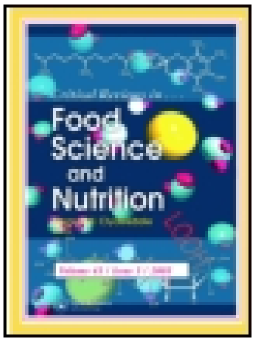

Taylor & Francis
Taylor & Francis Group

Critical Reviews in Food Science and Nutrition

ISSN: 1040-8398 (Print) 1549-7852 (Online) Journal homepage: http://www.tandfonline.com/loi/bfsn20

Non-destructive and Rapid Evaluation of Staple Foods Quality by Using Spectroscopic Techniques: A Review

Wen-Hao Su, Hong-Ju He & Da-Wen Sun

Non-destructive and Rapid Evaluation of Staple Foods Quality by Using Spectroscopic Techniques: A Review

Wen-Hao Su, Hong-Ju He, Da-Wen Sun*

Food Refrigeration and Computerised Food Technology (FRCFT), School of Biosystems Engineering, Agriculture & Food Science Centre, University College Dublin (UCD), National University of Ireland, Belfield, Dublin 4, Ireland

Abstract

Staple foods including cereals, legumes, and root/tuber crops dominate the daily diet of human by providing valuable protein, starch, oils, minerals and vitamins. Quality evaluation of staple foods is primarily carried out on sensory (e.g. external defect, colour), adulteration (e.g. species, origin), chemical (e.g. starch, protein), mycotoxin (e.g. *Fusarium* toxin, aflatoxin), parasitic infection (e.g. weevil, beetle), and internal physiological (e.g. hollow heart, black heart) aspects. Conventional methods for the quality assessment of staple foods are always laborious, destructive and time-consuming. Requirements for online monitoring of staple foods have been proposed to encourage the development of rapid, reagentless and non-invasive techniques. Spectroscopic techniques such as visible/infrared (VIS/IR) spectroscopy, Raman spectroscopy, nuclear magnetic resonance (NMR) spectroscopy and spectral imaging, have been introduced as

* Corresponding author. Tel.: +353 1 7167342; Fax: +353 1 7167493.
E-mail address: dawen.sun@ucd.ie (D.-W. Sun).
URLs: http://www.ucd.ie/refrig, http://www.ucd.ie/sun (D.-W. Sun).

promising analytical tools and applied for the quality evaluation of staple foods. This review summarizes the recent applications and progresses of such spectroscopic techniques in determining various qualities of staple foods. Besides, challenges and future trends of these spectroscopic techniques are also presented.

Keywords: VIS/IR, Raman, NMR, spectral imaging, rapid evaluation, staple foods

1. Introduction

Staple foods are dominant constituents of human diet and are consumed in a large scale worldwide because of their high energy and nutrition. Cereals, legumes, and root/tuber crops are three kinds of the most important staple foods, accounting for approximately 90% of the world's food calories intake (Bender and Smith, 1997). Cereals (e.g. maize, rice, wheat, barley, sorghum, millet, rye and oats) are rich in vitamins, minerals, carbohydrates, fat, oils and protein and provide over half of energy consumed by humans. Legumes mainly include soybean, pea, bean, lentil, lupin, tamarind, carob, mesquite and peanut, and carry protein contents varying from 17% to 40%, higher than that of cereals (7-13%) (Naerstad *et al.*, 2012). Legumes are widely used as a main source for oil production and also a valuable source of complex carbohydrates, dietary fibre, vitamins and minerals. Root/tuber crops are plants containing starchy roots, tubers, rhizomes, corms and stems, which hold high contents of carbohydrates, calcium and vitamin C. The Food and Agriculture Organization (FAO) has distinguished several primary root/tuber crops, which are potatoes, sweet potatoes, yautia, cassavas, taro and yams. In recent years, the total production of staple foods has increased rapidly and generated tremendous growth in their market values. The overwhelming majority of staple foods need to be stored before consuming. The shelf-life of staple foods after harvest could be from a few weeks to more than one year depending on the food variety and storage conditions. However, the sensory and chemical attributes of postharvest staple foods changes continuously during storage and transportation

 ACCEPTED MANUSCRIPT

(Sowokinos, 1978, Hou and Chang, 2004), in particular, staple foods often suffer from damages caused by sprouting (Fu *et al.*, 2014), toxigenic fungi (Kirinčič *et al.*, 2015, Pitt *et al.*, 2013), parasitic insect (Trematerra *et al.*, 2003), and black heart (Novy, 2014) during storage and transportation. In addition, food adulteration is also considered to have great health risk, which is difficult to detect (Tahkapaa *et al.*, 2015). Therefore, it is necessary and important to conduct quality evaluation of staple foods.

Currently, staple food quality are evaluated on sensory (e.g. external defect, colour), adulteration (e.g. species, origin), chemical (e.g. starch, protein), mycotoxin (e.g. *Fusarium* toxin, aflatoxin), parasitic infection (e.g. weevil, beetle), and internal physiological (e.g. hollow heart, black heart) aspects, as shown in Fig. 1. Conventional analysis and detection methods such as high performance liquid chromatography (HPLC) and thin-layer chromatography are still widely used in the staple food industry for detecting food quality. However, these conventional operations are normally destructive, manual, time consuming and laborious. Therefore, in recent years, novel spectroscopic techniques such as visible/infrared (VIS/IR) (Xu *et al.*, 2014, Barbin *et al.*, 2015), Raman spectroscopy (Corvucci *et al.*, 2015), nuclear magnetic resonance (NMR) (Ohtsuki *et al.*, 2015) spectroscopy and spectral imaging (Nogales-Bueno *et al.*, 2015) have been developed and applied for the quality evaluation of staple food. Although reviews have been published on the use of these novel techniques, they focused on only one or two such spectroscopic techniques for a particular type of products such as fruit (Magwaza *et al.*, 2012),

ACCEPTED MANUSCRIPT

vegetables (Pu *et al.*, 2015), milk (Domingo *et al.*, 2014), oil (Dais and Hatzakis, 2013), and fish and meat (Herrero, 2008, Kamruzzaman *et al.*, 2015). Therefore, in the current review, a comprehensive description of the spectroscopic techniques for determining the sensory, adulteration, chemical, mycotoxin, parasitic infection, and internal physiological aspects of different staple foods is presented, the challenges and future trends of these techniques are also discussed.

2. Spectroscopic techniques

VIS and IR spectroscopy including visible/near-infrared (VIS/NIR), mid-infrared (MIR) and far-infrared (FIR) spectroscopy are well-established non-destructive analytical techniques allowing for the reliable, direct, and fast determination of several properties without sample pre-treatment (Sun, 2009). VIS spectra cover the spectral range from 380 to 780 nm, indicating different perceived information. NIR spectroscopy (780-2500 nm) is more useful for quantitative analysis of complex mixtures. This is mainly due to that NIR spectra provide feedbacks of spectral stretching and bending of the chemical bonds involving C-H, N-H, S-H and O-H. NIR range is usually divided into short-wave near-infrared (SW-NIR) spectral region (780-1100 nm) where transmittance measurements are usually carried out with weak absorptions, and long-wave near-infrared (LW-NIR) spectral region (1100-2500 nm) where most of the incident radiation is diffuse reflectance. Compared with NIR, MIR spectra arise from more abundant spectral information of intensities and frequencies. MIR is commonly used to determine the chemical functional groups of a sample in both qualitative and quantitative way. The MIR spectra locate in 2500-25000 nm and can be divided into three wide regions: X-H stretching region (2500-3571 nm), triple bond region (4348-4762 nm), and fingerprint region (5556-12500 nm). FIR spectroscopy is an efficient substitution approach usually used in transmission and attenuated total reflectance modes for the characterization of the chemical compounds that is inactive in the MIR region. Raman spectroscopy is used to observe vibrational, rotational, and other low-frequency modes in a system and provide a fingerprint by which molecules can be identified and localised (Jääskeläinen *et al.*, 2013). It relies on inelastic scattering or Raman scattering of monochromatic light, usually produced from a laser in the visible, near infrared, or near ultraviolet range. NMR is a powerful technology and has been applied for food traceability and authenticity without altering the sample and producing hazardous wastes (Laghi *et al.*, 2014). NMR spectroscopy offers the possibility to measure structural information of molecules quantitatively where the atoms are contained with an intrinsic magnetic moment and angular momentum. Low-field proton nuclear magnetic resonance (^{1}H-NMR) relaxation is always used to identify and quantify a large number of compounds simultaneously. Spectral imaging technique is a combination of spectroscopy and imaging where some spectral information are located at every spot in a scene, which means that spectral imaging technique can be used for the simultaneous acquisition of spatial images and spectral information (Sun, 2010). The spectral imaging systems obtain three-dimensional datacubes with two dimensions (x, y) representing spatial images and the third dimension (λ) representing spectral information. Spectral imaging technique that mainly includes full spectral imaging, hyperspectral imaging (HSI) and multispectral imaging, is one of the most novel and promising analytical technique. Full spectral imaging treated as the successor of HSI extracts information that is only in the spectral curves excluded signal, noise, redundant bits, while HSI considered as multispectral imaging with numerous bands obtains data as contiguous spectral bands (Bolton, 2004). The number of wavelength bands captured by spectral imaging system is diverse, normally several hundred bands for HSI and about 5-20 wavelengths for multispectral imaging. HSI has already been used to fast acquire both spectral and spatial information of food samples (Bannon, 2009). Fig. 2 shows the flow chart of HSI technique for the evaluation of staple food quality. By integrating the main features of imaging and spectroscopy, spectral imaging can be applied for the quantitative prediction of the inherent chemical and physical properties as well as their spatial distribution simultaneously. Currently, most of imaging spectrometers are designed to record spectral information in VIS and NIR region (400-2500 nm) (Ustin *et al.*, 2004). Table 1 summaries the application, principles and main instruments of these spectroscopic techniques.

