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A Brief History of Whiskey Adulteration and the Role of Spectroscopy Combined With Chemometrics in the Detection of Modern Whiskey Fraud

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Review

A brief history of whiskey adulteration and the role of spectroscopy combined with chemometrics in the detection of modern whiskey fraud

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Abstract: Food fraud and adulteration is a major concern in terms of economic and public health. Multivariate methods combined with spectroscopic techniques have shown promise as a novel analytical strategy for addressing issues related to food fraud that cannot be solved by the analysis of one variable, particularly in complex matrices such distilled beverages. This review describes and discusses different aspects of whisky production, and recent developments of laboratory, in field and high throughput analysis. In particular, recent applications detailing the use of vibrational spectroscopy techniques combined with data analytical methods used to not only distinguish between brand and origin of whisky but to also detect adulteration are presented.

Keywords: whisky; fraud; chemometrics; spectroscopy

Introduction

Whisky is a distilled alcoholic beverage produced from fermented grain mash where various grains are used for different varieties (e.g. barley, corn, rye, and wheat). This alcoholic beverage is generally classified by their country of origin, the nature of the grain, storage conditions and the type of blends. The production of this type of alcoholic beverages was first reported in Ireland in the *Annals of Clonmacnoise* from 1405 whereas in Scotland the early records dates from 1494 [1, 2]. Irish and Scotch remain the two main European whiskies to this day. Other major producers include the United States, Canada and Japan.

This review describes and discusses different aspects of whisky production, and the recent development of laboratory, in field and high throughput analysis. In particular, recent applications detailing the use of vibrational spectroscopy techniques combined with data analytical methods used to distinguish between brand and origin of whisky as well as to detect adulteration are presented.

History, origin and economic impact

Whisky is legally defined under European Community Council (ECC) regulation no. 1576/89 [3]. The regulation first defines a spirit drink (Article 2) as an alcoholic beverage that is *(a) intended for human consumption; (b) possessing particular organoleptic qualities; and (c) having a minimum alcoholic strength of 15% vol and contains a distillate of a naturally fermented agricultural product. None of the alcohol contained* *in a spirit drink shall be of synthetic or non-agricultural origin [Article 3(4)]. The nature of the raw material that may be considered agricultural in origin is contained in the Treaty on the Functioning of the European Union (TFEU) in Annex I* [4].

Within this definition Scotch and Irish whiskeys are further circumscribed, as they are both internationally recognised by Geographic Indication [2, 3]. According to the European Union (EU) definition [Article 4, Annex II] [4] *(a)* Whisky or whiskey is a *spirit drink produced exclusively by (i) distillation of a mash made from malted cereals with or without whole grains of other cereals, which has been: – saccharified by the diastase of the malt contained therein, with or without other natural enzymes, – fermented by the action of yeast; (ii) one or more distillations at less than 94.8% vol., so that the distillate has an aroma and taste derived from the raw materials used, (iii) maturation of the final distillate for at least three years in wooden casks not exceeding 700 L capacity. The final distillate, to which only water and plain caramel (for colouring) may be added, retains its colour, aroma and taste derived from the production process referred to in points (i), (ii) and (iii). (b) The minimum alcoholic strength by volume of whisky or whiskey shall be 40%. (c) No addition of alcohol as defined in Annex I(5)4, diluted or not, shall take place. (d) Whisky or whiskey shall not be sweetened or flavoured, nor contain any additives other than plain caramel used for colouring.*

Thus 'Scotch whisky' must be produced and matured in oak casks for a minimum of three years in Scottish distilleries from one of five designated regions Speyside, Highlands, Lowlands, Islay & Campbeltown [5-7]. Since 2005, the Scotch whisky definition was refined to include five distinct categories, determined by its production process in the whiskey industry; (I) Single Malt (SM) Scotch Whisky - distilled at a single distillery (i) from water and malted barley without the addition of any other cereals, and (ii) by batch distillation in pot stills; (II) Single Grain (SG) Scotch Whisky - distilled at a single distillery (i) from water and malted barley with or without whole grains of other malted or unmalted cereals, and (ii) which does not comply with the definition of SM.; (III) Blended, a blend of one or more SMs with one or more SGs; (IV) Blended Malt (BM) Scotch Whisky, a blend of SMs distilled at more than one distillery; and (V) Blended Grain (BG) Scotch Whisky a blend of SGs distilled at more than one distillery [6-8]. The bulk of the Scotch whisky is blended from 60% to 70% grain whisky and 30 to 40% malt whiskies. This blended whisky usually contains up to 40 individual malts which are blended to produce a consistent brand flavour. Every component of the blend must be matured for the minimum period or the specified date indicated on the bottle [5-8].

Irish whisky on the other hand is produced from either malted barley or a mixture of malted and un-malted other cereals and barley of which a minimum of 25% must consist of malted barley. The combination of the use of partially malted barley and a specialised processing approach. This involves the drying of the malt in closed kilns rather than over open peat fires and the application of a triple distillation process, the first of which produces 'low wines' a pot still distillate, which is re-distilled in another pot still to produce 'feints', before being placed in a Coffey still for the final distillation. It is this production process that gives Irish whiskeys their smooth and natural flavour. It is unique [2, 6, 9, 10] particularly in comparison to whiskies from other regions.

American whisky developed under an alternative legislative framework [9, 11, 12], US whisky is broadly defined as the distillate of a fermented grain mash at less than 95% alcohol. Consequently, US whisky consist of a broader range of distinct products in comparison to the Irish and Scotch spirits. There are six major types, rye, rye malt, pure malt, wheat, Bourbon and corn, all of which are produced from a different type of cereal grain. The exact type of grain and its required percentage (not less than 51%) in the mash used to produce the whiskey product are governed by Title 27 of the U.S. Code of Federal Regulations [11, 12]. All US whisky must also conform to additional standards outlined by title 27 of the U.S. Code of Federal Regulations, and so they must be distilled to not more than 80% alcohol by volume, to ensure the proper flavour profile; producers are prohibited from adding any colourings, caramel or flavour additives and finally, all of these whiskies (with the exception of corn whisky) must be aged in charred new oak container. There is no minimum period of aging specified, which creates opportunities for distilleries to differentiate their product based on the aging process. One such distinction is a 'straight whisky'. For a given whisky to be designated thus, it should not be blended with any other spirit, be no more than 80% alcohol by volume and aged for a minimum of two years [2, 11, 12]. There are several other types of American whisky, which do not specify a dominant grain. These include Blended Whisky, a Blend of Straight Whisky, Light Whisky (one which has been distilled at greater than 80% alcohol by volume) and Spirit Whisky (where a 'neutral spirit', a non-flavoured *alcohol* of 95% is mixed with at least 5 percent of a particular type of whisky).

