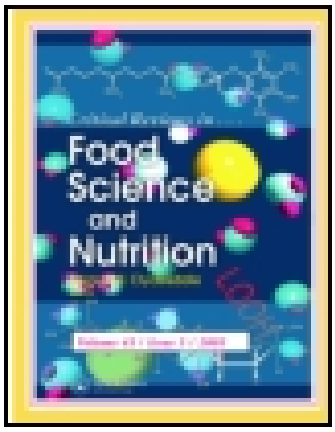

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Detection of Adulterants and Contaminants in Liquid Foods - A Review

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Milk and fruit juices have paramount importance in human diet. Increasing demand of these liquid foods has made them vulnerable to economic adulteration during processing and in supply chain. Adulterants are difficult to detect by consumers and thus necessitating the requirement of rapid, accurate and sensitive detection. The potential adulterants in milk and fruit juices and their limits set by different regulatory bodies have been briefly described in this review. Potential

advantages and limitations of various techniques such as physicochemical methods, chromatography, immunoassays, molecular, electrical, spectroscopy with chemometrics, electronic nose and biosensors have been described. Spectroscopy in combination with chemometrics has shown potential for rapid, precise and sensitive detection of potential adulterants in these liquid foods.

Keywords: Milk, fruit juices, chromatography, immunoassay, spectroscopy, chemometrics, biosensors

INTRODUCTION

Milk and fruit juices undergo numerous processing steps which makes them the most vulnerable for adulteration and contamination before reaching to consumers. High protein content in milk and vitamins in fruit juices make them specifically essential for certain groups of consumer's viz. women, children and the elderly; thereby playing an important role in feeding the population. Unscrupulous practice of adulteration has been prevalent since ancient times, and has increased manifold in recent time (International Dairy Federation, 2008). Dairy products (milk, ice-cream, yogurt, butter, cheese, etc.) are in tremendous demand and are sold at premium prices. Therefore, from an economic point of view, it could be attractive to modify its composition and replace part of it with other dairy or non-dairy ingredients leading to adulteration (De La Fuente and Juarez, 2005; Arvaniyoyannis and Tzouros, 2005; Moore et al., 2012). Similarly juices of costly fruits are adulterated with similar looking low value juices to get quick economic benefit. The objective of this review is to bring all information on adulteration of milk and fruit juices at one platform and to discuss the feasibility of different methods employed for their detection.

Milk Adulteration

Adulteration of milk products have found its way in common man's lives and have resulted in death of many adults and children. Three lakh cases of sick infants were reported in China due to contaminated milk formula which included six deaths and 860 hospitalizations by 2008 (BBC, 2008; Gossner et al., 2009; Yan et al., 2009). The infants were found to be exhibiting urinary problems, renal blockages and kidney stones which otherwise are rare in case

of infants due to adulteration with an organic base chemical melamine. Melamine is widely used in plastics, adhesives, dishware and whiteboards (Andersen et al., 2008; Garber, 2008) but their use in food products is not approved by FAO/WHO/ CODEX Alimantarius. Still due to its high nitrogen content, it is being added to milk illegally to make it appear high in protein content as standard protein determination assays (e.g. Kjeldahl) cannot differentiate between protein nitrogen and non-protein nitrogen. Reviews have been carried on the melamine adulteration scandal, including how it unfolded and the worldwide consequences generated in its aftermath (Sharma and Paradakar, 2010) and its implications for food safety regulations (Pei et al., 2011). In a more recent case in 2012, around 100 Chinese citizens were arrested for trying to sell milk contaminated with melamine and cyanuric acid. Cyanuric acid is used to clean swimming pools and added illegally to milk to increase its protein content which subsequently causes renal failures. Chinese authorities reported melamine concentration in adulterated milk 500 fold higher than the permissible limit. On a similar note, a milk packaging factory was sealed by the Pakistan officials in Multan district in 2012 as the milk was found adulterated with a detergent named sinopol and glycerol monocarpylate to increase its volume. These chemicals were found to be harmful for human consumption particularly for children and pregnant women (The Express Tribune, Pakistan 2012).

In National survey on milk adulteration conducted by Food Safety and Standards Authority of India (FSSAI) in 2011 in 33 states, about 68% of the total 1791 samples did not conform to the quality standards. About 46% of these non-compliant samples exhibited low SNF (solid-not-fat) content due to the dilution with water, the most common adulterant of milk. Viscosity, thickness and SNF of diluted milk are adjusted by adding adulterants such as salt,

detergents, glucose, starch etc. The survey also checked the presence of various neutralizers, hydrogen peroxide, sugar, urea, formalin, vegetable salts and skimmed milk powder. Nearly 548 samples contained skim milk powder whereas glucose was present in 477 samples. The Food and Drug Administration (FDA) officials and local police busted a milk adulteration racket in August 2012 at a Government chilling plant in Eastern Maharashtra known to receive about 5000 litres of milk daily. It was found that 2000-3000 litres of water, nonpermissible malto-dextrin powder, syrup liquid and other chemicals were added daily in pure milk (CNBC, 2012). Lack of hygiene, sanitation and improper cleaning in milk handling and packaging industries was also found to be one of the reasons of presence of detergents in milk. These detergents cause food poisoning, gastrointestinal disorders and their high alkaline levels damage body tissue.

Urea is a natural component of milk that usually comes from the grass or feed given to dairy cattle. Normal permissible limit for urea is 10-16mg/dl (Abdallah et al., 2008) and if present in more than this concentration, is presumed to be added externally. Adulteration of milk by urea is common to increase its shelf life and SNF. Although it gives a rich white look to milk, its addition renders it poisonous for the consumer due to severe health problems caused by its consumpsion such as indigestion, acidity, ulcers, etc (Trivedi et al., 2009) Addition of urea may also lead to vomiting, nausea, gastritis and may ultimately lead to kidney failure. Formalin which is used as a milk preservative can cause liver damage whereas caustic soda harms the mucosa of the food pipe. The sodium containing chemicals are harmful for the people suffering from hypertension and heart ailments. Due to compositional similarity and price difference, adulteration of butter with margarine is another health threat for consumers in developing countries.

Milk Contamination

Milk is considered an excellent medium for microbial growth, thus high standards of hygiene are required in its production and distribution chain. Though most of pathogens, viruses and bacteria are killed by pasteurization, there may be microbial contamination during processing. Major foodborne outbreaks associated with consumption of milk due to pathogens include *Listeria monocytogenes, Escherichia coli* 0157:H7, *Salmonella*, *Campylobacter*, *Staphylococcus aureus*, etc. (Wells et al., 1991; Bleem, 1994; Bean et al., 1996; Ryser, 1998; Coia et al., 2001; Chyea et al., 2004). Presence of these pathogenic bacteria in milk may prove fatal especially for those individuals who still drink raw milk. Therefore, microbial quality of raw milk is considered decisive for milk processing. Different regulating authorities have defined the limits for microbial counts for accepting raw milk samples in a dairy plant (Table 1). Aflatoxins, group of toxic and carcinogenic secondary metabolites produced by *Aspergillus flavus* and *Aspergillus parasiticus* (Wood, 1992; Rustom, 1997), pose another major health and economic problems worldwide, as evidenced by their frequent detection in milk and other agricultural commodities (Li et al., 2001; Creppy, 2002; Hussain et al., 2008; Wu et al., 2009; Wild and Gong, 2010; Hussain et al., 2010; Iqbal et al., 2011). These toxins account for losses of millions of dollars annually in human health, animal health, and condemned agricultural products, disposal of contaminated foods and feeds, and investment in research and applications to reduce severity of the mycotoxin problem worldwide (Shane, 1994; Vasanthi and Bhat, 1998). Aflatoxin B1 (AFB_1), the most commonly found toxin, has been classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC) (IARC, 2002). In a study conducted in and around Ludhiana city in India, 21 of 28 dairy feed samples from farms

were found to be contaminated with AFB1 at levels ranging from 50 to 400μg/kg (Dhand et al., 1998). AFB1 is metabolized to aflatoxin M1 in mammals on consumption of AFB1-contaminated feed which is then secreted in their milk (Chopra et al., 1999; Ruangwises and Ruangwises, 2010). AFM1 is as acutely hepatotoxic as AFB1 but its carcinogenicity is approximately 2-10% of AFB1 (Carnaghan et al., 1963; Creppy, 2002; Asi et al., 2012). It was initially classified by IARC as an agent in Group 2B, with a possible carcinogenic effect on humans (IARC, 1993) but later reclassified as Group 1 carcinogenic agent due to its high toxicity (IARC, 2002). Moreover AFM1 is a very stable aflatoxin as it is not destroyed by storage and processing, such as pasteurization, autoclaving or other methods used in production of fluid milk (Tajkarimi et al., 2008). Since consumption of milk and milk products is quiet high particularly among infants and young children, the risk of exposure to AFM1 is of great concern (Baskaya et al., 2006). Researchers have shown the hazardous effects of AFM1 through dairy products (Galvano et al., 1996) using diverse methods (Kim et al., 2000; Pathirana et al., 2010) and many countries have therefore prescribed their regulatory limits to avoid excessive exposure to humans and animals (Table1).

Fruit juice Adulteration

Adulteration of fruit juices and their products is also widespread. The juices are primarily adulterated adding water, sugar, cheap juices, corn syrup, cheap solid materials etc. Majority of solid ingredients are carbohydrates, namely malic acid which is generally added to low acidity apple juice concentrate to produce more valuable high acid juice. Presently most packaged commercial fruit juices are manufactured from fruit concentrates. To prepare a juice concentrate, water is removed from the fruit juice and shipped to various locations, where the concentrate is

diluted with local water in a processing unit to a concentration approximating the original juice and is distributed for consumption. In some cases producers add sugar and water to juices to increase the volume and control the sweetness, without labeling them on packets. As sugar and water are cheap ingredients there addition often leads to a considerable profit to the trader. Several colorants and cover up compounds are added to cover the adulteration practice. These reconstituted juices, although are not much harmful for the health, but they lack the wholesome freshness and nutrition of the freshly squeezed juices thus compromising with consumer's money and trust.

