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The synthesis of phosphor ethers: who was Franz Anton Voegeli?

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The synthesis of the first organophosphate cholinesterase inhibitor (tetraethyl pyrophosphate, TEPP) is credited to the French organic chemist Philippe de Clermont (1831–1921) and to the Russian chemist Wladimir P. Moshnin from Moscow, both working in the laboratories of Adolphe Wurtz in Paris. In his publications de Clermont describes however not only the TEPP synthesis but also that of the related compound triethyl phosphate (TEP). TEP was previously synthesized by the Swiss chemist Franz Anton Voegeli (1825–1874), working in the laboratory of Gustav Magnus in Berlin. While TEPP is a potent organophosphate cholinesterase inhibitor with an IC_{50} in the low nanomolar range, TEP has no anticholinesterase activity up to millimolar concentrations. Therefore de Clermont and Moschnin are indeed the fathers of the first organophosphate cholinesterase inhibitor (TEPP), but are not entitled to claim paternity of the first compound in the class of phosphoric acid esters (TEP), an honor which belongs to Franz Anton Voegeli.

1. Introduction

For people interested in the history of organophosphorus cholinesterase inhibitors Bo Holmstedt's chapter in Koelle's Textbook "Cholinesterases and Anticholinesterase Agents" is the "must read" monograph (Holmstedt 1963). It contains a detailed account of the synthesis of the first organophosphate cholinesterase inhibitor, tetraethyl pyrophosphate (TEPP) by the French organic chemist Philippe de Clermont (1831–1921), working in the laboratories of Adolphe Wurtz (1817–1884) in Paris (Fig. 1). Paragraphs from the two de Clermont's publications dealing with TEPP are reproduced by Holmstedt, including de Clermont's acknowledgment that TEPP was actually synthesized earlier by another student of Wurtz, named Moschnine (de Clermont 1854, 1855). Holmstedt concluded his remarks by noting that "*Nobody knows who Moschnine was*".

This missing piece in the puzzle of early organophosphate history was recently put in place by the publication in this Journal of an overview of the life and achievements of the Russian chemist Wladimir P. Moshnin from Moscow, who studied chemistry with de Clermont in Gießen, Germany, and followed him to Paris to work in the Wurtz laboratories (Petroianu 2008).

Recently, few people have actually read these de Clermont publications. Those who did may have noticed that de Clermont describes not only the TEPP synthesis but also that of the related compound triethyl phosphate (TEP, Fig. 2). New questions therefore arise, such as to why did Holmstedt choose to ignore TEP and who was the first one to have synthesized TEP. This brief account is an attempt to answer these questions.

2. Investigations, results and discussion

2.1. Why did Holmstedt choose to ignore TEP?

TEPP's prominence is related to the extreme toxicity of most organophosphates, the substance class to which TEPP belongs. TEPP is considered nowadays to be the

Fig. 1:
Philippe Henri Arnout de Clermont (seated) with his coworkers in his Paris laboratory. Original photo in the possession of the family [Antoinette F. de Ferrière]