Commercial distilleries began producing scotch in the late 18th century, despite its first being recorded in the 1492 Exchequer Rolls of Scotland. As of 2018, the Scottish Parliament recognised 245 distilling related businesses. The Distillers Company (DCL) is a dominant player in the industry since the "Big Amalgamation" the merger of the 'Big Five' brewing houses Buchanan, Dewar, Walker, Haig, and Mackie in 1925 [6-8, 12, 13].

American Whisky was first produced in the states of Virginia, Maryland and Pennsylvania in eastern United States around late 18th century and was originally a predominately rye-based spirit. Early distillers were often farmers who produced and distributed whiskey as a supplementary income. In 1791, Alexander Hamilton, the U.S. Secretary of the Treasury, in an effort to generate revenue, established a 25% tax on whiskey distillers. The majority of distillers operated small production facilities and the federal tax was greatly opposed. This opposition became known as "The Whisky Rebellion" when it was necessary for the federal government to send troops to enforce the tax [14]. This resulted in a larger number of producers relocating West, most notably to Kentucky. Over time, the number of states producing whiskey increased, including Tennessee which produced the famous Jack Daniel's brand. America's whisky industry suffered repeated setbacks, including a 13-year Prohibition on alcohol between 1922 and 1933, which barred production of all alcohol; the supporters of prohibition saw alcohol as a major catalyst for the ills experienced in the society. By the 1933, however, it became apparent that prohibition was going to remain a noble experiment. The popularity of whiskey grew reaching its heyday in the 1950s in the U.S. before falling out of favour. Today, whisky popularity is resurging as established brands such as Jack Daniel's and Jim Beam offer single-barrel whisky aimed at connoisseurs and new distilleries are appearing annually [11, 14, 15].

Conversely, the number of Irish distilleries remains limited compared to the number producing scotch and American whiskies. Prior to the 1900s Irish whisky led the world's spirits trade until a perfect storm of the newly formed Irish Republic's national politics, the American prohibition, and technology decimated the industries producers. At the turn of the last century the Irish whiskey industry was at its pinnacle, with 88 licensed distilleries producing an estimated 12 million cases primarily for export. This coupled with the impact of the Irish War of Independence and Civil War and the fledgling Irish State's economic policies debilitated the industry. Arguably however, far more devastating was the reluctance of Irish whisky producers to adopt and capitalise on the invention of the column still, which allowed for the easier production of palatable spirits which Scottish distillers producing whisky blends incorporated readily. This ultimately handed an overwhelming advantage to Scottish whisky producers [16, 17]. By the 1930s there were only five active Irish distilleries, Old Bushmills, Jameson, John Powers, Cork (Paddy) and Tullamore Dew. In 1966, all bar Old Bushmills combined to constitute the Middleton centre in Cork. In 2007 there were only four distilleries – Old Bushmills, Middleton, Cooley and Kilbeggan in operation [1, 9, 10]. By the end of 2019 there were 56 revenue registered Irish Whiskey Producers in Ireland ~~(**Table 1**)~~.

~~Table 1: List of Revenue registered Irish Whiskey producers in Ireland (2019)~~

~~1. Admiralford Ltd., 113-119 IDA Industrial Estate, Waterford~~
~~2. All Technology Ireland Ltd., Pearse Lyons Distillery, James Street, Dublin 8.~~
~~3. Ballykeefe Distillery, Kyle, Cuffsgrange, Co. Kilkenny.~~
~~4. Blackwater Distillery, Church Road, Ballyduff, Co. Waterford.~~
~~5. Clonakilty Distillery Ltd., Dunowen Farm, Ardfield, Clonakilty, Co. Cork.~~

~~6. Clonakilty Distillery Ltd., The Waterfront, Clonakilty, Co. Cork.~~
~~7. Cooley Distillery, Riverstown, Dundalk, Co. Louth.~~
~~8. Copeland Distillery, 43 Manor Street, Donaghadee, Co Down, BT210HG.~~
~~9. Dingle Distillery, Ballinaboula, Dingle, Co. Kerry.~~
~~10. Dingle Distillery, The Old Mill, Dingle, Co. Kerry.~~
~~11. Drioglann Shliabh Liag, Line Road, Carrick, Co. Donegal.~~
~~12. Dublin Liberties Distillery, 33 Millstreet, Dublin 8.~~
~~13. Great Northern Distillery, BAK Bulk Services, RED Barnes, Drumcar Road, Dunleer, Co. Louth.~~
~~14. Great Northern Distillery, Carrick Road, Dundalk, Co. Louth.~~
~~15. Great northern Distillery, Christianstown, Readypenny, Co. Louth.~~
~~16. Great Northern Distillery, Cliven, Rathescar, Co. Louth.~~
~~17. Great Northern Distillery, Oberstown, Ardee, Co. Louth.~~
~~18. Great Northern Distillery, Rathlust, Ardee, Co. Louth.~~
~~19. Great Northern Distillery, Snic Hill, Ardee, Co. Louth.~~
~~20. Great Northern Distillery, Tullamore, Co. Offaly.~~
~~21. Hanlon Transport Ltd., Greenore Ind. Est., Dundalk, Co. Louth.~~
~~22. Hazlewood Demesne Ltd., Hazlewood House, Hazlewood Avenue, Sligo.~~
~~23. Irish Distillers, Midleton Distillery, Midleton, Co. Cork.~~
~~24. Irish Distillers, Robinhood Road, Clondalkin, Dublin 22.~~
~~25. Kilbeggan Distillery, Main Street, Kilbeggan, Co. Westmeath.~~
~~26. Killowen Distillery Ltd., 29 Kilfeagan Road, Rostrevor, Co. Down, BT3434W.~~
~~27. Lambay Irish Whiskey, Lambay Island, Malahide, Co. Dublin.~~
~~28. Liam Connaughton & Sons, Grand Canal Quay, Dublin 2.~~
~~29. Niche Drinks Company Ltd., 10 Rossdowney Road, Londonderry, BT476NS.~~
~~30. O'Connell Heritage Ltd., Lakeview House, Fossa, Killarney, Co. Kerry.~~
~~31. P.J. Rigney Distillery, The Food Hub, Drumshambo, Co. Leitrim.~~
~~32. Piranha Beverages Ltd., 3 Turvey Business Park, Turvey, Donabate, Co. Dublin.~~

