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Counterfeiting of milk and dairy products, and analytical methods suitable for the detection of counterfeiting

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1. SUMMARY

In the first part of this review article, the authors write about food counterfeiting in general, and then seek answers to the following questions: Are foods counterfeited today? What does food counterfeiting means and how to combat it? What official measures should be taken when food counterfeiting is detected? What sanctions can be imposed in case of food counterfeiting? In the context of counterfeiting milk and dairy products, the authors of the article report on the counterfeiting of milk from various animal breeds, such as buffalo, goat's and sheep's milk, as well as breast milk using cow's milk, and also on mixing soy milk to cow's milk. They also describe the detection of whey and buttermilk in milk, the determination of whey protein in dairy products, the analysis of milk produced from milk powder and other possibilities of milk and dairy product counterfeiting. Finally, they report on the detection of other fats in milk, butter and ghee (a traditional Indian butter formula made from buffalo milk – the Editor), the watering of milk, the determination of the extent of heat treatment of milk and dairy products, and the detection of the amount of spoiled milk unfit for consumption. The manuscript also describes the principles of analytical methods suitable for the detection of counterfeiting.

2. Introduction

Food counterfeiting is as old as human food production itself. The primary purpose of counterfeiting is to obtain illicit (illegal) profit. The earliest written records of food counterfeiting remain from ancient times, when Hammurabi's laws already prohibited the sale of low quality or overly expensive beers. Anyone who violated the king's laws could face severe punishment, as food counterfeiting could cost him his life **[1]**. We have written records of wine being counterfeited, primarily watered in the Roman Empire, which was also severely punished **[2]**. Unfortunately, food counterfeiting is still present in a number of countries today, and so the authorities taking action against it have developed procedures that may be suitable for the detection of counterfeit foods and provide information on the very fact of counterfeiting **[3]**.

The appearance of counterfeiting of milk is characteristic of newer times, because the watering of milk is simple to carry out: water is cheap and easily accessible. Before the 1800s in England, the counterfeiting of milk with well water was an almost common practice, and it only became less frequent when, at the end of the century, methods were developed to detect milk counterfeiting **[4, 5]**. Milk counterfeiting still does go on, in some countries and regions it is a daily practice to mask watering by adding salt, and in some cases cooking oil and detergents are added to increase the fat content of the milk **[3]**.

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Very expensive cheeses made from milk are also counterfeited in significant quantities. The first counterfeiting, memorable in in the history of food production, took place in the United States in the 1870s, when it was found out that high quality Wisconsin cheeses were counterfeited with cheap fats, such as lard, to increase their weight. After the fact of counterfeiting had come to light, the export of these cheeses fell, they lost their reputation, and it took decades for them to recover **[3]**. Counterfeiting has not ceased to this day; there are many examples of imitating expensive cheeses, but in many cases their quality is not even close to that of the high quality cheeses, which have been matured for several years and are sought after by consumers.

3. Are foods counterfeited today?

The answer is obviously yes: there are regularly news in the media about food counterfeiting, it is enough to think of the recent scandals when honey was counterfeited with high fructose corn starch hydrolyzate until the experts were able to develop the appropriate method to detect the above-mentioned foreign substance in honey **[6]**. There were two counterfeiting scandals in relation to wines recently. In Austria, four wineries have tried to produce more full-bodied wines using antifreeze containing ethylene glycol, which causes severe poisoning in the human body. As a result of this case, Austrian wines disappeared from the shelves of European supermarkets for a long time **[7]**.

Unfortunately, a few years after the Austrian case, a memorable wine counterfeiting scandal took place in Hungary as well, when a winemaker tried to "improve" Eger bull's blood with glycerol. Although glycerol, which is also found originally in wine, is not toxic to the human body, but a glycerol content above the permissible tolerance limit is considered to be counterfeiting **[8]**. Certain beverages can be easily counterfeited by the addition of food concentrates, especially sugar solutions, diluted with the proper amount of water. For example, the ratio of glucose to sucrose to fructose in orange juice is 1:2:1, so this food is counterfeited with invert sugar extracted from sugar beet, because its ratio of sugars is the same as that of orange juice. In the case of these foods, to maintain the correct acid-sugar ratio, besides sugar, various organic acids are also added **[9]**.

In parallel with the appearance of counterfeiting, professionals are constantly working on the development of methods to detect the fact of counterfeiting, in order to reduce it. For example, the invert sugar used for counterfeiting with sugar solution also contains trisaccharides, which may be a marker for counterfeiting. The malic acid used to adjust the acid ratio of orange juice is available in the DL version because of its industrial manufacture, while orange juice only contains the natural L version. In synthetic products, the D:L ratio is 1:1, so if this type of artificial „apple juice" is added to the orange juice, then the presence of D-malic acid indicates counterfeiting. The D stereoisomer of malic acid is now readily detectable in foods using either enzymatic methods or high performance liquid chromatography, for example **[10]**.

Of course, many other similar counterfeiting methods could be described, but this is not possible due to the limited scope of our review. The examples listed above suggest that almost any food can be counterfeited, and there may be serious cases where the substances used for counterfeiting are extremely harmful to the human body, and may even be fatal. One such example was the counterfeiting of Hungarian ground paprika with lead oxide in order to make the color of the product more desirable **[11, 12]**. In China, infant formulas and dog foods were counterfeited with melamine to adjust the apparent crude protein content of the formulas and dog foods, measured on the basis of their nitrogen content, to the required value. Consuming the products counterfeited with melamine has resulted in the deaths of many infants and animals **[13]**.

According to our own records, the following cases of food counterfeiting have been discovered in recent years, in Hungary as well: vegetable fat was detected in milk powder; foreign sugar was mixed with honey, sweetener with powdered sugar; date of minimum durability was falsely indicated; meat products made from poultry were falsely labeled; bakery products were prepared and mineral water was produced using unregistered methods; raw milk and smoked finished products were manufactured without authorization; foods intended for public consumption were marketed in a prohibited way following slaughter in an unauthorized facility.

