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HISTORY AND BIOGRAPHY

On the Pelletier and Caventou (1817, 1818) papers on chlorophyll and beyond

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Abstract
The first use of the word 'chlorophyll' (chlorophile or chlorophyle in the French original) appeared in two papers by Pierre-Joseph Pelletier and Joseph Bienaimé Caventou, pharmacists in Paris who isolated and studied the green pigment from plants. Here, we provide English translations of their 1818 note and the slightly longer 1817 paper. Historical context is provided including a timeline of key discoveries in chlorophyll chemistry pertaining to photosynthesis.

Keywords Chlorophyll · Hans Fischer · Alexander Krasnovsky · Eugene Rabinowitch · Richard Willstätter · Robert Woodward

Introduction

The plant sciences have touched nearly every aspect of humanity and consequently have an extraordinarily rich history. Understanding of the history of the plant sciences can be enriched by bringing to the fore key studies that may be little appreciated in the modern era. For example, as to who was the first to ask and answer the question concerning where the oxygen comes from; the answer given by De Fourcroy, in 1787, was "water"—long before anyone had said so; this answer was given in a book, in French (for its English translation, and discussion, see Joliot et al. 2016). The answer to the question of who first named chlorophyll is Pelletier and Caventou in 1817 and 1818. A beautiful account of the lives and scientific contributions of Pelletier and Caventou, both pharmacists, has been provided by Delépine (1951), but the word chlorophyll is mentioned only once therein, obscured by the focus on their isolation of quinine and other medicinal agents from plants.

The present paper begins by consulting Rabinowitch (1945, 1951, 1956) to highlight historical accomplishments concerning 'chlorophyll' in photosynthesis. We then present a translation, in English, of a note by Pelletier (1818) on the green matter of the leaves. A translation is provided (along with notes concerning selected terms in early nineteenth century French) in the Supplementary Material of the slightly longer, preceding paper by Pelletier (1817). The present paper concludes with a brief timeline for perspective concerning key events in chlorophyll chemistry particularly in relation to the understanding of photosynthesis.

What did Eugene Rabinowitch and a few others say about 'chlorophyll'?

A background on the life and discoveries of Eugene Rabinowitch (1898–1973), the prophet of everything related to photosynthesis, has been published (Govindjee et al. 2019). It was Rabinowitch and Weiss (1936) and Porret and Rabinowitch (1937) who had discovered the reversible oxidation and reduction of chlorophyll in solution, by using iron compounds in the reaction medium! They clearly demonstrated the existence of light-oxidized chlorophyll. It was much later that Bessel Kok (1957) showed that chlorophyll (Chl) *a*, located in the reaction center of Photosystem (PS) I was oxidized, i.e., P700 became P700$^{+\bullet}$. Thus, the Rabinowitch and

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Weiss reaction of chlorophyll (later shown to be a dimeric special pair of chlorophylls; see McElroy et al. 1972; Norris et al. 1974) represents the primary photochemical reaction of photosynthesis. For the suggestion of another chlorophyll *a* (P680-P690) in PS II, see Krey and Govindjee (1964), and Rabinowitch and Govindjee (1965); soon thereafter, it was discovered in the laboratory of Horst T. Witt (see Dőring et al. 1967; and proven not to be an artifact, in his Lab, by Govindjee 1970).

The Pelletier (1817) paper was indeed mentioned by Rabinowitch (1945) as a true "philosopher's stone" of organic synthesis, a veritable "elixir of life" (see Dutrochet 1837).

In contrast to the reversible photochemical oxidation of chlorophyll *a* (see above), Alexander Krasnovsky (1948) discovered a reversible photochemical reduction of chlorophyll using ascorbic acid. This reaction was named the Krasnovsky reaction. For further information on the work of Krasnovsky, see Karapetyan and Govindjee (2014). In this reaction, photoreduction of chlorophyll was shown to be accompanied by the formation of a 'pink' chlorophyll absorbing in the green region of the spectrum, the structure of which was elucidated by Scheer and Katz (1974); in darkness, it regenerated chlorophyll. Although chlorophylls in both P700 and P680 are oxidized in light, neighboring chlorophylls are reduced at the same time, serving as one electron acceptors. In PSI, it has been shown be a Chl *a* molecule (for a review, see Mamedov et al. 2015). Further, the Krasnovsky reaction has the potential of being exploited for 'artificial photosynthesis'.

