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Study of Extent of Removal of Lead, Copper, and Zinc in the Tap Water Using Tea Leaves

Dissertation Submitted in Partial Fulfillment for the Degree of Bachelor of Science (Health) in Forensic Science

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CERTIFICATE

This is to certify that the dissertation entitled

"Study of the Extent of Removal of Lead, Copper, and Zinc in Tap Water by Using Tea Leaves"

is the bona fide record of research work done by

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ABSTRACT

The extent of removal of copper, lead and zinc in tap water by using tea leaves had been studied. The tap water usually contains some amount of copper, lead and zinc. The concentration of the copper, lead and zinc in the tap water was determined by atomic absorption spectrometer. After that, tea was prepared using the same tap water. Then, the concentration of the metals in the tea was determined by using the atomic absorption spectrometer again. The concentration of the metals in the water before and after preparing the tea was compared. It was expected that the tea leaves will remove some amount of the metals of concern. The results showed that tea leaves in tea-bags could remove 21.2 % of lead and 63.1 % of zinc; medium tea leaves could remove 18.2 % of lead and 75.1 % of zinc; and large tea leaves could remove 16.5 % of lead and 83.1 % of zinc. However, the results have shown that the concentration of copper in the tea has increased after the tea was prepared. The concentration of the copper has increased by 0.041 mg/L, 0.019 mg/L, and 0.013 mg/L in tea prepared using tea-bags, medium-sized tea leaves and large tea leaves, respectively.

INTRODUCTION

Research has been done by the Australian, Chilean and US scientists to find out the extent of removal of heavy metals by the coffee-brewing process and how much of the metal is trapped in the coffee grounds. This research has shown that coffee grounds have a remarkable ability to mop up the heavy metal atoms. Scientists discovered that normal filtered coffee can remove 78 to 90 percent of copper and lead from the water using three different commercial brands of coffee. The researchers also suggested that there is also possibility that tea-bags and tea leaves can remove the heavy metals in the water (David Icke's Medical Archive, 2000). Copper, lead, and zinc are the three common contaminants that can be found in the tap water. If it is possible, then drinking tea will provide a safer and a healthier drink. This could add more to the numerous medicinal benefits of tea. Thus, it is the objective of this study to find out the extent of removal of metals in the tap water by tea leaves.

Copper is a reddish metal which occurs naturally in rock, soil, water, sediment, and air (The Official Internet Site for the Wisconsin Department of Natural Resources, 2003). Copper is widely used in piping and fittings in household plumbing materials (University of Nebraska-Lincoln Extension, 2005). Copper is also used to make electrical wiring, valves, cooking utensils, coins and building materials (World Health Organization, 2004). Copper compounds are used in fungicides, algicides, insecticides, and wood preservatives and in electroplating, azo dye manufactures, engraving, lithography, petroleum refining and pyrotechnics (World Health Organization, 2004).

Most copper compounds found in air and soils are strongly attached to dust or are embedded in minerals. In water, copper is primarily present in complexes or as particulate matter, or dissolved in water and not attached to other particles (World Health Organization, 2004).

The fate of elemental copper in water is complex. Surface oxidation of copper produces copper (I) oxide or hydroxide. In most instances, copper (I) ion is subsequently oxidized to copper (II) ion. Copper (II) ion is the more common oxidation state. However, copper (I) ammonium and copper (I) chloride complexes are stable in aqueous solution. Copper (II) ion will form complexes with hydroxide and carbonate ions. The formation of insoluble malachite $[Cu_2(OH)_2CO_3]$ is a major factor in controlling the level of copper (II) ion in aqueous solution. Copper (II) ion is the major species in water at pH 6; at pH 6 to 9.3, aqueous $CuCO_3$ is prevalent; and at pH 9.3 to 10.7, the aqueous $[Cu(CO_3)_2]^{2-}$ ion predominates (World Health Organization, 2004).

Copper may occur in the drinking water by corrosion of copper household plumbing. Corrosion of plumbing is far the greatest cause for concern. High levels of copper occur if corrosive water comes in contact with copper plumbing and copper containing fixtures in a water distribution system (Environmental Protection Agency, 2006).

