(Wongsaipun et al., 2021)	Thai Jasmine Rice (423)	3 type rice	400 – 2498	Normalization	PLSR	RMSEP = 2.6; R ² p = 0.98	
7	(Castro et al., 2021)	Saffron (38)	Onion, Calendula, Pomegranate and Turmeric	1000 – 2500	2 nd dev SG + SNV	MCR-ALS	RMSEP = 0.8 - 2.3	
8	(<i>Liu et al.</i> , 2021)	Infant formula (200)	Hydrolyzed leather protein and melamine	900 – 1700	1 st dev	CNN	R ² p=0.96 – 0.99	
9	(Aykas and Menevseoglu,	Powdered Pistachio	Powdered green pea and peanut	2500 – 15385	2 nd dev SG + Smoothing	PLSR	rval = 0.99	
	2021)	(19)		1351 – 2551			rval = 0.99	
10	(<i>Masithoh et al.</i> , 2021)	Arenga pinnata	Coconut sugar	1000 – 2500	MSC	PLSR	RMSEP = 12.42	
		sugar (187)		2500 – 15385	Normalization		RMSEP = 6.95	
11	(Genis et al., 2021) (Silva et al., 2020)	(143)	spinach nut	908 – 1695		PLSR	4.69 – 7.87	
12	(Silva et al., 2020)	chicken (150)	Eeer, pork	908 – 1676	MSC		-4.7	
13	(<i>Yang et al.</i> , 2020)	honey (93)	Five different syrups	400 – 2500 1100 – 2500	2 ^m dev SG	PLSK	3.61	
14	(<i>Rukundo et al.</i> , 2020)	Dried turmeric powder (120)	Metanil yellow	780 – 2500	1 st dev SG	PLSR	RPD = 10.3	
15	(Uysal and Boyaci, 2020)	Liquid egg (100)	Water	1000 – 2500 2500 – 25000	Baseline, autoscale, smoothing, 1st	PCR PCR	RMSECV = 0.8 - 0.74 RMSECV = 0.12 - 17.4	
16	(<i>Ndlovu et al.</i> , 2019)	Unripe banana flour (82)	Wheat flour	447– 1005	2 nd dev SG	PLSR	RPD = 12.02	
17	(<i>Kar et al.</i> , 2019)	Turmeric powder (200)	Corn starch	1000 – 2500	SNV + 1 st dev SG	PLSR	RMSEP = 0.26; R ² p = 0.99	
18	(<i>Pereira et al.</i> , 2019)	Butter oil (33)	Soybean oil	833 – 2500 2500 –	Raw	PLSR	RPD = 21.68 RPD = 12.27	
19	(Yasmin et al., 2019)	Cinnamon Powder (195)	Lower quality cinnamon Powder	25000 1000 - 2500 2857 - 15285	2 nd dev SG	PLSR	$R^{2}p = 0.97;$ RMSEP = 2.2 $R^{2}p = 0.96;$ RSEP = 2.5	
20	(<i>Lukacs et al.</i> , 2018)	Whey protein	Urea, L-taurine, L- histidine	800 – 2750	Smoothing, SNV,	PLSR	$R^{2}p > 0.98$	
21	(<i>Da Silva Dias et al.</i> , 2018)	Raw milk (50)	Water	1200, 1450, 1530	Raw	MLR	R ² _p = 0.96, RMSEP = 0.018	
22	(<i>Picouet et al.</i> , 2018)	Sunflower oil (138)	Mineral oil	1000 – 2200	Baseline, MSC, SNV	PLSR	RMSEP = 0.23 – 1.26	
23	<i>(Kar et al.</i> , 2018)	Turmeric Powder (248)	Metanil yellow	1000 – 2500	1 st dev SG	PLSR	$R^2p = 0.91$	
24	(<i>Correia et al.</i> , 2018)	Arabica coffee (125)	Robusta coffee, corn, peels, and sticks	908 – 1676	1 st dev SG	PLSR	RPD = 64.23	

