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Herb and Spice Fraud; the Drivers, Challenges and Detection

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Abstract

The global herb and spice industry, valued at approximately US$4 billion, continues to grow. This industry is continuously under threat from criminals dealing in economically motivated adulteration. Opportunities for criminals to adulterate herbs and spices can occur at any point along the long and complex supply chains. This review looks at the cases and effects of adulteration in the herb and spice industry, and analytical methods being used to detect it and ultimately prevent it. The economy and consumer confidence can be negatively affected following a food fraud scandal. Fraud may also pose a health risk to consumers, even though it is economically motivated, such as the case with nut protein in cumin and paprika. Therefore, for these reasons, rapid screening techniques are required to detect and help prevent fraud from occurring in the industry. Advances in technology has resulted in an increase in the use of spectroscopic techniques being used alongside chemometrics for the detection of adulteration in the herb and spice industry. Also, improvements in DNA analysis and mass spectrometry are providing faster and cheaper methods of adulteration detection. These advancing techniques aim to protect the herb and spice industry and its consumers from fraud by detecting, deterring and therefore preventing adulteration.

1. Introduction: The Herb and Spice Industry

According to the International Trade Centre, the world market for herbs and spices is valued at US$4 billion, and is expected to grow to US$6.5 billion in the near future. The Asia-Pacific region is expected to have the fastest growing market in the world. Over the 2015-2020 period, a Compound Annual Growth Rate of 7% is predicted for this region (CBI, 2016). In the EU, imports of herbs and spices amounted to 533 thousand tonnes, a value of €1.9 billion in 2014, with a slow but steady market growth (CBI, 2015a). Dried herbs and spices are sold mainly in three main markets in the EU; retail, catering/food service, and the largest category that accounts for 50-60% of trade, is food manufacturing (International Trade Centre, 2006).

The consumption of herbs and spices in the EU increased at a rate of 1.7% per year between 2010 and 2013, with the greatest of these consumers in Western Europe. There is an increasing popularity for their use, with ready-made meals, health awareness and food innovation on the rise (CBI 2015a). From a global perspective, the main consumers of herbs and spices are Asian and European; however, the US consumers are also becoming increasingly interested in herbs and spices (AMCHAM and Trade USA, 2015). The supply is not expected to keep up with future demand of herbs and spices worldwide, therefore, prices will rise (CBI, 2016).

The EU produced just 137 thousand tonnes of herbs and spices in 2013; however, it imported over three times this amount. Just 2% of the world's herbs and spices are produced in Europe, 81% in Asia, 12% in Africa, and 3.7% in Latin America and the Caribbean. North America and Oceania produce <0.1% of global production (CBI 2015a). The volume of imports of herbs and spices in the EU grew by 3.8% between 2010 and 2014, even throughout the economic recession. The value of imports increased by 10% per year in the same period, and the volume of imports did not drop when prices rose (CBI, 2015a). In the US, the increasing demand is satisfied with imports also, as it is not traditionally a producing market for herbs and spices (AMCHAM and Trade USA, 2015).

Direct imports from developing countries (according to OECD DAC list) in 2014 amounted to 57% of total EU imports with 302 thousand tonnes or €1 billion imported. China is the largest supplier to the EU (35% of the total imports from outside the EU), followed by India (17%), Vietnam (11%), Indonesia (6.9%), Brazil (5%) and Peru (2.6%) (CBI, 2015a). Asian exporters accounted for 90% of the US imports in 2014 with the leading exporters in descending order being, China, India, Turkey, Spain and Peru (AMCHAM and Trade USA, 2015).

The imported volume of crushed and ground herbs and spices in the EU increased from 23% in 2010 to 31% in 2014. This increase can be due to the desire for ready-meals and easy cooking methods that are becoming more popular with busy lifestyles. The processing of these products allows suppliers in developing countries to add value and increase margins in their products. Asian countries process these products more than other countries. As Asia is the largest global producer and has a large domestic market for its products, it is more capable of investing in processing techniques. EU companies however, still dominate the market for processed herbs and spices (CBI, 2015a).

There is a hesitance to buy such processed herbs and spices, as there is a higher risk of opportunity for adulteration (CBI, 2015a). An important reason for adulteration is economic gain (CBI, 2015b) and the increase in demand for these products, along with the increase in prices cannot be ignored as being a possible motivation for adulteration. The threat of fraud is a concern in the growing herb and spice industry, with valuable products at risk.

2. Food Fraud and Economically Motivated Adulteration

The overall areas of concern with food protection are combined in the model, 'The Food Risk Matrix'. The food risk matrix aids the understanding of the role of food fraud in the context of other food protection issues such as food quality, safety and defence as seen in Figure 1 (Spink and Moyer, 2011). In this study of spices and herbs, food fraud is the area of concern being focused on. As can be seen from the food risk matrix, food fraud is an intentional act for economic gain. This is in contrast to food safety issues and food quality issues, which are unintentional acts that may cause harm, or food defence, which is an intentional act aimed at causing harm (Spink and Moyer, 2011).

Along with the food safety issues such as microbiological, chemical and physical hazards in the food chain (Bouzembrak and Marvin, 2016), there is an increasing need to combat the rising threat of food fraud. Food fraud is a 'collective term used to encompass the deliberate and intentional substitution, addition, tampering, or mispresentation of food, food ingredients, or food packaging; or false or misleading statements made about a product, for economic gain' (Spink and Moyer, 2011). Those who commit the crime usually do not want to cause a public health risk, but want to go unnoticed, and continue with their economic gain. It is also difficult to measure the occurrence of food fraud, as the consumer is unlikely to notice the product they have bought is fraudulent (Johnson, 2014). Food fraud may also continue to occur unnoticed until a public health incident occurs; however, food fraud is never a "victimless crime" (Elliott, 2014). As well as industry, the consumer is the victim as they purchase the food that is not what it claims to be, as with the case of the oregano scandal in 2015 (Black, Haughey, Chevallier, Galvin-King and Elliott, 2016). This was an example of the consumer being deprived of the product (100% oregano) they thought they were buying.

Food fraud is a broad term that encompasses the term 'economically motivated adulteration' (EMA) (Spink and Moyer, 2011). The US Food and Drug Administration (FDA) defined EMA as "the fraudulent, intentional substitution or addition of a substance in a product for the purpose of increasing the apparent value of the product or reducing the cost of its production, i.e. for economic gain" (FDA, 2009). There is more incentive to adulterate more costly food products with cheaper alternatives (Lakshmi, 2012), and as herbs and spices are valuable products, they are at high risk. The act of adulterating food products, although carried out with economic or financial motivation, can have an effect that can often lead unintentionally to a public health threat as a possible added substance may be unconventional (Spink and Moyer, 2011). Adulteration can also negatively affect the food industry and consumer trust (Bo, 2010, Spink and Moyer, 2011).

3. Effects of Food Fraud on the Economy and Consumer Trust

It is not known how common the occurrence of food fraud is, although food fraud is estimated to cost the global food industry US$40 billion dollars per year according to John Spink, (PwC and SSAFE, 2016) and US$10 to 15 billion dollars per year according to Grocery Manufacturers Association (GMA). The cost of one incident to a company can be between 2% and 15% of annual revenue (GMA and Kearney, 2010).

