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Combining classic and novel tools in the study of Historical Collections of Chinese Materia Medica in the Netherlands

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Chapter One

General Introduction

History of Chinese *materia medica* (CMM)

Traditional Chinese medicine (TCM) is an experience-based healthcare system that has been practiced for thousands of years in China. It focuses on aspects of prevention and personalized treatment of human diseases and continues to hold significant relevance in the modern era (Hu, 2015). Chinese *materia medica* (CMM) refers to a significant component of TCM that consists of various natural substances, primarily derived from plants, animals, and minerals, for therapeutic purposes according to TCM principles. Based on Chinese philosophy and clinical experiences, the theories and methods of Chinese *materia medica* uses were gradually formed and recorded in written works (Zhang et al., 2010). The first treatise on Chinese *materia medica* is the *Shen nong ben cao jing* (*The Divine Husbandman's classic of materia medica*). The knowledge of *materia medica* presented in this treatise, which had been transmitted orally from ancient times (c. 6000 BC), was first written down in the Eastern Han Dynasty (25-220 AD). Although the original edition was lost, the contents of *The Divine Husbandman's classic of materia medica* were preserved and recreated as early as the Song dynasty (960-1280 AD) (Yang, 1998). The treatise records 365 types of CMM, which are classified into three categories (top grade, medium grade and low grade) based on their medicinal effects and toxicity (Teng, 2019; Zhao et al., 2018). The *Xin xiu ben cao* (*Newly revised materia medica*), completed in 659 AD by Su Jing et al., depended on the administrative power of the Tang dynasty, and became the earliest officially compiled treatise on *materia medica* in Chinese history. The treatise consists of 844 CMM, among which 144 are newly added (Teng, 2019). The monograph *Hai yao ben cao* (*Materia medica from overseas*), written by Li Xun in the 10th century, delves into the *materia medica* from the southern coastal area of China and from overseas regions. This demonstrates that traditional Chinese medicine has an inclusive and open-minded approach to the medicinal properties of natural substances, even when those substances originate outside of China. The *Ben cao gang mu* (*Compendium of materia medica*), a monumental masterpiece on CMM compiled by Li Shizhen in 1578, identified 1,892 medicinal drugs and listed 11,096 prescriptions. The book also covers discussion on topics such as botany, zoology, mineralogy, physics, astronomy, chemistry, metallurgy, geology, meteorology, etc. Charles Darwin, the British evolutionary biologist, hailed the book as an “ancient Chinese encyclopedia” (UNESCO, 2016). The *Ben cao gang mu shi yi* (*Supplement to Compendium of materia medica*) was written by Zhao Xuemin in 1765. In this work, 921 medicinal products were described, including 716 new additions not previously mentioned in older works. The treatise is compiled especially for the supplements and corrections of errors in the *Compendium of materia medica* (Li Shizhen, 1578). Furthermore, the work extensively documents folk herbal medicines and medicinal products from foreign lands (Teng, 2019). The *Zhong hua ben cao* (*Chinese materia medica*) was completed in 1999 with the efforts of more than 100 experts in China. It contains more than 20 million Chinese characters, records 8,980 types of CMM and adds such information as chemical components, pharmacological agents and clinical reports. This is a comprehensive work, reflecting the development of the CMM in the 20th century (Teng,

2019; Zhonghua Bencao Edit Committee, 1999). The Chinese Pharmacopoeia (ChP), compiled by the Pharmacopoeia Commission of the Ministry of Health of the People's Republic of China, is a legal code with the national legal authority that records drug standards and specifications. The first edition of the Chinese Pharmacopoeia was published in 1953, and since 1985, a new edition has been released every five years. The latest version currently available is the one issued in 2020.

