CONTAMINATION ASSESSMENT AND OPTIMIZATION OF COAGULATION-FLOCCULATION-SEDIMENTATION WATER TREATMENT PROCESS FOR KOU RIVER IN NORTHERN TANZANIA

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A Dissertation Submitted in Partial Fulfillment of the Requirements for the Degree of Master's in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology

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ABSTRACT

The Babati Urban Water Supply and Sanitation Authority (BAWASA) in northern Tanzania supplies water from the Kou River to Magugu town for domestic purpose, where its Fe (3.64 mg/L) and turbidity (87 NTU) concentrations were found in excess beyond their acceptable limits. A coagulation-flocculation-sedimentation water treatment plant was planned to be installed for treating the water, where the system's performance is negatively influenced due to inadequate applications, hence requiring optimization. The quality of sediment and water of the Kou River was examined for physicochemical parameters and 9 metallic elements. The water did not meet the requirements for aquatic, biological life, and irrigation purposes for at least 1 metallic ion. Furthermore, pollution levels of the 9 metals in the river sediments were evaluated using 6 pollution indices and a moderate contamination was found, hardly posing ecological risk. A coagulation-flocculation-sedimentation water treatment system was optimized to respond to coagulant dose, pH, mixing rates, mixing time, and settling time in the removal of Fe and turbidity using jar-test. The optimum dose and conditions were: 30 mg/L alum, an initial mixing rate of 150 rpm for 5 minutes, slow mixing rate of 20 rpm for 30 minutes and settling time for 30 minutes which was validated using scale-up pilot test and resulted in 98.25% turbidity and 97.5% Fe removal. Furthe research on developing enhanced time-saving optimization method and developing a dose indicator technology to assign alum dose requirement based on the fluctuating characteristics of the raw waters (turbidity) is recommended.

DECLARATION

I Netsanet Muluneh Gebreyohannes do hereby declare to the Senate of Nelson Mandela African Institution of Science and Technology that this dissertation is my original work, and it has neither been submitted nor being concurrently submitted for degree award in any other institution.

Netsanet Muluneh Gebreyohannes

Name and Signature of the Candidate Date

The above declaration is confirmed by:

Prof. Revocatus L. Machunda

Name and signature of Supervisor (1) Date

Dr. Mwemezi J. Rwiza

Name and signature of supervisor (2) Date

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CERTIFICATION

The undersigned certify that they have read and found that the dissertation conforms to the standard and format acceptable for submission. Therefore, do hereby recommend for acceptance of dissertation entitled "*Contamination Assessment and Optimization of Coagulation-Flocculation-Sedimentation Water Treatment Process for Kou River in Northern Tanzania*", in Partial Fulfilment of the Requirements for the award of Degree of Master's in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology.

Prof. Revocatus L. Machunda

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DEDICATION

This work is dedicated to my beloved father “Gideon Birhanu” whom our Arusha family lost due to the Covid-19 pandemic. May God rest his soul in heaven!

My parents: My mom Etaferahu Amare, my dad Muluneh Gebreyohannes, all my families and my best friend Mr. Leule Getenet.

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LIST OF ABBREVIATIONS AND SYMBOLS

AAS	Atomic Absorption Spectroscopy
APHA	American Public Health Association
BAWASA	Babati Urban Water Supply and Sanitation Authority
BCR	Basalt Columbia River
BCR	Community Bureau of Reference
BDL	Below Detection Level
BF	Bioavailability Factor
CF	Contamination Factor
DDS	Darakuta Downstream Station
DO	Dissolved Oxygen
DUS	Darakuta Upstream Station
EC	Electric Conductivity
EDTA	Ethylene Diamine Tetra Acetate
ED-XRF	Energy-Dispersive X-Ray Fluorescence
EF	Enrichment Factor
EPA	Environmental Protection Agency
FAO	Food and Agriculture Organization
Hrs.	Hours
IUCEA	Inter University Council for East Africa
mCd	Modified contamination degree
MDL	Machine Detection Limit
N/A	Not Available
NM-AIST	Nelson Mandela African Institution of Science and Technology
NOM	Natural Organic Matter
NQEp	Standards of provisional environmental quality
NTU	Nephelometric Turbidity Unit
OFAT	One Factor at a Time
RAC	Risk Assessment Code
RI	Potential Ecological Risk Index
RPM	Revolutions Per Minute
SDG	Sustainable Development Goal
SEQ-Eau	the French system for the evaluation of the quality of water streams

TBS	Tanzania Bureau of Standards
TDS	Total Dissolved Solids
THM	Trihalomethanes
TMEs	Trace metallic elements
UN	United Nations
USEPA	United States Environmental Protection Agency
USGS	United States Geological Survey
WHO	World Health Organization

CHAPTER ONE

INTRODUCTION

1.1 Background of the Problem

Water is essential for sustaining healthy functioning of an ecosystem, reduction of poverty and socio-economic progress (Funtowicz *et al.,* 1998). One of the sustainable development goals (SDGs) set by the United Nations in 2015 was to ensure availability and sustainable management of water and sanitation for all by 2030 (Sørup *et al.,* 2020). However, the global status of water quality is negatively affected because of natural and anthropogenic factors on water sources (Khatri *et al.,* 2017).

The major notable factors which impair the quality of water are sourced from geologic formations and rapid population growth that increase demands for industrial, agricultural, and domestic applications. Urbanization, intensive agriculture, recreation and the manufacturing industry have been reported as key determinants of water quality degradation through the introduction of different pollutants, including trace metallic elements (TMEs) (Sørup *et al.,* 2020). Copper, iron, manganese, chromium, selenium, and zinc are some of the TMEs which bioaccumulate in aquatic ecosystems, body tissues and organs by entering the food chain from polluted sediment and water. Although some of these TMEs are essential for human and ecological health at limited levels, excess intake may result in acute and chronic health problems (Jaishankar *et al.,* 2014).

Assessing pollution levels of water and sediments is important for understanding the purpose-specific suitability of water bodies and maintaining a healthy environment. For this reason, various standard guidelines and tools have been developed to assess the quality of water and sediments in the aquatic ecosystems (Elamassi, 2012). Several contamination indices have also been made available to assess the pollution level of TMEs in sediments and to indicate anthropogenic influences (Caeiro *et al.,* 2005). Moreover, various studies have provided extraction methods to quantify TMEs' mobility and bioavailability, which are core factors that determine their potential risks on living organism (Alhadrami *et al.,* 2016).

There are several technologies available globally subdivided under 3 major classes (Conventional strategies, biological strategies and membrane technology-based strategies) that are capable of overcoming the varying stresses on water. These include coagulation-flocculation-sedimentation process, trickling filter, ultrafiltration, microfiltration, reverse osmosis, filter media separation, adsorption, electrocoagulation/floatation, and oxidation/precipitation (Khatri *et al.,* 2017).

Waters of undesirable characteristics are treated using those facilities designed to protect human and the ecosystem from harmful and toxic elements. Coagulation-flocculation-sedimentation system is one of the most commonly applied water purification techniques, especially in developing countries due to its low cost, ease of maintenance and the material's local availability (Bitton, 2014). The system is widely used in municipalities for treating domestic water because of its effectiveness in the removal of various water impurities such as turbidity, color, dissolved organics as well as inorganics (Katrivesis *et al.*, 2019). However, the performance of coagulation-flocculation-sedimentation treatment plants widely varies with water characteristics, requiring specific optimization of chemical doses and operating conditions, including the initial pH, mixing rate and time, and settling time (Piri *et al.,* 2011). This makes optimization a priority before application in full-scale treatment plants (Widiyanti, 2019a).

The Kou River is a river system in the Manyara region of northern Tanzania, where water is mainly used for irrigation, forestry, agriculture, and domestic consumption. The Babati Urban Water Supply and Sanitation Authority (BAWASA) supplies domestic water from Kou River to the Magugu community in the Babati district of the Manyara region, which has a population of over 26,000. The Authority runs periodic monitoring for selected water quality parameters and, recently, the river was found to have high turbidity and Fe levels. For this reason, a coagulation-flocculation-sedimentation water treatment plant was planned to be constructed and it required optimization, specifically for the treatment of Kou River water. Therefore, this study aimed at assessing the contamination of water and sediments, and to optimize a coagulation-flocculation-sedimentation water treatment system for the Kou River.

1.2 Statement of the Problem

BAWASA (the Authority) periodically monitors water quality of the Kou River at the water collection point for selected parameters where turbidity and Fe have been found in excess compared to the acceptable limits for drinking water. For the Magugu town community and for the water Authority, excessive Fe and turbidity in water has been a problem. The water Authority is, therefore, planning to install a treatment facility in the form of a coagulation-flocculation-sedimentation system. Although this kind of system is known to be effective for the removal of turbidity, color, biological matter, dissolved organic materials and inorganics, its performance varies with operating conditions, chemical dosage, and water type, hence requiring optimization (Piri *et al.,* 2010; Katrivesis *et al.,* 2019). This work was set out to comprehensively assess the level of contamination of Kou River water and sediments, and to optimize the coagulation-flocculation-sedimentation system for the water treatment.

1.3 Rationale of the Study

Water is essential for sustaining healthy functioning of an ecosystem, reduction of poverty and socio-economic progress (Funtowicz *et al.,* 1998). One of the sustainable development goals (SDGs) set by the United Nations in 2015 was to ensure availability and sustainable management of water and sanitation for all by 2030 (Sørup *et al.,* 2020). However, the global status of water quality is negatively affected because of natural and anthropogenic factors on water sources (Khatri *et al.,* 2017). The Kou River is a river system in the Manyara region of northern Tanzania, where water is mainly used for irrigation, forestry, agriculture, and domestic consumption. The Babati Urban Water Supply and Sanitation Authority (BAWASA) supplies domestic water from Kou River to the Magugu community in the Babati district of the Manyara region, which has a population of over 26,000. The Authority runs periodic monitoring for selected water quality parameters and, recently, the river was found to have high turbidity and Fe levels. For this reason, a coagulation-flocculation-sedimentation water treatment plant was planned to be constructed and it required optimization, specifically for the treatment of Kou River water. Therefore, this study aimed at assessing the contamination of water and sediments, and to optimize a coagulation-flocculation-sedimentation water treatment system for the Kou River

1.4 Research Objectives

1.4.1 Main Objective

The main objective of this study was to conduct quality assessment and optimization of a coagulation-flocculation-sedimentation water treatment process for Kou River.

1.4.2 Specific Objectives

The following were the specific objectives:

(i) To assess the water quality of the Kou River.

(ii) To assess the pollution status of the Kou River sediments with respect to selected TMEs.

(iii) To optimize a coagulation-flocculation-sedimentation water treatment system suitable for treatment of the Kou River water.

1.5 Research Questions

(i) What is the water quality of the Kou River compared to selected standards?

(ii) What is the pollution status of the selected TMEs in the Kou River sediments?

(iii) What are the optimum conditions for a coagulation-flocculation-sedimentation process in treating the Kou River water?

1.6 Significance of the Study

The Kou River is an important river system in the Manyara region of northern Tanzania, where water is mainly used for irrigation, forestry, agriculture and domestic consumption. The Magugu community in the Babati district of the Manyara region with a population of more than 26 000 depends on the river for domestic use. However, the river water was found to be problematic due to its high turbidity and Fe concentration in reference to the WHO and TBS drinking water acceptable limits (World Health Organization [WHO], 1984; Tanzania Bureau of Standards [TBS], 2003). Although the BAWASA authority in the region plans to install a water treatment facility, i.e., a coagulation-flocculation-sedimentation water treatment system, an effective process for treating various impurities including turbidity and inorganics, its treatment performance is largely influenced by several factors, thus requiring optimization (Piri *et al.*, 2010; Katrivesis *et al.*, 2019). Therefore, the present study should provide quality data on the optimum chemical dosing and conditions of a coagulation-flocculation-sedimentation process for the treatment of the Kou and other river waters with similar characteristics. The findings in this study should recommend the Babati and other water authorities facing similar river pollution, and policy makers on the nature and interaction of pollutants, and possible pollutant sources for mitigation actions, and better environmental protection and conservation.

1.7 Delineation of the Study

The study comprehensively assessed the level of contamination of Kou River water and sediments, with respect to physicochemical properties and selected trace metallic elements, i.e., Cu, Cr, As, Zn, Mn, Co, Ni, Pb, and Fe and optimized a coagulation-flocculation-sedimentation system for the river water treatment. Only 9 metallic elements among the most common surface water pollutants were selected and studied in the research due to time and resource limitations.

CHAPTER TWO

LITERATURE REVIEW

2.1 Introduction

Rivers which are the main sources of water for socio-economic activities and sustaining healthy function of ecosystems in Africa have lost their natural water qualities as a result of both natural and anthropogenic factors (Subramaniam & Rimayi, 2018). The Kou River is among Tanzanian rivers found in Manyara region, which is mainly utilized for domestic use, agriculture, irrigation, and forestry. The river originates from Mbulu district and it is typified by huge human population whereby activities such as livestock keeping, farming, minerals mining and small-scale industries are practiced (Raphael, 2018). However, various parameters including trace metallic elements (TMEs) are potential contaminants of rivers through anthropogenic activities like irrigation, agriculture, horticulture, livestock keeping, industrial wastes and landfill leachates (Akhtar *et al.*, 2021).