The motive for counterfeiting is always financial gain. False ingredients are in many cases difficult to detect, because they are often unknown to professionals; inspectors do not necessarily suspect their presence. A good example of this is melamine, which until 2007 was not considered as a contaminant or a substance used for counterfeiting until it became detectable in dog food, and until, in 2008, it caused mass deaths when mixed into infant formulas and other dairy products. It was later discovered that melamine had been used for counterfeiting since 1979 to achieve higher apparent protein content, but this remained hidden from both consumers and researchers until 2007. Counterfeiting with melamine was not suspected by anyone, because the detection of this compound was not part of routine quality control. The food counterfeiting warning system cannot be designed to detect an unmanageable number of potential counterfeiting substances. We believe that the most effective way to combat food counterfeiting in the practice of food quality control is to continuously monitor in the laboratory the amount of components that a good quality food must contain. A well-designed analysis can detect both known and unknown counterfeiting ingredients, which is a great

advantage in an environment where we do not know what kind of dangerous counterfeit product might be encountered.

4. Counterfeiting of milk and dairy products and their detection

To detect counterfeiting, various analytical methods, most often large instrument techniques are used by professionals **[3]**. Since the description of the diverse practices of counterfeiting would require the review of scientific literature that would fill several libraries even in the case of the most important staple foods, only the sophisticated methods of counterfeiting milk and dairy products are presented in this paper, and the often seemingly complex analytical steps by which counterfeiting can be detected **[14, 15, 16]**.

High quality milk and dairy products are free of impurities, unpleasant odor and taste and pathogenic microorganisms; its somatic cell count and total microbial count must not exceed the permitted values; it must not contain foreign water and any foreign substance, it has a good smell and a taste characteristic of milk. Its composition complies with the Hungarian Food Codex regulation for milk and dairy products and, in the case of products not produced according to the food codex, with the specification stated on the product data sheet. Its antibiotics and other toxic contaminant content must not exceed the values laid down in European Union laws currently in force **[17, 18, 19]**.

In the case of raw milk intended for marketing, the addition of any other ingredient (mainly water) to milk or the removal of any ingredient (mainly fat) is considered counterfeiting **[20]**. Counterfeiters most often add water or skimmed milk to milk or extract a significant portion of its original fat content **[3, 21, 22]**. This type of counterfeiting can be detected by measurement of density, checking the freezing point or by determining the fat content. Milk contaminated, infected with dirty water, cleaning agents, plant residues, animal hair, dust, other contaminants, animal urine or faeces can be tested organoleptically and distinguished from high quality milk **[17]**. However, to bring to light counterfeiting that cannot be detected by organoleptic methods, chemical analytical test methods are required. The authorities supervising the marketing of milk in several countries have introduced a scoring system that penalizes the presence of factors that impair the quality of milk and, consequently, provides a lower income for farmers who produce inadequate milk. Particular attention is paid to the contamination of milk with antibiotics, radioactive substances, chlorinated hydrocarbons and heavy metals **[17, 20, 23]**.

4.1. Milk from different animal breeds and their counterfeiting

The mixing of cow's milk and buffalo milk, or cow's milk, goat's milk and sheep's milk is a common practice worldwide. In the food law system of the European Union this is not allowed, the mixing of different types of milk is considered counterfeiting. Cow's milk is most often counterfeited with goat's milk, but it also happens often that otherwise high quality goat's milk is counterfeited with water or cow's milk **[24, 25]**. When goat's milk is counterfeited with cow's milk, its nutritional value remains unchanged, and if the amount of cow's milk added does not exceed 15%, its detection in goat's milk is difficult. The mixing of milks causes organoleptic defects during the production of cheeses, because different types of milk give cheese different flavor and taste, moreover, milk from a foreign species can trigger an allergic reaction in the body of the consumer **[26]**.

4.1.1. Analysis of the composition of mixed milk samples from different animal breeds

Several methods have been developed by researchers to detect counterfeiting by mixing different types of milk **[27]**. Lees et al., Aranda et al. **[28]**, Bitri et al. **[29]** and Castro et al. **[30]** reported immunoassay methods. Cartoni et al. **[31]**, Kaminarides and Koukiassa **[32]**, Lee et al. **[33]** and Műller et al. **[34]** used gel electrophoresis. Špoljarić et al. **[35]** and Mayer et al. **[36]** separated the casein fractions by isoelectric focusing, and milk samples from the different animals were identified based on this. Milk samples were analyzed on the basis of their long-chain fatty acid content by Gutiérrez et al. **[37]**, by gas chromatography by Cartoni et al. **[38]** and by examining their casein monopeptide content using high performance liquid chromatography by Ferreira and Oliveira **[39]**, and their composition was determined on these basis.

Different types of milk can also be identified by differences in their chemical composition and UV spectra **[17]**, but the different fatty acid composition of cow's milk and goat's milk and the difference in their indexes calculated from the fatty acids can be helpful in the detection of counterfeiting **[38]**.

Short-chain fatty acids and the index values calculated from their concentrations are particularly suitable for the detection of dairy product counterfeiting, and gas chromatography analysis have also show that since cheeses made from goat's milk and sheep's milk have different short-chain fatty acid patterns that those made from cow's milk, the different cheeses can be easily distinguished from each other using these index values **[37, 38]**. The average ratio of lauric aid to capric acid in cheese made from cow's milk is 1:1.16, in goat cheese it is 1:0,46, while in sheep cheese it is 1:0.58. This ratio can provide information on the amount of cow's milk in goat or sheep cheese. The mixing of cow's milk to goat's milk can also be detected on the basis of the β-carotene content, since this compound is not found in goat's milk **[40]**; the addition of 20% goat's milk to cow's milk can already be detected on the basis of the UV spectrum.

