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**Determination of heavy metal concentration of
surface water in Nalaikh area and the study of its
reduction methods**

Bachelor Thesis

by

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Statutory Declaration

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List of abbreviation

GMIT = German Mongolian Institute for Resources and Technology

NUM = National University of Mongolia

SGS = Standard Global Services

MNS = Standard of Mongolia

WHO = World Health Organization

EPA = Environmental Protection Agency

DSMA = Disodium methyl arsonate

MSMA = Monosodium methanearsonate

DNA = Deoxyribonucleic acid

MSSP = Moringa stenopetala seed powder

BPP = Banana peel powder

PKS = Palm oil kernel shell

FTIR = Fourier-transform infrared spectroscopy

SEM = Scanning electron microscope

EDX = Energy Dispersive X-Ray Analysis

XRD = X-ray diffraction analysis

ICP-AES = Inductively coupled plasma atomic emission spectrometry

ICP-MS = Inductively coupled plasma mass spectrometry

NA = not available

mg/L = milligrams per liter

Ppm = one part per million

Atm = atmosphere (standard atmosphere of pressure)

g/cc = gram per cubic centimeter

µg/L = micrograms per liter

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Abstract

This study aims to assess heavy metal pollution in surface water in Nalaikh, Mongolia. Sampling locations are taken from Bus Lake, polishing pond in Gorod WWTP, runoff, and shallow lagoon. Bus lake was used for traditional treatment and some types of birds such as gulls etc inhabit and drink. The runoff and shallow lagoon were only taken for observing the content of arsenic and chromium.

The research work continued from November 2021 to April 2022. In this thesis study, the determination of arsenic concentration was examined in laboratories by using spectrophotometric methods and ICP-MS equipment. The first arsenic concentration examination was determined in the laboratory with ICP-MS equipment in November and February (SGS laboratory). Overall of arsenic concentration determined with licensed laboratory is 0.32875 ppm, 0.018ppm, 1.1 ppm and 0.0083 ppm, respectively. Overall of arsenic concentration determined with spectrophotometric method is 0.32 ppm, 0.4095 ppm, NA, and 0.2135ppm, respectively. The most polluted location is Gorod's polishing pond. It is because the wastewater from WWTP adds up to the pond. The second polluted location is Bus lake. Thus, in order to reduce arsenic concentration, the first used method is adsorption and the adsorbent material is spent coffee ground. The reason why using the spent coffee ground for adsorbent is environmentally friendly and is a secondary raw material. Unfortunately, this method is not appropriate for arsenic concentration and also it increases the other content of samples. Thus, the electrocoagulation method was more efficient for arsenic concentration moreover for heavy metals. The test for reducing the content of As (V) by electrocoagulation is successfully carried out in solution. The As (V) value's 99.1% to 99.5% precipitated.

Keywords: Arsenic, Chromium, Water, adsorption, electrocoagulation, pH, time

1. Introduction

1.1. Background

Arsenic is found in nature in 3 allotropic compound forms and it contains more than 200 minerals. And also known as one of the most toxic pollutants in the water environment. If arsenic content exceeds (0.01 mg/L), it happens to irritate human skin, eyes, mouth, throat, and lungs and may cause some kinds of cancer such as lung, skin, liver, kidneys, and prostate cancers. These statements are from the Mongolian Agency for standard metrology MNS 4586: 1998. Furthermore, stomach pain, diarrhea, nausea, and vomiting are very common symptoms of poisoning when someone has acute arsenic poisoning. It is necessary to reduce arsenic content and detoxify the wastewater for processing and ore and mineral mining because of mining wastes from mining rehabilitation and environmental pollution. There are ways to remove arsenic content from the aquatic environment which are coagulation, adsorption, chemical precipitation, electrocoagulation, membrane technology, ion exchange, and biological processing. From those methods, reducing the number of heavy metals using the adsorption method is environmentally friendly and usable because the adsorbent materials can be secondary raw and used materials such as spent coffee ground, sawdust, etc. Unfortunately, these methods are mainly for a small amount of water and it is increasing other content of samples. Thus, scientists' research papers that have focused on the reduction of arsenic content in the aquatic environment by using the electrocoagulation method to remove other contaminants have huge advantages such as the electrocoagulation method's equipment is very easy, low cost, and it continuously creates coagulants by using anode soluble in a reactor. However, there are a few problems that occur in the electrocoagulation process. It is difficult to determine the composition of precipitation, properties, kinetics, and mechanisms of the process. The exact right design of electrocoagulation reactors can avoid those problems.

Chromium is found in soil, cliffs, plants, animals, and even in people. It does not occur in pure form. One of the unique properties of chromium is its ability to present as the very finest molecule. Chromium can be used for steel and other similar metallic products and also restrain corrosion. Chromium has a color compound that is used for making some kinds of dyes and pigments, also tan leather, preserving the wood. The most common usage of chromium is tanning leather in industries. There are two general forms of chromium which are trivalent chromium [Cr(III)] and hexavalent chromium [Cr(VI)] in our daily surroundings. Trivalent chromium appears in vegetables, meats, and food supplies as vitamins and minerals. On the other hand, hexavalent chromium is very toxic. Even

humans can be poisoned with chromium through the air as inhale. Based on studies of workers in chromium processing factories, hexavalent chromium produces carcinogens due to chronic inhalation exposures. It may cause damage to the liver and kidneys and also cancer. The chromium content standard is 0.1 mg/l or 100 ppb for total chromium in the EPA drinking water standard. Total chromium includes all forms of chromium. The few ways to remove chromium content from water are adsorption, chemical precipitation, electrocoagulation, ion exchange, electrodialysis, and membrane separation. But in hexavalent chromium, the separation process is very hard because it completely dissolves into the water. Thus, in chromium, the separation process must be done with electrocoagulation, ion exchange, electrodialysis, etc.

Overall, the diseases caused by heavy metals are very dangerous and acute. Moreover, it can affect soil and other environmental aspects within the locations, if the heavy metal concentration is too high. Thus, this thesis work aims to determine the exact amount of arsenic and chromium content in Nalaikh's surface and water and find out how much arsenic contents occur in those surface waters. In addition, an examination of those contents was done in SGS imme Mongolia LLC and Khanlab LLC Analyzing Minerals Laboratory, and find out which reduction method is suitable for arsenic concentration removal.

1.2. Study aim and objectives

The main aim of this thesis is to determine the accurate arsenic and chromium content near the Nalaikh area, especially around Bus Lake.

The specific objectives of the study are:

Objective 1: Make quantitative analysis

Objective 2: To find out which method is suitable for arsenic concentration

Objective 3: To find out which reduction method is suitable for arsenic concentration

2. State of art




2.1. Properties of arsenic and chromium in nature

Arsenic

Arsenic is the periodic table's 33rd element which is included in the nitrogen group (Group 15 [Va] of the periodic table) and also atomic weight is 75. It has a metallic appearance with a gray form and can be recognized in 3 species: white (As_4O_6), yellow (As_2S_3), and red (As_4S_4). Arsenic has 4 electron shells with 33 protons and 42 neutrons within the nucleus. It is also known as a unique semimetal. Arsenic has a variety of properties in different temperature ranges, for example, It is solid at room temperature, and melts at 36 atm which is equivalent to 814 °C. It can be also used as electrons because arsenic is a common n-type dopant in semiconductors (1).

As for arsenic compounds, yellow (As_2O_3) arsenic trioxide has a low density which is around 3.9g/cc, high volatile, and more metallic. And also it is very brittle, a good conductor of heat, and a poor conductor of electricity. This kind of arsenic is not common in nature. Red(As_4S_4) pararealgar is constantly found with metal sulfides. It is brittle, irregular and uneven shaped, light yellow colored, and has less density than yellow arsenic which is 3.52g/cc. Arsenic trioxide (As_2O_3) is a transparent crystal or white powder that is slightly soluble in water. White (As_4O_6) arsenious oxide is a white amorphous solid that is very soluble in water, forming arsenic acid (1). The allotropic forms in nature are shown in Table 1.

Table 1: Allotropic transformations of arsenic

Gray		<ul style="list-style-type: none"> ● Most common type ● Stable ● Semiconductor ● Brittle
Yellow		<ul style="list-style-type: none"> ● Adult-oriented rock ● Similar to volcanic wax ● Unstable
Black		<ul style="list-style-type: none"> ● Transparent as glass ● Low conductivity ● Brittle

The arsenic species is widespread in nature and currently has hundreds of species of arsenic compounds that have been detected and recorded. Arsenic can appear in soil, and minerals, and also it can be found in air, water, and land through wind, dust, runoff water, and some industrial activities such as ore smelting, etc. Thus, it can occur in an environment with an inorganic or organic form with different compounds. Organic arsenic exists in fish and shellfish. Inorganic arsenic exists in soils, sediments, and groundwater which is a more common type. It is widely used in the production of pesticides, herbicides, and in the manufacture of alloys. Three valence arsenic oxide (As_2O_3) has long been known as the "King Poison". Arsenic's natural compound forms are single valent arsenic (arsenic), trivalent arsenic (arsenide), and pentavalent arsenic (arsenate)(2). Other arsenic compound forms are shown in Table 2(3).

