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Jean Baptiste Alphonse Chevallier. Science applied to public health and social welfare

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Key words: analytical chemistry, arsenic poisoning, biological chemistry, falsification of foodstuffs, hygiene, legal medicine, occupational medicine, pharmacology, plant extracts, public welfare, toxicology, vinegar.

RESUMEN. Jean-Baptiste-Alphonse Chevallier (1793-1879), farmacéutico francés, realizó importantes investigaciones en química analítica, biológica y legal. Separó y estudió los principios activos presentes en muchas plantas. Sus más grandes contribuciones fueron en las áreas de salud e higiene públicas, protección al obrero, medicina ocupacional, adulteración de alimentos, medicinas y productos comerciales, métodos de identificación y legislación de protección al consumidor.

ABSTRACT. Jean-Baptiste-Alphonse Chevallier (1793-1879) was a French pharmacist who carried on significant research in analytical, biological, and legal chemistry. He isolated and studied the active principles present in many plants. His most important contributions were in the areas of public health and hygiene, protection of the worker, occupational medicine, adulteration of foodstuff, medicine, and commercial products, methods for their identification, and consumer protection legislation.

Life and career¹⁻⁴

Jean-Baptiste-Alphonse Chevallier was born in Langues, Lorraine, on July 19, 1793. In 1807, at the age of 14 he moved to Paris and began working in the *Muséum d'Histoire Naturelle* as a laboratory assistant to Louis Nicolas Vauquelin (1763-1829), André Laugier (1770-1832) and Pierre François Guillaume Boullay (1777-1869). At the age of 17, he was called into the Army, was wounded at the Battle of Leipzig (October 16-19, 1813), and left unfit for further military service. Chevallier returned to Paris to work as an intern in the pharmacies of the Saint-Lois La Pitié and the Midi Hospitals while continuing his pharmacy studies under Boullay. In 1822, he completed his studies at the *École de Pharmacie* de Paris and sometime afterwards he opened his own pharmacy at the place de point Saint-Michel, which he kept until 1835. During this period he began to publish the results of his first researches; mainly in the area of plant extracts. These were good enough to gain him an appointment to the pharmacy section of the *Académie* of Medicine in 1824 and to the public health section of the *Académie* in 1835, and to the Council on Hygiene and Health of the Seine department (1831). He was appointed chevalier to the *Legion d'Honneur* and afterwards promoted to officier. In 1825, he

became editor of the *Journal de Chimie Médicale, de Pharmacie et de Toxicologie*, to which he contributed a large number of papers. The editorial board of this journal included some of the most famous chemists and pharmacists of this time, among them Nicolas Jean-Baptiste Guibourt (1790-1887), Jean-Sébastien-Eugène Julia de Fontanelle (1790-1842), Jean Louis Lassaigne (1800-1859), Laugier, Mathieu Joseph Bonaventure Orfila (1787-1853), and Anselme Payen (1795-1878). According to Gallard,² an oversight accident of one of his students led him to abandon prematurely his business and dedicate himself to teaching and research; in 1835 he accepted a position as assistant professor of pharmacy at the *École de Pharmacie* and opened his own analytical laboratory. His numerous publications in the area of legal medicine and toxicology led him to be one of the founders of the *Société de Médecine Légale* (1868), of which he was one of its first vice presidents (1869). Chevallier died in Paris on November 29, 1879, at the age of eighty-six years. Chevallier's contributions covered a very ample range of subjects: plant extracts, analytical biological, general, and vegetable chemistry, hygiene, pharmacology, toxicology, legal medicine, public welfare, work safety, adulteration of foodstuffs, medicines, and commercial products. Together with his colleagues he took an active part in helping to identify the possible cause of death in many crimes and accidents. Chevallier separated and identified the active principle in many plants;⁵⁻¹⁷ in the area of public health he studied the manufacture and use of hypochlorites,¹⁸⁻²³ and worked on the sanitation of the Saint-Martin Canal, the sewers of Paris, London, and Montpellier, and the Bicêtre;²⁴⁻²⁷ in the area of occupational medicine he studied the health situation of workers in the printing area,²⁸ poisoning caused by lead,^{29,30} iodine and bromine,³¹ Paris green,³² quinine sulfate,³³ toxic colors in toys,³⁴ potassium bichromate,³⁵ copper,^{36,37} phosphorus,³⁸ and arsenic.^{33,40} In the area of adulteration of foodstuff^{41,42} we can mention that of vinegar,^{43,44} cane sugar,⁴⁵ coffee and chicory coffee,^{46,47} and milk.^{48,49} Chevallier published a history of the distribution of water in Paris since the year 360⁵⁰ and with Gustave Lagneau (1827-1896) an extensive study of the population movements in the city of Paris.⁵¹