Fruit juice contamination

Food borne illnesses have been reported due to consumption of contaminated fruit juices mainly affecting the gastrointestinal tract (Mosupye and Holy, 2000; Lewis et al., 2006; Chumber et al., 2007; Ghosh et al., 2007; Tambekar et al., 2009). Major bacterial pathogens reported to be associated with fruit juices are *E. coli* 0157:H7, species of *Salmonella* and *Shigella* and *S. aureus* (Buchmann et al., 1999; Sandeep et al., 2001; Barro et al., 2006; Lewis et al., 2006). Presence of mycotoxins, the toxic secondary metabolites produced by molds is another major concern in quality control of fruit juices. Recently, occurrences of patulin in apple juices have drawn attention of food technologists and processors throughout the world. Patulin causes serious health problems (De Champdoré et al., 2007; Gashlan, 2008; Saxena et al., 2009) because it is a toxic secondary metabolite produced by a wide range of fungal species of *Penicillium* and *Aspergillus* genera, principally by *Penicillium expansum*, which is a common contaminant of damaged fruit such as apples (Shephard and Leggott, 2000). Since, patulin is considered as a major worldwide problem, international recommendations and regulations have

been made for its maximum permissible levels (Table 1) in apple and apple products (Weigert et al., 1997; Trucksess and Tang, 1999; Pienontese et al., 2005; González-Osnaya, 2007).

These adulterants/ contaminants in milk and fruit juices pose enormous risk to human health. It is further multiplied if the consumers belong to vulnerable / immunocompromised group. It may take a form of epidemic. To avoid such situation, it is crucial to detect them at the early stage prior to their circulation in the market to guarantee both authenticity and compliance with the product specification and consumers safety. Various new and increasingly sophisticated techniques have been experimented and developed by researchers all over the world due to increased consumer awareness of food safety and quality issues (Jha and Matsuoka, 2000,2004a; Reid et al., 2006). Adulteration and misrepresentation are often difficult to detect because of sophisticated methods used by unscrupulous food processors. Analytical techniques that have been applied for food authentication includes chromatography (gas and liquid), chromatography combined with mass spectroscopy (MS), isotopic ratio mass spectroscopy, nuclear magnetic resonance (NMR), ultraviolet–visible light spectroscopy (UV/VIS), electronic nose, polymerase chain detection and enzyme-linked immunosorbent assay (ELISA) (Reid et al., 2006). These methods although sensitive are time consuming, expensive, require extensive sample preparation, some require hazardous chemicals as well as skilled and experienced staff and are mostly available only in large analytical laboratories (Ulberth and Roubicek, 1995; Heussen et al., 2007). Spectroscopy including near-infrared (NIR), mid-infrared (MIR) and Raman is rapidly becoming an important tool for rapid, simple, sensitive, and high-throughput analysis of food components (Jha et al., 2010; 2012). Advances in instrumentation and chemometric pattern

recognition techniques have made it possible to extract information related to the composition of food components from complex spectra (Narsaiah and Jha, 2012; Jaiswal et al., 2012a, b).

DETECTION METHODS

Detection of adulterants and contaminants in food samples has been a matter of concern for food technologists, regulatory authorities and food industries/processors due to increase in incidences of food borne diseases. In current scenario when the processed products are gaining popularity, we need rapid and simple detection techniques for timely detection of causative agents to save lives.

Physicochemical methods

Chemical methods have been successfully used to detect adulteration and contamination in milk and juices (Bector et al., 1998; Paradkar et al., 2000a) both qualitatively and quantitatively. It is based on the chemical reaction between adulterants/contaminants and a chemical reagent which is measured colorimetrically or a change in physicochemical parameters like degree brix to detect addition of water in fruit juices in food matrix. These methods have been used to detect the adulterants (urea, sodium chloride, sodium bicarbonate, sucrose, vegetable oil, detergent) in milk samples (Bector et al., 1998, Paradkar et al., 2000a; Abdallah et al., 2008). The physicochemical methods have been reported to detect as low as 2% urea in natural milk sample. The presence of detergents, which is another major adulterant in milk, has been detected colorimetrically (Paradkar et al., 2000b) while other adulterants such as hydrogen peroxide, carbonate and bicarbonate and flour have been checked qualitatively using various chemical methods (Ayub et al., 2007). Chemical methods have also been used to determine natural components such as fat (Pearson, 1976), protein (AOAC, 2000), total solids, water and

ash (AOAC, 2000). A summary of physicochemical methods used for detection of adulterants and contaminants in milk and juices is presented in Table 2.

Detection of food borne microorganisms, using conventional microbial identification techniques, is based on culturing the microbes from food samples followed by a series of biochemical, physiological and serological tests. These methods target specific biochemical properties of microbes. One of the key factors governing biochemical reactions that occur both outside and inside the cells is presence of enzymes which helps in identification and classification of microbes. Selective growth media for isolation of a particular microbe have been constituted based on biochemical properties. The selective agent may be a protein source, often a hydrolysate of casein, and a fermentable sugar, like lactose or glucose, occasionally an indicator, like neutral red or bromo cresol purple. Several specific media such as Tryptic soy agar, Sorbitol MacConkey Agar, Rainbow Agar, Biosynth Culture Medium; Biosynth, Staad and Fluorocult (Manafi and Kremsmair, 2001), have been used for selective isolation of common human pathogen *Escherchia coli* O157:H7, causing diarrhea, haemorrhagic colitis and haemolytic uraemic syndrome (HUS). Even strain differentiation could be achieved using differential selective media such as Fraser enrichment broth (FB), UVM modified enrichment broth (UVM), morpholinepropanesulfonic acid buffered *Listeria* enrichment broth (MOPS-BLEB), selective enrichment broth (SEB) and Palcam broth (PB) (Liamkaew et al., 2012) for *Listeria* in suspended cell culture. Recently scientists from National Dairy Research Institute (NDRI), Karnal, India have developed a micro-technique for detection of *Listeria monocytogenes* in milk within 24hours. It is based on the principle of targeting specific marker enzyme (s) of target bacteria, leading to release of free chromogen following “enzyme-substrate reaction” that is

visually detected by colour change after initial enrichment of the bacterium in selective medium. A semi quantitative method for detection of aflatoxin M1 in milk has also been developed using spore immobilized ampoules and chromogenic substrate (Kumar et al., 2012). It is reported to detect the presence of AFM1 higher than the permissible limit set by CODEX. A spore based biosensor was also developed for detection of *Enterococc*i, in which *Enterococci* specific marker enzyme acts on germinogenic substance of the spores coated on a gold chip to release germinant which further lead to germination mediated concomitant enzyme activity, the product of which could be detected by fluorescence spectroscopy.

These methods are promising, inexpensive and give both qualitative and quantitative information of the tested components. Their drawbacks include laborious sample preparation, trained man power and many of them use polluting chemical reagents.

Chromatographic- techniques

Mass Spectrometry(MS)

Chromatography due to its high separation efficiency tool has become an important separation technology in the food compositional analysis, especially for trace contents such as pesticide residues, food additives, etc. Chromatograph makes separation based on the difference of distribution coefficient and adsorption ability of material in two phases. This technique includes thin layer chromatography, liquid chromatography (LC) using different separation methods (reverse phase, ion exclusion, ion chromatography) and detectors (refractive index, UV absorption, amperometric) and gas chromatography. Gas chromatography (GC) takes gas as mobile phase while liquid chromatography takes liquid as mobile phase. Thus the separation velocity of GC is higher than that of LC (except high performance liquid chromatography

(HPLC). GC has higher sensitivity than LC, whereas application domain of later is wider than former which could only test 20% of organics with low boiling point.

Thin layer chromatography (TLC) is a technique used to separate mixtures based on the competition of the solute and the mobile phase for binding places on the stationary phase. TLC plate known as the stationary plate is a sheet of glass, plastic, or aluminium foil coated with thin layer adsorbent material usually silica gel, aluminium oxide, or cellulose (blotter paper). After application of sample on the plate, a solvent or solvent mixture (known as the mobile phase) is drawn up the plate via capillary action. Simplicity and speed of this technique has made it useful in monitoring various chemical reactions, microbial assays and detection of mycotoxins (Abramson et al., 1989; Paul et al., 1997; Kamkar, 2005; Tchana et al., 2010). TLC is often used as a reference method for screening assay of mycotoxins. Densitometry (Leming et al., 1993) and fluorodensitometry following the formation of fluorescent derivatives (Durakovic et al., 1993) have facilitated its use in quantification of spot intensities of mycotoxins. Nowdays, a number of enhancements to automate the different steps and increase the resolution achieved for more accurate quantization of desired compounds have been made to the original TLC method. Coupled with effect directed analysis (EDA), TLC is an important tool in food, pharmaceutical and phytochemical analyses. In EDA, there is no need to analyze all components in a sample, but only the relevant (for example, harmful or bioactive) compounds are detected using a bioassay coupled to TLC. These compounds act as the target analyte as well as unknown metabolites, side products, process contaminants, adulteration products and residues. Bioassays used with TLC include microbiological assays (bioautography), bacterial assays (the Camag BioLuminizer

bioluminescence detector utilizing *Vibrio fischeri*) and biochemical detections using enzymes, including immunoassays (Morlock and Schwack, 2010).