The economic effect of a food fraud scandal can be detrimental to a company and the industry in which it occurs. Many factors need to be considered when accounting for financial loss of a food fraud scandal. These costs can include the cost of a 'product recall or withdrawal, incident investigation, liabilities, lost sales, drop in share price'. These costs are also driven by the 'size of the product footprint, scale of the incident, toxicity of the adulterants, applicable regulations' (GMA and Kearney, 2010). In 2004, the scandal involving Sudan dyes in spices cost US$418 million (GMA and Kearney, 2010).

Current costs for companies includes conducting a food fraud vulnerability assessment plan (PwC and SSAFE, 2016). A single food fraud scandal can cause long-term industry wide losses, destroy valuable brands, close export markets and damage trust in public institutions. Significant investment is required to obtain effective strategies for supply chain risk. Addressing and preventing the food fraud risks aids economic growth, the movement of food through supply chains, and consumer confidence (PwC and SSAFE, 2016).

De Jonge et al. (2004) defines consumer confidence “as the consumers’ general expectation that food products will not cause any harm to their health or to the environment”. Evidence of good communication and risk management improves consumer trust (de Jonge, Frewer, van Trijp, Ja Renes, de Wit and Trimmers, 2004). An increase in food safety issues has reduced consumers’ confidence in the food industry (Grunert, 2002).

Consumers want improved traceability, clear and correct labelling, shorter supply chains, use of local ingredients, more attention to personal communication and reassurance, and information about the origin of products (Barnett et al., 2016). There are regulatory bodies in place to control the risks of fraud and to protect the consumer from being a victim of food fraud.

4. EU and US Regulations to Control Fraud in the Herb and Spice Industry

In the General Food Law Regulation (EC) 178/2002 (EU, 2002), the general principles and requirements of food law and procedures of food safety are outlined. With regard to the consumer’s interest, the General Food law aims to prevent, “fraudulent or deceptive practices, the adulteration of food, and any other practices which may mislead the consumer”. The European Food Safety Authority (EFSA) was established legally in 2002 under the General Food Law, following a number of food crises in the late 1990s. EFSA provides scientific advice and communicates risks within the food chain.
In the United States, the FDA and the US Department of Agriculture (USDA) are the principle federal agencies working on food safety. Border protection and import authorities, as well as food safety, food defence and food quality authorities broadly look after food fraud across a number of federal agencies (Johnson, 2014). The primary food safety law administered by the FDA is the Federal Food, Drug and Cosmetic Act (FFDCA) (FDA, 1938). This act tightened control over food, drugs, and consumer protection, and gave the government enforcement ability. The Food Safety Modernization Act was then passed by US

congress (FDA, 2011). This Act amended Section 415 of the FFDCA with the aim to prevent rather than respond to contamination and outbreaks.

Specific organisations have become involved in the protection of the herb and spice industry. The European Spice Association (ESA) is a non-profit organisation made up of national federations of the spice industry from the EU, Turkey and Switzerland. It has an aim to protect the industry and its members with regard to processing, packaging, quality assurance, food safety and marketing in the herb and spice industry. The American Spice Trade Association (ASTA) works similarly in the US, to ensure clean and safe spices, and enhance the industry and the business interests of its members. The ESA has a set maximum level of 2% w/w extraneous matter in herbs and 1% w/w maximum level in spices in the Quality Minima Document (ESA, 2015) whereas the ASTA has set a level of extraneous matter at 0.5-1% w/w (ASTA, 2011a). One of the difficulties in keeping the herb and spice industry free from fraud, is the issue of long industry supply chains that can exist over many countries.

5. Herb and Spice Industry Supply Chains

Supply chains in the herb and spice industry tend to be long, complex and can pass through many countries. Such complexities present many opportunities for criminals to carry out EMA. The stages of the supply chain can include grower, collector, primary processor, local traders, secondary processor, exporter, importer, trader, processor/packager, food manufacturer/retailer/wholesaler, and finally the consumer (Figure 2). At any stage of this supply chain, a number of fraud opportunities can occur including misrepresentation, adulteration and substitution (BRC-FDF-SSA, 2016).

“Fraud control measures” can be implemented in companies to detect fraud opportunities or motivations that may occur either internally, or externally of the company (PwC and SSAFE, 2016).The processing and manufacturing needs to be carefully monitored to ensure food protection. Cleanliness and protection of the product from contamination and adulteration is vital. The cost of maintaining these standards can be high. The blending and packaging stage provides an early opportunity for adulteration and needs to be carefully monitored. In more modern processing plants, the product is often enclosed during this process. In addition, careful monitoring is required for the preparation of ready meals i.e. precooked meals, and other food products that have herbs and spices added to them towards the end of the supply chain.

The ESA Adulteration Awareness Document (ESA, 2014) advises companies on ways to prevent adulteration: 1. “Evaluation of the supply chain” (knowing the history of the supply chain, adherence to legal requirements, traceability, adherence to HACCP (Hazard Analysis and Critical Control Points) and adherence to accreditation standards), 2. “The nature of the material” (whole or ground, botanical species and commercial grade), 3. “Product testing” (there is a range of methods being developed for the rapid and accurate detection of fraud). It is important to have these precautions in place for both industry and the consumer, however, cases of adulteration continue to occur, and there may be useful lessons in reviewing old examples of adulteration.

6. Economically Motivated Adulteration in the Herb and Spice Industry

A large global industry such as the herb and spice sector is under constant threat from fraudsters. With valuable condiments such as saffron, oregano, vanilla, turmeric and paprika, substantial amounts of money can be made by carrying out adulteration of these products at the expense of the consumer and potentially the reputation of food businesses. The long, complex supply chains and the increase in crushed and ground herbs and spices provide excellent opportunities for EMA. However, other vulnerabilities that may affect the chances of adulteration include seasonality and availability of the crop, weather events, cultural and geo-political events, economic indicators, food safety laws, prevalence of corruption and advances in technology to mask fraud (BRC-FDF-SSA, 2016). The 2016 garlic crop had potential to become vulnerable to adulteration following severe weather events of heavy rain and snow in late 2015, causing a surge in the price of garlic. (Terazono, Li and Hornby, 2016). This surge in the price caused stockpiling of garlic. Circumstances such as these can all provide motivation for adulteration. Preventative measures can include; knowing product specification, supplier assurance, product type (ground and crushed and where did this process take place), knowing the supply market and being aware of vulnerabilities in the supply chain. Verification and testing can be carried out to confirm the preventative measures are effective. This can involve devising representative sampling and inspection programmes for products, a suitable testing strategy that meets objectives, a test method in an accredited laboratory, and supply chain verification measures which may include pre-delivery of samples prior to purchase for approval, or evidence of authenticity from an accredited laboratory (BRC-FDF-SSA, 2016). The prevention of fraud is not in detecting each individual fraud and controlling one type, but reducing the vulnerabilities, as the fraudsters are always evolving and looking for their next crime (Spink and Moyer, 2013). The herb and spice industry has been a victim of EMA on numerous occasions. Table 1 focuses on examples where substitution adulteration occurred with various herbs and spices.

