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REVIEW PAPER

The adulteration of food, lessons from the past, with reference to butter, margarine and fraud

H. Deelstra · D. Thorburn Burns · M. J. Walker

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Abstract The history of food adulteration and fraud and attempts at their control from the Middle Ages to date is traced for Belgium and for the UK with special reference to butter and margarine. The development of analytical procedures for the authentication of milk fat is outlined, from those based on the characterisation of fatty acids derived from milk fat in the nineteenth century to chromatographic methods in the next century and the recent rapid spectroscopic approaches. The importance of adequate surveillance programmes to reduce the incidence of food fraud is stressed.

Keywords Butter · Margarine · History · Adulteration · Fraud

Introduction

Food adulteration and fraud and attempts at their control have a long history. Herein is traced that for Belgium and the UK with special reference to butter and margarine. Trade in butter was radically affected by the introduction of margarine in Europe in the late 1860s, followed by its large-scale manufacture in the USA. The adulteration of fats and oils remains a problem for analytical chemists, as well as for the control authorities, when food supplies increase globally at a slower rate than urban populations in many countries. The development of analytical and legal control measures from the Middle Ages to current practice is reviewed in order to provide recommendations for the protection of consumers and food chain integrity in the twenty-first century.

History of the control of adulteration of food in Belgium

The development of food law in Belgium

In the Middle Ages, groups of traders formed Guilds, which exerted a powerful influence on the regulation of commerce of food. In Antwerp, there is a copy of the oldest known “Keure”, an official collection of laws on foods published in 1312 [1]. This document is remarkable in the number and the details given of the practical provisions made for the protection of the consumers’ material interests and health. For five centuries, the Guilds and similar corporations expanded food control rules in Belgium and in other parts of Europe. However, the revolution of 1789 in France swept away their Guilds and the associated hierarchies, and the freedom of industry and trade was proclaimed. In many other countries in the same period, initiatives in food control were taken over by the states’, or by local or municipalities’ authorities [2].

After 1830, the year of Belgian independence, increase in industrialisation and urbanisation facilitated the adulteration of foods. The local authorities then enlisted the help of chemistry professors in the Belgian universities, as they were experts for legal purposes; e.g. C. A. Bergsma (1798–1859) was asked to control bread production in Ghent, as

H. Deelstra
University of Antwerp, Campus Drie Eiken, Universiteitsplein,
2610 Wilrijk, Belgium

D. T. Burns (✉)
School of Chemistry and Chemical Engineering, The Queen’s
University of Belfast, Belfast BT9 5AG, Northern Ireland, UK
e-mail: d.t.burns@qub.ac.uk

M. J. Walker
Laboratory of the Government Chemist, Queens Road,
Teddington, London TW11 0LY, UK

was F. M. L. Dony (1822–1896) of the University of Ghent who was also an expert of the adulteration of flour who published on this in 1847, 1849 and 1852. In Antwerp, a military pharmacist, F. L. Acar (1809–1881), published the first Belgian book on food adulteration, "Traité des Falsifications des substances alimentaires et les moyen de les reconnaître" in 1848 [3].

Food adulteration was a key topic at the International Congresses on Hygiene in 1851 and 1852 organised by the Belgian National Health Council, established on 15 May 1849. The second conference, from 20 to 23 September 1852, was one of the first International Congress in Belgium with a large number of foreign participants (57 of the 300 were non-Belgians). There were four scientific sections; two of analytical interest, namely the second, on drinking water and the fourth, on the adulteration of foods [4].

The first legal initiative by the Belgian government was the Law of March 17th 1856, stating that adulteration of food was a crime. The law was mainly intended to prevent unfair competition [5]. However, mirroring the UK situation at the time, a major impediment to its enforcement was the absence of control organisations, necessary to prove scientifically that adulterations had taken place.

The position was rectified largely as a result of efforts by Jean Baptiste Depaire (1842–1910), a pharmacist, the "Belgian father of food chemistry". In 1864, he became Professor in the Pharmacy School at the University of Brussels. He was a member of the Academy for Medicine (1855), of the National Health Council (1855) and was an elected member of the Brussels community council in 1854. He was able to convince the Mayor of Brussels, Charles De Brouckère (1798–1860), on 10 May 1856, that a laboratory for food control should be established [6, 7], and this was established in the City Hall of Brussels, but was reorganised and moved in 1871 to a building close by; the first director was H. Bergé (1835–1911), Professor at the University of Brussels. Following this first Food Control Laboratory, several others were established in Belgium [5].

Nineteenth century Belgian pharmacists made important contributions to food chemistry shown by their contributions, such as in the Journal de Pharmacie d'Anvers (established in 1845). Food adulteration was extensively covered in the 6th International Pharmaceutical Congress, from 31 August to 6 September 1885 in Brussels. The Chairman of this Congress was Désiré Henri Van Bastelaer (1823–1907) and the General Secretary, Ernest Van de Vyvere. There were 728 participants from 25 countries, 231 were non-Belgians [8]. An important new aspect was the attention to questions in the domains of pure and applied chemistry and biology, e.g. the falsification of foodstuffs and the quality and characteristics of drinking water alongside pharmaceutical education and the creation of a Universal Pharmacopoeia [9]. A report on adulteration of butter by Eugène Joseph Nihoul (1857–1894), a pharmacist, was discussed in one of the four specialised sections (others addressed professional affairs (2) and biological topics). Nihoul had been asked, prior to the Congress, to look for techniques to determine whether butter was adulterated with margarine, because as he noted, the commerce of butter in the country of Herve was the only means of existence for the local farmers [10]. Pharmacists welcomed the expansion of their activities into these new areas. The General Secretary stated, "We are happy with the extension, which has been given to the international congresses by discussing apart from the former subjects, also to include those problems, dealing with the public health and hygiene" [11].

Dairy chemistry was a key part of studies at the first State Agricultural Institute, established in Gembloux in 1860. In 1870, the Catholic University of Louvain started a Higher School for Agriculture. Practical agricultural chemistry received its greatest stimulus from the foundation of an Agricultural Research Institute at Gembloux in 1871 where in addition to academic, research assistance was given to farmers and cattle breeders. In 1872, Arthur Petermann (1845–1902), an authority on agricultural chemistry, became director of this institute, previously working in the Agricultural Institute at Weende near Göttingen under the direction of William Henneberg (1825–1890), a former pupil of Justus von Liebig (1803–1873). After the establishment of the Institute of Gembloux, Petermann became involved in the creation of other State Agricultural Laboratories [12].

"Butter is without any question the most counterfeited food" was the opening sentence [13] at a lecture at the symposium organised at the "Grand Concours International of Sciences and Industry" in Brussels, 1–9 September 1888 [6]. Belgian chemists interested in food adulteration and analysis met collectively for the first time [14] at the symposium dealing specifically with food adulteration and deterioration. This followed from extensive discussions about public health at previous congresses (Brussels, 1875 and 1885; Amsterdam, 1879; Geneva, 1882; The Hague, 1884 and Vienna 1887) and the efforts of the dynamic Belgian delegate, Théodore Belval (1832–1897), a pharmacist (also a PhD in sciences). Besides butter and margarine, the "Grand Concours" also dealt with the analysis of beer, wines, drinking water, mushrooms, syrups, milk, meat, alcoholic beverages and pepper. J. B. Depaire stressed in the first lecture "The necessity to take measures in order to restrain the adulteration of foods", and these included the need at university level for specific training in Food Chemistry [15].

At the "Grand Concours", an exemplary and temporary food control laboratory was set up on the advice of "eminent chemists" from the four Belgian Universities, namely

Armand Jorissen of Liège (1853–1920), Edouard Dubois of Ghent (1842–1892), Jean Baptiste Depaire of Brussels and Charles Blas of Louvain (1839–1919) [16].

On 30 March 1887, a few chemists in the sugar industry involved their colleagues in discussions on the harmonisation of a method for the determination of sugar in beet. In due course, the sugar chemists considered it would be useful to enlarge their group with chemists of other disciplines. They decided to create a general chemical society, established 4 August 1887, as the Association Belge des Chimistes [17, 18]. During the meeting of 5 September 1888, one member proposed a separate section about the adulteration of foods. At the first meeting, 20 March 1889, a tremendous interest was evident. Eighty-six new members joined, including 45 pharmacists. Several future professors in food chemistry were present. Désiré Alexander-Henri Van Bastelaer and A. Jorissen were elected as President and Secretary, respectively. The members of the section "food" became very active in their section and in the whole association [19].