~~33. Powerscourt Distillery Ltd., Powerscourt Estate, Enniskerry, co. Wicklow.~~
~~34. PRL Group Logistics, Chancellors Mills, Talbots Inch, Freshford Road, Kilkenny.~~
~~35. PRL Group Logistics, Talbots Inch, Freshford Road, Kilkenny.~~
~~36. R & A Bailey, Western Estate, Nangor Road, Dublin 12.~~
~~37. Rademon Estate Distillery Ltd., The Distillery, Rademon Estate, Ballynahinch Road, Co. Clare.~~
~~38. Renegades Waterford Distillery, 9 Mary Street, Waterford.~~
~~39. Rober A Merry & Co. Ltd., Cashel Road, Clonmel, Co. Tipperary.~~
~~40. Roe & Coe Distillery, St. James Gate, Dublin 8.~~
~~41. Roe & Coe, Visitor Experience Centre, St. James Gate, Dublin 8.~~
~~42. Royal Oak Distillery Ltd., Royal Oak, Co. Carlow.~~
~~43. Slane Castle Whiskey Ltd., Slane Castle Distillery, Slane, Co. Meath.~~
~~44. Stafford Bonded Warehousing Ltd., Killasem, Ballygarran, Co. Waterford.~~
~~45. Stafford Wholesale Ltd., Sinnottstown Lane, Drenagh, Co. Wexford.~~
~~46. Stafford Wholesale Ltd., Waterford Airport Business Pk, Ballygarran, Co. Waterford.~~
~~47. Teeling Whiskey, 13-17 Newmarket, Dublin 8.~~
~~48. Terra Spirits & Liqueurs, Baileboro, Co. Cavan.~~
~~49. The Chapel Gate Irish Whiskey Co., Gowerhass, Cooraclare, Co. Clare.~~
~~50. The Connacht Whiskey Co. Ltd., Beeleek, Ballina, Co. Mayo.~~
~~51. The Echlinville Distillery, 62 Gransha Road, Kircubbin, Newtownards, BT221AJ.~~
~~52. The Old Bushmills Distillery, Jameson Distillery, Bow Street, Dublin 7.~~
~~53. Watercourse Distillery, The Jameson Experience, Midleton, Co. Cork.~~
~~54. West Cork Distillers Ltd., Market Street, Skibbereen, Co. Cork.~~
~~55. West Cork Distillers Ltd., Marsh Road, Skibbereen, Co. Cork.~~
~~56. William Grant & Sons, Tullamore Distillery, Clonminch, Tullamore~~

The global whisky market size was valued at USD $57.96 billion in 2018 and it is projected to reach USD $89.60 billion by 2025. This growth is driven by multiple factors, including, increasing disposable income, consumer preferences and changing lifestyles [15, 18-20]. Scotland's brewing and distilling sectors play a vital role in the Scottish economy, and in 2019 the spirits industry contributed approximately 3% to total Scottish GDP. Moreover, since 2000 the spirits/distilling sector has contributed an average of 2.8% with a high of 3.3% in 2013 to total GDP [19].

In 2018, the US spirits industry gained market share over beer and wine, with sales rising seven-tenths of a point to 37.4% of the total beverage alcohol market. This was the ninth straight year of record spirits sales and volumes, reflecting continued market share gains. Supplier sales were up over 5.1%, rising from USD $1.3 billion to a total of USD $27.5 billion [20].

In 2019, Ireland's total agri-food sector exports amounted to €14.5 billon, with the food and beverages sector accounting for 21% of all industrial turnover and 23% of all manufacturing industry turnover. This represents a 67% increase in export values compared to 2010. International exports account for 31% and makes Ireland's Food and Drink industry the most global indigenous industry exporting to 180 markets worldwide [18]. Growth in Irish alcohol exports grew 8% in 2019 (€1.45bn) with Irish Whisky accounting for 50% of the €137m in beverage export growth. In 2019, Irish whisky exports increased 11% from the previous year contributing to an overall value of €727m, accounting for an overall climb of 370% between 2010 and 2019. Domestically, this growth is underpinned by new distillery openings and increased development of whisky heritage tourism [18].

Brand recognition is central to whisky global market growth, following a trend of 'drink less but better'. As imitations are often a response to increased demand, the growing global market for whisky has sparked concerns within the Industry that counterfeit and adulterated products may infringe on laws governing labelling and sales [12, 21-24]. The International Chamber of Commerce's 2017 report titled 'The Economic Impacts of Counterfeiting and Piracy' estimates that the global economic value of counterfeiting and piracy costs could hit USD $1.9 trillion by 2022. This, combined with the additional negative impacts of counterfeiting and piracy such as displaced economic activity, investment and public fiscal losses, the overall impact on the economy would be an estimated loss of USD $4.2 trillion from the global economy, while also endangering 5.4 million legitimate jobs in the sector [25].

Adulteration, fraud and public safety

The rebranding of lower quality commercial whiskeys as top-shelf products can significantly damage a producer's reputation and bottom line. In 2018 the BBC [26] and other media outlets [27, 28] reported that a third of commercial Scotch whiskies tested were fraudulent. Of greater concern is the potential risk to consumers and their health [29-35]. Such incidences as the 'Czech Republic methanol poisonings' of September 2012, where 38 people in the Czech Republic and 4 people in Poland died as a result of methanol tainted bootleg spirits [34]. Several poisoning incidents were reported in Iran with the poisoning of 768 people (including 96 deaths) by illicit and non-standard alcoholic beverages; 62 people (11 fatalities) were poisoned with methanol laced counterfeit spirits in Shiraz in 2004 and 694 (6 deaths) and poisonings recorded in Rafsanjan, Iran in 2013 [31]. More recently toxic moonshine was reported to have killed 154 people in India in two separate incidents in 2019 [36]. In March 2020, Iranian media reported that nearly 300 people have been killed and more than 1,000 sickened by drinking methanol laced bootlegged spirits, in the mistaken belief that it was effective against Covid-19 [37].

Other dangers to public health from the illicit production of spirits include the addition of industrial alcohol, the presence of chemicals used to denature industrial alcohol and the resultant contamination (e.g. ethyl acetate, which can cause irritation of the digestive tract) [38]. Ingestion of toxic concentrations of some of these chemicals can result in pronounced acidosis accompanied by cardiovascular shock and cause central nervous depression. Lower volumes of such adulterants can cause headache, nausea, fatigue, and dizziness.

High levels of chloroform are also often detected in illegally produced alcoholic products [39], most likely as a result of counterfeiters adding hypochlorite to the fake spirits in an attempt to remove denatonium benzoate, a widely used denaturant with a characteristic bitter taste, from denatured alcohol, via the addition of hypochlorite [40]. Ingestion of chloroform can result in damage to the central nervous system (brain), liver, and kidneys of unwitting consumers [41].

An additional danger to the public is the leaching of toxins from the improvised illegal distillation tools utilised by counterfeiters, particularly as the illicit stills and other production materials are often unfit to come into contact with food products. Genuine producers carry out testing to ensure that there is no unwanted contamination from beverage contact materials. Illegal producers are either unaware or indifferent to the potential of harmful toxins may be present in their illegal product. This was highlighted in a new report by Lachenmeier [42], which showed that a large number of fruit spirits in the Slovak Republic and Hungary were contaminated with the heavy metal elements lead and cadmium.

