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Timeline and bibliography of early isolations of plant metabolites (1770–1820) and their impact to pharmacy: A critical study

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ABSTRACT

Plant metabolites became objects of chemical research for pharmaceutical and medicinal reasons. The period of pure plant substances in chemistry started 1770 with isolation of tartaric acid from wine (wine in pharmacy is a plant-derived preparation). Carl Scheele isolated 7 plant acids: tartaric, benzoic, citric, oxalic, malic, glucuronic and gallic. The era of alkaloids started 1803 when narcotine was discovered and published. Since that time, pharmacists and toxicologists began to recognize alkaloids (or substances regarded as such) as highly active principles responsible for their powerful, thus easily-observed actions to humans and test animals. By 1820 when solanine was isolated, pharmaceutical chemistry has dealt with increasing number of natural plant-derived substances as organic medicines or reagents.

The following historical facts have been unknown: Scheele's tartaric acid was introduced officially as a medicinal substance as early as in 1775, benzoic, citric and oxalic acids became official by the end of the 18th century. Morphine was effectively published in 1806 (not 1804), hence the first alkaloid known in isolated state is narcotine (published 1803, official since 1827). Morphine became official in French pharmacy in 1818. And, 1814 is the year when 2 first toxicological accounts on plant-derived acids (oxalic and tartaric) appeared. Practical use in therapy, sometimes soon after discovery, inspired practical pharmacy and stimulated the progress of toxicology.

We studied the earliest 50 years of plant metabolites isolations era. A revised bibliography and a timeline chart for 24 plant substances from this period is provided. Plants from original publications are taxonomically identified.

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1. Introduction

A historian of pharmacy is usually perplexed when one attempts to precisely recall famous facts from the history of discoveries of pharmacologically active plant substances. We are well familiar with some of the highlights as the revolutionary discovery of morphine by Friedrich Sertürner and the date 1804 associated with this event. Even if this date is well known, we have no idea what was the second or the third of alkaloids discovered and isolated. Finding proper relevant citations and source publications is astoundingly hard. It is also surprising that Sertürner had a noble and great predecessor who influenced the pharmaceutical chemistry. This was Carl Scheele: some plant metabolites he had isolated, managed to become official substances in pharmacy yet a long time before morphine. Details on the history of first isolations of plant metabolites for pharmaceutical practice are reminded in relatively few (and usually old) publications. Coming across original literature reveals data overlooked or misquoted today. All these facts will be presented in this paper.

To study the timeline of chemical or pharmaceutical or toxicological knowledge we shall assume that a date of *effective publication* is the only valid method to resolve the priority of discoveries — rather than a date of actual discovery declared in manuscripts, lab notebooks, memories, letters or diaries. This is a standard approach for setting the priority of discoveries in any other branch of science.

This rule is commonly broken in case of Sertürner's results. In a common sense, he discovered morphine in 1804. But there is no full publication about it until 1806, and by 1805 only preliminary observations have been issued. In historiography of pharmacy and chemistry, Carl Scheele's discoveries are not displayed properly and his publications in Swedish are never cited by historians as sources. Some other substances were discovered "gradually" that is in 2–3 steps, e.g. cytisine and narcotine, or published in 2 journals (strychnine, delphinine,

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veratrine), so they have more than one discoverer or more than one valid printed source reference.

2. Aims of the work

The objectives of this study are: 1) to divulgate correct bibliographical entries of publications in which the discoveries of first pure plant metabolites were announced. The time range was set as 1770–1820 which is the oldest and most obscure period in this branch of science; 2) to specify the original plant stock (and the plant species) which became the original natural source of the respective isolated substance; 3) to cite first official pharmacopoeias in which plant metabolites were included: either as official medicinal substances or as official reagents; 4) to find first toxicological monographs of the abovementioned substances.

The knowledge about the actual original plant species which yielded the research material is much more than a beloved and traditional question of old pharmacognosy and toxicology. We shall be aware that in old times, specifying the species of the original plant was a way of standardization in pharmacy [6]. In the turn of the 18th and 19th centuries, a pharmacist and a chemist were hardly able to say more about the herbal stock than the plant name it had been harvested from, and the geographical region it had been shipped from.

Knowing the date a substance became *official* is valued in the history of pharmacy and pharmaceuticals.

3. Materials and methods

3.1. Source publications

Citations of publications and names of substances were queried for (by author name or by Latin or national chemical name) in a number of publications available online. Several secondary sources, such as old monographs (e.g. [4]), pharmaceutical catalogues (as [26]), bibliographies [76], historical studies [23,77] contained also some scattered historical information which we could follow. All found bibliographical entries of original publications on plant metabolites were successfully found by us in digital libraries. Each such an original communication on plant metabolite was then thoroughly read whether it contained requested pharmacognostical information: plant names, names of isolated substances.