3. Applications

3.1 Sensory aspects

Sensory attributes of staple food involving colour, shape, hardness, texture and external defect are significant components of food quality. The generation of visual colour is due

 ACCEPTED MANUSCRIPT

to the reflection of different wavelengths in visible light region. Shape as the general profile of a product is usually determined by dimension, weight or volume, and can affect consumers' preference selection and final consumption. Hardness is one of the foremost determinants of food quality reflecting the texture and moisture content of food species. In addition, external defects arose either in growth period or postharvest are another very common sensory attribute and affect food quality seriously. Therefore, accurate and fast prediction of these sensory attributes is the first principal concern for the agriculture and food industry.

The sensory attributes including colour and texture have been determined by VIS and NIR spectroscopy. In the research of Mendoza *et al.* (2014), VIS/NIR spectroscopy (400-2500 nm) were effectively used to qualify the intact dry beans by predicting canning quality traits including colour and texture, and the canned beans were classified into two sensory categories of "acceptable" and "unacceptable" by a linear discriminant model, with an ordinary classification accuracy of about 72.60 %. With the improvements in sensor and instruments, it is believed that VIS and IR spectroscopy techniques have the potential to meet the application requirements. Sensory evaluation of staple foods using HSI systems mainly are focused on the determination of colour, hardness and external defects. Specifically, the colour of fresh and dried soybeans was predicted by VIS and LW-NIR HSI (400-1000 nm) during drying process (Huang *et al.*, 2014). Based on an active contour model to automatically segment soybean spectral images, this HSI technique was more effective to acquire the mean reflectance of an image and measure the image entropy parameters. Later, a partial least squares regression (PLSR) model was established to detect the colour of the processed soybeans, with a good coefficients of determination (R^2) of 0.74. In the recent study of Wang *et al.* (2015), who discriminated rice samples by HSI in the range of 400-1000 nm based on colour and shape as shown in Fig. 3, five shape features (minor axis length, major axis length, perimeter, length-width ratio and eccentricity) and one colour feature (chalkiness degree) were used as the input of back propagation neural network (BPNN) models, which achieved a high classification accuracy of 94.45%. During the same time, Mahesh *et al.* (2015) evaluated the hardness of wheat samples by HSI (960-1700 nm) using PLSR and principal components regression (PCR), with PLSR performing better than PCR. Surface defects of potato were extensively determined by HSI as well. Xing *et al.* (2010) applied a HSI system (400-1000 nm) to classify sprouted and severely sprouted wheat kernels from sound ones. The ratio value of 878/728 nm was used to rapidly distinguish the sprouted from non-sprouted kernels. An obtained high accuracy of 94% demonstrated the good ability of HSI for the detection of sprouted wheat kernels. On the other hand, a LW-NIR HSI system (900-1700 nm) was employed to classify potato scabs, and an accuracy of 97.10% based on the support vector machines (SVM) classifier was obtained (Angel *et al.*, 2011a).

3.2 Adulteration aspects

Adulteration refers to the low-grade substances mixed into the original superior substances, which can destroy the completeness of the original products and reduce the nutritional value or quality of the food products. The appearance of most adulterated material looks very similar to the original products, thus it is difficult to distinguish them with the naked eye when mixed together. On the other hand, determining the species and origin of staple foods is very useful to protect consumers from potential fraud because the geographic indications of food product cannot be confirmed only from the food labels. Therefore, reliable adulteration analysis is necessary for the guarantee of product quality.

The adulterations of origin and species have been determined by different spectroscopic techniques. Transmission Raman spectroscopy was effectively utilized for the discrimination of rice samples according to geographical origin with PCA and linear discriminant analysis (LDA) (Hwang *et al.*, 2012). Similar results were obtained in the research of Feng *et al.* (2013), who combined Raman spectroscopy with multivariate data analysis methods including PLS-DA, K-nearest neighbor (KNN), and SVM for discrimination of rice samples from diverse regions of China, with the overall accuracy of more than 90%. It was concluded that Raman spectroscopy with chemometric techniques can be applied to rapidly discriminating adulteration of geographical origin in staple foods. ^{1}H NMR spectroscopy has also been used to discriminate wheat flour samples of three different locations (Lamanna *et al.*, 2011). The rate of correctly assigned samples achieved 80% using a simple linear model. Different types of rice samples and their geographical origin were also effectively discriminated based on the ^{1}H NMR spectroscopy and multivariate data analysis (Monakhova *et al.*, 2014). Both PLS-DA and the independent component analysis (ICA) gave the high rate of correct discrimination (0.96). In general, Raman and NMR spectroscopy could rapidly and accurately determine adulteration of staple foods.

The feasibility of NIR spectroscopy for discriminating viable-germinating corns and soybeans from dead seeds was tested by Esteve Agelet *et al.* (2012). Dead corn kernels suffered from heat were discriminated by partial least squares discriminant analyses (PLS-DA), with the accuracy of 99%. Haughey *et al.* (2013) applied NIR spectroscopy 833-2632 nm to detect adulteration of soya by analysing the spectra of de-hulled soya, soya hulls and toasted soya contaminated with melamine. The R^2 ranging from 0.89 to 0.99 were achieved by PLSR and PCR algorithms. Liu *et al.* (2013) distinguished the adulterated cheaper potato and sweet potato starch from non-adulterated lotus root powder by Fourier transform mid-infrared (FT-MIR) spectroscopy, with high R^2 of 0.96 to 0.99. In addition, Kouvoutsakis *et al.* (2014) successfully combined FT-IR spectroscopy with discriminant analysis for the geographical differentiation of dried lentil seeds.

For imaging spectroscopy, Serranti *et al.* (2013a) investigated the potential of HSI (1000-1700 nm) to identify different types of wheat kernels of vitreous and yellow berry. Better results were acquired by selecting three narrow intervals (1209-1230 nm, 1489-1510 nm and 1601-1622 nm) rather than using the full wavelength range. Meanwhile, an innovation based on HSI (1006-1650 nm) to accurately identify oat and groat kernels was validated by Serranti *et al.* (2013b). The accuracy approached 100% using PCA and PLS-DA. Recently, Mishra *et al.* (2015)

detected peanut traces of 0.10% in wheat flour with R^2 of 0.95 by NIR HSI in combination with PCA. All of these spectroscopic techniques showed a very good performance in the detection of different forms of food adulteration.

3.3 Chemical aspects

The food composition is the basis for determining the nutritional value and overall acceptance, as chemical components are intrinsic reasons affecting food quality. The highest percentage of chemical composition in cereals and legumes is carbohydrate followed by crude protein, moisture, crude fibre, ash and fat, while in root tubers, moisture is the highest. Starch is a kind of carbohydrate consisting of helical amylose and the branched amylopectin. Amylose content varies widely within a single species and the following ranges of amylose have been reported: maize 20-36%, sorghum 21-35%, wheat 17-29%, barley 11-26%, rice 8-37%, pea 34-37%, potatoes 18-23% (BeMiller, 2009). Starch properties, such as the proportion of amylose and amylopectin, have great influences on the quality of food such as bread and noodles (Hug-Iten *et al.*, 2003). Proteins are the principal structural and functional components in staple foods. Protein functional properties including elastic-plastic and extensibility are those physicochemical properties that contribute to the quality and organoleptic attributes of staple food product. Moisture as another major component is presented both as a solvent held by molecular forces, and as water of hydration held by hydrogen bonding. The water content of staple foods during storage strongly influences the keeping quality and the success of subsequent germination. The fibre in staple foods mainly contain cellulose, hemicellulose, lignin, pectin and gums, which could help to lower serum cholesterol levels but not provide energy in humans. By contrast, lipids are the most concentrated form of energy yielding more than twice as much energy per gram as either carbohydrates or proteins. The ash of food is the inorganic residue remaining after the organic matter has been burnt off. The importance of ash content is that it gives an idea of the amount of mineral elements presented in the food sample. The common method for moisture determination in staple foods is direct drying with oven or Karl Fisher titration, both of which are low-efficiency (Corpaş *et al.,* 2014). The crude fat can be determined by extracting the dried food material with ethyl ether or petroleum ether. The crude protein content is traditionally determined by quantifying the organic nitrogen content using the Kjeldahl and Dumas methods (Jung *et al.,* 2003). As most of these existing methods for detecting the major components of staple foods are destructive, time-consuming and require long time for sample preparation, development of rapid non-destructive detection methods will be an advantage.

VIS/IR spectroscopy techniques provide a great potential for the determination of chemical component in staple foods. Nie *et al.* (2012) showed that VIS/NIR spectroscopy in the range of 400 to 1000 nm could be used as a non-destructive technique to determine the poisonous phytohaemagglutinin in beans by controlling the boiling time of yardlong beans. NIR and MIR spectroscopy techniques were also applied and compared in another research to determine the chemical attributes including moisture, protein, lipid and ash contents in 20 varieties of soybean by Ferreira *et al.* (2014), who indicated that MIR and NIR techniques generated consistent PLSR models with good predictive abilities. The total time required for the preparation and determination of samples was less than 5 min, compared to the 10-16 h required by the reference methods. Ferreira *et al.* (2013) also examined the ability of FT-NIR spectroscopy to estimate the concentration of moisture, protein, lipid, ash and carbohydrate of Brazilian soybeans. Based on PLSR, the best results were found in protein (R^2 = 0.81) and moisture (R^2 = 0.80) contents, followed by lipids, ashes and carbohydrates. Moreover, López *et al.* (2013) accurately predicted the crude protein content in potato by NIR spectroscopy (1100-2300 nm). PLSR was applied to develop the calibration model, with high correlation coefficients ranging from 0.86 to 0.95. Later, Rady *et al.* (2014) demonstrated a potential of VIS/NIR spectral interactance for rapid in-field measurements of chemical compositions in two species of potato tubers. In their study, the VIS/NIR interactance system (446-1125 nm) was shown to yield the best correlation for predicting glucose, sucrose and soluble solids in sliced samples. In addition, chemical and enzymatic compositions including carotenoids, flavonoids, anthocyanins, phenolics, dioscin and catalase in staple foods were screened using NIR or MIR spectroscopy combined with chemometric tools such as PCA, hierarchical clustering analyses (HCA), PLS-DA and SVM, generating good predictive results (Uarrota *et al.*, 2014, Sánchez *et al.*, 2014, Kwon *et al.*, 2015).