Scientific contribution

Chevallier was a very prolific writer, he published, alone or with some of his collaborators, several hundreds of papers in the areas of plant extracts, analytical biological, general, and vegetable, chemistry, hygiene, pharmacology, toxicology, legal medicine, public welfare, work safety, falsification of foodstuffs, medicine, and commercial products. Some of these were also published as books.^{20,21,38,41,44,47,48,52-57} A paper by Sicard gives a detailed list of the publications.²

Plant extracts

Chevallier's initial research activities, alone or with Lassaigne, were in the field of plant extracts; some his results are as follows:

Arundo donax (giant reed, Provence reed)

The fresh roots of this plant were known to have a sweet flavor and had been used for a long time as a medicine for stopping milk secretion. The roots, treated with boiling water, yielded a brew having a bitter agreeable taste, which turned red litmus paper; produced a flocculent precipitate when treated with silver nitrate and a trifle one when treated with barium nitrate. An alcoholic extraction, followed by analysis of the residue and evaporation of the extract, indicated that the roots of the cane contained a slightly bitter mucous extract, an aromatic bitter resinous matter having properties similar to

those of the aromatic resinous matter of vanilla, malic acid, an essential oil, a nitrogen-containing substance, a substantial amount of sugar, potassium malate, nitrate, sulfate and phosphate, calcium sulfate, and silica.⁵

Chenopodium vulvaria

According to Chevallier and Lassaigue most of the vegetables, which had been analyzed thus far showed the presence of an acid. They had analyzed the composition of *Chenopodium vulvaria* (stinking goosefoot), a plant, which spread a smell of rotten fish when touched, and found it to contain a free alkali, which seemed to be ammonia. Their results indicated that the plant produced white fumes when in contact with vapors of nitric acid. Crushing the plant with boiling water left a green precipitate and a yellow green solution, which turned blue litmus paper previously reddened by an acid. This solution released a strong smell of ammonia when treated with KOH, did not precipitate a solution of barium nitrate but produced an abundant precipitate with silver nitrate, limewater, lead acetate, chlorine, gallnut, and concentrated sulfuric and nitric acids. A large amount of the plant, distilled (in the absence of water) in a water bath yielded a milky liquor, having a smell of rotten fish stronger than that of the plant itself. Treatment of this liquor with different reagents indicated that it contained ammonium bicarbonate (which Chevallier and Lassaigue believed it was liberated by a putrefying animal matter present in the live plant), albumin, osmazone (the sapid part of meat), a small amount of an aromatic resin, a bitter substance soluble in water and alcohol, a large amount of potassium nitrate, and traces of potassium acetate, sulfate, and tartrate.¹¹

***Chelidonium glaucum* (yellow horned poppy)**

Another work was related to the nature of the stomach disturbance principle present in yellow horned poppy.¹² After testing in many animals the action of the plant juice and its water and alcoholic extracts, Chevallier and Lassaigue concluded that the active principle should actually be considered a diuretic. Crushing the plant with boiling water left a yellow green extremely bitter solution, which reddened blue litmus and precipitated a solution of lead acetate, ammonium oxalate, silver nitrate, limewater, gallnut, chlorine and nitric acid. When heated, a thin layer appeared at its surface, which increased in volume and thickness as the heating was continued. Eventually the layer precipitated as a brown flocculent matter. Distillation of the latter produced an alkaline product having a strong fetid smell, similar to burnt horns. All these results indicated that the precipitate was albumin containing a small amount of a calcium salt. Evaporation of floating liquid to a syrupy consistency led to the precipitation of a yellow white substance that was shown to be calcium citrate. The remaining liquid was further concentrated by evaporation and extracted with alcohol. The alcoholic extract was evaporated to dryness yielding a yellow brown resinous material having a highly disagreeable taste. Further extractions with alcohol and water and analysis of the fractions, led to the conclusion that the juice of yellow horned poppy contained a deep yellow bitter resin, a yellow orange gum-resinous nauseous bitter substance; a mucilaginous substance, albumin, malic acid, calcium citrate and phosphate, potassium nitrate and chloride, and a small amount of silica.¹²

***Chara vulgaris* (Muskgrass)**

Chevallier and Lassaigue became interested in chara (a freshwater green algae containing a viscous substance in its inside) because of the particular characteristics it

presented when in contact with air: the green color disappeared; the plant became white and went through a substantial reduction in volume.¹³ They macerated a certain amount of wet chara and filtered the yellow juice that separated. This liquid was tasteless, did not color litmus paper, and produced a precipitate with nutgalls, silver nitrate, ammonium oxalate, and barium nitrate. The green matter remained in the filter paper and was found to be insoluble in water and soluble in ether. Addition of water to the ethereal solution produced a green-flaked precipitate of oily composition and tasting like fish. When treated with sulfuric acid, the chara effervesced strongly releasing CO₂. The resulting liquid was diluted with water and treated with KOH. The resulting brown flocculent gelatinous precipitate was insipid and burned over incandescent carbon it released a smell like burnt horn. The resulting cinder contained calcium sulfate and chloride, and sodium chloride.