The compounds which are vaporized without decomposition are separated and analyzed using GC based on the comparison of their retention time. In this chromatography technique, the mobile phase is an inert and non-reactive carrier gas and the stationary phase is a microscopic layer of liquid or polymer on an inert solid support, inside a piece of glass or metal tubing called a column. The gaseous compounds being analyzed interact with the walls of the column, which is coated with a stationary phase. This causes each compound to elute at a different time, known as the retention time of the compound. GC in combination with chemometric analysis has successfully been applied to authenticate several fruit juices (Del Castillo et al., 2003a; 2003b; Reid et al., 2004) and to determine triacylglycerol profiles in milk and non-milk fat. The linear discriminant analysis allowed the correct classification of 94.4% of the samples (raw milk fat) with levels <10% of adulteration (pork lard, bovine tallow, fish oil, peanut, corn, olive and soy) (Gutiérrez et al., 2009). GC has also been used for authenticating different dairy products by analyzing quantity and quality of flavour constituents (Ulberth and Roubicek, 1995; Viallon et al., 2000; Sag˘dıc et al., 2004) besides detecting mycotoxins. Several GC based methods have been developed for the determination of mycotoxins (Tarter and Scott, 1991; Sheu and Syhu, 1995). Researchers have utilized derivatives of mycotoxins to ensure adequate chromatographic separation and detection. Llovera et al., (1999), reported detection of as low as 4mg/L of patulin in apple juice without any derivatisation based on MS detection of electronic pressure controlled on-column injection of samples. The Microbial Identification System (MIS) is another example of fully automated GC analytical system used to identify bacteria and fungi based on their unique

fatty acid profiles (Lambert et al., 1986). Advantages of GC are short separation time, high resolution and high sensitivity. Development of many types of sensitive detectors has enhanced the sensitivity of gas chromatography than most high performance liquid chromatography (HPLC) detectors. Disadvantages of GC in analysis of complex food samples are extensive cleaning and fractionation to remove the non-volatile substances or other disrupting chemicals in food to prepare a relatively simple mixture, so that it can be entered into the analysis system without too much damage to the chromatography column and detector. In addition, GC methods can only solve about 20% organic analysis as it needs esterification and other pretreatments to reduce the boiling point. Improper handling at any step would results in the loss or degradation of the samples.

High performance liquid chromatography (HPLC) has been used for compounds not volatilized readily. It is considered an instrumental technique of analytical chemistry instead of a gravitimetric one because solvent is forced under high pressure (up to 4.05×10^7 pascals) for getting high speed leading to much better separation of the components of a mixture. Smaller particle size for the column packing material gives much greater surface area for interactions between the stationary phase and the molecules flowing past it. It is the most frequently and widely used analytical method with high sensitivity and reasonably low detection limit (Trucksess, 1998). The sample preparation for HPLC analyses however is very crucial for efficient detection of target analyte. Any impurity leads to broader peaks and/or multiple peaks thereby leading to false positives or mis-identification. Researchers across the globe have continuously modified the steps involved in HPLC analysis to achieve high resolution and specificity (Mo¨ller and Josefsson, 1980; Forbito and Babsky, 1985; Brause et al., 1996).

Depending on the physical and chemical attributes of the analyte of interest different detectors have been used for detection of different adulterants/contaminants (Stroka et al., 2000; Scudamore., 2005). HPLC with a fluorescent detector is one of the most commonly used method for monitoring AFB1 in foods and feed (Izquierdo et al., 2005; Lattanzio et al., 2007; Khayoon et al., 2010). Patulin in apple juice has been detected and quantified by the same technique within a concentration range from 0.057 to 0.104g/ml. (Gashlan, 2009). It has also been successfully used to determine authenticity in fruit juices by oligosaccharide profiling (Low, 1996; Low et al., 1999; Hammond, 2001; Pan et al., 2002; Castillo et al., 2003). Organic acids in fruits i.e. citric acid, malic acid and ascorbic acid and sugars such as sucrose, glucose and fructose are easily identified by HPLC (Kelebek et al., 2009). Anthocyanins which are a natural component of fruit juices have been quantified using this technique to check the adulteration of berry juices by other cheap juices (Stoj et al., 2006).

HPLC technique has been successfully applied in milk authentication and detection of different adulterants. Analysis of the whey protein *b*-lactoglobulin has enabled detection of the adulteration of ovine and caprine cheese with bovine milk (Ferreira and Cacote, 2003) at levels as low as 2% v/v and of caprine milk with bovine milk (Chen et al., 2004). Several advancements in chromatographic techniques in recent years have increased their ease of application to industrial food authentication. Venkatasami and Sowa (2010) developed acetonitrile-free, reverse phase high performance liquid chromatography (HPLC) method for determination of melamine in dry and liquid infant formula. The limit of detection (LOD) and limit of quantification (LOQ) values were $0.1\mu gmL^{-1}$ and $0.2\mu gmL^{-1}$, respectively. Sun et al. (2010) also developed a sensitive and validated method for determination of melamine residue in

liquid milk based on reversed phase high performance liquid chromatography-diode array detection (RP-HPLC-DAD) with solid phase extraction (SPE). The method could permit the detection of melamine residues at levels as low as 60µg/kg in different liquid milks. Several HPLC based analytical methods have been designated as the AOAC official methods (AOAC, 2000). One of the major drawbacks of this technique is that the identity of interfering compound is often unknown, and in such cases liquid chromatography-mass spectrometry is more useful to avoid reporting false positive results. The application of liquid chromatography is greatly broadened with continuous development in workstation software and collaboration of mass spectrometry.

Organic mass spectrometry is generally used in tandem with gas chromatography or liquid chromatography for food testing. In this method, the samples are first ionized and then separated according to their mass charge ratio (m/z) when they pass through the electric and magnetic field. Mass spectrometry has the advantage of high sensitivity, less sample consumption and rapid analysis. The combination of GC/LC with mass spectrometry improves selectivity and allow quantitative determination and confirmation of adulterants in milk and milk products (She et al., 2008; Miao et al., 2009; Lutter et al., 2011). Gas chromatography/isotope dilution mass spectrometry (GC/IDMS) has been reported to be highly accurate in spite of the challenges associated with complexity of the sample matrix. Urea concentration in milk and milk powder was measured using this technique (Dai et al., 2010). Use of time-of-flight mass spectrometry coupled with GC (GC-TOFMS) has further reduced the time of analysis. This technique however, has found limited applications to date in the area of food authenticity, principally due to the cost of acquiring and running a GC-MS system. Vaclavik et

al. (2010) used fast semi-automated method employing direct analysis in real time ion source coupled to TOFMS for analysis of melamine and cyanuric acid in milk powder and milk based products. The sample extraction procedure in this method did not involve any prior chromatographic separation and achieved limits of detection 170 and 450μgkg^{-1} for melamine and cyanuric acid, respectively. Binary milk mixtures were also analyzed directly using Matrix-assisted laser desorption/ionization time of flight mass spectrometry (MALDI-TOFMS) with chemometrics. The basis for differentiation using spectra was different ratio of protein structures (β-lactoglobulin and α-lactalbumin) in different types of milk. Despite the milk processing, which may affect the protein structure, MALDI-ToF-MS was able to achieve high accuracy levels of adulteration in cow milk (Nicolaou et al., 2011).

Pyrolysis mass spectrometry (PyMS) in combination with mass spectrometry has been evaluated to detect the fat content and adulteration in mixture of milk of binary species such as of cow's and ewe's, cow's and goat's (Goodacre, 1997). This technique has also been applied in the quantitative assessment of the adulteration of orange juice with sucrose (Goodacre, 1997). Although the combination of mass spectrometry with other techniques increased detection sensitivity and accuracy, however use of this application is limited mainly due to expensive equipment, complicated analysis of the results and infeasibility for onsite detection owing to their long incubation time and excessive washing steps.

Immunoassays

Immunoassays quantify a specific substance at very low concentrations using an immunological reaction. Antigens associated with organisms difficult to culture such as hepatitis B virus and Chlamydia trichomatis have been detected using this technique.

Radioimmunoassay, Enzyme immunoassay, immunoprecipetin, fluorescence immunoassays are some of common examples. The majority of reported studies on immunological techniques for food authentication involve the use of Enzyme linked immunosorbent assay (ELISA). This technique involves cultivation of antibodies or antisera that are capable of binding to a protein of interest, thereby enabling the detection of that protein, both qualitatively and quantitatively. Enzyme-linked immunosorbent assay technique has been widely used to detect the occurrence of Aflatoxin B1 in the animal feed (Lee et al., 2004; Kolosova et al., 2006; Tudorache and Bala, 2008) and Aflatoxin M1 (AFM1) in infant milk products, liquid milk and UHT milk samples (Rastogi et al., 2004; Kang'ethe and Lang'a, 2009; Gundinic and Filazi, 2009; Gomaa and Deeb, 2010; Mohammadian et al., 2010; Pathirana et al., 2010). ELISA was compared with HPLC to detect the minimum level of occurrence of aflatoxin M1 (AFM1) in pasteurized milk and dairy products. The limits of detection of AFM1using ELISA and HPLC were found to be 2 pg/ml and 10 pg/ml, respectively (Kim et al., 2000). Commercial ELISA kits for detection of AFB1 are also available; for example, Romer labs (MO, USA) and R-Biopharm Inc. (Germany) have commercialized ELISA kits with a detection limit of 1.0 - 40ng/g and 0 -50ng/g, respectively (Huybrechts, 2011). Availability of immunochemical methods has led to development of many rapid and sensitive methods for monitoring and quantifying AFB1 in contaminated foods and feeds (Lee et al., 2004; Kolosova et al., 2006; Tudorache and Bala, 2008). Recently Liu et al. (2013) reported a highly sensitive and rapid competitive direct enzyme linked immunosorbent assay (cd ELISA) and a gold nanoparticle immunochromatographic strip methods for screening of AFB1 in food and feed samples. The strip has been reported to detect as low as 2.0ng/ml of AFB1 in food and feed samples.

ELISA-based techniques have been reported to detect the presence of vegetable proteins in milk powder (Sanchez et al., 2002); differentiate milk from different species (Bania et al., 2001; Moatsou and Anifantakis, 2003; Hurley et al., 2004). Yin et al. (2010) developed an indirect competitive ELISA (icELISA) method based on preparation of monoclonal antibodies (MAbs) to detect melamine in raw milk, milk powder and animal feeds. The detection limits of melamine in milk, milk powder and feeds were 0.1, 0.2 and 0.5mg L^{-1}, respectively. The specificity of technique for detection of melamine in milk was further enhanced (Lei et al., 2011) using three haptens of melamine with different spacer arms prepared from different plate coating antigens. The limit of detection was 8.9ng mL^{-1}. The developed icELISA showed a much lower cross-reactivity to cyromazine, a fly-killing insecticide widely used in vegetables compared to the ELISA results previously reported. Recoveries obtained from milk samples in this study were in agreement with those obtained using the HPLC-MS method, indicating the detection performance of icELISA could meet the requirement of residue limit set by the Codex Alimentarius Commission. This technique holds much potential for authentication of food products but till date limited advances have been made in extending its authentication capabilities. The disadvantages of the ELISA approach include initial difficulty in producing an antibody specific to a particular protein. However, this is a relatively minor difficulty to overcome when the selectivity of the technique is taken into account. Similar to chromatography and mass spectrometry, ELISA approaches are also infeasible for on-site detection owing to the long incubation time, excessive washing steps, and complexity in applying instrumentation.