ACCEPTED MANUSCRIPT

It was concluded that VIS/IR spectroscopy techniques can be effectively applied to monitor the quality of staple foods.

FT-Raman spectroscopy as a promising tool was used to rapidly determine cassava starch crystallinity, with a strong linear correlation (R^2 = 0.99) between crystallinities and integrated areas of the skeletal mode Raman band at 208333 nm (Mutungi *et al.*, 2012). Then, Lee *et al.* (2013) developed an optimal prediction model for efficiently determining the protein and oil contents in soybeans using a dispersive Raman spectroscopy 5556-50000 nm. The R^2 of the optimal PLSR model were 0.92 and 0.87 for crude protein content and crude oil content, respectively. Besides, NMR spectroscopy is also widely used to detect different chemical components in staple foods. Based on ^{1}H and ^{13}C NMR spectroscopy, the main chemical compositions including protein, dietary fibre and unsaturated fat in marama beans from different geographical sites and harvest years were determined by Holse *et al.* (2011). Compositions of amino acids, fatty acids, sugars, elements, and isoflavones in several soybean varieties were effectively performed by ^{1}H NMR spectroscopy combined with PCA (Jiao *et al.*, 2012, Song *et al.*, 2013). Recently, a comparative study was conducted using NMR to detect water-absorption in six rice varieties (Horigane *et al.*, 2014). In their research, moisture distributions were compared based on signal intensity profiles of the presence and absence of a white core, difference in harvest years, varieties of the parent and their offspring, and difference in polishing yield. It was concluded that NMR was a powerful technique for estimating the varietal differences of the water absorption properties of cereals and can be used for aptitude evaluation of cereal varieties for a wide range of final uses, such as boil-cooking, brewing, and flour milling for bread and noodles.

HSI systems have been applied for predicting alpha-amylase activity and the content of amino acid, sweetness, glucose, moisture and sucrose in staple foods. Based on PLSR method, the alpha-amylase enzyme activity levels in wheat kernels were predicted from the spectral information, with the highest R^2 of 0.88 (Xing *et al.*, 2011). Moreover, for prediction of amino acid content and sweetness on the basis of sucrose, glucose, fructose and nitrogen concentrations in soybeans, an early study was completed by Monteiro *et al.* (2007) using a nonlinear regression model based on the second derivative transformed dataset. A better accuracy of 0.74 was obtained, followed by sucrose, nitrogen and fructose, with the accuracy of 0.73, 0.61 and 0.60 respectively, which indicated the potential of HSI in the assessment of chemical composition. After a few years, another research on the prediction of glucose, sucrose, soluble solids and other ingredients in potatoes were accomplished based on HSI (400-1000 nm) (Rady *et al.*, 2014). The glucose was predicted with a greater accuracy of 0.74, but the sucrose and soluble solid content revealed weak correlations with the accuracy of 0.57 and 0.36. Moisture content is another vital parameter used to estimate staple food quality. Huang *et al.* (2014) designed regression models for predicting the moisture content in soybeans during the drying process by HSI (400-1000 nm). Better prediction result for moisture content was achieved using the automatic segmentation and

PLSR technique, with a high R^2 of 0.94. Later, the moisture content in peanut was determined by PLSR and exhibited a good performance with R^2 of 0.91 by selecting six special wavelengths in 400-1000 nm of HSI (Jin *et al.*, 2015).

3.4 Mycotoxin aspects

Foods contaminated by moulds and related toxins generated during post-harvest handling or storage can lead to significant losses in nutrient composition and market value, causing severe food safety concern. Mycotoxins such as aflatoxin and *Fusarium* toxin are toxic secondary metabolites and are well-known carcinogen associated with liver and lung cancer in humans. It has been estimated that up to 25% of the world's crops grown for animal feed and human consumption may be contaminated with mycotoxins (Hussein and Brasel, 2001). Mycotoxins levels for staple food products have been strictly specified by EU (Cheli *et al.*, 2014). Actually, aflatoxin level of 20 ppb (parts per billion) in food and 100 ppb in feed are allowed for interstate commerce in the USA (Yao *et al.*, 2013a). According to a report from FAO, about 1000 million metric tons of food are spoiled globally each year due to mycotoxins (Bhat *et al.*, 2010). Therefore, successful detection of different fungal infection levels can make a great contribution to controlling diseases in agricultural crops and reducing food safety risks. Nowadays, the methods available for the toxic detection and quantification mainly include thin-layer chromatography and HPLC (Ahmed *et al.*, 2014), which require the destruction of samples and are costly and time consuming.

Rapid and feasible detection of mycotoxins for staple foods can be widely found based on IR spectroscopy. Sirisomboon *et al.* (2013) detected total fungi and yellow-green *Aspergillus flavus* infection in rice using NIR spectroscopy (950-1650 nm) coupled with PLSR, but the accuracies for both total fungi and yellow-green *A. flavus* infection were not high. Then, Giacomo and Stefania (2013) applied PLSR with full cross validation to screen the maize contaminated by fumonisins based on NIR spectroscopy, with a high R^2 of 0.91. This means that NIR could be used as a suitable alternative tool to detect fumonisins infection. Afterwards, an automated differentiation of peanut kernels contaminated with aflatoxin and non-aflatoxin strains was achieved by Kaya-Celiker *et al.* (2014) using FT-MIR 2500-16000 nm coupled with attenuated total reflectance unit. Classification was performed to separate the "Acceptable" stream (aflatoxin ≤ 20 ppb) from "Mildly" (20 < aflatoxin < 300 ppb), "Highly Toxic" (300 < aflatoxin < 1200 ppb) and "Highly Moldy" (aflatoxin > 1200 ppb). Within the fingerprint region (5556-12500 nm), R^2 of 99.98% was achieved to predict both *A. flavus* and *A. parasiticus* species with different levels based on PLSR model.

Using Raman technique to measure mycotoxins has long been the goal of researchers. Liu *et al.* (2009) applied Raman technique for non-destructive screening deoxynivalenol (vomitoxin) contaminated wheat and barley by PCA. The advantages of this technique included the use of a 1064 nm NIR excitation laser that reduced interference from fluorescence of biological compounds as well as the use of an intensity-intensity algorithm at two unique frequencies.

ACCEPTED MANUSCRIPT

Raman spectroscopy was also effectively used to classify a large number of aflatoxin-contaminated maize samples by Lee *et al.* (2014a). PLSR model showed a high R^2 arranging from 0.94 to 0.96 for predicting aflatoxin concentration. There was no significant difference between predicted values using Raman spectroscopy and reference values using HPLC. Meanwhile, Lee *et al.* (2014b) employed a novel surface-enhanced Raman technique for aflatoxin detection in maize. The MLR model showed the highest predictive accuracy with R^2 of 0.94 over other quantification models. Compared to other spectroscopic techniques, this technique seemed to provide more comprehensive information on the structural and electronic properties of aflatoxin molecule.

Spectral imaging has a great potential to be a more rapid, objective means in the future and has already been applied to indirectly identify fungal infected staple foods. Liu *et al.* (2010) successfully discriminated fungal infection in healthy, light, moderate, and serious levels in rice panicles using HSI (350-2500 nm). The overall accuracy applying a learning vector quantization neural network classifier was 100%, better than a LDA model with the accuracy of 92%. Researches also focused on classifying corn kernels (Yao *et al.*, 2013a) and maize kernels (Yao *et al.*, 2013b) infected with *A. flavus* using fluorescence hyperspectral imagery. It was concluded that the binary encoding means had a better performance with the accuracy of 87% or 88% with 20 ppb or 100 ppb threshold for corn kernels, and a higher classification accuracy of 94.40% with 100 ppb on the germ side, better than the endosperm side for maize kernels based on the discriminant analysis. Then, Shahin *et al.* (2014) quantified mildew damage in wheat by analysing two kinds of spectral data from a HSI system (400-950 nm) and a FT-NIR analyser (1000-2500 nm). It was found that HSI system were more effective (accuracy of 96%) for the classification than NIR spectroscopic technique. Recently, Kandpal *et al.* (2015) utilized HSI (1100-1700 nm) technique coupled with PLS-DA model to determine aflatoxin B_1 on corn kernels, with the highest accuracy of 96.9%. Similarly, PLSR was also used to monitor *A. flavus* growth on stored rice from the HSI reflectance spectra (400-1000 nm) (Siripatrawan and Makino, 2015). A R^2 of 0.97 was obtained, which also demonstrated that HSI can rapidly and effectively classify staple foods with different levels of fungal infection.

3.5 Parasitic infection aspects

The contamination caused by parasitic insects could downgrade staple foods and lower the market value. Parasitic insects stored in staple foods not only directly feed, but also produce heat and moisture due to their metabolic activity, which can cause the development of insect-induced localized hotspots and spoilage. Insect damage results in loss of weight, nutrients, and germination ability, and increases the susceptibility to contamination during storage. Stored products are vulnerable to both external infection of adult insects such as *Oryzaephilus surinamensis* and internal infection from insects such as *Sitophilus granarius*. For parasitic insects including sweet potato weevil (*Cylas formicarius elegantulus*), alterations in the external appearance are minimal and the primary damage is done throughout the inside of the product.

Internal damage is due to the presence of immature insects (eggs, larvae, and pupae) in seed, such as rice weevil (*Sitophilus oryzae*) and lesser grain borer (*Rhyzopertha dominica*). These insects can continue their destructive activity inside the kernel for four to seven weeks and visible damages of seed are not apparent until adults emerge with an exit hole left (Singh *et al.*, 2010). Moreover, some secondary feeders such as rusty grain beetle (*Cryptolestes ferrugineus*) and red flour beetle (*Tribolium castaneum*) cause external infection by feeding on the damaged or broken grains.