Consequently, fraud, particularly in the distilling sector is causing increasing levels of concern. It is an incredibly lucrative business, with perpetrators profiteering at comparatively lower risk as the legal repercussions are much more lenient than those for other illegal activities, such as drug trafficking [21-23, 30, 35, 43, 44]. It is apparent that without a proper verification technique that derives from the beverage itself rather than some externally affixed marker or associated paperwork (e.g. blockchain), the system will always be vulnerable to the inclusion of illegal or otherwise non-compliant material [45-48].

In order to assess the composition and identity of the beverage directly, the development of rapid and non-destructive analysis methods are critical for the future of the whisky industry. In addition, methods to verify the compliance of producer declarations regarding origin and source, as defined and requested by quality assurance standards in the production value chain will be of benefit. The current trend in analysis (as well as in all fields of research in food fraud) is towards fast, simple and reliable analytical techniques with the potential to partly or fully replace the complex and expensive reference methods that dominate the landscape [49-55]. The traditional chromatographic based techniques are expensive, time consuming and require highly trained operators.

In order to preserve and protect the premium status of its merchandise the global whisky industry must assure product safety and quality. This requires not only continuous monitoring~~, and~~ but also the development of analytical systems aimed at safeguarding consumer confidence in whisky and related spirit drinks. Therefore, significant research on flavour and quality, consumer safety and anti-counterfeiting/authenticity is now being carried out. Moreover, in recent years there has been an observable effort by researchers and stakeholders within the industry to develop new technologies and processes aimed at anti-counterfeiting and authenticity checking, supported by initiatives like the pan-European food integrity project [56]. Key to this effort is the development of sensors and rapid methods for the analysis of suspect products, particularly those that are field-portable and can be used at point-of-sale or distribution [44, 53, 57].

Standard methods of analysis

The authentication of spirit and alcoholic beverages, and the detection of counterfeits is an arduous task. Their chemical profiles are dominated by two major constituents, ethanol and water, which can often mask adulterants or other constituents present in the liquid product. This has required exhaustive method developments in the area of beverage analysis to date, to ensure that trace levels of adulterant constituents can be well separated from the dominant ethanol and water to allow for their characterisation and quantitation. Analysts rely on other flavour providing compounds, generally at trace concentrations (ppm and ppb) to definitively identify and differentiate between samples. However, the cost of such analysis is high, as state of the art, highly selective and sensitive instrumentation is required. Moreover, exceptionally skilled staff are required to maintain the instrumentation, conduct the analysis~~,~~ and develop and optimise testing protocols.

A variety of these analytical methods that are currently employed to ensure the safety, quality and authenticity of spirits are summarised in **Table ~~2~~1**. These methods are utilised to ensure that a given sample is consistent with the production requirements legislated by either the EU, Commission Regulation (EC) No 2870/2000 [58, 59], or the AOAC International Official Methods of Analysis [60] mandated by the United States Alcohol and Tobacco Tax and Trade Bureau.

Table ~~2~~1: Standard analytical methods utilised and their application [58-60]

Analytical technique	Indicative data or analyte	Authenticity issue / information

Densitometry	Alcohol Strength (not suitable for spirits with significant levels of dissolved solids e.g. sugars)	Dilution
Distillation and Densitometry	Alcohol Strength	Dilution
Gas Chromatography with flame ionisation detector (GC-FID)	~~Major~~ Volatile Compounds~~ngeners~~	Category and brand discrimination
GC-FID	Denaturants (Methanol, isopropanol, methyl ethyl ketone etc.)	Detection of non-potable alcohol
UV-Vis Spectroscopy (UV-Vis)	Spectroscopic profile	Brand discrimination
Ultra-High-Performance Liquid Chromatography - UV-Vis (UHPLC-UV)	Maturation Congeners	Category discrimination, lack of maturation, addition of flavouring
pH	pH	Lack of maturation
Atomic Absorption Spectroscopy (AAS)	Trace Metals	Brand Discrimination
Ion Chromatography (IC)	Trace Metals	Brand Discrimination
Inductively coupled plasma - optical emission spectrometry (ICP-OES)	Trace Metals	Brand Discrimination
Inductively coupled plasma - Mass Spectrometry (ICP-MS)	Trace Metals	Brand Discrimination
Ion Chromatography-Pulsed Amperometry Detection (IC-PAD)	Sugars	Addition of sweetening
Ultra-High-Performance Liquid Chromatography – Reflective Index Detection (UHPLC-RI)	Sugars	Addition of sweetening, brand discrimination
Gas Chromatography - Mass Spectrometry (GC-MS)	Flavourings, Denaturants, Fingerprinting	Brand discrimination, addition of flavourings, detection of non-potable alcohol
Liquid Chromatography - Mass Spectrometry (LC-MS)	Flavourings, Denaturants, Fingerprinting	Brand discrimination, addition of flavourings, detection of non-potable alcohol
Nuclear Magnetic Resonance spectroscopy (NMR)	Ethanol	Botanical origin of ethanol, detection of synthetic alcohol
14C dating by Liquid Scintillation Counting or Accelerator Mass Spectrometry	Ethanol	Date of production

The alcohol content of a whisky is measured to ensure that quality standards are met and to ensure product integrity. Its measurement is also necessary since there is a minimum alcohol strength requirement in the legislation which genuine products must meet consequently a strong indication of counterfeit products. If the alcohol content of the sample falls below the minimum alcohol strength limit and/or a definitive difference between the measured alcoholic strength and the stated label value is often an indication of some manipulation of the original product. The accepted reference methods associated with alcoholic strength exploit the liquids density. Gas chromatographic (GC) methods coupled with a variety of detectors are utilised to monitor most of the major volatile congeners and denaturants present in alcoholic beverages. These include but not only limited to acetaldehyde and ethanal, 1-propanol, 1-butanol, 2-butanol, 2-methyl-1-propanol, 2-methyl-1-butanol and 3-methyl-1-butanol, ethyl acetate and methanol. Similarly, GC-MS and LC-MS are useful for the detection and quantification of both volatile and non-volatile flavouring additive compounds.

Counterfeiters commonly add sugars fraudulent products in an attempt to improve their taste and mimic the natural sweetness of a genuine product. However, the sugar profile of a suspect material will differ significantly to that of a genuine product. For example, genuine Scotch whisky products contain considerably less sucrose than glucose and fructose. Chromatographic methods such as UHPLC-RI, IC and IC-PAD are utilised to measure trace levels of individual sugars present naturally in certain spirits in order to define appropriate sugar profiles which can be later utilised to detect adulteration.

Current trends within the industry are focused on the potential of testing not only during the production process but at multiple key points in the supply chain. This has prompted research in the application of alternative analysis approaches with an emphasis on field based rapid, portable, user – friendly (i.e. for the non- specialist) options.

