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RECENT ADVANCES IN *GILE MAKHTUM* (SEALED EARTH); AN ANTIDOTE IN UNANI MEDICINE A MINI REVIEW

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Review Paper

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ABSTRACT

The medical use of clays/earths and other minerals may be traced to prehistoric times. The use of ochre for a wide variety of ailments, including for intestinal problems as well as for various eye ailments was recorded in Ebers Papyrus. *Gile Makhtum* is an earth brought from a hilly area located at Milos island of Greece. Makhtum is an Arabic word which means stamped or sealed. Due to the soft nature of *Gile Makhtum* it can be stamped easily. In earlier times *Gile Makhtum* was sold in the market in the form of cakes stamped with the seal of the head of Artemis (Greek goddess) for authenticity. That's why this clay was named *Gile Makhtum*. From 13th to 14 th centuries *Gile Makhtum* (terra sigillata) was the most sought after drug due to its efficacy against poisons and a number of diseases including plague during epidemics. Terra sigillata was included in the first edition of the Pharmacopoeia of the Royal College of Physicians of London, in 1618. Its temperament is cold and dry in third degree having *Tiryaq samoom* (antidote of poison), *Daf-e-Humma Wabaiya* (Antipyretic to epidemic fever), *Mufarreh* (Exhilarant), *Muqawwi-e-Qalb* (Cardiotonic), *Muqawwi-e-Meda* (stomachic), *Qabiz* (Astringent), *Habis-ud-Dam* (Styptic), *Mujaffif* (Desiccant), *Mugharri* (Agglutinant) and *Mundamile qurooh* (cicatrizant) actions. The chemical composition of *Gile Makhtum* is Silicates-37.23%, Ferric oxide-4.08%, Aluminum oxide-13.51%, Calcium oxide-22.90%, Magnesia and alkali oxides-1.50%. This clay is supposed to be the first drug used on humans for clinical trials. Wendel Thumblardt, a German prisoner, was the first human volunteer for testing the theriac effect of terra sigillata, against mercury sublimate. He drank a draught of the clay mixed with wine after consuming mercury sublimate and he survived. As per agreement with the authorities he was freed from prison. This review will be helpful to the students, researchers for exploring other actions as mentioned in literature by modern methods.

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INTRODUCTION AND DESCRIPTION

The medical use of clays/earths and other minerals may be traced to prehistoric times. The first recorded use of medicinal clay goes back to ancient Mesopotamia. The uses of particular types of clays and earths are still prevalent worldwide as therapeutic agents in the traditional medicine of different countries. The use of ochre for a wide variety of ailments, including for intestinal problems as well as for various eye ailments was recorded in Ebers Papyrus. [1,2] *Gile Makhtum* (terra sigillata/sealed earth) is one of the clay which is used to prepare various compound formulations in Unani Medicine.

Gile Makhtum is an earth brought from a hilly area located at Milos/Melos island of Greece which is having smooth surface and devoid of grass and rocks called *Buhaira*. On this hilly area there is a temple of goddess Artemis. In this temple the priest is usually a woman. The people living there had a habit of sacrificing goats in respect of goddess. Due to the blood of sacrificed goats, the color of the upper surface of land becomes red. The color of land underneath is usually pale. The lady priest of the temple in the hilly area usually collects this earth in the faith of its healing properties and brings it to the town, stir it in water to form a solution and leave the solution for

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some time to settle down. The supernatant water was discarded and the residual thick, fatty and viscid substance yielded a wax like clay/earth. Then cakes of this earth were prepared. Then she stamped the cakes with the seal of goddess Artemis. This stamped earth was then sent to the kings of Rome and Greece. [3] This clay is also known as *Tin Kaahini* because in ancient times it was discovered by a soothsayer lady (Kaahina). It is also called *Tin Maghra Milonia* as it was considered a variety of red ochre (Geru) because of its red/pink color. The difference between red ochre and *Gile Makhtum* is that *Gile Makhtum* does not stain the hands on touching between fingers. According to Dioscorides, it is collected from the caves located in Island Lemnos. Then it is kneaded with goats' blood and tablets are made. The tablets were then sealed with the figure of a goat. According to Jalinus (Galen) *Gile Makhtum* is of three kinds; one medicinal which only one priest might touch, another is a true Reddle (red ochre) used by carpenters, and third one is white and having cleansing nature and used by those who cleaned linen and woolen clothes. [3, 5-10] *Terra sigillata* is considered an early trademarked drug which was first used on the Greek island of Lemnos in around 500 BCE. It was stamped with a seal of authenticity for trade and marketing purposes. [12]

MATERIAL AND METHODS:

The present review includes the information available in Unani classical literature for its complete description viz. temperament, actions, therapeutic, and dosage uses etc. For History, pharmacological actions and other aspects to prove the importance of *Gile Makhtum* the information available in computerized databases such as Medline, PubMed, Research gate, Google Scholar and Science-direct has been included.