Historically, and philosophically, it is important to mention that it was Louis N.M. Duysens (1921–2015) who in his doctoral dissertation (Duysens 1952) had coined the term "P" for the reaction center pigments that serve as the electron donor in the primary reaction of all photosynthetic systems, and was the first one to have discovered P890 in *Chromatium vinosum*, and P870 in *Rhodobacter sphaeroides*; see Duysens et al. (1956). For further information on the work of Duysens, see van Grondelle and van Gorkom (2014) and Govindjee (2016).

Today, the structure and function of chlorophyll are well known, and many books exist on it (see e.g., those edited by Vernon and Seely (1966), Scheer (1991), Grimm Porra Rüdiger Scheer (2006), and Ameen et al. 2022); also an excellent review on this topic is also available (Tamiaki 2022). For all aspects of the structure and the function of chlorophylls and bacteriochlorophylls, readers are referred to the masterpiece by Blankenship (2001) on the molecular mechanisms of photosynthesis.

These introductory remarks are closed by showing, in Fig. 1, pictures of both Pierre- Joseph Pelletier (1788–1842) and Joseph Bienaimé Caventou (1795–1877). Some information on their life and work beyond that described by Delépine (1951) is available on web sites (https://en.wikipedia.org/wiki/Pierre_Joseph_Pelletier, https://en.wikipedia.org/wiki/Joseph_Bienaim%C3%A9_Caventou), but still their contributions concerning chlorophyll appear little appreciated. We now turn to their paper where they coined the name "chlorophyll" (chlorophile or chlorophyle in the French

Fig. 1 Left: Pierre-Joseph Pelletier (22 March 1788–19 July 1842). Right: Joseph Bienaimé Caventou (30 June 1795–5 May 1877). They are oil paintings by Catherine Buisson, 1930, after Elisa Desrivières, 1870. Sources: <https://wellcomecollection.org/works/k5cxvenq> and <https://wellcomecollection.org/works/wqp8arqf>)

original) for the green pigment of plants and, as known today, present in all oxygenic photosynthesis. When anoxygenic photosynthesis is included, more than 50 chlorophylls and bacteriochlorophylls are now known to be present, all sharing the same basic tetrapyrrole structure and including not only the above-mentioned photochemically active pigments in the reaction centers, but also the much more abundant pigments responsible for light-harvesting in most photosynthetic organisms.

A translation, from French to English, of the Pelletier and Caventou (1818) paper follows

On the Green Matter of Leaves

By Mr. Pelletier and Mr. Caventou.

We have obtained this material by treating the thoroughly pressed and washed pulp of several herbaceous plants with cold distilled alcohol; after evaporation of the alcoholic liquid, it remained a dark green substance that was resinous in appearance, which, when crushed to powder and treated with hot water, acquired a high degree of purity by losing a small quantity of brown colored material. The green matter, thus obtained, showed all its already known properties: it dissolved completely in alcohol, ether, oils, alkalies; but chlorine immediately destroyed its green color, and produced a yellow flaky matter (Proust, *Journ. de Phys.*[this paper was published, in 1794, by Joseph Louis Proust, a French chemist, and dealt with the color of Prussian blue].

The green material, when exposed to air for several weeks, showed no alteration at all. When heated, it softens; but it does not melt; it decomposes at higher temperature but releases no scent of ammonia. It burns with flame; and leaves a voluminous charcoal that inflames/ignites easily.

Hot water dissolves a little bit of it; however, by cooling the sample, the green material partially precipitates. Acetic acid is the only fatty acid which dissolves it significantly.

Concentrated sulfuric acid dissolves it in the cold without alteration: water precipitates a fraction of it; the rest can be obtained by saturating the acid. Over time, the green material decomposes. The alcoholic solution shows no changes upon addition of an equal amount of sulfuric acid.

Hydrochloric acid, and especially nitric acid, destroy the green matter; with the latter, it neither yields mucic acid nor oxalic acid.

Iodine destroys the color of the green matter, just as chlorine does, but more slowly.

If, after mixing an alkaline earth or metallic salt to the alcoholic solution of the green matter, one adds an alkali or a carbonate, an abundant precipitate is formed, which, in most cases, entrains the green matter into a composite. We can prepare green lacquers inexpensively by adding to the juice of plants, obtained by pressing them out thoroughly, an earthy salt which is decomposed by alkali or carbonates. Almost all the lacquers we have prepared were stable in the light for several weeks: we note mainly those that we had obtained from common meadow plants, hemlock, elderberry, and alfalfa.