The year 1890 was important for food chemistry in Belgium, since in that year three important laws were established. The first law (April 10th) started the complete reorganisation of university education; research was to become more important, and it also revised the curricula, especially those for pharmacy when study of the adulteration of food became mandatory. The same year, Law of October 17th, at each Belgian University-specific professors for these courses were nominated. Finally, the law of August 4th concerning food adulteration was promulgated, which gave the minister permission to take all necessary measures to combat food adulteration. This law was so fundamental that it remained unaltered until 1964 (Law of June 20th, 1964) [20].

Also in 1890, the powerful and influential organisation, the "De Boerenbond", was created along with their publication, "De Boer" (The farmer). De Boerenbond was a strong association of catholic farmers and was very well organised. This association played an important role in the struggle against the falsification of butter [21, 22].

At nearly every meeting of the Food Section in the Association Belge des Chimistes, the adulteration of butter and its control were discussed.

A very surprising achievement of the young Belgian Association of Chemists was the organisation of the first International Congress for Applied Chemistry in 1894 [23, 24]. Every section of the Association was, during the preparation of the Congress, to propose questions on topics of importance to all chemists in Europe. The Food Section raised five questions. The first was what are the current good techniques to determine the adulteration of butter? Jules Victor Wauters (1852–1949) was made responsible to produce a report "on the determination of analytical techniques to identify the purity of butter". After discussions during the Congress, Wauters' proposed eight methods to evaluate butter fatty acids were accepted [25]. In the overview (see Appendix 1) of the chemical methods, the iodine number of Arthur von Hübl (1853–1932) was included because vegetable oils containing unsaturated fatty acids were used in the production of margarine [26].

Adulteration of butter

It is convenient to divide the history of the adulteration of butter into two periods; those prior to and after the commercial manufacture of margarine. Margarine was invented by Hippolyte Mège Mouriès (1817–1880), who in 1869 patented his process in France and in England. In 1871, he sold his knowledge (there was no patent law in the Netherlands at that time) to the Dutch firm of Antoon Jurgens (1805–1880) and his British, American and Prussian patents to various buyers in 1873 and 1874 [27 (a)]. Over the years, the manufacture of margarine increased almost exponentially [27 (b)] using a variety of raw materials [27 (c)].

Before the production of margarine

In 1848, Acar [3 (a)] noted that the composition of authentic butter was not always the same. However, in some samples of commercial butter cheaper substances could be found, e.g. chalk and potato starch. These adulterations are easy to detect; by melting the so-called butter in water at 60 °C, the butter rises to the surface and the foreign substances precipitate. If part of the deposit is boiled in water, starch can be detected by the addition of iodine solution. The other part of the deposit when treated with vinegar gives a strong effervescence if chalk was present. The calcium acetate produced could be confirmed by the addition of ammonium oxalate to precipitate calcium oxalate. Acar considered it was not important to analyse for the presence of natural colours such as saffron, the sap of yellow carrots and other artificial colourings added to give butter a nice yellow colour, because they were without danger. If other substances were used to adulterate butter, chemistry could not help. It was up to the buyer of butter to use his mouth to taste the falsification [3 (a)].

After the start of the production of margarine

With the arrival of margarine, a new phase arrived in the counterfeiting of butter. Not only did sellers try to sell margarine as if it were butter, but mixing much cheaper margarine into the butter became very important and nearly became established as a regular industry. Falsely labelled "butter" flooded into Belgium and such fraud existed all over Europe.

Falsification of butter in Belgium had an important political impact, because the Catholic Party had an absolute majority in the Parliament. From 1884 to 1914, all governments in Belgium were constituted with Catholic Ministers. These governments were obliged to defend the economic interests of the farmers, their key electorate.

The Directors of the State Agricultural Laboratories were asked to study the falsification of butter. We can read in an extract of the minutes of January 1886 they proposed the method of Otto Hehner (1853–1924) [28, 29] that for 5 g of butter, the percentage of insoluble fatty acids should be 89 %. Butters with higher percentages should be considered as non-pure, and the Emile Reichert method (1838–1894) [30] should be used for confirmation [31].

The activities of Belgian chemists before the creation of the "food adulteration" section in the Belgian Association of Chemists (1887) are not well documented. In preparation for the "Grands Concour" in 1888, all Belgian chemists working in the food control laboratories were asked for the numbers of samples of foods analysed and those detected adulterated during the last decade. The average percentage of foods adulterated included: milk: 55 %; butter: 24 %; chocolate: 23 %; syrups and jams: 20 %; olive oil: 20 %; wines: 15 %; vinegar: 12 % and pepper: 10 % [32].

A. Jorissen reviewed methods for the confirmation of foreign fats in butter at the "Grands Concour" and at the first meeting of the "food adulteration" section of the Association [33, 34]. He insisted that different methods should be used if conclusions on samples were to be regarded as definitive. He noted that due to the variability of milk fat composition in pure butter in samples from Belgium and other countries, there was not always proof of adulteration even if the presence of insoluble fatty acids or of volatile fatty acids seems to give the impression that small quantities of foreign fats had been added. Hence, it is scientifically inadmissible to evaluate the degree of a sample's adulteration by volatile fatty acid values alone. He reported that the method of Hehner was used very much in Belgium, but not in Germany. In the case of pure butter, the Hehner value was 88.5 % or more. He then gave information on the Emile Reichert and Emerich Meissl (1855–1905) value [35], later called the Reichert–Meissl–Wollny value after Rudolf Wollny (1846–1901) who slightly modified the Reichert–Meissl value in 1887 [36]. This method was used very much by German chemists and those from other countries. This value is an indicator of how much volatile fatty acid soluble in water can be liberated from fat through saponification. In 1888, A. Jorissen analysed 20 different butter samples and found an average of 24 for the Reichert–Meissl value [34]. Another method to identify adulteration is that of C. Königs [37] who recommended the use of the specific gravity at 100 °C. For margarine, the values lie between 0.859 and 0.861 and for butter between 0.866 and 0.868. Jorissen mentioned Köttstorfer's (also given as Koettstorfer's) mean molecular weight method [38] which he rejected, stating that the Meissl method was the most adequate.

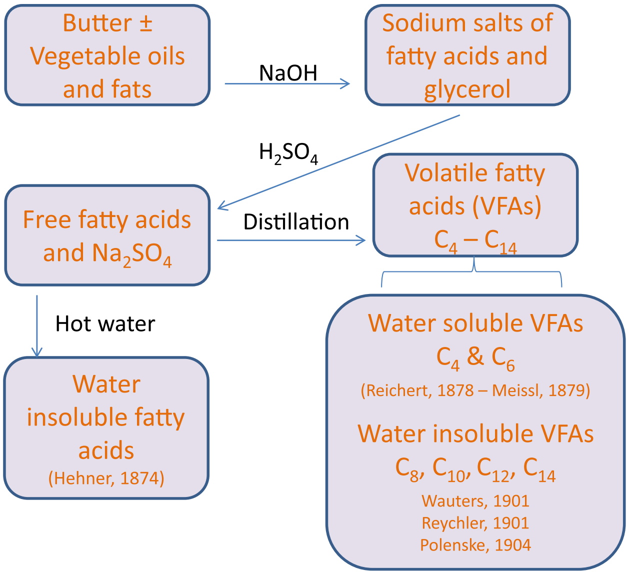

Fig. 1 Schematic diagram illustrating the chemistry of butter authenticity testing prior to gas chromatography

Appendix 1 gives a summary of the physical and chemical methods used at the period for characterising fats and oils, mentioned herein. The relationships between methods based on the volatile fatty acids content are shown diagrammatically in Fig. 1.

Figure 1 shows the relationships between the chemical methods used to determine the authenticity of butter prior to the development of gas chromatographic methods.

In a letter to the President of the Food Section, Auguste Zune (living in Paris) proposed a special commission of the Food Section to review the methods for the analysis of butter and give recommendations on the interpretation of the method's limit values [39]. On 18 October 1890, A. Jorissen supported Zune's proposal as did D. Van Bastelaer, President of the Food Section, saying "Our section is a true/active commission for the study of the analysis of butter, because this question is on the agenda of each meeting and most of the members have already presented research on this subject" [40]. The members of this Commission were A. Bilteryst, Jules Wauters and Armand Jorissen. The complete report on the research on the presence of foreign fats in butter was published in September 1893, in the Bulletin [41]. This report was discussed during the meetings of 18 November 1893 and 12 May 1894. It was concluded that four different measurements should be carried out before there should be doubt on the purity of butter, and these were the melting point and the way of the melting, the specific gravity at 100 °C, the index of refraction and finally, added in the last meeting, the Meissl value which should not be lower than 26. If there was doubt, the method of Hehner and others should be used before it is possible to declare a sample of butter as abnormal or suspect.

