A review of Fourier Transform Infrared (FTIR) spectroscopy used in food adulteration and authenticity investigations

Reema Valand, Sangeeta Tanna, Graham Lawson, Linda Bengtström

School of Pharmacy, Faculty of Health and Life Sciences. De Montfort University; Leicester LE1 9BH, UK

Corresponding author:

Linda Bengtström

School of Pharmacy, Faculty of Health and Life Sciences

De Montfort University; Leicester LE1 9BH, UK

linda.bengtstrom@dmu.ac.uk

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Abstract

The increasing demand for food and the globalisation of the supply chain, have resulted in a rise in food fraud, and recent high profile cases, such as the Chinese milk scandal in 2008 and the EU horsemeat scandal in 2013 have emphasised the vulnerability of the food supply system to adulteration and authenticity frauds. Fourier Transform Infrared (FTIR) spectroscopy is routinely used in cases of suspected food fraud as it offers a rapid, easy and reliable detection method for these investigations. In this review we first present a brief summary of the concepts of food adulteration and authenticity as well as a discussion of the current legislation regarding these crimes. Thereafter, we give an extensive overview of FTIR as an analytical technique and the different foods where FTIR analysis has been employed for food fraud investigations as well as the subsequent multivariate data analysis have been applied successfully to investigate case of adulteration or authenticity. Finally, we give a critical discussion of the applications and limitations of FTIR, either as a standalone technique or incorporated in a test battery, in the fight against food fraud.

Key words: FTIR, food fraud, food adulteration, food authenticity, spectroscopy

Introduction

With the increasing globalisation of the food supply chain, with several stakeholders in between ‘farm to fork’, and the intrinsic vulnerability of such a system, food quality and safety have become of increased concern for consumers, food producers and governments (US Food and Drug Administration, 2009; European Commission, 2018). Scandals such as the addition of melamine in baby formula in China in 2008 (Pei et al., 2011), the European horse-meat scandal in 2013 (Premanandh, 2013), and the issue of peanuts and almonds found in ground cumin and paprika in Europe and the US in 2015 (Agres, 2015), all illustrate the negative impact of food fraud, including both adulteration and authenticity.

Although there is no harmonised definition of food fraud in the EU, it is generally accepted by the member states that food fraud are committed ‘intentionally for financial gain through consumer deception’ (European Commission, 2018). The criminal activity of intentionally adulterating food has gradually become more prevalent due to the associated incomes (Galvin-King, Haughey and Elliott, 2018). Food adulteration can be defined as the process in which the quality of food is purposefully degraded either by the addition of low-grade quality material or by extraction of valuable ingredients (Spink and Moyer, 2011). Nowadays, the most common foods categories susceptible to any type of food fraud are in descending order; olive oil, fish, organic foods, milk, grains, honey and maple syrup, coffee and tea, spices, wine and certain fruit juices (Moore, Spink and Lipp, 2012; Hoffman, 2013a). The so-called premium food products, such as meat (Rohman *et al.*, 2011; Nunes *et al.*, 2016a), spices and herbs (Lohumi *et al.*, 2017a; Wielogorska *et al.*, 2018), honey (Amiry, Esmaili and Alizadeh, 2017), olive oil (Rohman and Man, 2010; Georgouli, Martinez Del Rincon and Koidis, 2017a), and coffee (Reis *et al.*, 2017) are particularly susceptible to adulteration, especially when they are produced and supplied through complex supply chains (Black *et al.*, 2016). Expensive commodities are vulnerable to economically motivated adulteration (EMA) due to the large incomes generated from these products (Galvin-King, Haughey and Elliott, 2018).

Conversely, cheap foods, such as milk and grains, are also susceptible to adulteration, mainly due to the low margins of profit (Smith, Manning and McElwee, 2017). For instance, the increased consumption of milk, due to its high nutritional value, has made it prone to fraudulent activity

(Handford, Campbell and Elliott, 2016). Additionally, 21st century agriculture faces multiple challenges due to increasing global population, such as an increasing demand on food production for humans and livestock as well as increased demands on feedstock for the bioenergy market (FAO, 2009). The agricultural sector also faces demands due to the developments in the agriculture-dependent developing countries and the adaptation to more efficient and sustainable production methods and climate change (FAO, 2009). All of these factors are important when understanding the reasons for food fraud. Although it is not currently known how common food fraud is, it is estimated that the cost for the global food industry could be as high as US\$40 billion dollars annually (PwC, 2016).

Depending on the adulterant used, food fraud could have severe adverse health effects, such as the development of cramps, nausea, diarrhoea, vomiting, nerve damage, allergic reactions and paralysis (Sicherer, Burks and Sampson, 1998). For instance, in 2015 there were reports of the addition of peanuts and almonds to cumin and paprika powder in the USA and Europe (Agres, 2015). The unintentional ingestion of these two allergens could potentially lead to severe or even lethal allergic reactions (Sicherer, Burks and Sampson, 1998). Other common allergens found in contaminated food are fruits, soybeans, fish, milk, egg, tree nuts, wheat and shell fish (Añíbarro, Seoane and Múgica, 2007) all of which can cause severe health problems.

The scandals concerning fraudulent foods have increased the pressure on food laboratories to develop fast and reliable screening methods for the detection of food fraud. One of the most commonly used screening techniques for food fraud today currently is Fourier Transform IR (FTIR) based on mid infrared (MIR) vibrational spectroscopy, as it offers a rapid and reliable detection method and in this review we describe the use of FTIR as a tool to investigate food adulteration and food authenticity. The first part of this review offers an overview of food adulteration and authenticity from a historical and current global perspective. The second part introduces FTIR as a screening technique and discusses the theory behind this technique. Finally, the third part of this review offers a critical discussion of the use of FTIR and multivariate analytical tools, on their own or as part of a battery of techniques as a method to investigate different food matrices.

This is to our knowledge the first critical review of FTIR as a tool to investigate food adulteration and authenticity since Rodriguez-Saona and Allendorf published their review in 2011 (Rodriguez-Saona and Allendorf, 2012) and considering the recent advances in the analytical techniques as well as the recent high profile food scandals, it is therefore a timely addition to the discussion of food authenticity and adulteration.