Etymology: Makhtum is an Arabic word which means stamped or sealed. Due to the soft nature of *Gile Makhtum* it can be stamped easily. In earlier times *Gile Makhtum* was sold in the market in the form of cakes stamped with the seal of the head of Artemis (Greek goddess) for authenticity. That's why this clay was named *Gile Makhtum*. [3]. The scientific name of *Gile Makhtum* is Terra sigillata which is a Latin word. The literal meaning of terra sigillata is sealed or stamped earth. It can be defined in two ways; (1) a type of earth or clay used medicinally and originated from the Greek islands. (2) The archaeologist defines it as a type of fine, red earth used in pottery. Roman pottery was made from the 1-3 AD in Gaul (France, Luxembourg, and Belgium, most of Switzerland, and parts of Northern Italy (Lombardy), the Netherlands, and Germany west of the Rhine) by using terra sigillata. [4] As it was obtained from a Greek Island of Lemnos, it was also called Lemnian earth/terra lemnia/limnia GI. [6] Pliny described it as Lemnia rubrica (Lemnian reddle) and also said it *Lemnia sphragis.* Sphragis is a Greek word which means sealed. [11] .

Historical aspect and varieties of *Gile Makhtum* prevalent in ancient times;

Jalinus (Galen) is supposed to have visited the island where this clay was found. An important man of the island, handed over a book written in ancient times about uses of the Lemnian earth. Galen took away 20,000 of these seals with the object to experiment with. [13].

From 13th to 14 th *terra sigillata* was the most sought after drug due to its efficacy against poisons and a number of diseases including plague during epidemics. Many other earths having similar properties as mentioned in books of that period were explored as an alternative to *terra sigillata*. Almost every European country was in search of a substitute for *terra sigillata.* Some remarkable varieties of terra sigillata may be described as under;

Terra Sigillata *Strigoniensis*; The yellow coloured Strigian earth found in Silesia having remarkable properties was made into tablets impressed with a seal representing three mountains. As it was obtained from a pit in a disused gold mine and likely to have gold particles so it was also known as Axungia Solis. Johannes Baptista Montanus (1498–1551 AD) a famous physician of Italy, described the properties of Strigian earth and said that it was initially tried for its theriac power on animals, and afterwards it was given to a man who had consumed a considerable quantity of corrosive sublimate. Montanus says that *terra sigillata Strigoniensis* is produced by rising vapors of the gold, which are assembled drop by drop and coagulated.

Terra Sigillata; a white earth of a chalky nature obtained from Malta. This was considered to be efficacious in cases of snake-bite in particular·

Terra Samia; obtained from the Island of Samos having similar nature.

Terra Sicula; obtained from Sicily.

Terra. Portugallica; obtained from Portugal having the impression of a rose.

Terra Chia and *Terra Cymolia*, both white earths.

Terra Lignicensis; having the impression of an eagle. Due to its excavation from the neighborhood of a silver mine that was supposed to contain a small amount of silver, it was also called Axungia Lunae.

Terra Sigillata of Italian origin known as Sessana, Toccarese, Oreana and Florentina. It was stamped with the arms of the Medici family. They were all popular as antidotes against poison.

Terra Sigillata Hierosolymitana; a white earth excavated in the vicinity of Jerusalem having the impression of a crucifix.

Terra Sigillata originated from England were two varieties red and white. No description of the impression or seal is reported. [13].

Terra Silesiaca; During the Ottoman rule in the 16th century the island of lemnos was under its administration and the extraction and distribution of *terra sigillata* was administered by the authorities in Istanbul. So due to the scarcity of *terra sigillata* in the markets of Europe led to doubts about its purity and authenticity. *Terra sigillata* obtained from Silesia (modern day Poland) known as *Terra Silesia* (or *Silesiaca*) was the best alternative for antidotes to 'poison' and preventives 'against the plague'. [22,23].