Based on the significantly higher riboflavin content and xanthine oxidase activity of cow's milk, enzymatic methods have also been developed for the detection of cow's milk added to sheep's milk, and thanks to these 2% cow's milk in sheep's milk can already be detected. The deficiency of the method is that heat treatment destroys enzyme activity, and so it cannot be applied in the case of heat treated milk.

The mineral content of sheep's, goat's and cow's milk is relatively constant, but the ratio of the different elements in various milks may be very different. The amount of minerals is also influenced by the technology: there are marked differences between cheeses made from different milks **[41]**. For example, the ratio of calcium to magnesium is 23.3 in cow's milk, but only 17.2 in sheep's milk. Based on this, dairy products made from the two types of milk can be easily distinguished. For example, professionals found differences in the K/Mg, Na/Ca, Cu/Zn and Cu/Na ratios of cow's, goat's and sheep's milk, and multivariate analysis of trace elements (Cr, Mn, Fe, Ni, Cu, Zn, Mo, Cd and Pb) could be used to distinguish between the milks of the different species **[41]**.

Researchers were able to distinguish cheeses made from the milk of different species by electrophoresis, for example, on the basis of the different mobility of various casein fractions (primarily κ-casein), but whey protein fractions were also found to be useful in this case **[42]**. Because the mobility of the α-casein and β-lactoglobulin fractions of cow's milk is significantly higher than that of goat's milk fractions, they are also suitable for the detection of counterfeiting **[28]**. Based on the αs_1-casein fraction of cow's milk, the addition of 5-10% of cow's milk to goat's milk is detectable, and the same can be said about the β-lactoglobulin fraction **[31]**.

In the case of cheeses, the α-casein fraction is significantly more sensitive than the β-lactoglobulin fraction. The reason for this is that, on the one hand, its concentration is low because it is removed from the dairy product during cheese making, and on the other hand it tends to precipitate, which also reduces its amount. Analyses related to α-casein are based on the assumption that its concentration in cow's milk s relatively constant, although some studies have shown that individual differences that also influence coagulation may be significant, making it difficult to detect less than 5% cow's milk in goat cheese **[32, 36]**.

Isoelectric focusing following the urea extraction of cheeses allows for a much more accurate determination of the amount of goat's milk in goat and sheep cheese on the basis of the para-κ-casein content, compared to the method based on α-casein. With the help of this method, using densitometric evaluation, 1-2% of cow's milk can be detected in sheep's milk and sheep cheese **[36]**.

High performance liquid chromatography (HPLC) is also suitable for the detection and quantification of a minimum of 2% of cow's milk added to goat's or sheep's milk. A minimum of 2.5% cow's milk can be detected in sheep's or goat's milk by immunodiffusion methods and immunoelectrophoresis **[43]**. The above methods are also suitable for determining the amount of cheese coming from cow's milk, provided that the amount of cow's milk added is at least 10%. For the detection of cow's milk in sheep's and goat's milk, radial immunodiffusion has previously been used by experts, but this technique has not gained much ground in practice. Rocket immunoelectrophoresis is also effective in detecting cow's milk in the milk of the other two species (when mixing 1-5% cow's milk to goat's milk), because there is no cross-reaction between the antibody and goat's milk. The method is applicable to both heat treated, homogenized and raw milk **[44]**.

Experts have also used the ELISA method with adequate efficiency to detect cow's milk in sheep's milk and sheep cheese, although a weaker immune response was obtained for gently and ultra-high temperature pasteurized milk, as well as sterilized milk, due to probable precipitation **[42, 43]**.

When comparing the methods it can be stated that electrophoresis, especially polyacrylamide gel electrophoresis (PAGE), gives more accurate and reliable results than immunoelectrophoresis or radial immunodiffusion. Using electrophoresis, the addition of 5% of goat's milk to sheep's milk can be detected with greater certainty **[45, 46]**.

4.2. Counterfeiting of buffalo milk with cow's milk

During the production of mozzarella (a typical Italian cheese), water buffalo milk is often counterfeited with cow's milk because of its low price. Electrophoresis can be used successfully to detect cow's milk added to buffalo milk, based on the electrophoretic mobility of the caseins. α- and β-caseins are the most suitable for the purpose, since their mobility differs most from each other. Of the casein fractions, the best results for both polyacrylamide and agarose gel electrophoresis were obtained in the case of αs_1-casein. The corresponding pairs of casein fractions can be found in cow's milk and buffalo milk; these can be separated by isoelectric focusing (IEF) **[47]**.

To distinguish between the two milks, the use of proteolytic enzymes was attempted by the experts, followed by the separation of the fractions. The difference in the electrophoretic mobility of the fractions obtained can also be utilized well in the detection of cow's milk in buffalo milk. Attempts were made to analyze the γ_2 and γ_3 casein fractions after plasmin administration using PAGE and IEF. This method has been shown to be suitable for the detection and quantification of one type of milk in another at a level of 1%, using the casein fractions already mentioned **[48, 49]**.

Researchers have also tried to use electrical conductivity to detect counterfeiting, based on the principle that the electrical conductivity of buffalo milk increases proportionally with the amount of cow's milk added **[50]**. The determination of the fatty acid composition of the fat may also be a suitable method; in this case, the palmitic and oleic acid content of the milk fat of buffalo milk in the liquid phase increases significantly as a result of the addition of cow's milk. These two fatty acids are extremely sensitive to mixing with cow's milk, and can be used to detect with high certainty the addition of 5% of cow's milk to buffalo milk. Since the fatty acid composition is also affected by the season, the region and the animal feed, it is advisable to carry out the comparison of the fat composition of the two species in different environments, and to establish a system of estimation of the proportion of cow's milk that takes into account local characteristics **[51]**.