Table 2: Arsenic forms of compound

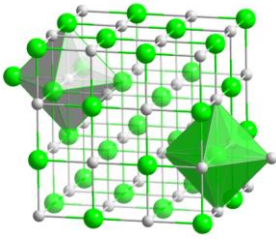
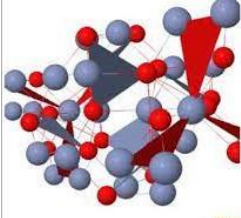
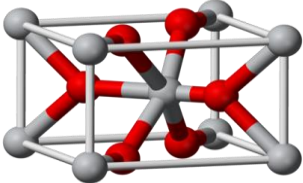
Name	Abbreviation	Chemical formula
Arsenite	As(III)	As(OH) ₃
Arsenate	As(V)	AsO(OH) ₃
Monomethylarsonic acid	MMA(V)	CH ₃ AsO(OH) ₂
Monomethylarsonous acid	MMA(III)	CH ₃ As(OH) ₂
Dimethylarsinic acid	DMA(V)	(CH ₃) ₂ AsO(OH)
Dimethylarsinous acid	DMA(III)	(CH ₃) ₂ As(OH)
Trimethylarsine oxide	TMAO	(CH ₃) ₃ AsO

Chromium

Chromium is the periodic table's 24th element which is the first element in group 6 (VIb) and also atomic weight is 52. It has a silvery metallic appearance with a gray form and is hard as solid. It is a very impermeable element. It is a very bright and glassy transition metal that is very brittle. Chromium(II) oxide (CrO) is an inorganic compound formed from chromium and oxygen.

Cr(II) compounds including oxides and dihalides have enough metallic chromium. Exposing chromium amalgam to air gives black powder CrO oxides that are not characterized. Cr(III) forms the most stable oxidation and its stable trivalent cation is found in a series of substituted inert metallic complexes plus water solution. Giving heat to ammonium dichromate or burning chromium in oxygen gives Cr₂O₃ oxide. The enlargement of chromium(III) to chromium(IV) is through the reaction of forming chromium(IV). These reaction reactivities may cause a reduction of chromium valence by the effect of stoichiometries. Thus, it gives acute results, a form of Cr(IV) in the aquatic environment(4). Chromium ionic structures are shown in Table 3:

Table 3: Chromium structures and characteristics

Chromium oxide		<ul style="list-style-type: none"> • CrO • Melting point is 300 °C • Black • Cubic structure
Chromium(III) oxide		<ul style="list-style-type: none"> • Cr₂O₃ • Melting point is 2,435°C • Insoluble in water • Dark greenish • Fine crystals
Chromium(IV) oxide		<ul style="list-style-type: none"> • CrO₂ • Melting point is 375 °C • Insoluble in water • Black • Ferromagnetic crystals

Chromium has 4 electronic shells with 24 protons and 28 neutrons within the nucleus. As for chemical compounds in chromium, it includes chromium oxide, chromium(VI) oxide, chromium halides, compounds of chromium(III), compounds of chromium(VI), and chromyl chloride (5). Chemical formula of chromium compounds are shown in Table 4:

Table 4: Chemical formula of chromium compounds

Name	Chemical formula	Potassium chromate (Cr(VI))	K ₂ CrO ₄
Chromium	Cr	Sodium chromate (Cr(VI))	Na ₂ CrO ₄
Chromium (III) chloride	CrCl ₃	Potassium dichromate (Cr(VI))	Na ₂ Cr ₂ O ₇
Chromium (III) oxide	Cr ₂ O ₃	Sodium dichromate dihydrate (Cr(VI))	Na ₂ Cr ₂ O ₇ *2H ₂ O

2.2. Heavy metals in water

Arsenic in environmental

Arsenic (As) is transported to land and water by soil and minerals through the air. Volcano explosions are another source of arsenic. Less amount of arsenic content is spread in the atmosphere from power plants and incinerators. Removing arsenic content completely is almost impossible but it can be stabilized by ligand exchange (6). Arsenic which is released from power plants and other combustion processes is not a recent concern. But these air particles are falling to the ground, then adsorbed by soil or absorbed into the soil by rain. Small and fine particles of arsenic can stay a very long time in open surroundings. Some of the common arsenic compounds are soluble in lakes, rivers, and groundwater through rain and snow or through industrial waste. Some of the arsenic accumulates at the bottom of lakes and rivers, some are transported by water. Most arsenic is found in soils or sediments (7). Currently, about 90 percent of arsenic is used to make wood resistant to decay (chromium-plated copper arsenate). In 2003, the United States banned the use of arsenic-containing wood and other products. However, wood factories still used arsenic in their products. Processed wood can be stored and used for a very long time, and it does not affect the structure of furniture made of processed wood. In the past, inorganic arsenic compounds were used as pesticides, mainly in cotton fields and gardens. Arsenic-containing organic compounds such as cacodyl acid, sodium methyl arsenate (DSMA), and monohydrate dimethyl arsenate (MSMA) are used to make pesticides and cotton. Organic arsenic compounds are used as animal feed additives. Small amounts of arsenic are added to other metals to form metal alloys or alloys. The most common use in arsenic alloys is in automotive lead-acid batteries. Another important use of arsenic compounds is in semiconductors and light-emitting diodes.

Main arsenic concentration comes from volcanic eruptions and volcanic ash is primary source. Other use and distribution of arsenic are shown in Table 5 (8):

Table 5: Use and distribution of arsenic

Main distribution	Volcanic eruptions and arsenic ores
Production	Smelting of metals (lead, gold, zinc, cobalt, nickel, etc.), metal alloys and wood production using copper arsenate, leathering
Nature	Volcanic eruptions and arsenic ores, groundwater and wastewater from gold mining
Goods and products	Teak wood, pesticides, herbicides, fungicides, sponges, dyes, pigments, leaded gasoline, etc.
Foods	Alcohol , cigars, seafood (especially shellfish).

Chromium in environmental

Chromium (Cr) has consequential effects on the environment, especially water and soil. Furthermore, some forms of chromium are very toxic to human health, even to fatal danger.

Chromium enters the water through natural processes such as weathering, leaching of soils, industrial operations, etc. And also drinking water can contain a significant amount of chromium which is allowed by the normal standard. And also these contained chromium is good for human health as is the role of some nutrients.

The amount of chromium content on the surface is between 200 and 600 µg/L. Rivers chromium contaminants are around 1 µg/L or more. Thus, the river barely contains chromium in its presence. In drinking water, chromium's possible forms are anionic trivalent $\text{Cr}(\text{OH})_3$ or hexavalent CrO_4^{2-} (9).

Chromium can be found almost everywhere in nature because of natural sources such as the composition of rocks and sediments. Chromium presence in soil is a combination of chromium(III) to chromium(IV). In some processing facilities, solid wastes can produce chromium. Chromium in the soil might remain for a few years. Chromium contamination of soil can differentiate the soil texture and area. In the major range of chromium in soil ranging from 1 to 1000mg/kg, about average value is 14 to 79mg/kg (9).

Humans can poison chromium by different forms of a compound such as oxidation state, etc. Some researchers' studies show that trivalent chromium has little or no toxicity on human health, whereas hexavalent chromium compounds are more toxic to human health by breathing.

Some types of chromium used in metal refining and alloys for example stainless steel contain 12-15 percent of chromium in its characteristic (9).

Around 20,000 tons of chromium are used for those products every year. The primary sources of hexavalent chromium in industrial wastewaters are tanning the leather and making dyes for painting. Chromium compounds are used as pigments, and chromium compounds are used to tan 90% of the leather. Chromium standard levels in wastewater are normally about 5 $\mu\text{g/L}$ (9).

2.3. Effect of Arsenic, Chromium in health and environmental

Arsenic

Under standard circumstances, people get a necessary amount of arsenic from food and drinking water. Fish, seafood, and other related marine environments are contaminated with arsenobetaine and arsenocholine. These substances are less harmful to the human body and easily excreted from the body. From human improper use, As (V) can be absorbed into the soil, then introverted into an organic form from inorganic form (10).

Studies have shown that plants are not resistant to arsenate (As (V)). Thus, if plants are contaminated with As(V), then they stop growing and begin to die (11).