Period 1890–1895

The Belgian Law on food adulteration passed 4 August 1890 was simple and short but modern in the sense that allowed the Minister in charge permission to promulgate Royal Decrees when necessary. The purpose of this law was to regulate the trade in food to protect public health and also to prevent food adulteration. The Minister should also regulate and set up laboratories to control food adulteration. The law contained only eight articles. The last article stated that the Government should inform the Parliament every 2 years concerning all measurements and results made to implement the law.

The first Royal Decree was that of 10 December 1890 on the trade of artificial butter [42]. This specific law was discussed in the Parliament at the same time as the discussion of the general and fundamental law. The dual discussion was aimed to show the Parliament how the general law should perform in future. The first law on the trade of butter and margarine was relatively short [42]. Article one defined margarine, stating that “margarine is an artificial butter, suggesting an analogy with natural butter and was not produced exclusively by means of milk”. The trade in margarine was described in detail (articles 2–5), and margarine should be sold in the form of cubes, to distinguish it from butter. Also the name “margarine” should be stated very clearly on the packing. In the Royal Decree of June 22nd 1891, the members of the Food Inspectorate were nominated [43]. Article eight of this decree concerned the unification of the analytical methods to be used in food adulteration [44]. On 16 May 1892, the Minister sent a circular letter to all the directors of the laboratories agreed by the inspectorate, to implement article eight, especially on the analysis of butter. In his first biennial report to the parliament gave the Government a detailed report on the views of experts on the analysis of butter on (i) choice of the methods, (ii) description of the method of analysis and (iii) the interpretation of the results and conclusions [45]. The experts were as follows P. Warsage, Ch. Masson, D. Crispo, M. de Molinari, A. Bilteryst, D. Van Bastelaer, H. Spinette, J. Wauters, A. Jorissen, M. Pirney and M. Mercier. All, except Pirney, were active members of the Belgian Association of Chemists. It is clear that there was considerable overlap of the members in Food Section of the Association and the persons giving advice to the Minister.

By official notice, the Minister of Agriculture L. De Bruyn (1838–1908) sent the report of the eleven experts to the National Health Council and the Inspectorate asking their opinion on it [46]. The National Health Council was very pleased with the report. They proposed that the Minister take it into account along with the resolutions which would be taken soon by the Food Section of the Belgian Association of Chemists [41]. J. B. André, Head of the Inspectorate, came to the conclusion that the best way to control the butter was to use simultaneously the methods of Reichert (Meissl and Wollny) and Hehner. When the values would be in the limits of 19–23 or 88–90, it should be necessary first to measure the specific gravity, then to use Köttstorfer’s method and finally the refractive index. Finally, he proposed that the Minister to invite experts to come to a final conclusion on how to distinguish between pure and suspect butter [47].

On 26 July 1893, eight experts on the adulteration of butter, seven analysts of other specialities and five delegates of the inspectorate met to discuss the unification of methods to investigate foreign fats in butter. First, they took note of the conclusions of the Food Section of the Belgian Association of Chemists [41], and then they adopted a schematic system of how to distinguish pure butter, abnormal butter and adulterated butter. It was clear that there was a range of values of the parameters between which it was not possible to judge whether the sample of butter was adulterated or not [48].[1]

Period 1895–1900

Since the Royal Decree of December 10th 1890 on the trade of artificial butter had not been efficient to prevent adulteration of butter, on 11 March 1895, a new Royal Decree on the trade of butter and margarine was promulgated. The first article as in the Decree of 1890 also gave a definition of butter, and butter was exclusively made of the fats of milk. Article two gave the conditions for the trade of butter and margarine; it was not permitted to colour margarine, and it was forbidden to sell butter and margarine in the same location. The locations where margarine was sold should clearly be indicated with a sign “Trade of Margarine” (at least 20 cm of height). The inscription of “Margarine” on the packing should be very clear (at least 2 cm in height) [49].

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[1] The full title of this specialised journal is Bulletin du service de surveillance de la fabrication and du commerce des Denrées Alimentaires, Compte rendu mensuel des mesures prises par le Gouvernement en exécution de la Loi du 4 août 1890. Published by Administration du Service Santé et de l’Hygiène publique du Ministère de l’Agiculture et des Travaux Publics, Bruxelles.

At the time, the courts were much preoccupied with cases in which the adulteration of butter which could not always be proven. By Ministerial Order on 5 August 1896, a Commission was formed to study all methods to prevent and to suppress the fraud from trade in adulterated butter. This Commission was composed of very influential members, the Chairman was Baron de Steenhault (1840–1906) of the Senate, and the members were Members of the Parliament, Professors and so on [50].

One chemist in the Belgian Association of Chemists, Leon Crismer (1858–1844), a Professor at the Military Academy, discovered a new method for the examination of fats, by determination of the critical temperature of dissolution. First, the temperature of fat in alcohol should be increased till the mixture is transparent. Then temperature is lowered till the first discernible turbidity, and this is the critical temperature of dissolution. The paper was sent to the Academy of Sciences. The first referee, Walter Spring (1834–1913), highly recommended publication in the Bulletin of the Academy and [51] the second referee, Louis Henry (1848–1911), agreed and it appeared quickly [52]. Crismer communicated further on the topic to the Food Section on 6 July 1895. This paper was accepted and published in the Bulletin of the Chemical Association [53]. This new and elegant analytical technique did not need any measuring of weight or volume; the analytical value being read from the thermometer. The alcohol used had a density of 0.8195 at 15.5 °C (with 9 % of water). This was necessary, because the butter should be dissolved under the boiling point of pure alcohol of 78 °C. The critical temperature for butter is then around 100 °C and for margarine between 120 and 124 °C. The only handicap for using this technique was that the method needed closed tubes (fusing the end of the glass tubes). Members of the Commission were very enthusiastic about this new technique [54]. Crismer improved his method to allow the use of open tubes. In a communication, 4 November 1895, to the Food Section, he demonstrated linearity between the critical temperature of dissolution of butter in alcohol and the water content of the alcohol. He showed it was possible to perform the test in open tubes, using nearly absolute alcohol (with 0.9 % of water) when the critical temperature of dissolution for butter was between 54 and 55 °C and for margarine between 74 and 78 °C [55].

At the last meeting of the Food Section, before the Association was reorganised into local sections, on 6 March 1897, Crismer gave an overview of all analytical techniques for butter. At the end, he declared “It is not possible to confirm adulterated butter by margarine, when it is mixed with butter at lower than 35 %. This difficult situation will go on till the moment a substance in margarine will be found easily to detect, but which, however, is difficult to eliminate” [56].

The difficulty in proving adulteration of butter below about 35 % admixture with margarine was communicated by the Minister of Agriculture, L. De Bruyne, to his colleague, V. Begerem (1853–1934), the Minister of Justice [57]. This and the fact that the composition of authentic butter was not always constant led the Minister of Agriculture to instruct his Inspectorate to take every 2 weeks for a complete year, samples of butter in all regions of Belgium, specifying the methods for analysis [58].

Meanwhile the search for a “marker” continued. At the general assembly of the Association of Belgian Chemists on 1 May 1898, J. Wauters discussed “the denaturising of Margarine”. He explained that the German Law of June 15th 1897 said that sesame oil (10 %) must be added margarine, and this could easily be detected with hydrochloric acid and an alcoholic solution of 2 % furfural (Baudouin reaction) [59]. Not unexpectedly, the opinions of German chemists and margarine manufacturers on this law were divergent [60]. In Belgium, the Commission created on 15 August 1896 proposed in 1897 a change to the law of March 11th 1895 that would prohibit margarine without the addition of phenolphthalein, amazing in view of its laxative characteristics. However, 7 voted against, 3 for this proposition and 3 abstained [61]. Thus, the Minister did not follow this proposition. Other possible substances were discussed such as diaminoazobenzol. Wauters came to the conclusion that this matter needed international agreement; otherwise margarine could continue to be added to butter. Indeed some manufacturers such as Samuel Van den Berg Jr. (1864–1941) in Oss, the Netherlands, made large resistances against the addition of any marker substance. Dutch delegates at the International Congresses on Dairy products in 1903, 1905 and 1907 continued resistance against adding sesame oil to margarine [29 d]. Wauters stressed in his lecture “that it was necessary to continue to battle against the adulteration of butter, but this should not be done in order to damage the production of such a useful product as margarine for the working class. The interests of agriculture should not, under any circumstances, prevail to the general interest” [60].