Table 1: Examples of Substitution Adulteration in the Herb and Spice Industry

Ingredient	**Adulterant**	**Reference**
Chilli	Oil, rice flour, bran	(The Express Tribune, 2016)
	Ziziphus nummularia fruits	(Dhanya, Syamkumar, Siju and Sasikumar, 2011a)
	Plant husks, rice powder, sawdust, stone powder	(The Hindu, 2008)
Oregano	Sumac, olive leaves	(Choice Magazine, 2016)
	Olive leaves, myrtle leaves	(Black, Haughey, Chevallier, Galvin-King and Elliott, 2016)
	Satureja montana L. and *Origanum majorana* L.	(Marieschi, Torelli, Bianchi and Bruni, 2011a)
	Cistus incanus L., *Rubus caesius L.* and *Rhus coriaria* L	(Marieschi, Torelli, Poli, Bianchi and Bruni, 2010)
Cumin	Almond, peanut, tree nuts, peach and cherry	(Garber et al., 2016)
	Fennel seeds	(John, 2012)
	Peanut shell	(Agres, 2015)
Black pepper	Chilli	(Parvathy, Swetha, Sheeja, Leela, Chempakam and Sasikumar, 2014)
	Buckwheat or millet	(ASTA, 2011b)
	Papaya	(Lakshmi, 2012)
Cinnamon	Coffee husk	(ASTA, 2011b)
Chinese star anise	Japanese star anise	(Perret, Tabin, Marcoz, Llor and Cheseaux, 2011)
Nutmeg	Coffee husks	(ASTA, 2011b)
Paprika	Almond	(Whitworth, 2015)
	White pepper, curcuma, barium sulphate, brick powder	(Lead Action News, 1995)
	Defatted paprika	(ASTA, 2011b)
	Paprika of inferior quality substituting paprika from the Protected Designation of Origin (PDO) 'La Vera' region.	(Hernandez, Martin, Aranda, Bartolome and Cordoba, 2007)
	Falsely declared Szegedi paprika substituted for *Szegedi Füszerpaprika* PDO	(Brunner, Katona, Stefanka and Prohaska, 2010)
Saffron	Saffron of unknown origin labelled as being cultivated in the PDO region in Spain can be used for substitution.	(Rubert, Lacina, Zachariasova and Hajslova, 2016)
	Beet, pomegranate fibres, dyed corn stigmas, red dyed silk fibres, safflower, marigold to red stigma	(Heidarbeigi, Mohtasebi, Foroughirad, Ghasemi-Varnamkhasti, Rafiee and Rezaei, 2015)
	Safflower, gardenia, meat fibres, gelatine fibres, curcuma, sandalwood, campeche wood powder, stigmas of other saffron types, flowers, starch, glucose	(Soffritti et al., 2016,Saffron in Europe-White Book,)
Turmeric	*Curcuma zedoaria, Curcuma malabarica*	(Dhanya, Syamkumar, Siju and Sasikumar, 2011b)
	Chalk powder	(Nallappan, Dash, Ray and Pesala, 2013)

The addition adulteration of colour to spices to improve their value is a common occurrence. Colour can influence the perception of food and stimulate appetite, therefore, increase the value of a product (Downham and Collins, 2000). The addition of colourants to foodstuffs dates back to at least 1500 BCE, and up until the middle of the 19th century, ingredients such as the spice saffron was added for a decorative effect in certain foodstuffs (Downham and Collins, 2000). Natural dyes were commonly used in food around this time, however, as the 1900s began, the use of synthetic dyes became the colouring of choice with ease of production, less expense and superior colouring ability (Downham and Collins, 2000).

As with other types of food adulteration, there is a likelihood that certain synthetic dyes may be a threat to public health, and historical records show that injuries and even death occurred following ingestion of toxic colourants (Downham and Collins, 2000). Allergic and asthmatic reactions as well as DNA damage have also been reported (Gray et al., 2016). Therefore, the use of most synthetic dyes is forbidden in Europe (Gray et al., 2016). The two main types of dyes that may be illegally added to food include azo dyes and triphenylmethanes (EFSA, 2005). Examples of these illegal azo dyes include Sudan I, II, III, IV, para red, orange II, methyl yellow and rhodamine B. Malachite green and its metabolite leucomalachite green are examples of triphenylmethane dyes considered genotoxic and/or carcinogenic (EFSA, 2005).

In May 2003, Sudan 1 was found to be illegally present in chilli powder and foods containing chilli powder in the EU (EFSA, 2005). Following this event, in 2005 and 2006, numerous tests were carried out for the presence of illegal dyes by the UK Food Standards Agency (FSA) (Oplatowska-Stachowiak and Elliott, 2017). Regulatory legislation was put in place following the scandal, and member states were required to monitor high risk products and provide analytical reports for the presence or absence of Sudan dyes as an emergency measure in the European Commission Decision 2005/402/EC (EU, 2005). This legislation was later repealed in the European Commission Regulation (EC) No. 669/2009 (EU, 2009) to a less intensive testing regime due to a reduction in the presence of Sudan dyes.

Legislation varies in different countries, which can cause problems for importers and exporters (Oplatowska-Stachowiak and Elliott, 2017). In the EU, Regulation (EC) No. 1333/2008 (EU, 2008) on food additives was developed "…with a view to… ensuring a high level of protection of human health and a high level of consumer protection …." With regard to food colours, there are currently 25 natural, and 15 synthetic dyes on Annex II of this regulation that can be allowed in food (Oplatowska-Stachowiak and Elliott, 2017). The US

FDA regulates food additives in the US. To indicate the variation between countries, three synthetic dyes approved in the US are not approved in the EU, and nine synthetic food colours in the EU are not approved in the US (Oplatowska-Stachowiak and Elliott, 2017). There is still a continued risk of adulteration with dyes in spices.

The results in Table 2 summarises reported cases of adulteration of spices with dyes from 2013 to 2017 in the US. In this work the most common dyes reported were Sudan 1 and Sudan 4. These results indicate that adulteration with dyes is ongoing. Continued surveillance of spices to detect and prevent adulteration with dyes is vital to the herb and spice industry as well as the safety of consumers. Health risks can occur alongside both substitution and addition adulteration. They can cause more than an economic threat to the consumer.

Table 2 Adulteration with Dyes as reported by Tarantelli and Sheridan (2014) and by Tarantelli (2017).

Spice	**Adulteration**
Red Pepper Chili powder	Sudan 1, Sudan 4, Metanil Yellow, Sudan 3, Oil Orange SS, Rhodamine B, Auramine O, Orange II, Dimethyl Yellow, Fast Garnet GBC, Malachite Green, Allura Red
Paprika powder	Sudan 1, Sudan 4, Acid Black 1, Orange II, Annatto
Turmeric powder	Sudan 1, Mentanil Yellow, Orange II, Lead Chromate
Sumac	Amaranth Red, Basic Red 46
Curry powder	Auramine O, Chrysoidin (Basic Orange II)
Saffron flower	Acid Orange II, Mentanil Yellow, Sudan I, Ponceau 4R, Ponceau 6R
Cayenne pepper	Crystal Violet
Five spice powder	Auramine O

7. Public Health Risks and Impact Due to Economically Motivated Adulteration

The main motivation for the addition to, or substitution of the authentic product is for economic reasons, however, with the cases outlined in Table 3, a number of health risks were a detrimental result of this criminal behaviour. There is an increasing concern over the introduction of hazards from food fraud. It is a constant and growing concern in the food industry, with greater actions needed to be put in place to detect it.

There are three types of food fraud risks that pose a threat to the public: 1. Direct: The consumer is put at immediate risk from a short-term exposure leading to acute toxicity or lethality, 2. Indirect: The consumer is put at risk over long-term exposure with potential chronic effects, 3. Technical: Food documentation may not be representative of the food content (Spink and Moyer, 2011). A serious example of a technical fraud risk could be an allergic reaction to an unknown product that has not been outlined in the label.