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Table 3

#	Source	Objective	Adulterant	Range of	The best of		Prediction
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results
25	(<i>Liu and Zhou</i> , 2017)	Apple juice (31)	Water	830 – 2490	MSC	SPA-PSO- PLS	R ² p = 0.99; RMSEP = 0.063
26	(<i>Bázár et al.</i> , 2016)	Honey (492)	High fructose corn syrup	1100 – 2500	Smoothing + SNV + 2 nd dev SG	PLSR	R ² cv = 0.987; RMSECV = 1 48
27	(<i>Dvorak et al.</i> , 2016)	Goat milk for cheeses (48)	Cow's milk	1000 – 2500	Raw	PLSR	$R^2cv = 0.783$
28	(Winkler-Moser et	Coffea arabica (84)	Corn	400 - 2500	1 st dev SG	PLSR	$R^2cv = 0.974$
29	(Kumaravelu and Gopal, 2015)	Honey (160)	Jaggery	400 – 2500	Smoothing + SNV	PLSR	$R_{p}^{2} = 0.99$
	(Mouazen and Al- Walaan, 2014)	Honey (345)	Glucose syrup	305 – 2200	SNV + 1 st dev SG + Smoothing	PLSR	R²p = 0.78, RPD = 2.06
30 31	(<i>Lohumi et al.</i> , 2014)	Onion powder (180)	Corn starch	1000 – 2500	SNV	PLSR	$R^2p = 0.90$
				2500 – 15385			R ² p = 0.98
32	(Vichasilp and Poungchompu, 2014)	Beef and chicken Meatballs (140)	Pork meat	1000 – 2500	Raw	PLSR	R ² v = 0.88 – 0.83
	(<i>Wang et al.</i> , 2014)	Òat Ílour (220)	Wheat flour	833 – 2500	2 nd dev SG	PLSR	RMSEP = 1.975
33 34	(Santos et al., 2013)	Bovine milk (744; 372 – 837)	Tap water, whey, synthetic milk, synthetic urine, urea, and hydrogen	1600 – 2400 2500 – 15385	Raw	PLSR	$R^2 v = 0.92$ $R^2 v = 0.92 - 0.98$
35	(<i>Öztürk et al.</i> , 2010)	Olive oil (160)	Soybean, cotton, corn, canola and	1000 – 2500	Raw	GILS	SEP = 2.93 – 5.86 rv = 0.90
36	(<i>Mishra et al.</i> ,	Honey (56)	Jaggery syrup	1380 -	Raw	PLSR	= 0.99 R ² v = 0.66
37	(<i>Pizarro et al.</i> , 2007)	Arabica coffee powder (191)	Robusta coffee powder	1100 – 2500	1 st dev SG + OWAVEC	PLSR	R ² p = 1
38	(Özdemir and Öztürk 2007)	Olive oil (52)	Sunflower and corn oil	1000 – 2500	Raw	GILS	$R^2p = 0.99$
39	(<i>Gayo and Hale</i> , 2007) 2007)	Atlantic blue crabmeat (110)	Blue swimmer crabmeat	400 – 2498	1 st dev SG	PLSR	R ² p = 0.98
40	(Cocchi et al., 2006)	Durum wheat	Bread wheat flour	400 – 2498	SNV	PLSR	RMSEP =
41	(Gayo et al., 2006)	Crab meat	Surimi-based imitation crab meat	400 – 2498	1 st dev SG	PCR	$R^2p = 0.99;$ SEP = 0.24
42	(Jha and Matsuoka, 2004)	Cow Milk (125)	Urea, NaOH, Oil, shampoo	700 – 1124	MSC	MLR	$R^2 v = 0.58 - 0.98$
43	(Uddin and Okazaki, 2004)	Fresh (162)	Frozen-thawed fish	1920 – 2350	2 nd dev SG	MLR	$R^2c = 0.95 - 0.99$
44	(<i>Maraboli et al.</i> , 2002)	Milk powder (155)	Vegetable proteins	1100– 2500	1 st dev SG	MLR	$R^2p = 0.993$
45	(Rodriguez- Saona et al., 2001)	Fruit juices (60)	Sugars	1000 – 2500	2 nd dev SG	PLSR	R ² p = 0.99
46	(<i>Wesley et al.</i> , 1995)	Olive oil (310)	Corn oil, sunflower oil, raw olive residue oil	800 – 2500	1 st dev SG	PLSR	rv = 0.8