Period 1900–1905

On 4 May 1900, a new Law on butter and margarine with several important innovations was promulgated by the Minister of Agriculture, Maurice Van der Bruggen (1852–1919), and the Minister of Justice, Jules Van den Heuvel (1854–1926) [62]. Article ten declared that edible fats, not butter, but having the appearance of butter, should be described as margarine. Article four stated that margarine should only be sold if it contained substances that easily distinguished margarine from butter, these should be fixed later, by Royal Decree. Article ten forbade the sale of abnormal butter. The chemical and physical values to identify abnormal butter were fixed by Royal Decree [63] (see Appendix 2, a).

The Royal Decree of October 31st 1900 on butter and margarine followed the recommendations of the National Health Council [64]. Article one obliged margarine factories to mix margarine with 5 % of sesame oil and 0.5 % of potato flour. Article three gave the values for the chemical and physical values for the identification of abnormal butter [65]. These values were chosen after the analysis of all samples obtained during the sampling campaigns during 1897 and 1898 [58]. Article three and the criteria for abnormal butter caused intense discussion among Belgian Chemical experts. J. Wauters, a recognised authority and acting as an intermediary with some members of the Parliament, told the Brussels Section of the Association of Belgian Chemists, 14 May 1900, that "the law of 1900 was not all that scientific and even unjust, because there were authentic butters produced at certain periods of the year with very low content of volatile fatty acids". Wauters explained the problems with the law in the Parliament and in the Senate and gave information on a judicial sentence in Ghent, where Professor Frédéric Swarts (1866–1940), in defence of so-called counterfeiters of butter, told the judges that science was not able to declare the butter to be adulterated, as a result of which the judges acquitted the defendants. Wauters explained that problems with abnormal butter were inevitable and the offence to sell this butter was unreasonable and unfair, because in the sampling campaign, 1897 and 1898, milk from some normal cows gave butter with very low Meissl numbers values. The Brussels Section members all agreed with Wauters, and his paper with annexes of the Law and the Royal Decree of 1900 was published [66]. The General Assembly of 27 January 1901, the Association, chair Professor L.L. De Koninck (1844–1921), confirmed the opinion of the Brussels Section opinions on this and sent their views to the Minister of Agriculture [67].

From 1901, the food control inspection at the border with the Netherlands examined all imported butter by the Reichert–Meissl method. For example, in 1904, 60 % of all imported butter (3,691,000 kg) had R–M values below 25. Data directly from the butter production sites gave average R–M values in the range of 30–32. At most courts, the Dutch importers were defended by Dutch chemists and because of doubt about the values of pure butter samples, most alleged counterfeiters were acquitted [68].

Meanwhile a new fat, coconut oil, was used for the production of margarine. W-G. Indemans, a pharmacist, communicated his doctoral thesis (Free University of Brussels) to the Association of Belgian Chemists. The point of fusion of coconut oil (25 °C) made this interesting for margarine (or vegaline) manufacturers and for the adulterators of butter. Coconut oil has an R–M value of 7.6 compared to 5.87 for normal margarine. Margarine containing coconut oil could be mixed with butter to give a product with R–M value of 26.5. In Indemans' view, to control the quality of butter only using the Reichert–Meissl value was thus inadequate [69].

Belgian food chemists continued to make important contributions to the better understanding of the composition of fats. J. Wauters communicated to the Brussels Section, 16 January 1901, new values for the identification of fats. For coconut oil, he found that a large fraction of the volatile fatty acids were not soluble in hot water (15.4), besides the soluble volatile fatty acids with R–M value 11.4. This volatile non-soluble fatty acid fraction for pure butter was 0.8–0.9, with R–M value of 26–28. A mixture of 75 % butter and 25 % coconut oil had a R–M value of 24.2 and a volatile non-soluble fatty acid fraction of 5.0 [70]. In the same year, Albert Reychler (1854–1938) noted the same observation in Paris [71]. Eduard Polenske (1849–1911) published on the determination of coconut oil in butter 2 years later in 1904 [72]. Henseval in 1904 gave a comparison of three analytical methods for the determination of volatile fatty acids not soluble in hot water [73].

Since the start of coconut oil in the production of margarine, it was easy to mix margarine containing coconut oil with butter, because the Meissl values of the Royal Decree of October 31st 1900 could be achieved [74]. With the advice of a special Commission and the discussions in the Parliament and the Senate, a new Law on butter, margarine and edible fats was promulgated on 12 August 1903 [75]. Apart from a new definition for butter and the way to trade in fats, there were not big changes. The term "abnormal butter" was eliminated and instead a new text appeared, "Butters, without being sure that they are not adulterated, may not be sold if their values are deviating from the values of the majority of pure butters". These values were stated by the Royal Decree on butter, margarine and edible fats of 20 October 1903 [76] (see Appendix 2, b). However, by 27 October 1904 the National Health Council discussed extensively to change the Royal Decree of October 20th 1903. The member proposed new values to declare a sample of butter adulterated [77]. On 21 November 1904, a new Law on butter, margarine and edible fats superseded the Royal Decree of 1903 [78, 79] (see Appendix 2, c).

History of the control of adulteration of food in the UK

Development of UK food law

As in the rest of Europe, in the Middle Ages groups of traders in various parts of the UK formed Guilds to regulate commerce in precious metals, spices and drugs and other high-value items. Prior to the first general food act, bread, tea, coffee, chicory, beer and wine were dealt with by special statutes, with the objects, primarily, to prevent defrauding the revenue and protecting purchaser's health [80, 81]; for example, for bread, "Assize of Bread and ale" ca.1266 [82], coffee act of 1718 [83], coffee and tea act of 1723 [84] and tea acts of 1730 [85] and 1776 [86]. This latter act is interesting as it made it possible for dealers or manufacturers of materials used for the adulteration of tea, although not dealers in tea, to be legally punishable.

By the early nineteenth century, various chemists and medical practitioners became concerned about the quality, contamination and adulteration of food. In 1820, Fredrick Accum published "Treatise on the Adulteration of Foods and Culinary Poisons" [87]. In 1850, Thomas Wakely, secretary and founder of the journal, The Lancet, set up "The Lancet Analytical Sanitary Commission" under the direction of Arthur Hill Hassall to investigate and report on the composition of food. Hassall was a skilled microscopist and the first UK analyst to investigate scientifically food adulteration. He reported on the analysis of 2063 foods and 324 drugs and found evidence of adulteration. The reports of the Lancet Commission were published in 1855 [88]. A Select Parliamentary Committee was then appointed to make further study, finding adulteration was wide spread and the health of the public was at risk. As a result, the first Adulteration Act was passed by Parliament in 1860 [89]. This Act was amended in 1872 [90] to include the duty to appoint Public Analysts [91, 92]. By 1874, there was general dissatisfaction with the working of the Acts of 1860 and 1872 by merchants and traders who claimed they were unfairly prosecuted and a second Parliamentary Select Committee was set up in 1874 and heard about the lack of a definition of "adulteration", poor quality of analyses and inexperience of some analysts. It was suggested that the analysts consult among themselves to sort out the difficulties. Following from the Select Committee's conclusions in 1875, a new Sale of Food and Drugs Act [93] was passed which included the mandatory appointment of Public Analysts. This was subsequently modified in 1879 [94] and in 1899 [95]. Further general Acts which affected the role and work of Public Analysts include the Merchandise Marks Acts, 1887, 1891 and 1894 [96] (to deal with the mislabelling of imported foods), Public Health Act, 1907 [97] (with additional regulations as to food) and the Fertilizer and Feeding Stuffs Act, 1906 [98] (bringing materials to improve crop yield and animal feeds into the food chain and the appointment of agricultural analysts). The special laws and regulations for dairy products, milk and butter, and margarine are dealt later.