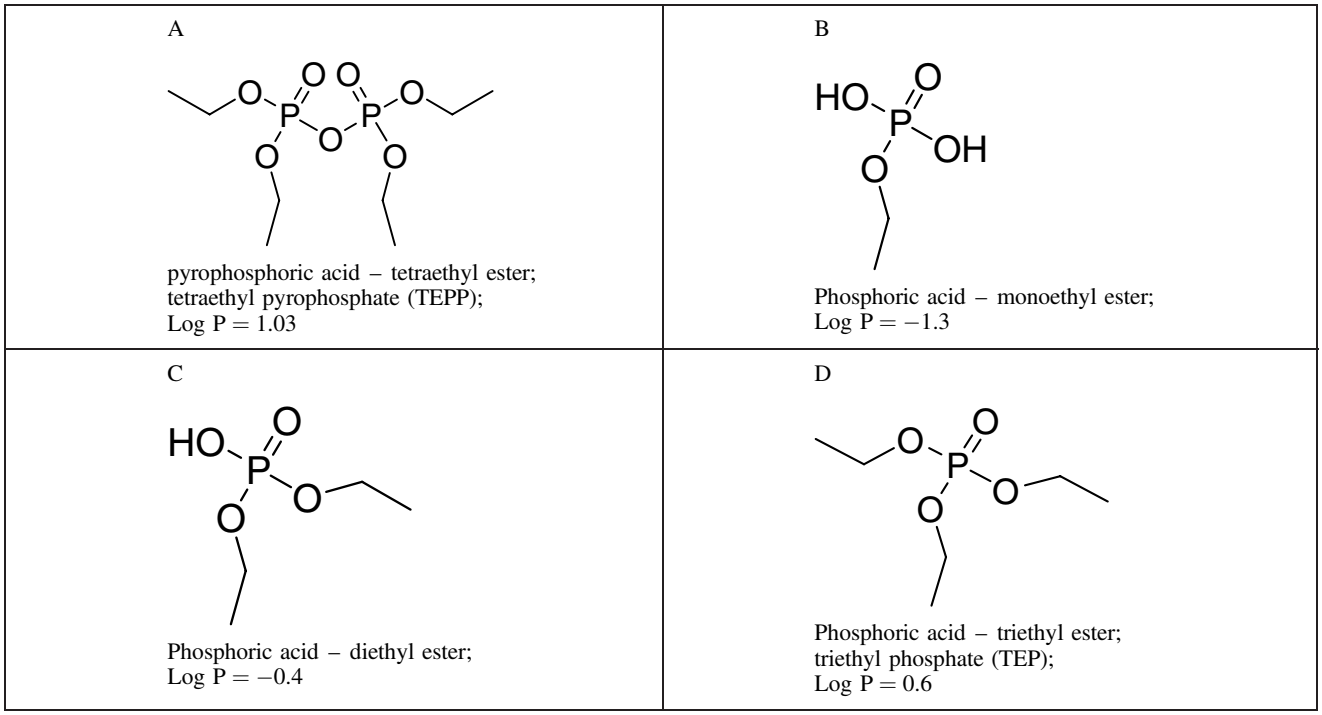


Fig. 2: Chemical structure of the compounds discussed
A: Tetraethyl pyrophosphate (TEPP) is the first organophosphate inhibitor of cholinesterase to be synthesized by the Frenchman Philippe de Clermont and the Muscovite Wladimir Moschnin, both *élèves* of Adolphe Wurtz in his Paris School of Chemistry
B: Phosphovinic acid or monoethyl hydrogen phosphate
C: Diethyl hydrogen phosphate was synthesized by Voegeli
D: Triethyl phosphate (TEP) is the first organophosphate ever to be synthesized by the Swiss Voegeli, working in Berlin in the laboratory of Magnus

first organophosphate inhibitor of cholinesterases. Of course neither the toxicity nor the mode of action of the new compound was known at the time, as evidenced by de Clermont's willingness to taste his product, which he describes as a sticky liquid with a burning taste and a peculiar odor (*"un liquide visqueux d'une saveur brulante (et) d'une odeur particuliere")*.

It was almost a century later, in 1932, that Willy Lange[1] and his graduate student Gerda von Krueger realized the toxicity of organophosphates: *"the fumes of these compounds have a pleasant, slightly aromatic odor. But a few minutes after inhalation there is a feeling of pressure to the larynx and difficulty in breathing. Then a disturbance of consciousness develops, as well as blurred vision and a painful oversensitivity of the eyes to light. Only after several hours do the problems wear off. They are apparently not caused by acidic products of a possible decomposition, but by the esters themselves. The effects are produced by very small amounts."* (Lange and von Krueger 1932).

The mechanism of toxicity would be recognized soon thereafter. According to Holmstedt (2000), it was Eberhard Gross[2] who identified inhibition of cholinesterases as the mechanism of toxicity of TEPP in 1939. Hans Gremels[3], working with nerve gases, came to the same conclusion in 1940 (Hahn 1970; Schmaltz 2006).

The same results concerning the mechanism of toxicity were obtained in 1941 on the other side of the English Channel by British researchers from the group led by Edgar Douglas Adrian, Nobel prize laureate (1932) and later Baron of Cambridge (1955), who were working with fluorophosphonates (Adrian et al. 1947).

Similar conclusions were also drawn in the Soviet Union during World War II by Aleksander G. Ginetzinsky (1895–1962) and his associate Zoya I. Barbashova (Rozengart 1996).