CNN = Convolutional neural network; GILS = Genetic inverse least squares; MCR-ALS = Multivariate curve resolution – alternating least squares; MLR = Multiple linear regression; PCR = Principal component regression; PLSR = Partial least squares regression; SVMR = Support vector machines regression

Combine qualitative and quantitative analysis of food and agro-products adulteration # Source Objective Adulterant Range of The best of Prediction spectral (Sample Pre-treatment results material Algorithm number) (nm) Accuracy = 1 (Kazazić et al., Butter (36) Pork fat, Margarine 900 -PLS-DA Raw 100% 2021) 1700 PLSR RPD = 5.24 -37.51 Accuracy = 95.4 - 100% $R^2 = 0.95 -$ MN + 2nd dev 2 (Amirvaresi et al., Saffron (120) C. sativus style, 833 -PLS-DA 2021) safflower, rubia and 2500 calendula PLSR 0.99

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#	Source	Objective	Adulterant	Range of	The best	t of	Prediction
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results
				2500 -		PLS-DA	Accuracy =
				25000			81.3 – 100%
3	(Hosseini et al.,	Sterilized milk	Sodium dodecyl	769 –	MN + SD scaled	PLS-DA	$R^2cv = 0.98$
	2021)	(11)	sulphate	2500	2 nd dev+SNV	PLSR	$R^2p = 0.96$
				2500 –	Smoothing SG	PLS-DA	$R^2cv = 0.94$
				16667		PLSR	$R^2p = 0.98$
4	(<i>Du et al.</i> , 2021a)	Camellia oil (130)	Corn oil, rapeseed oil and sunflower oil	1000 – 2381	1 st dev SG	DA	Accuracy = 96.7%
					SNV + 1 st dev SG	PLSR	RMSEP = 4.98
5	(Le Nauven Doan	Green tea (475)	Sugar and glutinous	900 -	SNV	SVMC	Accuracy =
-	et al., 2021)		rice flour	1700	-	S//MD	97.47%
6	(Vitalis at al	Tomato naste	Ground paprika	740	1 st day SG +		IP > 0.94 Precision -
0	(<i>Vitalis et al.</i> , 2020)	(57)	seed Corn starch	1700	MSC	LDA	78 64% -
	2020)	(07)	Sucrose Salt	1700	MOO		97 65%
			Cuorose, Cuit			PLSR	RMSECV =
						I LOIR	0.23 - 0.89
7	(Temizkan et al	Yoghurt (100)	Several fat-free	1000 -	MN + MSC	SIMCA	Specificity -
'	(101112Kall 01 al., 2020a)	rognart (100)	LIHT	2500		OINIOA	100%
	20200)		onn	2000	MN + 1 st dev SG	PLSR	RPD = 4.35
					+ MSC	LOIT	N D = 1.00
				2500 -	MN + 2 nd dev SG	SIMCA	Specificity =
				15385		Cilvio, (100%
				10000		PI SR	RPD = 4.65
8	(Mabood et al	Fresh milk	Urea	1000 -	Baseline	PLS-DA	$R^2 = 0.97$
Ŭ	2020)	samples (162)	oroa	2500	Daoonno	PLSR	$R^2 = 0.98$