Spectroscopic methods and the use of chemometrics

Spectroscopy techniques have shown considerable promise in the fight against counterfeit and fraud, as they are non-destructive, non-invasive and possess unique analytical capabilities, the development of a materials chemical 'fingerprint'. Their usefulness is further enhanced by the development of chemometric or multivariate analysis methodologies which allow the rapid identification and classification of similar samples using their molecular properties (e.g. fingerprint) [61-64].

Spectroscopy methods and techniques are often the preferred analytical approach for the qualitative and quantitative characterisation of chemical mixtures, as a large amount of data can be generated in a rapid and non-invasive manner. However, interpreting the data to form a clear and concise conclusion from such analysis is not always straightforward. The use of certain techniques, like UV-VIS spectroscopy can lead to spectral response overlaps with overlaps with other components in the whisky, which has very many trace components that can carry over from the malts/grains in the distillation process. These can inhibit the determination of an individual component (or adulterant) concentrations in the sample being tested. Therefore, the precision and accuracy of identification can be challenging because of the similarity of many spectral responses [65]. Consequently, analysts will often apply a work around, which might include the addition of a component to interact with the adulterant you wish to identify so that its response can be well separated out and measured. However, the majority of spectroscopic 'fixes' or sample pre-treatments, to aid in the extraction of results from the spectral data work less well than is ideal. That being said, there is a considerable wealth of "information" gathered in a spectral scan that is not used for identification or measurement. Scientists have begun to look at this unused data to determine if some data points can be used to elicit different patterns that could be used to verify the measurements of similar species better and without the need for a second type of confirmation test to be conducted. This type of forensic investigation of all of the spectral data is commonly referred to as a chemometric study. It relies heavily on the use of mathematics and statistics in interpreting the data to provide definitive results. While chemometrics was first mooted back in 1995, it took almost twenty years for spectroscopic instrumentation to be fitted with effective and reproducible software tools to allow researchers to incorporate chemometrics into the processing of their spectral data to give absolute verifiable identification and quantitation of chemical components that would otherwise have been missed [66-68].

The combination of scientific analysis with software tools underpinned by mathematical systems is of enormous use to those companies trying to track fraudulent products. It is timely now that as the number of whisky producers is on the increase that adulterant measurement and analysis has become more robust. The integration of chemometrics with spectroscopy allows the analyst to better mine the data and extract relevant information for the generation of more confidence in a specific result. While chemometric software can certainly add more certainty to analysis results it is still challenging where one is trying to measure whether a single or small amount of an adulterant is present or not in a sample that already has many components present natural. Food and beverages are examples of such complex samples, and the data may have to be analysed at different spectral wavelengths or channels to be of use. The data generated often has a high number of correlations from one measurement channel to the next and from one chemical species to the next over those same channels. This high serial correlation decreases the use of much the data and this can be a limiting factor. However, all is not lost, as the data results can be refined using chemometric software to allow for such redundancy of data. Nowadays, spectroscopic instruments have inbuilt chemometric methods which are extremely efficient at extracting unique and redundant information from multichannel data such as spectra [61-64].

The field of chemometrics is still evolving and consequently it defintion requires continued modification to allow for its development, the international chemometrics society defines chemometrics as the chemical discipline that exploits mathematical and statistical methods to design or select optimal measurement procedures and experiments to provide maximum chemical information by analysing chemical data [62].

~~For example, it is very hard to find a wavelength channel in a standard UV – Vis spectrum to distinguish between alanine and glycine, both essential amino acids in foodstuffs, as chemically they are very similar, and both tend to absorb over the same sets of wavelengths. However, because chemometrics expects this correlation, it allows the analyst to take advantage of the correlations similarity or redundancy to increase the methods precision, similar to the manner that a mean takes advantage of the redundancy of a set of numbers. This is referred to as multivariate analysis.~~

Recent innovations in adulterant analysis

The recent literature presents a number of spectroscopic techniques for the rapid and more reliable identification of adulterants in whisky **(Table ~~3~~2)**. ~~In all cases the studies~~ A~~a~~ variety of spectroscopic techniques~~, not just the traditional UV VIS spectroscopy,~~ have been combined with multivariate analysis software tools to **(I)** characterise whisky from different geographical origin,; **(II)** provide key information to indicate differing maturation process (e.g. maturation time); and **(III)** to detect fraud or the presence of an adulterant. Some highlights from the literature are described in more detail below.

Table ~~3~~2: Application of Spectroscopy and Chemometrics

Technique	Application	Number of samples	Validation Method	Reported Classification	Ref
UV-Vis (PCA)	Authentication of Scotch Whiskies	Ref set 50 Test set 35	Complimentary gas chromatographic authentication	100 %	69
UV-Vis (PLS-DA)	Discrimination & identification of Scotch whiskies	Ref set 164 Test set 73	Two independent data sets not part of the reference set	Ref 98.6 % Test 93.1 %	70

UV-Vis (PLS-DA)	Discrimination of whiskies	27	N/A	N/A	71
NIR (PCA/SIMCA)	Authentication & provenance of whiskies	Ref set 40 Test set 69	Cross Validation	100 %	72
FT-IR (PLS-DA)	Discrimination & authentication of whiskies	200	Validation set containing 25 % of samples	96.3 %	73
NIR (Machine Learning)	Determination of ethanol & methanol concentration	44	Leave one out Cross Validation	100 %	74
Raman (Machine Learning)	Discrimination & identification of Scotch whiskies	6 classes (400 samples)	5-fold cross validation	70 – 90 %	75
ATR-IR (PLS-DA)	Discrimination & identification of whiskies and other spirits	Ref set 85 Test set 23	Validation set 43	≥ 96 %	76
FT-IR (PCA)	Authentication of whiskies and detection of methanol.	150	Cross Validation	≥ 97 %	77
FT-IR/UV-Vis (PLS-DA)	Authentication & provenance of whiskies	11	Cross Validation	100 %	78
Raman/NIR (PLS-DA)	Discrimination & authentication of whiskies	114	N/A	100 %	79
Raman (PC-DFA)	Discrimination & authentication of whiskies	144	N/A	100 %	80

MacKenzie and Aylot [69], reported the development of a novel spectroscopic method for Scotch whisky brand authentication. The UV-Vis based technique clearly distinguished between genuine Scotch samples and counterfeits, the majority of which were a combination of cheap local alcohol flavoured with a smaller proportion of the genuine whisky and colour. The authors also reported the method's ability to classify various Scotch whisky brands. It was illustrated that the UV-Vis technique combined with chemometric analysis could be used as complimentary method to the traditional GC authentication methodology. This study highlighted some distinct advantages of the spectroscopy approach, including, the portability of the handheld spectrophotometer which enabled field-testing. The spectroscopic method was also quicker (i.e., sample could be analysed in less than a minute compared to a GC analysis time of approximately 20 minutes), was more cost and resource effective when in compared to the standard methods [69].