Methods have been developed based on the antibody produced by the casein micelles of buffalo milk in rabbit, as well as on carotene content; the latter uses the fact that the carotene content of buffalo milk is significantly lower than that of cow's milk. Compared to cow's milk, buffalo milk contains more lactenin and less agglutinin, which may also be the basis of distinction **[52, 53]**.

The milk of different animal species can also be distinguished on the basis of various volatile components. For example, in cow's, goat's and sheep's milk, dimethyl sulfone accounts for 25% of all volatile components, while this proportion is only 4% in buffalo milk, which can also be used to distinguish between the two milks of different origin. 3-Methylbutanal is only found in buffalo milk, phenylacetaldehyde and benzaldehyde in high concentrations in goat's milk, while 2-methylketones and 1-octene-3-ol in high concentrations in buffalo milk. Phenylethanol cannot be detected in sheep's milk and goat's milk at all, but it is present in buffalo milk at a concentration one hundred times higher than that in cow's milk. The methods described above can all serve as the basis for potential analytical methods **[54, 55]**.

4.3. Counterfeiting of breast milk with other milks

In the flocculation test routinely used in milk analysis, casein type proteins are precipitated at 37 °C and whey proteins at 60 °C using a calcium acetate solution of suitable concentration, but it does not react with human milk or colostrum. If flakes precipitate from breast milk, it also contains cow's milk **[58]**. Cow's milk added to breast milk can also be detected by a saturated copper sulfate solution containing 4% cadmium sulfate, in the reaction of which a precipitate is formed if the breast milk contains cow's milk. The watering of breast milk becomes detectable by the increase in freezing point, but this method is for information purposes only, as the freezing point may vary from person to person, and even from time to time in the case of the same person **[57]**.

Cow's milk added to breast milk is relatively easy to detect based on differences in the properties of the protein fractions of breast milk and cow's milk. Since β-lactoglobulin is not present in breast milk, its presence is a clear indicator of counterfeiting. The α-lactalbumin in the whey protein fraction and the κ-casein in the casein fraction were also found to be suitable for proving counterfeiting. With the help of these protein fractions, the addition of 1% of cow's milk to breast milk can already be detected. The method used is PAGE and IEF **[59]**.

The free amino acid and taurine content of breast milk is significantly higher than that of cow's milk. While the taurine content of breast milk is 33.5 µmol/100 ml on average, that of cow's milk is only 1.9 µmol/100 ml, while the concentration of glutamic acid is 262.7 µmol/100 ml and 28.8 µmol/100 ml, respectively. Based on these averages, cow's milk in breast milk can be detected, as it reduces the amounts of both taurine and free glutamic acid significantly. Taurine and free glutamic acid contents can be determined by ion exchange column chromatography with post-column derivatization with ninhydrin or by high performance liquid chromatography and pre-column derivatization **[60]**.

4.4. Soy milk in cow's milk

Today, soy milk and soy protein receive a lot of attention both from an economic and nutrition point of view. This is particularly true for developing countries, where there is a shortage of high quality protein of animal origin, and soy protein can be used to replace, substitute or supplement it. In addition, soy milk and dairy-like substances made from it are ideal nutrients for vegetarians and people with milk protein allergy. However, it is not easy to find analytical methods that can detect soy protein added to milk, because the addition of 10-20% of soy milk to cow's milk does not change the organoleptic properties of either yogurt or cheese. The addition of 20% of soy milk does not change the curdling time, but in the case of larger quantities, longer curdling times are expected **[61, 62]**.

The similarities in their structure pose a particular difficulty for analysts when it comes to detecting soy proteins in a dairy product. Several methods have been developed for this purpose: sodium dodecyl sulfate polyacrylamide gel electrophoresis (SDS-PAGE), various serological methods and peptide analysis. These types of analysis are based on the differences in the protein content of soy milk and cow's milk **[62]**. PAGE, with the application of a pH 8.6 TRIS (tris(hydroxymethyl)aminomethane) buffer, can be used to separate six fractions of cow's milk and nine fractions of soy milk. The electrophoretic mobility of the soybean globulin fractions is greater than that of the corresponding milk protein, κ-casein, but less than that of γ-casein. This method makes it possible to detect the addition of 2% of soy milk

to cow's milk. In addition to the above-mentioned methods, PAGE, SDS-PAGE and HPLC are also useful for the detection of soy proteins. With these methods, the addition of 5% of soy milk to cow's milk can be detected with high certainty and its quantity can be determined **[63]**.

When evaluating the peaks obtained in the HPLC analysis with the help if a calibration curve, it can be stated that more than 1% of soy milk in cow's milk can be detected with high confidence. The disadvantages of these methods are that they are expensive, required skilled personnel and expensive instruments, whereas ELISA methods are significantly cheaper and can also detect soy milk in excess of 1%. In addition to soy milk, coconut milk added to cow's milk can also be detected by the above methods **[64]**.

4.5. Detection of whey and buttermilk in milk

Due to the increased production and consumption of cheese today, the amount of whey remaining in the factories has also increased, the use and disposal of which often presents difficulties. Whey powder made from whey is significantly cheaper than skimmed milk powder, but its use is limited, due to its high lactose content **[38]**. According to manufacturing specifications, skimmed milk powder may only be made from skimmed milk **[105]**, it cannot contain rennet, and cannot contain dry matter from whey or buttermilk. In many parts of the world, buttermilk remaining after the production of butter from sweet cream is added as a powder to skimmed milk powder; various methods have been developed by the experts to detect this. Counterfeiting can be traced on the basis of the amount of the whey protein fraction and lactic acid, which is positive if it exceeds 150 mg/100g, and on the basis of the ash content, which is positive if it exceeds 8%.

Methods that can be used to detect buttermilk powder include electron microscopy, because the particles have different surfaces if the powder is made from skimmed milk or buttermilk, and the acid precipitation test, during which casein micelles, whey proteins and the membranes of the large amounts of fat globules in buttermilk behave differently.