9	(Leng et al.	Minced beef	Pork and Duck meat	800 -	Raw	DA	Accuracy =
Ũ	2020)	(150)	r ont and Duoit mout	1852	num	BA	91.5 - 100%
	_0_0)	(100)			Raw	PLSR	RMSEP = 7.27
10	(Paraira at al	Goot milk (112)	Cow milk	1000	Pow		
10	(<i>F el ell'a el al.</i> , 2020)	Guat milk (112)		2500	Naw	FL3-DA	100%
	2020)			2300	Moving mean +	SPA	RPD - 10
					Baseline offset	01A	
11	(Weng et al	Minced heef	Reef loin heef	1000 -	SG smoothing	CNN	Accuracy =
•••	2020)	(240)	heart beef tallow	2500	ee onlood ling	onni	99%
	=0=0)	(=)	and pork loin	2000	CARS	RF	RMSEP =
							2.145
12	(Biancolillo et al	Egg pasta	Turmeric	1000 -	MSC	PLS-DA	Precision =
	2020)	(100)		2500		-	97.5%
	,	()			SNV	PLSR	RMSEP = 0.11
13	(Oliveira et al.,	Paprika powder	Potato starch,	900 -	Auto-scaling	PLS-DA	Specificity =
	2020)	(315)	acacia gum and	1700	Ū		90%
	,	. ,	annatto		Smoothing + 1 st	PLSR	RMSEP = 0.95
					dev SG		– 1.74
14	(Kene Ejeahalaka	Fat-filled milk	Melamine, urea and	850 –	2 nd dev SG +	SIMCA	Sensitivity =
	and On, 2020)	powder (150)	4 different vegetable	2500	EMSC		85%
			oils			PLSR	$R^2p = 0.96$
15	(Lima et al.,	Black pepper	Starch cassava,	1100 –	Raw	O-PLS-DA	Specificity =
	2020)	and Cumin	corn flour	2500			100%
		(130)				PLSR	RPD = 2.24 –
							7.01
16	(Aliaño-González	Honey (68)	Inverted sugar, rice	400 -	Raw	LDA	Precision =
	<i>et al.</i> , 2019)		syrup, brown cane	2500			100%
			sugar and fructose			PLSR	RMSEP = 3.89
			syrup				
17	(Zaukuu et al.,	Paprika powder	Corn flour	750 –	Smoothing +	LDA	Accuracy =
	2019)	(54)		1700	MSC		95.55%
						PLSR	$R^{2}cv = 0.98;$
							RMSECV =
					_		1.71
18	(Ferreiro-	Honey (22)	High fructose corn	400 –	Raw	PCA-LDA	Accuracy =
	González et al.,		syrup	2500			100%
	2018)					PLSR	$R^2p = 0.99$,
		o :	o <i>"</i>	4465	and L as		RMSEP = 4.71
19	(Quelal-	Cocoa powder	Carob flour	1100 –	2 nd dev SG +	PLS-DA	Accuracy =
	Vásconez et al.,	(234)		2500	OSC	B I 65	100%
	2018)				OSC	PLSR	R [∠] p = 0.97,
		.	0	1000	D <i>"</i>		RMSEP = 3.2
20	(Mabood et al.,	⊢ruit juice (198)	Saccharin	1000 -	Baseline	PLS-DA	$R^{2}CV = 0.98$
	2018)			2500	correction +	PLSR	R²p = 0.97
.		Min and the f	Anadhanh		Smoothing SG	0)////0	Desiste
21	(Rady and	Winced beef	Another beef		Normalization +	SVMC	Precision =
	Aaeaeji, 2018)	(1697)			1° dev SG		100%