All water is corrosive toward copper to some degree, even water termed non-corrosive or treated water to make it less corrosive. Corrosiveness toward copper is more in acidic water. The corrosive properties of water vary. Factors causing corrosion include acidity (low pH), high temperature, low total dissolved solids contents and high amounts of dissolved oxygen or carbon dioxide (Environmental Protection Agency, 2006).

At levels above 2.5 mg/L, copper imparts an unpleasant, metallic bitter taste in the drinking water. Blue-green stains on porcelain sinks and plumbing fixtures could also be seen which occurs from copper dissolved in tap water. Staining of laundry and sanitary ware occurs at copper concentration above 1 mg/L (World Health Organization, 2004).

Copper also is an essential nutrient for the body. USA and Canada has recently established a recommended dietary allowance (RDA) for adults of 900 µg per day. Dosage for children are 340 µg/day for the first three years, 440 µg/day for ages 4 through 8, 700 µg/day for ages through 9 to 13 and 890 µg/day day for ages through 18. World Health Organization (WHO) estimated that average copper requirements for adults are 12.5 µg/kg of body weight per day and about 50 µg/kg of body weight per day for infants (World Health Organization, 2004).

The acute lethal dose for adults lies between 4 and 400 mg of copper (II) ion per kg of body weight. At lower doses, copper ions can cause symptoms of typical food poisoning such as headache, nausea, vomiting and diarrhea. Individuals ingesting large doses of copper are presented with gastrointestinal bleeding, hematuria, intravascular hemolysis, methemoglobinanemia, hepatocellular toxicity, acute renal failure and oligouria (World Health Organization, 2004).

Lead is one of the contaminants that may be found in tap water. Lead is one of the commonest heavy elements found on earth, accounting for 13 mg/kg of the earth's crust. Lead can be found in natural deposit such as in air, soil, dust, food and water (World Health Organization, 2003).

Lead is commonly used in household plumbing materials and water service lines (Environmental Protection Agency, 2006). Lead is also used in the production of lead acid batteries, solder, alloys, cable sheathing, pigments, rust inhibitors, ammunition, glazes and plastic stabilizers (World Health Organization, 2003).

Lead normally binds to soils when it is released to land, and does not migrate to ground water. In water, it binds to sediments. Lead does not accumulate in fish, but it does in some shellfish, such as mussels (Environmental Protection Agency, 2006).

Lead is primarily present in tap water as a result of corrosion of household plumbing systems such as the pipes, solder, fittings, or service connection to homes containing lead (World Health Organization, 2003). Corrosion of plumbing is by far the greatest cause for concern (Environmental Protection Agency, 2006). PVC pipes also contain lead compounds that can be leached from them and result in high lead concentrations in drinking water (World Health Organization, 2003).

All water is corrosive to metal plumbing materials to some degree (Environmental Protection Agency, 2006). The amount of lead dissolved from the plumbing system depends on several factors such as the presence of chloride and dissolved oxygen, pH or the acidity of water, the temperature of water, water softness, and standing time of the water or how long the water stays in the pipes (World Health Organization, 2003). The most corrosive waters are those of at low pH, high carbon dioxide content, and low mineral salt contents (World Health Organization, 2003).

In 1986, JECFA (Joint FAO/WHO Expert Committee on Food Additives) established a provisional tolerable weekly intake of 25 µg of lead per kg of body weight for infants and children, which is equivalent to 3.5 µg/kg of body weight per day, As infants are considered to be the most sensitive subgroup of the population, this guideline value will also be protective for other age group (World Health Organization, 2003).

Signs of acute intoxication may be seen when the blood lead levels are in the range of 100-120 µg/dL in adults and 80-100 µg/dL in children. The signs of acute intoxication include dullness, restlessness, irritability, poor attention span, headaches, muscle tremor, abdominal cramps, kidney damage, hallucinations, loss of memory, and encephalopathy. Signs of chronic lead toxicity including tiredness, sleeplessness, irritability, headaches, joint pain, and gastrointestinal cramps, may appear in adults at blood levels of 50-80 µg/dL. After one to two years of exposure, muscle weakness, gastrointestinal symptoms, lower scores on psychometric tests, disturbances in mood, and symptoms of peripheral neuropathy were observed in occupationally exposed populations at blood lead levels of 40-60 µg/dL (World Health Organization, 2003).