Dynamic changes

Due to its long-standing history and extensive documentation, traditional Chinese medicine (TCM) still plays an important role in the Chinese healthcare system nowadays (Wang et al., 2017), and therefore Chinese *materia medica* (CMM) as well. The total output value of the pharmaceutical industry based on TCM was RMB 786.6 billion (approx. € 102.18 billion) in 2015, accounting for 29% of the total revenues generated by the country's pharmaceutical industry based on synthetic medicine (Xu and Xia, 2019). In 2017, the total fiscal appropriation in China's TCM agencies was c. € 5.98 billion. The number of students in TCM universities specializing in traditional Chinese medicine, was 693,267 in 2017, while the numbers of TCM agencies and practitioners were 54,243 and 217,118 respectively in the same year (Wang et al., 2021). The development of traditional Chinese medicine has received and will continue to receive, strong support from the Chinese government, such as evidenced by the *Outline of the Strategic Plan on the Development of Traditional Chinese Medicine (2016–2030)* and the *13th and 14th Five-Year Traditional Chinese Medicine development plan* (Wang et al., 2021). Furthermore, the popularity of TCM and CMM is also increasing outside of China. For example, the import and export values of Chinese herbal medicine products to the European Union (EU) reached approximately 850 million USD in 2017 (Wang et al., 2022). With such massive support and worldwide demand, traditional Chinese medicine will continue to grow in the near future.

A pivotal aspect attributing to the remarkable success of TCM (and therefore its natural ingredients) is the continual inheritance of valuable historical experiences, accompanied by ongoing growth and changes. The majority of CMM are plant-based products (Leon and Lin, 2017). Through consistent testing and verification by Chinese practitioners and clients, certain plants have demonstrated their ability to cure diseases (Zhong, 2016). Consequently, essential information about species, such as the plant's vernacular name, place of origin, morphological characteristics, and other relevant details has been documented to ensure that future generations use the correct species for their medication (Zhonghua Bencao Edit Committee, 1999). Medicinal plants like *Panax ginseng* C.A.Mey., *Angelica sinensis* (Oliv.) Diels and *Astragalus mongholicus* Bunge have been used for more than 2000 years (Goldstein, 1975) and are still used today (Chinese Pharmacopoeia Commission, 2020). On the other hand, *Panax notoginseng* (Burkill) F.H.Chen, *Codonopsis pilosula* (Franch.) Nannf. and *Stellaria dichotoma* var. *lanceolata* Bunge, 'only' have a few hundred years of medicinal history, and remain in current usage (Xie, 2008). Chinese *materia medica* is not limited to

the consistency of the taxonomic identity of the ingredients over time but also encompasses the persistence of accumulated knowledge. For instance, the entry of ginseng roots (*Panax ginseng*) in the *Divine Husbandman's classic of materia medica* (25-220 AD) comprises only 47 words, while the entry in the *Compendium of materia medica* (Li, 1578) contains nearly 10,000 words and more than 200,000 words in the modern treatise on *Chinese materia medica* (Zhonghua Bencao Edit Committee, 1999).

Based on both the positive and negative impact of certain CMM ingredients, certain changes have taken place in the course of history. An example is the goji berry (with the pharmaceutical name *Lycii Fructus*), which has been used as medicine to nourish the liver and kidney (according to the theory of TCM), and to improve vision since the 2nd-3rd century (Zhonghua Bencao Edit Committee, 1999). The morphological description in the *Ben cao tu jing* (*Illustrated Classics of materia medica*), compiled by Su Song in the 11th century, identified the biological origin of the goji berry as *Lycium chinense* Mill. (Xie, 2008). However, in the 16th century *Compendium of materia medica* (Li Shizhen, 1578), another species (*Lycium barbarum* L.) was also recognized as a source of goji berry. Due to the superior quality of *L. barbarum*, it became the only official species included in the 2000 edition of the Chinese Pharmacopoeia (Chinese Pharmacopoeia Commission, 2000; Xie, 2008; Zhonghua Bencao Edit Committee, 1999).