The food industry is in need to seek rapid and automatic methods for detecting parasitic insect infestation in staple foods, especially the internal infestation, which is more difficult to inspect. The spectral information of HSI arising from the reflectance or absorbance of the insect-infested food kernels would potentially carry the information about internal infestation, which is more effectively used for discrimination of healthy cereals from insect-damaged cereals than other spectroscopic techniques. Singh *et al.* (2009) used LW-NIR HSI (900-1700nm) to discriminate insect-infested wheat kernels from healthy ones, with an accuracy of just over 85% based on LDA and quadratic discriminant analysis (QDA) classifiers. In order to improve the accuracy, NIR HSI system (700-1100 nm) combined with a colour imaging system was used to detect wheat kernels infested by four kinds of parasitic insects including rice weevil (*Sitophilus oryzae*), lesser grain borer (*Rhyzopertha dominica*), rusty grain beetle (*Cryptolestes ferrugineus*), and red flour beetle (*Tribolium castaneum*) (Singh *et al.,* 2010). It was concluded that based on

QDA classifier, the highest accuracy achieved was 96.40% for healthy and over 91% for insect-damaged wheat kernels. HSI technique can also be successfully applied to detect parasitic infestation in legumes. For instance, NIR HSI techniques in the range of 960 to 1700 nm were used to detect soybeans infested by egg, larval, and pupal stages of *Callosobruchus maculatus* along with uninfested and completely damaged (hollowed-out after emergence of adults) soybeans. As a result, pair-wise linear discriminant analysis (PWLDA) classification models yielded over 86% and 87% classification accuracy for uninfested and infested seeds (Chelladurai *et al.*, 2014). In another study, uninfested mung bean kernels and kernels infested with different extents of cowpea weevil (*Callosobruchus maculates*) were also effectively inspected by NIR HSI system (1000-1600 nm) (Kaliramesh *et al.*, 2013), and the average classification accuracies of more than 85% and 82% were obtained by statistical classifiers for identifying uninfested and infested mung bean kernels, respectively. The results showed that mung beans with pupal and adult stages of infestation had higher classification accuracies than the egg and larval stages of infestation. Furthermore, using HSI, an automatic threshold segmentation based on the iteration method and an optimal wavelength selection method were investigated by providing a new approach for the real-time and online detection of insect-infested vegetable soybean (Ma *et al.*, 2014). The classification results indicated that the normal bean samples were 100% correctly classified based on the method of automatic extracting the region of interest, for the insect-damaged samples with 91.7% accuracy being

ACCEPTED MANUSCRIPT

found. Additionally, Nansen *et al.* (2014) acquired HSI data (392-889 nm) from samples of field peas (*Pisum sativum*) with and without pea weevil (*Bruchus pisorum*) infestation. The classification performance of 100% was obtained with a combination of reflectance values in two spectral bands (641 nm and 868 nm), which also indicated that multispectral imaging with only two wavelengths would be more effective in detecting the weevil infestation.

3.6 Internal physiological aspects

Internal physiological disorders are caused by the abnormal growth pattern of staple food crops. This is due to adverse environmental conditions such as deviation from normal state of temperature, oxygen, moisture, nutrient, harmful gases and inadequate supply of growth regulators. The symptoms are typically expressed as hollow heart, black heart and internal brown spot in root tubers. Hollow heart in tubers may be caused by an excess of nitrogen fertiliser during the growth (Sparrow and Chapman, 2003). When tubers begin to grow rapidly in growth period, the tuber pith maybe die or pull apart leaving a void in the centre. This disorder can make the tubers unattractive and reduce the storage time as well as affect the quality of chips. Blackheart in tubers is due to hypoxia during growth or in storage. Tubers harvested from wet areas with high or low temperatures easily carry blackheart. In addition, internal brown spot of tuber is characterized by an internal necrosis of medullary tissue that greatly reduces culinary value of tuber. Some other physiological injuries from frozen tubers to chilling injury are usually caused by prolonged exposure to freezing temperatures during postharvest period.

Corresponding symptoms range from grey and black patches in the tuber tissue to a brown discolouration of the vascular ring. These physiological disorders are not visible and have a strong impact on root/tuber quality and value.

Some researchers reported to detect hollow hearts in potatoes by X-ray examination (Finney and Norris, 1978), but failed using acoustics method (Elbatawi, 2008) as the tiny hollow hearts cannot be detected and the noise is difficult to insulate. Besides, those methods are intensively dependent on the orientation of the potato. Nevertheless, spectroscopic techniques have been proved effectively for non-destructive predicting of internal physiological disorders in terms of black heart, hollow heart and internal brown spot in root tubers. Based on VIS/NIR transmission spectroscopic technique (513-850 nm), Zhou *et al.* (2015) compared three different morphological correction methods coupled with PLS-DA and PCA for detecting black heart in potatoes. Height corrected transmittance was found to have the best performance. With six wavelengths (711, 817, 741, 839, 678, and 698 nm) being selected, the overall classification rate of black heart for validation was 96.53%.

HSI was used to detect hollow heart and internal brown spot of tubers. Angel *et al.* (2011b) applied HSI technique (1000-1700 nm) to detect the presence of the hollow heart in potato tubers. The correct recognition rate of 89.10% was achieved by SVM and different image processing techniques. Similarly, internal brown spot in potato tubers were externally and non-destructively measured using time-resolved reflectance spectroscopy (540-900 nm) (Vanoli *et al.,* 2012). By detecting internal healthy and black spot tissues, the most sensitive wavelength for detection of internal brown spot was found at 690 nm.

4. Challenges and future trends of non-destructive techniques

Spectroscopic techniques including VIS/IR spectroscopy, Raman spectroscopy and NMR spectroscopy and spectral imaging have been successfully applied for staple foods quality evaluation and inspection as illustrated in Table 2. By guaranteeing the quality of food products with high precision and efficiency, these spectroscopic techniques proved to be non-invasive and cost-effective could be widely implemented in the food industry for automatic detection of cereals, legumes and root/tuber crops. These techniques require minimal or no sample preparation or destruction, which avoids potential pollution as well. However, some challenges still impede the applications of these spectroscopic techniques. Concerning the VIS/IR spectroscopy, it focuses only on a small portion of the sample and does not provide information on spatial distributions of whole specimen. Besides, it is difficult to detect objects with small size. With respect to Raman spectroscopy, it requires high-stability laser sources and sensitive amplification equipment to detect the weak signal that is easily interfered by biological fluorescence, which also makes Raman instruments cost more. Future work should focus on developing low-cost laser sources and amplification instruments. The NMR spectroscopy used for staple foods quality evaluation is also not cheap and it takes more time to interpret the complex spectra. Regarding the spectral imaging, especially full spectral imaging and HSI, a spectral image contains much more information than a single color image, which indicates that it needs lots of energy to extract the characteristic information from the lengthy and detailed spectral data. Compared with HSI, the advantages of full spectral imaging are the simplification of data storage, processing and interpretation, and the automatic removing of noise and redundant information, which means that full spectral imaging may have a potential in the rapid detection of food samples with improvements in technology in the near future (Bolton, 2004). Using HSI system to select several optimal wavelengths, the inexpensive multispectral imaging system can be developed to meet the industrial speed demand. The separation of different wavelengths for multispectral imaging can be generally done using either filters or instruments that are sensitive to specific wavelengths. Therefore, there is an inevitable trend for multispectral imaging with only a few important bands instead of full wavelengths in the non-destructive and rapid evaluation of food quality. With respect to these spectroscopic techniques, innovative calibration and prediction models with higher accuracy should be developed by eliminating data redundancy more conveniently for determining food quality in the future. Furthermore, in order to improve the model robustness and extend application boundary, the developed models should be evaluated using the same quality attribute (e.g. starch, protein, sprouting, and black heart) with different varieties of one specific staple food species. With the lowering of equipment price as well as progresses in developing instruments and novel algorithms, these non-destructive and rapid techniques will be more valuable for the food industry.

5. Conclusions

Spectroscopic techniques have been developed in the past decade, and these techniques have been extensively applied for the quality analysis of staple foods. In this review, several spectroscopic techniques including VIS/IR spectroscopy, Raman spectroscopy, NMR spectroscopy and spectral imaging are described in their great potentials in quality and safety evaluation of various staple foods. VIS/IR spectroscopy as a vital versatile technique, which could rapidly provide extensive information correlated well with sensory, adulteration, chemical, mycotoxin and internal physiological aspects of staple foods, and is suitable for measuring the quality of staple foods related to appearance, colour, texture, adulteration of species and origin, mycotoxin, black heart, carbohydrate, moisture, protein, lipid and ash content. NIR spectroscopy is becoming increasingly recognized as a complementary method for providing extensive information on staple food quality. Raman spectroscopy as a powerful tool is valuable to monitor the adulteration aspects related to geographical origin and different categories of kernels, chemical aspects including starch crystallinity, protein and oil contents, and microbiological aspects such as deoxynivalenol and aflatoxin. NMR spectroscopy has also been considered as an effective technique to evaluate staple foods focusing on adulteration aspects, and chemical aspects including amino acids, fatty acids, sugars, isoflavones and moisture distributions. As a more novel technique, HSI that integrates the advantages of spectroscopy and computer vision, has wider applications for quality inspection of staple foods in all kinds of quality aspects mentioned above. As a simplified form of HSI, multispectral imaging technology only holds a few important bands and will have greater potential in the field of non-destructive testing. These application results showed that spectroscopic techniques together with appropriate multivariate methods enabled a better control and analysis of staple foods quality.

Acknowledgements

The authors would like to acknowledge the UCD-CSC programme supported by University College Dublin and Chinese Scholarship Council.

References

Ahmed, N., Khan, S. H., Anjum, M. A. & Rehman, A. (2014). A cost effective preparative thin layer chromatography cleanup method for high performance liquid chromatography analysis of aflatoxins B1, B2 and G2. *Advancements in Life Sciences,* **2,** 1-4.

Angel, D.-N., Arno, F., Pilar, C., Esteban, V.-F. & Manuel, F.-D. (2011a). Common Scab Detection on Potatoes Using an infrared hyperspectral imaging system. *Image Analysis and Processing,* **6979,** 303-312.