Food adulteration and authenticity

Legislation and industry standards

Food fraud is a collective term that encompasses the deliberate substitution, addition, tampering or misrepresentation of food, food ingredients or food packaging, or false or misleading statements made about a product for economic gain (Spink and Moyer, 2011; Lakshmi and Pradesh, 2012). Within the EU there is currently no harmonised definition of food fraud, however, there are four operational criteria for establishing food fraud (European Commission, 2018). These include the violation of the EU Food Law, intention, economic gain and deception of customers (European Commission, 2018). In the General Food Law Regulation EC 178/2002 (European Commission, 2002), the broad principles of food safety and regulation are outlined. For instance, under Article 8 – ‘Protection of consumers’ interests’, the General Food Law states its aim to prevent ‘fraudulent or deceptive practices’ and ‘the adulteration of food’ (European Commission, 2002). Furthermore, the EC 178/2002 regulation also established the European Food Safety Authority (EFSA) to provide scientific advice (European Commission, 2002) following some high profile European food scandals during the previous two decades.

In the United States (US), the two primary agencies for food safety, and thus for food fraud and authenticity, are the US Department of Agriculture (USDA) and the US Food and Drug Administration (Johnson, 2014). The primary legislation concerning food safety; governing food, drug and consumer protection, is the Federal Food, Drug and Cosmetic Act (FFDCA) from 1938 (United States Congress, 1938). In 2011, the Food Safety Modernization Act (FSMA) was signed into law,

awarding the FDA further authorities such as a mandatory recall authority (United States Congress, 2011), deemed necessary due to reported incidents of foodborne illnesses during the early 2000's.

It is important to emphasise that not all levels of contamination are considered intentional adulteration. Most national and international legislations as well as industrial organisations today have a threshold for extraneous matter to allow distinction between accidental and intentional contamination. For instance, the threshold for gross adulteration for horsemeat in the UK is when food items contain 1% weight by weight (w/w) or above of extraneous material (Food Standards Agency, 2015). In the US, the FDA have established the Food Defect Action Levels in order to distinguish between 'natural or unavoidable' defects, where adulteration is one of many causes, for a range of commodities (US FDA, 2018). An example of an industry organ with its own thresholds is the European Spice Association (ESA), with a maximum of 2% w/w for extraneous material for herbs and 1% w/w for spices (ESA, 2015). However, with recent developments in increasing sensitivity of the methods employed, such as DNA tests and analytical chemical methods, to detect food fraud, very low concentrations of a contaminant could be detected (Galvin-King, Haughey and Elliott, 2018). These thresholds are therefore in place to differentiate between contamination and intentional adulteration, and are set based on the limit of detection for the particular analytical technique used to detect the adulteration (Downey, 2016).

Testing for food authenticity is important to ensure that food offered for sale or sold is of the nature, substance and quality expected by the purchaser (Defra et al., 2014). If manufactures fail to correctly and honestly state what ingredients are present within the food item, it becomes deception and is then classified as food crime (Defra et al., 2014). There are therefore harmonised regulations in the EU on food labelling, presentation and advertising aim to protect consumers and facilitate trade inside and outside Europe (European Commission, 2000). In 2006 the European Council Regulation introduced three different types of quality logos for ensuring authenticity of agricultural products and foodstuff (The Council of the European Union, 2006). Two of these logos - the Protected Designation of Origin (PDO) and the Protected Geographical Indication (PGI) - have a specific link to the region where the product comes from, while the third one - the Traditional Speciality Guaranteed (TSG) logo

highlights a traditional production process (The Council of the European Union, 2006). In practicality, this directive regulates food or drinks that are produced must adhere to a precise set of specifications to be labelled with any of the three logos (The Council of the European Union, 2006). As an example, as wine is a premier agricultural product of the EU and exported worldwide, the EU has taken serious action to maintain the reputation of the wine producing companies and other food manufacturers. Since 2011, the EU also has specific regulations for wine authenticity since 2011. Wine adhering to the specifications set by the framework is now categorised according to PDO and PGI (The European Commission, 2014b, 2014a).

Food adulteration throughout history

One subcategory of food fraud is Economically Motivated Adulteration (EMA), which can be defined as the intentional contamination of food for increased profit (Spink and Moyer, 2011). Not only is EMA a fraudulent activity that might harm consumers trust in a specific food category or company (GMA and Kearney, 2010; Kendall *et al.*, 2018), it might also directly or indirectly have adverse effects on consumers health by the addition of potentially harmful substances to the food (Spink and Moyer, 2011). However , EMA is by no means a contemporary phenomenon and is likely to be as old as the food processing and production systems themselves (Ellis *et al.*, 2012). Historical examples of EMA are the addition of lead to soured wine in ancient Rome, or adding the drug *cocculus indicus* in order mask the smaller amounts of hops and malt used during brewing or mixing alum with flour in bread to make it whiter (Evershed and Temple, 2016).

The first researcher to apply scientific methods to detect food adulteration was Frederick Accum, who made the first serious attempt to expose both the extent and dangers of food adulteration in 1820 (Accum, 1820). However, more than half a century after Accum first published his book, food adulteration was still so rife that a journalist for the *New York Times* wrote that most people were not aware of ‘what abominable messes they are constantly putting down their throats under the most innocent disguises’ (Evershed and Temple, 2016).

Even though modern food adulteration is usually more sophisticated than these historical examples, it is no less prevalent, and only in the last decade, there have been several high profile cases linked to food adulteration and authenticity. In 2008, elevated levels of melamine was found in the products of 22 Chinese dairy companies (The Associated Press, 2008), and this contamination led to the death of six infants and the hospitalisation of over 52,000 small children (Pei *et al.*, 2011). Melamine is an organic base which is used to make plastics, fertilisers and concrete, and can cause kidney stone formation that in certain risk groups can trigger acute renal failure (Skinner, Thomas and Osterloh, 2010). When melamine is added to food products, it mimics a higher protein content of a diluted food (Pei *et al.*, 2011), and this unapproved enhancement of the milk is a case of the devastating consequences EMA can have (Lakshmi and Pradesh, 2012). The melamine scandal in China also illustrates the vulnerability of food safety regulation if official controls do not efficiently assure the implementation of existing legislation (Pei *et al.*, 2011).

Another highly publicised food scandal is the so-called ‘2013 horsemeat scandal’ in Europe in which food products marketed as beef were found to contain high levels, sometimes up to 100%, of undeclared or improperly declared horse meat (BBC News, 2013). This case, where cheaper horsemeat was mislabelled as expensive beef, highlighted several vulnerabilities within the European food supply chain (Department of Agriculture Food and and Marine, 2013; Elliott, 2013). For instance a complex network of traders and grocers, which may contain farmers, primary processor, local traders, secondary processor, exporter, importer, trader, processor/packager, retailer and finally consumer, cause a lack of transparency in this part of the supply chain (Department of Agriculture Food and and Marine, 2013; Elliott, 2013; Galvin-King, Haughey and Elliott, 2018). In the aftermath of the horsemeat scandal, it was concluded that the control of food safety also need to involve checks on food authenticity (Department of Agriculture Food and and Marine, 2013).