The counterfeiting of pasteurized milk is also a major problem in many countries. Since the price of whey is low and its organoleptic properties are not significantly different from those of milk, it is possible to to obtain illegal economic benefits by counterfeiting milk with whey **[65]**. However, the amount of whey in milk can be determined by the casein/whey protein ratio. Casein in the milk sample can be determined by precipitation at pH 4.6. Whey protein remains in the phase above the precipitate. The casein and phosphorus contents of milk are closely related, since only casein can bind phosphates by an ester linkage. One can thus draw conclusions regarding the casein content from the phosphorus content, and from the casein content regarding the counterfeiting of milk with whey **[65]**.

4.5.1. Whey protein in dairy products

It is important to know how much milk dry matter the various dairy products contain, and how much of this is whole milk powder. Frozen dairy products should contain at least 10% fat and 20% dry matter. In addition, when investigating counterfeiting, the ratio of whey protein and casein in the sample should be determined. For this purpose, for example, to determine the protein content of ice creams, the so-called dye-binding methods are suitable, but they give slightly different results than the conventional Kjeldahl method. Casein is difficult to separate from whey protein, because they precipitate together after various heat treatment processes, and so they are practically inseparable **[39]**.

In order to determine these two proteins, either the complex they form must be disrupted, or other solutions, such as estimation based on phosphorus content, must be employed. Since phosphorus is only bound to casein, the amount of casein can be estimated on the basis of the phosphorus/nitrogen ratio, even in complex matrices such as ice cream **[66]**. Radial immunodiffusion can also be used to estimate the amounts of casein and whey protein. The determination of the casein content based on the phosphorus content can be applied to both sodium caseinate and processed dairy products **[67]**. The addition of whey powder, buttermilk powder or caseinate to skimmed milk can be detected with the help of the cysteine-cystine (S-S) complex or sialic acid. Cysteine and cystine contents can be measured by the modified ninhydrin reaction or by ion exchange column chromatography. The amount of SH groups in normal skimmed milk powder is 86.4 µg/g protein on average, showing a linear increase on the addition of whey or whey protein. The addition of 10% of whey protein to skimmed milk powder significantly increases the concentration of SH groups, so the amount of added whey or whey protein can be easily determined by this method. If the cysteine to cystine ratio in the sample is greater than 3 and the sialic acid content exceeds 3%, the addition of whey protein can be considered as proven. HPLC and gel electrophoresis can also be used, but these techniques are much more expensive **[39]**.

The amount of added whey protein can also be determined on the basis of the amino acid composition, if it is equal to or greater than 10%. This method is unaffected by whether the whey protein is denatured or not, or whether there has been heat treatment or not. Researchers have also attempted the determination of glycomacropeptides by liquid chromatography or spectrophotometry, but a number of erroneous results have been obtained due to bacterial contamination. However, favorable results have been achieved by de-

tecting the addition of whey powder produced using rennin to sweet buttermilk powder. Test results were most reliable when analyzing whey powders **[68]**.

Comparing the analytical methods listed above, it can be concluded that the HPLC method outperforms all others in terms of both reliability and sensitivity, and can be used to detect the addition of even 0.5% of sweet whey powder to the given dairy product on the basis of the analysis of the protein fractions **[69]**.

The sweet whey powder produced during cheese making contains much more of the water-soluble molecules also found in milk, therefore it has higher lactose, sodium, potassium and chloride contents. As a result, milk made from milk powder will have a significantly lower freezing point when whey powder is added. Using regression equations, the amount of added whey powder can be calculated from the decrease in the freezing point.

There are other known methods for determining the amount of added whey powder, but these require complicated sample preparation procedures and therefore are not widespread in practice. Fourier-tansform infrared spectroscopy can be suitable to discriminate between proteins **[70]**.

4.5.2. (Reconstituted) milk made from milk powder

During the production of milk powder some of the proteins are denatured, which can be used to detect reconstituted milk. No differences could be detected between normal and reconstituted milk either by the dye-binding method or gel electrophoresis. However, based on the ratio of β-casein to α-lactalbumin, the addition of 25% of reconstituted milk to normal milk can be detected **[71]**.

It has been determined by experts using electron microscopy that reconstituted milk contains aggregates with diameters greater than 500 nm, which are not present in normal milk **[72]**. Doerr et al. also experimented with the addition of resazurin, which gives different colors in the case of the two milks due to the total reducing capacity of milk. It is assumed that even if the density and freezing point values are within the expected range, the nitrate content of reconstituted milk will be higher than that of the extremely low nitrate content of normal milk because of the nitrate content of the dilution water used. If the nitrate content is on average 1 mg/kg higher than that of milk in general, then the milk is likely to contain reconstituted milk. During this determination, nitrate is converted into nitrite, which can be measured with adequate precision using a chemiluminescent method **[73]**.

4.5.3. Other possibilities of milk and dairy product counterfeiting

If the manganese content of milk is high, counterfeiting with calf feed can be suspected, as its manganese content can be as high as 10-15 mg/kg, while that of milk is only 0.021 mg/kg on average **[74]**. Supplementation of pure milk with milk containing vegetable proteins can be detected by measuring the nitrogen content of the whey protein after the precipitation of the casein present.

The addition of raw milk to pasteurized milk can be detected by measuring the activity of the phosphatase enzyme **[75]**, while the authenticity of mozzarella cheese can be verified by scanning electron microscopy or scanning calorimetry, because counterfeit products contain fat globules that are not present in the authentic cheese **[76]**.

Glucose, cane sugar, urea or ammonium sulfate are added to milk to hide the fact that it is diluted with water. These substances can also prevent the increase in freezing point, so sophisticated analytical methods are needed to detect counterfeiting. Because of the milk sugar originally present in the milk, sugar added to milk can only be detected using several separation technique, in particular by liquid chromatographic separation; in this case, not the total amount of sugars, but individual sugars are determined by the researchers. Sugars are hydrolyzed by invertase enzyme, and the glucose and fructose produced can be determined by a rapid method using the glucose oxidase peroxidase test **[61]**.