The professional organisation of public analysts

As a result of the suggestion made in the deliberations of the Parliamentary Select Committee of 1874 that analysts consult among themselves to resolve the problems identified, two Public Analysts, namely C. Heisch and G. W. Wigner, sent invitations to 77 analysts with official appointments to a meeting on 7 August 1874. Twenty-five attended and an Organising Committee was set up under T. Redwood as Chairman to consider a constitution, rules and suggestions for standards for "The Society for Public Analysts" (SPA), which was formed on 1 December 1874 [99]. In 1907, the Society changed to become "The Society for Public Analysts and Other Analytical Chemists" and in 1954 to "The Society for Analytical Chemistry" (SAC) [100]. Today, after its centenary in 1974 and amalgamation with the Chemical Society it is the "Analytical Division of the Royal Society of Chemistry" [101]. Since 1954, the affairs of Public Analysts have been dealt with by the "Association of Public Analysts". One of the first tasks of Council of the SPA was to find clear definitions for the adulteration of foods and of drugs and compositional standards for milk, tea, cocoa and vinegar.

The official referee analyst

The Select Committee of 1874 set up to discuss improvements to the 1872 Act suggested a central body to act as referee in cases where the certificate of a Public Analyst was contested. That this was to be the Inland Revenue Laboratory at Somerset House was not agreed by all Public Analysts because not all the Somerset House staff were trained chemists [99 (a), 102, 103 (a)]. Nonetheless, the 1875 Act appointed the "Chemical Officers of Somerset House" as referees and the referred cases were discussed in the Annual Reports of the Commissioners of Inland Revenue [104]. In early reports, frequent reference was made to staff training [103 (b)] and to research and method development. The referee analyst function is now carried out by the Government Chemist and his designated officers, and the laboratories annual reports continue to outline key cases and LGC contributions to method development [105].

The Analyst uc T and A

Papers on the analysis of dairy products and the dissemination of information on adulteration in the UK and beyond were in The Analyst, a new journal quickly set up by SPA, appearing first in March 1876. The early issues contain a large number of papers on dairy products. Many of the early Public Analysts made contributions to the analysis of dairy products, notable among these are Charles Alexander Cameron (1830–1921) [106], Robert Rattray Tatlock (1837–1934) [107], Alexander Wynter Blyth (1844–1921) [108], Alfred Henry Allen (1846–1904) [109] and Otto Hehner (1853–1924) [110], all in time, became Presidents of the SPA.

The extent of adulteration in the UK

The extent of the adulteration of food including dairy products in the various parts of the UK is available in the Public Analyst Quarterly Reports to their Local Government Board, and summaries of some of these appeared in the early issues of The Analyst [111].

Annual overviews were provided by Wigner for most years from 1875 to 1881 [112], giving the number and types of samples examined, the numbers of items found adulterated and the number of convictions in each local authority. In 1882, he reported "The usual although very unpleasant sequence to my annual report is to point out that there are seven counties and 43 towns, besides one Metropolitan District, St. Martins, in which the Act has been absolutely ignored, and nothing whatsoever was examined during the year. And in addition, there are seven counties and 26 towns where the amount of work performed has been utterly inadequate to the number of inhabitants" [112].

Much of the same material was reported, but from a different view point, in the Annual reports of the Local Government Board for England and Wales [113]. Another regular early feature in The Analyst were the almost monthly "Law Reports", with brief details of police-court decisions on food and drug fraud and adulteration, building up case law [114]. These developed into "Law Notes" in May 1888 [115]. Some of the more important cases, for example, the butter prosecutions at Cheltenham, were reported in detail [116].

The work load and the attitude of the local authorities can be illustrated in detail using the data for Belfast, from an archive of cuttings (Belfast Newsletter and a few from the Irish Times) and, in their absence, manuscript summaries for Summons Court cases, for 14 April 1900–19 September 1904 [117]. There were at least 199 (about 4/month) successful prosecutions for milk (watered/skimmed or containers incorrectly labelled) and buttermilk (watered) and 30 for selling margarine as butter or butter with excess water. In 1904, a few cases involved the illegal use of formalin or boric acid, in milk. Also in 1904, butter was not the only fat to be adulterated, in three cases of "dripping" (fat rendered from meat on roasting) were adulterated with up to 30 % cotton seed oil, the sellers were fined heavily. Mr. J. Burke J. P. stated in his judgement, "He fully agreed with those public men in the City who had called upon the magistrates to punish frauds of this kind with a heavy hand".

From the start, the SPA took an interest in overseas food analyses and legislation, for example in Canada [118, 119], America [120, 121], France [122, 123] and Holland [123]. Relevant to the current review is the mention by Diehl in the report from the American National Board of Health of the recent demise in Brussels of "an extensive establishment for adulteration detection, carried out with great skill" [120]. The lengthy report on adulteration in New York of butter mentions the work of English and French chemists on the topic [122], illustrating the relative ease of transfer of technical data in the nineteenth century. The reports from France [122, 123] and Holland [124] give details of the processes, equipment and samples examined in Municipal Laboratories. Not every report was on serious science; for example, attention was drawn to adulteration in Paris by an amusing abstract of an article by Cochin in Revue des deux Mondes [125] about a meal, each course being made from adulterated items. The importance of the transfer of technical data was recognised by the SPA by the "Monthly record of analytical researches into food" and "….into drugs" in The Analyst from November 1884 [126], in due course broadened to include "improvements in analytical topics" [127] which by 1954 developed into a journal, namely Analytical Abstracts.

From the 1920s, the annual reports of official laboratories in countries of the former British territories or protectorates, most having senior staff trained in the UK [128], were of interest to SPA members for employment prospects in the depression post-World War I as well as for technical information. Summaries of such reports appeared in The Analyst [129].

The expertise within the SPA, soon after its establishment, was such that in 1887 they were consulted by the German Government through its Commissioner, Dr. Rottenburgh, on food adulteration and the legal machinery for its suppression, in view of contemplated German legislation. Replies to 27 questions raised were prepared and put to members for ratification, comment or revision, before handing to Dr. Rottenburgh. The questions, the replies and outcome are in The Analyst [130–132].

The proposed America Adulteration Bill was brought to the attention of the Society in 1879 [133] as were details of the state bills for New York [134], Michigan [135] and the specific legislation for butter and cheese in Boston [136].

In the period from 1876 to 1900, 204 articles on milk and 78 on butter appeared in The Analyst. Particular attention was given to new methods and the evaluation of continental methods for butter and to the translations of key papers such as those by Koettstorfer [137] and Wollny [138]. Following publication of Reichert's method, Allen drew attention to its advantages [139, 140] as did others [141–143] and Smetham noted an improvement to the Koettstorfer process for use in the presence of mineral oil [144].

Over the years, numerous papers were published on the composition of genuine and wholesome UK butter, imported butters and samples of abnormal butter originating in Sweden (shipped via Copenhagen) [145–148], Denmark [146], Holland [149], Egypt [150], Ireland [146, 151, 152], and on variability within a specific herd of cows [153], changes in the composition of butterfat subsequent to variation in animal feed [154–158] and the effects of storage for long periods [159–162]. Although not of commercial interest, studies were made on Irish "bog butter" [163, 164], the most recent, on Scottish bog butter using a combination of molecular and isotopic techniques [165].

In addition to the analytical and compositional problems of abnormal butters, PAs and others had to develop skills to deal with butters adulterated with other fats such as coconut oil [166–173] and with margarine, and margarine with added butter [174–180] and the recognition of hydrogenated oils [181].

UK legislation with regard to butter and margarine

Margarine was imported until 1889 when Otto Mønsted, a Dane, built Britain's first margarine factory in Manchester. In 1887, two Bills were introduced to Parliament to deal with the importation, manufacture and sale of butter substitutes. The Bills were referred to a Parliamentary Committee, attended by the SPA President, A. H. Allen and O. Hehner [182]. The result, a single Act for the better Prevention of the Fraudulent Sale of Margarine in 1887 [183–185], which defined butter and margarine and stated every box margarine should be branded in capital letters, not less than three quarters of an inch square and all retail packages to have wrappers with Margarine in capital letters not less than a quarter inch square.

This legislation for butter and margarine was amended in 1899 by an act to amend the law relating to the sale of food and drugs [186] in addition to clauses concerned with milk, and Clause 8 restricted the permitted amount of butterfat in margarine to be below 10 %.

To prevent differences and dissentions in prosecutions under Clause 8, and regard to the natural variations in the composition of butter, the Principal Chemist of the Government Laboratory, Dr. Thorpe, set up a committee to discuss with him and colleagues and agree a method to estimate the proportion of butterfat in margarine: a move designed to break down the barriers and reduce disputes between the PAs and his laboratory [187]. In due course, the joint committee produced data to relate the Reichert-Wollny number of a mixture to the percentage of butterfat present, and their chosen method was endorsed by the SPA [188].