Another example of historic changes in TCM owing to later discoveries on quality is the root of *Lithospermum erythrorhizon* Siebold & Zucc., with the pharmaceutical name *Arnebiae Radix*, which has been used for over two millennia to clear heat, cool and activate the blood (Zhonghua Bencao Edit Committee, 1999). In recent decades, *Arnebia euchroma* (Royle ex Benth.) I.M.Johnst. and *A. guttata* Bunge have also been utilized for the same purposes. In the Chinese Pharmacopoeia 2000 edition (ChP 2000), all three species were acknowledged as botanical sources of *Arnebiae Radix*. With the release of ChP 2005, *L. erythrorhizon* was subsequently excluded from official sources. This decision was made after the discovery that *A. euchroma* and *A. guttata* exhibit superior quality attributes, rendering them more suitable candidates for anti-microbial applications because of their higher shikonin content (Kumar et al., 2021; Xie, 2008).

Historic change in Chinese *materia medica* can also be the result of negative side effects. Since the earliest records in *The Divine Husbandman's classic of materia medica* nearly 2000 years ago, *Akebia quinata* (Thunb. ex Houtt.) Decne. has consistently been recognized as the authentic species for the pharmaceutical product *Akebiae Caulis*. In modern times, caused by an increased demand for *Akebiae Caulis*, another plant with a similar appearance and higher yield, *Aristolochia manshuriensis* Kom., has been used as a substitute (Xie, 2008; Zhu, 2002). However, carcinogenic aristolochic acids have been discovered in species of the *Aristolochia* genus. The use of *A. manshuriensis* in TCM has raised serious health concerns due to its content of aristolochic acid I and II (Arlt et al., 2002). These compounds are responsible for causing interstitial renal fibrosis which can progress to end-stage renal failure in affected

patients (Lord et al., 1999). In 2003, China's National Medical Products Administration banned *A. manshuriensis*, and the Chinese Pharmacopoeia removed it from the 2005 edition (Kim et al., 2013).

Changes in Chinese *materia medica* have been the result of the substitution of imported ingredients as well. *Draconis Sanguis*, commonly known as dragon's blood, refers to the prepared resin of the fruit of *Daemonorops draco* (Willd.) Blume. This resin has been utilized for nearly 1500 years as CMM. Due to its origin in Southeast Asia, Chinese consumers historically relied exclusively on resin imports. However, since the 1970s, Chinese researchers have actively sought alternatives because importing alone is no longer sufficient to meet consumer demands (Xie, 2008). In this pursuit, the ethanol extract obtained from the resinous wood of *Dracaena cochinchinensis* (Lour.) S.C.Chen emerged as a substitute for *Draconis Sanguis* (Xie, 2008). Until recently, there was an ongoing controversy regarding the potential use of *Dracaena cochinchinensis* as a substitute for *Daemonorops draco* to relieve pain, stop bleeding, and promote wound healing in traditional Chinese medicine (Xie, 2008; Zhang et al., 2019). Therefore, in the most recent edition of the Chinese Pharmacopoeia, *Daemonorops draco* remains the exclusive source of *Draconis Sanguis*, while *Dracaena cochinchinensis* is excluded as an ingredient (Chinese Pharmacopoeia Commission, 2020).

After undergoing extensive practice and verification, any beneficial improvements resulting from these changes in ingredients will eventually be acknowledged as positive changes. They will serve as supplements to the existing CMM, or gradually evolve into independent new drugs. Conversely, if any harmful shifts are identified, they will be classified as negative changes and will be prohibited and eventually eliminated.

Research on the dynamic changes in Chinese *materia medica*

An important reason for studying the dynamic changes of CMM use is to ensure the safety of medications (Zhao et al., 2006). There have been issues with drug safety caused by a change from *Akebia* species to *Aristolochia* species. Unfortunately, until 2006, herbal preparations made with the toxic *Aristolochia* species instead of the *Akebia* species could still be found in the Netherlands (Martena et al., 2007).