Turbidity is a common problem in surface waters, which is an indication of the occurrence of suspended sediments. Suspended solids affect the growth of aquatic plants by blocking light penetration, harm the health of aquatic organisms, and carry pathogens and pollutants such as bacteria and TMEs (WHO, 2017). The quality of several surface water bodies has been exacerbated due to the presence of undesirable levels of TMEs and turbidity, and Kou River is no exception.

Environmental toxicity caused by TMEs contamination notably occur via sediments and water, which are the major components of a riverine system that consequently pose danger to human health and aquatic life. The TMEs in sediments and water can enter the food chain and bioaccumulate in body organs and tissues (Jaishankar *et al.,* 2014). It is worth noting that some TMEs such as Cu, Cr, Co, Zn, Fe and Ni are essential minerals for proper body growth at low concentration, however, high doses result in acute and chronic health problems when consumed by human and aquatic life (Balali-Mood *et al.,* 2021; Wongsasuluk *et al.*, 2021; Zaynab *et al.*, 2022). For instance, TMEs like As and Pb have been reported to impede the functions of organism at low concentrations (Jaishankar *et al.,* 2014; Raychaudhuri *et al.*, 2021). Therefore, understanding the quality and pollution status of water bodies help to take measures for environmental protection and conservation where the safety of the ecosystem can be preserved (Apitz *et al.*, 2006).

There are several water treatment techniques made available under 3 major classes (conventional strategies, biological strategies and membrane technology-based strategies) including trickling filter, ultrafiltration, microfiltration, reverse osmosis, filter media separation, adsorption, electrocoagulation/floatation, oxidation/precipitation as reviewed by researchers, each of them having their own merits and demerits. Combination of some of these methods within a treatment plant in an optimized manner has been identified to have an outstanding removal efficiency (Khatri *et al.*, 2017). However, coagulation-flocculation-sedimentation process is the most applied technique in most developing countries due to its low cost, ease of maintenance and materials local availability (Bitton, 2014).

Coagulation-flocculation-sedimentation system is one among the commonly applied water treatment techniques at medium–large scale water purification plants to sustain a healthy ecosystem (Ratnayaka *et al.,* 2009). The BAWASA authority in Manyara region in Tanzania also plans to apply the same technique for Kou River water treatment to be supplied to the Magugu community. Nonetheless, although this technique is effective in the removal of color, dissolved organic materials, inorganic pollutants and turbidity, its performance efficiency often fluctuates depending on several factors which urges for optimization (Widiyanti, 2019a).

Factors that affect the performance of coagulation, flocculation-sedimentation process include dose of chemical, initial pH, mixing conditions and type of the raw water to be treated (Widiyanti, 2019a). Understanding the characteristics of the water is important for running successful optimization because it is a priority factor which determines the performance of the treatment system (Su, 2019). Optimization is usually conducted using jar test, with a requirement for validation using scale-up pilot tests for reliability in the performance of obtained results on full-scale plants (Jan *et al.*, 2007).

2.2 Water Quality Assessment

Water and sediment quality guideline values for various parameters are established at both national and international levels to protect human health and aquatic life and to sustain the desired use of water bodies (Elamassi, 2012). Water sources that vitally support lives of diverse habitats should be examined for their suitability with respect to purpose-specific guideline values as listed below.

2.1.1 Tanzania Bureau of Standards and World Health Organization Drinking Water Quality Standards

Access to safe drinking-water is essential as a health and development concern at national, regional and local levels. The WHO drinking water quality guidelines are addressed principally to water and health regulators, policymakers and their advisors, in order to assist in the development of national standards (WHO, 1984). The TBS drinking water quality standard specifies the permissible concentration level of pollutants in the water used for drinking (TBS, 2003).

2.1.2 Water Quality Standards for Agriculture by the Food and Agriculture Organization of the United Nations

The FAO of the UN developed agriculture water quality standard for guiding farm and task managers, experts and engineers in assessing and analyzing potential issues related to water quality. The recommendation was developed to indicate potential problems and feasible regulation on use of the water for agriculture (Maleki *et al.,* 2013),

2.1.3 United States Environmental Protection Agency Aquatic Life Quality Guidelines

The EPA aquatic life standard is developed to recommend the amount of chemicals that can exist in surface waters without harming plants and animals. The EPA standard is established to protect both freshwater and saltwater organisms from long-term and short-term exposure (USEPA, 2014).

2.1.4 The Nqep and SEQ-Eau Surface Water Quality Standards

The circular of 7 May 2007 DCE/23 defining ‘standards of provisional environmental quality (NQEp)’ for surface fresh waters” and “the French system for the evaluation of the quality of water streams (SEQ-Eau) recommended by the water agencies for fresh waters” were established and introduced to recommend the advisable concentrations of chemical elements in surface and stream waters with an objective of maintaining healthy ecological status (Khaled-Khodja *et al.,* 2018).

2.3 Sediment Quality and Contamination Assessment

2.3.1 Sediment Quality Standard Guidelines

There are various standard guidelines at both national and international levels for the evaluation of sediment's pollution status based on the impacts on environmental health. The "SEQ-Eau standard for biological life" and "the USEPA sediment quality criteria" are few among many established sediment quality guidelines (Dasar *et al.*, 2009; Zoynab *et al.*, 2013; Khaled-Khodja *et al.*, 2018).

2.3.2 Pollution Indices

Pollution indices are tools which are used for the evaluation of the level of contamination in sediments (Joanna, 2018). The following lists are some of the commonly applied pollution indices which have been developed to inspect the contamination of TMEs in sediments. Explanations and interpretations of each pollution index is given in Table 4 under the methods section.

(i) Geo-Accumulation Index (I_{geo})

Is a tool to identify extent of metals contamination in sediments. In the I_{geo} calculation (Table 4), the background value is multiplied by a constant of 1.5 to compensate possible variations due to lithogenic effect (Engdaw *et al.*, 2022).

(ii) Enrichment Factor (EF)

Is a pollution index to assess possible anthropogenic impacts on sediments. It indicates the intensity of influence from human activities on the presence of a metal in sediments (Birch, 2013).

(iii) Contamination Factor (CF)

The CF, also known as single pollution index, is applied to identify element's contamination in sediments. It is calculated by comparing metal concentration with its background value (Singh *et al.*, 2017). It is an easy and worthwhile tool to monitor single metal contamination by classifying into 4 groups from low to very high level (Ajani *et al.*, 2021).

(iv) Modified Degree Of Contamination (Mcd)

Is a generalized tool that determines the overall degree of TMEs contamination of a specific site which incorporates as many metals as desired with no limitation (Zarei *et al.,* 2014).

(v) Potential Ecological Risk (RI)

Was introduced by Hakanson for determination of the extent to which ecological risk is posed by the presence of heavy metals in sediments. It is calculated from the single ecological risk factor of each metal depending on their toxicity response coefficients (T_r^i) (Zhu *et al.,* 2012; Chen *et al.,* 2020).

2.3.3 Sequential Extraction of Trace Metallic Elements in Sediment

Total metal concentration in sediments is not always indicative of actual risks to living organism; rather, it is their mobility and bioavailability which are the main factors that influence TMEs contamination (Namieśnik & Rabajczyk, 2010; Alhadrami *et al.,* 2016). The TMEs bioavailability and toxicity are determined using bioavailability factor (BF) and Risk Assessment Code (RAC) respectively by involving the proportion of the different fractions of metals in the calculations. The fractions of metals can be sequentially extracted using Tessier Sequential extraction method (Tessier, 1979) that fractionate TMEs species into four and the Community Bureau of Reference (BCR) (Delil & Köleli, 2018) that speciate the TMEs species in to five species. Depending on their mobility, six fractions of metals are stated in Table 1.

Table 1: Description of the sequentially extracted fractions of metals

TMEs Fractions	Description	Reference
Water soluble	Metals bound in an active form, dissolved and very weakly bonded, great mobility and bioavailability.	(Bożym, 2017)
Ion-exchangeable	Weakly bonded through electrostatic interaction or ion exchange, great mobility, and bioavailability during slightly acidic conditions.	Bakircioglu *et al.* (2011)
Carbonate bound	Bound to carbonates, phosphates and sulfates, potentially mobile and bioavailable at low pH.	Baran and Tarnawski (2015), Bożym (2017)
Fe-Mn bound	Reduceable fractions, become potentially mobile and bioavailable through coprecipitation and adsorption in conditions of low oxidation and reduction potential, and are considerable constituents of sediments.	Okuku *et al.* (2010)
Organics and sulfide bound	Oxidizable fractions, metals bound to organic matter and sulfides, released through decomposition of organic matter when oxidized conditions predominate.	Devi and Bhattacharyya (2018)
Residuals	Metals bound to the crystalline matrix, non-bioavailable and no potential ecological risk.	Al-Mur (2020)

2.3.4 Leaching kinetics of Trace Metallic Elements in sediments

The leaching kinetics of TMEs is related to their mobility and toxicity where the information provides a broad understanding on their rate and degree of release during variations in natural conditions (Violante *et al.,* 2010). To estimate the leachability potential of elements from sediments over a particular period, different leaching solutions such as acid rain have been utilized. Kinetic experiments help to determine the time required to reach an equilibrium (Lee *et al.*, 2012).

2.4 Water Treatment Techniques

Various surface water treatment techniques have been made available under 3 major classes (Conventional strategies, biological strategies and membrane technology-based strategies) over the globe. Technologies such as trickling filter, ultrafiltration, microfiltration, reverse osmosis, coagulation-flocculation-sedimentation, filter media separation, adsorption, oxidation/precipitation, and electrocoagulation/floatation have been reviewed by researchers as treatment methods, each of them having their own merits and demerits. Combination of some of these methods within a treatment plant in an optimized manner has been identified to have an outstanding removal efficiency (Khatri *et al.,* 2017). Among these methods, the coagulation-flocculation-sedimentation water treatment technique, a technique which requires optimization specific to the raw water to be treated, is the most commonly applied technique in most developing countries due to its affordable cost, ease of maintenance and materials local availability (Piri *et al.,* 2010; Katrivesis *et al.,* 2019).

2.5 Optimization of Coagulation-Flocculation-Sedimentation System

2.5.1 Coagulation-Flocculation Definition and Principle

Coagulation-flocculation-sedimentation-filtration system is a commonly applied solid-liquid separation procedure in water treatment. Coagulation-flocculation and sedimentation are important initial stages in the treatment of most surface waters (Ratnayaka *et al.,* 2009). Coagulation and flocculation are interconnected phrases which explain the formation of colloidal particles to be removed by sedimentation. The term “coagulation” means the destabilization of colloidal particles by a method which weaken the energy forces around particles that restricts them from bonding together to form a bigger mass, i.e., micro flocs with the addition of chemical coagulant. On the other hand, flocculation refers back to the agglomeration of micro-flocs together to form visible suspended particles which later grow in to bigger settleable flocs. The process by which the flocs settle to the bottom of the water is called sedimentation (Taşdemir, 2012).

The coagulation-flocculation-sedimentation processes is capable of treating turbidity, color, biological matter, and other dissolved organic materials as well as inorganics such as iron precipitates in water with great efficiencies (Katrivesis *et al.,* 2019). However, the removal of pathogens after this process is not satisfactory where a later stage of filtration and disinfection is required for removing those water defects. In the coagulation-flocculation-sedimentation stage, the removal of viruses and bacteria ranges between 27-84% and 32-87 % respectively (Conventional water treatment: Coagulation and filtration fact sheet). Thus, the flocculation process is crucial because it reduces the demand for chlorine disinfectants in the later stage which could result in formation of trihalomethanes (THM) through reaction with natural organic matter (NOM) present in the water (Ratnayaka *et al.,* 2009).