Angel, D.-N., Arno, F., Pilar, C., Esteban, V.-F. & Manuel, F.-D. (2011b). Non–destructive Detection of Hollow Heart in Potatoes Using Hyperspectral Imaging. *Computer Analysis of Images and Patterns,* **6855,** 180-187.

Bannon, D. (2009). Hyperspectral Imaging Cubes and Slices. *Nature Photonics,* **3,** 627-629.

Barbin, D. F., Kaminishikawahara, C. M., Soares, A. L., Mizubuti, I. Y., Grespan, M., Shimokomaki, M. & Hirooka, E. Y. (2015). Prediction of chicken quality attributes by near infrared spectroscopy. *Food Chemistry,* **168,** 554-560.

BeMiller, J. N. (2009). Starch: Chemistry and Technology Third Edition Preface to the Third Edition. *Starch: Chemistry and Technology, 3rd Edition***,** Xvii-Xviii.

Bender, W. & Smith, M. (1997). Population food and nutrition. *Population Bulletin (Washington),* **51,** 1-48.

Bhat, R., Rai, R. V. & Karim, A. A. (2010). Mycotoxins in Food and Feed: Present Status and Future Concerns. *Comprehensive Reviews in Food Science and Food Safety,* **9,** 57-81.

Bolton, J. F. (2004). Full spectral imaging: a revisited approach to remote sensing. In: Remote Sensing. Pp. 243-251. International Society for Optics and Photonics.

Cheli, F., Battaglia, D., Gallo, R. & Dell'Orto, V. (2014). EU legislation on cereal safety: An update with a focus on mycotoxins. *Food Control,* **37,** 315-325.

Chelladurai, V., Karuppiah, K., Jayas, D. S., Fields, P. G. & White, N. D. G. (2014). Detection of Callosobruchus maculatus (F.) infestation in soybean using soft X-ray and NIR hyperspectral imaging techniques. *Journal of Stored Products Research,* **57,** 43-48.

Chen, J., Ren, X., Zhang, Q., Diao, X. & Shen, Q. (2013). Determination of protein, total carbohydrates and crude fat contents of foxtail millet using effective wavelengths in NIR spectroscopy. *Journal of Cereal Science,* **58,** 241-247.

Corpaş, L., Hădărugă, N. G., David, I., Pîrşan, P., Hădărugă, D. I. & Isengard, H.-D. (2014). Karl Fischer Water Titration− Principal Component Analysis Approach on Wheat Flour. *Food Analytical Methods,* **7,** 1353-1358.

Corvucci, F., Nobili, L., Melucci, D. & Grillenzoni, F. V. (2015). The discrimination of honey origin using melissopalynology and Raman spectroscopy techniques coupled with multivariate analysis. *Food Chemistry,* **169,** 297-304.

Cozzolino, D., Roumeliotis, S. & Eglinton, J. (2014). Evaluation of the use of attenuated total reflectance mid infrared spectroscopy to determine fatty acids in intact seeds of barley (Hordeum vulgare). *LWT - Food Science and Technology*, **56,** 478-483.

Sirisomboon, C. D., Putthang, R. & Sirisomboon, P. (2013). Application of near infrared spectroscopy to detect aflatoxigenic fungal contamination in rice. *Food Control*, **33,** 207-214.

Dais, P. & Hatzakis, E. (2013). Quality assessment and authentication of virgin olive oil by NMR spectroscopy: a critical review. *Analytica chimica acta*, **765,** 1-27.

Demir, P., Onde, S. & Severcan, F. (2015). Phylogeny of cultivated and wild wheat species using ATR–FTIR spectroscopy. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, **135,** 757-763.

Domingo, E., Tirelli, A. A., Nunes, C. A., Guerreiro, M. C. & Pinto, S. M. (2014). Melamine detection in milk using vibrational spectroscopy and chemometrics analysis: A review. *Food Research International*, **60,** 131-139.

Elbatawi, I. E. (2008). An acoustic nimpact method to detect hollow heart of potato tubers. *Biosystems Engineering*, **100,** 206-213.

Esteve Agelet, L., Ellis, D. D., Duvick, S., Goggi, A. S., Hurburgh, C. R. & Gardner, C. A. (2012). Feasibility of near infrared spectroscopy for analyzing corn kernel damage and viability of soybean and corn kernels. *Journal of Cereal Science*, **55,** 160-165.

Feng, X., Zhang, Q., Cong, P. & Zhu, Z. (2013). Preliminary study on classification of rice and detection of paraffin in the adulterated samples by Raman spectroscopy combined with multivariate analysis. *Talanta,* **115,** 548-55.

Ferreira, D. S., Galão, O. F., Pallone, J. A. L. & Poppi, R. J. (2014). Comparison and application of near-infrared (NIR) and mid-infrared (MIR) spectroscopy for determination of quality parameters in soybean samples. *Food Control,* **35,** 227-232.

Ferreira, D. S., Pallone, J. A. L. & Poppi, R. J. (2013). Fourier transform near-infrared spectroscopy (FT-NIRS) application to estimate Brazilian soybean [Glycine max (L.) Merril] composition. *Food Research International,* **51,** 53-58.

Finney, E. E. & Norris, K. H. (1978). X-Ray Scans for Detecting Hollow Heart in Potatoes. *American Potato Journal,* **55,** 95-105.

Fu, B. X., Hatcher, D. W. & Schlichting, L. (2014). Effects of sprout damage on durum wheat milling and pasta processing quality. *Canadian Journal of Plant Science,* **94,** 545-553.

Gaspardo, B., Del Zotto, S., Torelli, E., Cividino, S., Firrao, G., Della Riccia, G. & Stefanon, B. (2012). A rapid method for detection of fumonisins B 1 and B 2 in corn meal using Fourier transform near infrared (FT-NIR) spectroscopy implemented with integrating sphere. *Food Chemistry,* **135,** 1608-1612.

Giacomo, D. & Stefania, D. (2013). A multivariate regression model for detection of fumonisins content in maize from near infrared spectra. *Food Chemistry,* **141,** 4289-4294.

ACCEPTED MANUSCRIPT

Haughey, S. A., Graham, S. F., Cancouët, E. & Elliott, C. T. (2013). The application of Near-Infrared Reflectance Spectroscopy (NIRS) to detect melamine adulteration of soya bean meal. *Food Chemistry,* **136,** 1557-1561.

Herrero, A. M. (2008). Raman spectroscopy a promising technique for quality assessment of meat and fish: A review. *Food Chemistry,* **107,** 1642-1651.

Holse, M., Larsen, F. H., Hansen, Å. & Engelsen, S. B. (2011). Characterization of marama bean (Tylosema esculentum) by comparative spectroscopy: NMR, FT-Raman, FT-IR and NIR. *Food Research International,* **44,** 373-384.

Horigane, A. K., Suzuki, K. & Yoshida, M. (2014). Moisture distribution in rice grains used for sake brewing analyzed by magnetic resonance imaging. *Journal of Cereal Science,* **60,** 193-201.

Hou, H. & Chang, K. (2004). Storage conditions affect soybean color, chemical composition and tofu qualities. *Journal of Food Processing and Preservation,* **28,** 473-488.

Huang, M., Wang, Q., Zhang, M. & Zhu, Q. (2014). Prediction of color and moisture content for vegetable soybean during drying using hyperspectral imaging technology. *Journal of Food Engineering,* **128,** 24-30.

Hug-Iten, S., Escher, F. & Conde-Petit, B. (2003). Staling of bread: Role of amylose and amylopectin and influence of starch-degrading enzymes. *Cereal Chemistry,* **80,** 654-661.

Hussein, H. S. & Brasel, J. M. (2001). Toxicity, metabolism, and impact of mycotoxins on humans and animals. *Toxicology,* **167,** 101-134.

Hwang, J., Kang, S., Lee, K. & Chung, H. (2012). Enhanced Raman spectroscopic discrimination of the geographical origins of rice samples via transmission spectral collection through packed grains. *Talanta,* **101,** 488-94.

Jääskeläinen, A.-S., Holopainen-Mantila, U., Tamminen, T. & Vuorinen, T. (2013). Endosperm and aleurone cell structure in barley and wheat as studied by optical and Raman microscopy. *Journal of Cereal Science,* **57,** 543-550.

Jiao, Z., Si, X.-x., Zhang, Z.-m., Li, G.-k. & Cai, Z.-w. (2012). Compositional study of different soybean (Glycine max L.) varieties by 1H NMR spectroscopy, chromatographic and spectrometric techniques. *Food Chemistry,* **135,** 285-291.

Jin, H., Li, L. & Cheng, J. (2015). Rapid and Non-destructive Determination of Moisture Content of Peanut Kernels Using Hyperspectral Imaging Technique. *Food Analytical Methods*.

Jung, S., Rickert, D. A., Deak, N. A., Aldin, E. D., Recknor, J., Johnson, L. A. & Murphy, P. A. (2003). Comparison of Kjeldahl and Dumas methods for determining protein contents of soybean products. *Journal of the American Oil Chemists Society,* **80,** 1169-1173.

Kaliramesh, S., Chelladurai, V., Jayas, D. S., Alagusundaram, K., White, N. D. G. & Fields, P. G. (2013). Detection of infestation by Callosobruchus maculatus in mung bean using near-infrared hyperspectral imaging. *Journal of Stored Products Research,* **52,** 107-111.

Kamruzzaman, M., Makino, Y. & Oshita, S. (2015). Non-invasive analytical technology for the detection of contamination, adulteration, and authenticity of meat, poultry, and fish: A review. *Analytica Chimica Acta,* **853,** 19-29.

Kandpal, L. M., Lee, S., Kim, M. S., Bae, H. & Cho, B.-K. (2015). Short wave infrared (SWIR) hyperspectral imaging technique for examination of aflatoxin B 1 (AFB 1) on corn kernels. *Food Control,* **51,** 171-176.

Kaya-Celiker, H., Mallikarjunan, P. K., Schmale, D. & Christie, M. E. (2014). Discrimination of moldy peanuts with reference to aflatoxin using FTIR-ATR system. *Food Control*, **44,** 64-71.