Cytisus laburnum

A short note published Charles Louis Cadet de Gassicourt (1769-1821) reporting that the seeds of *Cytisus laburnum*⁵⁸ had strong emetic and purgative properties, led Chevallier and Lassaigne to try to separate the active principles contained in the grains.¹⁴ They reported that an aqueous infusion of the seeds had a slight yellow color; it turned red litmus and produced a precipitate when treated with gallnuts, nitric acid, lead acetate, silver nitrate, and limewater, and ammonium oxalate. No reaction was observed when treated with barium nitrate. Heating the infusion led to precipitation of a flocculent coagulum, having all the properties of coagulated albumin. According to Chevallier and Lassaigne, these results indicated the presence of a large amount of an animal matter (containing nitrogen).

The seeds were now boiled with alcohol and the tincture filtrated. On cooling, a fatty material separated as grainy flakes. The remaining liquid was evaporated to the consistence of an extract and then digested with water. The remaining green residue had all the properties of chlorophyll. The aqueous yellow solution, having a nauseating bitter smell, was mixed with lead in order to precipitate the albuminous matter, which it contained, and then treated with a stream of hydrogen sulfide to throw down the excess of lead, which it contained. The filtrated liquid was evaporated leaving as residue the peculiar emetic principle of these seeds. This substance had a disagreeable taste; eight grains of it swallowed at intervals occasioned vertigos, strong spasmodic contractions, flushing of the face, increased the velocity of the pulse, and occasioned violent vomiting. These symptoms lasted two hours, and left the person who had swallowed the substance in a state of considerable debility. The active principle had a greenish yellow color, it was not precipitated by acetate of lead, but it was by lead subacetate. It was also precipitated by silver nitrate, ammonia oxalate, and barium chloride. Chevallier and Lassaigne remarked these last three reagents did not precipitate emetine, the peculiar principle of ipecacuanha.¹⁴

Further chemical analysis indicated that the seeds of the *cytissus laburnum* contained a fatty matter of a greenish-white color, albumen, a green vegetable coloring matter, malic and phosphoric acids, potassium and calcium malate, and a small amount of silica.¹⁴

The active principle, which Chevallier and Lassaigne named *cystisine*, was purified by mixing the alcoholic extract with basic acetate of lead, followed by treatment with hydrogen sulfide, concentration by evaporation, extraction with alcohol, and concentration by evaporation. Cystisine appeared as a colorless crystalline mass, subliming undecomposed, having a strongly alkaline reaction, and deliquescing in the

air. Cystisine proved to be fatal to many animals of different species: Death occurred after vomiting and convulsions.¹⁴

Chevallier and Lassaigue found that the flowers of arnica (*Arnica Montana*) also contained a nauseous bitter substance similar to cytisine.¹⁶

Berries of *Taxus baccata* (yew)

In another work Chevallier and Lassaigue studied the composition of the berries of yew (*Taxus baccata*), which on expression liberated a transparent viscous and mucilaginous juice having a sweet sugared taste.¹⁵ Mixing the juice with alcohol produced a flocculent precipitate, soluble in water and having the properties of a gum. The liquid fraction was concentrated to a syrupy consistency; its analysis indicated the presence of malic and phosphoric acids. Chevallier and Lassaigue concluded that the berries contained a sugary non-crystallizable matter, gum, malic and phosphoric acids, and a carmine-colored fatty material (soluble in alcohol and insoluble in water).¹⁵

Additional sources studied included the fungus *Telephora coerulea*; Virginia snakeroot (*Aristolochia serpentaria*), common mallow (*Malva sylvestris*), field bindweed (*Convolvulus arvensis*), and hedge bindweed (*Convolvulus saepium*).^{7-10,17}

Disinfection with chlorine compounds

Chevallier carried on several detailed studies on the preparation and use of hypochlorites for disinfecting purposes. In the first of these, he described the manufacture of solutions of *chlorure de chaux* (lime chloride, calcium hypochlorite) for neutralizing putrid miasmas.¹⁸ As warned by Chevallier, the name *chlorure de chaux* should not be confused with *chlorure de calcium* (calcium chloride). Smithson Tennant (1761-1815) prepared dry calcium hypochlorite for the first time in 1798 and then used its solutions for bleaching purposes (cotton, clothes, starch, paper paste, etc.). Afterwards, Antoine Labarraque (1777-1850) applied it successfully to the disinfection of putrefying animal substances. These and other results had clearly shown the advantages of using calcium hypochlorite for neutralizing putrid emanations, without the inconveniences, which resulted from using chlorine fumigation. Chevallier suggested that the best way of preparing a solution of calcium hypochlorite was dissolving 100 g of the dry salt (marking 90 to 100 degrees in Gay-Lussac's chlorometer) in 1000 g of ordinary water. The solid salt was first ground in a mortar and the appropriate amount of water was added slowly. The resulting solution was then filtered and stored in well-closed bottle; it had a density of 2.421 at 0 °C and contained 32 g of chlorine, marking 90⁰ in Gay-Lussac's chlorometer.¹⁸