Addition of table salt to milk up to 0.4% does not cause a change in taste, but at the same time, 13% of water can be added to the milk without significantly changing its freezing point. To reduce acidity and increase shelf-life, counterfeiters sometimes add ammonia solution, or sometimes sodium bicarbonate or antibiotics. The addition of 0.3% sodium bicarbonate to milk allows it to be diluted with 10% of water without significant changes in the measurable parameters **[77]**.

4.5.4. Other fats in milk, butter and ghee (a traditional Indian butter formula)

Since milk fat is one of the most expensive fats, counterfeiting it with other cheap fats occurs almost everywhere in the world. Most often vegetable oils, particularly linseed oil and beef tallow are used. In many countries, various methods have been developed by the experts to detect butter counterfeiting. Most of the methods are based on the determination of the structure of triglycerides, on fatty acid composition analysis, on the measurement of unsaponifiable lipids (sterols, sterol esters, tocopherols, carbonyl compounds) and the analysis of various physical properties **[78]**.

The most promising method for the detection of counterfeiting is based on the analysis of triglycerides; during this, with the help of triglycerides with different carbon atom numbers, milk fat can be easily separated from other fats and the addition of already

5-10% of foreign fat can be detected with great certainty. Researchers have developed different formulas to be able to discover not only the fact of counterfeiting, but also the type of fat used to counterfeit the given milk fat. These methods are based on the fact that only milk fat contains butyric acid, caprioc acid, caprylic acid and capric acid, and therefore it has a much higher concentration of lower carbon triglycerides than other fats **[79]**.

However, the results obtained should be treated with caution, as not only the fatty acid composition but also the triglyceride composition may vary with the season, the region and the lactation state. For example, winter milk contains more short and medium chain triglycerides than summer milk. Ultraviolet light absorption was not successful in the detection of vegetable oils in milk fat, but the measurement of the concentration of butyric acid was successful, as was the gas chromatographic separation on a capillary column, whereby not only fatty acids but also stereoisomers (cis, trans, cis-trans, cis-cis, trans-trans, etc.) could be determined. Infrared spectroscopy was also used effectively to identify the latter **[80]**.

Parodi and Dunstan were able to detect 0-30% of cotton seed oil added to butter by the infrared spectroscopy of trans-unsaturated fatty acids. Trans-unsaturated fatty acids occur naturally in milk fat, but they are not present in naturally occurring, non-hydrogenated (catalytic hydrogenation) vegetable oils, so measuring the concentration of trans-unsaturated fatty acids allows for the detection of counterfeiting of butter. The results obtained once again should be treated with caution as the amount of trans fatty acids may be affected by the trans fatty acid content of the feed and the biohydrogenation processes that take place in the rumen of the cow **[81]**. Microorganisms in the rumen are capable of saturating unsaturated fatty acids and synthesizing trans isomers from cis isomers, and even of producing conjugated double bonds from isolated double bonds, resulting in the formation of the cis9, trans11 conjugated linoleic acid, considered to be extremely useful for humans, and its other positional isomers **[82]**.

Various indices of pure, unadulterated milk fat are determined by the experts during classification with the help of fatty acids. By comparing the fatty acid composition of the counterfeit sample with that of the pure sample, counterfeiting can be confirmed and researchers can also obtain information on the substance used to counterfeit the given butter. Butyric acid, caproic acid and cholesterol were determined by Japanese researchers by gas chromatography, and the data obtained were used to conclude counterfeiting. Based on the ratio of butyric acid to caproic acid, they were able to detect counterfeiting when beef tallow or coconut fat transesterified with butyric acid was added to the butter, sitosterol content had also been used previously to detect counterfeiting **[83]**.

Although seasonal and geographical differences may be significant in terms of the composition of milk fat, they become almost negligible when the fatty acid compositions and cholesterol contents of butter and other fats and oils, used for counterfeiting, are compared **[78]**. Particularly useful in detecting the counterfeiting of butter is the determination of the lauric acid-capric acid, myristic acid-caprioc acid and myristic acid-lauric acid ratios. The following oils and fats are regularly used to counterfeit butter:

Vegetable fats. The fatty acid composition, monoglyceride and triglyceride content of of milk fat is so different from those of other fats that counterfeiting by not only vegetable fats but animal fats can be detected easily by measuring these components **[84]**. In view of the differences between the varieties, climatic conditions and geographical locations, vegetable fats can be detected in milk fat with high certainty based on the lauric acid-capric acid ratio. 10% of coconut fat, palm or rapeseed oil, as well as 5% of soy oil added to milk fat can be easily detected based on long and medium chain triglycerides and sterols **[78]**.

Partially hydrogenated vegetable fats can be detected in cheese by gas chromatographic separation, based on the fatty acid composition. Of the fatty acid indices, the ratio of butyric acid to oleic acid is the most sensitive to counterfeiting, because vegetable oils contain a lot of oleic acid, but practically no butyric acid. This method cannot be used in the case of coconut fat, which contains relatively little oleic acid **[85]**.

Ghee butter is counterfeited with vegetable fat made from the fruit of the Phulwara tree grown in India, because its color and texture are very similar to those of ghee, but its price is significantly lower. Its quantity is determined by the TLC analysis of the triglycerides. Since this is a vegetable fat, counterfeiting can also be detected by measuring the cholesterol content. Cholesterol or phytosterol measurements can be used to detect any kind of vegetable fat, since the vast majority (more than 99%) of the sterol content of butter is cholesterol; no other type of sterol compound is present in it in detectable amounts. Cotton seed oil contains mainly β-sitosterol **[83]**, but also some γ-sitosterol and stigmasterol, so counterfeiting with vegetable oil is clearly indicated by a decrease in the concentration of cholesterol in the counterfeit food, and also an increase in the concentration of plant sterols **[86]**. The method is unaffected by the refining, deodorizing or steaming of fats, but animal fats with similar cholesterol content cannot be detected in milk fat by this method. More than 2% of maize or rice oil, more than 5% of cocoa butter, rapeseed, sesame, soybean, linseed or hazelnut oil, more than 20% of coconut fat or palm oil and more than 35% of palm kernel oil can be easily detected in butter using the above method. Garcia et al. **[80]** used the MALDI-QTOF MS technique with sufficient efficiency to detect the counterfeiting of milk powder with vegetable oils and fats.