Kirinčič, S., Škrjanc, B., Kos, N., Kozolc, B., Pirnat, N. & Tavčar-Kalcher, G. (2015). Mycotoxins in cereals and cereal products in Slovenia – Official control of foods in the years 2008–2012. *Food Control,* **50,** 157-165.

Kouvoutsakis, G., Mitsi, C., Tarantilis, P., Polissiou, M. & Pappas, C. (2014). Geographical differentiation of dried lentil seed (Lens culinaris) samples using Diffuse Reflectance Fourier Transform Infrared Spectroscopy (DRIFTS) and discriminant analysis. *Food Chemistry,* **145,** 1011-1014.

Kwon, Y.-K., Jie, E. Y., Sartie, A., Kim, D. J., Liu, J. R., Min, B. W. & Kim, S. W. (2015). Rapid metabolic discrimination and prediction of dioscin content from African yam

ACCEPTED MANUSCRIPT

tubers using Fourier transform-infrared spectroscopy combined with multivariate analysis. *Food Chemistry,* **166,** 389-396.

López, A., Arazuri, S., Jarén, C., Mangado, J., Arnal, P., Galarreta, J. I. R. d., Riga, P. & López, R. (2013). Crude Protein Content Determination of Potatoes by NIRS Technology. *Procedia Technology,* **8,** 488-492.

Laghi, L., Picone, G. & Capozzi, F. (2014). Nuclear magnetic resonance for foodomics beyond food analysis. *Trac-Trends in Analytical Chemistry,* **59,** 93-102.

Lamanna, R., Cattivelli, L., Miglietta, M. L. & Troccoli, A. (2011). Geographical origin of durum wheat studied by H-1-NMR profiling. *Magnetic Resonance in Chemistry,* **49,** 1-5.

Lee, H., Cho, B.-K., Kim, M. S., Lee, W.-H., Tewari, J., Bae, H., Sohn, S.-I. & Chi, H.-Y. (2013). Prediction of crude protein and oil content of soybeans using Raman spectroscopy. *Sensors and Actuators B: Chemical,* **185,** 694-700.

Lee, K.-M., Herrman, T. J. & Yun, U. (2014a). Application of Raman spectroscopy for qualitative and quantitative analysis of aflatoxins in ground maize samples. *Journal of Cereal Science,* **59,** 70-78.

Lee, K. M., Herrman, T. J., Bisrat, Y. & Murray, S. C. (2014b). Feasibility of Surface-Enhanced Raman Spectroscopy for Rapid Detection of Aflatoxins in Maize. *Journal of Agricultural and Food Chemistry,* **62,** 4466-4474.

Liu, J., Wen, Y., Dong, N., Lai, C. & Zhao, G. (2013). Authentication of lotus root powder adulterated with potato starch and/or sweet potato starch using Fourier transform mid-infrared spectroscopy. *Food Chemistry*, **141,** 3103-3109.

Liu, Y., Delwiche, S. R. & Dong, Y. (2009). Feasibility of FT-Raman spectroscopy for rapid screening for DON toxin in ground wheat and barley. *Food Additives and Contaminants Part a-Chemistry Analysis Control Exposure & Risk Assessment*, **26,** 1396-1401.

Liu, Z.-Y., Wu, H.-F. & Huang, J.-F. (2010). Application of neural networks to discriminate fungal infection levels in rice panicles using hyperspectral reflectance and principal components analysis. *Computers and Electronics in Agriculture*, **72,** 99-106.

Ma, Y., Huang, M., Yang, B. & Zhu, Q. (2014). Automatic threshold method and optimal wavelength selection for insect-damaged vegetable soybean detection using hyperspectral images. *Computers and Electronics in Agriculture*, **106,** 102-110.

Magwaza, L. S., Opara, U. L., Nieuwoudt, H., Cronje, P. J., Saeys, W. & Nicolaï, B. (2012). NIR spectroscopy applications for internal and external quality analysis of citrus fruit—a review. *Food and Bioprocess Technology*, **5,** 425-444.

Mahesh, S., Jayas, D., Paliwal, J. & White, N. (2015). Comparison of Partial Least Squares Regression (PLSR) and Principal Components Regression (PCR) Methods for Protein and Hardness Predictions using the Near-Infrared (NIR) Hyperspectral Images of Bulk Samples of Canadian Wheat. *Food and Bioprocess Technology*, **8,** 31-40.

Mendoza, F. A., Cichy, K., Lu, R. F. & Kelly, J. D. (2014). Evaluation of Canning Quality Traits in Black Beans (Phaseolus vulgaris L.) by Visible/Near-Infrared Spectroscopy. *Food and Bioprocess Technology,* **7,** 2666-2678.

Mishra, P., Herrero-Langreo, A., Barreiro, P., Roger, J. M., Diezma, B., Gorretta, N. & Lleó, L. (2015). Detection and quantification of peanut traces in wheat flour by near infrared hyperspectral imaging spectroscopy using principal-component analysis. *Journal of Near Infrared Spectroscopy,* **23,** 15-22.

Monakhova, Y. B., Rutledge, D. N., Rossmann, A., Waiblinger, H. U., Mahler, M., Ilse, M., Kuballa, T. & Lachenmeier, D. W. (2014). Determination of rice type by H-1 NMR spectroscopy in combination with different chemometric tools. *Journal of Chemometrics,* **28,** 83-92.

Monteiro, S. T., Minekawa, Y., Kosugi, Y., Akazawa, T. & Oda, K. (2007). Prediction of sweetness and amino acid content in soybean crops from hyperspectral imagery. *ISPRS Journal of Photogrammetry and Remote Sensing,* **62,** 2-12.

Mutungi, C., Passauer, L., Onyango, C., Jaros, D. & Rohm, H. (2012). Debranched cassava starch crystallinity determination by Raman spectroscopy: Correlation of features in Raman spectra with X-ray diffraction and 13C CP/MAS NMR spectroscopy. *Carbohydrate Polymers,* **87,** 598-606.

ACCEPTED MANUSCRIPT

Naerstad, R., Dees, M. W., Le, V. H., Holgado, R. & Hermansen, A. (2012). Occurrence of Skin Blemish Diseases (Scab and Scurf) in Norwegian Potato Production. *Potato Research,* **55,** 225-239.

Nansen, C., Zhang, X., Aryamanesh, N. & Yan, G. (2014). Use of variogram analysis to classify field peas with and without internal defects caused by weevil infestation. *Journal of Food Engineering*, **123,** 17-22.

Nie, P., Wu, D., Yang, Y. & He, Y. (2012). Fast determination of boiling time of yardlong bean using visible and near infrared spectroscopy and chemometrics. *Journal of Food Engineering*, **109,** 155-161.

Nogales-Bueno, J., Rodriguez-Pulido, F. J., Heredia, F. J. & Hernandez-Hierro, J. M. (2015). Comparative study on the use of anthocyanin profile, color image analysis and near-infrared hyperspectral imaging as tools to discriminate between four autochthonous red grape cultivars from La Rioja (Spain). *Talanta*, **131,** 412-416.

Novy, R. (2014). Traditional Breeding and Cultivar Development. *The Potato: Botany, Production and Uses*, 272-289.

Ohtsuki, T., Sato, K., Abe, Y., Sugimoto, N. & Akiyama, H. (2015). Quantification of acesulfame potassium in processed foods by quantitative H-1 NMR. *Talanta,* **131,** 712-718.

Pitt, J. I., Taniwaki, M. H. & Cole, M. B. (2013). Mycotoxin production in major crops as influenced by growing, harvesting, storage and processing, with emphasis on the achievement of Food Safety Objectives. *Food Control*, **32,** 205-215.

Pu, Y. Y., Feng, Y. Z. & Sun, D.-W. (2015). Recent Progress of Hyperspectral Imaging on Quality and Safety Inspection of Fruits and Vegetables: A Review. *Comprehensive Reviews in Food Science and Food Safety*, **14,** 176-188.

Rady, A. M., Guyer, D. E., Kirk, W. & Donis-González, I. R. (2014). The potential use of visible/near infrared spectroscopy and hyperspectral imaging to predict processing-related constituents of potatoes. *Journal of Food Engineering*, **135,** 11-25.

Sánchez, T., Ceballos, H., Dufour, D., Ortiz, D., Morante, N., Calle, F., Zum Felde, T., Domínguez, M. & Davrieux, F. (2014). Prediction of carotenoids, cyanide and dry matter contents in fresh cassava root using NIRS and Hunter color techniques. *Food Chemistry*, **151,** 444-451.

Serranti, S., Cesare, D. & Bonifazi, G. (2013a). The development of a hyperspectral imaging method for the detection of Fusarium-damaged, yellow berry and vitreous Italian durum wheat kernels. *Biosystems Engineering*, **115,** 20-30.

Serranti, S., Cesare, D., Marini, F. & Bonifazi, G. (2013b). Classification of oat and groat kernels using NIR hyperspectral imaging. *Talanta*, **103,** 276-84.

Shahin, M. A., Symons, S. J. & Hatcher, D. W. (2014). Quantification of Mildew Damage in Soft Red Winter Wheat Based on Spectral Characteristics of Bulk Samples: A Comparison of Visible-Near-Infrared Imaging and Near-Infrared Spectroscopy. *Food and Bioprocess Technology,* **7,** 224-234.

Singh, C. B., Jayas, D. S., Paliwal, J. & White, N. D. G. (2009). Detection of insect-damaged wheat kernels using near-infrared hyperspectral imaging. *Journal of Stored Products Research,* **45,** 151-158.

Singh, C. B., Jayas, D. S., Paliwal, J. & White, N. D. G. (2010). Identification of insect-damaged wheat kernels using short-wave near-infrared hyperspectral and digital colour imaging. *Computers and Electronics in Agriculture,* **73,** 118-125.

Siripatrawan, U. & Makino, Y. (2015). Monitoring fungal growth on brown rice grains using rapid and non-destructive hyperspectral imaging. *International Journal of Food Microbiology,* **199,** 93-100.