The short-term effects of lead may include interference with red blood cell chemistry, delays in the normal physical and mental development in babies and young children, slight deficit in attention span, hearing, and learning abilities of children, and slight increases in the blood pressure in some adults. The long-term effects of lead can cause stroke, kidney disease, and cancer (World Health Organization, 2003).

Zinc is another contaminant that may be found in tap water. Zinc is a metal normally found in small amounts in nature (Illinois Department of Public Health, 2006). Most zinc ores found naturally in the environment is in the form of zinc sulfide (World Health Organization, 2003).

Zinc is used in many commercial industries. Zinc is used in the production of corrosion-resistant alloys and brass. Zinc is also used for galvanizing steel, producing alloys, and for serving as an ingredient in rubber, ceramics, and paints. Zinc oxide, for example, used in rubber as a white pigment, is the most widely used zinc compound. Zinc carbamates are used as pesticide (World Health Organization, 2003).

Zinc may be found in the drinking water if the water is stored in metal containers or flow through zinc-coated pipes (Environmental Bureau of Investigation, 2006). In tap water, the zinc concentration can be much higher as a result of the leaching of zinc from piping and fittings (World Health Organization, 2003). The level of dissolved zinc may increase as the acidity of water increase. The most corrosive waters are those of low pH, high carbon dioxide content, and low mineral salts contents (World Health Organization, 2003).

Zinc is an essential nutrient needed by the body for growth, development of bones, metabolism and wound healing. In 1982, JECFA proposed a daily dietary requirement of zinc of 0.3 mg/kg of body weight and a provisional maximum tolerable daily intake of 1.0 mg/kg of body weight. The daily requirement for adult humans is 15-22 mg/day (World Health Organization, 2003).

Acute toxicity of zinc arises from the ingestion of excessive amounts of zinc salts, either accidentally or deliberately as an emetic or dietary supplement. Vomiting usually occurs after the consumption of more than 500 mg of zinc sulfate. Fever, nausea, vomiting, stomach cramps and diarrhea occurred 3 to 12 hours after ingestion. Drinking large amount of zinc for longer periods may cause anemia, nervous system disorders, and damage to the pancreas (World Health Organization, 2003).

Tea is one of the most common beverages consumed by most populations. Indeed, apart from water, tea is one of the most natural beverages available in the market. Tea has been consumed socially and habitually by people since about 3000 BC. There is a possibility that tea-bags and tea leaves can remove the heavy metals in the tap water. This means that drinking tea will provide a safer, healthier drink. This could add more to the numerous medicinal benefits of tea.

Camellia sinensis is the tea plant, the plant species whose leaves and leaf buds are used to produce tea (Wikipedia, 2006).

Kingdom	: Embryophyta (Plants)
Phylum	: Angiosperms
Class	: Eudicotyledons
Order	: Ericales
Family	: Theaceae
Genus	: *Camellia*

Tea plant originates from the high regions such as South-west China, Myanmar, and North-east India. However, today tea is cultivated throughout the world, in tropical and subtropical regions. The producing countries include Southeast Asia, China, India, Sri Lanka, Myanmar, Japan, Seychelles, Tanzania, Kenya, Malawi, and Argentina (Zürich & Frick, 2002).

Tea plant is a small evergreen shrub which grows to about 16 meters tall that is usually trimmed to below two meters when cultivated for its leaves. The tree has a strong taproot. The flowers are yellow-white, 2.5 to 4 cm in diameter with 7 to 8 petals. The leaves are 4 to 15 cm long and 2 to 5 cm broad (Wikipedia, 2006).

Figure 1: Camellia sinensis (L.) Kuntze

Source: http://en.wikipedia.org/wiki/Camellia_sinensis.htm

Primarily, tea is drunk as black tea. Other important teas to the world markets are green tea, and Oolong tea. Recently, organic green tea is manufactured in increasing quantities. Instant tea has also begun to manufacture in increasing quantities (Zürich & Frick, 2002).

The seeds of *Camellia sinensis* can be pressed to yield tea oil and may be used as a sweetish seasoning and cooking oil. Steam distillation of black tea yields an essential oil. This tea extract can be used as a flavor in alcoholic beverages, frozen dairy desserts, candy, baked goods, gelatins, and puddings. Refined tea seed oil is used in the manufacture of sanctuary or signal oil for burning purposes. The oil is a non-drying oil and is not subject to oxidation changes, thus making it very suitable for use in the textile industry; it remains a liquid below -18°C. Tea is also a potential source of food colors such as black, green, orange or yellow (Duke, 1983).