One of the more recent cases of EMA with potentially severe adverse health effects for the public is the adulteration of ground cumin and paprika sold in Europe and the US by the addition of peanuts and almonds (Agres, 2015). The unintentional ingestion of peanuts or almonds could potentially lead to severe or even lethal allergic reactions (Sicherer, Burks and Sampson, 1998) and detection of

such adulterations is of high interest. This is also an example whereby adulteration associated with EMA is driven by crop failures (Bawden, 2015). Whenever there is a crop failure of a certain commodity, that commodity shows an increased susceptibility to food fraud, similar to any food with a sudden increase in price or demand (Bawden, 2015; Black *et al.*, 2016).

Yet another example of adulteration of a food product in high demand is the dilution of expensive extra virgin olive oil (EVOO) with cheaper vegetable and seed oils (Dalmia, 2015). This also results in a lower nutritional value further adding to the fraudulent nature of the adulteration (Dalmia, 2015). The adulteration of olive oil using unhealthy substitutes is therefore considered a threat for public health (Ok, 2017). The increased global demand for olive oil coupled with the recent years of drought in the Mediterranean regions, has seen a steep rise in the prices of EVOO (Schrieberg, 2017), making adulteration particularly profitable. Adulteration of EVOO has therefore recently been recognised as a common problem for regulatory agencies, oil suppliers and consumers (Dalmia, 2015). For instance, in 2012, two Spanish businessmen were sentenced to prison after they were found trading hundreds of thousands of litres of EVOO adulterated with sunflower oil (70-80%) (Henley, 2012).

Food authenticity as an emerging issue

The authenticity of food can be understood as how honestly and accurately the food item has been marketed and trading of inauthentic foods is currently a large economic problem (Hansen, 2015). In order to protect consumers and implement food laws, it is important to verify the labelling of food in relation to composition, processing or origin (Primrose, Woolfe and Rollinson, 2010). The determination of authenticity is important so that consumers can make informed choices in relation to health concerns, religious beliefs or lifestyle choices (Primrose, Woolfe and Rollinson, 2010). Food products that are marketed claiming to contain natural ingredients or originating from a special location produced using local and traditional methods become difficult to verify unless thoroughly checked (Hansen, 2015). Trading of inauthentic foods is currently a large economic problem (Hansen, 2015).

Food authenticity is a growing concern due to the consequences of deceptive marketing can have on consumer health and trust. Moreover, the country or region of origin of certain commodities or

products that meet the standards of organic farming, such as saffron (Anastasaki *et al.*, 2010), olive oil (Gouveinhas *et al.*, 2015), honey (Gok *et al.*, 2015) as well as wine (Cozzolino *et al.*, 2009a) and wine derived products (Ríos-Reina *et al.*, 2017) will greatly impact the retail price. This increased price makes these products vulnerable to food fraud due to the economic gains associated with the substitution with a cheaper ingredient. For instance, the price of saffron is very much dependent on the country of origin (Anastasaki *et al.*, 2010), and there have been reports showing that more than 50% of saffron labelled as the prestigious and Protected Designation of Origin (PDO) Spanish saffron could be grown and/or processed in other countries, such as Iran (Govan, 2011; Rubert *et al.*, 2016).

Similarly, olive oils from certain PDO regions or countries, such as Sabina (Lazio), will also fetch a higher price due to their unique qualities in flavour and/or production technique (Bevilacqua *et al.*, 2012a). The verification of authenticity of a food product is however a complex issue, where the entire food product is usually analysed in a non-targeted analysis (Bevilacqua *et al.*, 2012a). In these cases, no foreign material has been added to the food product and only small chemical differences in several variables between the authentic and the fraudulent product are analysed, which requires multivariate statistical techniques, such as partial least squares (PLS), principal component regression (PCR), principal component analysis (PCA) or Soft Independent Modeling Class Analogy (SIMCA) to distinguish between foods with very similar chemical composition (Granato *et al.*, 2018).

From the cases presented above it is clear that food fraud is neither a new phenomenon, nor something that is expected to decrease within the foreseeable future. Even if the food safety legislation has become more stringent in modern times, there is need for official controls using scientific methods to ensure that these regulations are implemented. It is clear that food fraud has been and will be posing a threat not only to the food industry in terms of loss of consumer trust and ultimately profits, but also, and more importantly, to consumers in the shape of potentially adverse health effects. These examples of food fraud also illustrate the need for a rapid, easy and reliable detection technique in order to analyse a larger subset of food for adulteration and authenticity, and thus discourage fraudulent activities.

FTIR vibrational spectroscopy as a detection technique

Introduction to infrared vibrational spectroscopy

Mid-IR (MIR) spectroscopy as an analytical technique measures the absorbance of IR radiation of dipole bonds within functional groups of molecules (Chalmers and Griffiths, 2002a; Gauglitz and Vo-Dihn, 2003). The absorbance of IR radiation excites the electrons in the bonds to a higher state of vibration (Gauglitz and Vo-Dihn, 2003), causing bending or stretching motions of the molecular bonds (Chalmers and Griffiths, 2002b). A molecule will absorb IR radiation only if the absorption of the particular wavelength causes a change in the at-rest and excited vibrational states (Amand and Tullin, 1999). Most chemical compounds, except for instance elemental diatomic gases such as N₂, H₂ and O₂, have a dipole moment and can be analysed by their characteristic infrared absorption (Amand and Tullin, 1999).

For organic compounds, characteristic stretching vibrations usually occurs between 4000-1500 cm⁻¹, in the so-called MIR region (Gauglitz and Vo-Dihn, 2003). However, below 1500 cm⁻¹, vibrations, generally bending, result in a characteristic fingerprint of the molecule or a large part of the molecule and can therefore give structural information useful for identification (Stuart, 2005). Moreover, the intensity of the absorbance is directly proportional to the concentration of a pure compounds or that particular feature when analysing mixtures (Gauglitz and Vo-Dihn, 2003), further making IR spectroscopy suitable for quantitative purposes.