Song, J., Liu, C., Li, D. & Gu, Z. (2013). Evaluation of sugar, free amino acid, and organic acid compositions of different varieties of vegetable soybean (Glycine max [L.] Merr). *Industrial Crops and Products,* **50,** 743-749.

Sowokinos, J. (1978). Relationship of harvest sucrose content to processing maturity and storage life of potatoes. *American Potato Journal,* **55,** 333-344.

Sparrow, L. & Chapman, K. (2003). Effects of nitrogen fertiliser on potato (Solanum tuberosum L., cv. Russet Burbank) in Tasmania. 1. Yield and quality. *Animal Production Science*, **43,** 631-641.

Sun, D.-W. (2009). Infrared spectroscopy for food quality analysis and control. Academic Press, Elsevier, San Diego, California, USA.

Sun, D.-W. (2010). Hyperspectral imaging for food quality analysis and control. Academic Press, Elsevier, San Diego, California, USA.

Tahkapaa, S., Maijala, R., Korkeala, H. & Nevas, M. (2015). Patterns of food frauds and adulterations reported in the EU rapid alert system for food and feed and in Finland. *Food Control*, **47,** 175-184.

Trematerra, P., Gentile, P. & Djikoloum, T. (2003). Traditional cereal storage and insect pests in some villages of southern Chad. *Advances in Stored Product Protection***,** 281-287.

Uarrota, V. G., Moresco, R., Coelho, B., da Costa Nunes, E., Peruch, L. A. M., de Oliveira Neubert, E., Rocha, M. & Maraschin, M. (2014). Metabolomics combined with chemometric tools (PCA, HCA, PLS-DA and SVM) for screening cassava (Manihot esculenta Crantz) roots during postharvest physiological deterioration. *Food Chemistry*, **161,** 67-78.

Ustin, S. L., Roberts, D. A., Gamon, J. A., Asner, G. P. & Green, R. O. (2004). Using imaging spectroscopy to study ecosystem processes and properties. *Bioscience,* **54,** 523-534.

 ACCEPTED MANUSCRIPT

Vanoli, M., Rizzolo, A., Spinelli, L., Parisi, B. & Torricelli, A. (2012). Non destructive detection of Internal Brown Spot in potato tubers by time-resolved reflectance spectroscopy: preliminary results on a susceptible cultivar. In Proc. International Conference of Agricultural Engineering, PF: Post Harvest, Food and Process Engineering.

Wang, L., Liu, D., Pu, H. B., Sun, D.-W., Gao, W. H. & Xiong, Z. J. (2015). Use of Hyperspectral Imaging to Discriminate the Variety and Quality of Rice. *Food Analytical Methods,* **8,** 515-523.

Wang, W., Heitschmidt, G. W., Ni, X., Windham, W. R., Hawkins, S. & Chu, X. (2014). Identification of aflatoxin B1 on maize kernel surfaces using hyperspectral imaging. *Food Control,* **42,** 78-86.

Xing, J., Symons, S., Hatcher, D. & Shahin, M. (2011). Comparison of short-wavelength infrared (SWIR) hyperspectral imaging system with an FT-NIR spectrophotometer for predicting alpha-amylase activities in individual Canadian Western Red Spring (CWRS) wheat kernels. *Biosystems Engineering,* **108,** 303-310.

Xing, J., Symons, S., Shahin, M. & Hatcher, D. (2010). Detection of sprout damage in Canada Western Red Spring wheat with multiple wavebands using visible/near-infrared hyperspectral imaging. *Biosystems Engineering,* **106,** 188-194.

 ACCEPTED MANUSCRIPT

Xu, W., Liu, X., Xie, L. & Ying, Y. (2014). Comparison of Fourier Transform near-Infrared, Visible near-Infrared, Mid-Infrared, and Raman Spectroscopy as Non-Invasive Tools for Transgenic Rice Discrimination. *Transactions of the ASABE,* **57,** 141-150.

Yao, H., Hruska, Z., Kincaid, R., Brown, R., Bhatnagar, D. & Cleveland, T. (2013a). Hyperspectral image classification and development of fluorescence index for single corn kernels infected with Aspergillus flavus. *Transactions of the ASABE*, **56,** 1977-1988.

Yao, H., Hruska, Z., Kincaid, R., Brown, R. L., Bhatnagar, D. & Cleveland, T. E. (2013b). Detecting maize inoculated with toxigenic and atoxigenic fungal strains with fluorescence hyperspectral imagery. *Biosystems Engineering*, **115,** 125-135.

Zhang, X. L., Liu, F., He, Y. & Li, X. L. (2012). Application of Hyperspectral Imaging and Chemometric Calibrations for Variety Discrimination of Maize Seeds. *Sensors,* **12,** 17234-17246.

Zhao, H., Guo, B., Wei, Y. & Zhang, B. (2013). Near infrared reflectance spectroscopy for determination of the geographical origin of wheat. *Food Chemistry,* **138,** 1902-1907.

Zhou, Z., Zeng, S., Li, X. & Zheng, J. (2015). Nondestructive Detection of Blackheart in Potato by Visible/Near Infrared Transmittance Spectroscopy. *Journal of Spectroscopy*, DOI: 10.1155/2015/786709.

Table 1. The application principles and main instrumental components of spectroscopic techniques.

Spectrum	VIS	SW-NIR	LW-NIR	MIR	FIR	Main components	Characteristics
Wavelength /nm	380 - 780	780 - 1100	1100 - 2500	2500 - 25000	25000 - 300000		
Wavenumber /cm^{-1}	26316 - 12821	12821 - 9091	9091 - 4000	4000 - 400	400 - 33		
VIS/IR spectroscopy	√	√	√	√	√	Spectrometer, computer	Identifying structural properties of chemical components of complex mixtures in a small area

Raman spectroscopy				√	√	Imaging spectrometer, CCD camera, probe, laser light source and laser focus module, computer	Providing a fingerprint by which molecules can be identified
NMR spectroscopy						Spectrometer, computer	Determining the structure, dynamics, reaction state, and chemical environment of molecules
Spectral imaging	√	√	√			Halogen lamps, spectrograph,	Collecting spectral information of

						CCD camera, translation stage, computer	chemical components of every location within an image obtained

VIS: Visible, SW-NIR: Short-wave near-infrared, LW-NIR: long-wave near-infrared, MIR: Mid-infrared, FIR: Far- infrared. NMR: Nuclear magnetic resonance.

Table 2. Overview of spectroscopic techniques for the quality evaluation of staple foods.

Quality aspects	Staple foods	Spectroscopy techniques	Wavelength range (nm)	Multivariable analysis	Accuracy	References
Sensory	Black bean	VIS/NIR	400-2500	PLSR	>72.60% (Appearance, colour, and texture)	Mendoza *et al.* (2014)
	Vegetable soybean	HSI	400-1000	PLSR	R^2 = 0.74 (colour)	Huang *et al.* (2014)
	wheat	HSI	960-1700	PLSR, PCR	R^2 = 0.67 (Hardness)	Mahesh *et al.* (2015)
	Rice	HSI	400-1000	PCA, BPNN	94.45% (Shape, chalkiness degree)	Wang *et al.* (2015)
Adulteratio	Maize	HSI	380-1030	PCA,	98.89%	Zhang *et al.*

n				KPCA, LS-SVM, BPNN	(Species)	(2012)
	Wheat	NIR	950-1650	PCA, LDA, DPLS	92.50% (Geographical origin)	Zhao *et al.* (2013)
	Soybean meal	NIR	833-2632	PCA, PLSR, PCR	R^2 = 0.89-0.99 (Melamine adulteration)	Haughey *et al.* (2013)
	Lotus root powder, potato starch, sweet potato starch	MIR	2500-20000	PCA	R^2 = 0.96-0.99	Liu *et al.* (2013)

	Rice	Raman	6250-100000	SIMCA, PLS-DA, KNN, SVM	＞90% (Geographical origins)	Feng *et al.* (2013)
	Oat	HSI	880-1720	PLS-DA	Almost 100% (Classification)	Serranti *et al.* (2013b)
	Lentil	MIR	4444-5814, 7843-10472	MA	100% (Geographical origin)	Kouvoutsakis *et al.* (2014)
	Rice	NMR	-	PLS-DA, ICA	0.96 (Geographical origins)	Monakhova *et al.* (2014)
	Wheat	MIR	2500-15385	HCA, PCA	87.14% (Species)	Demir *et al.* (2015)
	Wheat flour	HSI	900-1700	PCA	R^2 = 0.95 (Peanut traces of 0.1%)	Mishra *et al.* (2015)

 ACCEPTED MANUSCRIPT

Chemical	Cassava	Raman	2857-33333	-	R^2 = 0.99 (Starch crystallinity)	Mutungi *et al.* (2012)
	Soybean	Raman	5555-50000	PLSR	R^2 = 0.92 (Protein), R^2 = 0.87 (Oil)	Lee *et al.* (2013)
	Millet	NIR	950-1650	iPLS, SPA, MLR	R^2 = 0.94 (Protein), R^2 = 0.92 (Total carbohydrates), R^2 = 0.70 (Crude fat)	Chen *et al.* (2013)
	Cassava	NIR	1100-2500	PCA, PLSR, SD	R^2 = 0.96 (Dry matter), R^2 = 0.92 (Carotenoids), R^2 = 0.96 (Carotene),	Sánchez *et al.* (2014)

					R^2 = 0.86 (Cyanogenic)	
	Vegetable soybean	HSI	400-1000	PLSR	R^2 = 0.94 (Moisture)	Huang *et al.* (2014)
	Soybean	NIR/MIR	1250-2500, 2500-25000	PLSR	R^2 = 0.72 (Moisture), R^2 = 0.73 (Ash), R^2 = 0.88 (Protein), R^2 = 0.81 (Lipid) for NIR; R^2 = 0.63 (Moisture), R^2 = 0.87 (Ash), R^2 = 0.91	Ferreira *et al.* (2014)