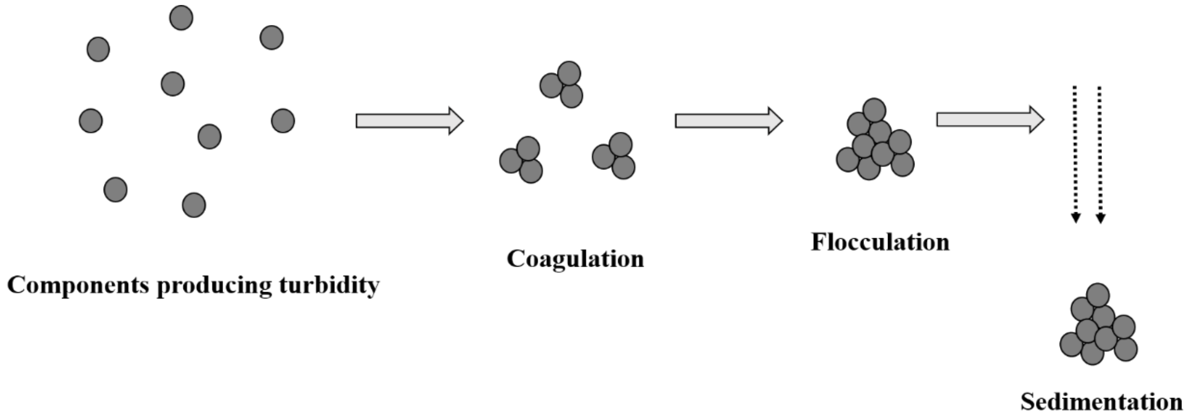


Figure 1: A diagrammatic representation of the coagulation-flocculation-sedimentation process (Obtained and modified from Conventional water treatment: Coagulation and filtration fact sheet)

Although the coagulation-flocculation-sedimentation water purification technique is known to be effective, its performance is negatively affected due to inadequate applications (Piri *et al.*, 2010). There are several factors which control the flocculation performance. Type of chemical coagulant and dose, Water alkalinity, pH of water, mixing conditions, temperature and flocs settling time are some of the factors which affect the effectiveness of the coagulation-flocculation process (Widiyanti, 2019b). Therefore, optimization of these parameters before application in full-scale water treatment plants is critical.

2.5.2 Factors Affecting Coagulation-Flocculation Process

(i) Dose of Coagulant Chemicals

A number of coagulants are used in water treatment including aluminum salts, iron salts, hydrated lime and magnesium carbonate. Among these, aluminum sulfate is a commonly applied coagulant chemical in most drinking water purification plants (Shewa & Dagnew, 2020). When aluminum sulfate ($Al_2(SO_4)_3$) is added to water, a chemical reaction takes place where the aluminum salt (Al III) dissociates, and trivalent aluminum (Al^{3+}) is produced as indicated below:

$$Al_2(SO_4)_3 \rightleftarrows 2Al^{3+} + 3SO_4^{2-} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

After dissociation, the aquometal complex ($Al(H_2O)_6^{3+}$), a predominant specie of aluminum salt in aqueous solution is formed from the hydration of trivalent aluminum. Then different soluble polynuclear and mononuclear species form and interact with the particles that are present in the water to form precipitates (Su, 2019).

Overall, the precipitation reaction when hydrated aluminum sulfate (alum) is added to water is provided as follows:

$Al_2(SO_4)_3 \cdot 14H_2O \rightarrow 2Al\,(OH)_3(s) + 6H^+ + 3SO_4^{2-} + 8H_2O$ …………………………… (2)

The dose of coagulant chemical is an important factor that determines the efficiency of coagulation-flocculation process. Underdosing results in insufficient floc formation whereas overdosing of the chemicals cause re-stabilization of particles, which leads to an increment in unsettleable suspended particles in the water (Widiyanti, 2019b). Besides, if the optimum dose and operating conditions are not met, health problems emerge as a result of residuals from the coagulant chemicals. For instance, excessive residual aluminum from aluminum sulfate coagulant in treated drinking water can cause Alzheimer disease in people (Katrivesis *et al.*, 2019). The EPA in 2017 reported that residual aluminum in treated water should fall in the range 0.05 and 0.2 mg/L recommending that 0.2 mg/L Al as secondary maximum pollutant level (Krupińska, 2020).

The required dose of chemical can be obtained not only in proportion to the suspended particles present in the water, but also on the conditions that could create a room for efficient performance of the coagulant chemical (Widiyanti, 2019b). Under optimum operating conditions, the chemical requirement could be minimized making it cost effective and health friendly. Though optimum doses of chemicals for a specific treatment system are determined through optimization experiments, there is still a challenge of overdosing and underdosing due to fluctuation of raw water characteristics especially turbidity over short period (Gauthier *et al.*, 2003). Therefore, periodic water quality monitoring and optimization is important because a single time optimized dose cannot be used permanently on waters with varying characteristics.

(ii) Initial pH of Water

The stabilization of particles and surface charges of coagulants is significantly affected by pH. Optimum pH conditions promote neutralization of negatively charged particles and facilitate formation of flocs (Widiyanti, 2019b). The suitability of pH condition varies for different purification plants depending on the target impurities in the water. For instance, low pH value facilitates efficient flocculation in the presence of natural organic matter; slightly acidic to neutral pH is suitable for algae removal; and neutral pH is desirable for the removal of inorganics (Naceradska *et al.*, 2019). Therefore, it is crucial to carefully adjust the initial pH of the water to optimum in relation to the removal of a specific impurity.

(iii) Mixing Conditions and Sedimentation

The first important stage of the coagulation-flocculation process after the addition of coagulant chemical is rapid mixing. In this stage, an intense, turbulent mixing for a short period of time is required in order to facilitate homogeneous distribution of the coagulant chemical in the water and to promote collision among particles. The collision of particles provides good coagulation and results in formation of micro-flocs. However, without fast enough mixing rate, overdosing of chemical could occur. The rapid mixing duration is another factor which influence the performance of the flocculation process. Although it is required to allow sufficient time for mixing, extended period than the optimum leads to poor flocculation performance (Taşdemir, 2012).

The second stage following the rapid mixing process is slow mixing. This step is crucial to accommodate flocculation by increasing collision rates between destabilized particles and flocs. At this stage, a slow to moderate mixing intensity must be maintained to prevent breakage of the already formed flocs. Once bigger settleable flocs are formed, sufficient time should be provided for sedimentation to accommodate solid-liquid separation (Taşdemir, 2012).

2.5.3 Optimization of the Coagulation-Flocculation-Sedimentation Process

Water treatment optimization is most often carried out using Jar test; inexpensive lab-scale batch test method for identifying suitable chemicals, doses and operating conditions without altering the behavior of full scale treatment plants (Zane, 2005). However, jar test also has the disadvantages of being time consuming and providing results that are sometimes inconsistent to the results obtained on full-scale plants (Mazille & Spuhler, 2014; Schott, 2020). In order to provide quality data for making informed decisions for transferable full-scale plant performance, it is important to validate the jar-test optimized conditions on Scale-up pilot tests (Jan *et al.*, 2007).

(i) Jar-test

A jar-test is a pilot scale test which represents a coagulation-flocculation-sedimentation process of a particular water treatment plant using a jar test equipment containing jars, impellers, impeller drivers and lab analysis equipment (Su, 2019). The jar-test is used to identify the most appropriate chemicals, doses and operating conditions for treating a specific raw water (Zane, 2005). Despite being widely used, this system faces challenges due to it's high time consumption and sometimes, inconsistency of results when applied on full-scale plants (Mazille & Spuhler, 2014).

Different optimization techniques have been developed for successful determination of suitable working conditions using jar test. The one-factor-at-a-time (OFAT) technique is a simple and commonly applied technique which is performed by varying each individual factor (dose, pH, rapid-slow mixing rate and time) in a single experiment at once where the other factors are kept constant. Following each experiment, the supernatant water is analyzed for the target impurity and the condition which provides better results is considered to be optimum (Dawood & Li, 2013). The Fig. 2 shows a typical jar-test apparatus used in the jar-test process.

Figure 2: A laboratory set up of the jar-test apparatus

(ii) Scale-Up Pilot Plant Test

Studies have pointed out that results obtained from jar tests may sometimes fail to correspond with the ones obtained up on application on full-scale treatment plants (Mazille & Spuhler, 2014; Schott, 2020). In this regard, conducting further validation experiment/test is critical to validate the optimized conditions and to provide quality data for transferable full-scale plant performance (Jan *et al.,* 2007). This can be done through scale-up-pilot-test which is a process of increasing the size of the batch testing operation for implementing similar process to larger output volumes (Ismail *et al.,* 2012; Crater & Lievense, 2018).

2.6 Sources of River Water Pollution

Natural and anthropogenic contamination and limited treatment practices affected the availability of clean and safe water in Africa (de Paul Obade & Moore, 2018). Some of the common sources of water pollution include industrial wastes, flooding during rainy seasons carrying wastes in to waters, deforestation, pesticides from agricultural cites, and herbicides and fertilizers (Owa, 2013). Turbidity is a common problem in most surface waters which is usually induced as a result of poor farming practices, wastewater discharge, phytoplankton, erosion during rainy seasons, inorganics, algae, urban runoff and sediment disruption (Karikari & Ansa-Asare, 2006). Metals contamination has also received a significant concern due to their toxicity, accumulation in habitats and their perseverance in the ecosystem. Bedrocks release through dissolution, runoff from banks and basins drain have been regarded as the prevailing sources for lithogenic contribution while primary man made sources may include mining and smelting operations, industries, fossil fuels combustion and influents from urban and industrial wastewater (Giri *et al.,* 2013). Sediments collected in river beds have been used to identify possible sources of the pollutants and their distribution where in most cases, both natural and anthropogenic activities discovered to have variable contributions in different seasons (Xiao *et al.,* 2013).

This study assessed the Kou River water and sediment quality and contamination. Furthermore, this study optimized a coagulation-flocculation-sedimentation process proposed for a water treatment system for the Kou River following the methods as presented in Chapter 3.

CHAPTER THREE

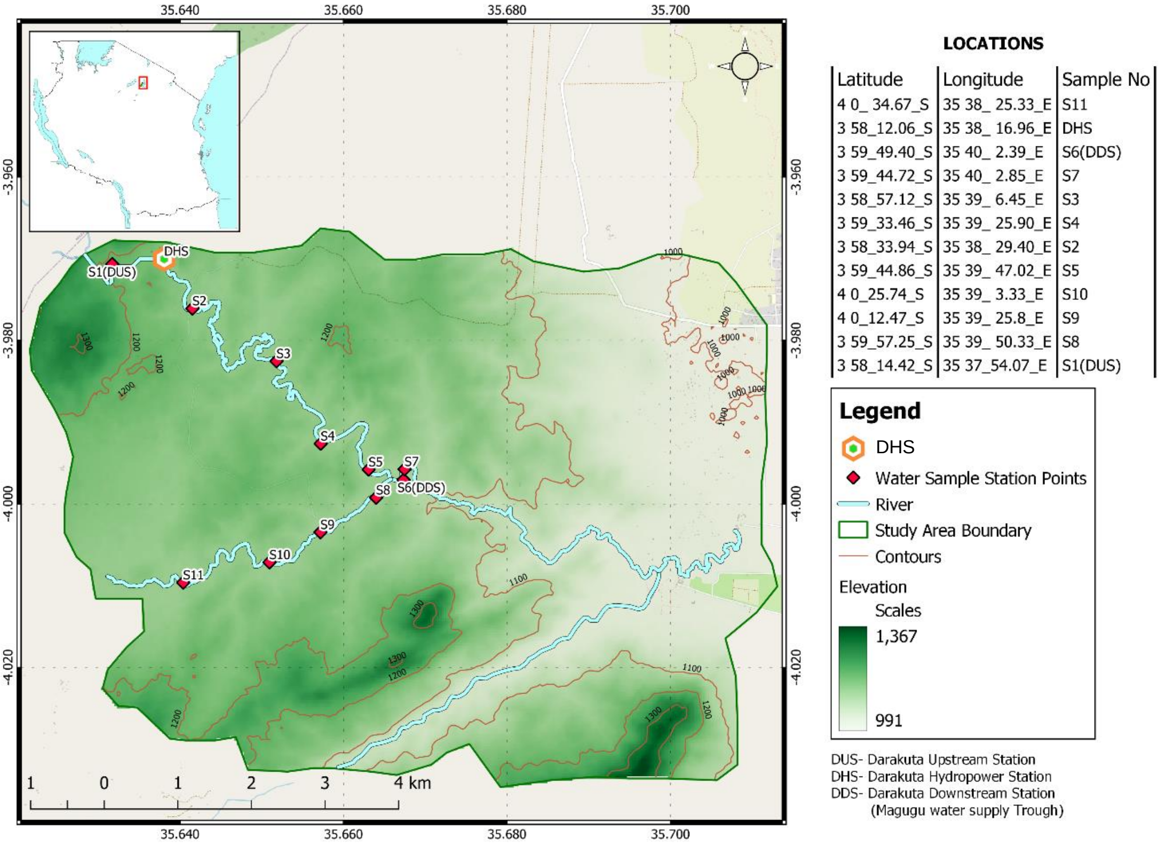
MATERIALS AND METHODS

3.1 Study Site

The Kou River is a perennial river located in Manyara region in Northern Tanzania (Fig. 3). Detailed features of the Kou River are provided in Table 2 in reference to the data provided by Babati Urban Water Supply and Sanitation Authority (BAWASA) and field observation.