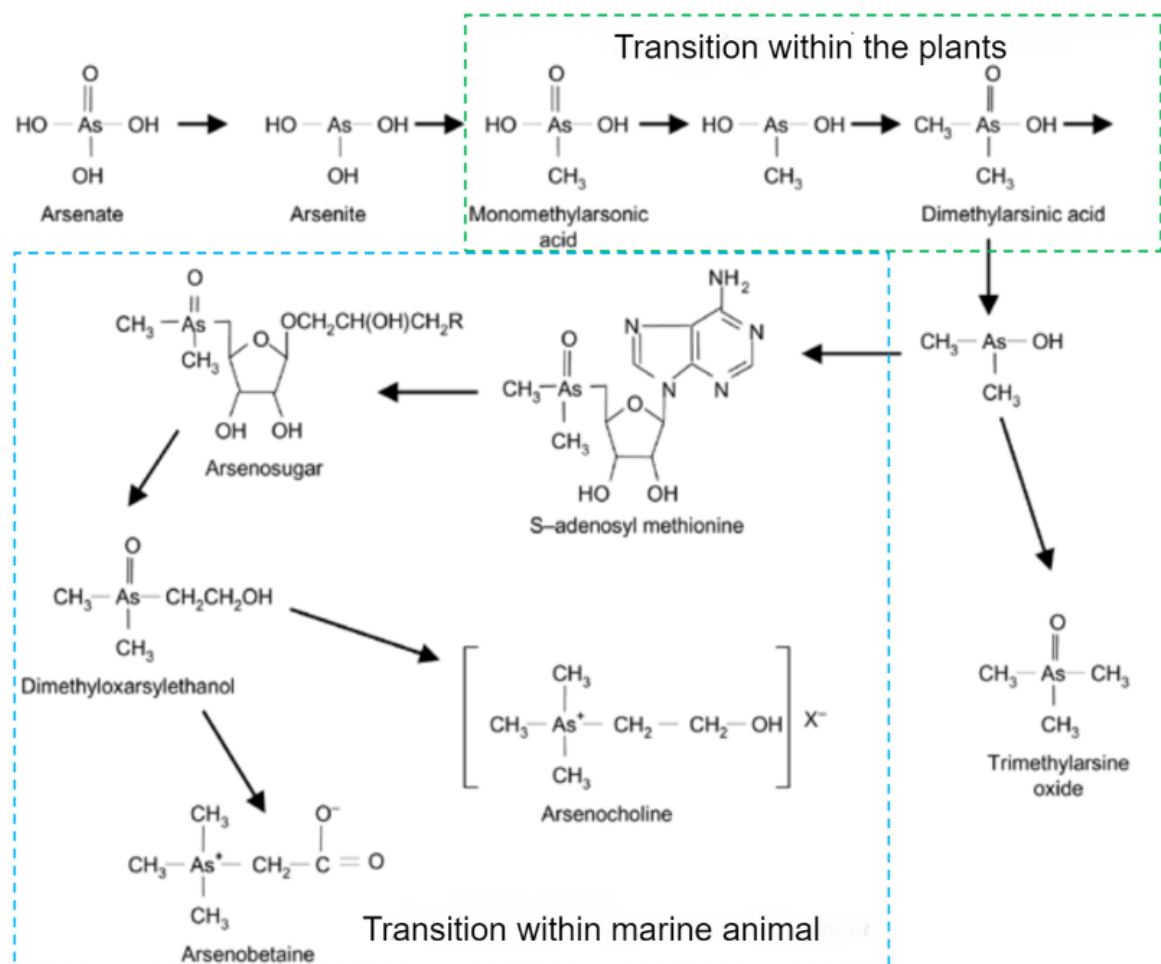


Figure 1: Arsenic transition within the plants and marine animal

About 60-90% of arsenic in the human body is absorbed through food and inhalation accumulates. It is possible to get infected through inhalation, ingestion, and skin contact. The absorbed arsenic spreads into human body parts such as liver, spleen and kidney

within 4 weeks and then eventually leaves the body. But little amount of arsenic stays in the body parts which are skin, hair, nails, bones, and teeth. If the human body is poisoned by arsenic more than WHO standard ($10\mu\text{g/L}$), then it happens to irritate skin, eyes, mouth, throat, and lungs. Furthermore, it causes stomach pain, diarrhea and vomiting as usual symptoms of poisoning. More serious damages are internal bleeding and endocrine injuries. In case of acute arsenic poisoning, then the nasal mucosa is damaged first and the skin color changes which is more green, and inflammation of the lungs. Also brain function damage and order of loss, arrhythmia and fatal injury(8).

Chromium

Our body needs a significant amount of chromium concentration. Additionally, chromium has a small amount of assistance to blood sugar levels. It helps diabetes, bipolar disorder, etc. Thus, some foods include chromium such as meats, grain products, fruits, vegetables, nuts, spices, brewer's yeast, beer, and wine. The reason why these foods include chromium content is depending on what is fertilized and used because soil and water can contain chromium. Depending on the concentration of chromium, the effect on the human body is different. Acute effects include irritation and inhalation problem, and serious effects through inhalation are asthma, chronic bronchitis, chronic irritation, chronic pharyngitis, chronic rhinitis, congestion and hyperemia, polyps in the upper respiratory tract, tracheobronchitis, and ulceration of the mucous nasal membranes with the perforation possibility of the septum. Chromium effects can occur on the skin in the form of irritation, dermatitis, and atopic dermatitis because of allergies. The atopic dermatitis and dermatitis symptoms include dryness, erythema, papules, small vesicles, and swelling. Based on studies of workers in chromium processing factories, hexavalent chromium produces carcinogens due to chronic inhalation exposures. It may cause damage to the liver and kidneys and cancers. It means chromium related to cancer happens in the respiratory system more specifically into lung, nasal and sinus cancers. Furthermore, chromium can damage the brain system and genes. Chromium reactions in the industry make hydroxyl radicals. The hydroxyl radicals cause DNA damage, and mutation and increase cancer possibility. DNA damages include DNA strand breaks, Cr-DNA adducts, DNA-DNA and DNA-protein crosslinks, and oxidative DNA damage. In addition, Hexavalent chromium makes tumors in the human body which causes cancer. Common symptoms of chromium poisonings are dizziness, general weakness, and eye irritation. Further symptoms are kidney, liver, gastrointestinal, cardiac, hematologic or reproduction disorders, growth problems, nasal perforation, and corneal injury (12).

These effects occur when arsenic and chromium levels are higher than MNS 4586:1998. More specifically, if heavy metals concentration is higher than 0.01

mg/L, then the these symptoms gradually observed such as skin irritation occurs when you constantly poisoned by heavy metals. MNS 4586:1998 standard shown in Table 6:

Table 6: Mongolian standards of arsenic

MNS 4586: 1998		
Heavy metals	Unit of measurement	Content
Arsenic	mg/L	0.01
Total chromium	mg/L	0.05
Hexavalent chromium Cr(VI)	mg/L	0.01

2.4. Methods to reduce pollution in the aquatic environment

The quality of fresh natural water is deteriorating due to improper use of water resources. It is necessary to improve water treatment technology to manage water usage of the population. Natural water pollution, especially groundwater and surface water pollution, is one of the biggest problems that affects all living organisms on Earth. Wastewater is essential for recycling and there are many ways to reduce it.

2.4.1. Chemical methods

One of the most commonly used chemical methods is chemical precipitation. It is very important to choose the right precipitator that interacts with contaminants in chemical precipitation. There have been many experiments with chemical processes based on ozone, silver, copper, ferrate, iodine, bromine, hydrogen peroxide, and potassium permanganate. However, the use of chemicals has huge disadvantages of generating chemical water(13).

2.4.2. Physical methods

Physical methods include simple mechanical methods such as settling and filtration. It is not suitable for the removal of As (V) because it is used in the primary treatment of wastewater. Among the most effective physical methods are ultraviolet and ultrasonic disinfection methods that have recently been studied worldwide (13). These methods are effective in killing simple microorganisms but not suitable for the removal of heavy metal contaminants from the environment.

2.4.3. Biological methods

Biological methods of wastewater treatments commonly use microorganisms such as algae and fungal bacteria, and the process can take place under two main conditions. These include aerobic conditions that continuously allow oxygen to pass through the air, and anaerobic conditions in which there is no oxygen. Aerobic methods are widely used in wastewater and surface water, and are effective. However, biological processes are time consuming and require large experimental sites(12).

2.4.4. Electrocoagulation method

The electrocoagulation method is an electrochemical process in which an anode dissolves to coagulant form in solution. Electrocoagulation is a method of effective removal of chemical contaminants from surface and drinking water, especially all types

of contaminants, including heavy metal ions, inorganic compounds, organic contaminants, and pesticides.

In this process, electrochemically dissolved manganese ions spontaneously interact with hydroxide ions (OH) in water to make insoluble metal hydroxides and coagulants. Monomer hydroxide metal species combine to form metal hydroxide $\text{Fe}(\text{OH})_2$, $\text{Fe}(\text{H}_2\text{O})_5\text{OH}_2$, $\text{Fe}(\text{H}_2\text{O})_4(\text{OH})_2$. These have the ability to destabilize substances or absorb contaminants (12). Also many processes can occur simultaneously such as coagulants and chemical contaminants during electrocoagulation, electrostatic interaction, adsorption of polymers, chemical interactions between pollutants and dissolved Fe^{2+} ions (13).

2.4.5. Adsorption method

Adsorption method is a removal process which can occur in solid-liquid, solid-gas, liquid-gas, and liquid-liquid forms. The absorption process takes place when heavy metal molecules or bulky molecules bind themselves to the surface of a solid substance. Nowadays, adsorption methods are commonly used in industrial processes and also it makes more than 99% efficiency. Thus, this type of process is used in wastewater treatment for heavy metal removal.

From a few studies on adsorption methods, highly porous adsorbents are important in the adsorption process and also the most efficient adsorbent is activated carbon. It is giving the best of results but economically unprofitable and resources are limited. Global faces water deficiency which means the wastewater needs to be recycled and reused to reduce the problem. Using secondary raw materials such as used coffee grounds, sawdust, etc can be adsorbent material, absorb most of the heavy metals and low cost adsorbents. Thus, the adsorption method is environmentally safe, cost effective and minimum waste disposal.

2.4.6. Kinetics of sedimentation process

The rate of a chemical reaction is a quantitative measure of the molecular interactions of the reactants. The rate of a chemical reaction depends on the external and internal factors in which the reaction takes place, but the rate of the reaction is affected by parameters such as the concentration of the reactants, ambient temperature, pressure, and catalyst.

2.5. Heavy metal studies

There are many studies that concentrate on the reduction of arsenic concentration in aquatic environments and also use adsorption methods to reduce it.

Maitlo et al. studied electrocoagulation using Fe electrodes to reduce the content of As in groundwater, creating $[\text{Fe}(\text{OH})_3\text{AsO}_4^{3-}]$ through an adsorption mechanism, but did not fully understand the kinetic patterns (14).