FTIR instruments utilise the interference between two IR beams to yield a signal, called an interferogram, which is a function of the change in path length between the two beams (Stuart, 2005). This is usually achieved using a Michelson interferometer, see Figure 1. The generated signal from a Michelson interferometer can then be decomposed into the frequencies that form a signal, using Fourier Transform algorithms (Stuart, 2005). The Michelson interferometer, first described in 1881 (Michelson, 1881, 1927), consists of a light source, such as a mercury arc, tungsten or a globar lamp, a semi-reflecting beamsplitter and two perpendicular mirrors, one stationary and one moving (Stuart, 2005; Albert, Albert and Quack, 2011a). In the first part of the interferometer a collimated IR beam is split with equal intensity by a beamsplitter (Gauglitz and Vo-Dihn, 2003). These divided beams are then reflected by both the stationary and moving mirrors back to the beamsplitter, where they recombine and interfere, see Figure 1 (Gauglitz and Vo-Dihn, 2003). This interference, either constructive or destructive in the recombined beam, are caused by the moving mirror producing a difference in path length (Stuart, 2005).

Figure 1. Schematics of a Michelson interferometer used in modern FTIR instruments. Adapted from Albert, Albert and Quack (2011)

Although the Michelson interferometer was developed in the late 1800's, it was not until the late 1970's the methodology revolutionised high resolution IR, when the bottleneck of computational power to perform an Fourier transformation on high resolution IR data was overcome (Albert, Albert and Quack, 2011b). The Fourier transformation convert the obtained interferograms, presenting intensity versus time, to an IR spectrum, showing intensity versus frequency (Gauglitz and Vo-Dihn, 2003). Prior to the computer revolution in the late 1970's, the resolution of most FTIR instruments were comparable to the dispersive instruments (Albert, Albert and Quack, 2011b). Currently, the resolution of an FTIR instrument is limited by the maximum path difference between the two beams reflected by the mirrors (Stuart, 2005), see Figure 1, which is proportional to the pathlength difference between the Michelson interferometer arms, so that a longer distance would increase the resolution (Gauglitz and Vo-Dihn, 2003). More precisely, the limiting factor to achieve even higher resolution with modern FTIR instruments is the precision of the optic and movement mechanisms for the moving mirrors in the interferometer (Stuart, 2005).

Analysis of foods adulteration and authenticity using FTIR

Foods analysed by FTIR

Traditionally, FTIR has been used to detect food adulteration in virgin oil (Rohman and Man, 2010; Georgouli, Martinez Del Rincon and Koidis, 2017b), honey (Rios-Corripio *et al.*, 2011; Gok *et al.*, 2015) and beef (Alamprese *et al.*, 2013; Nunes *et al.*, 2016b). However, recently it has also been used to analyse adulteration and authenticity in spices and herbs, such as saffron and oregano (Black *et al.*, 2016; Lohumi *et al.*, 2017b; Petrakis and Polissiou, 2017; Wielogorska *et al.*, 2018), authenticity of wine and wine based products (Cozzolino *et al.*, 2009b; Ríos-Reina *et al.*, 2017) and nectars and jams (Miaw *et al.*, 2018). All in all, IR spectroscopy has been used to detect fraud of many of the food products listed as most at risk (Hoffman, 2013b), emphasising the effectiveness of FTIR analysis in the battle against EMA and authenticity frauds.

Sampling techniques for food analysis using FTIR

Analysis of solid foods in FTIR can be carried out using alkali halide discs made from potassium bromide (KBr), as this simple inorganic salt do not generate any vibrations in the MIR region (Nyquist and Kagel, 2012). Solid materials can therefore be analysed by mixing the analyte with KBr and subsequently pressing the mix to a fused disc which can be introduced into the light beam of a spectrophotometer (Simmons, 1960; Gauglitz and Vo-Dihn, 2003). The use of KBr discs is seemingly the preferred method in some studies, such as for the detection of fraud in saffron samples (Anastasaki *et al.*, 2010; Ordoudi *et al.*, 2017; Petrakis and Polissiou, 2017), see Table 1. One of the main disadvantages by using KBr discs is the issue of reproducibility, as conditions such as matrix ratios and homogeneousness needs to be exactly the same for each sample. Even with the addition of internal standards, the discs and mulls must be prepared under precisely the same circumstances to avoid any changes in path length and thus response (Stuart, 2005). Moreover, KBr pellet method is susceptible to interfering moisture effects, that may cause water absorption peaks in the infrared spectrum or fluctuations in the baseline due to scattering caused by the clouding of KBr pellets (Stuart, 2005). Furthermore, due to the mixing of analyte and KBr this sample preparation technique is destructive.

As seen in Table 1, using Attenuated Total Reflectance (ATR) FTIR seem to be favoured nowadays when investigating food authenticity or adulteration, most likely due to its minimal, sometimes non-existent, sample preparation. In ATR FTIR the sample is pressed against a crystal, usually made from germanium, diamond or zinc selenide (Gauglitz and Vo-Dihn, 2003). The technique operates by measuring the changes that occur in a internally reflected IR beam when the it comes into contact with a sample surface (see Figure 2) (Kazarian and Chan, 2013). When the IR beam travels inside the crystal a standing wave of radiation, called the evanescent wave, is created (Gauglitz and Vo-Dihn, 2003). ATR has in recent years revolutionised solid and liquid sample analyses by FTIR as it combats the most challenging aspects of infrared analyses, namely sample preparation and spectral reproducibility (Kazarian and Chan, 2013), thus increasing the reproducibility of the analyses. This method does not only minimise issues with reproducibility it also allows for a rapid analysis. As with any screening technique, ATR also has its limitations, caused by the optical contact between the sample

and the ATR crystal (Lindenberg *et al.*, 2012). This is of particular concern when solid samples are analysed, and in order to overcome this issue the accessories have devices that clamp the sample to the crystal surface and apply pressure (Lindenberg *et al.*, 2012).

Figure 2. Schematics of an Attenuated total reflectance (ATR) cell.

Multivariate analysis of FTIR results

Two of the most common multivariate analyses employed for the quantification of adulteration is partial least squares (PLS) and principal component regression (PCR). These statistical techniques are normally used for calibration purposes and uses the full spectra region rather than smaller region of pre-specified wave numbers (Tasumi, 2015). Both these methods also use statistically independent covariates from lower dimensions (Granato and Ares, 2014). The main difference between PLS and PCR is that while PCR seeks a high variance distance between the space of the covariates, PLS seeks the space between covariates that will predict the best outcome, using the concentration information (Tasumi, 2015). To establish calibration curves, PLS is therefore more suitable for analysing noisy spectra when the noise is random, whereas PCR is more appropriate for spectra where a systematic noise is observed (Tasumi, 2015).