Several varieties of *Camellia sinensis* are used for tea production. *Camellia sinensis var. assamica* is used to produce Assam tea and most Ceylon teas are from this plant. The Assam tea is purely a tropical crop, and reacts sensitively to drought and cold. The Assam tea produces malty and earthy drinks. *Camellia sinensis var. sinensis* is used to produce the Chinese tea. Chinese tea is especially suited to hilly regions and resistant to drought, and can tolerate short periods of frost. *Camellia sinensis var. parvifolia* is used to produce the Cambodian tea (Wikipedia, 2006).

In broad terms, there are three types of tea, each of which is produced by a different processing method. The three types of tea are black tea, green tea, and Oolong tea. Black tea is a fully fermented tea (Zürich & Frick, 2002). The tea leaves are fermented in cool, humid rooms until the entire leaf is darkened (Tourle, 2004).

Green tea is a non-fermented tea. Fermentation is suppressed by deactivating polyphenols. Polyphenols in the fresh leaves are inactivated through heat treatment in pans or with steam. This causes the leaves to retain their olive green color. Only then the product is rolled and dried (Zürich & Frick, 2002). Rolling is the process by which the cellular structure of the picked leaves is slightly broken to release essential oils and promote oxidation (In Pursuit of Tea, 1999).

Oolong tea is a partly fermented tea. The fermentation process is halted at an earlier stage (Zürich & Frick, 2002). The leaves are wilted in sunlight, bruised and allowed to partly ferment until reddening of the leaf edges occurs (Tourle, 2004).

Tea leaves contain many compounds such as polyphenolic compounds, carbohydrates, proteins, lignin, ash, amino acid, lipids, organic acids, chlorophyll as well as caratenoids and volatile substances. Among polyphenols, catechins are the most abundant compounds. The most important catechins are epigallocatechin gallate (EGCG), epigallocatechin (EGC), epicatechin gallate

(ECG) and epicatechin (EC). Other compounds that present in tea are gallocatechin, epigallocatechin digallate, 3-methylepicatechin gallate, catechin gallate, and gallocatechin gallate. Although all tea types have anti-oxidizing properties, the efficacy decreases substantially as the variety of tea becomes darker. This is due to lower contents of anti-oxidizing polyphenols remaining in the leaves (Łuczaj & Skrzydlewska, 2005).

To demonstrate the ability of the tea leaves to remove some of the heavy metals in the tap water, tea was prepared using tap water. Three types of tea leaves were used for this experiment:

- Large tea leaves or Guangdong Shou Mei tea
- Medium-sized tea leaves or Pu-er tea
- Tea leaves in tea-bags or Boh tea which has a fine texture

Shou Mei, or longevity or long life eyebrow, is considered as a lower rank white tea (WhiteTeaGuide.com, 2006). Shou Mei is produced from naturally withered upper leaf and tips. It is mostly grown in the Fujian Province or Guangxi Province in China (Wikipedia, 2006).

Shou Mei tea is a fruity furry white tea that is a chaotic mix of tips and upper leaf. It has a stronger flavor than other white teas, similar to Oolong tea. The tea is slightly darker in color, but it should still have a proportionally green color (Wikipedia, 2006).

Figure 2: Shou Mei tea leaves before preparing the tea

White tea is very different from other types of tea such as green or black tea. The leaves are not steamed, or pan-fired, or fermented and fired. This tea is special because the leaves are plucked from a special varietal tea bush called *Narcissus* or chaica bushes. The leaves then are naturally withered and dried in the sun. If mechanical drying is required, it is a baking process at temperatures less than 40°C. The tea is dried naturally using sunlight or lower temperature to preserve tea polyphenols. The leaves are often picked when the buds are tightly enclosed in new leaves and while still covered with fine silvery hair. These leaves maintain the silky white hairs that denote new growth. Since they undergo little processing, these hairs are often intact in the final product. Many believe that the more downy the leaves, the better quality and more delicate the tea (In Pursuit of Tea, 1999).

Pu-er tea is a fermented tea, named after Pu'er country in Yunnan, China. It is an unusual tea, because unlike other teas which are consumed shortly after production, Pu-er tea can be over 50 years old and is usually aged at least one to four years. Over this time it acquires a very strong and rich in earthy robust flavor (Wikepedia, 2006).