Table 2: Detailed geographical and water flow parameters for the Kou River found by the present study

Kou River	Description
Type	Perennial
Origin	Nou Hills, Mbulu district
Flow through districts	Mbulu-Babati
Estimated length	40 km
Average width	3.70 km
Average annual depth	2.50 m
Maximum depth	3.00 m
Flow rate	1.32 m^3/s
Water collection site	Babati district: Darakuta Ranch
Site observations	Darakuta Ranch: 7000 ha, supports ranching, wildlife, agriculture, mini-hydropower plant (300 kw/h), domestic water supply



LOCATIONS

Latitude	Longitude	Sample No
4 0_ 34.67_S	35 38_ 25.33_E	S11
3 58_12.06_S	35 38_ 16.96_E	DHS
3 59_49.40_S	35 40_ 2.39_E	S6(DDS)
3 59_44.72_S	35 40_ 2.85_E	S7
3 58_57.12_S	35 39_ 6.45_E	S3
3 59_33.46_S	35 39_ 25.90_E	S4
3 58_33.94_S	35 38_ 29.40_E	S2
3 59_44.86_S	35 39_ 47.02_E	S5
4 0_25.74_S	35 39_ 3.33_E	S10
4 0_12.47_S	35 39_ 25.8_E	S9
3 59_57.25_S	35 39_ 50.33_E	S8
3 58_14.42_S	35 37_54.07_E	S1(DUS)

DUS- Darakuta Upstream Station
DHS- Darakuta Hydropower Station
DDS- Darakuta Downstream Station
(Magugu water supply Trough)

Figure 3: Map of Tanzania (top left inset) showing the Kou River and sampling points used in the present study (Gebreyohannes *et al.*, 2022)

3.2 Sample Collection

Sample collection from the Kou River was undertaken at two different periods. The first sample collection for purpose of assessing the Kou River quality was undertaken from 3rd - 6th March, 2021. The collection of water and sediment samples were conducted in Kou River and an Unnamed tributary in Darakuta Ranch, Babati district. Kou river water and sediments were sampled at seven sampling stations while that of the unnamed tributary was sampled from four sampling sites. Both water (triplicate samples) and sediment (compound samples) samples were collected from same points in the river. 1 liter polyethylene plastic bottles were used for water sample collection after thorough rinsing with acid. The sampled waters were filtered using membrane filtration assembly of conical shape using 0.45 membrane filter papers. Filtered samples were preserved to pH less than 2 using a few drops of concentrated nitric acid, transported to NM-AIST laboratory in an ice-cool box and stored in refrigerator at 4°C until analysis. Surface sediment samples were collected at 7 cm depth from the top surface of the river bed using sediment sampler and plastic spatulas. Then they were carried to the laboratory in ice-cool box in polyethylene bags and dried at 105°C in oven and prepared for TMEs analysis.

The second water sample collection for running an optimization experiment was carried out on February 3rd, 2022. Water samples were collected from the point where the BAWASA authority collects water for Magugu town domestic water supply. Ten pieces of thoroughly rinsed 20-litre jerricans were used to collect the samples.

3.3 Physicochemical Properties Analysis in Water and Sediments

3.3.1 Physicochemical Properties Analysis

All samples from the eleven sampling stations of the Kou River were analyzed for physicochemical properties listed below. The analysis was caried out based on “the standard methods for examination of water and wastewater” (American Public Health Association [APHA], 2017). Dissolved oxygen (DO), pH, turbidity, electrical conductivity (EC) and Total dissolved solids (TDS) were analyzed on site using the Hanna Instrument Multiparameter (HI-9829). The EDTA Titrimetric method was used for hardness analysis using the DIT 50 Behrotest hardness analysis device.

3.3.2 Trace Metallic Elements Analysis in Sediments and Water

Spectro Xepos™ Energy-Dispersive X-Ray Fluorescence (ED-XRF) machine was used for analysis of TMEs in sediment samples guided by SPECTRO XEPOS user manual. Dry weight of the samples (4 g of each), cellulose binder (0.9 g) was measured using the electronic beam balance and set together with four 3 mm radius spherical balls in bowl. Then grounding and homogenization of the mixture was done using an equipment called “Fritsch Pulverisette™ pulverizer” at 150 rpm and for 15 minutes. The mixture from this procedure was then poured into the lapped thrust piece and pellets were formed by pressing the mixture using hydraulic Retsch™ with a pressing pressure of 12 tons. Four replicate pellets prepared out of each sample, measured using ED-XRF and X–Lab Pro™ software used for quantification/analysis of the elements of concern.

The USEPA procedures manual was used for the analysis of TMEs in water. Filtered water samples from the Kou River downstream and upstream stations namely Darakuta downstream (DDS) and Darakuta upstream (DUS) stations were analyzed for Mn, Co, Fe, Pb, As, Cr, Zn, Cu and Ni which are among the most common metal pollutants in surface water bodies. WFX-210 atomic absorption spectrometer (AAS) machine was used to analyze Mn, Pb, Co, Zn, Cr, and Ni following EPA Method 1620 (Mahugija, 2018). Each element analysis utilized five machine calibration standards. Element specific operating conditions of the machine such as lamp current, spectral bandwidth, acetylene pressure, height of burner, air pressure and flow, and wavelength were adjusted, and analysis took place. Palintest Arsenator (PT981) digital Arsenic test kit was used for arsenic analysis in water using the procedures manual for digital arenator. Copper and iron were analyzed using DR 2800 by Method 8506 & 8026 and Method 8008 respectively. Calorimetry technique was followed for this purpose as per the provided procedures manual (APHA, 2017).

3.4 Quality Control

Analytical grade reagent chemicals were used for all the experiments that were computed for TMEs analysis. Standards solutions were prepared in laboratory to calibrate the atomic absorption spectrometer (AAS) machine. The equation “MDL = 3.14S” was used to establish the machine detection limit (MDL) in mass-spectrometry and calorimetry analysis methods where “S” stands for standard deviation of seven replicates of a specific standard. 3.14 represents a *t* value for 6 degrees of freedom (7-1), at 99% confidence level, from one-sided *t* distribution table. The MDL for Zn, Pb, Cu, Cr, As, Co, Ni and Mn was 1.75, 1.75, 70, 10.5, 3.5, 8.75, 17.5 and 3.5 μg/L respectively. Samples were diluted before analysis to eliminate the matrix effect that was pre-identified by standard addition method. Basalt Columbia River (BCR-2) certified reference material was used for the validation of the sediments analysis method where the percent recovery was maintained between 90% - 110% with analysis of 4 replicates as provided in Table 3 (United States Geological Survey [USGS], 1998).

Table 3: Measured data and the analytical results of the certified reference material (BCR-2)

Metal	Certified value	Measured value	Recovery (%)
Fe (%)	9.65±0.15	9.27±0.05	96.06
Al (%)	7.14±0.10	7.79±0.43	109.10
Mn (mg/kg)	1520.00±60.00	1482.00±16.50	97.50
Cr (mg/kg)	17.00±2.00	18.61±0.70	109.47
Co (mg/kg)	37.00±3.00	39.98±1.50	108.05
Zn (mg/kg)	127.00±9.00	121.62±3.42	95.76
Cu (mg/kg)	19.00±2.00	17.59±0.75	92.58
Pb (mg/kg)	11.00±2.00	12.09±1.70	109.90

3.5 Water Quality Assessment

Being used for several purposes, Kou River water quality was assessed with respect to “aquatic life quality guidelines” (USEPA, 2014), “the circular of 7 May 2007 DCE/23 defining ‘standards of provisional environmental quality (NQEp)’ for surface fresh waters” and “the French system for the evaluation of the quality of water streams (SEQ-Eau) recommended by the water agencies for fresh waters” (Khaled-Khodja *et al.,* 2018) and “water quality standards for agriculture” by FAO of the UN (Ayers & Westcot, 1995; Maleki *et al.,* 2013).

3.6 Sediment Quality and Contamination Assessment

3.6.1 Sediment Quality Guidelines

Sediment’s pollution status based on their impacts on environmental health was assessed using “SEQ-Eau standard for biological life” and “the USEPA sediment quality criteria” standard guidelines (Dasar *et al.,* 2009; Zoynab *et al.,* 2013; Khaled-Khodja *et al.,* 2018).

3.6.2 Sediment Pollution Indices

Among several sediment contamination indicating tools, six pollution indices were computed aiming on evaluating the risks of TMEs in the Kou River sediments. Their detailed descriptions and elucidation are given in Table (4).

Table 4: Explanation and interpretations of sediment pollution indices[a] used in the present study

Equation	Where	Interpretation	
		Sediment contamination status	
Geo-accumulation index $I_{geo} = Log_2(\frac{C_n}{1.5B_n})$	Cn: metal concentration in sample Bn: Background metal concentration 1.5: correction factor	$I_{geo} < 0$	Uncontaminated
		$0 \leq I_{geo} < 1$	Uncontaminated to moderate
		$1 \leq I_{geo} < 2$	Moderate
		$2 \leq I_{geo} < 3$	Moderate to heavy
		$3 \leq I_{geo} < 4$	Heavy
		$4 \leq I_{geo} < 5$	Heavy to extreme
		$I_{geo} \geq 5$	Extreme
		Enrichment	
Enrichment factor $EF = \frac{(C_n/C_{RE})_{Sample}}{(C_n/C_{RE})_{Background}}$	C_{RE}: reference element (Al, due to its conservative nature and stability	$EF < 2$	Deficit to minimum
		$2 \leq EF < 5$	Moderate
		$5 \leq EF < 20$	Significant
		$20 \leq EF \leq 40$	Very high
		$EF > 40$	Extremely high
		Degree of contamination	
Contamination factor $CF = \frac{C_n}{B_n}$		$CF < 1$	Low degree
		$1 \leq CF < 3$	Moderate degree
		$3 \leq CF < 6$	Considerable degree
		$CF > 6$	Very high
		Degree of contamination	
Modified contamination factor $mCd = \frac{(\sum_{i=0}^{n} CF_i)}{n}$	CF_i: Contamination factor of metal i n: number of metals	$mCd < 1.5$	Nil/very low
		$1.5 \leq mCd < 2$	Low
		$2 \leq mCd < 4$	Moderate
		$4 \leq mCd < 8$	High
		$8 \leq mCd < 16$	Very high
		$16 \leq mCd < 32$	Extremely high
		$mCd \geq 32$	Ultrahigh
		Ecological risk	
Ecological risk $E_r^i = T_r^i \times CF$	T_r^i: toxicity risk coefficient of metal i	$E_r^i < 40$	Low
		$40 \leq E_r^i < 80$	Moderate
		$80 \leq E_r^i < 160$	Considerable
		$160 \leq E_r^i < 320$	High
		$E_r^i \geq 320$	Very high
		Potential ecological risk	
Potential ecological risk $RI = \sum E_r^i$		$RI < 150$	Low
		$150 \leq RI < 300$	Moderate
		$300 \leq RI < 600$	Considerable
		$RI \geq 600$	High

[a]Obtained from (Hakanson, 1980; Kowalska *et al.*, 2018; Shilla & Shilla, 2021; Gebreyohannes *et al.*, 2022)

3.6.3 Sequential Extraction Procedure

Tessier and BCR sequential extraction methods were applied to extract the different fractions of metals. The 1 g by dry weight of sediment was agitated in 12.5 ml of leaching solution for extraction and extracted in centrifuge tube at 4000 rpm. The extracted supernatant was then filtered and analyzed for the element of concern using DR2800 spectrophotometer. Each extraction procedure was performed in triplicates for valid results. The step-by-step methods are illustrated as follows:

(i) Water Soluble (F1)

12.5 mL of distilled water was mixed with 1 g of sediment and agitated for 4 h at room temperature.

(ii) Ion-Exchangeable (F2)

The 12.5 mL of 1 M Magnesium chloride at pH 7 was mixed with the residue from step 1 and agitated for 1 hr.

(iii) Carbonate Bound (Acid Soluble) (F3)

The 12.5 mL of 0.11 M acetic acid solution was mixed with the residue from step 2 and agitated for 16 hrs.

(iv) Fe-Mn Oxide Bound (Reducible) (F4)

The 12.5 mL 0.2 M ammonium oxalate and oxalic acid solution at pH 2.5-3 was agitated with the residue from step 3 for 4 hrs.

(v) Organic Bound (Oxidizable) (F5)

The 10 mL of 8.8 M hydrogen peroxide solution (solution I) where pH adjusted to 2-3 with concentrated nitric acid and added to the residue from step 4 in tube and digested at room temperature for 1 hour with occasional shaking. Digestion continued with 85°C for the following 1 hour, the volume was minimized to few ml. Then, 10 mL solution I was added and heated for 1 h at 85°C and volume minimized to few mills. Sample was diluted to 40 mL with 1 M ammonium acetate solution after cooling (Tessier, 1979; Pueyo *et al.,* 2008; Sutherland, 2010; Hossain *et al.,* 2019).