The desirability of regulations under Clause 4 of the 1899 Act was discussed by a Committee of the Board of Agriculture who made three recommendations [189]:

1. That the figure 24 arrived at by the Reichert-Wollny method should be the limit below which a presumption should be raised that butter is not genuine.
2. That the use of 10 % of sesame oil in the manufacture of margarine should be made compulsory.
3. That steps be taken to obtain international cooperation.

The next legislation was the Butter and Margarine Act 1907 [190, 191] which stood until 1938 [192]. The Act omitted the suggested use of sesame oil as a marker for margarine and the recommended value for the Reichert-Wollny number. Importantly in clause 32, it fixed the maximum permitted amounts of water in butter and in margarine at 16 and 24 % for products known as "milk blended butter".

Current international definitions of butter, margarine and derived products

The current Codex Alimentarius standard, CODEX STAN 279-1971, sets out the global standard for butter and its description, "Butter is a fatty product derived exclusively from milk and/or products obtained from milk, principally in the form of an emulsion of the type water-in-oil", and the compositional criteria (minimum milk fat content of 80 % m/m, maximum water content of 16 % m/m and maximum milk solids-not-fat content of 2 % m/m). These standards essential for the microbiological safety, organoleptic qualities and consumer preferences of what we call butter are reflected in many national food laws. They are built upon the foundations described earlier, herein. The methods of sampling and analysis are set out in CODEX STAN 234-1999.

Modern thinking on butter and its competitor products has been influenced by at least four considerations:

1. A level playing field for agricultural including dairy products,
2. Protection of traditional products such as butter,
3. Avoidance of confusion in the minds of consumers,
4. Improved nutrition.

The last area addresses concerns on increasing obesity and poor cardiovascular health by preference for less-energy dense products, avoidance of saturated fatty acids, cholesterol and trans-fatty acids and phytosterol esters for lowering plasma cholesterol levels.

There are obvious tensions between some of the above considerations, and the market in other yellow fats and spreadable fats has diversified considerably aided by more flexible sectoral regulations. For many years, the European Union main legislation on these matters included Council Regulation (EC) No 2991/94 of 5 December 1994 laying down standards for spreadable fats and Council Regulation (EEC) No 1898/87 of 2 July 1987 on the protection of designations used in marketing of milk and milk products. Both these have since been repealed and replaced by Council Regulation (EC) No 1234/2007 of 22 October 2007, which established a common organisation of agricultural markets and specific provisions for certain agricultural products. This is a complex measure concerned mainly with agricultural markets in a variety of sectors including dairy products but also wine, rice, meat species and many others [193].

Regulation 1234/2007 defines "butter" as the product with a milk fat content of not less than 80 % but less than 90 %, a maximum water content of 16 % and a maximum dry non-fat milk material content (e.g. milk proteins) of 2 % and "margarine" as the product obtained from vegetable and/or animal fats with a fat content of not less than 80 % but less than 90 %. A diverse range of other products are allowed on sale subject to clear labelling [194]:

(a) milk fat products such as butter, but also "dairy spread *X* %, 62 % < *X* < 80 %", "three quarter fat butter" or "reduced fat butter" with a milk fat content ≥ 60 % but ≤ 62 %, "dairy spread *X* %" or "reduced fat dairy spread *X* %", with a milk fat content of >41 % but <60 % and finally "half fat butter" or "low fat butter" or "light butter" with a milk fat content of ≥ 39 % but ≤ 41 %.
(b) vegetable and/or animal fat products with a milk fat content not more than 3 % of the fat content, for example margarine, lower fat analogues such as fat spread *X* %, "three quarter fat margarine" down to "minarine", "halverine" "light" or "low fat margarine" with fat contents in the ranges as under (a) and
(c) mixed milk fat and vegetable/animal fat products with milk fat content between 10 % and 80 % of the fat content, for example the "blended spreads *X* % (where *X* is the total percentage fat content) and again with sales designations and a range of fat contents mirroring those under (a) above.

The Spreadable Fats (Marketing Standards) and the Milk and Milk Products (Protection of Designations) (England) Regulations 2008, No. 1287, implemented in England the provisions of Regulation 1234/2007 with equivalent measures in the other devolved countries of the UK. These UK regulations also required the addition of standard amounts of vitamins A and D to margarine; however, this requirement has been dropped as part of the UK Government's commitment to reducing the number of regulations which industry and traders have to navigate. There was some concern that dropping the requirement would potentially lower vitamin D intakes and therefore increase vitamin D deficiency in the population. Government estimated that only a small number of small producers in the UK make a fat spread that would legally qualify as margarine. Therefore, almost all fat spreads made in the UK do not need to meet the vitamin A and D fortification requirements, but producers continue to do so on a voluntary basis [195]. The revocation of the vitamin requirements was put into effect by the Environmental Noise, Site Waste Management Plans and Spreadable Fats, etc. (Revocations and Amendments) Regulations 2013.

Products such as "brandy butter" (and the sherry or rum equivalents) are dealt with by Regulation 445/2007 and must contain a minimum of 20 % milk fat [196].

Summary of current methods for detecting the adulteration of butter

A comprehensive review of modern techniques has not been attempted, however, based on experience an indication is given, with appropriate literature references, of the main techniques applicable and those in current routine use.

It is a tribute to nineteenth century chemists that the major constituents of butter and vegetable oils were well established by the time Martin and Synge first published on gas–liquid chromatography [197], GC, which has since proved highly effective in the determination of the constituents of butter and its adulterants. In 1952, James and Martin further developed GC by separating the volatile fatty acids from formic to dodecanoic [198]. However, it was 16 years before GC was applied routinely to the analysis of butterfat and its adulterants in official food control laboratories in the UK. The Reichert–Meissl approach, known in the UK as the Reichert, Polenske and Kirschner, RPK, method persisted alongside GC for many years. For example, there were extensive experimental details in the 1991 edition of "Pearson", the food analyst's vade mecum, on the RPK method [199]. A British Standard method for RPK based on previous standards was published as late as 1976 [200] and was only withdrawn in 2009, due to the lack of use [201].

Modern analyses of butter begin with sampling, warming and shaking the sample at 32–35 °C in a closed sample jar to produce a homogenous fluid emulsion. Analysis is recommended for fat, moisture, milk solids other than fat (milk solids-not-fat, MSNF), salt (sodium and or chloride), lactose, mineral matter, rancidity parameters, acidity, diacetyl, additives and contaminants. The authenticity of butter depends on confirmation that the lipid content is cows' milk fat. If not, or for mixed fat products, it is important to estimate the relative proportions of milk fat and non-milk fat. Both the historical RPK and modern chromatographic approaches start from anhydrous fat isolated from the sample.

Kirk and Sawyer reviewed chromatographic methods to verify the authenticity of milk and butterfat, noting developments in the preparation of fatty acid methyl esters for GC [199]. Longer chain fatty acids are often diagnostic, e.g. the ratio of C12 to C10 acids is about 1.1 for butterfat but over 8 for many fats and oils used to make margarine while the C14/C12 ratio should be about 3–4 for butterfat. However, such ratios can be influenced by the animals' diet or oxidation of the fat during storage. As the short chain fatty acids, C4 to C10 are synthesised in the mammary gland they are less influenced by diet and the principle such acid, butyric (butanoic) acid, is relatively characteristic for milk fat, corresponding with the determination of volatile water-soluble fatty acids in the Reichert value (although the latter also includes caproic acid). The determination of butyric acid by GC of methyl butyrate is now well established. An IUPAC method, based on that of Christopherson and Glass, uses 2M methanolic potassium hydroxide to hydrolyse and methylate the fatty acids which are extracted into heptane for GC [202]. Phillips and Sanders described a simple method for the free butyric acid which is also widely used and begins by saponifying the anhydrous fat with 0.5 M ethanolic potassium hydroxide. After evaporation of the ethanol, addition of 5 % aqueous phosphoric acid precipitates the longer chain fatty acids and butyric acid can be determined in the filtrate by GC [203].