Martins and co-workers [70], determined that UV – Vis spectroscopy combined with partial least squares discriminant analysis (PLS – DA) modelling was an efficient method for discriminating between seven brands of whisky. The method proposed by the authors was also very useful for the detection of adulterants in other spirits. The method was able to differentiate between all genuine samples and detected the counterfeit samples with correct identification rates of between 93 – 100% (depending on the brand).

Similarly, Joshi et al [71], also reported the successful application of UV-Vis combined with chemometrics to classify whisky samples from several geographical regions. The authors reported that PLS – DA models correctly classified 100% of the whisky samples belonging to the USA and Canada and 98% of those belonging to Scotland and Ireland respectively. Moreover, Joshi and co-workers also determined that the scanning temperature of the whiskey samples did not impact the UV – Vis spectra of the sample and therefore the classification rates. However, they do recommend that if an analytical protocol to analyse this type of alcoholic beverages will be developed to target authenticity, integrity, or country of origin in a consistent manner it would be appropriate to define an appropriate scanning temperature for quality assurance and certification purposes [71].

Infrared spectroscopy both near (NIR) and mid (MIR) combined with chemometrics has proven to be a popular technique for determining whisky quality either solely or in unison with other spectroscopy methods. Pontes et al [72], developed a classification method for distilled alcoholic beverages and verification of adulteration, with water, methanol and industrial ethanol, using NIR spectroscopy and chemometric methods such as principal component analysis (PCA) and soft independent modelling of class analogy (SIMCA). The authors reported that their strategy was an effective tool in the classification and verification of adulteration in whisky, brandies, rums and vodkas. Pure and adulterated samples were successfully classified (100% at the level of 95% of confidence). Other benefits of the approach include, direct sample analysis, and pre-treatment required; use of small volumes allowing for high sample analysis throughput; and no additional use of reagents thereby reducing costs; can be carried out by untrained personnel to name a few, thereby, making this strategy suitable for screening analysis to verify adulteration of alcoholic beverages [72].

Sujka and Koczon [73] have reported the development of a rapid, simple, and non-destructive analytical procedure for the discrimination and authentication of whisky samples originating from Scotland, Ireland and USA using MIR spectroscopy combined with multivariate analysis models. The procedure was also found to be useful for identifying the whiskies time of maturation (two, three, six and twelve years). The authors describe the construction of eight discriminant models which allowed analysts to distinguish Scottish, Irish, and American whisky samples. As well as completely differentiating between beverages matured for 2 – and 3 – years from those aged for between 6 – and 12 – years. The authors also reported 100 % accuracy when discriminating between American and Scottish whiskies.

Large and co – workers [74] demonstrated the ability to determine the alcohol concentration non – invasively in arbitrary bottles using NIR spectroscopy in combination with machine learning. While the authors reported that the determination of ethanol concentration was possible with high accuracy the determination of methanol concentrations within a consistent overall alcohol level was more difficult. Backhaus et al [75] combined chemometrics with NIR spectroscopy to classify the age, maturing cask, distillery and product variety of Scotch with very high accuracy. The authors also highlight that the technique reduced overall cost and processing time of analysis.

Mid-infrared spectroscopy was also reported by Picque et al [76], to analyse and discriminate between Cognacs and other distilled drinks including whisky, bourbons, and counterfeit products. Chemometrics was applied by the authors to the spectral data with good levels of accuracy, and 96% of samples in the test set were correctly assigned to Cognacs and non-Cognacs by PLS – DA. The authors also have come up with a means of applying a sequence of combined analytical techniques to provide enhanced accuracy for the discrimination between Cognacs. They propose that a single chemometric process could be used to the combined data outputs of IR, UV-vis, NMR and GC analysis, coupled with neural network information could further enhance the determinations of counterfeit products from Cognac and other products [76].

Chen et al [77] have employed chemometrics and IR spectroscopy integrated with information from digital labelling to develop a means of rapidly detecting fraudulent liquors, for the presence of methanol, which is the most important and difficult adulterant to detect with accuracy. The spectral bands of methanol were labelled using iterative discrete wavelet transform for classification, and PCA and PLS analysis were then applied to discriminate problematic samples using the iterative discrete wavelet transform filtered signals. By using digital pre-processing methods, the authors could extract spectral features of methanol from the alcoholic drinks in the presence of a diverse array of uncontrolled matrix effects. The technique boasted a recognition accuracy of higher than 97.0%, with each measurement taking 3 min, illustrating the promise of the tool. The authors also indicated that the method could be extended to detect of other targeted volatile substance in complex matrixes. In a 2017 study Wiśniewska and colleagues [78] utilised headspace mass-spectrometry (HS-MS), MIR an UV – Vis to authenticate whisky samples from multiple origins and ways of production (Irish, Spanish, Bourbon, Tennessee whisky and Scotch). The authors used PLS-DA to build classification models which fully classified the five groups of whisky samples. The authors also reported that it was also possible to differentiate samples within this product class, demonstrating that production processes were impactful on the quality of the spirits [78].

Recently work by Ellis et al [79, 80], has investigated the use of Raman spectroscopy combined with chemometrics as a means for rapid *in situ* through-container analysis of whisky samples; the authors report detection of multiple chemical markers known for their use in the adulteration and counterfeiting of Scotch whisky, and other spirit drinks without any physical contact with the sample; with the ability to discriminate between and within multiple well-known Scotch whisky brands, and the detection of methanol concentrations well below the maximum human tolerable level of 2% v/v.

Conclusion

The implementation and adoption of spectroscopy techniques combined with chemometrics allows for the rapid and non-destructive analysis, characterisation and detection of fraud in whiskies. The most promising and significant developments point to the use of NIR, MIR and Raman spectroscopies combined with data mining tools as the means for analysis of fraudulent whisky and related beverages, giving greater confidence in quality evaluation and adulterant analysis. It has been also demonstrated by several authors that both the accuracy and robustness of the methods described are comparable to those obtained by traditional analytical tools such as GC-MS techniques. The field of study however is still in its early stages and it should be noted that the application of calibration models requires continuous validation and as it is the critical step to ensure the robustness of the method.

References

1. Murphy, J.P., *Poitín: A Spirit of Rebellion and Inspiration.* 2016.
2. Piggott, J., *Whisky*, in *Current Developments in Biotechnology and Bioengineering*. 2017, Elsevier. p. 435-450.
3. Parliament, E. and Council, *Regulation (EC) No 110/2008 of the European Parliament and of the Council of 15 January 2008 on the definition, description, presentation, labelling and the protection of geographical indications of spirit drinks and repealing Council Regulation (EEC) No 1576/89.* Off J Europ Union, 2008. **39**: p. 16-54.
4. Union., E., *Consolidated version of the Treaty on the Functioning of the European Union (TFEU).* , E. Union, Editor. 2016: Official Journal of the European Union, C202 (7.6.2016), 47-388.