Wang et al. (2011) developed a fluorescence polarization immunoassay (FPIA) based on a polyclonal antibody for determination of melamine in milk. Three fluorescein-labeled melamine tracers with different structures and spacer bridges were synthesized. The limit of detection (10% inhibition) of FPIA was 9.3ng mL^{-1} and the IC 50 (50% inhibition) value was 164.7ng mL^{-1}. The antibody in the FPIA showed 21.2% cross-reactivity to the fly-killing insecticide cyromazine, but had no cross-reactivity to other natural structurally related compounds. The results were confirmed by high performance liquid chromatography-mass spectrometry.

Presence of whey in processed and raw milk has also been detected using Immunochromatographic assay of glycomacropeptide (GMP) also known as caseinomacropeptide (CMP) (Oancea, 2009). GMP is a bioactive 64 amino acids residue glycopeptide released enzymatically in whey from k-casein by the action of chymosin during cheese making (Eigel et al., 1984). Addition of whey in milk due to its low cost though does not cause any health hazard, supplementation of infant formula with GMP enhance the absorption of trace minerals leading to excess dietary intake (Kelleher et al., 2003).

Molecular methods

Molecular methods have successfully been used as a tool for enhancing sensitivity and specificity of authenticity testing and microbial contaminant identification. Polymerase Chain Reaction (PCR), an invitro technique based on the principle of DNA polymerization reaction by which a particular DNA sequence is amplified several folds, has brought revolutionary change in detection, identification and characterization of microbial contaminants (Settanni et al., 2005). PCR-based technologies such as PCR-RFLP (Restriction fragment length polymorphism) and

randomly amplified polymorphic DNA (RAPD) have successfully been used for differentiation and identification of microbial isolates **(**Guo et al., 2008; Shao et al., 2008). Another variant of PCR is Real Time-PCR which enables detection and quantification simultaneously. It has been successfully applied for the specific detection and quantification of goat's milk adulteration with cows' milk in the range of 0.5–100% using standard curve with R^2 value more than 0.99 (Dąbrowska et al., 2010). Multiplex PCR (MPCR) further offers added advantage of simultaneous detection of multiple microorganisms by simultaneous amplification of more than one locus in a single reaction (Chamberlain et al., 1988). Here several specific primer sets are combined into a single PCR assay. MPCR is one of the fastest culture-independent approaches for strain specific detection in complex matrices. Like the majority of molecular techniques, MPCR commonly targets the 16S rRNA gene, the gene most widely used to infer phylogenetic relationships among bacteria (Rosselló-Mora and Amann, 2001). This technique is widely applied in the various fields of microbiology for rapid differentiation of pathogenic and non-pathogenic microorganisms.

PCR based techniques have been successfully reported to detect pathogenic microorganisms such as *E. coli* (Osek and Gallien, 2002), *Clostridium perfringens* (Baums et al., 2004), *Salmonella enterica* (Lim et al., 2003), *Staphylococcus aureus* (Kwon et al., 2004) and multiple species of single genera like *Campylobacter* spp. (Harmon et al., 1997; Houng et al., 2001; Cloak and Fratamico, 2002; Wang et al., 2002; Klena et al., 2004), *Listeria* spp. (Graves et al., 1991; Brosch et al.,1996) without compromising accuracy. Similarly non-pathogenic microorganisms such as starter cultures used in food fermentations (Aquilanti et al.,

2006; Valmorri et al., 2006) and probiotic strains (Yost and Nattress, 2000; Ventura et al., 2001; Mullié et al., 2003) could be differentiated using MPCR.

The major drawback of PCR technology is its inability to distinguish between dead and live cells and DNA from dead cells can still be amplified after a significant time interval (Lei et al., 2008). Inhibitory compounds such as high amounts of fat and protein in the food samples, further limit the application of PCR technology. Efficient procedures therefore are required for successful elimination of such inhibition for routine use of this technology. Nonetheless, environmental conditions affecting somatic cell numbers in milk, such as animal mastitis or changes from raw to heat-treated milk may adversely affect PCR results, even though a few recent techniques have suggested that PCR is unaffected by these conditions (Lopez-Calleja et al., 2005). At the moment PCR lacks certain practicality for routine commercial use but with further improvement it could become a very good option in future.

Electrical methods

The electrical properties of liquid food products under controlled conditions mainly depend on parameters of the measuring current (voltage, frequency, impulse, shape and type of electric current), experimental conditions particularly temperature and different chemical components in raw material and their degree of dissociation(Zywica et al. 2005; Jha et al 2011).

Milk is characterized by good ionic conductivity owing to its high water and mineral salts content. Milk conductivity is determined using two main parameters viz. impedance the main component of which is resistance and admittance the main constituent of which is conductance. The relationship between milk constituents (key attributes of its quality) and electric properties has been studied and applied for quality evaluation. Zhuang et al. (1997) showed a statistically

significant ($P < 0.05$) correlation between protein content of a commercial whey powder determined by means of electric conductance and Kjeldahl's method. Investigations into the impact of water, fat and protein contents on the specific thermal conductivity of different dairy products (cheeses, yoghurts, butter and margarine) demonstrated that the values of thermal conductivity increased with increase in water content and decreased along with increasing fat and protein content (Tavman and Tavman, 1999; Żywica et al., 2012). Mabrook and Petty (2002) used the method of electrical admittance spectroscopy to study the water and fat content of milk. Over the frequency range 5 Hz to 1 MHz, the electrical circuit was dominated by a single time constant. To eliminate the effect of electrode polarization, the conductance of milk was measured at high frequencies where it showed a saturation value. The characteristics at 100 kHz and 8°C for all milk samples revealed a linear decrease in conductance with increasing water content. Admittance data for full fat, semi-skimmed and skimmed milk showed an increase in milk conductance with decreasing fat content. Linear correlation between conductance of milk and its water content has been applied in detection of adulterations through water addition. The characteristics at 100 kHz and 8°C for all skimmed milk samples revealed a linear decrease in conductance with increasing water content over the entire range of water concentrations. In contrast, the conductance of full fat milk showed a decrease only at added water concentrations higher than 10%. At lower added water concentrations, the full fat milk exhibited an anomalous conductivity maximum at 2–3% added water (Mabrook and Petty, 2003a). Mabrook and Petty (2003a) developed a novel method to detect added water to full fat milk using single frequency electrical conductance measurements. Electrical resistance and conductivity has also been

studied as a means to detect freshness and adulteration of eggplant fruits (Jha and Matsuoka, 2004b) and milk (Mabrook and Petty, 2003b), respectively.

Conductance of milk is chiefly due to presence of ions (Na^+, K^+, Cl^-). Since the salt content of pure milk remains constant, it exhibits a constant conductance under controlled conditions of temperature and measurement parameters. Any addition of adulterant leads to change in salt concentration which results in variation in conductance of milk samples with frequencies ranging from 20Hz to 1MHz. Based on this, Sadat et al (2006) detected adulteration of pure milk by detergents and synthetic milk using alternating current (AC) conductance measurement from 20 Hz to 1 MHz. Since the natural milk samples have been found to exhibit higher conductance at 100 kHz than synthetic milk, the addition of synthetic milk resulted in decrease in conductance with respect to the added concentration to natural milk. The conductance has been found to decrease at the rate of 0.13mS with 10% increase in the percentage of synthetic milk at 8^oC. Conductance of milk samples has been found to decrease with increase in temperature.

Dielectric properties, or permittivities, are intrinsic properties that determine the interaction of electromagnetic energy with milk when subjected to dielectric heating. The dielectric properties of milk had also been successfully used for quality evaluation of milk. Open-ended coaxial-line probe method associated with network analyzers or impedance analyzers is a useful technique to determine dielectric properties, i.e. dielectric constant, dielectric loss factor and loss tangent of materials, especially liquid foods, according to the reflection coefficient at the material-probe interface. Guo et al. (2010) proposed the use of loss factor as an indicator in predicting milk concentration and freshness. The dielectric properties of

water-diluted cow's milk with milk concentrations from 70% to 100% stored during 36h storage at 22°C and 144h at 5°C were measured at room temperature for frequencies ranging from 10 to 4500 MHz using open-ended coaxial-line probe technology, along with electrical conductivity and pH value. The raw milk had the lowest dielectric constant when the frequency was higher than about 20MHz, and had the highest loss factor at each frequency. The highest linear coefficient of determination, 0.995, between the milk concentration and the loss factor at 915MHz was observed. The change tendency of the loss factor was inversed to pH during milk storage with the best linear correlation (R^2 = 0.983) at 1100MHz. Das et al. (2011) successfully differentiated using impedance sensor synthetic milk and original milk when the synthetic milk is reconstructed by adding minimum 15% of adulteration. The measurement is based on the change in constant value of phase angle of the impedance sensor due to the change of ionic property of adulterated milk. It was observed that the sensor can identify 5% of liquid-whey adulteration, 10% tap-water adulteration, and 0.6 mg of urea per ml of milk.

Change in the electrical properties of the medium due to the breakdown of complex molecules into charged catabolic products (Ur and Brown, 1975) has enabled electrical methods' wide application in dairy microbiology. Commercially available instruments like the Bactometer (Gnan and Luedecke, 1982; Suhren and Heeschen, 1987) and the Malthus system (Fung, 1994) measures the concentration of microorganisms growing in a medium by measuring the electrical properties of the medium. These instruments have been used for detecting specific microorganisms in milk (Easter and Gibson, 1989; Kowalik and Ziajka, 2005), milk powder (Neaves et al., 1988) and yoghurt (Pirovano et al., 1995), for monitoring the quality of raw milk

(Senyk et al., 1988), estimation of microorganisms in raw and pasteurized milk and cheese (Vasavada, 1993), and for predicting the shelf-life of milk (Visser and De-Groote, 1984; White, 1993). Requirement of a large bacterial population (1×10^6–1×10^7 cfu/ml) for efficient detection is however the major drawback of this technology (Vasavada, 1993, Adams and Moss, 2000).