The detection of undeclared nut protein in cumin and paprika in 2015 was one case where adulteration did not result in just economic losses (Garber et al., 2016). This crime had serious consequences for public health and strengthened the demand for food protection. With food allergies affecting approximately 3-4% of the adult population, an estimated 0.6% are allergic to peanut and 0.5% allergic to tree nut (Sicherer and Sampson, 2006). All products that come into contact with nut protein need to be labelled accurately as the risk of an unsuspecting sensitive individual coming into contact with this can be fatal. In a study by Bock, Muñoz-Furlong, and Sampson (2001), it was found that out of 32 fatal cases of anaphylaxis from 1994-1999, 94% of the cases were caused by peanut or tree nuts, indicating that the vast majority of food induced anaphylaxis is caused by these foodstuffs. The adulteration of spices with nuts is a serious public health risk for susceptible individuals.

Chinese star anise (*Illicium verum*) is infused in teas to relieve the symptoms of colic in children. The adulteration of Chinese star anise with Japanese star anise (*Illicium anisatum*) has in previous years resulted in the intoxication of children. Japanese star anise looks similar to Chinese star anise, and they are often even more difficult to distinguish as they can be sold in broken or ground form. Therefore, chemical analysis is required to distinguish them. Japanese star anise contains neurotoxins and can result in a child having neurological and gastrointestinal problems (Perret, Tabin, Marcoz, Llor and Cheseaux, 2011).

Papaya seeds have been used to adulterate and bulk black pepper. However, these papaya seeds can cause liver and stomach problems, and therefore pose a health risk to the unsuspecting consumer (Lakshmi, 2012).

Turmeric can contain various adulterants that threaten public health. Yellow chalk powder has been used to add bulk to turmeric as it is a cheap material (Nallappan, Dash, Ray and Pesala, 2013, Food Safety and Standards Authority of India, 2012). This adulterated product however can cause swelling of the face, loss of appetite, nausea and vomiting (Nallappan, Dash, Ray and Pesala, 2013). *Curcuma zedoaria* can be used to adulterate turmeric (Dhanya, Syamkumar, Siju and Sasikumar, 2011b), and was found to have toxic effects in rats and chickens by Latif et al. (1979) if not processed properly. Lead chromate added to turmeric was used as a dye as well as a bulking powder. Over exposure to lead can cause delayed mental and physical development (Food Safety News, 2016).

In a case reported in the Times of India (John, 2012), poor grade fennel seeds were coated with waste marble dust and dye, and mixed in with the cumin product. In this case, it was the treatment of the fraudulent product that caused the public health risk rather, than the fennel seeds themselves.

The use of other plant cuttings such as olive leaves in the adulteration of oregano (Black, Haughey, Chevallier, Galvin-King and Elliott, 2016) can also pose a health risk to the consumer. As these leaves are not produced for consumption, it is unknown how these cuttings may be treated. In the case of olive leaves in particular, evidence of pesticides can be found (Elliott, C- personal communication). Pesticide residues pose a health risk, and hazards such as toxicity, carcinogenicity and mutagenicity are associated with them (WHO, 2010).

There are many possible risks with food adulteration. Therefore, it is vital that there is adequate policing of the supply chains and the food industry to deter and try to prevent any fraud before it is too late.

Table 3: Examples of Economically Motivated Adulteration with Possible Health Impact

Herb/Spice	**Adulterant**	**Possible Health Impact**	**Reference**	**Type of Food Fraud Risk**
Cumin, Paprika	Nut protein	Anaphylaxis	(Sicherer and Sampson, 2006,Garber et al., 2016)	Direct
Chinese star anise	Japanese star anise	Neurological and gastrointestinal problems	(Perret, Tabin, Marcoz, Llor and Cheseaux, 2011)	Direct
Black pepper	Papaya seeds	Liver and stomach problems	(Lakshmi, 2012)	Direct
Turmeric	Yellow chalk powder	Face swelling, loss of appetite, nausea, and vomiting	(Nallappan, Dash, Ray and Pesala, 2013)	Direct
	Curcuma zedoaria	Toxicity in rats and chickens	(Latif, Morris, Miah, Hewitt and Ford, 1979)	Direct
	Lead chromate	Delayed mental and physical development	(Food Safety News, 2016)	Indirect
Cumin	Fennel seeds coated with marble dust and dye	Possible health risk from the use of dye and marble dust	(John, 2012)	Indirect
Oregano	Olive leaves	Presence of pesticides-Toxicity, carcinogenicity, mutagenicity	(WHO, 2010)	Indirect

Illegal dyes are a constant threat to the international food industry and are found intermittently, as indicated by the alerts in Rapid Alert System for Food and Feed (RASFF). Examples from RASFF and the possible health impacts can be seen in Table 4.

It is vital that authentication testing is carried out to detect cases of economic fraud and to verify that preventative measures are effectively in place (BRC-FDF-SSA, 2016). This prevention not only maintains quality and consumer trust, but also helps to prevent the possibility of public health risk (Lohumi, Lee, Lee and Cho, 2015).

346 Table 4 The Possible Health Impacts of Common Illegal Dyes

Common Illegal Dyes	Possible Health Impact	Examples of Spices
Sudan 1	Genotoxic and carcinogenic in rats	Cayenne pepper, Turmeric, Chilli, Paprika, Curry
Sudan 4	Potentially genotoxic and possibly carcinogenic	Curry, Turmeric, Chilli, Paprika, Sumac
Para Red	Potentially genotoxic and possibly carcinogenic	Chilli, Cayenne pepper, Paprika
Orange II	Potentially genotoxic, insufficient data on carcinogenicity	Chilli, Safflower, Sumac, Paprika
Methyl Yellow	Possibly carcinogenic to humans (IARC, 1975)	Curry
Rhodamine B	Potentially genotoxic and potentially carcinogenic	Sumac, Chilli, Paprika, Turmeric, Curry

(RASFF portal, EFSA, 2005)

8. Analytical Methods for the Detection of Adulteration in Herbs and Spices

Fast, reliable and competent analytical techniques are what is required to confirm the authenticity of food with this increasing trend of food adulteration (Lohumi, Lee, Lee and Cho, 2015).

According to the database records collected by Moore, Spink and Lipp, (2012) from 1980 to 2010, the top two methods used for detecting food adulteration were liquid-chromatography and infrared spectroscopy. Visual inspection and microscopy are common methods used to detect adulteration in herbs and spices as reported by the British Retail Consortium, the Food and Drink Federation, and the Seasoning and Spice Association in 'Guidance on Authenticity of Herbs and Spices' (BRC-FDF-SSA, 2016). However, it requires highly trained analysts and analysis can take a long time, therefore research is continuously being carried out to develop new methods for the detection of adulteration in herbs and spices. Fraudsters tend to be one-step ahead of the food safety agencies but also, techniques for food adulteration are becoming more and more advanced (Lakshmi, 2012). Recent analytical methods for the detection of adulterants are listed in Table 5.

8.1 DNA Analysis

DNA analysis is increasingly being used in the fight against food fraud as advances in methods provide cheaper, more efficient and accurate means of detection of fraud. It can be seen from Table 5 that DNA analysis plays an important role in the detection of substitution adulteration in herbs and spices. In recent years, Sequence Characterised Amplified Region-Polymerase Chain Reaction (SCAR-PCR) and DNA barcoding are becoming desirable methods for the detection of food adulteration.