The year of discovery of a plant substance was understood by us as the year of effective publication of results in print.

3.2. Plant species

The botanical identities of original plants (which yielded their medicinal stocks for the original researchers) were ascertained on the basis of botanical names in the source publications. These names were provided by the original authors in their national languages (e.g. in Swedish, French, Portuguese or German), as a neoLatin apothecary name or as a Latin botanical binomial. A currently accepted taxonomical name was found by us on the basis of scientific and vernacular synonyms, and is hereby cited in accordance with *The Plant List* database [68].

3.3. The terminology: "official", "pharmacopoeial", and "a monograph"

A drug or a preparation (e.g. a chemical substance or its isolation method) was regarded *official* when it was included in a national or a local pharmacopoeia. In our query, for the earliest "pharmacopoeial" records, neither *universal pharmacopoeias* nor *dispensatories* nor *commentaries on pharmacopoeias* nor *proposals/projects of such* were taken into consideration because works of such kinds have never been normative for any land or state [6]. The same applies to a series of French works entitled *Pharmacopée raisonnée* ('a rational pharmacopoeia') which were intended as theoretical and practical treatises on pharmacy and therapy.

We queried pharmacopoeias for substances described as official ones. Such designation is indicated when a pharmacopoeia contains one of the following: 1) a monograph of a substance, 2) a method of isolation of a substance in a drugstore laboratory, 3) a passage of text dealing with main physical and pharmacological properties recognized for a substance. We understand a *pharmaceutical monograph* as a short, more or less normative text characterizing a substance or methods of its isolation for officinal purposes. Such a text is usually placed in a chapter on *materia medica*. In contrary, when a substance was mentioned in a pharmacopoeia only as a constituent of an official medicinal stock (e.g. a remark that apples contain malic acid), such a record was ignored. Toxicological descriptions of a substance usually contain its physical and chemical characteristics together with the action onto human and animal organisms.

Some special historical sources were treated in a different way according to their authors' intentions. Namely, some books are comments on pharmacopoeias, like Riegel [63] which is not an official book but in which the author clearly states (on page v) that he recorded "simples" (that is simple substances or the *materia medica*) which were available in Prussian apothecaries in his time. Other kind of sources we used were some early medical formularies or pocket-books for physicians. They are testimonies of real usage of a plant substance in therapy (as a simple medicine or in a compound drug) by practitioners in medicine.

Eventually, we paid special attention to Trommsdorff's [69–73] dictionaries because all works by this prominent scientist influenced a lot the early 19th-century pharmacy and seem to have really stimulated further research and clinical applications (for example, Trommsdorff corresponded with Sertürner). It is important that Trommsdorff (1805–1813) highlighted Scheele's authorship of discoveries of plant acids.

Years of discovery (isolation), incorporation to pharmaceutical and toxicological sources were plotted on a timeline chart (Fig. 1) and discussed, especially in relation to medical practical formularies: Fuller [13], Magendie [25], Ebermaier [12], Haden [17,18].

3.4. Full citations

We placed the bibliographical citations for published discoveries in the main text of our paper in order to present them in full, unshortened forms. All these records were retrieved by us from original publications which we had found and read. Data missing in an original publication but compulsory in bibliographies are added in [ ], e.g. a record "Pelletier [P.-J.]" means that Pierre-Joseph Pelletier's given names (or initials) were not printed in the article head. An unknown initial is marked as [n]. A record *Kongl*[*iga*] means that this old Swedish word for 'royal' was abbreviated in the journal title pages as *Kongl*. Original spelling and capitalization of publication titles is retained, e.g. *strychine* vs. *strychnine*; *asparagus* vs. *Asparagus*; *antidysenterica* vs. *anti-dysenterica*; *Henningssche* vs. *Hennings'sche*.

4. Results

4.1. 1770–1800: Carl Wilhelm Scheele's organic acids

Among natural substances isolated by Carl Wilhelm Scheele, the following ones had a plant origin:

4.1.1. Tartaric acid (published 1770)

It was obtained by Carl Wilhelm Scheele from tartarus. Crystals of tartarus grow in wine during its storage (wine in pharmacy is a plant-derived preparation treated as fermented juice of grapes, which are fruits of common grape vine *Vitis vinifera* L.). This Scheele's discovery was kindly summarized by his teacher, Anders Jahan Retzius, in a

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