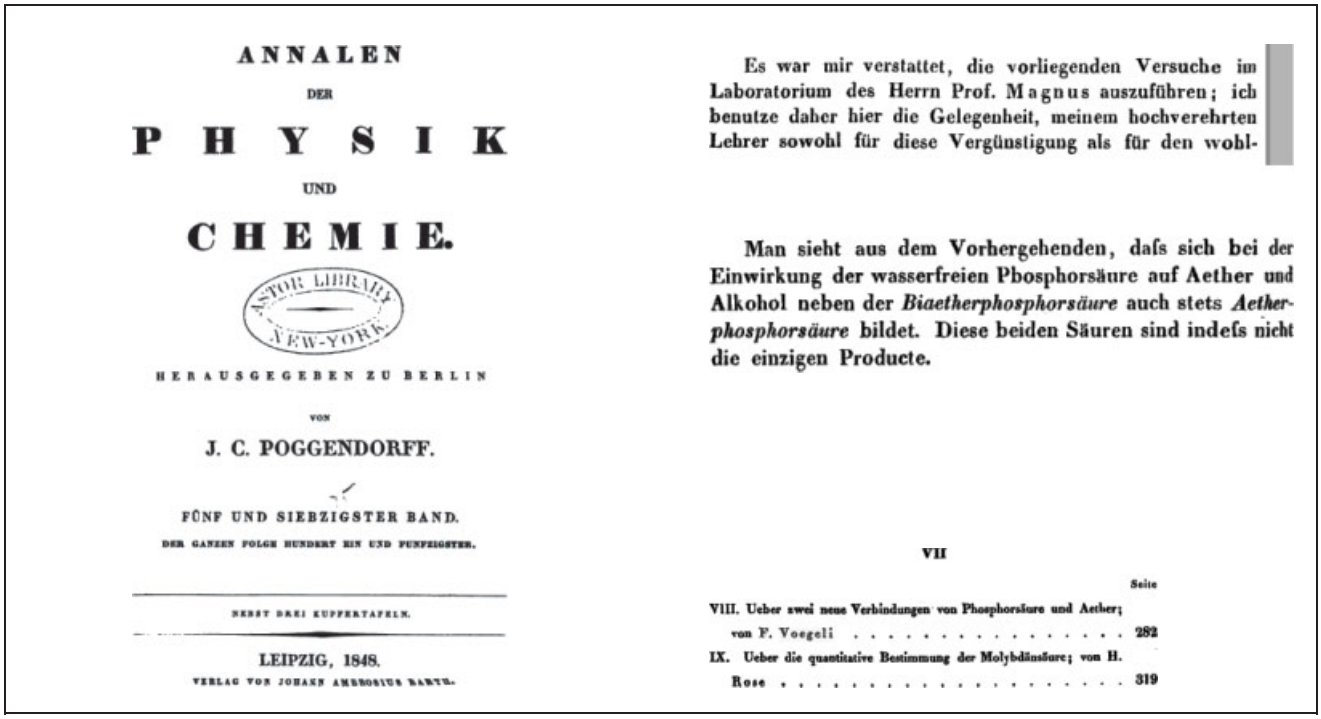
While TEPP is a potent organophosphate cholinesterase inhibitor with an IC_{50} in the low nanomolar range, TEP has no anticholinesterase activity up to millimolar concentrations (Gumbmann and Williams 1970; Petroianu, unpublished data).

Holmstedt's decision not to mention TEP was based on his knowledge that TEP has no anticholinesterase effect; therefore such a compound was of no interest to him in the context of the history of cholinesterase inhibitors. From the perspective of the history of organophosphates however TEP deserves the same attention as TEPP does.

2.2. Is de Clermont the first one to have synthesized TEP?

The synthetic approach used by de Clermont (TEPP and TEP) and Moschnin (TEPP) was made possible by the work of Alexander Williamson (1824–1904), an English chemist trained in Germany (in Heidelberg and Gießen) who became Professor at the University College in London. In the early part of the 19th century, the direct reaction between the "spirit of wine" (ethanol) and acids was the only known method of generating the elusive "ether" (a name applied to any volatile with a mostly pleasant odor, including compounds that nowadays would be called esters). Williamson serendipitously discovered a new way to produce ethers using ethyl iodide and potassium salts (nowadays called the Williamson reaction) thereby demonstrating, that ethers are not dehydrated alcohols (Wiliamson 1851, 1852). In addition, he was the first to correctly present the composition of ether (Priesner 1986).

The birthplace of triethyl phosphate (TEP) is the chemistry laboratory of Gustav Magnus (1802–1870), Professor of Physics and Technology at the University of Berlin. Magnus' interest in ether was only fleeting, but he super-



ANNALEN
DER
PHYSIK
UND
CHEMIE.

HERAUSGEGEBEN ZU BERLIN
VON
J. C. POGGENDORFF.

FÜNF UND SIEBZIGSTER BAND.
DER GANZEN FOLGE HUNDERT EIN UND FUNFZIGSTER.

MEBST DREI KUPFERTAFELN.

LEIPZIG, 1848.
VERLAG VON JOHANN AMBROSIUS BARTH.

Es war mir verstattet, die vorliegenden Versuche im Laboratorium des Herrn Prof. Magnus auszuführen; ich benutze daher hier die Gelegenheit, meinem hochverehrten Lehrer sowohl für diese Vergünstigung als für den wohl-

Man sieht aus dem Vorhergehenden, dafs sich bei der Einwirkung der wasserfreien Phosphorsäure auf Aether und Alkohol neben der *Biaetherphosphorsäure* auch stets *Aetherphosphorsäure* bildet. Diese beiden Säuren sind indefs nicht die einzigen Producte.

VII

Fig. 3: Cover page of Poggendorf's Annalen containing Voegeli's paper titled "About two new combinations of phosphric acid and ether". The author writes "*Permission was given to me, to perform the following experiments in the laboratory of Professor Magnus; I take therefore the opportunity to publicly express the most warm gratitude to my highly respected teacher, for both this favor and the benevolent advice, through which he offered substantial support to me.*" He continues " *One can see from the previous that from the action of water-free phosphoric acid on ether and alcohol in addition to diethyl phosphoric acid also always ethyl phosphoric acid is created*

vised the work of the Swiss student Franz Anton Voegeli, who was to become the father of both diethyl and triethyl phosphate (Voegeli 1848; Hofmann 1870, Fig. 2). His Ph.D. thesis (Dr. phil.) entitled "*De duobus novi acidi phosphorici cum aethere connubiis*" was published in 1848 in Berlin and reprinted in German in the Annalen der Physik und Chemie (Fig. 3).