SCAR-PCR is an advancement on the use of Random Amplified Polymorphic DNA (RAPD) markers in DNA analysis. RAPD analysis is considered a useful starting point as it has low operating cost and can distinguish between botanical varieties (Marieschi, Torelli and Bruni, 2012, Marieschi, Torelli, Poli, Sacchetti and Bruni, 2009). Although RAPD markers are a fast and cheap method, their downfall is that repeatability is low and exchanging results between laboratories creates difficulties (Babaei, Talebi and Bahar, 2014). This problem with RAPD markers was corrected with the development of SCAR primers and this increased specificity and reliability (Paran and Michelmore, 1993). The use of SCAR-PCR was observed by

Marieschi, Torelli & Bruni (2012) for the detection of bulking agents in saffron, where, the method screened large batches with a fast, reliable sensitive and low cost screening method. The detection of adulteration of oregano with *Cistus incanus* L., *Rubus caesius* L., and *Rhus coriaria* L., was carried out by Marieschi et al. using RAPD (2009) and subsequently with SCAR-PCR (2010) to improve the robustness of the method.

Other SCAR-PCR methods include the detection of olive leaves, *Satureja montana* L., and *Origanum majoranan* L. in oregano (Marieschi, Torelli, Bianchi and Bruni, 2011a,Marieschi, Torelli, Bianchi and Bruni, 2011b) , the presence of *Curcuma zeodoaria*/*Curcuma malabarica* in turmeric (Dhanya, Syamkumar, Siju and Sasikumar, 2011b) and the presence of plant based materials in chilli (Dhanya, Syamkumar, Siju and Sasikumar, 2011a). The development of a SCAR and Internal Transcriber Spacer (ITS) region multiplex PCR method allowed the detection of both the adulterant safflower and the spice saffron in the one analysis (Babaei, Talebi and Bahar, 2014). It is evident that the use of SCAR-PCR has potential for EMA adulteration detection in a number of herbs and spices. SCAR-PCR is a sensitive method with detection limits at 1% for the adulteration of oregano with *Cistus incanus* L., *Rubus caesius* L., and *Rhus coriaria* L. (Marieschi, Torelli, Poli, Bianchi and Bruni, 2010), 1% for the detection of olive leaves in oregano (Marieschi, Torelli, Bianchi and Bruni, 2011b) and a limit of detection (LOD) of 10g/kg for the presence of *Curcuma zeodoaria*/*Curcuma malabarica* in turmeric (Dhanya, Syamkumar, Siju and Sasikumar, 2011b) indicate this. However, a limitation of SCAR-PCR is the need for sequence data for the PCR primers design (Ganie, Upadhyay, Das and Prasad Sharma, 2015).

DNA barcoding is a relatively new method that was firstly developed by Hebert et al. (2003). It is based on the variability within a standard region of the genome, the 'DNA barcode' (Hebert, Cywinska, Ball and deWaard, 2003). It has become increasingly used since its development, and there is successful evidence of this method in the detection of adulterants in herbs and spices. This method has been used for the detection of adulterants in saffron (Huang, Li, Liu and Long, 2015, Jiang, Cao, Yuan, Chen, Jin and Huang, 2014), and chilli adulteration in black pepper (Parvathy, Swetha, Sheeja, Leela, Chempakam and Sasikumar, 2014). DNA barcoding is a fast, reliable sensitive method for a wide range of food commodities, and even strongly processed foods (Galimberti et al., 2013). There is also the possibility of building reference databases to improve the chances of it becoming a routine test for food quality, and traceability (Galimberti et al., 2013).

DNA purity and integrity are concerning with regard to DNA barcodes, which, can be a limitation of the test. Poor quality DNA may reduce amplification success of DNA barcodes. (Huang, Li, Liu and Long, 2015). DNA barcoding also relies on the availability of sequence libraries to reference against (Ellis, Muhamadali, Allen, Elliott and Goodacre, 2016).

Whole genome sequencing is becoming a possibility and it has potential for the detection of food adulteration with Next Generation Sequencing (NGS). However, so far, little work in this area has been carried out with the complex work flow and high costs associated with this method (Burns et al., 2016).

The methods for the detection of adulteration in herbs and spices using DNA analysis described are qualitative. Quantitative methods often result in high measurement uncertainty, although advancements in PCR technologies are improving in this way (Burns et al., 2016). Overall, the limitations with DNA analysis may include poor integrity and purity of the DNA, poor efficiency of the extraction, and the risk of contamination is a concern with these methods (Burns et al., 2016). Also, low level accidental contamination can be misinterpreted as intentional substitution.

8.2 Mass Spectrometry

Mass Spectrometry (MS) is a powerful tool in the fight against food fraud, and in many industries, it is considered the gold standard technique. Methods include Gas Chromatography (GC-MS), Liquid Chromatography (LC-MS), Isotope Ratio (IR-MS) and Inductively Coupled Plasma (ICP-MS). Once a targeted method is developed, mass spectrometry can provide a highly specific and sensitive technique that can quantify known analytes to sub-μg concentrations (Ellis, Muhamadali, Haughey, Elliott and Goodacre, 2015). Although an expensive technique that requires significant expertise and laboratory surroundings, it is highly regarded as a confirmatory technique.

In the study by Black et al. (2016), Liquid Chromatography coupled to High Resolution Mass Spectrometry (LC-HRMS) was used as part of a two-tier approach to detect the presence of adulterants in oregano with LC-HRMS used as a confirmatory technique. The analysis was untargeted, and with the use of Principal Component Analysis (PCA) and Orthogonal Partial Least Squares- Discriminant Analysis (OPLS-DA) chemometrics, biomarkers specific to the classes (oregano and various adulterants) were identified. The identification of such biomarkers allowed further developments in the detection of adulteration with targeted mass spectrometry (Wielogorska et al., 2018). Wielogorska et al. used targeted FTIR (Fourier

Transform Infrared) and LC-MS/MS to quantitatively detect adulteration in oregano. The studies by Black et al. (2016) and Wielogorska et al. (2018) were an improvement on the work of Bononi and Tateo (2011) as they identified biomarkers for a number of adulterants, as well the development of a quantitative method. In the work by Bononi and Tateo, a targeted method was developed for the detection of a characteristic marker of olive leaves, the phenolic compound oleuropein, in both oregano and sage with the use of Liquid Chromatography-Electrospray Ionization Mass Spectrometry (LC-ESI-MS/MS). This compound oleuropein was later found to be also present in myrtle leaves by Wielogorska et al. (2018). Similarly, the use of untargeted Ultra High Performance Liquid Chromatography coupled to High Resolution Mass Spectrometry (UHPLC-HRMS) merged with chemometrics, OPLS-DA proved to be a successful powerful tool in determining products from the PDO of saffron (Rubert, Lacina, Zachariasova and Hajslova, 2016). Falsely declared saffron from a PDO can be used in substitution of the authentic product.

GC-MS is another method that has been used to detect possible adulterants. This was the case with the study carried out by Ma et al. (2015) when investigating detection methods for known fruit adulterants in fennel seed. Essential oils of fennel seed and two adulterants were profiled, and distinct differences between fennel seed and two of its adulterants were observed. Bononi, Fiordalise and Tateo (2010) were able to use GC-MS to detect olive leaves in oregano and sage by using GC-MS with a detection limit of 1%. The benefits of this method included the ease of use and reproducibility of the results. However, with regard to the detection of adulteration in herbs and spices, an issue that may occur with the use of GC-MS is that, only the volatile oils are investigated. Therefore, the addition of volatile oils to a product may cheat the GC-MS adulteration detection method.