The detection of counterfeiting can also be based on the fact that the proportion of total hydrocarbons and sterols in the unsaponifiable fraction is completely different in bacon, margarine and ghee. Bacon and margarine contain 20 to 30 times as much hydrocarbon as bovine ghee, and 10 to 15 times as much as buffalo milk ghee. Based on the above, using the regression equations calculated, lard or margarine added to ghee can be detected with high confidence **[87]**.

The various vegetable oils also contain compounds that are present only in the particular type of oil and not in other ones. An example for this is the presence of sesamin and sesamol in sesame oil, the detection of which, together with a high tocopherol content, clearly indicates counterfeiting. Counterfeiting can also be detected by differential scanning calorimetry and differential thermal analysis, but these methods are not widespread in practice. Alcohol-soluble and alcohol-insoluble triglyceride contents are also suitable for distinguishing and for the detection of counterfeiting **[88]**.

Animal and marine fats. The detection of animal body fats in butter is difficult because they have many properties in common. It can even be stated that if buffaloes are feed with cotton seed cake, their milk fat will be similar to the butter counterfeited with animal fat. Since animal fats are difficult to detect in milk fat, a number of methods have been developed by the experts, and these have been applied more or less successfully **[89]**.

Researchers investigated the different solubilities of milk fat and animal fats in a 3:4 mixture of acetic acid and ethanol using the following techniques: measurement of „butyric acid number"; analysis of the critical melting temperature (ghee between 49.5-53.5 °C, tallow between 70-73 °C); measuring the fat content precipitated and not precipitated by urea; fluorescence, during which counterfeit ghee exhibited a blue fluorescence, while the original, unadulterated ghee showed a pale green fluorescence; various chromatographic techniques were used mainly to determine triglycerides or some fraction, but most often fatty acid composition, which was then used to estimate the various fats added to the butter by forming indices **[84, 89]**.

From the point of view of applicability, the most useful of these indices are the stearic acid-oleic acid ratio, the ratio of total saturated and total unsaturated fatty acids, the palmitic acid-stearic acid ratio, and the ratio of saturated and unsaturated triglycerides **[90]**. Experts have also tried enzymatic methods, namely the analysis of free fatty acids remaining after the application of the lipase enzyme,as well as the determination of 2-monoacylglycerol, which showed that short chain fatty acids in triglycerides are less resistant to lipase attack than long chain ones **[91]**. Butter and lard can be distinguished by UV spectrum analysis in the 220-420 nm range, while butter and tallow could not be distinguished **[92]**.

After separation by chromatography, fish oil and butter are easily distinguished by their different fluorescent signals. It was easy to separate and distinguish 5-20% of dolphin oil from butter by the distillation and chromatographic determination of volatile fatty acids **[93]**. Counterfeiting of butter with triacetin or hydrogenated dolphin oil cold be detected by measuring the conductivity of the volatile distillate, since the conductivity of pure butter is lower than that of the counterfeit one, due to the higher concentrations of acetic acid and isovaleric acid in dolphin oil **[94]**.

Butter is also considered to be a counterfeit product if it is made fro the milk of different animal species or if the milk fat itself is modified by some technological intervention. When utter is produced from blended milk of different ruminant species, it is almost impossible to detect, because even gas chromatographic fatty acid analysis is not sensitive enough to distinguish the counterfeit product from the real dairy product. In India, large quantities of hydrogenated vegetable oils are used, which is a major source of counterfeit ghee. Since the degree of hydrogenation can now be controlled precisely, this type of counterfeiting is difficult to detect even with sensitive gas chromatography techniques **[95]**.

4.5.5. Watering of milk and its detection

The watering of milk can be detected easily by determining the freezing point, because water increases the original freezing point of milk. 3% of water added to milk can be detected with a high degree of certainty on the basis of freezing point using a thermistor cryoscope **[96]**.

The freezing point of milk can be ascertained to the next one thousandth of degree using a Beckmann cryoscope. The freezing point of milk varies between -0.53 and -0.56 °C. If the milk tested has a freezing point greater than -0.53 °C, it is counterfeited with water. If the freezing point of milk increases from -0.53 °C to -0.27 °C, the extent of dilution is estimated to be between 2 and 50%, so this method not only detects the fact of counterfeiting, but also also provides data on the amount of water added **[97]**.

The osmotic pressure of milk is mainly due to lactose (4.6-4.9% in cow's milk), secondly to the sodium and potassium ions, followed by the rest of the minerals, because the effect of the other components on the osmotic pressure is negligible. The hydrolysis of lactose to glucose and galactose significantly reduces the freezing point (by -0.274 °C) and increases the osmotic pressure. Therefore, when lactose is hydrolyzed, counterfeiting of milk with a moderate amount of water cannot be detected, because the freezing point does not change **[98]**.

Surface tension and viscosity measurements, absorbance measurement, at a wavelength of 280 nm, of the filtrate remaining after trypsin digestion and

trichloroacetic acid precipitation, and the analysis of nitrate ions, which is a clear sign of dilution, were also used to detect the water content of milk. Refractometric analysis of the filtrate remaining after ultracentrifugation can be used to detect the watering of breast milk. Thermistor cryoscope and vapor pressure thermometer were also used to detect the watering of milk, but these methods have not gained widespread use in practice **[98]**.