Another pivotal reason is to prevent the misdirection of new drug development. It is widely recognized that the 2015 Nobel laureate Dr. Youyou Tu, a Chinese pharmaceutical chemist who discovered artemisinin, drew inspiration from ancient Chinese medicine books, specifically the *Handbook of Prescriptions for Emergencies* by Ge Hong (284-346 AD). This led to the development of the effective antimalarial drug artemisinin, derived from *Artemisia annua* L. (Tu, 2011). The original text in this handbook states: "A handful of *qinghao* (sweet wormwood) immersed with two litres of water, wring out the juice and drink it all" (Ge Hong, 284-346 AD, p. 92). However, the text lacks additional details about the plant's botanical identity. Therefore, prior to initiating the antimalarial substance research, Tu conducted meticulous textual research to confirm the botanical identity of the *qinghao*. This was

necessary due to historical confusion about the plant's Chinese name and five potential *Artemisia* species that could be meant by this name. Subsequent research results proved that the content of artemisinin in *Artemisia annua* L. was significantly higher than the five other species in the same genus (Tu, 2017).

The study of Chinese *materia medica*, as an independent discipline of a vital part of traditional Chinese medicine, has a history of over two thousand years. Investigating its dynamic changes can show a process of refinement, which helps prevent errors from accumulating over the extensive course of development. This endeavor ensures the sustainability of the discipline. However, research on the dynamic changes of Chinese *materia medica* heavily relies on textual research. Given the extensive history of traditional Chinese medicine, many treatises related to CMM from various historical periods have been preserved. Despite certain earlier works, such as *The Divine Husbandman's classic of materia medica*, providing overly simplistic descriptions of herbal medicine and lacking clear botanical characteristics of herbal ingredients, these valuable historical treatises still provide substantial foundations for the studying of the dynamic changes in CMM (Chen and Huang, 2005; Tu, 2017; Xie, 2008). Still, there remains quite some uncertainty on the plant species used in historic recipes documented in TCM herbals, as botanical descriptions are minimal and illustrations are lacking (Xie, 2008).

Historical collections of Chinese *materia medica*, which hold samples of medicinal specimens, offer tangible evidence of the dynamic changes within the field (Brand et al., 2017; Zhao et al., 2015). When compared to textual research on ancient Chinese herbals, these physical specimens present a more intuitive and compelling perspective, as they allow for scientific identification. As shown in Figure 1.1, both textual and specimen research provide substantial support for exploring the dynamic changes in CMM. While each of these methods possesses distinct strengths and weaknesses, integrating these approaches has the potential to enrich our understanding of the ongoing 'evolution' of CMM.

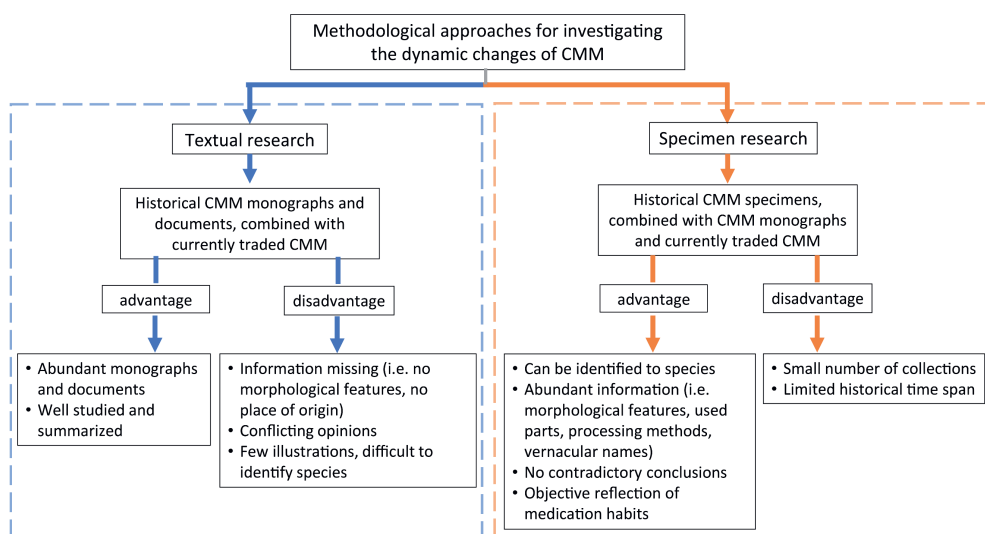


Figure 1.1 Flow chart of the methodology for investigating the dynamic changes of CMM.