Ulberth compared three analytical approaches for butyric acid by GC of the free acid, GC of methyl butyrate and GC with headspace sampling. The headspace method consisted of weighing the fat into a headspace vial, adding the sodium methoxide trans-esterification reagent, crimp-sealing the vial and automated headspace GC. A reference material (anhydrous milk fat) having a certified content of butyric acid was used to check the accuracy of the methods. All methods returned results within the uncertainty range of the certified value. The methyl butyrate method had highest precision, followed by the free butyric acid method and the headspace sampling procedure. Relative standard deviations for repeatability were 0.38, 0.86 and 1.48, respectively. The headspace method had the highest sample throughput (8 samples per hour), while the methyl butyrate method only allows one sample per hour [204].

Butyric acid can also be determined by liquid chromatography, LC, directly, [205] or after derivatisation as its phenylethyl or p-bromo phenylacyl ester. Boley and Colwell have compared the GC and LC methods finding both equally facile and reliable [206]. Other techniques, such as capillary zone electrophoresis [207] and gas chromatography–mass spectrometry, GC–MS [208], have been successfully applied to free butyric acid and methyl butyrate, respectively. Hydrogenated fats/oils, often used in non-dairy spreads, are detectable by the presence of trans-fatty acids [199].

Kirk and Sawyer recommended average butyric acid content for authentic butterfat or milk fat of 3.6 % in the range 3.2–4 % [199]. The average value of 3.6 % is supported by other authorities such as IUPAC, method 2.310 (1987) [209].

Commission Regulation (EC) No 900/2008 (latest version 2011) on methods of analysis for imports of certain agricultural products specifies 4 % methyl butyrate, equivalent to 3.45 % butyric acid, in milk fat and butterfat [210].

The adulteration of butterfat can also be detected by the presence of the sitosterols and stigmasterol (phytosterols) which occur in vegetable oils but are absent in butterfat where cholesterol is the major sterol. The most common method for the determination of total sterols content in food is by direct saponification, extraction of the unsaponificable residue into a non-polar solvent and GC with flame ionisation detection, FID. LC methods are also applicable, e.g. reverse-phase LC with isocratic elution (methanol and water, 95:5), and detection at 205 nm for the determination of cholesterol and other sterols in milk and dairy products has been shown to be a fast and reliable method [211]. The amounts and profile of tocopherols in a fat can yield some information on its origin, e.g. butterfat with low α-tocopherol can be distinguished from soya oil which has higher amounts of gamma- and delta-tocopherol and low amounts of α-tocopherol [208].

In addition to the determination of the component fatty acids after hydrolysis and methylation, it is possible to examine the intact triglycerides or triacylglycerol, TAG, in fats. An international reference method, ISO 17678/IDF 202:2010, for the determination of milk fat by GC of TAG is available [212]. Vegetable and animal fats such as beef tallow and lard can be detected. Using defined triglyceride relations, the integrity of milk fat can be determined. The method is applicable to bulk milk, or products made thereof, irrespective of feeding, breed or lactation conditions. In particular, it is applicable to fat extracted from milk products purporting to contain pure milk fat with unchanged composition, such as butter, cream, milk and milk powder. However, the method is not applicable to milk fat:

(a) From bovine milk other than cow's milk;
(b) From single cows, due to individual variabilities;
(c) From cows which have been fed an exceptionally high amount of pure vegetable oils such as rapeseed oil;
(d) From colostrum;
(e) Subjected to treatment such as removal of cholesterol or fractionation;
(f) From skim milk or buttermilk;
(g) Extracted by acid hydrolysis or detergent methods. In the latter case, substantial quantities of partial glycerides or phospholipids can pass into the fat phase. Consequently, the scope of ISO 17678/IDF 202:2010

excludes certain products, particularly cheese, whose ripening process can affect the fat composition [212].

Derewiaka et al. [208] also discussed TAG analysis by reverse-phase LC with diode array detection, DAD, and identification of the TAG by LC-atmospheric pressure chemical ionisation (APCI)/MS.

TAG analysis has been successfully applied to the determination of milk fat and cocoa butter in chocolate. The approach is based on comprehensive databases covering the TAG composition of a wide range of samples of authentic milk fat, cocoa butter and cocoa butter equivalents, CBEs, using over 900 gravimetrically prepared reference mixtures. An algorithm allows quantification of the milk fat content in chocolate was. A complex multivariate statistical procedure allows quantification of CBEs in milk chocolate. In a validation trial, with twelve laboratories, no false-positive or false-negative results were reported [213]. Further information and a "toolkit" are on the website of the European Commission, Directorate-General Joint Research Centre, Institute for Reference Materials and Measurements, IRMM [214]. A fast TAG method using of a short apolar open-tubular capillary column has been described by Destaillats et al. [215].

Useful reviews of chromatographic techniques applied to milk fat and oils that have authenticity are by Derewiaka et al. [208] and Cserháti et al. [216].

There is an extensive literature on non-chromatographic approaches to ascertain the authenticity of milk fat and butterfat. Near-infrared spectroscopy (NIR) and Fourier transform infrared (FTIR) spectroscopy are well known for the rapid appraisal of food authenticity [217]. For both, model building, training and validation require extensive sets of samples of butter, oils and blends thereof. Heussen et al. [218], using principal components analysis and partial least square discriminant analysis, assessed the performance of NIR as less good than GC of butyric acid but this disadvantage can be outweighed by shorter measurement times and the lower operator skills required. Temperature-controlled attenuated total reflectance-mid-infrared (ATR-MIR) spectroscopy combined with multivariate analysis has been applied as a simple and rapid method for the determination of butter adulteration [219]. With the advent of cheaper and reliable instruments, Raman spectroscopy has been applied to butterfat. Beattie et al. have shown that Raman spectra of melted butter samples could be used to predict fatty acid profiles and bulk parameters normally obtained by standard analyses, such as the iodine value and solid fat content [220]. Although the prediction errors for the abundances of each of the fatty acids in a sample were much larger with Raman than with a GC analysis, the accuracy was acceptable for quality control applications. This result, and because Raman spectra can be obtained with no sample preparation and with 60-s data collection times, means that high-throughput, online or on-site Raman analysis of butter samples should be possible.

Koidis et al. [221] have discussed the limitations of chromatographic methods and the emergence of spectroscopic methods coupled with multivariate data analysis for the identification of extracted and refined vegetable oils in admixture and an approach to meet the EU legislational demands for correct vegetable oils labelling, due to be introduced in December 2014 [222].

Lastly, as an example of the sophistication of modern adulteration Picariello et al. [223] recently reported the use of a synthetic TAG to adulterate butterfat, a practice difficult to detect with the classical methods of analysis, especially when the adulterating TAGs contain fatty acids in proportions similar to those of genuine butter. They applied mono-dimensional 13C NMR spectroscopy to determine the distribution of butyric acid on the glycerol backbone to provide a diagnostic differentiation of genuine butter from mixtures with synthetic TAG. The method achieved reliable quantification up to 2.5 % w/w of adulterating fats (LOQ) and can detect quantities as low as 1 % w/w of synthetic TAGs blended with butter (LOD).

Conclusions

Food adulteration, fraud and their control have a long history. The recent horsemeat episode [224] shows that historical problems can re-arise causing massive loss of consumer confidence with significant economic consequences. Review of the history of food authenticity in Belgium and the UK, with reference to butter and margarine, allows some general conclusions.

It should be remembered that even after individual fatty acids were identified, it was impossible in the nineteenth century to identify the individual fatty acids in complex mixtures [65]. Chemists working on the adulteration of butter at the turn of the nineteenth/twentieth centuries dealt with significant problems due to the lack of specific and selective methods. Fats are complex mixtures of triacylglycerols, with a wide spectrum of fatty acids, and other lipid components. The fatty acid composition of butter is influenced by many factors including that of the producing animal's diet. Only after the introduction of chromatographic methods in the 1950s was it possible to separate and to quantify the individual fatty acids.

Viewed alongside modern chromatographic techniques, the ingenuity and experimental skills of the nineteenth century chemists mentioned earlier, such as Crismer, Reichert and others, in analysis of butter and vegetable oils are admirable, involving both the partial separation of complex mixtures and the elucidation of structures. The research and the dedication of chemists in this early period in combating the major problems of food adulteration demonstrated the value of food analysis to society. Such work was an important step in the development of chemistry as a profession.

As a result of developments in food chemistry, 1888–1894, a sound scientific approach was arrived at to begin to appraise butter for adulteration. The methods adopted were robust, and their limits were recognised including the need to protect honest traders from suspicion arising from natural composition variability of milk fat. The timescale of the developments is impressive by modern standards and even today, and it is seldom possible to prove adulteration of a natural product by one method alone.