3.6.4 Leaching Kinetics Experiment

Natural rainwater was used as the extractant for conducting leaching experiment. 12.5 mL of the rainwater was used to extract metals of concern from 1 g dry weight sediment. The mixture was agitated with 30 rpm at room temperature for 1 hour, 6 hours, 12 hours, 24 hours, 48 hours, 72 hours, 6 days, 9 days, 12 days, 15 days, 18 days, 21 days, 24 days, 27 days and 1 month. The leachates were then centrifuged at 4000 rpm and filtered before being analyzed for the TMEs of interest.

3.7 Optimization of Coagulation-Flocculation Sedimentation System

Experiments were based on the One Factor at a Time (OFAT) jar testing procedure. The main parameters of the optimization procedure were turbidity, residual aluminum and other TMEs which were found in excess for drinking water. The initial concentration of each parameter was not influenced (kept constant) throughout the process. The experiment was divided in to five parts and additional control experiment was conducted to see the effect of pH, mixing conditions, and settling on the removal of impurities without chemicals addition. The supernatant water after the sedimentation process of each experiment was analyzed to identify the best-performing dose and conditions. The factors considered in the optimization experiment were:

(i) Optimal dose of coagulant chemical.

(ii) Optimal operating pH.

(iii) Effect of rapid-slow mixing rate.

(iv) Effect of rapid-slow mixing time.

(v) Effect of settling time.

(vi) Control experiment (pH, mixing conditions and sedimentation effect) without addition of coagulant chemical.

The data obtained from these experiments was utilized to identify the optimum operating conditions and was presented in the results and discussion section.

3.7.1 Kou River Water Characteristics

Before the optimization experiment was conducted, the characteristics of the Kou River water at the water supply collection point was determined to identify the target elements to be removed by the treatment plant (Table 12). The water quality was assessed using WHO and TBS drinking water quality guidelines (WHO, 1984; TBS, 2003).

3.7.2 Basic Chemical Reagents and Source of Water

The coagulant chemical used in this experiment was aluminum sulfate hydrate ($Al_2SO_4{}^3.xH_2O$), which was purchased from HACH Company distributors in Dar-es-salaam. The analytical chemicals used for pH adjustment were sodium hydroxide (NaOH) and nitric acid. All the experimental water was collected from Kou River domestic water supply collection point and the quality of the water for running the optimization experiment was not influenced.

3.7.3 Main Equipment for Optimization Process

The standard jar testing device was used for jar testing and the impellers and jars were thoroughly rinsed with distilled water before each experiment. The turbidity meter, pH sensor and the spectrophotometer devices were calibrated every time before being used for measurement.

3.7.4 Jar-Test Optimization

(i) Optimal Dose of Coagulant

For determination of optimal dose of the coagulant chemical (alum), eight different concentrations (2.5 mg/L to 50 mg/L) were tested. The initial operating conditions were, pH = 7.7 (the waters natural pH), rapid-slow mixing rate 150-20 rpm, rapid-slow mixing time of 5 min - 30 min and settling time of 15 min (Taşdemir, 2012; Oraeki *et al.,* 2018). After this procedure, the dose of alum which performed better were chosen to be applied in the following experiments.

(ii) Optimal pH

Determination of optimum pH was carried out by varying the initial pH of the water between 5 and 9. The result which was chosen from the first experiment was applied for chemical dosing.

The rest of the initial working conditions were maintained from the first experiment except for pH. After this procedure, the optimum working pH was identified.

(iii) Effect of Rapid-Slow Mixing Rate

The previously identified optimum dose and pH along with the other working conditions from experiment 1 and 2 were maintained for this experiment. The effects of rapid mixing at 50 rpm, 75 rpm, 100 rpm and 150 rpm and slow mixing rates of 10 rpm, 20 rpm, 30 rpm and 40 rpm were evaluated in this experiment. The mixing rate with better performance were selected and applied in the following experiments.

(iv) Effect of Rapid-Slow Mixing Time

The selected dose and operating conditions from experiments 1, 2 and 3 were maintained where the rapid mixing time of 1 min, 3 min and 5 min and slow mixing time of 10 min, 20 min, 30 min and 40 min were evaluated. The mixing time which performed well was maintained in the remaining experiments.

(v) Effect of Settling Time

The flocculation-sedimentation performance was examined at different settling times, i.e., 15 min, 30 min and 1 hr. All the other working conditions were maintained from the results of the previously conducted experiments.

(vi) Control Experiment

To observe the independent effects of pH and the mixing and settling conditions on the removal of Fe and turbidity from the river water, a control experiment with pH variation in the range of 5 to 9 was conducted without the addition of any coagulant chemical.

3.7.5 Scale-up pilot test

To validate the Jar-test-optimized conditions, a scale-up pilot test system was developed with 10 liters capacity (Fig. 4). The scale-up system was designed based on the jar-test optimized conditions. The materials used for the scale-up pilot test system development are described as follows:

(i) 10 litter buckets: 3 pieces of 10 litter buckets for main tank, rapid mixing tank, slow mixing and settling tank.

(ii) Plastic water pipes: to transfer water from one tank to another.

(iii) Basin: to store the final treated water.

(iv) Solenoid valves: time monitored valves to allow water retention and flow from one tank to another.

(v) Stepper motors: for accommodating the rapid and slow mixing conditions.

(vi) Motor drivers: to drive the mixing motors to rotate with the desired rates.

(vii) Arduino software: to program and run the code for accommodating the optimized conditions in the developed system.

(viii) Standing desks: wooden desks of different heights to allow gravity flow of water from one tank to another.

(ix) Pilot-test configuration: step-by-step

Three 10-litre tanks, each with solenoid valves and two with mixing motors were connected one after another with plastic water pipes to facilitate continuous coagulation-flocculation-sedimentation process. The three tanks are named as Main Tank (Tank 1), Rapid Mixing Tank (Tank 2) and Slow Mixing Tank (Tank 3). Main Tank has a solenoid valve which is monitored to allow passage of 10 liters of water to the rapid mixing tank. The second (Rapid Mix Tank) consists of small hole for manual chemical addition, motor for flash mixing and a solenoid valve to allow the water to pass to the slow mixing tank upon completion of the rapid mixing stage. The third (slow mixing tank) consists of motor for slow mixing purpose and a solenoid valve which opens to allow water to pass to the last treated water storage basin. The sedimentation stage takes place in the slow mixing tank where the water is provided sufficient time to settle after the slow mixing motor stops rotating for a certain time. Finally, the water is allowed to transfer to the final treated water receiving basin. The mixing rates, mixing time and the duration of water flow from one tank to another are controlled through the Arduino programming software. The system was designed using a SolidWorks software and presented in Fig. 4.

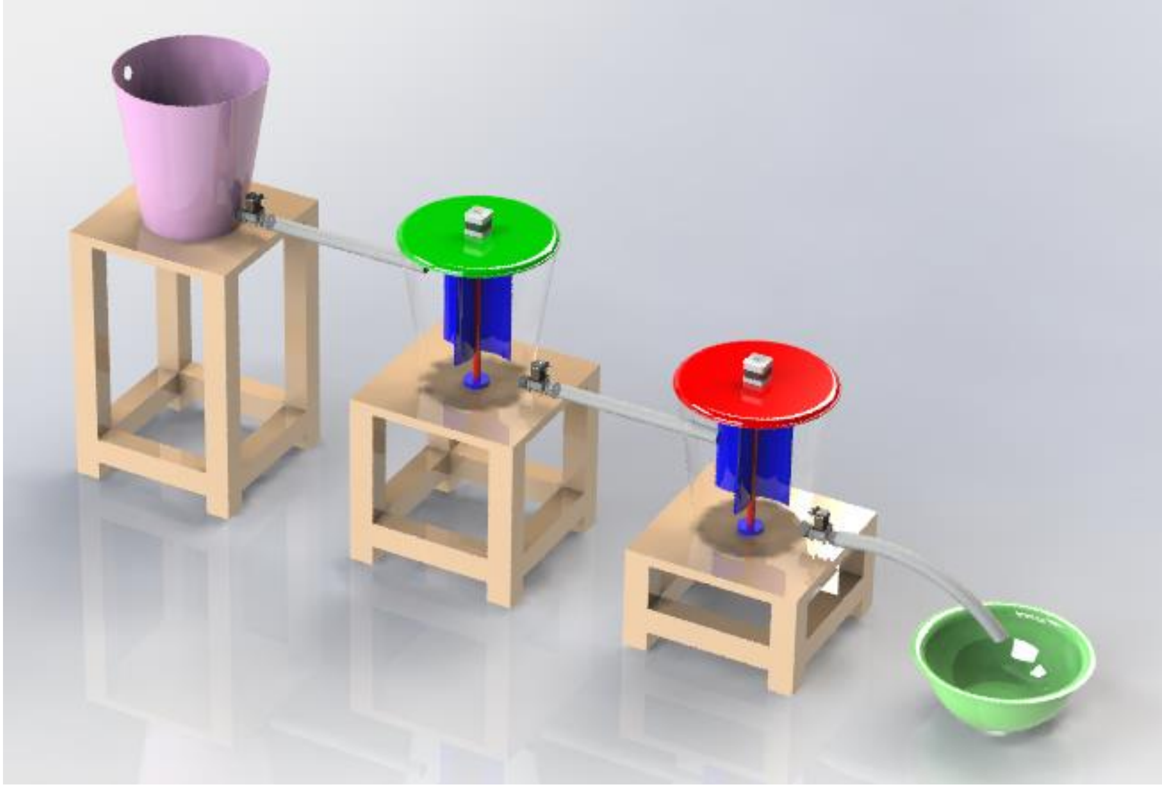


Figure 4: **Design of the scale-up coagulation-flocculation-sedimentation pilot plant**

The pilot plant Showing raw water receiving main tank (pink tank), rapid mixing tank with rotating motor and impeller (green-lidded tank), slow mixing tank with rotating motor and impeller (red-lidded tank), and treated water receiving basin (green basin), with water flow pipes connected from one tank to the other. The wooden desks hold the tanks at varying heights to maintain gravity water flow.

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Water Quality Assessment of Kou River

Tables 5 and 6 show the water quality results for the Kou River and the recommended limits for the various purpose-specific criteria for each water quality parameter analyzed. The Kou River meets the SEQ-Eau minimum limits for DO, EC, pH, temperature, and turbidity of biological life (Table 5). However, as with the other criteria used, some TMEs in the Kou River water showed higher concentrations than recommended (Table 6).

Eight TMEs (Zn, Cr, Co, Ni, Fe, Mn, As and Cu) with concentrations ranging from 0.0043 to 5.48 mg/L were detected in DUS and DDS stations. All TMEs except Co exceeded the NQEp standard limit for biological life. The Mn (0.28 & 0.27 mg/L), Zn (0.56 mg/L), Cu (0.29 mg/L & 0.91 mg/L), Ni (0.04 mg/L & 0.1 mg/L), Fe (4.12 mg/L & 5.09 mg/L) concentration far exceeded the NQEp standard. Among the detected TMEs, Zn, Ni and Cu showed a significant increase beyond the biological lifespan cutoff of SEQ-EAU.

Under USEPA standards, Cr, As, Pb and Cu were allowed for aquatic organisms at the two Kou River stations, but Ni, Mn, Co, Fe and Zn exceeded their limits. Regarding the use of the river water for agriculture, only Co (0.46 mg/L & 0.57 mg/L), Fe (5.09 mg/L) and Cu (0.91 mg/L) are above acceptable levels of metals. Thus, there is possibility of acceptability of Kou River water for irrigation in terms of TMEs concentration exclusive of Fe and Co.