In a following publication, Chevallier described the construction and operation of the equipment for manufacturing large amounts of dry calcium hypochlorite.¹⁹ The reactor was a silica brick box, 1 m x 1 m x 1.8 m, hermetically close to avoid chlorine leaks, provided with a glass window allowing the operator to judge the advance of the reaction by the color of the internal vapors. Inside the box were a number of plates, arranged one on top of the other, for holding the calcium carbonate. Chlorine gas was admitted through a tube located in one of the walls; its flow-rate was adjusted to avoid an exaggerate increase in temperature because then the product would contain calcium chlorate, inappropriate for bleaching and disinfection. The excess chlorine left through another tube in the walls and was absorbed in limewater. The resulting solution contained calcium hypochlorite, which could also be used for the main purposes of the apparatus.

¹The structure of apparatus for preparing chlorine varied from one installation to the other; generally it was constructed of lead. The pertinent chemical reaction was carried on decomposing sodium chloride with sulfuric acid or hydrogen chloride, in the presence of manganese dioxide. The quality of the calcium hypochlorite thus produced was established using Gay-Lussac's chlorometer.¹⁹

In a later publication, Chevallier described an American apparatus designed to produce concentrated solutions of chlorine, useful for sanitation purposes. It consisted basically of a rotating horizontal drum, fed with a mixture of powdered red lead oxide, water, sodium chloride, and concentrated sulfuric acid. The drum was rotated during half-an-hour and then the contents filtered. The concentrated chlorine solution was appropriate for bleaching purposes using the Claude-Louis Berthollet's (1748-1822) method.²²

Chevallier published two books summarizing his experiences using chlorine and hypochlorites for disinfection, sanitation, and industrial purposes, as well as medical ones, particularly phthisis (pulmonary tuberculosis), syphilis, animal bites, ringworms, gangrene, chilblains, etc., elimination of odors, cleaning of sewers, removal of bodies from their tomb, etc. The books described the best procedures and most economical for preparing the hypochlorites and their different applications.^{20,21}

Falsification of foodstuffs

The subject of falsification of foodstuffs, medicines and chemicals was one of the most investigated by Chevallier. Here it is described a few of them.

Vinegar

As a result of a task commissioned to him by the Council de Salubrité, Chevallier decided to investigate the purity of the vinegar sold in Paris and the means used to adulterate it. Cleverly, he requested from a poor woman to buy for him vinegar in 120 shops. Their analysis indicated that 97 were pure vinegar, 17 were vinegar adulterated with sulfuric acid, three contained acrid substances, two contained copper, and one contained lead. From conversation with the sellers he found that many of them did not know that addition of foreign substances to vinegar was a crime; others thought that by making their product stronger, they were perfecting it (and even bought recipes for doing so), and others practiced it in order to sell it for a more competitive price. Whatever the reasons, Chevallier realized that it was necessary that the health authorities should have (and make public) procedures for identifying the adulteration of vinegar; that pharmacists should be encouraged to analyze substances, which were assumed to be falsified and that severe laws should be enacted to punish those who endangered public health.⁴³

Adulteration of vinegar with sulfuric acid was an old practice and several procedures had been proposed for detecting it. The best one was based on the addition of an aqueous solution of barium chloride. A small amount of a yellow flocculent precipitate

[1] Gay-Lussac's chlorometer, or bleaching power measurer, was an apparatus used to quantify the bleaching power and chlorine content in any of the common combinations of this element with the alkalis or alkaline earths. It was designed on the basis that chlorine destroys ten times its volume of a solution of indigo of suitable strength. These ten volumes were themselves divided into hundred parts; the decolorizing of one of these parts constitutes what was called one degree, and indicated in the chloride the presence of 1 L chlorine/kg. The best chloride decolorized 100 parts, that is, it marked 100 degrees, and contained 100 L chlorine/kg.

was formed if the vinegar did not contain sulfuric acid; otherwise there was an abundant white green precipitate, even when the vinegar contained only 1 % of sulfuric acid. The amount of sulfuric acid present was determined from the weight of the precipitate. Chevallier suggested that a very simple qualitative procedure for detecting the presence of sulfuric acid was to evaporate to dryness a sample of the suspected vinegar. If the vinegar was pure, the emerging vapors did not present any irregular characteristics but if it contained sulfuric acid, at the end of the evaporation the vapors were white and very dense and suffocating.⁴³

Similar procedures were available for detecting the presence of hydrogen chloride: A sample was distilled until all the HCl had passed over and the distillate treated with silver nitrate. A white precipitate of silver chloride appeared if the sample contained HCl; this precipitate was soluble in ammonia.