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#	Source	Objective	Adultoront	Banga of	The best of		Prodiction
#	Source	(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results
		,		200 – 1100, 900		PLSR	RPD = 1.64 – 1.98
22	(<i>Mabood et al.</i> , 2017b)	Camel milk (54)	Cow milk	- 1700 1000 - 2500	1 st dev SG	PLS-DA PLSR	$R^2 = 0.97$ $R^2 = 0.92;$
23	(<i>Mabood et al.</i> , 2017a)	Camel milk (54)	Goat milk	700 – 2500	Baseline correction +	PLS-DA PLSR	$R^{2} = 0.97$ $R^{2} = 0.94$
24	(<i>Liu et al.</i> , 2017)	Honey (360)	High-fructose corn syrup, maltose	1000 – 2500	Norris + 2 nd dev	PLS-DA	Accuracy = 86.3% - 96.1%
			syrup		Norris + 1 st dev	PLSR	R²p = 0.9 – 0.98
25	(<i>Liu and Zhou</i> , 2017)	Infant formula (170)	Hydrolysed leather protein powder	900 – 1700	MSC + 1 st dev SG	SIMCA SVMR	Accuracy = 98.21% RPD = 7.42
26	(<i>Alamprese et al.</i> , 2016)	Minced beef meat (198)	Turkey meat	800 – 2667	SNV	PLSDA	Sensitivity = 0.84
						PLSR	R ² p = 0.884; RMSEP = 10.8
27	(<i>Capuano et al.</i> , 2015)	Skim milk powder (384)	Whey, starch, maltodextrin,	400 – 2498	SNV + 2 nd dev SG + mean centering	SIMCA PLSR	Accuracy = 82.42% $R_{p}^{2} = 0.93 -$
28	(Kuswandi et al., 2015)	Beef meatball	Pork meat	850 – 2000	1 st dev SG	LDA	0.98 Accuracy = 100%
29	(<i>Luqing et al.</i> , 2015)	Roasted green tea (150)	Sugar and glucose	800 – 2500	SLB, Min/max	PLSR PLS-DA	$R_{p}^{2} = 0.97$ Accuracy = 96 - 100%
30	(<i>Teye et al.</i> , 2014)	Fermented cocoa beans	Unfermented cocoa	1000 – 2500	SNV SNV	PLSR SVMC	$R^{2}p = 0.99$ Accuracy = 100%
	,	(132)			Selection wavelength using Si-PLS	PLSR	rp = 0.98; RMSEP = 1.68
31	(<i>Alamprese et al.</i> , 2013)	Minced beef (242)	Turkey meat	800 – 2667	SNV		Accuracy = 71.2% $P^2 = 98.13$
32	(Morsy and Sun,	Minced beef	Pork, fat trimming	400 -	2 nd dev SG,	PLS-DA	Accuracy =
	2013)	(191)	and offal	2500	SNV, Moving average	PLSR	100% R²p = 0.82 – 0.96
33	(<i>Zhao et al.</i> , 2013)	Beefburger (164)	Offal	850 – 1098	2 nd dev SG, MSC, Baw	PLS-DA	Accuracy = 88.9 – 95.5% RPD – 1.5 –
24	(liu at al. 2010)	Fishmool (276)	Molomino	000	2 nd doy SC 1		2.3 Accuracy –
34	(<i>Liu et al.</i> , 2010)	Fishineai (270)	Melannine	2500	2 st dev SG + Smoothing 1 st dev SG +	PLS-DA	$R^{2}p = 0.98 -$
					Smoothing + SNV		0.99; RMSEP = 0.38 – 0.24
35	(Kasemsumran et al., 2007)	Cow milk (90)	Water and Whey	1100 – 2500	MSC + 2 nd dev SG MSC	PLS-DA PLSR	Accuracy = 86.73 - 100% $R^2 = 0.99$
36	(<i>Kelly et al.</i> , 2006)	Honey (179)	Beet invert syrup and High fructose	1100 – 2498	Raw	SIMCA	Accuracy = 100%
07		Apple Ivies	corn syrup	100	MSC, 2 nd dev SG	PLSR	$R^2 = 0.72 - 0.79$
37	(<i>Leon et al.</i> , 2005)	Apple Juice (450)	Fructose, glucose, sucrose	400 – 2498	MSC	PLS-DA PLSR	Accuracy = 86 - 100% r = 0.77 - 0.94
38	(Downey and Kelly, 2004)	Strawberry and raspberry	Apples purees	400 – 2498	SNV + 2 nd dev SG	SIMCA	Accuracy = $75.1-95.1\%$
39	(<i>Paradkar et al.</i> , 2002b)	Maple syrup (272)	Cane and beet invert syrups, cane and beet sugar	1100 – 1660	1 st dev SG	PLS-DA PLSR	Accuracy = 98.39% $R^2v = 0.83 - 0.000$
			SOIUTIOUS	2500 – 25000		PLS-DA	Accuracy = 100%
40	(Contal et al., 2002)	Strawberry and raspberry purees (344)	Apples purees	400 – 2500	Raw	PLSR SIMCA PLSR	$R^2v = 0.99$ Accuracy = 79.07 - 94.77 rv = 0.98 - 0.99