2.3. Who was Franz Anton Voegeli?

Franz Anton, born October 22nd, 1825 hails from an old and well off Zurich merchant family. He was the second of four male children born to Johannes Vögeli (1798–1865) and Emerentiana Elisabetha Holzhalb (1805–1863): Johann(es) (1824–1900), Franz Anton (1825–1874), Arnold (1826–1915) and Friedrich Emil (1828–1847). The most prominent of the brothers was actually not Franz Anton but his junior Arnold.

Arnold (Fig. 4) entered at the age of sixteen the K & K (Imperial and Royal) Military Engineer Academy in Vienna where he excelled as a student (second best in his class). He served in the Austrian Army until 1861 which he left in the rank of a captain and after having received the Military Cross of Merritt (Militär-Verdienstkreuz). Subsequently he joined the Swiss Army where he rose through the ranks up to colonel and division commander. Involved in Zurich politics, he became in 1865 member of the city council (Stadtrat) and later of the canton council (Kantonrat). As President of the Committee in charge of the Swiss exhibition in Zurich 1883 (Landesausstellung) he oversaw important development works around the Zurich Lake; for his contribution, he received the Gold Merritt Medal of the city of Zurich. Based on the success of the Zurich exhibition he was appointed Swiss General Commissioner for the 1889 Paris World Exhibition.

His brother Franz Anton married in 1858 Charlotte Amalie Schweizer (1836–1898), the daughter of the famous theologian Alexander Schweizer (1808–1888), priest and professor of practical theology in Zurich. Five children were born to the couple: the two boys (and probably one of the girls) died at an early age so that only Emma Johanna (1859–1914), and Charlotte (1867–1917) reached adulthood. Charlotte married in 1887 a moderately renowned Zurich architect, Conrad von Muralt (1859–1928); the Villa Abegg, that now houses the Zurich University Centre for Ethics, and the Villa Seerose in Horgen are examples of his work. The couple had two children: Alex (n 1888) who became a physician and Franz (1890) who became a professor of agricultural engineering in Munich.

The last couple of years in his life Franz Anton lived in the Seidengasse 17 in Zurich; he died April 30th, 1874 before reaching his fiftieth birthday and was buried at the private family cemetery.

After the early years at school (probably in Zurich), in winter (Wintersemester) 1842–1843, two of the Voegeli brothers, Johann(es) (Jean) und Franz Anton (Francois) studied at the Faculty of Letters and Sciences of the Lausanne Academy, the precursor of the University of Lausanne (Fig. 5). From winter 1843 to March 1845 Franz Anton was registered (*immatrikuliert*) at the University of Zurich, with the registration number (Matrikelnummer) 967. Subsequently he worked as a doctoral student in the chemistry laboratory of Gustav Magnus, at Berlin University where he received, in 1848, his doctoral title (Dr. phil). The twenty-six page dissertation can be seen at the Museum for Medical History (Medizinhistorisches Museums) of Zurich University and at the State Library (Potsdamer Strasse) in Berlin.

In Lausanne Franz Anton became a member of Zofingia, the oldest Swiss student fraternity (Studentenverbindung)