Chevallier also provided procedures for identifying the presence of other acids seldom used with vinegar, such as nitric, oxalic and tartaric acids; and acid substances. In the later case it was enough to evaporate the vinegar to a high consistency and taste the residue.⁴³

The presence of lead or copper in the vinegar was the result of poor fabrication technique or inadequate selection of the materials used for storing it. The presence of these metals was easily demonstrated by (1) streaming hydrogen sulfide through the vinegar, (b) addition of ammonia (in the presence of copper the color of the vinegar became blue); (c) addition of potassium ferrocyanate (a chestnut colored precipitate was formed in the presence of copper); and (d) potassium chromate (a yellow precipitate was formed in the presence of lead).⁴³

Chevallier also described several techniques for determining the concentration of the vinegar with the help of KOH, sodium bicarbonate, calcium carbonate, ammonia and simply by measuring its density with an acetimeter.⁴³

This paper about vinegar falsification was the first of many others on the subject, which culminated in the publication of a book.⁴⁴

Cane sugar

According to Chevallier, adulteration of cane sugar and beet sugar with glucose (obtained from potatoes starch) was a long known and spread fraud, driven by the large difference in price with glucose, 56 cents for 0.5 kg raw sugar against 12 cents for the same weight of glucose.⁴⁵ Chevallier's initial labors to find an adequate process for detecting and quantifying the foreign substance had initially failed, but in 1842 he reported to the *Société d'Encouragement* that he had succeeded in his efforts with a method based on the color developed when treating the sugar with a concentrated solution of KOH. Boiling molasses treated with the alkaline solution they acquired a brown color while molasses mixed with a small amount of glucose (2 to 3 %) turned deep black. In 1841, Charles Frédéric Kuhlmann (1803-1881) had reported a similar process to the *Société d'Encouragement* of Lille. Chevallier's procedure consisted in boiling in a glass tube a test solution containing 5.8 g of sugar, 1 g of KOH and 33 g of water. A slight change in color was observed when the sugar was pure; if the sugar was mixed with glucose, there was a clear change in color, whose intensity increased significantly for higher glucose contents. Chevallier found that his procedure was not only appropriate for sugar of different sources (e.g. white sugar, beet sugar, and Indian sugar) but that it also allowed to determine approximately the amount of foreign substance by the intensity of the color developed. In his publication he provided a table of the color achieved for a wide range of proportions, for each of the above three sugars, starting from a mixture containing 100 parts of the pure material and 18 of KOH (118

parts in total), down to 50 parts of sugar, 50 parts of glucose, and 18 parts of KOH (118 parts in total). To assure the validity of his test, he requested from Lassaigne to repeat it; Lassaigne reported that using a solution of KOH and cuprous sulfate allowed determining the presence of $1/1000^{\text{th}}$ of glucose in the sugar. Chevallier went one step further and suggested that the people involving in this testing prepared a series of sealed test tubes containing different amounts of cane sugar and glucose, arranged in proper color gradation, for a quick estimation of the amount of impurity.⁴⁵

Chicory coffee

According to Chevallier, fabrication of an artificial coffee based on the roasted roots of chicory originated in Holland where it was practiced for more than one century until 1801 when some French industrialists imported the manufacturing process.⁴⁶ The roots were first washed to eliminate the soil, cut into fine pieces, dried, roasted in iron cylinders until they achieved the desired color, and then ground and packaged. The powder was mixed with a variable proportion of coffee and the resulting mixture had a bitterness, which originated from the chicory and that consumers considered refreshing. Eventually it was not mixed with coffee, and brewed alone. Ultimately, the manufacture of chicory coffee became an important French commercial activity with an annual production of about 6000 (1839). This prosperity led promptly to adulteration of product by addition of many cheaper materials, which although did not affect the health of the consumers, they represented a criminal offense.⁴⁶

Chevallier listed the following as the most common adulterations: (1) addition of ground bricks, ocher, soil, and sawdust. These substances were easily detected by calcination and analysis of the cinders, and comparison against a true sample; (2) addition of coffee dregs. Detection of the foreign material was very easy. The suspected mixture was dried well and the resulting powder thrown on top of water. The particles of chicory coffee absorbed water and dropped to the bottom while the dregs remained on the surface; (3) addition of toasted substances which could be recognized with the help of an aqueous solution of iodine (giving a blue tint) or iron persulfate (giving a black tint), e.g. bread, the remains of semolina, vermicelli, acorn, cereals, legume seeds, and old bark; (4) addition of cacao hulls. In this case the falsification was detected with the help of a magnifying glass; and (5) addition of roasted sugar beets or carrots. No known detection procedure was available for the latter.⁴⁶