#	Source	Objective	Adulterant	Range of	The best	t of	Prediction
		(Sample number)	material	spectral (nm)	Pre-treatment	Algorithm	results
41	(Paradkar et al.,	Maple syrup	Corn syrups	2500 –	Raw	PCA-DA	Accuracy =
	2002a)	(54)		25000			96.20
						PLSR	R ² p = 0.98
42	(Murray et al.,	Fish meal (136)	Meat and bone meal	1100 –	MSC	PLS-DA	Accuracy =
	2001)			2500			98.55%
					2 nd dv SG + SNV	PLSR	$R^2 = 0.94$
43	(Ding and Xu,	Beef	Mutton, pork, skim	400 –	SNV + 2 nd dev	k-NN	Accuracy =
	2000)	hamburgers	milk powder, or	2500	SG		92.7%
	,	(194)	wheat flour			PLSR	$R^2 v = 0.74 - 1$
44	(Thyholt et al.,	Beef (350)	Pork, mutton	780 –	1 st dev SG +	QDA	Accuracy =
	1997)			2500	Smoothing		98.53 – 100%
					-	PLSR	r = 0.68 - 0.94

O-PLS-DA = Orthogonal partial least squares – discriminant analysis; PCA-LDA = Principal component analysis – linear discriminant analysis; QDA = Quadratic – discriminant analysis; RF = Random Forest; SPA = Successive projections algorithm

Adulteration in livestock products

Adulteration of livestock products occurs often and considerably threatens human health and safety when other substances are added for specific purposes. *Liu et al.* (2021) reported machine learning in the form of a CNN architecture in tandem with near infrared spectroscopy data to predict hydrolysed leather protein and melamine in infant formula. Their result can predict adulterated and unadulterated milk R² up to 0.99%. Furthermore, Mabood also developed a method using near infrared spectroscopy in tandem with multivariate analysis to detect the mixture of camel milk with goat milk. They used PLS-DA to authenticate pure and adulterated milk and PLS to quantify adulteration levels with RMSE of 0.08% and 1.10%, respectively. Unfortunately, the model of this study still found inconsistent accuracy at the adulteration limit of 0.5% for authentication.

Even more amazing, *Karunathilaka et al.* (2018) proposed a methodology to rapidly evaluate commercial milk powders to determine if they are original or may include known or unknown adulterants using SIMCA classification algorithm. They claim that the classification models produced 100% sensitivities using benchtop spectrometers to detect milk powder fraud and are not limited only to specific types of known adulterants. This shows that using near infrared spectroscopy with the appropriate processing method will provide very precise and fast evaluation results for fraudulent food and agro-products.

Another issue in the livestock product is meat adulteration. Unscrupulous traders adulterate meat products with another adulterant (cheaper meat, animal offal, spoiled meat, and non-meat chemical synthetic materials) for profiteering purposes. Hence, *Zhao et al.* (2019) report the VIS-NIR technique to predict beef adulteration with spoiled beef using the LS-SVM algorithm. They declare that applying LS-SVM in the spectral range of 496 to 1000 nm can predict spoiled beef with an error prediction of approximately 5.67%. Weng et al. [52] conducted another research on the detection of adulteration meat using VIS-NIR spectroscopy was conducted by *Weng et al.* (2020) with minced beef samples. They used a spectral range of 350–2500 nm and claimed to detect minced beef mixed with pork and beef heart with error predictions of approximately 2.145% and 2.758%, respectively. These studies show that the application of VIS-NIR spectroscopy coupled with chemometrics can be powerful for the fast and accurate detection of adulterated livestock products.