Unfortunately, the investigation of the ‘evolution’ of CMM through historical specimens remains limited due to the scarcity of well-documented historical specimen collections, resulting in a considerable gap when compared to the findings derived from textual research.

Historical collections of Chinese *materia medica* in the Netherlands

The current Ph.D. thesis is built on research conducted on historical collections of Chinese *materia medica* in the Netherlands. A considerable number of CMM specimens are housed within the *materia medica* collections of Naturalis Biodiversity Center (Leiden), the Boerhaave Museum (Leiden), the Utrecht University Botanic Gardens, and the Utrecht University Museum. These specimens were transported from Southeast Asia to the Netherlands between the 1850s and the 1970s, facilitated by the trade in commercial plantation crops and other valuable goods, such as medicinal plant material, from Southeast Asia to the Netherlands that lasted for hundreds of years (Cook, 2007; van Andel et al., 2018). Some historic CMM specimens are held by private collectors in the Netherlands. Although these specimens have undergone preliminary identification and their basic classification has been established by either the vendors or the buyers of these collections, these historical specimens have never undergone systematic scientific research and remain inaccessible to the public. Studying these historical specimens of plants, animals and minerals will enable us to gain a deeper understanding on the dynamic changes in CMM over time, from a physical specimen perspective, bridging the gap between specimen research and textual studies, contributing to a comprehensive understanding of CMM in various aspects:

1. Identify the biological origin. The morphological descriptions of medicinal plants in historical CMM treatises are often generalized. For certain herbal medicines, the accurate biological source cannot be determined solely based on the descriptions provided in these treatises (Zhonghua Bencao Edit Committee, 1999). Furthermore, these comprehensive treatises tend to record the official species or widely accepted species of medicinal plants, but region-specific medicinal plants may be missing. By utilizing physical specimens, alternative methods such as microscopic and molecular identification can be employed to ascertain their biological origins if morphological characteristics are not visible to the naked eye (Han et al., 2018; Zhao et al., 2007). Certain CMM specimens may represent regional substitutions, which provide information about geographical variation in traditional Chinese medicine with regard to ingredients.

2. Examine the medicinal part. Although many recipes prescribe the whole plant as medicine, the majority of CMM consists of specific plant parts or related substances, such as resins or distilled extracts. The CMM specimens visually display the medicinal parts of plants. By examining specimens from different periods, it is possible to determine whether there have been any changes in the medicinal parts prescribed in herbal, zoological or mineral preparations.

3. Clarify confusing vernacular names. Historical CMM specimens usually have labels attached, bearing their common names, or the entire collection includes a catalogue containing the names of each medicine. The physical specimens, combined with the vernacular name(s), can help to clarify the confusion caused by multiple medicines sharing the same vernacular name or one medicine referred to by various names. Moreover, certain vernacular names that are region-specific also assist us in determining the area of origin for these historical specimens.

4. Determine the processing method. Some CMM are processed before being used in clinical applications, such as stir-frying. Historical specimens can directly demonstrate the processing methods used for the samples. By comparing specimens from different periods, it is possible to discover the dynamic changes in the processing methods.

5. Identify commonly used medicines from a physical specimen perspective. In general, the majority of CMM treatises are comprehensive, aiming to encompass as much pertinent information as possible (Zhonghua Bencao Edit Committee, 1999). Specimens in historical collections tend to consist of commonly used and easily obtainable items. By comparing specimens from different periods, it becomes possible to identify medicines that were commonly employed during specific timeframes, thus gaining insight into the dynamic changes of CMM.