.5. Lee, K.Y.M., et al., *Origins of flavour in whiskies and a revised flavour wheel: a review.* Journal of the Institute of Brewing, 2001. **107**(5): p. 287-313.

6. Lyons, T.P., *Production of Scotch and Irish whiskies: Their history and evolution*, in *The Alcohol Textbook*. 1999, Nottingham University Press Nottingham, UK. p. 137-164.
7. Storrie, M.C., *The scotch whisky industry.* Transactions and Papers (Institute of British Geographers), 1962(31): p. 97-114.
8. Bower, J., *Scotch Whisky: History, Heritage and the Stock Cycle.* Beverages, 2016. **2**(2): p. 11.
9. González-Arjona, D., et al., *Authentication and differentiation of Irish whiskeys by higher-alcohol congener analysis.* Analytica Chimica Acta, 1999. **381**(2-3): p. 257-264.
10. Quinn, D., *Irish whiskey*, in *Whisky*. 2014, Elsevier. p. 7-16.
11. Lyons, T.P., *North American whiskies: a story of evolution, experience, and an ongoing entrepreneurial spirit*, in *Whisky*. 2014, Elsevier. p. 39-48.
12. Podvia, M.W., *Bourbon and the Law: A Brief Overview.* Unbound: Rev. Legal Hist. & Rare Books, 2015. **8**: p. 5.
13. Horrocks, S.M., *The History of the Distillers Company, 1887-1939: Diversification and Growth in Whisky and Chemicals.* Business History, 1996. **38**(4): p. 99-101.
14. Tachau, M.K.B., *The whiskey rebellion in kentucky: a forgotten episode of civil disobedience.* Journal of the Early Republic, 1982: p. 239-259.
15. Rarick, C.A. and C.C. Mich, *The American whiskey renaissance: The rebirth of an American spirit.* Journal of the international academy for case studies, 2015. **21**(3): p. 149.
16. Humphreys, M., *An Issue of Confidence: the Decline of the Irish Whiskey Industry in Independent Ireland, 1922-1952.* Journal of European Economic History, 1994. **23**(1): p. 93.
17. Weir, R.B., *In and Out of Ireland: The Distillers Company Ltd. and the Irish Whiskey Trade 1900–39.* Irish economic and social history, 1980. **7**(1): p. 45-65.
18. BordBia, *Irish Food, Drink & Horticulture. Export Performance & Prospects. 2019 - 2020*. 2019.
19. O'Connor, A., *Brewing and distilling in Scotland - economic facts and figures*. 2018, Scottish Parliament Information Centre (SPICe).
20. States, D.S.C.o.t.U., *2018 Annual Economic Briefing Presentation*. 2019, Distilled Spirits Council of the United States.
21. Chaudhry, P.E., et al., *Preserving intellectual property rights: Managerial insight into the escalating counterfeit market quandary.* Business Horizons, 2009. **52**(1): p. 57-66.
22. Green, R.T. and T. Smith, *Executive insights: Countering brand counterfeiters.* Journal of international Marketing, 2002. **10**(4): p. 89-106.

23. Kotelnikova, Z., *Explaining counterfeit alcohol purchases in Russia.* Alcoholism: clinical and experimental research, 2017. **41**(4): p. 810-819.
24. Neufeld, M. and J. Rehm, *Effectiveness of policy changes to reduce harm from unrecorded alcohol in Russia between 2005 and now.* International Journal of Drug Policy, 2018. **51**: p. 1-9.
25. Commerce, I.C.o., *The Economic Impacts of Counterfeiting and Piracy*. 2017, International Chamber of Commerce
26. BBC *Third of rare Scotch whiskies tested found to be fake*. BBC News, 2018.
27. Cameron, C., *Dram and blast: third of vintage Scotch whisky found to be fake*, in *The Times*. 2018.
28. Carrell, S., *Rare whisky market flooded with fakes, says dealer*, in *The Guardian*. 2018.
29. Neufeld, M., et al., *Surrogate alcohol containing methanol, social deprivation and public health in Novosibirsk, Russia.* International Journal of Drug Policy, 2016. **37**: p. 107-110.
30. Neufeld, M., H.-U. Wittchen, and J. Rehm, *Drinking patterns and harm of unrecorded alcohol in Russia: a qualitative interview study.* Addiction Research & Theory, 2017. **25**(4): p. 310-317.
31. Aghababaeian, H., L. Araghi Ahvazi, and A. Ostadtaghizadeh, *The methanol poisoning outbreaks in Iran 2018.* Alcohol and alcoholism, 2019. **54**(2): p. 128-130.
32. Ahmad, K., *Methanol-laced moonshine kills 140 in Kenya.* The Lancet, 2000. **356**(9245): p. 1911.
33. áJohn Dennis, M., *Recent developments in food authentication.* Analyst, 1998. **123**(9): p. 151R-156R.
34. Diviák, T., J.K. Dijkstra, and T.A. Snijders, *Poisonous connections: a case study on a Czech counterfeit alcohol distribution network.* Global Crime, 2020. **21**(1): p. 51-73.
35. McKee, M., R. Adany, and D.A. Leon, *Illegally produced alcohol*. 2012, British Medical Journal Publishing Group.
36. Humayun, S.G.J.G.H., *Toxic moonshine kills 154 people and leaves hundreds hospitalized in India* 2019, CNN.
37. Roger, C., *Social Media Misinformation That Led to 300 Deaths in Iran Claimed That Drinking Methanol was a Cure for COVID-19*. 2020.
38. da Silva, A.R., et al., *Biochemical and hematological effects of acute and sub-acute administration to ethyl acetate fraction from the stem bark Scutia buxifolia Reissek in mice.* Journal of ethnopharmacology, 2014. **153**(3): p. 908-916.
39. Jackson, D.S., D.F. Crockett, and K.A. Wolnik, *The indirect detection of bleach (sodium hypochlorite) in beverages as evidence of product tampering.* Journal of forensic sciences, 2006. **51**(4): p. 827-831.
40. Kwiatkowski, A., et al., *Detection of denatonium benzoate (Bitrex) remnants in noncommercial alcoholic beverages by raman spectroscopy.* Journal of forensic sciences, 2014. **59**(5): p. 1358-1363.
41. Gemma, S., L. Vittozzi, and E. Testai, *Metabolism of chloroform in the human liver and identification of the competent P450s.* Drug metabolism and disposition, 2003. **31**(3): p. 266-274.
42. Lachenmeier, D.W., *Is there a need for alcohol policy to mitigate metal contamination in unrecorded fruit spirits?* International Journal of Environmental Research and Public Health, 2020. **17**(7): p. 2452.
43. Bonar-Bridges, J., *The Proof Is on the Label-Protecting Kentucky Bourbon in the Global Era.* Ky. J. Equine Agric. & Nat. Resources L., 2015. **8**: p. 491.
44. Soon, J.M. and L. Manning, *Developing anti-counterfeiting measures: The role of smart packaging.* Food Research International, 2019. **123**: p. 135-143.
45. Cozzolino, D., *Editorial overview: Innovation in food science—food fraud.* Current Opinion in Food Science, 2016(10): p. iv-v.
46. Cozzolino, D., *Food Adulteration*, in *Spectroscopic Methods in Food Analysis*. 2017, CRC Press. p. 353-362.