Adulteration in flour products

The detection of fraud in flour products ingredients has become an even more important topic since flour products, such as bread and other bakery products, are widely consumed as primary foods. Many consumers lost trust in the food they were buying and the food industry identified that more rapid measures in terms of the evaluation of its product had to be put in place. Frequently adulteration is achieved in high-value food items and those that come through complex supply chains. The flour product that comes from food is likely more highly vulnerable to adulteration due to the complexity of the characteristics, and it is widely used for products such as bread. To address this, cutting-edge methods must be easy to use, fast and inexpensive, especially for the flour industry. The most interesting method today is the application of food fingerprinting as a detection method by IR technology. At least in the last five years, durum wheat flour, banana flour, einkorn flour, wheat flour, barley flour and cassava flour were among the flour products found to be the most commonly adulterated and the researchers have studied how to detect it using IR spectroscopy technology.

In old studies, *Cocchi et al.* (2006) ever studied the use of near infrared spectroscopy to quantify the adulteration level of durum wheat flour using the PLS algorithm. The authors claim near infrared spectroscopy

data can show durum wheat flour adulteration using SNV pre-treatment. In another study by *Ndlovu et al.* (2019) considered VIS-NIR spectroscopy to detect adulteration of unripe banana flour with wheat flour.

They found that the PCA model could successfully separate samples of pure and contaminated banana flour. PLSR model also could quantify the level of adulteration. Both results of this study indicate that NIR and VIS-NIR spectroscopy could monitor the quality of flour in retail markets for the purpose of product verification.

In a recent study by *Ayvaz et al.* (2021a), near infrared spectroscopy is suggested to detect adulteration of einkorn flour with wheat flour and presents a correlation coefficient of 0.94 to 0.99. The lowest correlation coefficient is found in the adulteration ratio of wheat flour less than 7% (w/w). IR spectroscopy was also used by *Aykas and Menevseoglu* (2021) to detect the mixing of powdered pistachio with powdered green pea and peanut. Infrared spectroscopy can be correctly predicted with a coefficient correlation of about 0.99.

Furthermore, Tao published a study on the detection of eight varieties of adulterants of cassava flour in wheat flour using micro-IR spectroscopy in the range of 1150–2150 nm. The classification of this study finding that the adulteration of wheat flour with cassava flour achieved 100% accuracy, yet the level adulteration of wheat flour with cassava flour (5% to 40% adulteration) only presented correct classification rates between 56.25% and 100%. The last but not least, study reported by *Xu et al.* (2013c) used near infrared spectroscopy in the 1000–2500 nm range to classify Chinese glutinous rice flour from extraneous adulterants and unwanted variations. This study found an adulteration specificity of 0.92 with one-class partial least squares algorithms.

Adulteration in liquid agro-product

Adulteration of liquid agro-products is valued in the same way as pure products, and there is a need for fast, easy, and precise analytical methods to assess their characteristics and originality. Popular liquid agro-products obtained in the form of naturally sweet and viscous products are honey, fruit juices, and vegetable oil.

According to Tan et al. (2021) and Contal, L. (2002), the chemical content of wild honey is correlated with the season, geographical region, storage method and harvesting method, which makes it very difficult to compare other types of honey. It also makes honey very susceptible to adulteration and is valued similarly to pure honey. Evaluation the feasibility of near infrared spectroscopy technology in the rapid detection and classification of adulteration of honey has been study by some researcher. Kelly et al. (2006) detect adulterated honey from beet invert syrup and high fructose corn syrup using near infrared spectroscopy (1100-2498 nm) with an accuracy between 9.0 and 11.9 (RMSE-CV). Furthermore, the same study was also conducted by Bázár et al. (2016) to detect corn syrup additives in honey using near infrared spectroscopy in the wavelength ranges 1300–1800 nm and reached an accuracy better than the previous study (RMSE-CV of 1.48). Besides, Ferreiro-González et al. (2018) used VIS-NIR spectroscopy (400-2500 nm) to predict honey adulteration with fructose-rich corn syrup and obtained an accuracy not yet better than Bázár et al. (2016) (RMSE-CV of 4.71). The most recent to conduct a similar study is Valinger et al. (2021a), which evaluated the feasibility of near infrared spectroscopy technology in the rapid detection of adulteration of honey with corn syrup. Unfortunately, the results indicate that the near infrared spectroscopy of adulterated honey can be modelled to detect fraud with an accuracy that is not yet better than the previous study. However, the interesting one in this study is that the adulteration of honey with water reported cannot be predicted with precision.