It follows from the facts presented in this note that the green matter of plants, incorrectly referred to as *starch* or *resin*, is a particular substance which must be classified among the highly hydrogenated plant substances; that it must be distinguished from the resins; that it comes close to several dyes, such as those of alkanet, turmeric, red sandalwood, and that it deserves, by its properties and the role it plays in plant function, to be considered as a *principe immédiat* of plants. We propose to give it the name chlorophyll (*chlorophyle*).

Comments on Pelletier and Caventou (1818)

Two terms in the aforementioned 1818 paper warrant comment. First, Pelletier (1818) here spelled *chlorophyll* as "chlorophyle," and one year earlier (Pelletier 1817) as "chlorophile"; today in French, however, it is spelled "chlorophylle". Second, the term *"principe immédiat"* was coined in the mid-eighteenth century (Venel 1751) to describe a genuine constituent, i.e., a substance pre-existing in a plant (or animal) that was isolated directly without adulteration and is "indissociable from its physiological function" (Tomic 2010, 2012). The terminology to describe and classify substances derived from natural sources was in great ferment in the first few decades of the nineteenth century. The term was replaced within a few years following the naming of chlorophyll as one of the "organic chemical species" (Tomic 2012).

Chemistry in the early nineteenth century must have been exhilarating, particularly in Paris, the capital of science during this era, where 100–200 pharmacists were engaged in isolating and characterizing compounds from natural sources (Tomic 2012). The number of organic compounds isolated by pharmacists and chemists exploded from ~ 100 at the time of Pelletier 1817 report to nearly 250 merely ten years later (Tomic 2012). A brilliant exposition of this "gestational period of organic chemistry" (1785–1835) has been provided by Tomic (2010, 2012); his writings capture the thrilling *esprit du temps* (i.e., Zeitgeist) among the Parisian pharmacists and chemists, and also provide in-depth illustration of the laboratory equipment for extraction, fractionation, and isolation typical of what Pelletier and Caventou would have had available to them.

Yet much of interest concerning the work of Pelletier and Caventou on chlorophyll may be lost to history. Pelletier was a brilliant student, receiving the "highest award in chemistry from the hands of Fourcroy" (Delépine 1951) in 1807 at L'Ecole de Pharmacy, the same Fourcroy (1755–1809) who was a contemporary of Lavoisier (1743–1794) and who in 1787 attributed water as the source of oxygen produced in photosynthesis (as stated in the introduction). Pelletier also received similar awards as a student in botany and natural history the following year (Delépine 1951). Fourcroy had deep interests in a rapprochement of the chemistry of the distinctly regarded animal and plant kingdoms and was renowned as a conceptualizer and educator (Tomic 2010, 2012). It is fascinating to wonder whether Pelletier's research in plant pigments was prompted by Fourcroy's intellectual preferences and earlier accomplishments in photosynthesis.

Pelletier and Caventou considered the chlorophyll isolated from plants to be pure, as indicated by their usage of the term *"principe immédiat."* The green pigments in plants are now well known to include chlorophyll *a* and chlorophyll *b* as well as the free base analog pheophytin *a*. The techniques that could provide information on the composition were hardly available in the early nineteenth century. Elemental analysis was an emerging method that would come into vogue in the 1830s (Tomic 2010); spectroscopic examination was several decades in the future (Stokes 1864); and methods of chromatographic fractionation would appear almost a century later (Tswett 1906). Hence, the sample of chlorophyll isolated by Pelletier and Caventou, understandably unknown to them, undoubtedly contained a mixture of chlorophyll *a* and chlorophyll *b* (perhaps along with trace amounts of pheophytin *a*) and carotenoids. Proof of the presence of two chlorophylls as well as several carotenoids in Pelletier and Caventou's "chlorophile" from plants was provided about a half-century later by Stokes (1864) and Fremy (1877).