Vibrational spectroscopy techniques

Vibrational spectroscopy is used to study a wide range of samples and is carried out from a simple identification test to an in-depth, full spectrum, qualitative and quantitative analysis. Samples may be examined either in bulk or in microscopic amounts over a wide range of temperatures and physical states (e.g., gases, liquids, latexes, powders, films, fibers or as a surface or embedded layer). The important vibrational spectroscopy techniques are mid-infrared (IR), near-IR and Raman spectroscopy. Near-IR spectroscopy measures the broad overtone and combination bands of some of the fundamental vibrations (Jha, 2010). Both mid-IR and Raman spectroscopy provide characteristic fundamental vibrations that are employed for the elucidation of molecule. These techniques have various advantages and disadvantages with respect to instrumentation, sample handling and applications.

Raman and mid-IR spectroscopy are complementary techniques and usually both are required to completely measure the vibrational modes of a molecule. Although some vibrations may be active in both Raman and IR, these two forms of spectroscopy arise from different processes and different selection rules. In general, Raman spectroscopy is best at symmetric vibrations of non-polar groups while IR spectroscopy is best at the asymmetric vibrations of polar groups. Their bands are characterized by their frequency (energy), intensity (polar

character or polarizability) and band shape (environment of bonds). Since the vibrational energy levels are unique to each molecule, the IR and Raman spectrum provide a "fingerprint" of a particular molecule. The frequencies of these molecular vibrations depend on the masses of atoms, their geometric arrangement, and the strength of their chemical bonds. The spectra provide information on molecular structure, dynamics and environment. Infrared spectroscopy deals with the infrared region of the electromagnetic spectrum that is energy with a longer wavelength and lower frequency than visible range.

Near infrared spectroscopy

Near infrared spectroscopy (NIRS) combined with chemometrics has been employed to study different constituents of milk viz. total solids, fats, protein, casein, urea nitrogen, lactose and somatic cells in reflectance mode in the wavenumber range of 4000-10000 (Inon et al., 2004). Apart from being nondestructive in nature, this technique also includes speed and enables simultaneous measurement of a number of constituents (Jha, 2010). Sato et al. (1990) evaluated the role of NIRS for detection of foreign fat adulteration in dairy products using the extracted fats from samples of butter and margarine mixtures and from milk and soymilk mixtures. They observed NIR absorptions specific to *cis*-unsaturation of fatty acid moieties (which is intrinsic to each oil) at 1164, 1660, 2144, and 2176nm. On this basis they suggested that the near infrared spectra around these wavelengths could be used as a simple index for fatty acid profiles in oil. The application of preprocessing techniques could further improve the detection limit (3% foreign fat) for butter and margarine mixtures. NIR spectroscopy has also been employed to detect and quantify presence of water and whey as adulterants in cow milk in a wavelength

region of 1100-2500nm using PLS models after data treatment (Kasemsumran et al., 2007). The same technique has subsequently been used for the detection of different adulterants in cow's milk such as vegetable oil, urea, NaOH and shampoo in the wavelength range of 700-1124.8nm successfully for content ranging from 0-25% (Jha and Matsuoka, 2004). The different adulterants show variations in spectra at different wavelength ranges and could be detected nondestructively at a concentration even less than 10% (Table 3).

Al Qadiri et al. (2007) used short wave NIR spectroscopy to monitor the quality loss of pasteurized skim milk stored at high temperatures. They utilized the change in properties of milk such as proteolysis, formation of metabolic byproducts, lipolysis leading to reduction in pH as an indicator of bacterial count. PLS correlated spectral data featured with bacterial counts and pH and yielded a high correlation coefficient. Differentiation in milk samples based on omega-3 fatty acids using NIR spectroscopy had showed the potential to be used as fast method to discriminate between the milk as well as adulteration of raw cow milk samples with powdered milk at various concentrations (Aulrich and Molkentin, 2009; Hsieh et al., 2011). Least square support vector machine (LS-SVM) has been applied to calibrate the prediction model for adulteration ratio.

NIRS combined with chemometric techniques has also been used to classify the pure bayberry juice and the one adulterated with different concentrations of water. After using multiplicative scatter correction and standard normal variate transformation to preprocess spectra, the results demonstrated that principal component radial basis function neural networks (PC-RBFNN) with optimum parameters can separate pure bayberry juice samples from water-adulterated bayberry, but cannot clearly detect water levels in the adulterated bayberry juice (Li-

juan et al., 2008). Adulterants such as high fructose corn syrup and sugars solution in apple juice have been easily detected using NIR and chemometric analysis (Leon et al., 2005). Glucose, fructose and sucrose in juices have been detected and quantified using NIR spectroscopy after comparing it with the conventional methods such as HPLC. Partial least squares regression (PLSR) was used to construct calibration models on data preprocessed using different techniques. The best models have been obtained on second derivative processed spectra, especially for sucrose (Xie et al., 2009).

Balabin and Smirnov (2011) concluded that near-infrared (near-IR/NIR) and mid-infrared (mid-IR/MIR) spectroscopy were effective, quick, sensitive, robust, and low cost methods to detect melamine in dairy products, such as infant formula, milk powder, or liquid milk. A level of detection below 1ppm (0.76 ± 0.11ppm) was reached with use of correct spectrum preprocessing (pretreatment) technique and multivariate algorithm like partial least squares regression (PLS), polynomial PLS (Poly-PLS), artificial neural network, support vector regression or least squares support vector machine. Mauer et al. (2009) also evaluated near and mid-infrared spectroscopy methods (NIR, FTIR-ATR, FTIR-DRIFT) for the detection and quantification of melamine in infant formula powder. PLS models were established for correlating spectral data to melamine concentration with R^2>0.99, RMSECV≤0.9, and RPD≥12. Factorization analysis of spectra differentiated unadulterated infant formula powder from samples containing 1ppm melamine with no misclassifications. These nondestructive methods require little or no sample preparation. The NIR method had an assay time of 1 min, and a 2 min total time to detection. The FTIR methods required up to 5 min for melamine detection.

Mid-infrared (MIR) spectroscopy

MIR spectroscopy is commonly used as fourier transform infra-red spectroscopy, which works on principle of molecular vibrations, occurring in the spectral range of 2,500-25,000nm. The absorption bands in this range are narrow and their resolution is high. So it is often used for the identification of unknown materials. Spectra are usually collected in reflectance, transmission, or emission mode. He et al. (2010) has evaluated a method using MIR spectroscopy with two-dimensional (2D) correlation to detect melamine, urea, glucose and tetracyclinehydrochloride in milk. Two-dimensional correlation spectra of the samples have high time resolution and provide information about concentration dependent intensity changes which are not readily accessible from one-dimensional spectra. The concentrations of other adulterants such as sodium bicarbonate, sodium citrate and whey have also been determined using the same technique (Cassoli et al., 2011).

The adulteration of fruit juices with a low cost juice is a common problem in juice industry as it is not easily detected by general analytical methods. FTIR spectroscopy coupled with chemometric techniques is however a promising tool to differentiate between different juices in a mixture based on absorbance in different range of spectral wavelength. Thus it has been used in various quality control labs to check the adulteration of fruit juices for example the adulteration of pomegranate juice concentrate with grape juice concentrate (Vardin et al., 2008) and apple juice with low cost juices (Kelly and Downey, 2005). Adulterants such as partially inverted cane syrup, beet sucrose, high fructose corn syrup and synthetic solution of fructose, glucose and sucrose have been successfully detected using FTIR-ATR (Kelly and Downey, 2005). FTNIR and multivariate techniques have been applied for the rapid determination of individual sugars (Luis et al., 2001), pectin and organic acids such as sucrose, glucose, fructose,

sorbitol, citric acid and malic acid (Irudayaraj and Tewari, 2003; Gamal et al., 2011) in fruit juices. The use of MIR and chemometrics to detect adulteration of apple juice with beet syrup and cane syrup gave correct classifications of 100 and 96.2% respectively (Sivakesava et al., 2001). A combination of FT-MIR spectroscopy and chemometric technique has demonstrated to be capable of classifying apple juice beverages based on percent of pure apple juice present (Gomez-Carracedo et al., 2004). Jha and Gunasekaran, (2010) determined added sugar content (ASC), total soluble solids (TSS) and real juice content (RJC) in fresh and commercial mango juice using Fourier transform infrared spectrometer equipped with attenuated total reflection and chemometrics. They showed three clear peaks in the spectra **(**Fig. 1) and clear differences in absorbance values of sucrose solution, unadulterated and adulterated juice samples. The entire spectral data could be classified into four groups using PCA (Fig. 2). They used partial least squares (PLS) discrimination and principal component analysis (PCA) to classify the samples with or without ASC. Best results from the PCA projection method at 5% significance level was found in the range of 1476 to 912cm^{-1} wavenumber after baseline offset correction (Table 4). ASC, TSS and RJC were predicted in the wavenumber range of 1476 - 912cm^{-1} using PLS (R=0.99) and three wavenumbers (1088, 1050, 991 cm^{-1}) using MLR (R=0.98).

Bacterial identification and differentiation by mid infrared spectroscopy (2500-5000nm) has been investigated since middle of the last century, but shortcomings in available instrumentation led to discontinuation of these studies for several decades. FTIR has been reported to discriminate different bacterial species in juices based on unique FTIR vibrational combination bands from mid-IR active components of bacterial cells in the ‘‘fingerprint region’’ at wavenumbers between 1500 and 800cm^{-1} (Al-Holy et al., 2006). The differentiation of

Escherichia coli O157: H7 from *Alicyclobacillus* spp. in apple juice has been done using FT-IR spectroscopy (4000–400cm^{-1}) and multivariate spectral analysis (Al-Qadiri et al., 2006). The second derivative transformation and loadings plot over the range of 1800–900cm^{-1} highlighted the most distinctive variations among bacterial spectra. Baldauf et al. (2007) developed classification models (Soft Independent Modeling of Class Analogy SIMCA), generated from derivatized infrared spectra for successful discrimination among *Salmonella* strains in tomato juice, eggs, milk, and chicken. The major discrimination was associated to stretching modes of O-specific polysaccharide chains of lipopolysaccharides. Koca et al. (2010) demonstrated the feasibility of temperature-controlled attenuated total reflectance-mid-infrared (ATR-MIR) spectroscopy combined with multivariate analysis technique namely partial least square regression (PLSR) as a simple and rapid method for determination of butter adulteration with margarines at ratios of 2.5%, 13% and 45%. PLSR models gave standard error of cross-validation (SECV) of <1.2% (v/v) and correlation coefficients (r) > 0.99.