Table 5: **Physicochemical properties of Kou River water and its tributary with their biological life recommended limits in SEQ-Eau quality system**

Stations	**Hardness (mg/L)**	**Temperature (°C)**	**EC (µS/cm)**	**DO (mg/L)**	**Turbidity (NTU)**	**TDS (mg/L)**	**pH**
Main river							
S1(DUS)	15.48±1.5	22.3±0.2	59.4±0.0	7.21±0.1	16.0±0.2	38.0±1.9	6.8±0.2
S2	17.87±1.7	23.8±0.1	58.9±0.3	7.49±0.2	14.0±0.8	37.7±0.6	6.8±0.1
S3	19.10±0.2	22.6±0.1	58.0±0.2	7.13±0.3	10.7±1.2	37.1±2.5	7.3±0.1
S4	19.10±0.1	22.3±0.4	63.5±0.0	7.32±0.2	16.0±0.5	40.6±2.1	6.9±0.1
S5	21.07±1.6	22.8±0.0	60.0±0.1	7.12±0.3	18.3±1.8	38.4±1.4	6.9±0.0
S6 (DDS)	21.07±1.6	24.4±0.2	64.9±0.0	6.91±0.2	13.7±0.9	41.5±0.5	6.3±0.0
S7	21.80±1.1	23.2±0.0	62.6±0.0	6.95±0.3	13.0±0.9	40.1±1.1	6.7±0.2
Average	19.07±1.4	23.06±0.6	61.04±1.8	7.16±0.1	14.52±1.69	39.06±1.1	6.83±0.21
Tributary							
S8	48.37±1.6	23.2±0.4	119.0±0.0	7.12±0.3	5.0±0.9	76.1±0.6	7.4±0.0
S9	41.77±1.5	21.5±0.2	95.8±0.2	6.79±0.2	18.3±1.9	61.3±0.5	6.9±0.1
S10	40.63±1.5	21.8±0.3	82.2±0.3	6.77±0.1	16.7±1.4	52.6±1.2	6.8±0.1
S11	36.15±1.5	22.7±0.5	96.9±0.0	6.70±0.1	13.0±1.2	62.0±1.9	6.9±0.1
Average	41.72±4.28	22.3±0.7	98.5±12.9	6.85±0.2	13.26±5.0	63.0±8.3	6.98±0.2
SEQ-Eau	-	21.5-25.5	3000	>6	35	-	6-9

Table 6: **TMEs concentration (mg/L) in Kou River water and their recommended limits in SEQ-Eau biological life, NQEp surface water, irrigation USEPA aquatic life quality standards water**

TMEs (mg/L)	**S1 (DUS)**	**S6 (DDS)**	**Agriculture (Ayers & Westcot, 1995; Maleki *et al.*, 2013)**	**NQEp**	**SEQ-Eau (Khaled-Khodja *et al.*, 2018)**	**Aquatic life (USEPA, 2014)**
Fe	5.090±0.4	4.120±0.5	5.000	0.20	N/A	1.000
Zn	0.560±0.1	0.560±0.0	2.000	0.008	0.002	0.120
Mn	0.270±0.0	0.2800±0.0	0.200	0.050	N/A	0.100
Cu	0.910±0.0	0.290±0.0	0.200	0.001	0.001	<2.000
As	0.008±0.0	0.008±0.0	0.100	0.004	0.010	0.150
Pb	BDL	BDL	5.000	0.0004	0.005	0.025
Cr	0.006±0.0	0.004±0.0	0.100	0.003	0.002	0.050
Ni	0.100±0.0	0.040±0.0	0.200	0.002	0.003	0.052
Co	0.570±0.1	0.460±0.0	0.050	N/A	N/A	0.004

N/A = guideline not available. BDL = Below detection level of the machine (0.0001 mg/L).

Among all metals detected, the only metal that was beyond all the three standard guideline values for biota, aquatic life and agriculture was Fe (Table 6). Moreover, it was among the three metals (Zn, Cu and Fe) that exceed their recommended biological life standard limits by more than 25 times. Fe concentration along the main river ranged from 4.1 mg/L to 5.38 mg/L, and 2.12 mg/L to 3.44 mg/L in the unnamed tributary (Table 6). This level was comparatively higher than the standard values set by USEPA aquatic standard (1 mg/L), agriculture water standard (5 mg/L) and NQEp for surface water quality (0.2 mg/L). Therefore, Kou River water was considered unsuitable due to the presence of Fe, especially in supporting aquatic and biological life.

The notable water quality deterioration of the Kou River with Fe can be traced back to the natural geological formations of the area or possibly to human activities. The Fe levels in excess of the recommended limit have been reported elsewhere and it has been stated that the TMEs may originate from geological deposits, domestic waste, agricultural and industrial activities (Wogu & Okaka, 2011; Akhtar *et al.,* 2021). A detailed study of sediments can identify metals contamination in water and stipulate their main sources, as sediments can act as an adsorbent stage for various substances, including TMEs, providing clues to the hydro-chemical activities behind the studied water features (Namieśnik & Rabajczyk, 2010). Therefore, in this study, the major TMEs and their probable sources were identified through in-depth sedimentary investigation.

4.2 Sediment Quality and Contamination Assessment

4.2.1 Quality Assessment Using Standard Guidelines

The list of metals such as Ni, Zn, Fe, Pb, Mn, Cr and Cu were examined in Kou sediments for the purpose of quality and risk evaluation. In relation to the SEQ-EAU guideline (Table 7), the sediments have been suitable for supporting biological life in terms of their Pb, As, Zn and Cu concentrations. However, it's Ni (38.32 mg/Kg & 39.72 mg/kg) and Cr (213.6 mg/kg & 176.6 mg/kg) concentrations were not acceptable at DUS and DDS stations respectively. The levels of Mn (896.6 mg/kg and 915 mg/kg) and Fe (51698 mg/kg and 52950 mg/kg) were outrageous; nonetheless, the SEQ-Eau standard does not have guideline value for these factors (Khaled-Khodja *et al.,* 2018). On the contrary, the US-EPA guideline classified the sediments as "non-contaminated" for Zn, Cu and Pb, and "slightly contaminated" for Ni. The metals such as Fe, Cr, and Mn were classified as "severely contaminated" in reference to this guideline (Table 8).

Thus, depending on the SEQ-Eau and USEPA criteria, the river sediments are contaminated with TMEs particularly Fe, Ni, Cr and Mn. Recall that among the four metals, Fe at elevated levels than all recommended levels have also been recorded in the Kou water, probably pointing out a common source.

Table 7: The concentration of TMEs in Kou River sediments (mg/kg) and SEQ-Eau biological life recommended limits

	Cu	As	Cr	Fe	Zn	Ni	Pb	Mn	Co
DDS	23.06	1.40	**176.60**	52950.00	71.72	**39.72**	23.30	915.00	32.88
DUS	24.66	1.42	**213.60**	51698.00	65.96	**38.32**	23.40	896.60	29.78
SEQ-Eau	32.00	9.80	43.00	N/A	120.00	22.00	35.00	N/A	N/A

Table 8: The concentration of TMEs in Kou River sediments (mg/kg) and USEPA sediment quality classification

TMEs	No Contamination	Sligh Contaminated	Severe Contaminated	DUS	DDS
Zn	<90	90 – 200	>200	65.96	71.72
Cr	<25	25 – 75	>75	213.60	176.60
Fe	<17 000	17 000 – 25 000	>25 000	51 698.00	52 950.00
Cu	<25	25 – 50	>50	24.66	23.06
Ni	<20	20 – 50	>50	38.32	39.72
Mn	<300	-	>500	896.60	915.00
Pb	<40	40 – 60	>60	23.40	23.30

4.2.2 Contamination Assessment Using Pollution Indices

Assessing sediment contamination using various pollution indicators provides a better outlook on the main sources of metals and the associated ecological risks. Therefore, exposure to TMEs and potential ecological risks in Kou sediments were evaluated using six pollution indicators. Detailed explanation of the indices is provided in Table 7. The next section presents the results of the analysis of the six pollutant indices in reference to Table 9.

(i) Geo-Accumulation Index

The Geo-accumulation Index (I_{geo}), an indicator of sediment load by TMEs, classifies sediment pollution level into 7 classes, from non-polluted to extremely polluted (Kowalska *et al.*, 2018). The calculated I_{geo} of Mn (0, 18 & 0.21), Co (0.78 & 0.92), Ni (0.46 & 0.51), Fe (0.16 & 0.19) and Cu (0.2 & 0.1) in DUS and DDS stations respectively indicated unpolluted to moderately polluted status ($0 \leq I_{geo} < 1$) (Table 5). The I_{geo} values for As, Zn, and Pb were less than zero. An I_{geo} value below zero represents uncontaminated status (Shilla & Shilla, 2021). This may indicate that there was no contamination for the three TEMs. In contrast, moderate to heavy Cr loading was observed in sediments of DUS and DDS stations. Mining activities; ashes from combustion of coal or municipal wastes; and fertilizers manufacturing are typical anthropogenic sources of the metal in water and sediments (Tumolo *et al.,* 2020).

(ii) Contamination Factor

Contamination factor (CF) is an indicator for monitoring individual metal contamination by comparing its concentration to the metal's background value (Shilla & Shilla, 2021). The CF of Mn, Pb, Ni, Cu, Fe, Zn and Co was observed as moderate ($1 \leq CF < 3$) at both stations. Of all the metals, the CF value for Cr was the highest with CF of 6.1 at DUS (very high contamination for CF>6) while the DDS-Cr contamination grade (CF=5.05) was considerable ($3 \leq CF < 6$). The minimum CF (CF>1) was identified for As at both DDS (CF=0.7) and DUS (CF=0.71). It was noteworthy that the contamination category of TMEs provided by CF was found to be higher than that of the Igeo category. This result coincided with other literatures that suggesting that CF values often provide higher metal contamination status than the Igeo Counterparts (Hołtra & Zamorska-Wojdyła, 2020).

(iii) Enrichment Factor

The degree of anthropogenic effects on the abundance of TMEs in sediment is identified by calculating the enrichment factor (*EF*) (Rezaei *et al.,* 2019). With reference to Table 6, it was observed that the man-induced enrichment of Cu, Mn and Fe was minimal in sediments of DUS and DDS. The *EF* values for As, Zn, and Pb were less than 1.5, which suggest that their presence may be due to natural processes. The Co's *EF* values at the two stations were 2.48 and 2.96 and *EF*s between 2 and 5 were considered moderate. The Ni was observed with moderate enrichment in DDS sediments (*EF* = 2.23), while minimum in DUS sediments (*EF* = 1.99). Significant effects that may be associated with anthropogenic activity ($5 \leq EF \leq 20$) were observed for Cr with *EF* 5.27 and 5.9 at the two stations. Coal incineration plants, municipal waste; mining activity and wastes from tanning industry are typical anthropogenic sources of Cr in surface waters (Abadin *et al.*, 2012).

Table 9: The concentration of TMEs in Kou River sediments (mg/kg) and their respective sediment contamination indices calculated by the present study

	Al	**Mn**	**Cr**	**Cu**	**Pb**	**Co**	**Ni**	**Zn**	**Fe**	**As**
DUS	80146.00	896.60	213.60	24.66	23.40	29.78	38.32	65.96	51698.00	1.42
DDS	74168.00	915.00	176.60	23.06	23.30	32.88	39.72	71.72	52950.00	1.40
UCC^a	77440.00	527.00	35.00	14.30	17.00	11.60	18.60	52.00	30890.00	2.00
I_{geo}(DUS)		0.18	2.02	0.20	-0.12	0.78	0.46	-0.24	0.16	-1.08
I_{geo} (DDS)		0.21	1.75	0.10	-0.13	0.92	0.51	-0.12	0.19	-1.10
$CF_{(DUS)}$		1.70	6.10	1.72	1.38	2.57	2.06	1.27	1.67	0.71
$CF_{(DDS)}$		1.74	5.05	1.61	1.37	2.83	2.14	1.38	1.71	0.70
$EF_{(DUS)}$		1.64	5.90	1.67	1.33	2.48	1.99	1.23	1.62	0.69
$EF_{(DDS)}$		1.81	5.27	1.68	1.43	2.96	2.23	1.44	1.79	0.73
E_r^i(DUS)		1.70	12.20	8.62	6.88	12.83	10.30	1.27	N/A	7.10
E_r^i(DDS)		1.74	10.09	8.06	6.85	14.17	10.68	1.38	N/A	7.00
$mCd_{(DUS)}$	2.13									
$mCd_{(DDS)}$	2.06									
$RI_{(DUS)}$	60.92									
$RI_{(DDS)}$	59.97									

UCC values were obtained from Wedepohl (1995). ^{a}UCC is background value of metals taken from the upper continental crust.

(iv) Modified Degree of Contamination

Modified contamination degree (mCd) is a tool that establishes the overall contamination level for a specific site depending on the Contamination Factor (CF) of the individual TMEs in sediments (Kowalska *et al.*, 2018). After calculation of the CFs of each metal at the two sampling stations, i.e. Co (2.57 & 2.83), Pb (1.38 & 1.37), As (0.71 & 0.7), Cr (6.1 & 5.05), Cu (1.72 & 1.61), Zn (1.27 & 1.38), Ni (2.06 & 2.14), Fe (1.67 & 1.71), As (0.71 & 0.7) and Mn (1.7 & 1.74), DUS and DDS stations had mCd values of 2.13 and 2.06 respectively. mCd between 2 and 4 indicates moderate pollution (Shilla & Shilla, 2021). Thus, a moderate contamination of the river with the metals was observed. Alike the results from I_{geo} and CF, Fe, Co, Ni, Cu, Cr and Mn were the major elements in charge of the overall sediment pollution.

(v) Potential Ecological Risk

Ecological risk index (E_r^i) for the nine TMEs in the sediments was calculated based on each metal's ecological risk factor using toxicity response coefficients (T_r^i) of each metal (Hakanson, 1980). The E_r^i classifies the ecological risk of metals as very high ($E_r^i \geq 320$), high ($160 \leq E_r^i < 320$), considerable ($80 \leq E_r^i < 160$), moderate ($40 \leq E_r^i < 80$) and low ($E_r^i < 40$). The results of this study (Table 6) showed that all the nine TMEs used in the calculations were categorized under low ecological risk class ($E_r^i < 40$). The extent to which the presence of total TMEs in sediments poses ecological risk was determined by the Potential Ecological Risk Index (RI) using the total E_r^i values of all TMEs. This index groups the risk levels of the TMEs as high ($RI \geq 600$), considerable ($300 \leq RI < 600$), moderate ($150 \leq RI < 300$) and low (RI<150), (Nkinda et al., 2020). The *RI* of sediments from DUS (60.92) and DDS (59.70) showed a low potential ecological risk of the existing metals (other than Fe).