PKS studies using bio-absorbent material which is from palm oil mills. Studies focus on these metals: Cr^{6+} , Pb^{2+} , Cd^{2+} and Zn^{2+} and each metal ion using different kinetic models like the pseudo-first order, pseudo-second order and parabolic diffusion models, but not useful in every country (15).

Critical reviews on green adsorbents for wastewaters aim to use low cost material such as fruits, vegetables, foods, agricultural residues and wastes and low cost sources to adsorb the wastewater pollutants. Pollutants are dyes, heavy metals, phenols, pesticides and pharmaceuticals. The reviewed materials are peanut husk charcoal, fly ash and natural zeolite (16).

To determine the arsenic content, this study's spectrophotometer apparatus was used to measure the blue complex colored solution which is prepared with ascorbic acid and ammonium molybdate (17).

Carbonaceous, fibrous solid agricultural waste is groundnut shell. To reduce hexavalent chromium concentration from aqueous solution, this study uses groundnut shells as adsorbent and uses isotherm models (18).

Table 7: Foreign researchers study

Articles	Brief introduction	Advantages and disadvantages	The contribution of this study to my thesis
Metal air fuel cell electrocoagulation techniques for the treatment of arsenic in water	Use electrocoagulation method to reduce As concentration of groundwater	Pros: Kinetic curve [Fe(OH) ₃ AsO ₄ ³⁻ Cons: As(V) is adsorbed on coagulants.	For research (mechanism of electrocoagulation)
Palm Kernel Shell as an effective adsorbent for the treatment of heavy metal contaminated water	Use palm oil kernel shell (PKS) to absorb heavy contamination	Pros: Biosorption mechanisms (using bio-adsorbent) Cons: Long detention time	For research (adsorbents information)
Green Adsorbents for Wastewaters: Critical reviews	Reviewed on different type of material's adsorption	Pros: Low-cost, environmental efficient Cons: Not explaining the procedure	For research (adsorbent materials information)
A rapid colorimetric method for measuring arsenic concentrations in groundwater	Determine arsenic concentration by using spectrophotometer	Pros: Spectrophotometer Cons: Toxic, less precise	The method to determine arsenic concentration in laboratory
Adsorptive removal of chromium(VI) from aqueous solution onto groundnut shell	Using Langmuir isotherm process to adsorb hexavalent chromium	Pros: Quantachrome analyze Cons: too related to time	For research

D.Nomin-Erdene's master's dissertation determined the appropriate conditions for the precipitation process of As (V) using Fe and Al electrodes, analyzed FTIR, SEM, EDX, and XRD in the precipitate, and assumed that FeAsO₄ minerals can be formed. The mechanism of the precipitation process has not been studied(19).

Case study of Enkhmunkh aim to determine water pollution of the Tuul river around the Tannery industry in Ulaanbaatar city. The amount of pollution was determined depending on the location (20).

Table 8: Mongolian researchers study

Articles	Brief introduction	Advantages and disadvantages	The contribution of this study to my thesis
Comparison of electrocoagulation with iron and aluminum electrodes to precipitate As (V) from the aquatic environment	A comparative study of Fe and Al electrodes was performed.	Pros: The analysis of FTIR, SEM and XRD results in the formation of FeAsO ₄ and AlAsO ₄ minerals and chemical interactions. Cons: Not fully studied the mechanism	Using electrocoagulation method to reduce concentration of As(V) from samples
Impact assessment of tannery industry on water quality in tuul river	Identify impact of industry area on environment and water quality	Pros: ARCGIS Cons: Not precise	Including information about tannery industry and environmental impact

3. Methodology

3.1. Research area

This dissertation work is focused on the Nalaikh region, especially near the Gorod area. Nalaikh is a remote district where industrial, mining, and manufacturing developed relatively in the past. Up until the present, the environmental wastes from industrial and mining operations are still harmful to human health and surroundings. Very toxic metal arsenic can be released from a volcanic eruption, inorganic fertilizers, industrial waste, mining industry, landfills, etc.

In 1922, the Nalaikh Coal Mine of State was established and it was the first mining operation in Mongolia. Then the mining operation was officially closed in 1991. However, after 1991 they were unofficially used by “Ninja”-miners up to 2019 (21). Mining activities are directly related to environmental impacts. These impacts are land changes, erosion in the soil, water quality, and air quality. Reasons for the high content of arsenic could be received from legal and illegal ground mining activities. Thus, Nalaikh arsenic concentration is derived from the mining industry and illegal mining activity. Nowadays, the illegal small-scale coal mining activities in the winter period are located between the horse farm and the Mongolian air military airport next to Bus Lake(22). Therefore, examined samples in this thesis work were collected from near those locations (Table 9).

Table 9: Locations of samples

Location	North	West
Bus Lake	107.341006	47.778318
Stream	107.233036	47.462485
Runoff	107.193657	47.465501
Gorodok lake	107.241573	47.470584
Public well	107.160952	47.472943
Household well	107.160497	47.474873



Figure 2: Location of samples on google earth

In 1958, Mongolia's first glass factory was established in Nalaikh. The factory supplied the domestic market with a variety of glassware, including vodka, beer, soft drinks, jams, and medicine. Depending on the processing of raw materials at that time, 42.5% of silica ore, 18.9% of sand, 11.8% of lime and those 73.2% were sourced from National sources, and 19.7% of sodium carbonate, 3.9% of magnesium, 2.4% of nitrate, 0.9% of arsenic and that 26.8% were sourced from import. Nalaikh's glass industry mainly produces green , and a small percentage of colorless and various colored glass. Unfortunately, this only glass industry closed in 1988. Thus, chromium is used as a coloring material to make green glass. In that case, this thesis work also focused on chromium contents near the Nalaikh area (23).

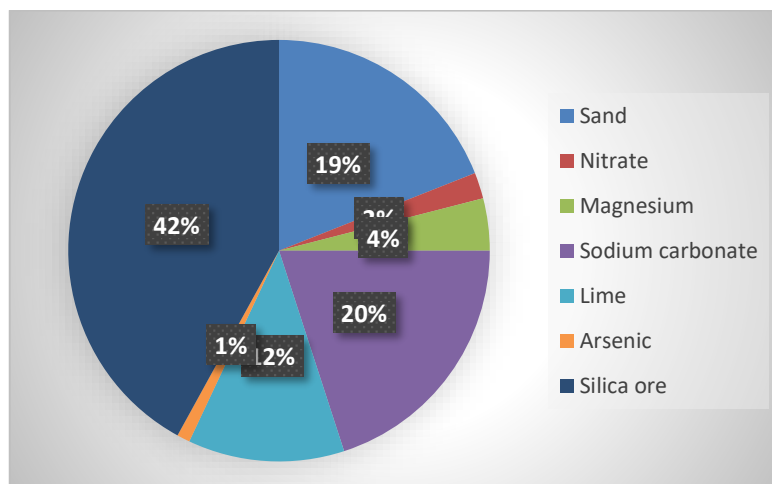


Figure 3: Nalaikh glass industry's contents

3.2. Sampling

One of the most important concepts of this dissertation is sampling. First of all, it's necessary to plan to take the sample: time planning, locations, and equipment (If you take the sample from solid water) (solid form of water shown in Table 9). Then prepare the sampling bottle (PET sample bottle: polyethylene terephthalate) which is rinsed with deionized water to clear contaminants and a permanent marker pen to differentiate the samples. The main procedure for sampling is taking the sample: rinse the PET sample bottles with site water 2-3 times (Must avoid unnecessary contaminants), then take aliquots of approximately 100 ml sample from each location into the sample bottles. Samples must be stored at 6-7°C. Lastly, these samples go to SGS and Khanlab LLC to be examined with ICP-MS and analyzed in the laboratory with a spectrophotometer. Water sampling flow chart is shown in Figure 4:

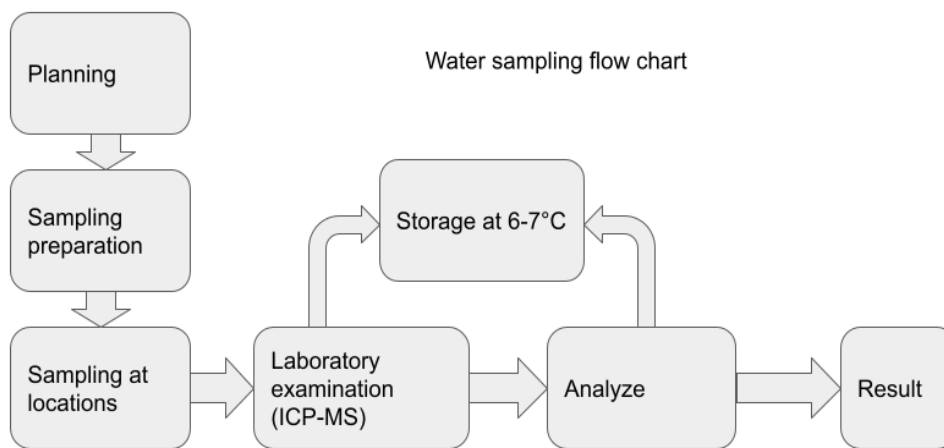


Figure 4: Water sampling flow chart

3.3. Laboratory materials

Information about tools and equipment used in determining the arsenic content.

Table 10: Equipments used in determining the arsenic content

No	Equipments	Accuracy	Mark	Country of origin
1	Spectrophotometer	±0.005	AELAB	China
2	Ovens	150°C	JKI	China
3	Fume hood	NA	LabTech	Indonesia

Information about chemicals and reagents used for arsenic analysis.