Recently attenuated total reflectance mid-infrared microspectroscopy (MIR microspectroscopy) was evaluated as a rapid method for detection and quantification of whey, hydrogen peroxide, synthetic urine, urea and synthetic milk adulteration in milk. Pattern recognition analysis by SIMCA showed tight and well-separated clusters allowing discrimination of control samples from adulterated milk. PLSR showed standard error of prediction (SEP) ~ 2.33, 0.06, 0.41, 0.30 and 0.014g/L for estimation of levels of adulteration with whey, synthetic milk, synthetic urine, urea and hydrogen peroxide, respectively (Santos et al., 2013). Fourier-transform infrared microspectroscopic (MIR microspectroscopy) instrumentation has facilitated the examination of small spatial regions thus allowing for sensitive, fast response detection,

high-fidelity and high-throughput measurements of spectral intensities of narrowly defined regions within larger sample areas (Bhargava and Levin, 2003). This technique has also been successfully tested for detecting subtle compositional differences between microorganisms at strain and serovar level (Grasso et al., 2011; Prabhakar et al., 2011).

Raman spectroscopy

Raman spectroscopy is used to study vibrational, rotational, and other low-frequency modes in a system. It relies on inelastic scattering or Raman scattering of monochromatic light, usually from a laser in the visible, near infrared, or near ultraviolet range. Being a scattering technique, specimens do not need to be fixed or sectioned. Raman spectra can be acquired even for a very small volume (<1µm in diameter). These spectra allow the identification of species present in that small volume. Water does not generally interfere with Raman spectral analysis. Thus, Raman spectroscopy is suitable for a vast range of liquid products. This is a huge advantage over FTIR. Another major advantage over FTIR spectroscopy is that this technique allows samples to be profiled through a variety of transparent materials, allowing analysis of food products through plastic or glass packaging. Major limitation is that the Raman effect is typically very weak as only 1 in every 106–108 photons are scattered inelastically. However, there are a number of ways to enhance the scattering involving resonance Raman (RR) spectroscopy, surface enhanced Raman scattering (SERS) and tip enhanced Raman scattering (TERS) which enhances Raman signals significantly (Hermann et al., 2011).

Raman spectroscopy in combination with chemometric analysis has also found application in dairy industries as fast and simple tool for determination of the unsaturation level of milk fat (El-Abassy et al., 2012). The Raman measurements have been performed directly on

liquid milk and on fat extracted from liquid milk. The Raman spectra taken from the extracted fat showed a higher resolution. The spectra directly obtained from milk and juice samples exhibit fluorescence background which limits the use of Raman spectroscopy. The flouresecnce in juice samples occur due to presence of various pigments. It has also been used to quantify the constituents and assess the nutritional parameters of infant formulae and milk powder (Moros et al., 2007) and different amounts of conjugated linoleic acid in cows' milk fat (Meurens et al., 2005, Bernuy et al., 2008). Melamine adulteration in milk powder was detected using a portable compact Raman spectrometric system (Okazaki et al., 2009). Melamine-free milk powder was used as blank control after verification by HPLC tandem mass spectrometry. Melamine fortified in milk powder was identified with good reproducibility by two characteristic vibration modes at 673 and 982cm^{-1}. The intensity of the first mode was used to quantify melamine adulterant in milk powder. A detection limit of 0.13% and a predictive partial least squares (PLS) model were obtained. As a fast-screening scheme, the method could be suitably applied to onsite quality control of dairy products and market surveillance. Furthermore, Raman spectroscopy has been used to characterize milk whey protein components (Liang et al., 2006) and Fourier Transform-Raman spectroscopy has been shown to be able to predict accurately the unsaturated fat content of clarified butter potentially for use in quality control (Beattie et al., 2004).

Nuclear Magnetic Resonance (NMR) spectroscopy

NMR techniques such as ^{1}H NMR, ^{2}H NMR mono or bidimensional (2D-NMR), and SNIF-NMR has complemented the analysis possibilities offered by IR spectroscopy. NMR spectroscopy can provide detailed information about the molecular structure of a food sample as it analyses the energy absorption by atomic nuclei with non-zero spins in the presence of a

magnetic field affected by the nuclei of surrounding molecules, which cause small local modifications to the external magnetic field. The application of NMR in food detection focuses on determination of food components viz. water, lipids, amino acids, carbohydrates. It is also used in analysis of microbial contamination, pesticide residues and non-destructive quality testing of fruit (Kotwaliwale et al, 2010). The advantage of NMR is that it is nondestructive, independent of the sample size and shape and has high accuracy and good repeatability. High-resolution NMR (HR-NMR) (frequencies above 100 MHz) has been applied more in food authenticity studies than low-resolution NMR (frequencies of 10–40MHz) as it is possible to obtain more detailed information regarding the molecular structure using former technique. The major disadvantage of HR-NMR is that it is one of the most expensive analytical techniques in terms of the initial capital outlay and running costs. Various fruit juice adulterants (sucrose, inverted beat sugar 50%, sodium benzoate, etc.) have been successfully detected using 1H NMR combined with principal component analysis (PCA) (Vogel et al., 1996). The detection of pulpwash, which is a serious quality control issue in production of orange juice, was also possible using a combination of H-1NMR and principal components analysis (Le Gall et al., 2001). The expensive equipment and complex data analysis limits application of this technique in food industry. It is difficult for ordinary food inspectors to master NMR for food detection.

Site-specific Natural Isotope Fractionation-NMR (SNIF-NMR) Spectroscopy and Isotopic Ratio Mass Spectrometry (IRMS)

SNIF-NMR and IRMS are the most sophisticated and specific techniques for determining food authenticity (Linda et al., 2006). These techniques exploit the phenomenon of isotopic fractionation. The effect of climatic and geographical conditions on the specific proportions of

particular isotopes of hydrogen and oxygen present in molecules is known as isotopic fractionation. Both techniques are capable of determining the exact proportion and location of specific isotopes within a food sample. SNIF-NMR method is based on the measurement of deuterium/hydrogen (D/H) ratios at a specific site in ethanol molecule to detect clandestine addition of sugar or modified sugar syrups in a juice or concentrate. Authentication of milk samples in terms of their geographic origin (Brescia et al., 2003) and feeding diet was successfully carried out using nuclear magnetic resonance and isotope ratio mass spectrometry (Renou et al., 2004). SNIF-NMR alone (Martin et al., 1996a) or SNIF-NMR and stable isotope ratio analysis mass spectrometry (SIRA-MS) (Martin et al., (1996b)) has been able to detect added sugar and assess authentication of fruit juices. Antolovich and Robards (2001) demonstrated that adulteration of orange juice with sugar can be detected using IRMS of delta ^{13}C values. A combination of SNIF-NMR sample preparation and IRMS analysis of $^{18}O/^{16}O$ ratio has been successfully used to detect adulteration of pure orange juice with orange juice made from concentrate at levels of 25% and above (Jamin et al., 2003). The addition of low cost sugar syrups to apple juice have been successfully detected using IRMS analysis of C^{12}/C^{13} and H^{1}/H^{2} ratios (Kelly et al., 2003). The major advantage of this technique is its capacity to be applied to any raw material or any food product (Remaud et al., 1997; Cordella et al., 2002). However, the high initial and operating cost of NMR and MS instruments, time consuming sample preparation for MS and need for a large knowledge base or database for validation purposes are the major disadvantages of these techniques.

Biosensors

International regulatory agencies have set considerably low acceptable limits for different adulterants/contaminants in milk and fruit juices. Conventional laboratory techniques although sensitive are expensive, require extensive sample preparation, laboratory infrastructure, personal training and have a limited throughput. Thus, there is a growing interest in biosensors due to their high specificity, convenience and quick response. A biosensor is a device that utilizes a biological recognition element which senses the analyte in sample by binding analyte on the reactive surface. The reactive surface of bio-recognition element generates a measured response upon binding with an analyte. The device and associated instrumentation convert analyte-binding events into qualitative or quantitative units of analyte concentration. Transducer is an important component in the instrumentation which is categorized into electrochemical, piezoelectric and thermometric and optical categories. In biosensor, a bio-receptor molecule is immobilized in a suitable matrix to form a biorecognition layer which is then placed in the immediate vicinity of a transducer. Thus, immobilization enables reuse of biomolecules and enhances their local concentration near the sensor probe. Optical biosensors show greater potential for the detection of pathogens (Narsaiah et al., 2012b), pesticides and drug residues, hygiene monitoring, heavy metals and other toxic substances in the food to check whether it is safe for consumption or not (Narsaiah et al., 2012a). Detection of *Enterococci* and *Listeria monocytogenes* in milk and milk products has been done by National Dairy Research Institute (NDRI), Karnal, India by using specific enzymatic reaction in selective media. The free chromogen released by the hydrolysis of chromogenic substrate complex by the activity of enzyme was detected colorimetrically. A spore based biosensor was also developed for detection of *Enterococci*, in which *Enterococci* specific marker enzyme acts on germinogenic substance of the spores coated on a gold chip to

release germinant which further leads to germination mediated concomitant enzyme activity, the product of which could be detected by fluorescence spectroscopy.

Antibodies have prominently been used in biosensor technologies due to their exceptional affinity, wide availability and adaptability to use in many formats. Biosensors have been used to check the adulteration of raw milk from a particular animal with raw milk from some other dairy cattle. Monoclonal antibodies (MAb) were raised against bovine k-casein and applied in an automated optical biosensor to create easy, fast, direct and inhibition biosensor immunoassays (BIA) for detection of cows' milk in the milk of ewes and goats (Willem et al., 2004). They have proven to be robust analytical tools for the automated immunochemical detection of different adulterants and contaminants in milk and milk powder (Haasnoot et al., 2006). An inhibition immunoassay for bovine K-casein has been evaluated for detection of cow's milk in ewe's and goat's milk and for detection of bovine rennet whey powder in milk powder. The sensor was also useful in control of fraudulent water additions to milk, simply by measuring differences in the bulk responses.