Another statistical approach, particularly employed for screening purposes, is principal component analysis (PCA) which is an exploratory method (Rios-Corripio *et al.*, 2011; Jaiswal *et al.*, 2015a; Ríos-Reina *et al.*, 2017). The aim of a PCA is to depict as much variability in a data set, such as an IR spectra, using as few variables as possible (Currell, 2015) and a linear combination of the original variables is used to generate new variables (Callao and Ruisánchez, 2018). The output of a PCA can then be visualised in a score plot, where the position of each sample is positioned according to its values

of the principal components (PCs), centralised around zero (Currell, 2015). A PCA approach is typically used to visualise the difference between suspected adulterated foods and pure foods, with the expectation that the two should form separate clusters in the resulting scatter plot. However, a major drawback of PCA is that this method generates PCs in an unsupervised manner (Vidal, Ma and Sastry, 2016). This unsupervised learning means that there is not a specifically predicted target variable, which is commonly used to find general patterns in the data set (Cady, 2017).

There is however, supervised classification techniques which aims to individually assign unknown samples to a predefined class based on sample characteristics (Callao and Ruisánchez, 2018). Examples of classification techniques commonly used for food fraud investigations are linear discriminant analysis (LDA), k nearest neighbours (kNN) and partial least squares discriminant analysis (PLS-DA), see Table 1. These techniques all require at least two defined classes, and will classify unknown samples to the class it most closely resembles based on a set of characteristics (Callao and Ruisánchez, 2018). However, a major drawback of these statistical methods is the need to add known adulterants to authentic foods when building the model, which means that they are not suitable for non-target analyses. Moreover, the detection of outliers within a dataset is more difficult using these methods, as all samples are assigned to a class (Currell, 2015; Callao and Ruisánchez, 2018).

Due to the drawback with both unsupervised and classification methods soft supervised learning models, such as Soft Independent Modeling Class Analogy (SIMCA), has in recent years gained attention for analysing suspected adulteration of food, see Table 1. In a SIMCA, the classification is made based on a set of PCs from training data, such as a calibration set (Salzer and Siesler, 2014). These classes are then applied on test data, where a class is independently assigned to the test data based on the closeness of that data to the classes in a multidimensional space (Salzer and Siesler, 2014; Callao and Ruisánchez, 2018). SIMCA has especially been shown to be useful when comparing known adulterated samples with unknown samples (Santos, Pereira-Filho and Rodriguez-Saona, 2013; Li *et al.*, 2015; Black *et al.*, 2016). As SIMCA only classifies the sample in one class, it is more suitable for cases where the adulterant is not known, and only the adulterated class needs to be

defined (Callao and Ruisánchez, 2018). However, due to this one class assignment the results can sometimes be ambiguous or inconclusive (Callao and Ruisánchez, 2018).

Moreover, some studies using FTIR to detect food adulteration have presented novel statistical tools, such as Continuous Locality Preserving Projections (CLPP) in recent years (Georgouli, Martinez Del Rincon and Koidis, 2017b) and hierarchical models (HM) (Gok *et al.*, 2015; Reis *et al.*, 2017) in recent years, to detect food adulteration based on FTIR data. CLPP can model mixtures of food and adulterant as data series rather than discrete data points, thus enabling a better visualisation of the classification compared to a PCA (Georgouli, Martinez Del Rincon and Koidis, 2017b). Just as PLS or PCR, HM is a type of linear regression model. However, in HM the variables fall into strictly hierarchical levels, also known as nested levels (Kery and Royle, 2016). Although HM has traditionally been used in biology to describe biodiversity (Kery and Royle, 2016), it can be used to classify non-adulterated versus adulterated food and even identify which particular adulterant has been used (Reis *et al.*, 2017). However, to be able to identify the specific adulterant it must be used when building the model, and thus HM is not suitable for non-target analysis.

369 **Table 1.** Summary of studies analysing different foods for adulteration and authenticity using FTIR as an analysis technique.

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Food	Adulterants	Determining adulteration	Sampling technique	FTIR wavelengths	Multivariate model	Comments	Study
Honey	Different Anatolian honey samples	N/A	ATR	Mid-IR (1800–750 cm ⁻¹)	Hierarchical clustering and PCA	Developed method able to differentiate between honey floral source	(Gok <i>et al.</i> , 2015)
	Inverted beet sugar adulteration	Scanned multiple samples of honey with different concentrations of beet invert sugar (0.5 to 25% w/w)	ATR	Mid-IR (950 – 1500 cm ⁻¹)	PLS and PCR	88-94% of the validation set were classified correctly	(Sivakesava and Irudayaraj, 2001)
	Corn syrup, cane sugar syrup	Honey samples from central Mexico were adulterated (0-90%)	ATR	Mid –IR (900–1140 cm ⁻¹)	PCA	Method able to discriminate between honey and cheaper syrup.	(Rios-Corripio <i>et al.</i> , 2011)
Olive oil	Sunflower oil, corn oil, and canola oil	Extra virgin olive oil was adulterated with varying concentrations of palm oil (1.0–50.0% wt./wt)	ATR	Mid –IR (1500–1000 cm ⁻¹)	PLS and PCR	Developed method able to discriminate between extra virgin olive oil and the adulterants tested	(Rohman and Man, 2010)
	N/A	47 samples of extra virgin olive oil from both PDOs and other origins were analysed	ATR	Mid –IR (4000–630 cm ⁻¹) NIR (10,000–4000 cm ⁻¹)	PLS-DA and SIMCA	NIR was found to provide better predictions than MIR, year of harvest were found to impact on spectra	(Bevilacqua <i>et al.</i> , 2012b)
	N/A	Monovarietal extra virgin olive oils from three cultivars (Portuguese)	ATR	Mid –IR (700–740, 950–1050, 1100–1250, 1350–1500, 1700–1800 and 2750–3000 cm ⁻¹)	PLS-DA and PCA	PLS-DA able to discriminate between samples from the three different cultivars	(Gouveinhas <i>et al.</i> , 2015)
	Sunflower, corn, soybean and hazelnut	Extra virgin olive oil samples were adulterated (0%, 5%, 10%, 30%, 50%, 75% and 100%)	KBr discs	Mid –IR (4000–500 cm ⁻¹)	LDA	Adulterant levels down to 5% could be detected using the method developed	(Lerma-García <i>et al.</i> , 2010)
	Hazelnut oil	Four extra virgin olive oil samples with different origins were adulterated with hazelnut oil (1–90%)	ATR	Mid –IR (4000–550 cm ⁻¹)	CLPP	Method developed can be used for screening purposes on even low levels of adulteration (1%). Raman spectroscopy also investigated.	(Georgouli, Martinez Del Rincon and Koidis, 2017b)
	Canola, hazelnut, pomace and high linoleic/oleic sunflower	Extra virgin olive oil was adulterated with varying concentrations of adulterating oil (5–40.0% wt./wt)	ATR	Mid –IR (4000–700 cm ⁻¹)	PLS	Method developed were able to discriminate between olive oil and adulterants and determine level of adulteration	(Maggio <i>et al.</i> , 2010)