Terra sigillata was included in the first edition of the *Pharmacopoeia of the Royal College of Physicians of London*, in 1618. Later in 1833 it was included in the *Pharmacopoeia Universalis of Jourdan* as Terra Sigillata, Terra Lemnia, or Lemnian Bole. Hasluck is said to be the last investigator to visit the site of *terra sigillata* in 1909. He was unable to obtain the specimens of it in the capital of the island, however he bought bowls of clay bearing the old inscription tin Maktum. [13]

Quality description: *Gile Makhtum* having bright red uniform color, smooth surface, sticking on tongue and lips, odor like that of *Soya* (dill), stops bleeding on sprinkling on wounds is considered best in quality. [3,5,7,9].

Figure 1: Gile Makhtum

Vernacular names *Arabic*: Tin Makhtum, Tin Kaahini, Tin Maghra Milonia *English*: Sealed earth/clay, stamped earth, Lemnian earth *Persian*: Gile Makhtum, *Latin*: Terra sigillata, terra Lemnia *Urdu*: *Gile Makhtum* [3, 5-10].

Temperament: Cold and Dry with three degree [3, 5-10].

Actions: As per Unani literature its action are; Mufarreh (*Exhilarant*), Muqawwi-e-Qalb (*Cardiotonic*), Muqawwi-e-Meda (*stomachic*), Qabiz (*Astringent*), Habis-ud-Dam (*Styptic*), Mujaffif (*Desiccant, Siccative*), Daf-e-Humma Wabaiya (*Antipyretic to epidemic fever*), Mugharri (*Agglutinant*), Tiryaq samoom (*antidote of poison*), Dafae taffun (*Antiseptic*), Mundamile qurooh (*cicatrizant / healing agent*). [3, 5-10].

Therapeutic Uses:

As a theriac when *Gile Makhtum* is taken with wine or used as a paint with vinegar, it counteracts all kinds of poisons and animal bites. If taken orally before or after poisoning it facilitates the body to expel the poison through vomiting. According to Galen, the use of a medicine prepared from a combination of juniper berry and *Gile Makhtum* in cases of bites by sea rabbit and stings of Spanish fly was useful to counter the poisonous effect. Such treatment expels the poisons instantly. The oral intake of this combination with wine is also useful in rabid dog bite cases. Local application of *Gile Makhtum* with vinegar followed by an application of the leaves of wild garlic and Qanturyun (*centaurea cyanus* L.) is useful in case of snake bite. Once one year old baby accidentally ate around 9 g of arsenic, the physician gave *Gile Makhtum* by dissolving in mothers milk. Soon after vomiting started and the arsenic came out of the body through vomiting. The process was repeated once again and thereafter vomiting with diarrhea ensued. Then the baby completely recovered from the poisoning effect. Here one has to be remembered that mostly boles/clay have an astringent effect on diarrhea whereas, *Gile Makhtum* acts as a purgative to counter poisonous effect. [3, 5-10].

As a hemostatic agent; Ibn Sina has quoted that, according to Paulos it is a very effective hemostatic agent. Mild roughness is seen in organs affected with hot swellings after its use due to its potent action. In fact it possesses cooling and agglutinant properties. Its use is effective in controlling bleeding from gums, oral cavity and hemoptysis. [3, 5-10].

As a wound healer and anti-inflammatory agent; Dusting of *Gile Makhtum* powder on fresh/old putrefied, bleeding/oozing wounds heals up early. It prevents burning caused by ulceration and heals such ulcers. Local application of its powder mixed with vinegar and cold water is useful in the initial stages of acute swellings. It protects the organs from an injury due to fall. It sets the broken bones, stops the flow of morbid matters towards the hands and feet and protects against degenerative changes. [3, 5-10].

As a cardiotonic agent; it is used with rose water for cardiotonic action. [3, 5-10].

Respiratory ailments; Its oral intake alone or with *Kateera* (gum tragacanth) stops catarrh and is useful in cases of phthisis and hemoptysis as it dries the lung wounds due to its desiccant property.

Intestinal ailments; Oral use of *Gile Makhtum* with *Aabe Bartang* (broadleaf plantain juice) or as an enema, is useful in malignant intestinal abrasions. This enema should be preceded by an enema with honey water and then an enema with salt water.