With the exception of Mn and Fe, the classifications of the contamination indices and the US EPA guideline were consistent. They all provided the sediments a no to moderate contamination status with Ni, Co, Zn, As and Cu whereas a considerable to severe contamination status with Cr. The Mn and Fe contamination level was no-to-moderate according to the pollution indices; however, a severe contamination by the two metals as well as Cr was indicated by the US EPA guideline. Although the Kou River sediments contained high concentrations of those metals, Fe was the only metal present at concentrations well above all recommended limits used for water quality assessment. Despite Cr considerably contaminating the river sediments, it was only slightly concentrated in the overlying water, probably as a result of its insoluble trivalent form.

In reference to the ecological risks of metals, the standard guidelines were inconsistent with the Ecological Risk Index ($E^i{}_r$). The US EPA and SEQ-Eau standards explain that Kou River sediments are inadequate for sustaining biological life in terms of Mn, Ni and Cr, but the ecological risk index indicated that the three metals to pose ecological risks is unlikely.

The US EPA guideline similarly indicated severe Fe contamination, but neither the $E^i{}_r$ index nor the SEQ-Eau standards established ecological risk levels for the metal. It is noteworthy that the US EPA's criteria classifies metals in sediments based on their capability to negatively affect overlying water quality (US EPA, 1998). Therefore, the heavy Fe load in the sediments and the undesirable Fe levels in water than the biological life, aquatic life and irrigation water recommended limits could indicate the metal's introduction from the sediments to the water and vice versa. Further experiments were conducted to determine its leachability and bioavailability in order to gain a deeper understanding of the potential ecological risks of Fe in sediments.

4.2.3 Fe Enrichment and Impacts

(i) Bioavailable Fractions of Fe

Analysis of total metal content is not considered a reliable indicator of their toxicity. By distinguishing different forms of the element in terms of physicochemical mobility and potential bioavailability, it sheds light on the state of pollution and its real impact on the environment (Olumayede & Ediagbonya, 2018). The Fe in the sediment was sequentially extracted into 6 categories of individual remobilization regimes under different environmental conditions. The six fractions were water soluble (F1), ion exchangeable (F2), carbonate binding (F3), Fe-Mn binding/reducing (F4), organic and sulfide binding/oxidizing (F5) and the residual fraction (F6).

Two stations were selected, upstream (Station 1) and downstream (Station 6), which is the water collection point for domestic supply, to study iron fractionation in river sediments. The proportions of the different Fe fractions in DUS and DDS sediments are provided in Table 10.

(ii) Water-Soluble Fraction (F1)

The water-soluble fraction is a dissolved fraction of the metal and is reported in most references as part of the interchangeable fraction. In this study, it accounted for only about 0.03% of the total iron content in river sediments (Table 10). Osakwe in his study also which showed that the proportion of Fe in water-soluble species was negligible, probably due to the easy uptake of the metal by the aquatic life (Osakwe, 2013).

(iii) Ion Exchangeable Fraction (F2)

These fractions are weakly bound metal species on sedimentary particles that can be separated through electrostatic interaction by altering the ion arrangement of water. The sorption-desorption process is governed by the ionic composition of the metal. The morphology adsorbed on the surface of the sediment is easily removed by this method, mainly by ion exchange. Under acidic conditions, the release of these fractions contributes to adverse environmental effects (Giraldo, 2018). The river sediments of this study contained only 0.01% ion-exchangeable iron which accounted the smaller portions than all other fractions (Table 10).

(iv) Carbonate Bound (F3)

This fraction is usually considered to be poorly absorptive. The metal in this fraction is bound to carbonate, phosphates, and sulfates. The F1 and F2 fractions are primarily mobile and biologically available, while the F3 fractions are more susceptible to certain environmental variations, most often acidic conditions (Li *et al.,* 2016). In this study, 0.22% and 0.01% Fe in the F3 form were observed at DDS and DUS stations.

(v) Fe-Mn Bound/Reducible Fraction (F4)

These fractions are not stable in environments of low redox potential (Eh). They are reduced under anaerobic conditions, facilitating metal adsorption during decomposition (Thomas, 2015). Since the two TMEs are common constituents of rocks and sediments, the concentrations of Fe and Mn are usually higher in the F4 fraction (Devi & Bhattacharyya, 2018). The Fe in this fraction (6.59% and 4.96%) in DDS and DUS sediments was the major fraction following the residual fractions.

(vi) Organics And Sulfide Bound/Oxidizable Fraction (F5)

The fractions in the F5 form are usually bound to organic and sulfide compounds. Environmental aerobic / anaerobic conditions play a major role in the oxidation of sulfides. This is directly related to the mobility of the metal fraction bound to the oxidizable phase (Lian Svendsen *et al.,* 2011). The metal in this fraction is likely to be released in an oxidized state as a result of mineralization, which later causes complexation and in accumulation in cells. In the case of DDS and DUS, Fe in the F5 fraction accounted for 0.87% and 2.74% of the total metal content, respectively.

(vii) Residual Fraction (F6)

The residual fraction is the remaining metal that is bound, immobilized, biologically inert by crystal structure and has no risk of environmental pollution (Borgese *et al.,* 2013). A high proportion (92%) of Fe was detected in this fraction, indicating a strong immobile bond in the lattice structure of the sediment.

Overall, the six Fe forms in the Kou River sediments followed the trend of residues (F6), Fe-Mn bound (F4), organic bound (F5), carbonate bound (F3), water soluble (F1) and ion exchangeable (F2). The first three Fe fractions (F1, F2 and F3) were insignificant but the reducible fraction after the residue was the predominant fraction. Higher reducible fractions than other weakly bound fractions indicate that the solubility of Fe in sediments was more closely associated with reduced environmental conditions. These observations are in good agreement with some findings that Fe content is often low in more soluble and exchangeable fractions (Klink *et al.*, 2019).

Table 10: Results of the sequentially extracted fractions of Fe (mg/kg and percentage) in the Kou River sediments

	Downstream (DDS)		Upstream (DUS)	
Fractions	**Fe fraction (mg/kg)**	**Percentage (%)**	**Fe fraction (mg/kg)**	**Percentage**
Water soluble (F1)	9.79	0.02	16.46	0.03%
Ion exchangeable (F2)	2.92	0.01	4.17	0.01%
Carbonate bound (F3)	118.96	0.22	5.42	0.01%
Reducible (F4)	3491.67	6.59	2562.50	4.96%
Oxidizable (F5)	458.33	0.87	1416.67	2.74%
Residual (F6)	48 868.30	92.29	47 694.80	92.25%
Total amount	52 950.00		51 698.00	

Bioavailability Factor

The bioavailability factor (BF) is the ratio of the content of the potentially mobile metal fraction to the total content of metallic elements (Al-Mur, 2020). The BF is calculated using Equation (3):

$$BF=\frac{C_{bio}}{C_{total}} \tag{3}$$

Where C_{bio} and C_{total} represent the concentration of potentially biologically available metals and the total concentration of metals in the analyzed deposits, respectively. In this regard, all non-residual fractions are considered to be biologically available forms because they are capable of being consumed by the organism (Al-Mur, 2020). The calculated BF of the river sediments was about 7.8%, indicating that about 92% of the total metal content is unlikely to seep into the environment. This indicates that sediments are unlikely to contribute significantly to the excess presence of iron in the river water.

Risk Assessment Code

The Risk Assessment Code (RAC) is a numerical expression used to assess the health risk of metals in sediments to living organisms. The classification is based on the metal percentages of the first three fractions (water-soluble, ion-exchangeable, and carbonate-bonded). These fractions are considered together with very weakly bound fractions and are readily available for biological uptake (Li *et al.*, 2018). According to this expression, a value less than 1% is harmless and is considered a very high environmental risk from 50%. Table 11 shows the interpretation of RAC and the results at the two stations.

Table 11: Interpretation of the Risk Assessment Code (RAC) based on the first three fractions of TMEs (water soluble, ion-exchangeable and carbonate bound) and the calculated Fe (%) results in the Kou River sediments

Risk Assessment Code (RAC)	Criteria (F1+F2+F3) (%)	DDS Fe (%)	DUS Fe (%)
No risk	<1	0.25	0.05
Low risk	1-10		
Medium risk	11-30		
High risk	31-50		
Very high risk	>50		

In reference to Table 11, the RAC value of Fe in the river sediments is less than 1%. Fe in the sediments has been shown to have an impact on ecosystem health based on USEPA sediment quality standard, but RAC suggests that it poses no environmental risk.

Leaching Kinetics of Fe

The TME's mobility and toxic effects in sediments may be related to their leaching kinetics and give information on the rate and extent of metal release with changes in natural conditions (Gismera *et al.,* 2004). Studies have used various chemical leachates such as acid rain to determine the leaching potential of elements with time. The time that is required for an equilibrium to occur can be identified by kinetic experiments (Lee *et al.*, 2012).

In this study, the tendency of release of Fe from the Kou River sediments was analyzed at various times (1 hour, 2 hours, 3 hours, 6 hours, 12 hours, 24 hours, 2 days, 3 days, 6 days, 9 days, 12 days, 15 days, 18 days, 21 days, 24 days 27 days and1 month) using natural rainwater with pH 6.2 ± 0.1 (Fig. 5). Experiments were performed using samples from the DDS station. The total Fe concentration in the experiment was 52,950 mg/kg. During the experiment, the stirring speed was set to 30 rpm.

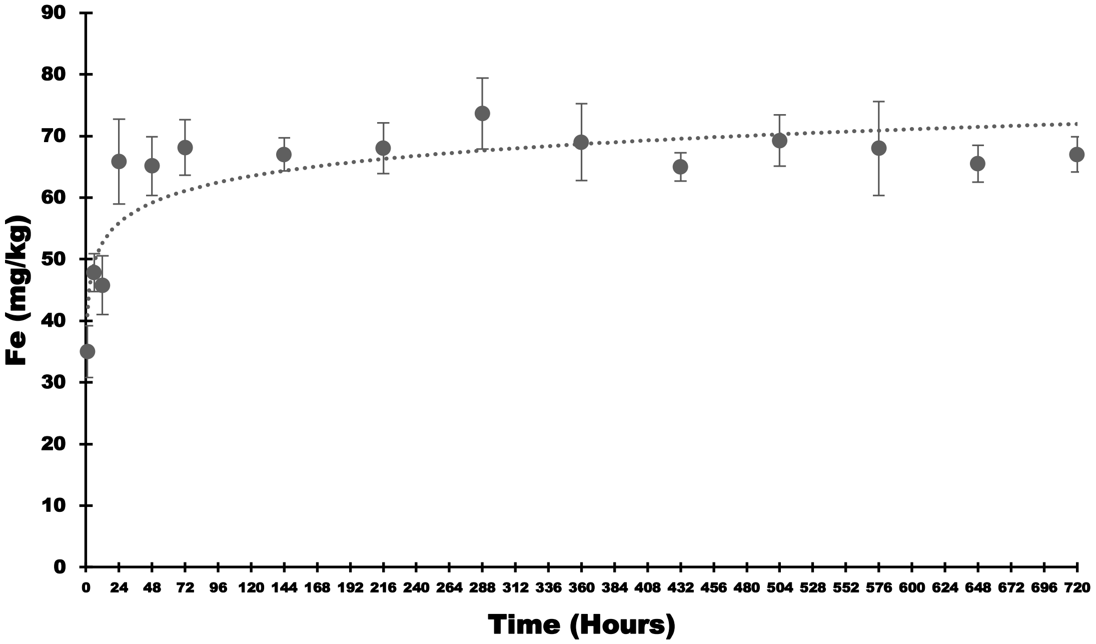


Figure 5: Kinetics of iron leaching from the Kou River sediment using rainwater

After the first one hour, 35 mg/kg Fe was released from the sediment (Fig. 5), which corresponds to 0.07% of the total metal concentration. A sharp increase has been seen after six hours of leaching with 47.8 mg/Kg Fe release (0.09%) where the pH dropped from 6.2 to 6.12. In the 12-hour experiment, there was little difference from the 6-hour extraction (45.85 mg/kg). The largest increase occurred in 24 hours, releasing 65.9 mg/kg (0.12%) of iron and lowering the pH to 6.09. After 24 hours, no significant difference was observed until 288 hours (12 days) of extraction. Maximum leaching (0.14%) was achieved at final pH 6. Fe leached after day 12 to 1 month showed no increase (0.12% to 0.13%) when the pH was maintained in the range of 6.0 to 6.13. This suggests that in situations where a heavy rain induces a pH change in the river, most of the soluble Fe phase will probably be leached from the sediment in the first 24 hours and may continue to be released to some degree up until days contact.