Beef	Injecting solutions of non-meat ingredients (NaCl, phosphates, carrageenan, maltodextrin)	Seized meat products from five slaughterhouses were analysed	ATR	Mid-IR (4000 to 525 cm ⁻¹)	Classification PLS-DA models	The addition of addition NaCl to bovine meat causes specific β -sheets vibrations of proteins	(Nunes <i>et al.</i> , 2016b)
	Pork	Analysis of fats from pork and beef mince meat	ATR	Mid-IR (1200–1000 cm ⁻¹)	PLS	Method able to quantify pork traces in beef meatballs	(Rohman <i>et al.</i> , 2011)
	Turkey mince	Minced beef adulterated with turkey meat (5-50% w/w)	ATR	Mid-IR (4000–400 cm ⁻¹)	PCA, LDA, and PLS	Combining information from UV/vis, NIR and MIR spectroscopy improved the confidence in the results	(Alamprese <i>et al.</i> , 2013)
	Horse meat, fat beef trimmings, and textured soy protein	Mixtures of beef minced meat and the adulterants were prepared (2–90% (w/w))	ATR	Mid-IR (4000–650 cm ⁻¹)	PCR and SIMCA	Method able to identify and quantify adulterant	(Meza-Márquez, Gallardo-Velázquez and Osorio-Revilla, 2010)
Saffron	Six characteristic adulterants of plant origin, i.e. <i>C. sativus</i> stamens, calendula, safflower, turmeric, buddleia, and gardenia	Saffron was adulterated with adulterants (5-20% w/w)	KBr discs	Mid-IR (4000–600 cm ⁻¹)	PCA and PLS-DA	Proposed method involved a three-step process for the detection of adulteration as well as for the identification and quantification of adulterants	(Petrakis and Polissiou, 2017)
	Tartrazine, sunset yellow along with propane-1,2-diol, propan-2-ol and acylglycerols	Synthetic commercial sample tested that mimicked the appearance and specific UV-Vis absorbance values of trade standard test	KBr	Mid-IR (4000–400 cm ⁻¹)	Linear regression	Multistep workflow using data acquired from HPLC-UV/Vis, UV-Vis, mid-infrared (FTIR) and nuclear magnetic resonance (NMR)	(Ordoudi <i>et al.</i> , 2017)
	N/A	Saffron from four countries, including from the region of Castilla- La Mancha	KBr discs	Mid-IR (4000–400 cm ⁻¹)	PCA and DA	Method able to correctly classify saffron from different countries in 90-98% of the cases depending on origin	(Anastasaki <i>et al.</i> , 2010)

	<i>Calendula</i> flowers or ground turmeric	Quantification of crocin in the samples	ATR	Mid-IR (1700–900 cm ⁻¹)	Target analysis, PLS	Validated against UV-Vis standard (ISO/TS3632)	(Lee, Htar and Akowuah, 2015)
Milk	Whey, hydrogen peroxide, synthetic urine, urea and synthetic milk	Milk was spiked with each of the adulterant at five different levels	ATR	Mid-IR (1400–1800 cm ⁻¹)	SMCA and PLS	Method able to discriminated samples based on levels of adulteration, and also type of adulterant	(Santos, Pereira-Filho and Rodriguez-Saona, 2013)
	Soymilk	Milk samples were adulterated with soymilk (2–40%)	ATR	Mid-IR (4000–500 cm ⁻¹ and 1680–1058 cm ⁻¹)	PCA	Method able to identify and quantify adulterant in ranges tested	(Jaiswal <i>et al.</i> , 2015b)
	Melamine	Milk samples were adulterated with melamine (0.0625–25% w/w)	ATR	Mid-IR (4000–650 cm ⁻¹)	PLS	Developed method able to distinguish between unadulterated milk samples and that containing 2.5 ppm MEL (threshold)	(Jawaid <i>et al.</i> , 2013)
Butter	Margarine	Butter was mixed with margarine (0–100% w/w)	ATR	Mid-IR (900–1800 and 2800–3040 cm ⁻¹)	PLS	Method able to predict adulteration with high confidence in selected adulteration ranges (0–5%, 0–25% and 20–60%)	(Koca <i>et al.</i> , 2010)
	Cheap animal fats; Lard	Butter adulterated with lard (0.5–80%)	ATR	Mid-IR (4000–650 cm ⁻¹)	PLS	Method determined for rapid identification of adulteration of butter with lard	(Nurulhidayah <i>et al.</i> , 2015)
	N/A	In total, 54 unsalted butter samples from three different regions of Morocco were tested	ATR	Mid-IR (4000–600 cm ⁻¹)	PCA, PLS and PLS-DA	PLS-DA were able to discriminate accurately between butter samples from the three different regions	(Bassbasi <i>et al.</i> , 2014)
Cheese	Formaldehyde	Soft white cheese were spiked with formaldehyde (1–100 mg/100 g)	ATR	Mid-IR (1600–850 cm ⁻¹)	PLS	Method developed suitable for determination of formaldehyde/formalin in cheese samples	(Alkhalf and Mirghani, 2017)
Oregano	Myrtle, olive leaves	Mixtures of oregano and each adulterant were prepared (0–100% w/w)	ATR	Mid-IR (4000–550 cm ⁻¹)	PLS and SIMCA	Liquid Chromatography High Resolution Mass spectrometry (LC-HRMS) to confirm oregano adulteration using biomarkers	(Black <i>et al.</i> , 2016; Wielogorska <i>et al.</i> , 2018)