Milk

In a paper published in 1856, Chevallier and O. Réveil wrote that although a chemical analysis allowed verifying if milk was a pure material or had been adulterated, to carry on the analysis required especial knowledge, laboratory abilities and instruments, which were not common. For these reasons, they thought appropriate to develop an instruction manual, which would allow ordinary citizens to carry on the necessary tests for detecting the falsification of milk.⁴⁹

Chevallier and Réveil wrote that the most common adulteration of milk practiced in France was addition of water. The municipal authorities had provided their inspectors with a galactometer (a sort of an hydrometer), an apparatus, which measured the density of milk, assuming that this parameter reflected the purity of the product. The stem of the galactometer carried five marks (0, 1, 2, 3, 4.). Mark 1 indicated pure milk; mark 2, 25 % dilution with water; mark 3, 33 % dilution, and mark 4, 50 % dilution. Most of the instruments available in the commerce were made of metal, which dented; many times the stem separated slightly from the ball, causing liquid entrance and falsification of the lecture, etc. Chevallier and Réveil, believed that this instrument was highly unreliable

and that the inspectors were not aware of its limitations. Some of them made their measurements on milk at room temperature, others on boiling milk, and others on milk added to melting ice. They also did not consider the circumstances that could affect the composition of milk; no attention was paid to the year season, to the grade of the milk (whole, skimmed, etc.), etc, etc. Failure in the results resulted in confiscation of the milk or prohibiting its sale.⁴⁹

Before describing their procedure, Chevallier and Réveil gave a short review of the composition and properties of milk. Basically, milk was an aqueous emulsion containing nitrogenous substances such as casein, albumin, lactose (milk sugar), a yellow and a red coloring matters, aromatic substances, and soluble and insoluble salts (magnesium and calcium). In this emulsion were suspended small spherical transparent globules of the fatty material that constituted butter. The casein was partly dissolved and partly present as very small globules. The accepted composition was 86.2 % water, 4.38 % butter, 5.27 % lactose, 3.8 % casein, and 0.27 % salts. From a chemical viewpoint, the important components were butter, casein, lactose, and water. Fresh milk was normally in an alkaline state but in contact with air, it easily became acid due to the transformation of a small amount of lactose into lactic acid. Fresh milk could also be acid, depending on the food given to cows. On boiling, milk did not coagulate but became covered by a layer of frangipane. Left to itself, milk separated into two layers, the upper one was unctuous and formed by larger butter globules mixed with milk (milk cream). The lower white layer, retaining a small amount of fatty material and having a density of 1.033, was called skimmed milk. Left alone, it coagulated under the influence of casein; part of the lactose converted into lactic acid, acetic acid, and sometimes alcohol and CO₂. The remaining clear yellow liquid was called serum.⁴⁹

The common adulteration procedure was to withdraw part of the cream and add water. This decreased the density of the milk and changed its flavor, so the operator corrected these defects by adding substances such as starch, flour, dextrin, gum tragacanth, sugar, glucose, caramel, molasses, gelatin, oil seeds, white of eggs, etc, etc.⁴⁹

The method they proposed required four instruments, a lactometer, a thermometer, a cream meter, and a saccharimeter. The manual they wrote gave detailed instructions about the use of these instruments and use of their results to determine the degree of purity of a given milk. The lactometer was an aerometer of ordinary shape, having a scale divided in two sections, giving the density of full milk and skimmed milk respectively. It was marked with the density of water (1000), of pure milk (1031), and skimmed milk (1033). The stem carried marks ranging from 14 to 42, corresponding to densities 1014 and 1042, respectively. Tables were provided for making the temperature corrections. The amount of sugar was determined by precipitating the serum with diluted sulfuric acid, followed by titration using the Fehling liquor. Commercial non-adulterated milk contained between 38.36 to 46.44 g sugar/L; any sugar giving a lower reading was supposed to be adulterated with water.⁴⁹

Chevallier summarized all his research results on the subject of falsification of foodstuff and chemicals in what is probably his most important book, a dictionary about the alteration and falsification of foodstuff, medicines, and commercial products, and the means of recognizing them.⁴²