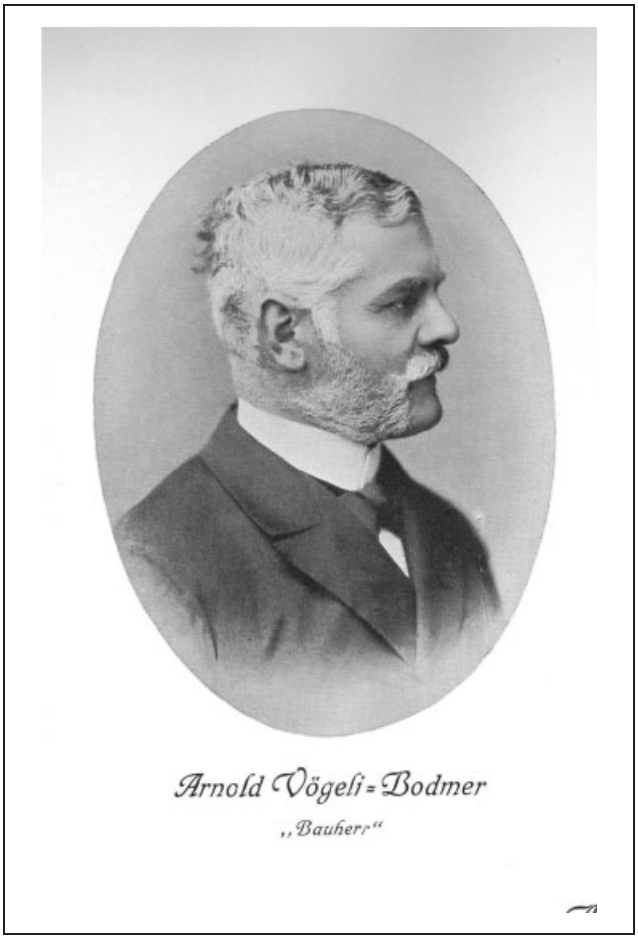


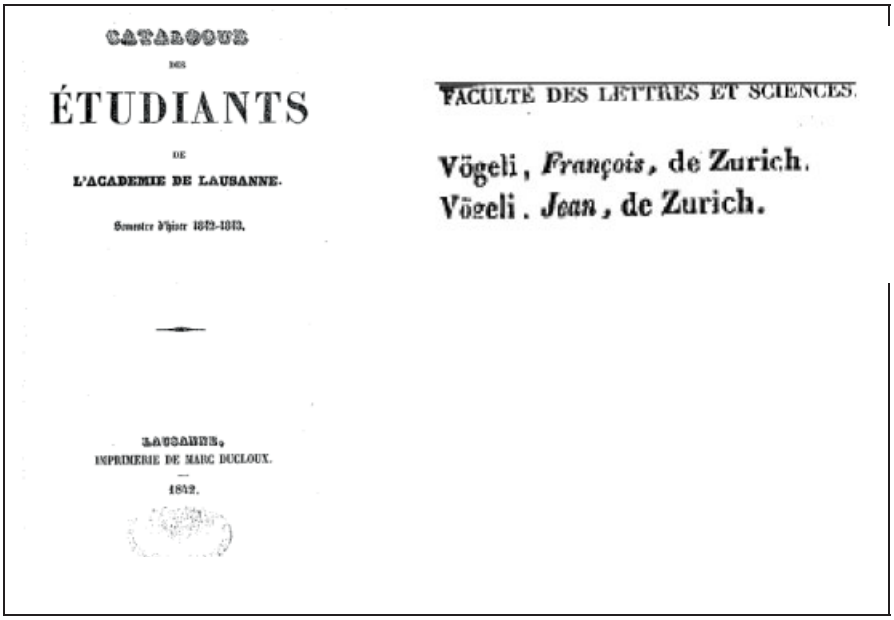
Fig. 4: Arnold Voegeli (1826–1915) Photo from “Hundert Jahre: Bilder aus der Geschichte der Stadt Zuerich in der Zeit von 1814–1914” published 1915 by Verlag Berichthaus

established in 1819. On November 10th, 1843 he transferred to the Zurich chapter of Zofingia and was registered under the number 623; he was active until 1846; the year he probably left for Berlin. After a hiatus of fifteen years Voegeli became active again and is one of the thirty-five senior members who established in Zofingen (the Swiss city which gave the organization its name) the “Alt Herren Bund” or Philisterium, an alumni network of previous Zofingia members that supported the student fraternity financially and otherwise (Ehinger 1994).

In 1848, the year he was granted the doctoral title, he became a member of the Natural History Society (Naturforschende Gesellschaft in Zürich), one of the oldest professional societies in the world, established in 1746. In the Journal of the Society (Mittheilungen der Naturforschenden Gesellschaft in Zürich) Heft III of 1848/1849 he published „Ueber zwei neue Verbindungen von Phosphorsäure und Aether“ essentially the same content as his publication in Annalen der Physik und Chemie. There is no information as to how long his membership continued but he is listed under number 70 in the 1859 membership directory as Dr. Voegeli F in Ravensburg. The members were numbered according to the year they joined the society: as an example Professor Schinz who joined the Society in 1799 had in the 1859 membership directory the number one, indicating the longest (living) membership.

With the doctoral title, Franz Anton’s interest in science diminished only to allow business activities to flourish. Together with his brother Johann(es) and with his father as a passive investor, Franz Anton applied in august 1851 for permission to establish a chemical factory in the city of Ravensburg, at the southern tip of the Kingdom of Wuerttemberg. His signature can be found on the application document (Fig. 6). Under King Wilhelm I, Wuerttemberg was one of the most progressive German states and the Central Board for Trade and Commerce (Koenigliche Zentralstelle für Gewerbe und Handel), which was created in 1848 under the leadership of von Sauter[4], was a strong supporter of economic activities. Another member of the Board was a friend of Magnus, Dr. Friedrich Ammermueller[5] (1809–1898), a teacher, chemist and physician of repute. Both the favorable attitude towards entrepreneurs in the country and the possible connection via Magnus to a key member of the Board might have influenced the choice of location for the future factory.

While the economic climate in Wuerttemberg was favourable, locally the opposition from the neighbors of the future chemical factory in Ravensburg was very strong. To settle the conflict, the Ministry of Internal Affairs asked for an expert opinion and a commission chaired by von


CATALOGUE

DES

ÉTUDIANTS

DE

L'ACADEMIE DE LAUSANNE.

Semestre d'hiver 1842-1843.

LAUSANNE,
IMPRIMERIE DE MARC DUCLOUX.

1842.

FACULTÉ DES LETTRES ET SCIENCES.

Vögeli, *François*, de Zurich.
Vögeli. *Jean*, de Zurich.