Paprika and chili powder	Sudan dye	Paprika powder were adulterated (0.02, -0.6% w/w)	ATR	Mid-IR (1400–1800 cm ⁻¹)	HLA/GO (hybrid linear analysis)	Technique determined to be cost-effective in comparison to main alternative technique, LC.	(Lohumi <i>et al.</i> , 2017b)
Onion powder	Starch	Samples were adulterated with starch (1-35% w/w)	ATR	Mid-IR (4000–650 cm ⁻¹), Near-IR (10000-4000 cm ⁻¹)	PLS	FT-NIR data was determined to have a higher predictive value than FTIR	(Lohumi <i>et al.</i> , 2014)
Coffee	Spent coffee grounds, roasted coffee husks, roasted corn, and roasted barley	Coffee adulterated with single or mixed adulterant (33.3-99% coffee in samples)	ATR and diffuse reflectance, DR	Mid-IR (4000–550 cm ⁻¹)	HM and DF	Method able to identifying adulterants in coffee, even in complex mixtures.	(Reis <i>et al.</i> , 2017)
Java tea (<i>Orthosiphon stamineus</i>)	N/A	Samples were tested based on their geographical origin and examined for a characteristic infrared spectrum	KBr discs	Mid-IR (4000–550 cm ⁻¹)	PLS and SIMCA	Sample origin seems to have more dominant effect to the chemical constituent of the plant compared to plant varieties	(Sim <i>et al.</i> , 2004)
Organic wine (Australian)	Non-organic wine (Australian)	Organic and non-organic wines (n=172) from eight different wine varieties, thirteen regions of Australia and vintages ranging from 200-2008 were analysed	Bacchus flow cell	Mid-IR (4000–400 cm ⁻¹)	PCA, discriminant PLS and LDA	DPLS was able to correctly assign 85% of all samples	(Cozzolino <i>et al.</i> , 2009a)
Wine vinegars (PDO)	N/A	In total 67 different wine vinegars belonging to different PDO categories were tested	ATR	Mid-IR (1800–900 cm ⁻¹)	PLS and PCA	Method able to distinguish between different wine vinegars aged different times	(Ríos-Reina <i>et al.</i> , 2017)
Walnut oil	Soybean oil	Four types of walnut oil was adulterated with three types of soybean oil (5– 50%, v/v)	ATR	Mid –IR (4000–650 cm ⁻¹)	PLS and SIMCA	Method could determine adulteration by soybean oil down to 10%	(Li <i>et al.</i> , 2015)
Avocado oil	Sunflower, canola and soybean oils	Mixtures of avocado oil and each adulterant were prepared (range between 2–50%) (v:v)	ATR	Mid-IR (4000–550 cm ⁻¹)	PLS and SIMCA	Developed method gave standard errors of prediction between 0.09 and 2.81 for the adulteration	(Quiñones-Islas <i>et al.</i> , 2013)
Nectars (Grape, orange, peach and passion fruit)	Syrup, apple and cashew juices	Nectars were adulterated according to a complex scheme	ATR	Mid –IR (4000–650 cm ⁻¹)	PLS	The mean relative errors of prediction using developed model varied from 3.0 to 6.7%	(Miaw <i>et al.</i> , 2018)
Jams and juices	Glucose syrup, synthetic flavour	Jams from fruits were prepared using 100% sucrose, 50% sucrose	KBr discs	Mid –IR (4000–400 cm ⁻¹)	Analysis of variance	Developed method able to distinguish adulterated jams	(Mohamed <i>et al.</i> , 2011)

(strawberry and apricot jam, orange, apple and strawberry juices)	and pigment (allura red and sunset yellow)	and 50% glucose syrup and 100% glucose syrup. Juices were prepared from fruits				and juices from non-adulterated ones	
Tea	Talcum powder	210 samples of tea powder with 13 dose levels of talcum powder (0.15-1.40 mg g ⁻¹)	KBr discs	Mid -IR (990–1055 cm ⁻¹)	PCA	Method developed able to detect talcum powder in tea	(Li, Zhang and He, 2016)

370

371 *Abbreviations: PCA: principal component analysis, PLS: partial least squares, PCR: principal component regression, PLS-DA: partial least squares discriminant analysis, SIMCA: Soft*

372 *Independent Modeling Class Analogy, LDA: linear discriminant analysis, CLPP: Continuous Locality Preserving Projection, HM: Hierarchical models and DF: data fusion*

FTIR in combination with other techniques

Several studies have combined FTIR analysis with other analytical chemical techniques such as UV-Vis, near-IR (NIR) or Raman spectroscopy (Bevilacqua *et al.*, 2012b; Alamprese *et al.*, 2013; Lee, Htar and Akowuah, 2015; Georgouli, Martinez Del Rincon and Koidis, 2017a; Ordoudi *et al.*, 2017), liquid chromatography coupled to either UV/Vis (Ordoudi *et al.*, 2017) or high resolution mass spectrometry (LC-HRMS) detectors (Black *et al.*, 2016; Wielogorska *et al.*, 2018) and DNA barcoding (Wielogorska *et al.*, 2018) in order to either compare a newly developed FTIR method with validated or standardised methods or to complement the FTIR analysis. For instance some studies have used FTIR analysis as a part of a spectroscopy test battery (Bevilacqua *et al.*, 2012b; Alamprese *et al.*, 2013; Georgouli, Martinez Del Rincon and Koidis, 2017a). Some of these studies have shown that combining FTIR with other spectroscopy techniques, such as UV-Vis and NIR, can improve the models for predicting adulteration of for example turkey mince in beef (Alamprese *et al.*, 2013). This was recently showcased in a case of saffron adulteration where the synthetic components used as adulterants were tailored to surpass the standard ISO 3632 testing solely relying on UV-Vis detection (Ordoudi *et al.*, 2017). This further emphasises the need to use a combination of techniques in order to detect the increasing and more sophisticated cases of food fraud.

FTIR in combination with other vibrational spectroscopy techniques

Although there are similarities between NIR and MIR there are also important differences which affect the ability to detect some adulterants. In general, the intensity of NIR (NIR, 780-2500 nm) absorbance is much lower than MIR making it more difficult to detect (Sun, 2009). Additionally, specific chemical groups are difficult to associate with a specific IR frequency due to the weak, broad overtone and combination bands associated with NIR absorbance (Sun, 2009). NIR is also not as sensitive as MIR spectroscopy because its radiation will penetrate the sample to a greater extent compared to MIR (Sun, 2009). However, in some studies where onion powder and olive oil were analysed, NIR was found to give better predictions of adulteration than FTIR (Bevilacqua *et al.*, 2012a; Lohumi *et al.*, 2014). This can most likely be explained by the fact that NIR is better suited to analyse highly absorbing bulk samples, such as powders and liquids, due to the lower intensity of the NIR absorbance compared to

MIR. FTIR has also been compared to Raman, specifically when the spectra were analysed using CLPP. For example Georgouli, Martinez Del Rincon and Koidis (2017) found that the prediction accuracy of adulteration of olive oil were similar between FTIR and Raman. One of the advantages of Raman compared to NIR and FTIR is that this type of spectroscopy does not exhibit interference from water, something that otherwise might be an issue when analysing foods without prior sample preparation.