ICP-MS along with PCA and Canonical Discriminant Analysis (CDA) was the method used by Brunner et al. (2010) to detect falsely declared Szegdi paprika (PDO). The Sr isotopic composition and the multi-elemental analysis is indicative of paprika from the region.

Upgrades in mass spectrometry involve the use of real time analysis of samples by directly introducing the samples to the mass spectrometer. Ambient mass spectrometry is a relatively new analytical technique that gives comparable results to conventional techniques without complex sample preparation (Black, Chevallier and Elliott, 2016). Examples of its use include the detection of the adulterant Japanese star anise in Chinese star anise using Direct Analysis Real Time-High Resolution Mass Spectrometry (DART-HRMS) (Shen, van Beek, Claassen, Zuilhof, Chen and Nielen, 2012) by detecting the presence of anisatin. Advances on this method involves the use of direct plant spray combined with orbitrap-HRMS (Schrage et al., 2013). This method can detect between the neurotoxic Japanese star anise and the Chinese star anise in seconds, and without sample pre-treatment. DART ionisation has slightly higher selectivity, no solvents added and the absence of high voltages when compared to direct plant spray. The benefits of direct plant spray over DART ionisation include the low cost, lower standard deviations and simplicity. Direct plant spray and DART ionisation techniques are more successful qualitative methods than quantitative methods (Schrage et al., 2013).

Currently the disadvantages of mass spectrometry in comparison to spectroscopy is the cost and the requirement of a laboratory setting and highly trained analysts. However, advances to overcome this are ongoing with aims to miniaturize the instrumentation, and for the data to be presented so that it is easily interpreted. However, these developments require further optimization and are not readily available (Ellis, Muhamadali, Haughey, Elliott and Goodacre, 2015). Similarly to spectroscopy, the validation procedure for non-targeted methods in mass spectrometry have not been standardised. This can reduce consistency between laboratories.

<u>8.3 Spectroscopy</u>

Vibrational spectroscopies, along with chemometrics, have become well known as rapid, non-destructive, fingerprinting techniques and are valuable screening tools in the detection of adulteration/authentication in the food industry. A range of spectroscopic analytical techniques used in the food industry include FTIR, Fourier Transform Near infrared (FT-NIR), Raman, Hyperspectral Imaging (HSI) (Lohumi, Lee, Lee and Cho, 2015) and Nuclear Magnetic Resonance (NMR) (Petrakis, Cagliani, Polissiou and Consonni, 2015).

In the detection of adulteration of herbs and spices for economic gain, a number of spectroscopic methods continue to be developed. Work has been carried out to develop competent models to detect cornstarch in garlic powder by FTIR (Lohumi, Lee and Cho, 2015) and onion powder by FTIR and NIR (Lohumi et al., 2014). Raman has also been used to detect cornstarch in onion powder and garlic or ginger powder (Lee et al., 2015, Lee, Lohumi, Cho, 김문성 and 이수희, 2014). Starch may be added to white powders such as garlic and onion powder to add bulk to the product. In these studies, a quantitative model was built using the algorithm Partial Least Squares Regression (PLSR) in chemometrics. The

Raman, FTIR and NIR spectral data based models described here are capable of detecting adulteration in onion powder, garlic and ginger with starch up to 35%.

In a study by Black et al. (2016) on the detection of adulteration in oregano, FTIR was used alongside the confirmatory technique LC-HRMS. Following the identification of biomarkers for both oregano and its adulterants, and the development of spectroscopic classification models using the unsupervised PCA and supervised OPLS-DA chemometric algorithms, a rapid screening method and confirmatory method was developed. The benefit of this method was that a number of different adulterants could be added to the database that was used to build the model. The developed screening technique therefore was robust and could identify numerous adulterants at each screening in the survey that was subsequently carried out. The results of the survey indicated that adulteration was ongoing, but also, it displayed the use of a rapid screening technique to help the fight against food fraud. Further development on these analytical techniques was carried out by Wielogorska et al. (2018) with the development of targeted quantitative methods using FTIR with PLSR and LC-MS/MS for the detection of adulteration in oregano.

Raman and FTIR methods analyse the sample in the mid infrared region of the electromagnetic spectrum. The spectral data consist of sharp bands representing inelastic scattering, or information on the fundamental vibrations of the sample respectively. This is in comparison to the vibrational overtones and combination peaks of the NIR, which does not provide as much information (Ellis, Muhamadali, Haughey, Elliott and Goodacre, 2015). However, in the detection of starch in onion powder, NIR with PLSR chemometric algorithm was determined the most suitable method by Lohumi et al. (2014). NIR has the ability to penetrate deeper into the sample and therefore is more suitable for bulk samples that have little or no sample preparation (Lohumi et al., 2014). Raman has advantages over NIR and FTIR as it is not affected by water, and inorganic materials can be analysed more easily. Analysis through packaging or glass is also a possibility (Lee et al., 2015). Recent improvements to Raman also include the use of Surface Enhanced Raman Scattering (SERS) and Spatially Offset Raman Spectroscopy (SORS) which has shown its ability to detect counterfeit products through packaging (Ellis, Muhamadali, Haughey, Elliott and Goodacre, 2015).

The use of Proton Nuclear Magnetic Resonance (^{1}H-NMR) combined with chemometrics (PCA, OPLS-DA, O2PLS-DA) was investigated and was proven successful at determining the quality and authenticity of saffron (Petrakis, Cagliani, Polissiou and Consonni, 2015). ^{1}H-NMR was shown to give reproducible results rapidly, however, sample pre-treatment, was more time consuming than required for other spectroscopic techniques, and this pre-treatment would require a laboratory setting and trained personnel. Therefore, further work carried out by Petrakis and Polissiou (2017) using DRIFTS on FTIR minimized the process of sample preparation and sample destruction and proved to be successful along with PLS-DA classification and quantitative PLSR models at detecting six known saffron adulterants (Petrakis and Polissiou, 2017).

Although these spectroscopy methods are often successful on their own, further developments are being made to improve the methods by; 1) combining data, 2) increasing sensitivity or 3) developing ways to analyse through packaging.

1) Combining data: Wang et al. (2014) carried out a study that improved FTIR and NIR results for the detection of the adulterant *Iuicium lanceolatum A.C. Smith* (ILACS) in Chinese star anise. This method involved combining the NIR and FTIR spectral data and the use of PCA and Linear Discriminant Analysis (LDA) chemometric techniques. Although the FTIR performed better than NIR in this study when analysed separately, the classification results from the combined approach proved to be even more successful.

2) Increasing sensitivity: Vermaak et al. (2013) used hyperspectral imaging with PCA and PLS-DA to distinguish between the neurotoxic Japanese star anise and Chinese star anise. This emerging method incorporates spectroscopy and imaging to produce both spatial and spectral data from a sample (Gowen, O'Donnell, Cullen, Downey and Frias, 2007). This method is also non-destructive and rapid with the added advantage that with the acquisition of several predictions on the sample, the statistics are better (Vermaak, Viljoen and Lindstrom, 2013). The quantification of adulterants, buckwheat or millet, in ground black pepper was carried out using FTIR and NIR with hyperspectral imaging with PLSR chemometrics. NIR with hyperspectral imaging was seen to produce the best calibrations which, in this case was largely to do with the larger sample area used with NIR, and the spatial information from the imaging system used with it (McGoverin, September, Geladi and Manley, 2012). Galaxy Scientific's Classical Least Squares (CLS)-based Advanced-ID algorithm has been developed to detect screening samples to a level as low as 0.01% (Galaxy Scientific, 2016). When it was used to detect paprika adulterants, it detected Sudan 1 dye at 0.1%, tomato skin at 0.5% and brick dust at 5%.