4.6. Determination of the degree of heat treatment of milk and dairy products

Because of the presence of potentially pathogenic microorganisms, milk must be heat treated. In the dairy industry, almost all milk and dairy products undergo some kind of heat treatment, and only a negligible proportion of traditional dairy products are made from raw milk. Heat treatment is sometimes insufficient to kill pathogenic germs, and sometimes, due to technological defects or deliberately, raw milk is mixed with pasteurized milk, which can be detected and the amount of raw milk added to heat treated milk estimated by the tests listed below **[98]**.

The Storch test may be used for milk which has been treated at a temperature above 80 °C or for more than 15 minutes at 75 °C, or for cream, sour milk and dairy products, cottage cheese and lump cheese made from such milk. The essence of the method is that the peroxidase enzyme present in raw or improperly heat treated milk, or in products made from such milk decomposes hydrogen peroxide, and the atomic oxygen liberated oxidizes N,N-diethyl-1,4-phenylenediamine hydrochloride to a gray or blue-gray compound **[99]**.

Quantitative determination of the phosphatase enzyme may be used for milk treated at a temperature below 80 °C, of above 75 °C for less than 35 seconds, or at 65 °C for 30 minutes, or for dairy products made from such milk **[100]**. In raw or insufficiently heat treated milk, or in pasteurized milk mixed with raw milk, or in products made from such milk, the phosphatase enzyme hydrolyzes disodium phenyl phosphate, the phenol liberated during the hydrolysis produces a blue color with 2,6-dibromoquinone-chloroimide, which is proportional to the amount of free phenol and can be measured by photometry **[101]**.

The phosphatase enzyme in raw or insufficiently heat treated milk, or in neat treated milk mixed with raw milk, or in dairy products made from such milk liberates ortho-cresolphthalein from hydrogen ortho-cresolphthalein phosphate, which gives a purple color at basic pH. The color indicates that there is phosphatase enzyme present in the sample, and so the sample did not undergo the desired heat treatment **[101]**.

4.7. Detection of abnormal milk from inflamed udders

Suitable for this purpose are the mastitest kits and the Whiteside test, which indicate the quantitative relationship between the nucleated cells (epithelial cells, leukocytes) in the milk, since the reagent releases the DNA from the nucleus and the extent of the reaction depends on the amount of this mucus-like substance **[102]**. Within three to five days after calving and during the last month of lactation, milk has a higher content of epithelial cells, so a positive reaction during these periods does not indicate udder disease. The two tests cannot be used for diagnostic purposes, but examination of the milk of the cows can be beneficial as even a mild reaction in the mixed milk indicates inflammation of the udders, poor udder health **[103]**.

4.8. Detection of the amount of spoiled milk unfit for consumption

The alizarin test, which is based on the detection of changes in the acidity and pH of the milk, is suitable for this purpose. The test can be used to isolate milk from an inflamed udder in the barn, but it can also be used to monitor changes during transport or storage. Since proteins in milk lose their original shape as a result of the increase in acidity, the pH change may also indicate whether the milk is suitable for the manufacture of dairy products such as UHT milk or milk powder. From the reaction of the alizarin indicator and the milk it can be concluded whether the pH of the milk has changed in the acidic or alkaline direction and how this change affects the technological properties of the milk **[101, 104]**.

5. Discussion

One can encounter news of food counterfeiting in the media almost every day. There is hardly any food that has not yet been counterfeited. In most cases, counterfeiters are one step ahead of the people who try to catch them. The many sophisticated ways of counterfeiting can only be combated by the development of anti-counterfeiting organizations all over the world, by the development of national anti-counterfeiting strategies, by the introduction of official measures to detect counterfeiting and the imposition of severe sanctions, and by sanctioning food manufacturers and distributors responsible for the counterfeiting in the cases brought to life by the authorities. In addition to the methods outlined in our paper, there is also a need for international cooperation against counterfeiting, for the harmonization of the strategies and measures, and for regular monitoring action.

In most cases, counterfeiting affects only the organoleptic and compositional characteristics of food and so they usually receive little media coverage, although the result of counterfeiting can sometimes be life-threatening or even fatal (such as the counterfeiting of infant formula with melamine).

In almost all cases, the purpose of counterfeiting is to obtain unlawful financial gains. Counterfeiters do not care, and often do not know, what the consequences of consuming their products are, their sole purpose is to maximize profits. Counterfeit components are often unknown, therefore they are often very hard to detect.

In the second part of our paper, in connection with the counterfeiting of milk and dairy products, it was shown that although counterfeiters are always a step ahead of the professional carrying out the control measurements, with the advancement of analytical chemistry and food analytics, methods that can detect the fact of counterfeiting are constantly being developed. With our paper, we hoped to draw attention to food counterfeiting, the fight against counterfeiting, consumer awareness and the protection of consumers from poor quality and counterfeit foods.

Finally, in connection with food counterfeiting, let us quote the essay of Béla Hamvas titled *„Roux soup”*:

„Food counterfeiting is undoubtedly the most serious of crimes. In some respects, it comprises treachery, blasphemy, poisoning, cheating and lying, and all this in an underhanded and hidden way; due to abominable greed it abuses the notion that if you are hungry, you have to eat. However terrible it is, DOSTOEVSKY is right: kick me, beat me, humiliate me, spit on me, just give me something to eat, to eat. There is only one thing more serious than food lie: false prophecy, which poisons people with false thoughts. We have almost lost the noble and real bread. There is hardly any greater shortage, and therefore the pain is deeper. Roux soup is so simple and modest that no one has thought of counterfeiting it. Maybe it's because it is so cheap that it's not worth it. In any case, our situation is not hopeless, we still have the music of Bach and our Palazzo Pitti, we have Velázquez and Hölderlin, normality has not completely disappeared as long as we have roux soup, potatoes and cooked rice **[106]**.”

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