Fig. 5:
Franz Anton and Johann(es) Voegeli at the Academie de Lausanne; kindly provided by Frédéric Duflon, Service des Archives, UNIL

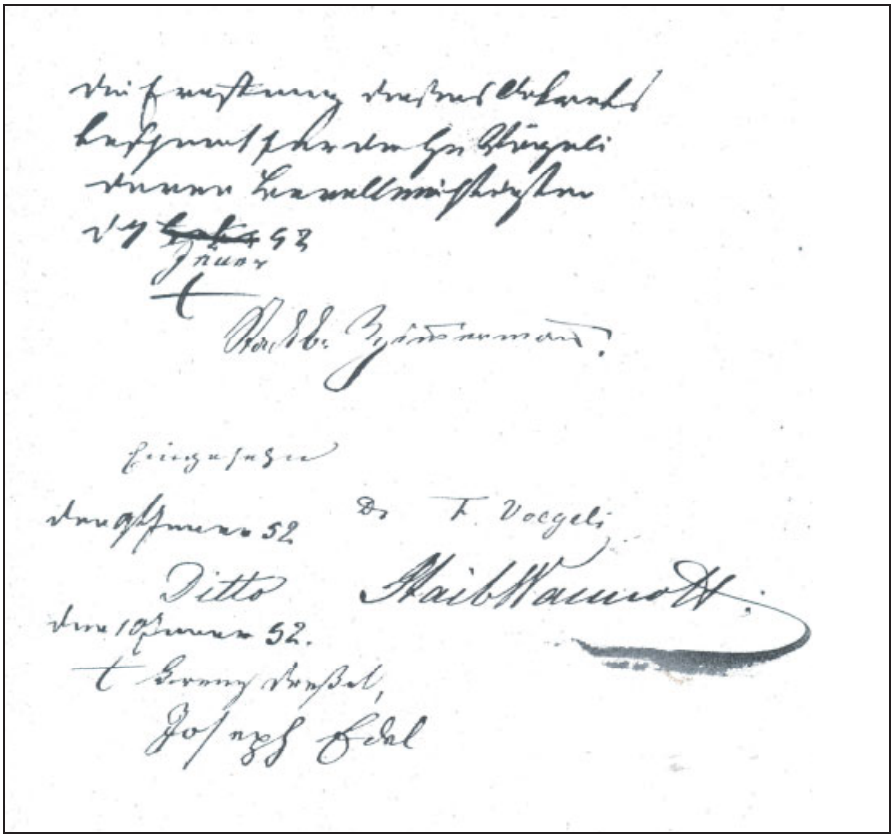
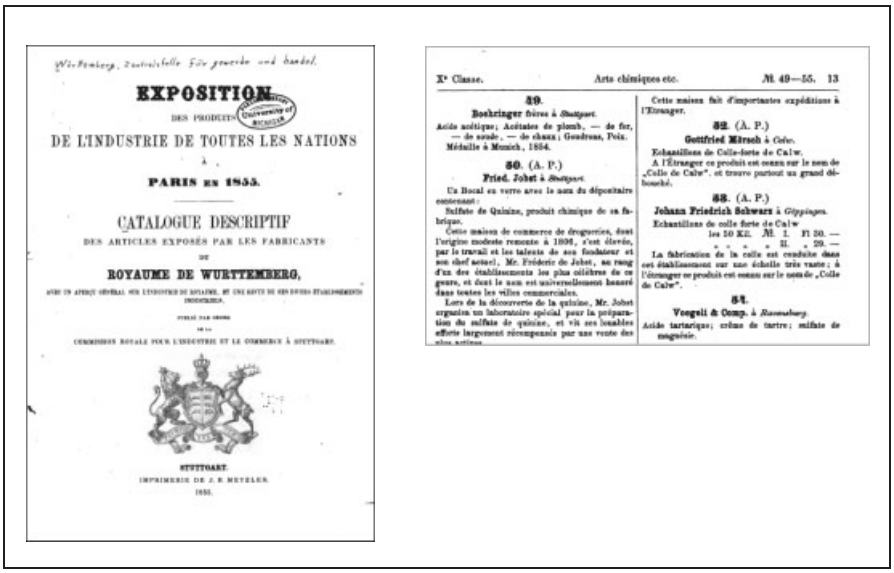


Fig. 6:
Signature of Franz Anton Voegeli

Sauter was constituted. Members were Dr. Ammermueller and the Central Board employee chemist Faisst[6] plus the external experts Bergrath (Mine Council) Bilfinger jr., Professor Karl Siemens from the Agricultural Academy Hohenheim[7] and the Stuttgart pharmacist Gayer[8] who previously owned a chemical factory.

The report, signed by von Sauter, stated *„Hiernach ist die Anlage der von Vögeli bei Ravensburg zu etablierenden Fabrik im allgemeinen Interesse als wünschenswert zu betrachten, da sie unter günstigen Verhältnissen entsteht, sowohl in Bezug auf die Lage des Ortes und den Preis den Kaufs hinsichtlich der Befähgung und der finanziellen Mittel der Gründer“* or „*Therefore the establishment by Voegeli of a factory in the neighborhood of Ravensburg is to be looked upon as desirable to the common interest, since it is created under favorable auspices, with respect to both location and purchase price and with respect to the ability and financial means of the founder*“.