ACCEPTED MANUSCRIPT

					(Protein), R^2 = 0.67 (Lipid) for MIR	
	Barley	MIR	2500-26667	PLSR	R^2 = 0.76 (Stearic), R^2 = 0.75 (Oleic), R^2 = 0.45 (Palmitic acids), R^2 = 0.89 (Total lipids)	Cozzolino *et al.* (2014)
	Yam	MIR	2500-25000	PLS-DA	R^2 = 0.72 (Dioscin)	Kwon *et al.* (2015)
	Peanut	HSI	400-2500	PLSR	R^2 = 0.91 (Moisture)	Jin *et al.* (2015)
Mycotoxin	Corn	VIS/NIR	650-2500	PCR, PLSR	R^2 = 0.92	Gaspardo *et*

					(Fumonisins B1 and B2)	*al.* (2012)
	Maize	HSI	400-700	C-VDA	94% (Fungal infection for 100 ppb)	Yao *et al.* (2013b)
	Peanuts	MIR	2500-16000	PLSR	R^2 = 99.98% (Aflatoxin)	Kaya-Celiker *et al.* (2014)
	Maize	Raman	2857-50000	PLSR	R^2 = 0.90-0.96 (Fungal aflatoxin)	Lee *et al.* (2014a)
	Maize	HSI	1000-2500	PCA, FDA	>88% (Aflatoxin B1)	Wang *et al.* (2014)
	Wheat	HSI	400-1000, 1000-2500	PLSR	96% (Mildew)	Shahin *et al.* (2014)
	Corn	HSI	1000-1700	PLS-DA	96.90%	Kandpal *et*

 ACCEPTED MANUSCRIPT

					(Aflatoxin B1)	*al.* (2015)
	Rice	HSI	400-1000	PLSR	0.97 (Fungal infection)	Siripatrawan and Makino (2015)
Parasitic infection	Vegetable soybean	HSI	400-1000	FRSTCA, SVDD	98.80% (Insect)	Ma *et al.* (2014)
	Field peas	HSI	392-889	LDA	100% (Weevil)	Nansen *et al.* (2014)
Internal physiological	Potato	HSI	900-1700	SVM	89.10% (Hollow heart)	Angel *et al.* (2011b)
	Potato	VIS/NIR	513-850	PLS-DA, PCA	96.53% (Black heart)	Zhou *et al.* (2015)

R^2: Coefficient of determination, LS-SVM: Least squares-support vector machines, DPLS: Discriminant partial least squares, iPLS: Interval partial least squares, PLSR: Partial least squares regression, HCA: Hierarchical clustering analyses, SPA: Successive projections algorithm, PCA: Principal component analysis, KPCA: Kernel principal component analysis, SIMCA: Soft independent modeling of class analogy, KNN: K-nearest neighbors, SVM: Support vector machine, PCR: Principal component regression, SD: Standard deviation, MA: Multivariate analysis, MLR: Multiple linear regression, LDA: Linear discriminant analysis, IS: Image subtraction, PLS-DA: Partial least squares discriminant analyses, FRSTCA: Fuzzy-rough set model based on the thermal charge algorithm, SVDD: Support vector data description, FDA: Factorial discriminant analysis, C-VDA: Cross-validation discriminant analysis, BPNN: Back propagation neural network, ICA: Independent component analysis.

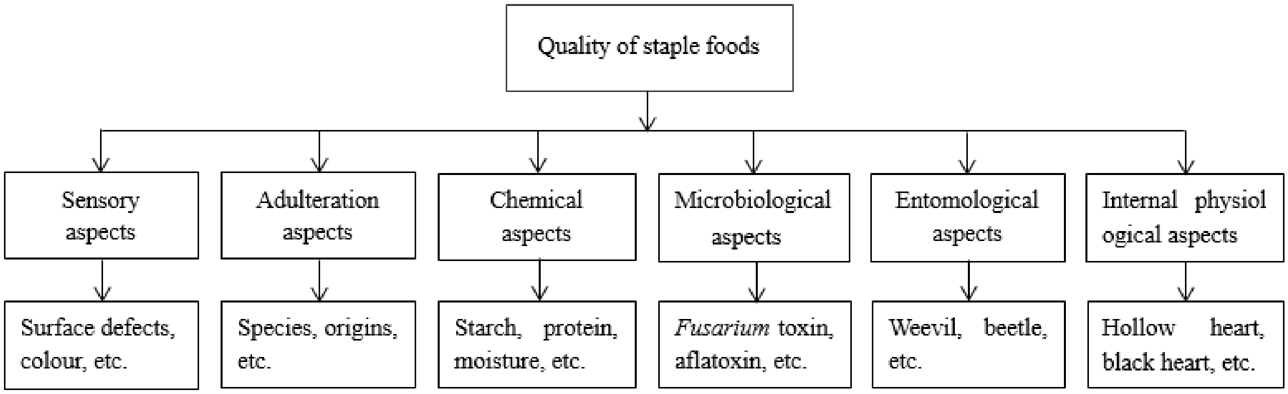

Fig. 1. Common quality evaluation of staple foods.

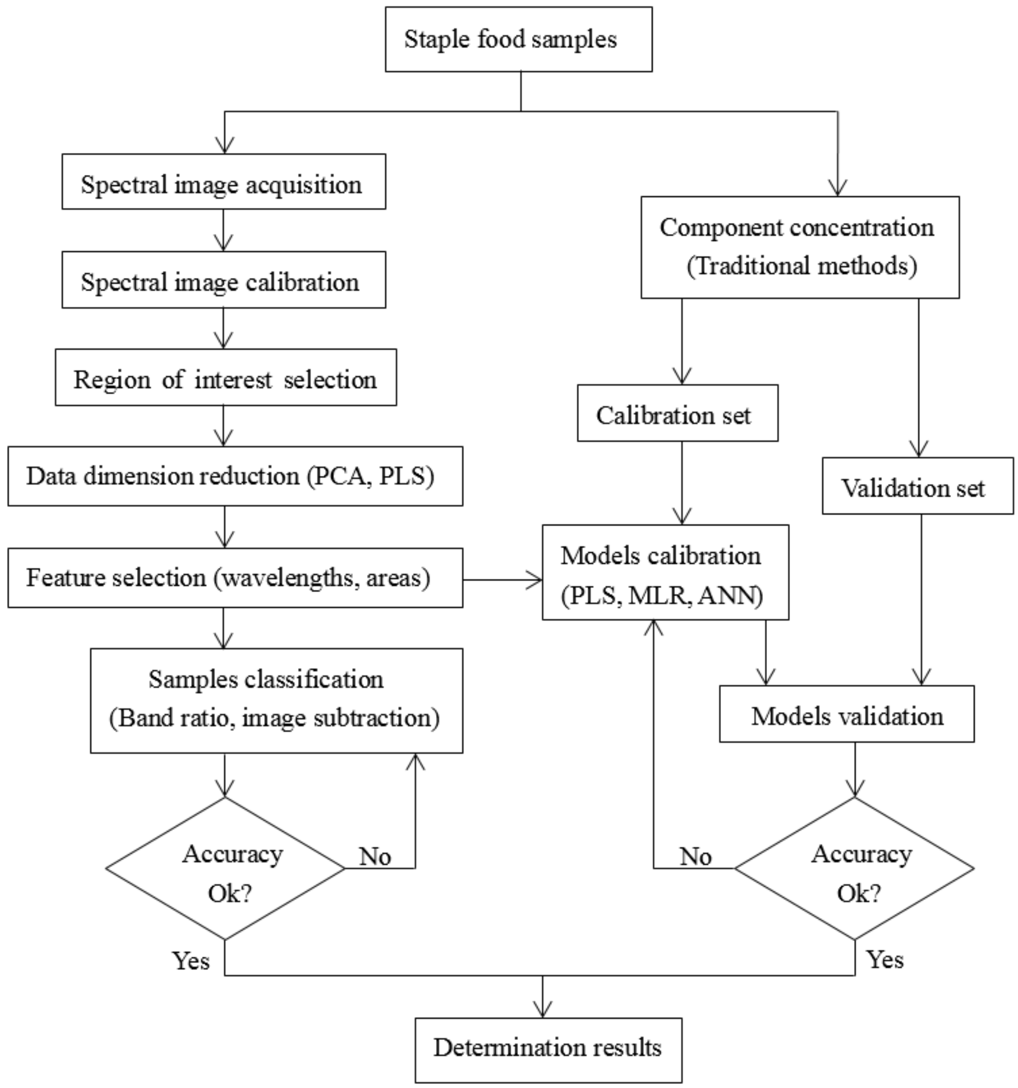

Fig. 2. Flow chart of evaluation of staple food quality by imaging spectroscopy.

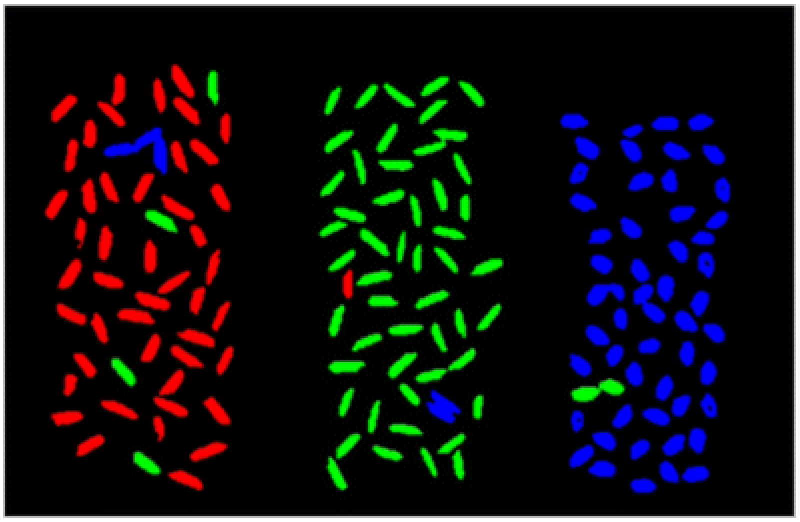

Fig. 3. Classification of rice variety based on colour and shape using hyperspectral imaging (*red* rice I, *green* rice II, *blue* rice III) (Wang *et al.*, 2015).