3) Analysis through packaging: Terahertz spectroscopy by Nallappan et al. (2013) was used to overcome the barrier of common packaging materials such as plastics and papers. This method is a promising non-intrusive technique that was used for the detection of yellow chalk powder in turmeric.

It is apparent that further improvements and developments are ongoing with the use of spectroscopy. Developments seen in benchtop spectroscopic instruments are also being transferred to handheld devices. An added benefit as discussed by Ellis et al. (2015) would be to use the advantages of the NIR and FTIR combined, and developed into a handheld device. Overall, the ability to transfer this technology to portable and handheld devices allows the user to determine authenticity in the field, and can focus on vulnerable points of the supply chain. This not only allows improvements in traceability and detection of fraud, but at a basic level, it can also act as a deterrent. If food fraud criminals are aware of this possibility, they may be less likely to take the risks of committing a crime in the first place.

Limitations of spectroscopy must not be overlooked. Spectroscopy is used as a rapid screening technique and therefore, further investigations may need to be carried out by confirmatory techniques that require more expertise, time and cost more, such as mass spectrometry. This is also true when building models using chemometrics, the purity of samples needs to be assured in order to build accurate models. Another limitation of spectroscopy, as a non-targeted method, is the lack of a standardised validation procedure for all laboratories.

Following a review of more than sixty scientific publications, Reinholds et al. (2015) found that spectroscopic techniques are the major analytical techniques used to determine adulteration of herbs and spices in high concentrations. Overall, these techniques provide a good first point of control in the fight against food fraud. Although the use of other confirmatory techniques such as mass spectrometry may be required in some circumstances, the bulk of screening herbs and spices for EMA is possible with spectroscopy.

Although not a spectroscopic technique, an analytical screening technique called the ‘electronic nose’, capable of detecting aroma fingerprints, was used alongside PCA and Artificial Neural Networks (ANN) to detect adulteration in saffron (Heidarbeigi, Mohtasebi, Foroughirad, Ghasemi-Varnamkhasti, Rafiee and Rezaei, 2015). This technique was found to be promising, as detection was possible at higher than 10% adulteration, enough to detect

EMA (Heidarbeigi, Mohtasebi, Foroughirad, Ghasemi-Varnamkhasti, Rafiee and Rezaei, 2015).

8.4 Combination of Detection Methods

In some circumstances, there is a need to use more than one technique to verify results. Along with the combination of methods already described by Black et al. (2016) the combination of microscopy and GC-MS was also carried out for the detection of adulteration of fennel seeds (Ma, Mao, Zhou, Li and Li, 2015). Screening tests are often carried out with rapid techniques, but they have their limitations. In 2014, the USA recalled over 675 products due to the presence of undeclared nut protein in cumin. In a study carried out by Garber et al. (2016), it reported failings in the antibody-assay based technologies involved in screening products for allergens. Although these methods are robust, and can detect as little as 1µg of allergen, they are not always specific to the allergen they are developed to detect. Therefore, with this analytical weakness, DNA and mass spectrometry based tests are often used for further investigations. With the use of DNA and mass spectrometry analysis, additional allergens were detected; however, further work on the development of biomarkers for accurate analysis of a range of possible allergens may improve detection (Garber et al., 2016). This case indicates the limitations of screening methods with single analyte testing in some cases, and the need for multiple testing methods to understand the adulteration further.

8.5 Chemometrics

Chemometrics is used to improve the chemical data obtained from analytical instruments and to correlate the properties of samples with the use of mathematics and statistical methods (Lohumi, Lee, Lee and Cho, 2015). Chemometrics has been used in the calibration analysis of spectroscopic and spectrometric data. It has been used with both targeted and untargeted methods to detect the presence of fraud in food or to determine authenticity (Reinholds, Bartkevics, Silvis, van Ruth and Esslinger, 2015). The use of pre-processing is carried out in chemometrics to amplify desirable information from raw data and reduce the effects of undesirable information in the spectra. There are three key stages in the use of chemometrics, data pre-processing, development of a robust model, and the validation of a model and the analysis of results (Lohumi, Lee, Lee and Cho, 2015). Two commonly used pre-processing techniques include scatter correction methods, and spectral derivatives. Scatter corrective techniques can include Multiplicative Scatter Correction (MSC), Standard Normal Variate (SNV) and, normalisation to reduce the effects of physical variability caused by scattering

(Rinnan, Berg and Engelsen, 2009). The two commonly used spectral derivatives are Norris-Williams (N-W) and Savitzky-Golay (S-G) (Rinnan, Berg and Engelsen, 2009). The spectral derivatives aim to smooth the spectra without reducing the signal to noise ratio in the spectra too much (Rinnan, Berg and Engelsen, 2009).

The analysis of adulteration using spectroscopy and in some cases mass spectrometry requires further investigation with chemometrics. The most common algorithms used for the determination of authenticity or the detection of fraud are the classification/discrimination algorithms such as the unsupervised PCA, and the supervised LDA, PLS-DA or OPLS-DA. For the quantification of adulterant in a sample, PLSR analysis is used frequently.

8.6 Detection Methods for the Addition of Illegal Dyes

An extensive review of detection methods for illegal dyes has been carried out by Oplatowska-Stachowiak and Elliott (2017). Liquid Chromatography is the most common method of detection of illegal dyes. Other chromatography techniques were used with various detection methods including voltammetric, spectrophotometric and capillary electrophoresis. The use of Enzyme-Linked Immunosorbent Assay (ELISA) is also a common method of detection in this field (Oplatowska-Stachowiak and Elliott, 2017).

678 Table 5 Examples of Detection Methods for Substitution Adulteration

Ingredient	**Adulterant**	**Reference**	**Detection Methods**	**Chemometrics**
Saffron	*Carthamus tinctorius, Chrysanthemum x morifolium, Zea mays, Nelumba nucifera*	(Huang, Li, Liu and Long, 2015)	DNA barcoding	
Black pepper	Chilli	(Parvathy, Swetha, Sheeja, Leela, Chempakam and Sasikumar, 2014)	DNA barcoding	
Saffron	Safflower	(Babaei, Talebi and Bahar, 2014)	SCAR and ITS Multiplex PCR	
Saffron	Saffron	(Jiang, Cao, Yuan, Chen, Jin and Huang, 2014)	Barcoding Melting Curve	
Chilli	Dried red beet pulp and powdered *Ziziphus nummularia* fruits	(Dhanya, Syamkumar, Siju and Sasikumar, 2011a)	PCR-SCAR markers	
Oregano	*Satureja montana* L. and *Origanum majorana* L.	(Marieschi, Torelli, Bianchi and Bruni, 2011a)	SCAR-PCR	
Oregano	Olive leaves	(Marieschi, Torelli, Bianchi and Bruni, 2011b)	SCAR-PCR	
Oregano	*Cistus incanus* L., *Rubus caesius* L. and *Rhus coriaria* L	(Marieschi, Torelli, Poli, Bianchi and Bruni, 2010)	SCAR-PCR	
Saffron	*Arnica montana* L., *Bixa orellana* L., *Calendula officinalis* L., *Carthamus tinctorius* L., *Crocus vernus* L., *Curcuma longa* L., and *Hemerocallis* sp.	(Marieschi, Torelli and Bruni, 2012)	SCAR-PCR	
Turmeric	*Curcuma zedoaria/Curcuma malabarica*	(Dhanya, Syamkumar, Siju and Sasikumar, 2011b)	SCAR-PCR	
Cumin	Almond, peanut, tree nuts, peach and cherry	(Garber et al., 2016)	DNA analysis, Antibody based technology, Microscopy, Mass spectrometry	
Saffron	Saffron of unknown origin labelled as being cultivated in the PDO region in Spain can be used for substitution.	(Rubert, Lacina, Zachariasova and Hajslova, 2016)	LC HRMS	PCA, OPLS-DA
Fennel seed	*Anethum graveolens* fruit (AGF) and *Cuminum cyminum* fruit (CCF)	(Ma, Mao, Zhou, Li and Li, 2015)	Light microscopy, fluorescence microscopy, GC-MS	
Chinese star anise	Japanese anise	(Schrage et al., 2013)	Plant spray DART-HRMS	
Chinese star anise	Japanese anise	(Shen, van Beek, Claassen, Zuilhof, Chen and Nielen, 2012)	DART-HRMS	
Oregano	Olive leaves, myrtle leaves, hazelnut leaves, sumac	(Wielogorska et al., 2018)	LC-MS/MS, FTIR	PLSR