Table 11: Reagents used for arsenic analysis

No	Reagents	Formula	Concentrate	Produced company	Country of origin
1	Sodium hydrogen arsenate	$\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$	≥98.0%	Unionlab	China
2	Ascorbic acid	$\text{C}_6\text{H}_6\text{O}_8$	≥99.7%	Xilong Chemical Co.,Ltd	China
3	Hexaammonium heptamolybdate tetrahydrate	$(\text{NH}_4)_6\text{Mo}_7\text{O}_{24} \cdot 4\text{H}_2\text{O}$	81%	Xilong Chemical Co.,Ltd	Russia
4	Sulfuric acid	H_2SO_4	95-97%	Sigma TEK	Russia

3.4. Solution preparation

In laboratory experiments, solution preparation is the most important concept. If the solution's molarity, percentage and concentration is more or less than the necessary amount, the whole experiment may fail. Thus, we need to prepare 0.15M sulfuric acid and 6% ascorbic acid.

To prepare 6% ascorbic acid, dissolve 11 gram of ascorbic acid into 1L distilled water.

To prepare 0.15M sulfuric acid, dissolve 14.7 gram of sulfuric acid solution to 1L distilled water.

The reason why using sulfuric acid is that ammonium molybdate doesn't dissolve in water but in acid. Hence, we add approximately 10 gram of dry ammonium molybdate into 250 mL of 0.15M sulfuric acid to get an ammonium molybdate solution. Then dilute the solution to 1L.

3.5. Laboratory spectrophotometer methodology

The important thing to determine arsenic concentration using a spectrophotometer is the standard curve. The spectrophotometric method for determining the content of As (V) in aqueous solution is based on the reaction of arsenic ions with ammonium molybdate in an acidic environment to form a blue complex (17).

From a solution of Na_2HAsO_4 solution with concentration, put the calculated necessary amount of As(V) concentration into a 25ml volumetric flask. Add 2 ml of ammonium molybdate solution (prepared in 0.15 M H_2SO_4 solution) and add 1 ml of 6% ascorbic acid solution and make up to volume with distilled water. Allow the prepared solutions to stand for 3 hours at room temperature until the color of the complex formed is constant (24). Then the absorption of 850 nm by spectrophotometric. Construct a comparison curve based on the results measured at the wavelength.

3.6. Electrocoagulation method as reduction

Electrocoagulation method to reduce and precipitate As (V) concentration in a water environment using an iron electrode. The instrument diagram is shown in Figure 5a.

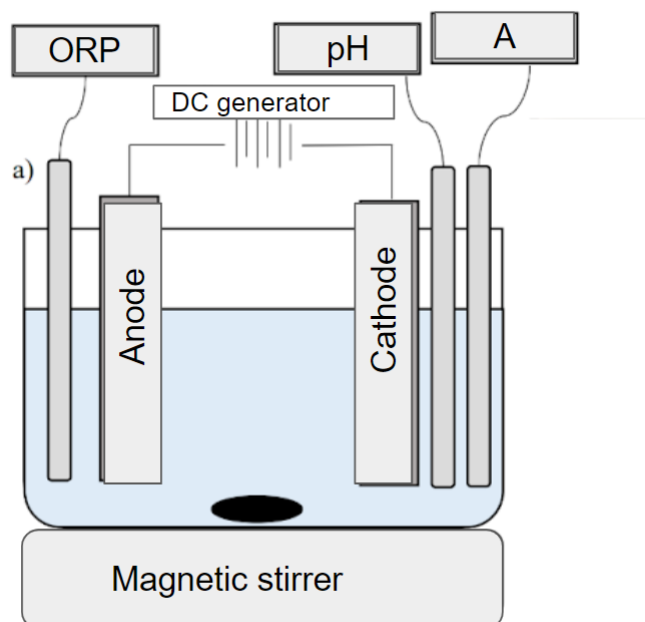


Figure 5: Electrocoagulation method's used instrument scheme

Here is shown how electrocoagulation method done in laboratory in Figure 5b.

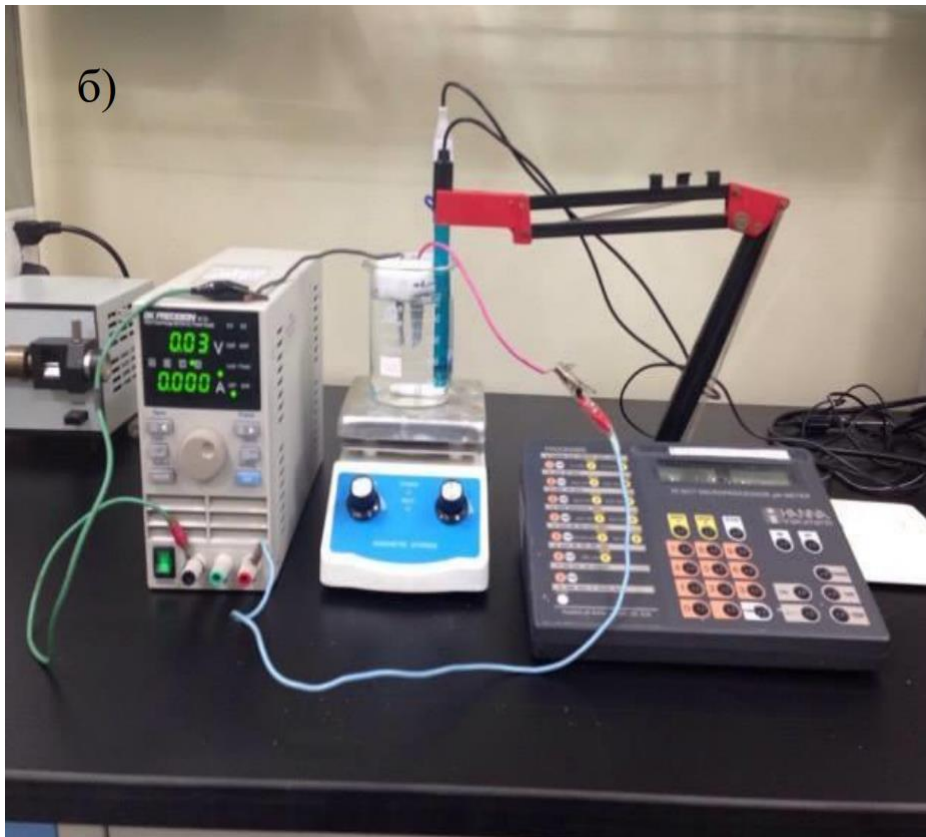


Figure 6: Electrocoagulation experiment in laboratory

3.7. Coffee and sawdust adsorbent

The use of food and secondary raw materials for removal is the very latest environmentally friendly method. Coffee ground and sawdust can be used as adsorbents and these are consumed in tons per year all around the world. The reason why they can be used as adsorbent is they have good filtration and adsorbent characteristics.

Coffee and sawdust for removal of arsenic concentration experiment all performed under the batch

system. First of all, we need 200ml of water sample in the beaker, then add around 15 grams of coffee ground and sawdust into the beaker. Thus the sample into a magnetic stirrer in a 150 rpm rotator for 2 hours. The solution color is slightly yellow and brown after 2 hours. In order to separate adsorbents from the solution, we use filter paper. The adsorbed samples go to the laboratory for examination with AAS.

4. Result and discussion

4.1. Results of sample done in a licensed laboratory

One of the important concerns in sampling is determining water states. The reason is that sample taking is very hard in wintertime. In Mongolia, winter has very harsh weather and temperature, and because of that rivers and lakes are overly deep-frozen. Here are the water states of sampling in Table 12.

Table 12: Water states of the sample's

Date	Bus Lake	Shallow lagoon	Runoff	Gorod's polishing pond
21 st Oct 2021	Liquid	Liquid	Liquid	Liquid
20 th Feb 2022	Solid	Solid	NA	Solid
22 nd May 2022	Solid	Solid	NA	Solid
17 th Apr 2022	Liquid	Liquid	NA	Liquid

October's sample was taken from Bus lake and runoff. February sample's taken from Bus lake, shallow lagoon, and polishing pond. Runoff sample dried in February because it is stagnant water. Results are shown in Table 13:

Table 13: Arsenic concentration results (October and February)

	October			February			
	As, ppm	Cr, ppm	Coffee adsorber for arsenic	As, ppm	Cr, ppm	Coffee adsorber for arsenic	Sawdust adsorber for arsenic
Bus Lake	1.1	<0.1	1.3	0.191	<0.1	0.0115	0.011
Runoff	1.1	<0.1	1.2	NA	NA	NA	NA
Shallow lagoon	NA	<0.1	NA	0.011	<0.1	0.0026	0.0045
Polishing pond	NA	<0.1	NA	0.012	<0.1	NA	NA

October laboratory examination was done in CGL LLC(Central geological Laboratory).

About October's sample, it was stored for about 3 months in the refrigerator. In that case, there occurred huge problematic examination results, even though it gave wrong results in the spent coffee adsorbent. Bus lake and runoff results increased by 0.1 ppm after the use of the spent coffee ground as an adsorbent. It might be because of the solution's condition(pH).

February laboratory examination was done in Khanlab LLC. This result is much more accurate than in October. Used adsorbent this month is coffee ground and additionally sawdust, both adsorbents gave successful results. Bus lake's arsenic concentration decreased by approximately 0.1795 (spent coffee ground), and 0.18 (sawdust). Stream's arsenic concentration decreased around 0.0084 (coffee ground), and 0.0065 (sawdust). After filtering with adsorbent, there exists a very less amount of arsenic concentration in both samples. Bus lake's arsenic concentration is much higher than the stream's. Therefore, the adsorbent work so well and efficiently. On the other hand, the result of chromium content in those 2 months is lower than 0.1 ppm. It is not precise because examined laboratory's given result range is 0.1 to 10000ppm.