Physical specimens represented in historical Chinese *materia medica* collections not only enhance understanding of its ‘evolution’ but also, owing to their time-honoured characteristics, offer valuable insights into other research fields. Delayed luminescence (DL),

for instance, has been developed as a rapid, direct, sample loss-free technique to measure the decaying ultra-weak luminescence exhibited by material after being illuminated by light (Sun et al., 2016a). Recently, some studies of CMM using delayed luminescence have proven successful in detecting variations caused by variations in growth conditions (Sun et al., 2016a), different processing methods (Sun et al., 2018; Sun et al., 2016b), and in the determination of authenticity (Sun et al., 2019). Therefore, delayed luminescence is a promising new method to measure the dynamic changes of CMM. In addition, using historical CMM can verify whether DL is a suitable tool for discriminating CMM storage time and explore the potential of DL for use in CMM quality control.

Research aims and outline of the thesis

The primary aim of this Ph.D. thesis is to explore and research the lesser-known historical Chinese *materia medica* specimens and collections housed in the Netherlands, with a focus on their physical attributes, in order to comprehend the dynamic changes of CMM. This objective is pursued by investigating the specific research questions listed below.

Which historical CMM collections and specimens are currently preserved in the Netherlands, and what are the taxonomic identities of the specimens, the utilized parts (plants, minerals or animals), and the processing methods of the specimens?

1. What differences can be observed between these historical specimens and currently traded Chinese *materia medica* in China and the EU?
2. How were historical CMM utilized by people in the past; for which illnesses or symptoms they were used?
3. How can these new findings be used to understand the dynamic changes in time of Chinese *materia medica*?
4. Can historical CMM offer valuable insights into the application of delayed luminescence technique?

In **Chapter 2**, research was conducted on a historical collection of nearly 400 specimens of Chinese *materia medica*. These specimens were originally gathered in Indonesia and later transported to the Netherlands during the late 19th century. Currently, the collection is preserved at the Utrecht University Museum in the Netherlands. The taxonomic identity, Chinese vernacular names, medicinal plant parts, and processing methods of the specimens were examined and compared with the descriptions in modern treatises. The physical evidence regarding changes in botanical identities, medicinal plant parts, adulterations, misidentifications, and the history of CMM itself is presented. The new findings in this chapter demonstrate that historical specimens can expand knowledge of CMM variations in space and time, while also revealing related information through these physical specimens. Several of the herbal ingredients and product names point towards an origin in southern China.

Chapter 3, a continuation of Chapter 2, focuses on a handwritten catalogue corresponding to this historical CMM collection from the late 19th century. The catalogue comprises over 400 entries, each containing a Chinese name, a phonetic Dutch transcription of the Chinese name, a brief description of the medicine's natural origin, the preparation method, and the medical indication for symptom or illness. The catalogue not only serves as a valuable historical record of CMM, contributing to our understanding of its dynamic changes but also reflects an individual physician's interpretation of Chinese medicine in a specific time and space. It gives us new insight into social and cultural aspects related to traditional Chinese medicine, but also to misinterpretations of TCM concepts in the Dutch translation of the recipes.

In **Chapter 4**, five sets of CMM collections from various periods have been selected as subjects for investigating the dynamic changes of CMM from the perspective of physical specimens. The samples encompass four historical CMM collections (one of which is preserved in the Utrecht University Museum as previously mentioned; another is a private collection in Leiden, and two are housed in the UK, as described in previously published research). The fifth collection represents contemporary marketed CMM in Europe. Through the analysis of these CMM collections, which span over 300 years and comprise more than 1,700 specimens, Fabaceae and Asteraceae emerge as the major sources of medicinal plants. Root/rhizomes and fruits/seeds are the most dominant medicinal parts.

Chapter 5 explores the value and contribution of aged CMM specimens beyond the study of dynamic change. In this chapter, the delayed luminescence (DL) technique was applied to test aged CMM specimens and their corresponding modern samples. The objective was to ascertain whether the DL technique can differentiate between CMM samples from different time periods and whether there is potential for the technique to emerge as a novel method for identifying the storage duration and quality control of Chinese *materia medica*.

In **Chapter 6**, the answers to the overall research questions are provided, followed by a discussion of how these new findings address the current gap in science regarding CMM dynamic change. Furthermore, the limitations of these studies are discussed, accompanied by suggestions for future research.

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