Figure 3: Pu-er tea leaves in the form of toucha that was used in this experiment

Puer tea consists of fine leaf particles that have been processed by double oxidation (Kasper, 1999). The leaves are plucked and converted as green or black tea by sun-drying or pan-drying. Once that is done, a special micro-fermentation agent is added. When the fermentation has been accomplished, the leaves are then either packed, or steamed and pressed into bricks called toucha (small, single serving pellets), or pressed into circular cakes varying diameters. Because of the additional processing step to the leaves, the leaves then changes over time through a natural fermentation process (In Pursuit of Tea, 2000).

The Boh tea or tea-bags consists of fine particle of tea leaves. The tea leaves are grown in Cameron Highlands, Malaysia. The leaves are of Assam varieties and are graded as the broken Orange Pekoe tea. It consists of a blend of generally undistinguished black teas (Boh, 2006). The term Pekoe describes the hairs on young tea leaves. The finest teas are produced from pickings of the first two leaves and a new leaf bud. The larger of the two leaves is called the pekoe leaf, and with time it became orange pekoe. Often the term broken Orange Pekoe is used in grading lower quality blends of loose leaf tea (In Pursuit of Tea, 2001).

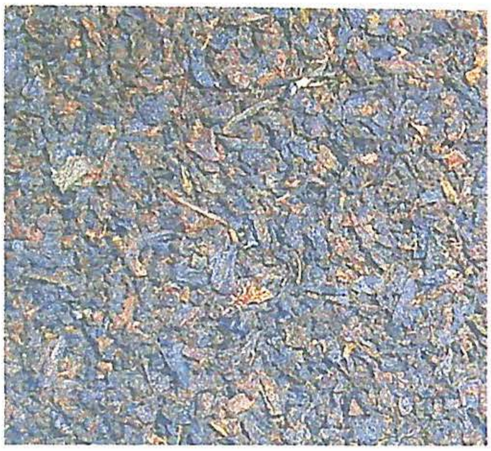

Figure 4: Tea leaves in tea-bag

In Malaysia and Southeast Asia region, Boh Plantations is acknowledged as the pioneer in tea cultivation. Boh Plantation Sendirian Berhad, a pioneer of tea cultivation in Malaysia, was established on 23 April 1929 by John Archibald Russell, the son of a British Government official. Boh Plantation's operations network is mechanized and automated. Application of fertilizers is carried out aerially, and leaf plucking is by hand-held machines. The Boh manufacturing plant is one of the most modern in the world, with computerized quality control at every processing stage (Boh, 2006).

Tea-bags consist of fine particle of tea leaves. These particles of tea is generally manufactured using cut, tear and curl processing, or better known as fannings or dust. The large surface area of the fine particles gives color to the hot water quickly. One of the benefits of the tea-bags is that the infusion of the tea can be controlled through water temperature, quantity of leaves and steeping time to achieve a pleasant cup of tea (In Pursuit of Tea, 2000).

To demonstrate the ability of the tea leaves to remove some of the heavy metals in the tap water, tea was prepared using tap water. The tap water usually contains some amount of copper, lead and zinc. Water samples that were used for this study were taken from the tap water at Desasiswa Murni, Universiti Sains

Malaysia, 16150 Kubang Kerian, Kelantan, and at D'Village Restaurant, a restaurant near the Health Campus.

The first step of this is experiment was to determine the concentration of the copper, lead and zinc in the tap water by atomic absorption spectrometer. The samples of tap water were then pretreated by nitric acid digestion, and metal levels were analyzed using atomic absorption spectrometer.

After determining the concentration of copper, lead and zinc in the tap water, the next step was to prepare tea. The tea was prepared using the same tap water. Three types of tea leaves were used for this experiment:

- Large tea leaves or Guangdong Shou Mei tea
- Medium-sized tea leaves or Pu-er tea
- Tea leaves in tea-bags or Boh tea which has a fine texture

After preparing the tea extract, it was then pretreated with nitric acid digestion. Pretreatment by nitric acid digestion was necessary to remove or to destroy all the organic materials in the sample before analyzing the sample using atomic absorption. Then, the concentration of the metals in the tea was determined by using the atomic absorption spectrometer again. The leaves should be able to remove some heavy metals in the tap water as expected.