FTIR in combination with hyphenated mass spectrometry (MS) techniques

Although FTIR analysis is a cheap and rapid screening technique that has been applied on many different food matrices, one drawback is that information regarding individual compounds or components in a complex mixture cannot be extracted. Therefore, it often needs to be complemented with other techniques such as hyphenated chromatography, for example LC coupled to high resolution MS (Black *et al.*, 2016; Wielogorska *et al.*, 2018) for confirmatory analyses in order to identify and quantify specific biomarkers, see Table 1. One advantage of combining FTIR with chromatography is that different compounds in a complex biological matrix such as foods, is first separated according to their physicochemical properties (Hird *et al.*, 2014). Hyphenated to mass spectrometry this becomes very a powerful as the mass-to-charge ratio of ions is detected making it possible to provide both quantitative and structural information about the compounds in the sample (de Hoffman and Stroobant, 2007). However, one of the disadvantages of these HRMS methods are that the data analysis is often complex and laborious.

Using LC-HRMS for screening or non-target analyses is also an important technique in other areas of food safety, for instance for the screening of pesticides (García-Reyes *et al.*, 2007; Malato *et al.*, 2011; Gómez-Ramos *et al.*, 2013), detection of contaminants in food packaging materials (Bengtström *et al.*, 2016; Pérez-Ortega *et al.*, 2016; Pieke, Smedsgaard and Granby, 2017) and identification of biomarkers for allergens (Korte, Lepski and Brockmeyer, 2016). With recent advances in non-target analysis of biomarkers in complex biological matrices using HRMS, it is expected that the two-tier approach using FTIR screening and LC-HRMS confirmation suggested by Black *et al.* (2016) and Wielogorska *et al.* (2018) for analyses of adulteration of oregano, will be implemented on a broader scale for analyses of food fraud.

One of the major drawbacks with all analytical chemical techniques is that contamination by any other biological species, such as other species of ground plant material in spices or substituted minced meat, is not easily identifiable by chemical profiles alone. Novel interdisciplinary strategies combining analytical chemistry such as FTIR screening and HRMS identification of biomarkers, with molecular biology, such as that suggested for detecting adulterating oregano with olive and/or myrtle leaves suggested by Wielogorska *et al.* (2018). DNA techniques such as DNA barcoding, real-time polymerase chain reaction (PCR) and Sequence Characterised Amplified Region (SCAR) PCR have been extensively used to detect food adulteration in spices such as oregano (Marieschi *et al.*, 2011b, 2011a), turmeric (Dhanya *et al.*, 2011) and saffron (Marieschi, Torelli and Bruni, 2012; Villa *et al.*, 2016) as well as meat products (Ali *et al.*, 2015; Hou *et al.*, 2015; Amaral *et al.*, 2017).

However, these methods all require access to species specific sequence data in order to design suitable primers to be used in the amplification process, ensuring the amplification of the correct DNA molecule. One approach to remove this limitation is to use next generation sequencing (NGS). However, NGS is an expensive technique that often involves complex workflows, limiting its use in routine analysis (Burns *et al.*, 2016). Although, these molecular methods are generally considered specific and sensitive they are still considered to be more qualitative than quantitative due to their high uncertainty in measurements (Burns *et al.*, 2016). The sensitivity of these techniques means that even low level of accidental contamination can be analysed. However, due to the high level of quantitative uncertainty, even these low-level contaminations can be interpreted as intentional, and thus fraudulent (Galvin-King, Haughey and Elliott, 2018).

Conclusions

The food production chain has become increasingly globalised, making food vulnerable to adulteration. There is therefore a growing demand on both authorities and food producers to develop fast and efficient methods for the comprehensive monitoring of food. Over the last decades, FTIR has proven itself to be a powerful tool for screening of foods for adulteration and authenticity. It is a fast, easy and generally cost effective method to detect food adulteration and in some cases it can also provide information about the geographical origin of certain foods which can be useful for investigating of authenticity. In many cases, FTIR analysis in combination with multivariate data analysis is sufficient to predict the level of adulteration. However, for some purposes, the use of FTIR as part of an extended test battery, such as other spectroscopy techniques, hyphenated chromatographic methods or DNA analysis, is needed to confirm authenticity or quantitative adulteration. Developing test batteries employing several different analysis methods is highly desirable as there are an increasing number of reports where of adulterating agents have been designed to comply with certain standardised analyses. Thus the latest development in the fight against food fraud emphasise the need to use at minimum of two techniques, based on different chemical principles, as is already standard in other areas of forensic science.

Ideally, confirmatory analysis using hyphenated MS techniques or nuclear magnetic resonance (NMR) should be used to identify any biomarkers that could potentially be quantified. For instance, recent developments in tentative identification, i.e. identification without certified reference materials, by HRMS are promising for these purposes and recent studies have shown the large potential of this two-tier approach for screening and confirmation of cases of food fraud. This two-tier strategy should therefore be used to detect fraud of more groups of food, especially for groups where the fraud is suggested to become increasingly sophisticated. Furthermore, recent advances in DNA sequencing, such as technical advances in NGS and more powerful bioinformatics tools for downstream analyses, have the potential to identify adulteration down to the species level without prior sequence knowledge. This is particularly useful in the case of suspected adulteration of mincemeat and ground spices and herbs, as the potential to add virtually any meat or plant material to these foods without chemical detection is large. Information gained from the combined use of analytical chemistry and DNA analyses

475 may even in certain cases be able used to trace the origin of any adulteration within the food supply
476 chain.

477 Food fraud by adulteration or authenticity is by no means a new phenomenon, but recent scandals
478 have illustrated the vulnerability of the modern and increasingly global food supply chain. Developing
479 rapid and easy screening techniques, using for instance FTIR, are therefore imperative to ensure future
480 food safety. With the increasing complexity of food fraud, future methods need to be able combine
481 rapid screening of large quantise of food as well as confirmatory analyses using for instance a
482 combination of detection techniques based on different chemical principles.

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