March and April samples were taken from Bus lake, shallow lagoon, and polishing pond. May and April's examinations were done in SGS LLC. This two months examination is more precise and accurate than the previous one. These two months examined chemicals are calcium (Ca), potassium (K), magnesium (Mg), sodium (Na), sulfur (S), arsenic (As), and chromium (Cr). There might be a difference in other elements before and after adsorption. Obviously, coffee ground adsorbent changes other elements. Calcium, potassium, magnesium, and sulfur concentration increased after coffee ground adsorption. Moreover, in April, there is no adsorption method result because it is not precise and also it said that spent coffee ground emits methane. This emitted methane was caused by global warming. Results are shown in Table 14:

Table 14: Arsenic and other contaminant results (May and April)

	March						
	Ca, ppm	K, ppm	Mg, ppm	Na, ppm	S, ppm	As, ppm	Cr, ppm
Bus Lake	11.99	3.304	3.76	8.79	9.136	0.004	0.000
Coffee removal for Bus Lake	15.46	244.4	26.35	6.814	12.04	-0.002	0.003
Shallow lagoon	9.94	5.952	2.11	4.83	4.638	0.003	0.001
Coffee removal for lagoon	23.14	210.9	34.45	52.04	61.19	0.002	0.005
Polishing pond	19.51	0.892	4.02	9.13	4.085	0.005	0.004
	April						
	Ca, ppm	K, ppm	Mg, ppm	Na, ppm	S, ppm	As, ppm	Cr, ppm
Bus Lake	14.46	5.24	34.65	87.75	75.93	0.02	0.001
Shallow lagoon	98.02	30.44	19.68	35.96	61.38	0.011	0.000
Gorod polishing pond	23.14	16.41	67.72	281.6	76.65	0.031	0.001

For overall, arsenic concentration of all samples are higher than MNS 4586:1998. Overall data results are shown in Table 15:

Table 15: Overall arsenic results of Bus Lake, shallow lagoon and polishing pond

Arsenic concentration,ppm	Bus lake	Shallow lagoon	Gorodok polishing pond
October	1.1	NA	NA
February	0.191	0.011	NA
March	0.004	0.003	0.005
April	0.02	0.011	0.031

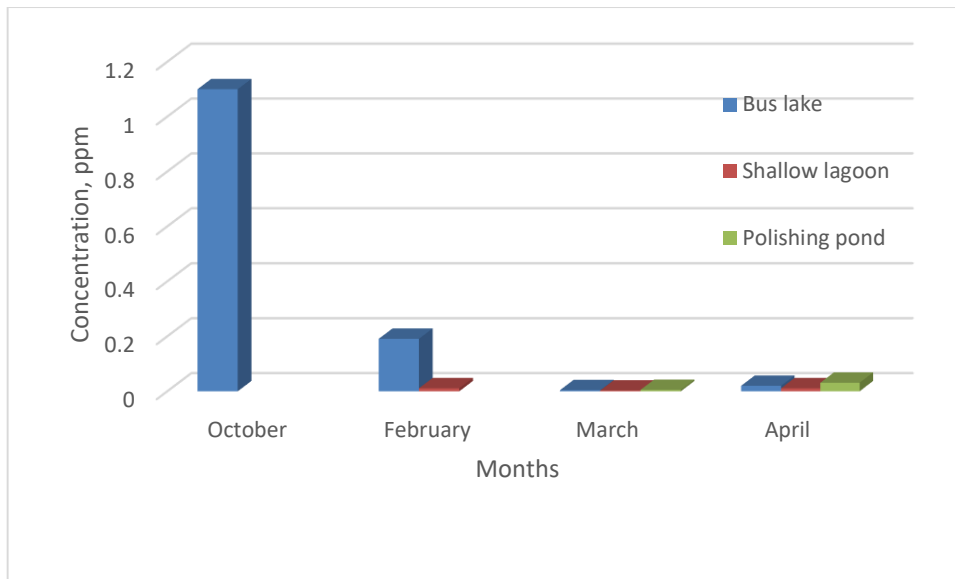


Figure 7: Overall arsenic concentration results

Bus lake's arsenic concentration decreased over the time (October to March), but increased in April. The reason why it might be April's sample is that it is in a fluid state.

The shallow lagoon's arsenic concentration value is very constant and lower because it is stagnant water. Moreover, the results are within the standards.

The polishing pond in Gorod WWTP is increased from March to April. This data shows the difference between seasonal changes.

4.2. Spectrophotometer results

In order to use the spectrophotometer method, there must occur a standard curve. In order to create a standard curve, prepare the concentrated solution. Then measure those solutions' absorbance. Thus, a standard curve is a relation between absorbance and concentration. Standard curve data is shown in Table 16, and the curve is shown in Figure 9.

Table 16: Concentrations of standard curve

C, ppm	A
2	0.0334
4	0.0504
5	0.0584
10	0.0683
15	0.0958
20	0.1241
25	0.1468
30	0.17
35	0.1873
40	0.1998
45	0.2174
50	0.2381
80	0.3645
100	0.4523

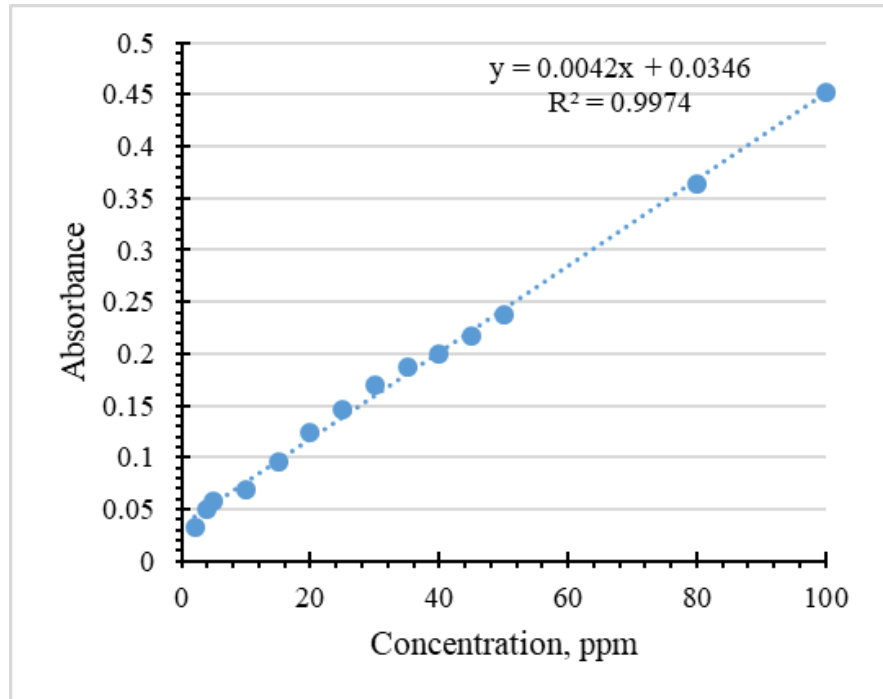


Figure 8: Standard curve of As(V)

The linear relationship of the standard curve to determine the content of As (V) was $A = 0.0042C + 0.0346$, and the regression coefficient was $R^2 = 0.9974$.

In the spectrophotometer method, we can find out the absorbance with wavelength of 850nm. After that, concentration can be found from absorbance's corresponding points. Or it can be found from linear equation. Here is spectrophotometric experiment's result in Table 17.

Table 17: Arsenic's spectrophotometer result

Sample	Wavelength of 540 nm	Arsenic concentration, ppm	Wavelength of 850 nm	Arsenic concentration ,ppm
Bus lake (March)	0.171	32.47	0.244	49.85
Shallow lagoon (March)	0.189	36.76	0.138	24.6
Polishing pond (March)	0.229	46.28	0.381	82.47
Bus Lake (April)	0.469	103.42	0.869	198.67
Shallow lagoon (April)	0.238	48.42	0.530	117.95
Polishing pond (April)	0.590	132.23	1.076	247.95

From table 17, spectrophotometer values are obtained, then we substitute those values into the standard curve's linear equation. These obtained arsenic concentrations are shown in Figure 10.

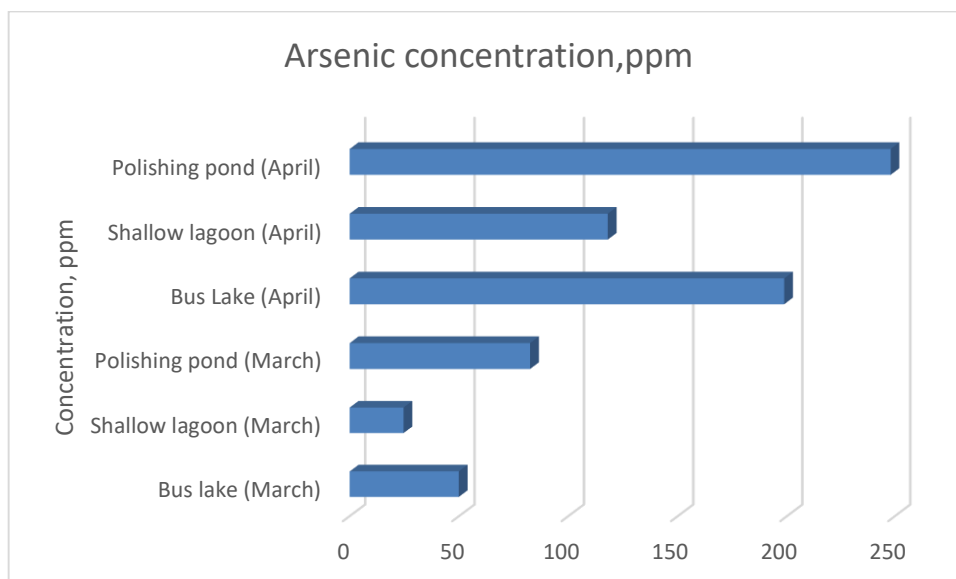


Figure 9: Arsenic concentration ni March and April with spectrophotometric method.