A lot of antibody based kits are available in the market for detection of mycotoxins in foodstuffs. The biosensors reported for mycotoxin analysis include optical sensors which are based on surface plasmon resonance, evanescent wave based fiber optic, chemiluminescence (Daly et al., 2000; ; Maragos, 2002; Yuan et al., 2009; Fang et al., 2011), quartz crystal microbalance based on surface acoustic wave (Hauck et al., 1998; Wang et al., 2009) and piezoelectric immunosensors (Jin et al., 2009). Recently biosensors based on amperometry principles (Parker and Tothill, 2009; Rejeb et al., 2009; Rameil et al., 2010) and electrochemical

impedance (Zaijun et al., 2010; Dinçkaya et al., 2011) have also come to light for mycotoxins detection.

Cell-based biosensors further offer many more advantages, such as high speed, low cost and considerable sensitivity, with the possibility to detect just a single target molecule (Kintzios, 2007). Välimaa et al. (2010) demonstrated the ability of detecting estrogenic mycotoxin residues in milk by utilizing bioluminescent whole cell biosensors, based on a genetically modified *Saccharomyces cerevisiae* strain. Recently Larou et al. (2013) developed a rapid novel biosensor system based on the Bioelectric Recognition Assay (BERA) for detection of aflatoxin M1 (AFM1). Membrane engineering was achieved by electroinsertion of AFM1 homologous antibodies into cell membrane. The BERA-based sensor was able to detect AFM1 rapidly (3min) at very low concentrations (5 pg/ml = 5 ppt). In addition, the assay was quite selective against other aflatoxins, such as aflatoxin B1 and ochratoxin A. Furthermore, the assay system allowed for high throughput AFM1 analysis (160 individual tests/h), due to its multiple cell electrode interface array. Although the elimination of possible matrix effects and its validation by assaying actual food samples still remains a challenge.

Fodey et al., 2011 developed a rapid and robust optical biosensor inhibition immunoassay method for the analysis of infant formula and infant liquid milk samples. A compound with a chemical structure similar to that of melamine was employed as a hapten to raise a polyclonal antibody and as the immobilized antigen on the surface of a biosensor chip. The sensitivity of the assay, given as IC50, was 67.9 $ngml^{-1}$ in buffer. The antibody did not cross-react with any of the byproducts of melamine manufacture; however, significant cross-reactivity was observed with the insecticide cyromazine of which melamine is a metabolite. When sample matrix was applied

to the assay, limit of detection <0.5 μgmL^{-1} was determined in both infant formula and infant liquid milk.

Electronic nose

Electronic nose technology is based on the detection of the volatile compounds present in the headspace of a food sample. It consists of an array of semi-selective gas sensors. It is a rapid technique with relatively small amount of sample preparation and can easily be used in conjunction with chemometrics in the authentication of a wide range of food types. Authenticity studies with electronic nose have also been successfully carried out for differentiation of determination of the geographical origins of Valencia orange juices (Steine et al., 2001) and Emmental cheese (Pillonel et al., 2003). An e-nose system has been used to classify off-flavours in milk (Marsili, 1999), which originate mostly from the metabolic and enzymatic activity of bacteria, as well as heat, light (photo-oxidation) and oxidation catalyzed by chemicals (Marsili, 2000, Ampuero et al., 2004). Korel and Balaban (2002) could quantify bacterial population in sterile, spiked (*P. fluorescence* and *Bacillus coagulans*) milk samples using e-nose and discriminant function analysis (DFA). Amari et al. (2009), developed e-nose system, which could be used to establish the shelf-life of raw milk. Electronic nose and tongue have been developed for aroma characterization and checking of briskness strength of black tea liquor, respectively (Bhattacharya and Bandhopadhyay, 2010). Major drawback of this technique is that sensors employed are not very selective for particular types of compounds thus preventing any real identification or quantitation of individual compounds present in a food matrix. Such a drawback has obvious implications for food authentication, as an adulterant could not be definitively identified. Despite its drawbacks, research on electronic nose technology holds much

potential for future development. The types of various techniques and their respective sensitivities are summarized in Table 5.

CONCLUSION

Milk is being adulterated using various chemicals and non-chemical substances; while fruit juices are mixed with cheaper similar liquid/juice. Sugars are also added without giving information of the same on labelling. Numerous researches have been carried out for development of detection protocols and methods for these adulterants and authentication of liquid foods. Analytical technique like chromatography has been successfully used for the purpose. It is, though sensitive, requires extensive sample preparation for complex food samples to avoid damage to the chromatography column and detector. In most cases identity of the interfering compound is unknown, resulting in misidentification of target compounds. Selectivity of liquid and gas chromatography has been enhanced by collaboration with the mass spectrometry, but their use is limited to large analytical laboratories due to high costs involved in acquiring and running such systems. Immunological techniques such as ELISA have been used extensively for food authentication and mycotoxin detection, but it involves initial difficulty in producing antibody specific to a particular protein. Polymerase Chain Reaction (PCR), an invitro technique based on the principle of DNA polymerization reaction, is widely used for rapid differentiation of pathogenic and non-pathogenic microorganisms. Its only drawback is its inability to distinguish between dead and living cells. Inhibitory compounds in food samples, further limit the application of PCR technology. The phenomenon of change in electrical

properties due to the breakdown of complex molecules of the medium into charged catabolic products has also been used in dairy microbiology for detecting specific microorganisms. Requirement of a large bacterial population for efficient detection is however the major drawback of this technology. Wet chemistry methods to detect and quantify adulterants and contaminants in foods although are simple and accurate; they need many steps involving use of various reagents and chemicals.

Electronic nose is another rapid, relatively cheap and easy to operate technique for authentication of a wide range of food types. This technique holds much potential for future development as it can easily be used in conjunction with chemometrics. If existing problems like low specificity and positioning of sensors are overcome, electronic nose could prove to be an efficient tool for food authentication and quality assurance in food industry. There is also a growing interest in biosensors due to their high specificity, convenience and quick response. Many antibody based immunosensor kits are already available in the market for the detection of mycotoxins in the foodstuffs which have sensitivity in the range of ng/ml and <100 colony-forming units/ml for bacteria. Biosensors however have certain limitations linked to antibody generation, as antibodies have short shelf-lives and are difficult to be chemically modified for incorporation into a biosensor.

Isotopic ratio mass spectroscopy and nuclear magnetic resonance spectroscopy techniques are nondestructive, independent of the sample size and shape and have high accuracy with good repeatability; but their application in industry can be made feasible only by lowering initial capital outlay and running costs. Spectroscopic techniques like near infrared, mid infrared and Raman spectroscopy have also demonstrated their potential as nondestructive, simple and

sensitive detection and quantification techniques for food adulterants and contaminants. These are fast, relatively less costly, and give high-throughput analysis of food components with little or no sample preparation. Advancements in instrumentation and chemometric pattern recognition techniques have further made it possible to extract information related to the composition of food components from complex spectra. Spectroscopic techniques provide fingerprints of different food products which can be used to detect different malpractices and thus assure quality. Spectroscopic techniques offer the greatest potential for development of cost effective and widely applicable food authentication tools to safeguard interests of both consumers and food industries. There is an urgent need of further research to make commercial grade equipment for onsite and online detection of adulterants and contaminants to save millions of lives from consuming adulterated foods.

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Table 1. Adulterants/contaminants and their permissible limits in milk and fruit juices

Food article	Adulterant/contaminant	Max. Permissible limit	Reference/publication	Website Accessed on 27.07.2012
Milk	Urea	7×10^5 μg/kg	Food Safety and Standard Authority of India (FSSAI)	www.fssai.gov.in
	AflatoxinB1	30 μg/kg	FSSAI	www.fssai.gov.in
		5 μg/kg	Codex	www.**codex**alimentarius.org
		2 μg /kg	European Communities	www.efsa.europa.eu/
	AflatoxinM1	0.5μg/kg	FSSAI	www.fssai.gov.in
		0.5μg/kg	USA Regulations	http://www.gipsa.usda.gov
		0.5μg /kg	Codex	www.**codex**alimentarius.org
		0.05μg /kg	European Communities	http://www.efsa.europa.eu
	Water	86-88%	Prevention of Food Adulteration act	www.fssai.gov.in
	Glucose/ Lactose	4.5-5.0%	Prevention of Food Adulteration act (FSS)	www.fssai.gov.in
	Melamine	1mg/kg	FDA, CODEX 2008	www.**codex**alimentarius.org
	Cyanuronic acid	1mg/kg	FDA, CODEX 2008	www.**codex**alimentarius.org
	Bacteria	10^5 bacteria ml^{-1}	EU Directive 92/46 EEC	www.efsa.europa.eu/
Fruit Juice (Apple& Mango)	Sugars	-	-	-
	Water	-	-	-
	Patulin	50μg/kg	FSSAI	www.fssai.gov.in
		50μg/kg	CODEX	www.**codex**alimentarius.org
		50μg/kg	USA	http://www.gipsa.usda.gov
	Corn Syrup	-	-	-
	Total Aflatoxins	20μg/kg	USA Regulations	http://www.gipsa.usda.gov
		30μg/kg	India	www.fssai.gov.in
		4mg/kg	European Communities	www.efsa.europa.eu/

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Table 2. Physiochemical methods for the determination of adulterants in milk and fruit juices