Based on this report, on December 30th, 1851 permission was granted to establish a chemical factory in Ravensburg licensed to produce tartar (Weinstein), tartaric acid (Weinsteinsäure), potassium salts (Kalisalze), sulfuric acid (Schwefelsäure), alum (Alaun; potassium-aluminium sulfate) and potassium cyanide (blausaures Kali). Indeed Voegeli & Comp. participated at the 1855 Paris industrial exhibition in the pavilion of the Kingdom of Wuerttemberg together with now famous names such as the Boehringer brothers (Fig. 7). However, while details are lack-


EXPOSITION

DES PRODUITS

DE L'INDUSTRIE DE TOUTES LES NATIONS

À

PARIS EN 1855.

CATALOGUE DESCRIPTIF

DES ARTICLES EXPOSÉS PAR LES FABRICANTS

DE

ROYAUME DE WURTTEMBERG,

STUTTGART.

X^e Classe. Arts chimiques etc. № 49—55. 13

49.

Boehringer frères à Stuttgart.

Acide acétique; Acétates de plomb, — de fer, — de soude, — de chaux; Goudrons, Poix. Médaille à Munich, 1854.

50. (A. P.)

Fried. Jobst à Stuttgart.

Un Bocal en verre avec le nom du déposituire contenant:

Sulfate de Quinine, produit chimique de sa fabrique.

Cette maison de commerce de droguerie, dont l'origine modeste remonte à 1826, s'est élevée, par le travail et les talents de son fondateur et son chef actuel, Mr. Frédéric de Jobst, au rang d'un des établissements les plus célèbres de ce genre, et dont le nom est universellement honoré dans toutes les villes commerciales.

Lors de la découverte de la quinine, Mr. Jobst organisa un laboratoire spécial pour la préparation du sulfate de quinine, et vit ses louables efforts largement récompensés par une vente des plus actives.

Cette maison fait d'importantes expéditions à l'Etranger.

52. (A. P.)

Gottfried Mörsch à Calw.

Echantillons de Calle-forte de Calw.

A l'Etranger ce produit est connu sur le nom de „Calle de Calw“, et trouve partout un grand débouché.

53. (A. P.)

Johann Friedrich Schwarz à Göppingen.

Echantillons de colle forte de Calw

les 50 Kil. № 1. Fl 30. —

„ „ „ 2. „ 29. —

La fabrication de la colle est conduite dans cet établissement sur une échelle très vaste; à l'étranger ce produit est connu sur le nom de „Colle de Calw“.

54.

Voegeli & Comp. à Ravensburg.

Acide tartarique; crême de tartre; sulfate de magnésie.

Fig. 7:
Voegeli & Comp. is participating at the 1855 Paris industrial exhibition in the pavilion of the Kingdom of Wuerttemberg

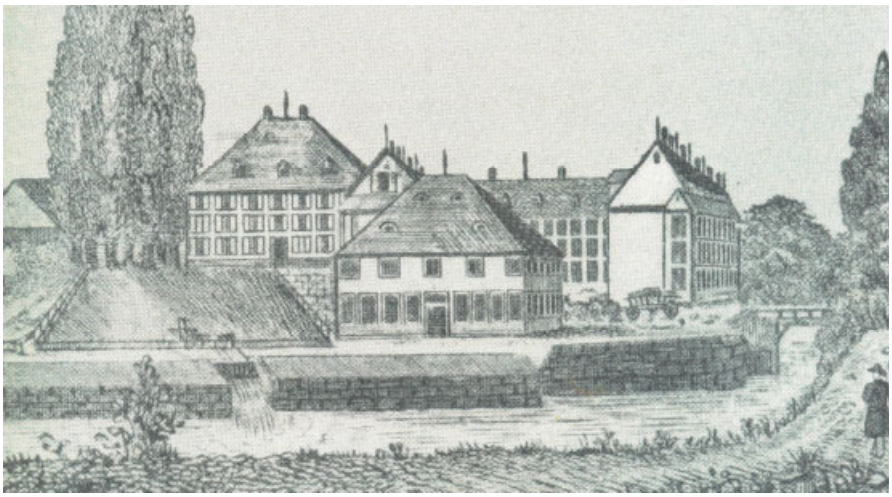


Fig. 8:
C. S. von Clais textile factory in Hard Wülflingen (since 1922 part of the city of Winterthur) operating the first mechanical, engine driven looms in Switzerland. Drawing from ≈ 1830

ing, thing did not go well; 1858 Voegeli returned to Switzerland and settled in Winterthur. In the 1863 Ravensburg list of businesses (Gewerbekataster) the Voegeli & Comp. chemical factory was not listed any more.