Merely two years after the reports on chlorophyll, Pelletier and Caventou isolated quinine from the bark of the chinchona tree (Pelletier 1820). Pelletier and Caventou are duly lionized for their isolation of quinine, which sparked the beginning of the modern era of medicinal chemistry and enabled improved therapy for malaria. The recognition includes an imposing statue in St.-Germain-des-Prés, the Latin quarter of Paris, and a French postage stamp (Delépine 1951). Pelletier and Caventou are particularly revered for their humility in making their discoveries on quinine freely available to mitigate the scourge of malaria. That humility also is seen in their papers on chlorophyll. In the first paper (1817, see the Supplementary Material), Pelletier and Caventou humbly state *"Nous n'avons aucun droit pour nommer une substance connue depuis long-temps, et à l'histoire de laquelle nous n'avons ajouté que quelques faits"* (We have no right to name a substance known for a long time, and to the history of which we have added only a few facts). The plaque on the statue in St.-Germain-des-Prés states *"Par leur précieuse découvert / par leur désintéressement / ils ont mérité le titre / de bienfaiteurs de l'humanité"* (for their precious discovery / for their selflessness / they have earned the title / of benefactors of humanity). The statue, of course, celebrates their discoveries about quinine. The same could be said for their findings on chlorophyll, which comprise a milestone in the extraordinarily rich field of plant biology.

A timeline for selected discoveries on chlorophyll

A timeline for selected discoveries in chlorophyll chemistry is provided here. The list is by no means complete—the chemistry of chlorophyll and its role in plant biology occupy a vast literature—but the following is a brief perspective on the chronology of key advances (leaving the previous half-century for a future retrospective).

1817/1818

Pierre-Joseph Pelletier (1782–1842) and Joseph Bienaimé Caventou (1795–1877) name the green pigment of leaves "chlorophyll".

1864, 1877

Sir George Gabriel Stokes (1819–1903) and E. Fremy show that the green material isolated from plants is a mixture of two green and two orange components (Stokes 1864; Fremy 1877), as stated above.

1906

The Russian botanist Mikhail Semyonovich Tsvet (Tswett; 1872–1919), performing for the first-time chromatography of green leaf extracts, using glass columns filled with different adsorbents, detected besides a major chlorophyll band a second smaller chlorophyll band, both known today as chlorophyll *a* and chlorophyll *b*, as well as several yellow pigments, known today as carotenes and xanthophylls (Tswett 1906).

1907/1913

Richard Martin Wilstätter (1872–1942) and Arthur Stoll (1887–1942) provided the first chemical structure of chlorophyll: $C_{55}H_{72}O_5N_4Mg$ (see Wilstätter 1907) Their book, written in German, "Untersuchungen über Chlorophylle" (Willstätter and Stoll 1913), also provides information on the earlier research on chlorophyll in the nineteenth century. Richard Willstätter and his group considerably extended the work of Tswett using many different plants as well as green algae and provided a rough estimation of the levels of these photosynthetic pigments. Willstätter's coworker Arthur Stoll later went into industry where he extended chlorophyll research considerably by preparing demetalated chlorophylls, pheophytins *a* and *b*, in kilogram amounts, and providing them to researchers including one of the authors (HS).

1940

Hans Fischer (1881–1945) provided the first synthesis of heme and showed that heme and chlorophyll were members of the same molecular family, i.e., cyclic tetrapyrroles, thereby showing aspects of the unity of animal and plant biochemistry; see Anonymous (1934); and Macdonald (1945). Also, see Inman (1935) for work till then on the relation of chlorophyll to photosynthesis.

1960

Robert Woodward (1917–1979) described a synthetic path to chlorophyll (Woodward et al. 1960), the full description of which was reported posthumously (Woodward et al. 1990) showing completion of 46 of the intended 49 steps (for a review, see Liu et al. 2018).

1967

Ian Fleming (b. 1935) determined the absolute configuration of chlorophylls *a* and *b* (Fleming 1967); also see his website: http://www-fleming.ch.cam.ac.uk/.

1973

Charles E. Strouse determined the X-ray structure of a chlorophyllide (Strouse 1973; Chow et al. 1975).

Three of the aforementioned scientists received the Nobel Prize for studies concerning the structure and chemistry of chlorophyll. They are: Richard Willstätter in 1915 (https://www.nobelprize.org/prizes/chemistry/1915/willstatter/facts); Hans Fischer in 1930 (https://www.nobelprize.org/prizes/chemistry/1930/fischer/facts); and Robert Burns Woodward in 1965 (https://www.nobelprize.org/prizes/chemistry/1965/woodward/facts). In addition, Johann Deisenhofer (b. 1943), Robert Huber (b. 1937) and Hartmut Michel (b. 1948) received the 1988 Nobel Prize for the structure of a bacterial reaction center, showing the structures and arrangement of the bacteriochlorophylls therein; see:

https://www.nobelprize.org/prizes/chemistry/1988/summary.

Chlorophyll is indeed one of the most important molecules on Earth that sustains our life!

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Declarations

Conflict of interest The authors declare no competing interests.

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