Food	Adulterant	Biochemical test		Reference
Milk	Urea	Urease+red litmus	Qualitativ	FSSAI
		Diacetyl method	Quantitati	Paradkar et al., 2000a
		Phenol red method	ve	Paradkar et al., 2000a
		DMAB method		Bector et al., 1998;
	Sodium chloride /Chlorine	Potassium iodide	Qualitativ	Souza et al., 2011
		Potassium chromate +		FSSAI
	Carbonates/bicarbonates	Rosalic acid test	Qualitativ	Ayub et al., 2007
	Starch/flour	Iodine test	Qualitativ	Ayub et al., 2007
		Anthrone method	Quantitati	Thimmaiah,1999
	Synthetic milk	Urease strips test	Qualitativ	FSSAI
		Diacetyl method,	Quantitati	Paradkar et al., 2000a
	Formaldehyde/Formalin	Nessler reagent test	Qualitativ	Souza et al., 2011
		Sulphuric acid test		FSSAI
	Sugar	HCl +Resorcinol test	Qualitativ	FSSAI
		DNS test	Quantitati	Thimmaiah,1999
		Phenol sulphuric acid	ve	Dubois et al.,1956,
		Anthrone method		Hedge and
		Nelson- Somogyi		Thimmaiah,1999
	Vegetable ghee/oil	HCl+ sugar test	Qualitativ	FSSAI ,
	Detergent/Shampoo	Shaking with water to	Qualitativ	FSSAI,
		methylene blue +	Quantitati	Paradkar et al., 2000b
	Water	Lactometer reading	Qualitative	FSSAI
	Lactose/glucose	diastrix strip test	Qualitativ	FSSAI
		Lactometer reading	Quantitati	FSSAI
	Hydrogen peroxide	Peroxide strips test,	Qualitative	Ayub et al., 2007,Souza
Fruit Juice–____	Sugar	HCl + Resorcinol test	Qualitativ	FSSAI
		DNS test	Quantitati	Thimmaiah,1999
		Phenol sulphuric acid	ve	Dubois et al.,1956 ,
		Anthrone method		Hedge and

Table 3: Wavelength ranges and correlation coefficients of different adulterants in milk (Jha and Matsuoka, 2004)

S.No.	Adulterant	Wavelength range (nm)	R (Calibration)	R (Validation)
1	Milk	926.634 – 939.409	0.89	0.89
2	Urea	996.599 – 1021.85	0.98	0.98
3	NaOH	945.788 – 977.599	0.95	0.86
4	Oil	926.634 – 961.708	0.89	0.74
5	Shampoo	933.024 – 945.788	0.69	0.58

Table 4: Classification of prepared and commercial samples to adulterated and unadulterated groups using PCA projection method at 5% significance level in wavenumber range 1476-912cm^{-1} after baseline offset correction (Jha and Gunasekaran, 2010)

Sample solution	Added sucrose	Total numbers of samples	Number of sample adulterated		Correct classification (%)
			Unadulterated	Adulterated	
Sugar solution	0	5	5	5	0
	1	5	5	5	0
	5	5	0	5	100
	9	5	0	5	100
	13	5	0	5	100
	17	5	0	5	100
	21	5	0	5	100
	25	5	0	5	100
Prepared juice-1	0	5	5	5	0
	1	5	5	5	0
	5	5	5	5	0
	9	5	0	5	100
	13	5	0	5	100
	17	5	0	5	100

	21	5	0	5	100
	25	5	0	5	100
Prepared juice-2	0	5	5	5	0
	3	5	5	5	0
	7	5	0	5	100
	11	5	0	5	100
	15	5	0	5	100
	19	5	0	5	100
	23	5	0	5	100
	27	5	0	5	100
Commercial juice-1	3.6	5	4	1	20
	7.1	5	0	5	100
	10.7	5	0	5	100
Commercial juice-2	12.8	30	0	30	100

Table 5. Various techniques and their sensitivity to detect adulterants/contaminants in milk and fruit juices

Adulterant ANALYTE	MATRIX	TECHNIQUE	SENSITIVITY	REFERENCE
Detergent/ shampoo	Milk	Colorimetric	0.02g/100ml (20mg/100ml)	Paradkar et al., 2000b
	Cow's milk	Near-IR	0-25g/100ml (0-25%)	Jha and Matsuoka, 2004
Synthetic milk	Milk	Colorimetric	0.02g/100ml (20mg/100ml)	Paradkar et al., 2000a
Urea	Milk	Colorimetric	0.02g/100ml (20mg/100ml)	Paradkar et al., 2000a, Bector et al., 1998; Abdallah et al., 2008
	Cow's milk	Near-IR	0-25g/100ml (0-25%)	Jha and Matsuoka, 2004
Whey powders	Milk	Optical sensor	0.006g/100ml	Nikoleli et al., 2010
		Immunochromatography	.0015-0.003 g/100ml(15-30µg/ml)	Oancea, 2009
		Mid-infrared (IR)/ FTIR	2.26g/100ml (2.26%)	Cassoli et al., 2007
		Near-IR	2.15g/100ml	Kasemsumran et al., 2007
NaOH	Cow's milk	Near-IR	0-25g/100ml (0-25%)	Jha and Matsuoka, 2004
Vegetable oil	Cow's milk	Near-IR	0-25g/100ml (0-25%)	Jha and Matsuoka, 2004
Carbonates/ bicarbonates	Milk	Mid-infrared (IR)/ FTIR	0.015g/100ml (0.015%)	Ayub et al., 2007
Milk from other animals	Milk	MALDI-MS	0-100% (Max error 2-10%)	Nicolaou et al., 2011
Bacteria	Milk	SWNIR	2.7-10.5$\log_{10}$ cfu/ml	Al Qadiri et al., 2008
L. monocytogenes	Milk	Biosensor	10-50 cells/25gm	Peng and Shelef, 2000
Aflatoxin M1	Milk and dairy products	High-performance liquid chromatography (HPLC)	10^{-9} g/100ml (10 pg/ml)	Kim et al., 2000
			13.1ng/L- 84.5ng/L	Pathirana et al., 2010

	Milk	ELISA	2×10^{-10} g/100ml (2 pg/ ml)	Kim et al., 2000
Aflatoxin B1	Milk	ELISA	1.8ppb	Kang'ethe and Lang'a, 2009
Patulin	Fruit Juice	Thin layer chromatography	2×10^{-6}g /100ml (20μg/l)	Scott, 1974
		GC	10^{-6}g /100ml (10μg/l)	Sheu and Shyu,1995
		HPLC	4.7×10^{-6}g /100ml (0.047μg/ml)	Gashlan, 2009,
		Near-IR	1ml/100ml	Kasemsumran et al.,2007
	Fruit Juice	Near-IR	10-20g/100gm	Li-juan et al., 2008
		Pyrolysis mass spectrometry	< 1g/100ml (<1%)	Goodacre et al., 1997
		Near-IR	10- 40g/100gm	Kelly and Downey, 2005a,b
		FT-NIR	0.8g/100ml	Luis et al., 2001
Corn Syrup	Fruit Juice	Mid-infrared (IR)/ FTIR	9.5gm/100ml (9.5%)	Kelly and Downey, 2005
Other juices	Fruit Juice	Mid-infrared (IR)/ FTIR	2-14 ml/100ml	Vardin et al., 2008

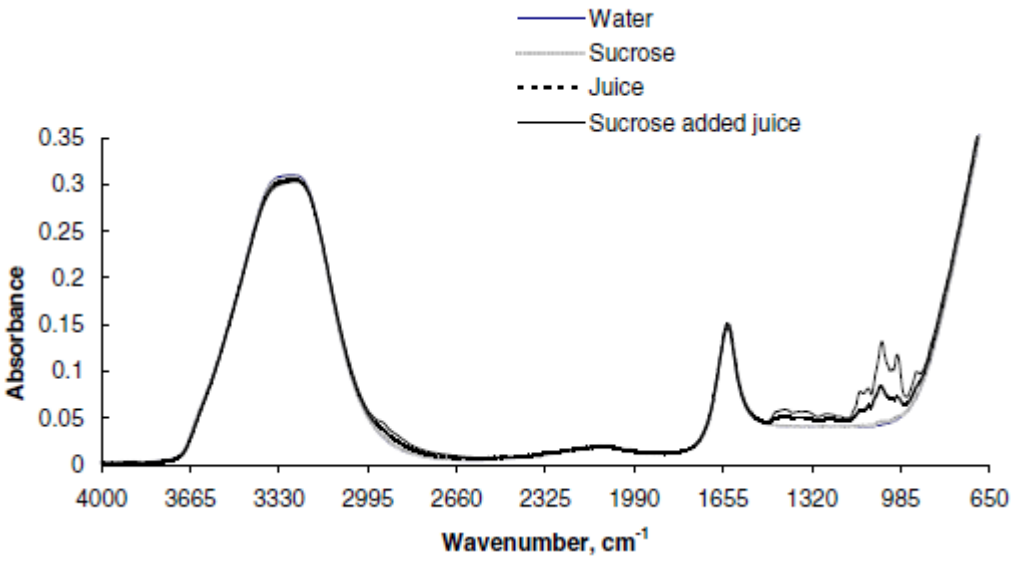

Fig.1. Typical spectra of water, sucrose, pure juice and sucrose added juice (Jha and Gunasekaran, 2010)

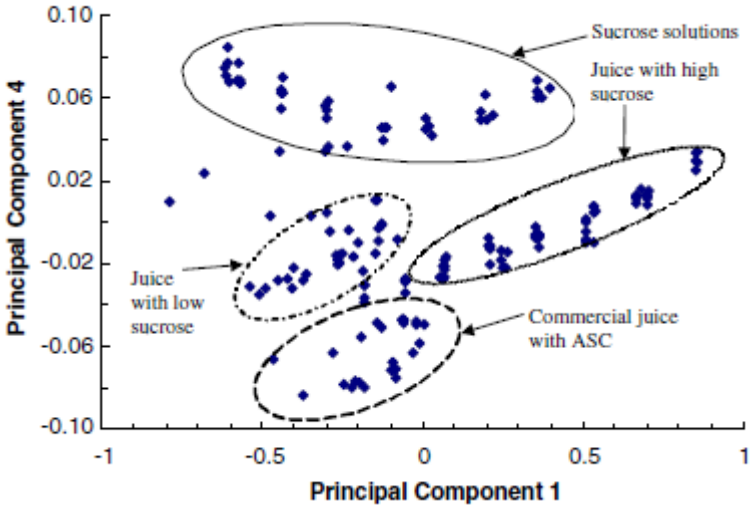

Fig.2. Principal component scores plot of 165 samples, ellipses show grouping of samples according to their nature (sugar solution, prepared and commercial juices) (Jha and Gunasekaran, 2010)