Oregano	Olive leaves	(Bononi, M.,Tateo, F., 2011)	LC-ESI-MS/MS	
Sage	Olive leaves	(Bononi, M.,Tateo, F., 2011)	LC-ESI-MS/MS	
Oregano	Olive leaves	(Bononi, Fiordaliso and Tateo, 2010)	GC/MS	
Paprika	Falsely declared Szegedi paprika substituted for *Szegedi Füszerpaprika* PDO	(Brunner, Katona, Stefanka and Prohaska, 2010)	ICP-MS	PCA, CDA
Oregano	Olive leaves, myrtle leaves, cistus, hazelnut leaves, sumac	(Black, Haughey, Chevallier, Galvin-King and Elliott, 2016)	FTIR , LC-HRMS	PCA, OPLS-DA
Garlic	Cornstarch	(Lohumi, Lee and Cho, 2015,Lee, Lohumi, Cho, 김문성 and 이수희, 2014)	Raman, FTIR	PLSR
Ginger	Cornstarch	(Lee, Lohumi, Cho, 김문성 and 이수희, 2014)	Raman	PLSR
Onion Powder	Cornstarch	(Lee et al., 2015,Lohumi et al., 2014)	Raman, FT-NIR, FTIR	PLSR
Saffron	*Crocus sativus* stamens, turmeric, safflower, gardenia	(Petrakis, Cagliani, Polissiou and Consonni, 2015)	^{1}H-NMR	PCA, OPLS-DA, O2PLS-DA
Saffron	*Crocus sativus* stamens, calendula, safflower, turmeric, buddleja, and gardenia	(Petrakis and Polissiou, 2017)	DRIFTS-FTIR	PLS-DA, PLSR
Chinese star anise	ILACS	(Wang, Mei, Ni and Kokot, 2014)	NIR/MIR	LDA, PCA
Chinese star anise	Japanese star anise	(Vermaak, Viljoen and Lindstrom, 2013)	SWIR-HIS	PCA, PLS-DA
Black pepper	Buckwheat or millet	(McGoverin, September, Geladi and Manley, 2012)	NIR hyperspectral imaging, FTIR	PLSR
Paprika	Tomato skins, brick dust	(Galaxy Scientific)	FT-NIR & Advanced-ID algorithm	
Turmeric	Yellow chalk powder	(Nallappan, Dash, Ray and Pesala, 2013)	Terahertz spectroscopy	
Saffron	Safflower dyed corn stigma	(Heidarbeigi, Mohtasebi, Foroughirad, Ghasemi-Varnamkhasti, Rafiee and Rezaei, 2015)	Electronic Nose	PCA, ANN

Conclusion

It is evident that EMA is a constant threat in the growing herb and spice industry. Cases of fraud have an economic impact on the industry as well as reducing consumer confidence. Potential public health risks following adulteration, such as the case of nut protein in cumin and paprika, are a major concern in the industry. Advances in DNA analysis include the use of SCAR-PCR and DNA barcoding provide faster and cheaper methods of analysis. Further advancement may include the use of NGS as it moves into the area of food fraud. Mass spectrometry, commonly used for the detection of food fraud is also improving by becoming faster and cheaper with the introduction of ambient techniques. Spectroscopic methods along with chemometric techniques are increasingly being used in the fight against food fraud and offer a rapid, robust screening technique that is cost effective and requires little expertise. There is an increasing need for screening techniques that can detect EMA over a range of products in the growing herb and spice industry.

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Captions

Figure 1 The Food Risk Matrix

(Spink and Moyer, 2011)

Figure 2 The Supply Chain Stages and Vulnerabilities within it for Herbs and Spices

(BRC-FDF-SSA, 2016)

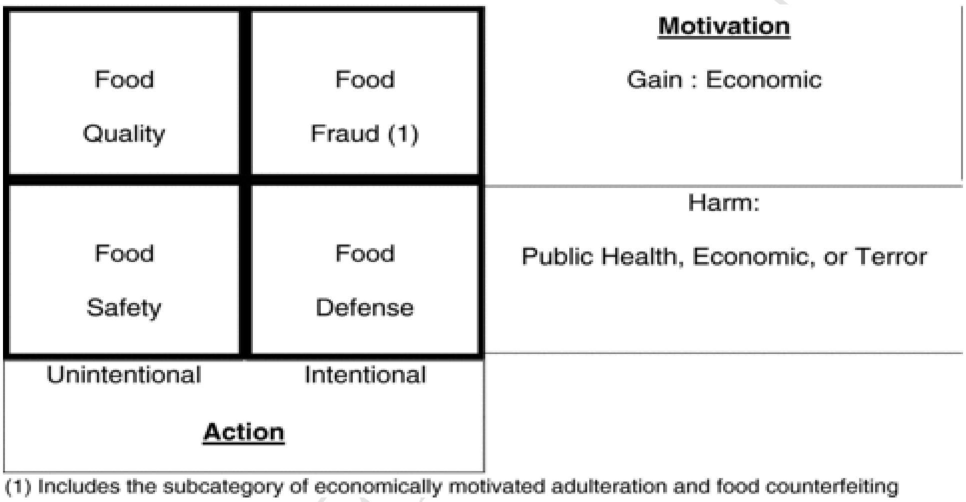

Figure 1


SUPPLY CHAIN STAGES
EXAMPLES OF VULNERABILITIES
GROWER
Adding non-functional parts of the plant
COLLECTOR
Loss of traceability
PRIMARY PROCESSOR
Adulteration at the grinding stage, (See Section 3)
LOCAL TRADERS
Deliberate misrepresentation
SECONDARY PROCESSOR
Adulteration (See Section 3)
EXPORTER
Purchase of low grade material / mislabelling
IMPORTER
Purchase of low grade material / mislabelling
TRADER
Purchase of low grade material / mislabelling
PROCESSOR/ PACKER
Substitution
FOOD MANUFACTURER/ RETAILER/ WHOLESALER
Knowingly placing mislabelled product on the market
CONSUMER

Figure 2