The next Voegeli business venture is a total departure from chemistry and has little relationship to his own expertise; In 1858 he became an "associé" of the C. S. von Clais textile factory in Hard Wülflingen (since 1922 part of the city of Winterthur) which operated the first mechanical, engine driven looms in Switzerland (Fig. 7). The factory was established in 1802 as one of the first in Switzerland to issue shares. While initially the enterprise was very successful, eventually problems arose and by 1841 ownership was entirely in the hands of colonel Carl Sebastian von Clais (1800–1858), the son of one of the founding shareholders. Von Clais died in 1858, and the ownership was transferred to his three sons, Carl, Arnold and Emil. They, however, lacked both the needed skills and unity to run the operation. While Carl favored selling the factory his views were overridden by Arnold and Emil who introduced Dr. phil. Franz Anton Voegeli as a partner. The fresh capital infusion was however insufficient to stop the decline of the factory and in 1866 bankruptcy was only serendipitously avoided when the Honegger brothers and Daniel Elmer-Wild decide to take over the property with liens of about 270 thousand for 341 thousand Swiss francs. The story of the textile factory in Hard Wülflingen is told in great detail by Emanuel Dejung and Max Ruoff (Dejung and Ruoff 1937, Fig. 8).

As to Voegeli himself, he moved subsequently to Grüningen looking for other investment options which however did not materialize. After that it became quiet around him, probably as a consequence of the losses he had incurred both in Ravensburg and in Hard. He passed away at the relatively young age of forty-nine.

In conclusion, de Clermont and Moschnin are indeed the fathers of the first organophosphate cholinesterase inhibitor (TEPP), but are not entitled to claim paternity of the first compound in the class of phosphoric acid esters (TEP), an honor which belongs to Franz Anton Voegeli. While this short biographical note does not exhaustively answer the question "who was Voegeli?" it should satisfy the idle curiosity of some. For those wishing to know more, it gives some direction for future investigations.

Acknowledgements: My gratitude to all those who helped gather the information and/or write the manuscript: G Ballak (Gemeinderatskanzlei Grüningen), C Battaglia (Stadt Zürich-Bevölkerungsamt), M Betschart (Stadtarchiv Winterthur), S Bolliger (Universität Zürich), F Duflon (UNIL, Lausanne), R Dresbach (Neustadt Weinstrasse), Dr R Dünki (Stadtarchiv Zürich), A de Ferriere (France), L Izzo (Calgary), P Jung (Zofingia-Zurich), G Kriegshäuser (Neustadt Weinstrasse), W Lammers (UAE University), W Raschle (Gemeindeamt des Kantons Zürich), Dr J Rauschert (Staatsarchiv Aargau), C Schummer (Universitaet Heidelberg Mannheim), Dr. A Schmauder (Stadtarchiv Ravensburg), Dr. H Stucki (Naturforschende Gesellschaft Zuerich), A Surber (Stadt Zürich-Bevölkerungsamt), B Weber (Zofingia-Zurich) – and to the miracle of Internet connectivity and Google Books.

[1] Willy Lange (1900–1976), who was married to Lilli Baermann, was forced by 1935 to leave the Nazi-dominated Friedrich-Wilhelms-University in Berlin, and the couple emigrated to the United States in 1939.

[2] Eberhard Rudolf Gross (1888– 1976) received his *venia legendi* in Heidelberg in 1922 under Albrecht Kossel. In 1926, he moved on to work for IG Farben with Schrader, and a year later he became a nontenured Professor (Weichardt 1976).

[3] Hans Gremels (1896–1949) received his *venia legendi* under Walther Straub in Munich in 1933 and went on to become *Ordinarius* (Professor and Chair) of the Pharmacological Institute in Marburg.

[4] Johann von Sautter (1807–1855); lawyer and chief government adviser (Oberregierungsrat), Director of the Central Office for Trade and Commerce up to his death 1855. His successor was Ferdinand Steibeis.

[5] Dr. Friedrich Ammermueller (1809–1898) was the son of Christoph F Ammermueller (1770–1852), a civil servant (Universitaets Kammeral Verwalter) at the University of Tübingen. Friedrich worked with Magnus and synthesized 1833 periodic acid (Überjodsäure) (Ammermüller F, Magnus G (1833) Pogg. Ann. 28: 614). Subsequently he taught in Reutlingen at a secondary school [Realschule; Oberlehrer und Vorsteher] from 1837 to 1852 whereupon he moved to Stuttgart. He was active in politics representing the Volkspartei and was elected to the Wuerttemberg Parliament (Landtag) and later to the German Zollparlament.

[6] The chemist Andreas Faißt (1821–1878) was a student of Hermann von Fehling (1811–1885) who was also an external adviser to the Board. Faißt left in 1853 the Board to become the Director of the sugar factory in Heilbronn. The Villa Faißt in Heilbronn, Cäcilienstr. 66 is a superb example of German architecture.

[7] Professor Karl Siemens daughter Antonie (1840–1900) married 13. July 1869 her distant relative Werner von Siemens.

[8] The Stuttgart pharmacist Gayer is most probably Carl Heinrich Geyer. He married in 1846 Julie Kreuser, the daughter of the pharmacist Christoph Heinrich Kreuser. Geyer and the brother of his wife Christian took over the Kreuser pharmacy in Stuttgart. Christian was a member of the Association for Patriotic Natural History in Wuerttemberg [Verein fuer vaterlaendische Naturkunde in Wuerttemberg]; members were also Ammermueller, Siemens, von Fehling and Bilfinger jr.

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