Arsenic testing

In 1836, James Marsh (1794-1846) published a paper describing a method for separating and identifying small amounts of arsenic from other substances with which it might be mixed.⁵⁹ According to Marsh, in spite of the improved methods that had been developed for detecting small amounts of the element in the content of the stomach, and

mixed with various other animal and vegetable matters, no analytical procedure had yet been developed that allowed its proper detection, particularly for forensic purposes. Marsh believed that one possible procedure could be based on the reduction of arsenic trioxide (named then arsenic) with nascent hydrogen and its following conversion into gaseous arsine (AsH_3). This gas would separate from the liquor spontaneously and could be collected and analyzed with any standard gas analysis equipment. Marsh brought these ideas into practice and developed the procedure and apparatus for separating very small amounts of arsenic from gruel, soup, porter, coffee, and other food liquors. His method was able to eliminate all the arsenic as arsine mixed with a little hydrogen. Since hydrogen was usually prepared by reacting metallic reacting zinc with sulfuric acid; Marsh warned that the operator had to be sure that the zinc and sulfuric acid he used were free or arsenic. The resulting arsine was burned and the arsenic recovered as metallic element if the flame was directed towards a cold surface, as arsenic trioxide if the flame was directed into a long tube, opened at both ends, or as a mixture of arsenic metal and arsenic trioxide if the flame was directed obliquely (at about 25°) into the tube, so as to graze it.⁵⁹

The Royal Society of Arts of London considered Marsh's procedure so valuable that awarded him the Large Gold Metal.

In the first part of a paper published in 1843, Cavallier made a long review of the information available about the reaction of arsenic (trioxide) with nascent hydrogen, the technique, and equipment developed by Marsh, and a summary of the many modifications and criticisms that had proposed.⁶⁰ Thus, he mentioned that the reaction between arsenic and hydrogen was discovered by Carl Wilhelm Scheele (1742-1786) in 1775, and further discussed by Joseph-Louis Proust (1754-1826) in 1798, Johann Bartholomäus Trommsdorf (1770-1837) in 1803, Friedrich Stromeyer (1776-1835) in 1806, Joseph-Louis Gay-Lussac (1778-1850) and Louis-Jacques Thenard (1777-1857) in 1808, Georges Simon Sérullas (1774-1832) in 1821, and Eugène Souberain (1797-1859) in 1830. Chevallier wrote that it was not possible to know if Marsh has based his procedure on the results reported by Sérullas, who had suggested that decomposition of arsine could be used in toxicology for determining the presence of arsenic or its compounds.⁶⁰

Many famous chemists commented Marsh's paper in a positive or critical manner, among them, Jöns Jacob Berzelius (1779-1848), Henri Braconnot (1780-1855), John Herapath (1790-1868), Justus von Liebig (1803-1883), Karl Friedrich Mohr (1806-1879), Mathieu Joseph Bonaventure Orfila (1787-1853), Thomas Thomson (1773-1852), and Heinrich August von Vogel (1778-1867). According to Liebig, Marsh's method had a sensibility that surpassed imagination as long as hydrogen was streamed slowly; while Thomson doubted its accuracy because he had found that antimony was also able to react with hydrogen, to form stibine (SbH_3), which on burning deposited either antimony or antimony trioxide. He believed that this fact disqualified the use of Marsh's procedure in legal medicine. Chevallier rejected this conclusion stating that the examiners could easily examine the product obtained after burning, to determine if it was arsenic, antimony, or a mixture of both. He listed a comparison of all the procedures that allowed differentiating between arsine and stibine. He went on to state that since 1838 he and other scientists involved in legal medicine had used successfully used Marsh's procedure in many cases of suspicion by means of arsenic. Among them, he mentioned Marie-Guillaume-Alphonse Devergie (1798-1879), Étienne Ossian Henry (1798-1873), and Ollivier d'Angers.⁶⁰

Anyhow, Chevallier believed that the apparatus designed by Marsh was hard to build and its use required too much time. For this reason he and his colleagues proposed a

very simple alternative construction, based on a gas-washing bottle; one of the entering tubes was used for introducing liquids and the other for releasing the arsine. The first step was to examine if the zinc contained arsenic. For this purpose, zinc filings were introduced in the open bottle, the bottle was then closed and sulfuric acid added; after a few minutes the gas tube was opened and the emerging arsine lighted over a porcelain plate. The latter did not stain if the zinc was free of arsenic and antimony. Zinc was now put in the clean apparatus and the liquid to be examined and sulfuric acid were now introduced with the help of funnel. The emerging gas was lighted over a porcelain plate or inside a tube, as before, and the stains were tested for presence of antimony and/or arsenic.⁶⁰

Chevallier used his apparatus to test several arsenic sulfides, prepared artificially or existing in nature, and reported that natural sulfides contained small amounts of arsenic trioxide.⁶⁰