REVIEW OF LITERATURE

There are a limited number of studies on the extent of removal of metals in tap water by using tea leaves. Most of the studies were done to discover the most effective technique for waste water treatment using tea waste.

Industrial and municipal waste water frequently contain metal ions. These metal ions can be harmful to aquatic and human health when present in sufficient quantity. Current methods for such waste water treatment include precipitation, coagulation, sedimentation, flotation, filtration, membrane process, electrochemical techniques, ion exchange, biological process, and chemical reaction. Each method has its merits and limitations in application. The adsorption process with activated carbon has attracted many scientists because of the effectiveness for the removal of heavy metal at trace ion levels. However, the process has not been used extensively because it is costly. For that reason, many studies have been done to discover the low cost materials that can be used as sorbent for metal removal from waste water. Among the materials that were investigated are the tea leaves for removal of toxic metal from waste water (Adeyiga *et al*, 1998).

In an experiment done by Tee & Khan (1988), the solid waste of commercially available tea leaves of local produce were found to be good sorbents of metal ions, especially lead(II), cadmium(II), and zinc(II) ions. The extent of the adsorption depends on pH, ionic strength, metal concentration, substrate concentration, and the presence of interfering ions and surfactants. Adsorption capacities of tea leaves were found to be 0.38 mmol/g for lead, 0.28 mmol/g for cadmium, and 0.18 mmol/g for zinc. Surfactants such as cetyl trimethylammonium bromide severely hindered the metal uptake while Triton X100 interfered moderately. It was also found that the anionic surfactant concentration increased while the enhancement effect decreases.

Çay *et al* (2004) had done a study to investigate the adsorption ability of Turkish tea waste, obtained from various tea-processing factories, for removal of copper(II) and cadmium(II) from a single or non-competitive and binary or competitive aqueous system. The tea waste was obtained from tea plants located in black sea region in Rize, Turkey. Prior to the experiments, hydrolysable tannins and other soluble and colored components were removed from the crushed tea wastes so that the tea waste was decolorized. Then, a known quantity of treated tea waste (0.05 g) was added in to 50 mL aqueous solution and stirred continuously. One hour was selected as the optimum contact time for single and binary solutions. Following adsorption, the aqueous phrase was separated from the tea waste by centrifugation. The results showed that industrial tea waste, which has a very low economical value, may be used effectively in the removal of copper(II) and cadmium(II) ions from aqueous system for environmental cleaning purposes. The main characteristics of tea waste produced as a by-product from tea-manufacturing processes all over the world is thought to be very similar to the tea-factory waste investigated here. Çay *et al* (2004), therefore, concluded that the results obtained from this study are likely to be valid for any other tea-factory waste used elsewhere.

Malkoc & Nuhoglu (2005) also had done a research on the ability of tea waste as an adsorption material. The research was done to study the possibility of using tea factory waste as an adsorbent material in nickel adsorption. The tea waste was obtained from tea plants located in black sea region in Giresun-Eynesil, Turkey. Prior to the experiments, other soluble dirt and colored components of the tea wastes were removed. Decolorized and cleaned tea wastes were dried at room temperature for few days. The tea wastes then were added into known nickel(II) solution. The effects of adsorbent dose, initial metal concentration, solution pH, agitating rate and temperature on the adsorption of nickel(II) on tea waste were studied. The following results were obtained:

- The capacity of adsorption of nickel(II) increased with increasing temperatures and pH. The maximum adsorption capacity was obtained at pH 4.0.
- Increase in the mass of adsorbent leads to increase in nickel(II) adsorption owing to corresponding increase in the number of adsorption sites.
- Zeta potential measurement indicated that negatively charge present in tea waste results in the electrostatic interaction between active sites and metal ions.
- From FTIR study, –OH, C–H, C=H and C–O groups present in chemical constituents of tea were the main functional groups which were responsible in metal binding.
- Tea waste which is a factory waste could be used as a potential adsorbent for the removal of nickel(II) ions from aqueous solution and is an inexpensive material for treating the industrial waste water.

In a research done by Murusegan *et al* (2006), pretreated and autoclaved tea fungal mat has been shown to have the ability to remove arsenic(III), arsenic(V), and iron(II) from ground water sample. Tea fungus, a waste produced