All samples concentration increased from May to April. Apparently, the polishing pond in Gorod WWTP concentration is the highest in those months. Then Bus Lake is the second most polluted location among these locations.

However, the As (V) value can be found using the spectrophotometric method. But these values were not equal on repeated results that were examined in a licensed laboratory with ICP-MS. The reason why those 2 results are different is that the content of phosphate is greater in samples. If phosphate concentration is high in sample's content, then the test results might be obtained wrong. It means samples have a huge amount of bacteria, algae, fungi, and etc. Thus, the spectrophotometric method is suitable for arsenic concentration.

Here is shown arsenic concentration's licensed laboratory results and spectrophotometric method in Table 18.

Table 18: Overall arsenic concentration result with 2 methods

Arsenic concentration,ppm	Bus lake	Shallow lagoon	Gorodok polishing pond
March	0.004	0.003	0.005
April	0.02	0.011	0.031
Spectrophotometric method			
March	49.85	24.6	82.47
April	198.67	117.95	247.95

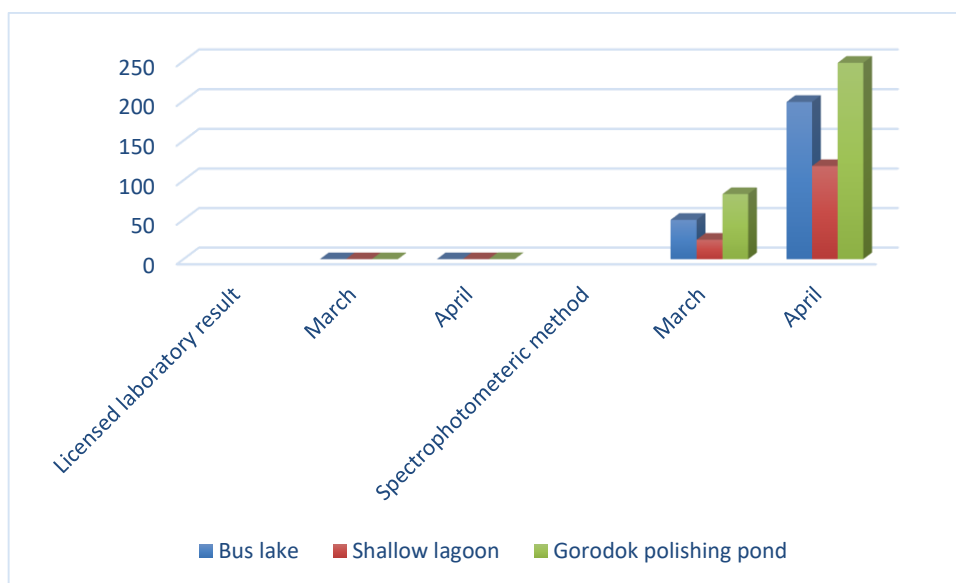
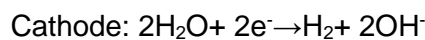
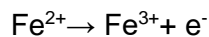
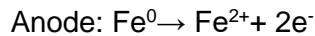


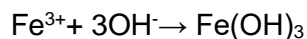
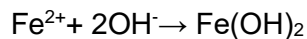
Figure 10: Overall results graph with 2 methods.

4.3. Results of electrocoagulation tests for solutions containing As (V) ions:

As a result of conducting electrocoagulation tests of As (V) ion-free solutions, performing chemical and surface analysis of coagulants, the process at the anode and cathode was determined, and also the reaction equation was summarized. For instance:



Fe^{2+} and Fe^{3+} ions from anode dissolution contain hydroxide ions from cathode reactions that create iron (II) and iron (III) hydroxides ($\text{Fe}(\text{OH})_2$, $\text{Fe}(\text{OH})_3$).



The precipitate of iron oxide (FeOOH) from the formed iron (III) hydroxide and this was similar to the experimental results of Chamteut et al (26).



Iron (II) hydroxide reacts with iron (III) hydroxide and creates iron oxide (magnetite, Fe_3O_4) precipitate.



Therefore, precipitation of iron oxides, FeOOH and iron oxyhydroxide and Fe_3O_4 interact with arsenic in solution. From this reaction precipitation occurs in solution.

The study of the effect of the pH of the initial solution on the electrocoagulation process with iron electrodes will be easier to further control the process and determine the mechanism.

To study the effect of the initial pH of the solution, the environment of the original solution of As (V) was adjusted with a 0.1 M solution of NaOH and HCl using an iron electrode.

The test was performed at all conditions (acidic, neutral and base).

It is shown in Figure 11:

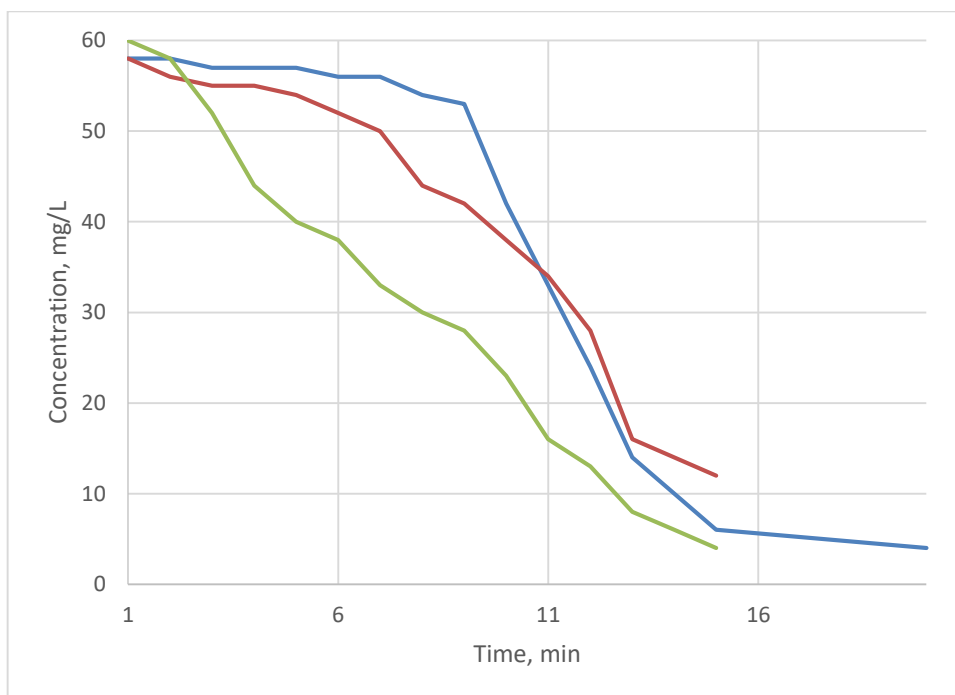
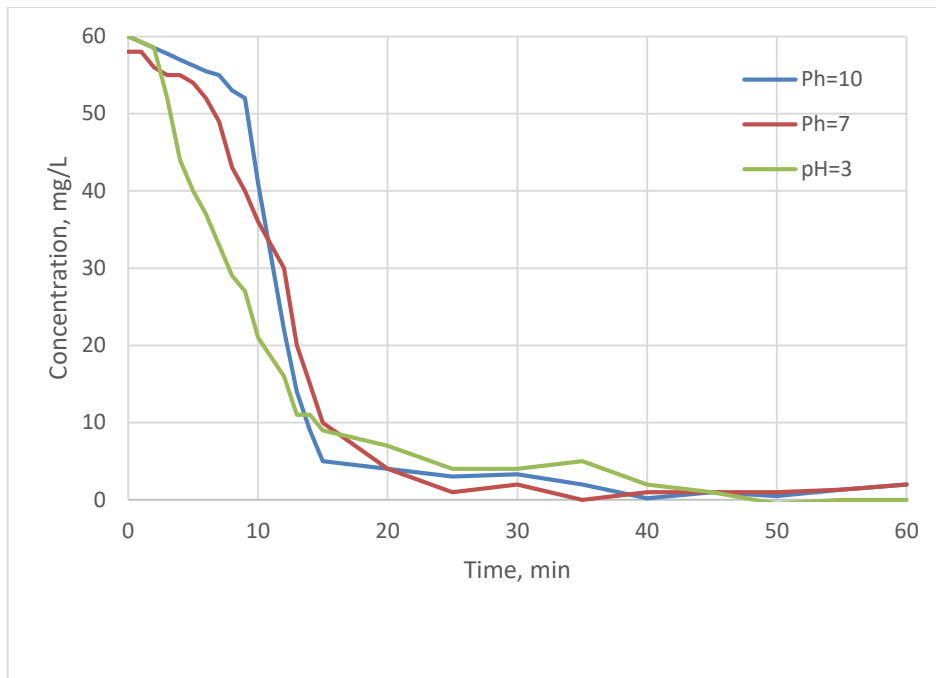


Figure 11: As(V) solution's relation between time and concentration

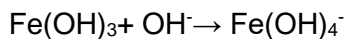
As shown in Figure 11, the kinetic curve of electrocoagulation with iron electrodes depends on the initial pH of the solution. When the initial solution is in base and neutral condition, the concentration of As (V) gradually decreases from 60 mg/L to 54 mg/LI in the first 0-8 minutes.