In a following paper, Chevallier reported his results about the presence of arsenic in different products.³⁹ The first example was related to the manufacture of sulfuric acid using the pyrites originating from Sicily. It was said that this acid contained arsenical derivatives and consequently, all other chemicals using this kind of acid in their manufacture (e.g. phosphoric and acetic acids, hydrogen chloride, etc.). The switch to this type of pyrites allowed a substantial reduction in the cost for producing sulfuric acid and had been powered by the large increase in the price of sulfur caused by its large use in disinfection of vineyards. The presence of arsenic in this acid had already been reported by several well-known chemists, among them, Pierre Jean Robiquet (1780-1840), Antoine Bussy (1794-1882), and Orfila. Orfila had suggested a procedure for purifying the acid by bubbling hydrogen sulfide through, followed by filtration to eliminate the precipitated arsenic sulfide, and followed by bubbling air to eliminate the excess of hydrogen sulfide. Chevallier remarked that the *Conseil d'Hygiène Publique et de Salubrité de Bordeaux* had made known to the French government the potential disadvantages of the use of arsenical sulfuric acid in the manufacture of food or medical preparations; as a consequence, Éugène Rouher (1814-1884), the minister of agriculture, commerce, and public works, had ordered (January 25, 1861) the health inspectors to check in their routine inspection of food and medical preparations factories, if this type of acid was being used.³⁹

Chevallier also wrote that arsenic had clearly been identified in phosphorus, phosphoric acid, hydrogen chloride, wood vinegar, sodium sulfate, and alum, prepared using sulfuric acid prepared from pyrites.⁶¹

Phosphorus poisoning

In 1830, Charles Marc Sauria (1812–1895) discovered the principle of the phosphorus match. After watching a demonstration of the reaction of sulfur mixed with potassium chlorate, Sauria experimented rubbing the prepared end of his match on a wall where there was some phosphorous. His match immediately ignited, and so did the development of the match industry. The poisonous effects of white phosphorus were noted as soon as its manufacture for matches started growing. Matches containing white phosphorus were dangerous to the workers manufacturing it and also to the general user when heated they release phosphorus fumes. Swallowing match heads also become a common method of committing suicide and swallowing match heads often poisoned small children. Those involved in the manufacture of the new matches were afflicted with *phossy jaw* with necrosis of the jawbone, the principal symptom of chronic phosphorus poisoning, and there was enough white phosphorus in one pack to kill a person.⁶²

In 1844, Gustaf Erik Pasch (1788-1862) invented the safety match; Patsch's idea was very simple: separation of the oxidizer and the tinder, thus removing the fire risk caused by self-ignition. The head of the match contained only potassium chlorate and no phosphorus so that ignition could only be achieved by striking the head on another surface (this is the safety match, as we know it today). A critical development took place in 1845 when Anton Schrötter (1802-1875) discovered red phosphorus, an allotropic form of the element. This discovery led to the fast development of the match industry in which white phosphorus was replaced by the red variety. In Sweden, the Lundström brothers, Carl Frans and Johan Edvard, developed and sold a safety match in which white phosphorus was replaced by red phosphorus, the phosphorus being confined to a strip on the box and the oxidizer used to tip the matches. These matches ignited perfectly and the friction surface did not deteriorate.⁶²

According to Chevallier and Poirier, the uncontrolled sale of red phosphorus was a menace to society; an easy source of the chemical was chemical matches prepared using this element.⁶³ The matches were treated with water and the solution used for criminal purposes. The poisoning effect was substantially stronger than that of arsenic; antidotes were readily available for poisoning caused by arsenic, copper, zinc, and lead, but none was known for phosphorus. In a booklet published on the subject, Chevallier and Abel Poirier gave a detailed historical review of the large number of cases of phosphorus poisoning (criminal, suicide, or accidental) that had occurred in France between 1824 and 1858, as well as a report of the death of different animals (birds, donkeys, dogs, etc.) caused by ingestion of pills or solutions containing phosphorus. Many fires were known to be a result of negligent use of chemical matches based on red phosphorus. In addition, the people working in factories manufacturing this kind of matches faced the possibility of becoming affected by jawbone necrosis, a fatal disease accompanied by intense pains. Chevallier and Poirier believed that most of these poisoning situations could be avoided by prohibiting the use of red phosphorus in the manufacture of chemical matches in their present procedure, by using the Lundström process, commercialized in France by the Coignet brothers. In this procedure a paste was prepared, which only ignites when rubbed against a small flat surface covered with red phosphorus.

In another publication, Chevallier gave a more detailed explanation of the dangers caused by phosphorus to the health of the workers working in its industry, and as source of many of the fires occurring in France.³⁸ Medical studies showed clearly that the manufacture of chemical matches using red phosphorus was an insalubrious industry, because it was not only the source of serious illnesses but also the cause of many deaths. Pregnant women handling the paste used to manufacture the matches always aborted or gave birth to a seriously ill baby.

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