Currently, laboratory GC analysis for butyric acid is the most straightforward means of authenticating the milk fat content of butter and detecting adulteration with foreign fats and oils. One should, as our nineteenth century predecessors knew, be cautious about individual samples with low butyric acid contents. The combination of a robust well-validated method with experience of the range of butyric acid and triacylglycerol concentrations in authentic butters provides a sound basis to detect fraud and reassure consumers. If suspicions are aroused, a combination of other analytical techniques alongside in-factory inspection and investigation of the supply chain should take place.

The rapidity and low sample preparation needs of spectroscopic techniques such as near infrared, Fourier transform infrared and Raman lend themselves to in-factory quality control, carried out by non-technical personnel. The robustness and mobility of these techniques suggest that on-site and point of inspection screening should be possible. Confirmatory analysis could then be obtained by conventional chromatographic techniques. If, as the horsemeat episode suggests, we need to apply intensive scrutiny to the food supply chain searching for fraud and criminality [225], rapid screening techniques will be required.

The notion of marker compounds, eventually rejected for the detection of margarine in butter, has found favour in other areas such as white alcoholic spirits, where some vodka manufacturers add innocuous traces of sugars and other similar compounds chemically to mark their brands [226].

Finally, some further lessons that the historical review indicates for the twenty-first century include as follows:

Natural biological variations in primary products such as fats, oils, meat and fish can subvert forensic efforts to control adulteration. Nevertheless, data collection, such as the Belgian authorities undertook in their sampling campaigns of 1897 and 1898, the EU dataset for milk fat and cocoa fat [214] and the series of investigations of the nitrogen content of flesh foods [227], shows that sufficient data can be used to yield forensically meaningful outcomes. Similar research leading to publically available datasets are needed for the nuclear and mitochondrial DNA content of flesh foods, and the isotopic and fatty acid profiles of farmed and wild fish [228].

International standardisation of methods, similar to that achieved for the characterisation of the fatty acids of milk fat, is vital for the control of food authenticity [225].

The involvement of research by third-level institutions on food adulteration and the enforcement of standards were clearly evident in Belgium in the nineteenth century; such input is again needed in the face of the ever increasing sophistication of food frauds.

Appendix 1: Overview of physical and chemical methods characterising edible lipids (fats and oils) mentioned in this paper

Physical methods:

1. Determination of the melting point.

This is the temperature at which fat/butter when heated very slowly becomes completely clear and liquid.

2. Determination of the specific gravity of fat/butter at 100 °C (Method of C. König (37)).

If the butterfat is pure, its specific gravity lies between 0.865 and 0.868.

3. Determination of refractive index with a butter refractometer

This can be done by Abbé refractometer, Zeiss butyrofractometer or refractometer of E.-H., Amagat and F. Jean.

4. Study by microscope of melted fat and the insoluble residue in ether and in alkali
5. Crismer number or critical temperature of dissolution of from butter or margarine fat (turbidity method)

L. Crismer discovered a completely new method for the examination of lipids when he developed the critical temperature of dissolution method. The temperature of fat in alcohol in a tube is increased until transparency of the mixture. After this transparency, the temperature should be dropped till the first discernible turbidity. This temperature is the critical temperature of dissolution. Because the boiling point of alcohol is 78 °C, it was necessary to work with closed tubes. Crismer proved that it was also possible to work with open tubes; however, in that case it was necessary to use nearly absolute alcohol [47, 50]. The values of the critical temperature of dissolution were for butter between 54 and 55 °C and for margarine between 74 and 78 °C.

Chemical methods:

1. The saponification or Köttstorfer number.

Saponification is the process of breaking down a neutral fat into glycerol and fatty acids (as soap) by treatment of the fat with alkali. The saponification or Köttstorfer number is the number of milligrams of KOH required to saponify 1 g of fat under specified conditions. This number is a measure to the average molecular weight of all fatty acids present. The smaller the number, the larger the fatty acid chain length [33].

2. The Hehner number

The Hehner number is the amount of insoluble fatty acids which is obtained from 100 g of fat after saponification and treatment with an acid. For pure butter, the Hehner number varies between 85.4 and 91.3 %, and the average may be taken as 87.5 % [27].

3. The Reichert–Meissl number

The Reichert–Meissl number is the volume of 0.1 N alkali, expressed in ml, which is required to neutralise the volatile and water-soluble fatty acids from 5 g of saponified fat. The Reichert–Meissl number for pure butter is high, because the volatile fats represent the acids soluble in water: butyric and caproic acid, and may vary between 19 and 34, but only in rare instances does fall below 24 [30, 31].

3. The Polenske number

The Polenske number is the volume of 0.1 N alkali in ml, which is required to neutralise the water-insoluble fatty volatile acids distilled from 5 g of saponified fat. The Polenske number in the case of pure butter is low, because the volatile acids insoluble in water include caprylic acid, capric acid, lauric acid, myristic acid, palmitic acid, stearic acid and oleic acid. For pure butter, the Polenske number varies between 1.5 and 3.5 [67].

Hence, the UK standard method referred to as the Reichert, Polenske and Kirschner, RPK, approach depends on the following definitions.

If M is the number of millilitres of 0.1 M aqueous alkali required to neutralise the fatty acids released on saponification and distillation from 5 g fat under the precise conditions specified in the method:

- The Reichert value, R, is M for the water-soluble volatile fatty acids,
- The Polenske value, P, is M for the water-insoluble volatile fatty acids, and
- The Kirschner value, K, is M for water-soluble volatile fatty acids that form water-soluble silver salts.

There are macro (5 g fat) and semi-micro (1 g fat) versions of the RPK procedure, the latter with a scaled down apparatus specified and 0.02 M NaOH or 0.01 M BaOH as titrant [191].

As with all biological products, there is a natural (including species) variation in R, P and K values which are also influenced by season, feeding and the nutritional and health status of the producing animals. Genuine milk fat seldom yields an R of less than 24, known relationships exist between R, P and K and their relative values can give clues as to the nature of any non-milk fat present. Equations have been established from which the percentage of milk fat in a fat sample can be estimated. If the RPK values are in the correct ratio, it is not safe to assume adulteration if they are somewhat low; positive proof of an adulterant should be sought [191].

4. Iodine Number

The iodine number is a measure of degree of unsaturation, which is the number of carbon–carbon double bonds in relation to the amount of fat or oil. The iodine number is defined as the grams of iodine absorbed per 100-g sample [25].

Appendix 2

a. *Royal Decree of October 31st 1900 (63) giving details of the Law of May 4th 1900, article 10 (62)*:

Article 3: Abnormal butter: Prohibition to sell

When in the sample at least two values of the following values are present, the butter should be declared abnormal.

- A refractive index (Abbe-Zeiss) 40 °C, higher than 1.4565;
- A critical temperature of dissolution in alcohol (99.1 g/L) higher than 59 °C;
- A specific gravity at 100 °C lower then 0.864;
- A value of volatile fatty acids (Meissl) lower than 25;
- A content of non-volatile fatty acids (Hehner) higher than 89.5 %;
- A value of saponification (Köttstorfer) lower than 221.

b. *Royal Decree of October 20th 1903 (76) giving details of the Law of August 12th 1903, article 11, section C (75):*

Article 4: Shall be considered as abnormal butter, if deviating from two of the values of the majority of pure butters, from the following values:

- A refractive index (Abbe-Zeiss) 40 °C, higher than 1.4565;
- A critical temperature of dissolution in alcohol (99.1 g/L) higher than 59 °C;
- A *specific gravity* at 100 °C lower than 0.864;

- A value of volatile fatty acids (Meissl) lower than 25;
- A content of non-volatile fatty acids (Hehner) higher than 9.5 %;
- A value of saponification (Köttstorfer) lower than 221.

c. *Law of November 21st 1904 (78)*

Changing article 11, section C of the Royal Decree of October 20th 1903 (76):

Article 2: changing article 4 of the Royal Decree of October 20th 1903 (76): Shall be considered as abnormal butter, when the values which are different for pure butter, an index of fatty volatile and soluble acids (Reichert–Meissl) lower than 28 and one of the other values:

- A refractive index (Abbe-Zeiss) 40 °C, higher than 1.4552;
- A critical temperature of dissolution in alcohol (99.1 g/L) higher than 57 °C;
- A specific gravity at 100 °C lower than 0.865;
- A content of non-volatile fatty acids (Hehner) higher than 88.5 %;
- A value of saponification (Köttstorfer) lower than 222.

These abnormal butters are declared illegal to sell.

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