However, a sharp decrease from 54 mg/L to 5 mg/L was observed between 8 and 15 minutes. When the initial solution was acidic, the concentration of As (V) decreased uniformly from 60 mg/L to 4 mg/L over a period of 0-15 minutes.

In acidic conditions: Due to the high content of H⁺ ions, the formation of OH⁻ ions from the cathode reaction creates negative-charged ions. At this point, the concentration of As (V) decreases because iron (II) ions are formed by dissolving the anode in the solution and interacting with As (V) and it creates precipitation. 6 minutes after the test starts, the solution begins to form a base during this formation process the iron ion interacts with the hydroxide ion to form a large amount of coagulants, and the formed coagulants interact with As (V).

In the neutral conditions: iron ions interact with additional OH⁻ ions which form from the cathode reaction in OH⁻, H⁺ equilibrium solution. This interaction process creates coagulation. The formed coagulants interact with solutions As content which make precipitation.

In base conditions: OH⁻ ions are formed in excess of OH⁻ ions from cathode reactions in solutions. These negative charges are iron hydroxides that are actively attracted to the positive iron ion charges from the anode dissolution. During time, coagulation forms and the reaction takes place under the continuous formation of hydroxide ions.



As the resulting Fe(OH)₄⁻ is repelled by the As (V) ion, so the concentration of As (V) gradually decreases. In order to study the mechanism of precipitation of As (V), the solution's relation between pH and time graph shown in Figure 12:

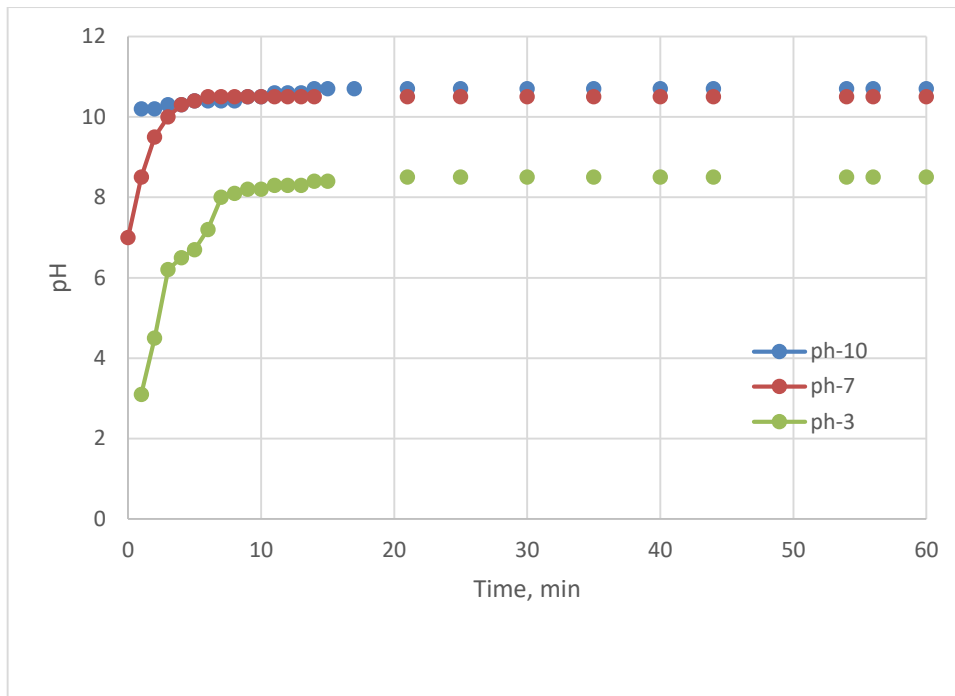


Figure 12: Relation between ph and time

As shown in Figure 12, the pH of the solution increased from 3 to 8.59 in the first 15 minutes in the acidic condition, and in the neutral and base condition, it increased up to 10.99. This is because of the formation of OH^- in cathode reactions. In acidic conditions, the H^+ ion dominates in solution, then cathodic reaction produces OH^- ions, which neutralize the solution for the first 6 minutes. pH level increases from 8.01 to 9.00 between 6 to 20 minutes. During the time, coagulants formed and interacted with the contaminant to stabilize the reaction. Thus, after 20 minutes, the whole solution stabilizes.

In order to confirm the interaction between As (V) and coagulants in the neutral condition, the electrocoagulation test was performed by adding As (V) to the As (V) initial solution after 5 minutes. The result of electrocoagulation of As (V) shown in Figure 13:

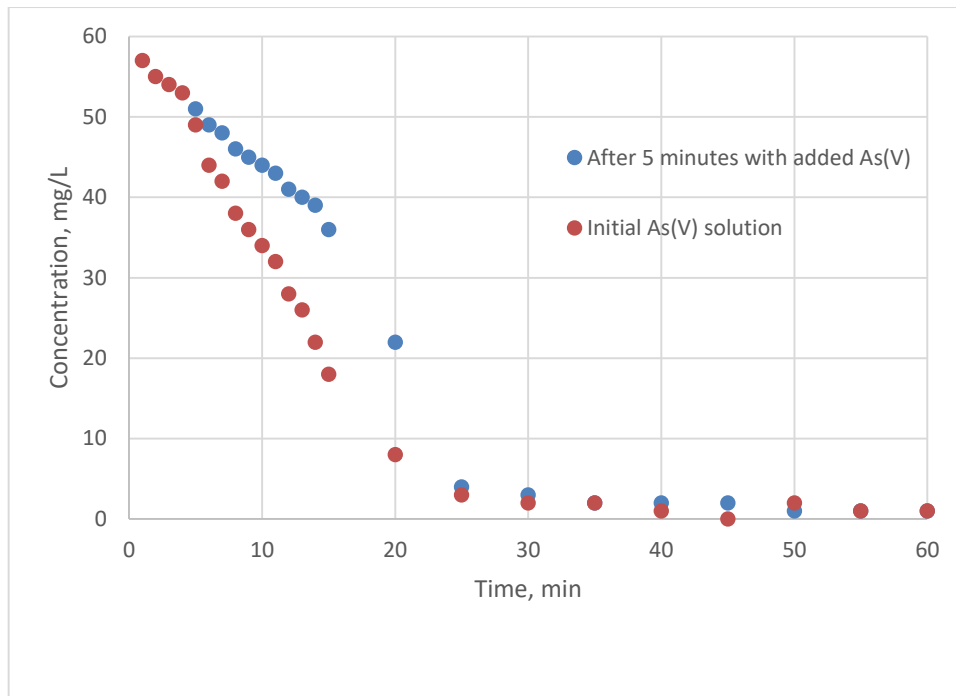


Figure 13: The result of electrocoagulation of As(V) in neutral condition.

As shown in Figure 13, electrocoagulation in the neutral state, the concentration of As (V) gradually decreases from 60 mg/L to 52 mg/L in the first 5 minutes. Then it rapidly decreases from 52 mg/L to 8 mg/L between 5 to 20 minutes. On the other hand, after adding As (V) after 5 minutes, the concentration of As (V) decreases from 50 mg/L to 43 mg/L in the first 11 minutes. Gradually decrease to 52 mg/L from 52 mg/L to 8 mg/L after 5 to 20 minutes. Then again decreases 43 mg/L to 8 mg/L between 12 to 20 minutes. These 2 results gave very similar sequences.

5. Conclusion

Arsenic concentration in Nalaikh's surface water monitoring were continued from November 2021 to April 2022 continued around 6 months

This experiment was successful. There occurs arsenic concentration in those locations sample. Unfortunately, arsenic concentration from those locations results are different in 2 methods. The reason why this differentiation is because of phosphate content in those samples. If phosphate concentration is higher in sample, thus there are too many algae and bacteria occurring. These bacteria and algae is one of the reason the result are wrong. In that case, spectrophotometric method is suitable for arsenic concentration in polluted water.

There was no chromium found in Nalaikh's surface water, as a result, it is not harmful to the human body. The chromium used by the factory that made green glass did not affect the soil and water resource of Nalaikh.

Using secondary raw material (coffee waste and sawdust) as adsorbent was not successful. The content of coffee waste decreased but increased the content of other elements. Moreover, using of coffee waste is not suitable for lake and river, so more advanced technologies such as electrocoagulation is needed.

Electrocoagulation with iron electrodes to precipitate As (V) from the water environment study and the chemical composition and morphology of the sediment determined. The test for reducing the content of As (V) by electrocoagulation is carried out in solution. The As (V) value's 99.1% to 99.5% precipitated.

6. Recommendation

Arsenic concentration in Bus Lake and polishing pond ni Gorod WWTP are too high. Thus, it must be detoxified with the right method. Highly recommend is to find out right reduction method for a huge amount of water such as lake, river, etc. Strongly needed in this pollution is biological and chemical methods which can used in lake.

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8. Appendices

Annex 1: Location: Bus lake's sample area



Annex 2: Location: Polishing pond in Gorod WWTP sample area



Annex 3: Location: Shallow lagoon's sample area