47. Ellis, D.I., et al., *A flavour of omics approaches for the detection of food fraud.* Current Opinion in Food Science, 2016. **10**: p. 7-15.
48. Manning, L., *Food fraud: Policy and food chain.* Current Opinion in Food Science, 2016. **10**: p. 16-21.
49. El Sheikha, A.F., *DNAFoil: Novel technology for the rapid detection of food adulteration.* Trends in food science & technology, 2019. **86**: p. 544-552.
50. Ellis, D.I., et al., *Point-and-shoot: rapid quantitative detection methods for on-site food fraud analysis–moving out of the laboratory and into the food supply chain.* Analytical Methods, 2015. **7**(22): p. 9401-9414.
51. Freitas, J.M., et al., *A portable electrochemical method for cocaine quantification and rapid screening of common adulterants in seized samples.* Sensors and Actuators B: Chemical, 2017. **243**: p. 557-565.
52. Li, B., et al., *Rapid detection of authenticity and adulteration of walnut oil by FTIR and fluorescence spectroscopy: A comparative study.* Food chemistry, 2015. **181**: p. 25-30.
53. Limm, W., et al., *A portable mid-infrared spectrometer and a non-targeted chemometric approach for the rapid screening of economically motivated adulteration of milk powder.* International Dairy Journal, 2018. **85**: p. 177-183.
54. Lohumi, S., et al., *A review of vibrational spectroscopic techniques for the detection of food authenticity and adulteration.* Trends in Food Science & Technology, 2015. **46**(1): p. 85-98.
55. Mu, T., et al., *Portable detection and quantification of olive oil adulteration by 473-nm laser-induced fluorescence.* Food analytical methods, 2016. **9**(1): p. 275-279.
56. Minnens, F., et al., *Ensuring the Integrity of the European food chain.*
57. de Oliveira, L.P., et al., *Forensics in hand: new trends in forensic devices (2013–2017).* Analytical methods, 2018. **10**(43): p. 5135-5163.
58. EC, *Commission Regulation (EC) no. 2870/2000 of 19 December 2000, Laying Down the Community Reference Methods for the Analysis of Spirits Drinks*. 2000, Official Journal of the European Communities Brussels.
59. Evans, L., *New law in the EU.* European Food and Feed Law Review, 2016. **11**(3): p. 226-228.
60. Latimer Jr, G.W., *Official methods of analysis of AOAC INTERNATIONAL 20th edition, Appendix D, Guidelines for collaborative study procedures to validate characteristics of a method of analysis.* Gaithersburg, MD, USA, 2016.
61. Adams, M.J., *Chemometrics in analytical spectroscopy*. 2007: Royal Society of Chemistry.
62. Geladi, P., *Chemometrics in spectroscopy. Part 1. Classical chemometrics.* Spectrochimica Acta Part B: Atomic Spectroscopy, 2003. **58**(5): p. 767-782.
63. Geladi, P., et al., *Chemometrics in spectroscopy: part 2. Examples.* Spectrochimica acta part b: atomic spectroscopy, 2004. **59**(9): p. 1347-1357.
64. Workman JR, J.J., et al., *Review of chemometrics applied to spectroscopy: 1985-95, Part I.* Applied Spectroscopy Reviews, 1996. **31**(1-2): p. 73-124.
65. Kemsley, E.K., M. Defernez, and F. Marini, *Multivariate statistics: Considerations and confidences in food authenticity problems.* Food Control, 2019.
66. Cozzolino, D., *The use of correlation, association and regression to analyse processes and products.* Mathematical and statistical methods in food science and technology. Wiley Blackwell, Chichester, 2014: p. 19-30.
67. Truong, V.K., et al., *From Academia to Reality Check: A Theoretical Framework on the Use of Chemometric in Food Sciences.* Foods, 2019. **8**(5): p. 164.
68. Wold, S., *Chemometrics; what do we mean with it, and what do we want from it?* Chemometrics and Intelligent Laboratory Systems, 1995. **30**(1): p. 109-115.

69. MacKenzie, W. and R. Aylott, *Analytical strategies to confirm Scotch whisky authenticity. Part II: Mobile brand authentication.* Analyst, 2004. **129**(7): p. 607-612.
70. Martins, A.R., et al., *Discrimination of whisky brands and counterfeit identification by UV–Vis spectroscopy and multivariate data analysis.* Food chemistry, 2017. **229**: p. 142-151.
71. Joshi, I., et al., *Influence of the scanning temperature on the classification of whisky samples analysed by UV-VIS spectroscopy.* Applied Sciences, 2019. **9**(16): p. 3254.
72. Pontes, M., et al., *Classification of distilled alcoholic beverages and verification of adulteration by near infrared spectrometry.* Food Research International, 2006. **39**(2): p. 182-189.
73. Sujka, K. and P. Koczoń, *The application of FT-IR spectroscopy in discrimination of differently originated and aged whisky.* European Food Research and Technology, 2018. **244**(11): p. 2019-2025.
74. Large, J., et al. *Detecting forged alcohol non-invasively through vibrational spectroscopy and machine learning*. in *Pacific-Asia Conference on Knowledge Discovery and Data Mining*. 2018. Springer.
75. Backhaus, A., et al. *Classifying Scotch Whisky from near-infrared Raman spectra with a Radial Basis Function Network with Relevance Learning*. in *ESANN*. 2012.
76. Picque, D., et al., *Discrimination of cognacs and other distilled drinks by mid-infrared spectropscopy.* Journal of agricultural and food chemistry, 2006. **54**(15): p. 5220-5226.
77. Chen, D., et al., *Detection of lethal fake liquors using digitally labelled gas-phase Fourier transform infrared spectroscopy.* Spectroscopy Letters, 2019. **52**(3-4): p. 204-210.
78. Wiśniewska, P., et al., *Authentication of whisky due to its botanical origin and way of production by instrumental analysis and multivariate classification methods.* Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 2017. **173**: p. 849-853.
79. Ellis, D., et al., *Rapid through-container detection of fake spirits and methanol quantification with handheld Raman spectroscopy.* Analyst, 2019. **144**(1): p. 324-330.
80. Ellis, D.I., et al., *Through-container, extremely low concentration detection of multiple chemical markers of counterfeit alcohol using a handheld SORS device.* Scientific reports, 2017. **7**(1): p. 1-8.