Fruit juice becomes a liquid food agro-product of the most common adulteration with artificial sweeteners, dilution with water, and fraud with low-quality or less-expensive fruit juice. Therefore, some researchers have developed a fast and low-cost method for inspecting fruit juice adulteration or dilution. In one study, *Mabood et al.* (2018) reported applications of near infrared spectroscopy (860–2500 nm) for classification of adulteration and non-adulteration in commercial fruit juices with precision between 0.067 to 0.169 (RMSE).

Adulteration in herbs and spices

Spices are highly valued agro-products because they are used in many in the world to flavour and preserve processed food. However, herbs and spices are extremely vulnerable to commercial gain motivated fraud including black pepper, garlic, saffron, and oregano.

Spices are high-value food components in weight units because they have desirable flavour characteristics and, therefore, are economically profitable targets for adulteration. To address this problem, Wilde and *Galvin-King et al.* (2021b) conducted a study on the feasibility of near infrared and infrared spectroscopy to detect adulteration in black pepper and garlic of adulterants. The developed model is claimed

to classify black pepper from its adulteration with a percentage of correct between 92% to 100%. Investigation of garlic adulteration detection using parameter validation in the form of fit measurement has an accuracy in the range of 98.5% to 99.4%.

Meanwhile, *Amirvaresi et al.* (2021) applied infrared spectroscopy to authentication saffron adulteration with accuracy classification between 81.3 to 100%. Unfortunately, detection limitations are only in the range of 1.0–3.1% (w/w) for each adulterant. Work has also been carried out by *Galvin-King et al.* (2020a), who have utilized infrared spectroscopy to identify the presence of adulterate powdered paprika with Varying seed or pod. Their model claims to predict component adulteration on powdered paprika with a coefficient of determination of about 0.94.

FUTURE PERSPECTIVES

Current studies indicate the potential of near infrared and infrared spectroscopy approaches for detecting the adulteration of food and agro-products. Such a breakthrough would undoubtedly support the further implementation of near infrared and infrared spectroscopy-based quality evaluation. The availability of multiple data sources and the fusion of multi-origin data affords a perspective for future research. The fusion of UV-VIS, near infrared, and infrared spectroscopy is the process of combining some spectral information to improve data quality and produce a high quality representation model (*Valinger et al.*, 2021a). Future studies may use sample adulteration from a different origin, variety, storage temperature, or even shelf-life when developing a model. With the increasing number and high quality of accessible samples, the future perspective for detecting the adulteration of food and agro-products possibly focuses on near infrared and infrared spectroscopy tandem with machine learning. The main advantage of the machine learning approach is decreasing the dependence on human domain knowledge by end-to-end analysis and the improved precision and generalizability.

CONCLUSIONS

In this paper, the feasibility of applying a non-destructive for detecting and discriminating food adulteration and agro-products is based on near infrared and infrared spectroscopy and various types of data analysis have been represented. Besides the non-destructive, the primary advantages of the analytical method are fast and economical, directing to cost-effective quality assurance of detecting such a key worldwide food and agro-products adulteration. Actually, once the chemometric model has been correctly calibrated, the time elapsed from the scanning of IR spectroscopy on the samples and their subsequent classification would only need a few seconds. Therefore, this approach could represent a concrete and effective answer to the need, claimed by industrial and agro-product producers, as well as by the Food Control Authority, for affordable, fast, and efficient technologies to evaluate food quality and authenticity. Furthermore, the results of the variable selection and discrimination of adulteration food and agro-products directly "in situ" to ensure authenticity and counteract adulteration. Last but not least, the promising results performed by the numerous laboratory model validation indicate the potential transferability of a near infrared and infrared spectroscopy-based method to various production food and agro-product sites.

In the future, although optimistic results were acquired in an investigation for fraud detection for food and agro-products today, it must be pointed out that the optical for near-infrared and infrared spectroscopy technologies applied remain pricey so far. To implement routine analyses in some food and agro-products, it is necessary to develop low-cost infrared optical technologies and have the same accuracy as those currently available.

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