As an anti-epidemic agent; Its oral intake alone or with *Kateera* (gum tragacanth) is useful in epidemic fever, plaque, bloody or bilious diarrhea, insect bite, air borne epidemic and dirty water borne symptoms. Its intake before or after onset of symptoms is helpful. [3, 5-10].

Potent Action: Tiryaq Sumoom (*Antidote to poisons*).[3, 5,8, 14,15,17].

Dosage: 1-3 g. [3, 5,8, 14,15-17].

Adverse Action: According to Unani literature *Gile Makhtum* can produce adverse effects on lungs and spleen. [3, 5,8, 14,15-17].

Correctives: To counter the adverse effects of *Kateera (gum tragacanth), rose and honey* has been recommended by Scholars [3, 5,8, 14,15-17] .

Substitute: In case of non-availability *Gile Armani* (Armenian bole) may be used as a substitute [3, 5,8, 14,15-17].

Important Formulations: Some important classical Unani formulations having *Gile Makhtum* as an ingredients are as follows: Mufarreh Motadil, Mufarreh Yaqooti Motadil, Majoon Muqawwi Reham, Sufoof Kishneez, Sufoof Ziyabetus[18] Habbe Teewaj, Mufarreh Azam [19] Jawarish Teewaj, Tirya al Tin [20] Majoon Murawweh ul arwah [21].

Chemical composition;
Many researchers have reported the chemical composition of *terra sigillata* in different periods which may be seen as below:

The chemical composition of *Gile Makhtum* (Lemnian Earth) as reported by Thompson as; Silicates-37.23%, Ferric oxide-4.08%, Aluminum oxide-13.51%, Calcium oxide-22.90%, Magnesia and alkali oxides-1.50%, Water and carbon dioxide-17.72%, Moisture as determined by heating at 1300-3.06% in 100 parts. [13] A study of '*Terra Sigillata*' medicinal tablet from a late 17th century Italian medicine chest was conducted for analysis of its chemical composition it was found that it contained three major components; kaolinite (67%), muscovite (16%) and Chlorite Group of silicate minerals (probably clinochlore) (10%). [22].

The six samples of *Terra Sigillata* available in the Pharmacy Museum of the University of Basel were analysed for their chemical composition. Out of six the red sample was found to contain 37.6% kaolinite, 41% illite, 17.7% quartz, and 3.8% hematite. The yellow grey contained 66% montmorillonite, 18.1% illite, 6.9% quartz, 9% albite. [24].

Pharmacological activities:
As a poison antidote; Wendel Thumblardt, a German prisoner is supposed to be the first human volunteer for clinical trial. To avoid his execution, he offered himself for trial of an antidote of poison as claimed by Andreas Berthold who had recently arrived in town with a remedy he called *terra sigillata*, or sealed earth. The condition he put before German authorities was that if survived from poison he would be freed. On the 25th January 1581, Thumblardt was given a fluid dram and a half of "mercury sublimate, mingled with the conserve of roses. The amount of mercury three times needed to kill an average man. He then drank a draught of the clay mixed with wine. The Thumblardt survived from poison and was freed. [22].

Antibacterial activity; antibacterial activity of six samples of *Terra Sigillata* available in the Pharmacy Museum of the University of Basel was carried out. The red sample only was found to be antibacterial against both Gram-positive and Gram-negative bacteria (*P. aeruginosa and S. aureus*). A yellow powder of Libya origin was found having mild antibacterial activity against one pathogen (*S. aureus*) only. [24].

CONCLUSION
The Unani system of medicine offers four kinds of treatment modalities including dieto-therapy,

pharmacotherapy, regimental therapy and surgical interventions. The clays have been in use for medicinal purposes for a long time but they remained largely unexamined in terms of their mechanism of action and potential benefits in medicine. The interest of scientists has been in the geochemical properties of clay minerals with respect to antibacterial and anti-inflammatory action. On the basis of previous research two historic healing clay "terra sigillata Silesiaca" deposits namely Boroszów and Pluznica have been confirmed in Silesia, which may be used in natural medicine. In this review etymology, history, actions, therapeutic uses, chemical composition and pharmacological activities have been explored. This review will be helpful to the researcher for planning further studies to explore antiseptic, cardiotonic, stomachic and antipyretic activity.

Conflict of Interest: none

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