4.2.4 Trace Metallic Elements Concentration in Kou River and Other Study Areas

The TMEs concentration in Kou and other rivers water and sediments have been provided in Table 12 for visualizing the TMEs pollution level of Kou River as compared to other rivers in different parts of the globe.

Table 12: Maximum concentration of TMEs in water (mg/L) and sediment (mg/kg) of Kou River and other rivers worldwide

	Rivers	**Kou (Tanzania)**	**Buriganga (Bangladesh)**	**Bartin (Turkey)**	**São Carlos, Urban (Brazil)**	**Voghji (Armenia)**	**Mvudi (South Africa)**
Ni	Water	0.100	0.400	0.008	0.069	0.003	-
	Sediment	39.72	59.00	31.00	43.04	18.80	-
Fe	Water	5.380	2.200	0.480	3.040	0.376	5.070
	Sediment	52950.00	80.20	29600.00	2508.23	20018.00	7460.00
Cr	Water	0.006	0.180	0.003	0.049	0.001	0.593
	Sediment	213.60	60.50	25.60	76.88	11.80	175.00
Zn	Water	0.560	0.990	0.010	0.410	0.100	0.548
	Sediment	71.72	60.50	94.00	190.65	92.30	1524.00
Cu	Water	0.910	0.990	0.004	0.029	0.083	0.567
	Sediment	24.66	85.20	42.00	90.90	620.00	5690.00
Co	Water	0.570	0.400	NA	0.110	0.003	-
	Sediment	32.88	11.28	10.30	17.50	13.80	-
Mn	Water	0.280	0.310	0.040	0.230	0.181	0.675
	Sediment	915.00	26.40	683.00	275.53	450.00	2160.00
As	Water	0.008	0.220	NA	-	0.004	-
	Sediment	1.42	26.40	6.17	-	29.70	-
Pb	Water	NA	0.521	NA	0.230	0.0005	0.046
	Sediment	23.40	80.20	21.00	28.29	40.50	7.95
References		Present study	(Bhuiyan *et al.*, 2014)	(Gunes, 2021)	(Chiba *et al.*, 2011)	(Gabrielyan *et al.*, 2018)	(Edokpayi *et al.*, 2016)

4.3 Optimization of Coagulation-Flocculation-Sedimentation System

4.3.1 Kou River Water Characteristics

TMEs and other water quality parameters of water collected from the Magugu water supply collection point were identified to determine for which parameter's removal the coagulation-flocculation-sedimentation system needs to be optimized. The TMEs and some physicochemical characteristics were analyzed by the present study while additional data were obtained from the BAWASA Head of water quality service. The water samples for this purpose have been collected on February 3rd 2022, where a heavy rain has been encountered in the area before sampling. The Kou water characteristics along with their respective TBS and WHO drinking water recommended limits are presented in Table 13 (WHO, 1984; TBS, 2003).

Table 13: Results of the physicochemical parameters of Kou River water and their respective WHO and TBS treated water recommended limits

TMEs	Fe	Zn	Mn	Cu	As	Pb	Cr	Ni	Co	Al
DDS	3.640	0.460	0.200	0.490	0.004	BDL	0.004	0.007	0.580	0.000
WHO	0.300	5.000	0.400	2.000	0.010	0.010	0.050	0.070	-	0.100
TBS	0.300	5.000	0.500	1.000	0.010	0.010	0.050	0.020	1.000	0.200

Others	Color (PtCo)	Turbidity (NTU)	pH	EC µS/cm	TDS mg/L	Temp. (°C)	Alkalinity (mg/L)	Total hardness (mg/L)
DDS	14	87.0	7.7	78.5	38	26	47.6	38.6
WHO	15	5.0	6.5-8.5	1400.0	500	20-35	200.0	500.0
TBS	15	5.0	5.5-9.5	2000.0	1000	20-35	-	300.0

Others	Phosphate (mg/L)	Calcium (mg/L)	Fluoride (mg/L)	Nitrate (mg/L)	Sulfate (mg/L)	Chloride (mg/L)	Magnesium (mg/L)	Nitrite-N (mg/L)
DDS	1.8	9.44	0.18	0.56	11	17.08	3.648	0.003
WHO	-	-	1.50	50.00	-	250.00	-	1.000
TBS	2.2	150.00	1.50	45.00	400	250.00	100.000	-

As can be seen from Table 13, all the parameters except Fe (3.64 mg/L) and turbidity (87 NTU) were already below the treated potable water recommended limits by both TBS and WHO. The heavy rain before the sample collection was considered as the possible reason for the elevated level of turbidity in the river water, where Fe was linked possibly to geologic formation of the area as explained in the previous section. High level of turbidity in Densu River of Ghana has also been reported as a result of transport of top soil form farming site in to the river through rain water after heavy rain (Karikari & Ansa-Asare, 2006).

Therefore, the coagulation-flocculation-sedimentation water treatment system required to be optimized for Fe and turbidity removal from Kou River water.

4.3.2 Jar-Test Optimization

The natural Kou River water was used for the optimization process throughout the experiment. The raw water had initial concentrations of 3.64 mg/L Fe and 87 NTU Turbidity which are the target impurities to be removed from the water by the coagulation-flocculation-sedimentation process.

(i) Optimal Dose of Coagulant Chemical (Experiment 1)

For Kou River water with 87 NTU turbidity and 3.64 Fe, the performance of eight different alum doses were evaluated to determine the optimum alum concentration. The working conditions of this experiment were as follows:

- Initial pH of 7.7 (River water pH).
- Rapid mixing at 150 rpm for 5 min.
- Slow mixing at 20 rpm for 30 min.
- Settling time provided 15 min.

Alum with concentrations ranging from 2.5 mg/L to 50 mg/L was dosed at different jars and the operating conditions were kept consistent. Poor performances of Fe and turbidity removal (below 35%) were observed with alum doses of 2.5 mg/L, 5 mg/L, 10 mg/L and 15 mg/L (Table 14). The removal efficiency increased dramatically with dose of 20 mg/L where 69% Fe and 60.44% turbidity was removed (Fig. 6). More than 95% Fe and Turbidity were removed with alum doses of 30, 40 and 50 mg/L. However, even though high treatment performances are achieved, careful monitoring on the concentration of residual aluminum in the treated water is essential because of its detrimental effects on health including Alzheimer’s disease. The residual aluminum from the 50 mg/L dosing was 0.236 mg/L while it was recommended to bring down its concentration in treated water to null as much as possible, 0.2 mg/L being the maximum recommended level by EPA and WHO (Lteef & Mahdi, 2018; Krupińska, 2020). Residual aluminum from the 30 and 40 mg/L dosing were acceptable (below 0.2 mg/L) whereas the 5, 10, 15 and 20 mg/L doses resulted in 0.33, 0.53, 0.65, and 0.67 mg/L Al respectively with lower Fe and turbidity removal performances. The reason behind the higher residual aluminum from low dosing was that low doses were insufficient to destabilize numerous particles to coagulate and to form large enough settleable flocs. Rather, small unsettleable flocs might have formed where Al also remained suspended in the supernatant water which counted as residual aluminum. The reverse was true for the higher doses which resulted in more than 95% treatment performance efficiency, showing that large settleable flocs were formed and most of the aluminum was settled out with the flocs, hence resulting in very low residual aluminum in the treated water. A similar result has been reported in a study conducted on Al Zubaidiyah water treatment plant with raw water having 80 NTU turbidity, where 30 mg/L alum dose performed well and the removal efficiency decreased with increasing alum dose beyond the optimum (Jaeel & Zaalan, 2017).

Therefore, the minimum dose which has performed well in the treatment process with acceptable residual aluminum was 30 mg/L. Nonetheless, as applying the list amount of chemical with good treatment efficiency is essential from both economic and health aspects, the concentration of alum just below the 30 mg/L, i.e., the 20 mg/L dose was also considered in the next experiment assuming that its performance may improve at different pH conditions. Therefore, 20 mg/L and 30 mg/L alum was used in the pH optimization experiment.

Table 14: Performance efficiencies of eight different alum coagulant concentrations used in the jar-test dose optimization process in removing Fe and turbidity from the Kou River water

Alum dose (mg/L)	Turbidity removal percentage	Fe removal Percentage	Residual Al	Final pH
2.5	26.10±4.3	31.32±2.3	0.194±0.3	7.5±0.3
5	26.90±2.3	30.77±3.7	0.333±0.0	7.3±0.1
10	28.05±3.1	17.58±4.4	0.534±0.1	6.9±0.1
15	33.68±3.4	24.18±1.9	0.652±0.1	6.8±0.2
20	69.00±1.7	60.44±3.2	0.687±0.1	6.6±0.2
30	98.85±1.0	96.43±1.6	0.107±0.0	6.3±0.1
40	96.30±0.8	97.53±0.9	0.101±0.1	6.1±0.0
50	97.80±0.6	98.63±0.9	0.236±0.1	5.6±0.1

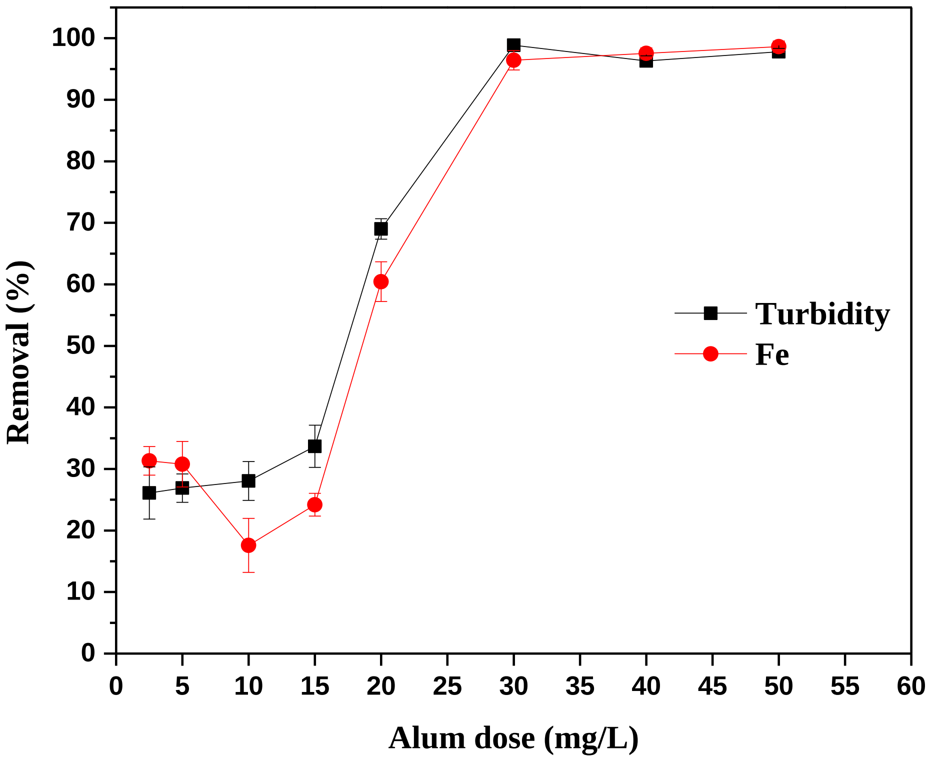


Figure 6: **Determination of the optimum dose of alum for Fe and turbidity removal from the Kou River water using jar-test**

(ii) Optimal pH (Experiment 2)

For this experiment, the optimized dose and working conditions of experiment 1 were applied except pH. The effect of initial pH ranging from 5 to 9 was evaluated at constant doses of 30 mg/L and 20 mg/L alum. At 30 mg/L dose, the removal of both Fe (89.29% & 92.03%) and turbidity (95.37% & 95.12%) was relatively lower at pH 5 and 9 respectively. Turbidity was removed 100% at pH 6, 7 and 8 while Fe removal accounted for approximately 98% (Fig. 7). Although a good performance efficiency at all the five pH levels was observed, the residual aluminum at pH 5 and 6 was too high (0.96 mg/L and 0.47 mg/L) whereas at pH 9, it was slightly higher (0.239 mg/L) than the undesirable level of residual Al (0.2 mg/L) recommended by EPA (Table 15). Therefore, at dose of 30 mg/L, pH approximately between 7 and 8 was deemed optimum to perform effectively in treating the Kou River water. Jaeel and Zaalan (2017) in their study reported that alum generally performs well in the pH range of 5-6, which was in contradiction to the present study. The current study claims that the pH condition suitable for removal efficiency is dependent on the varying characteristic of the water.

Table 15: The effect of initial pH values in the removal of Fe and turbidity from the Kou River water using jar-test with 30 mg/L alum dose

Alum dose (mg/L)	pH	Turbidity removal percentage	Fe removal Percentage	Residual Al	Final pH
30	5	95.37±2.3	89.29±2.8	0.960±0.0	4.7±0.1
30	6	100.00±0.0	97.80±1.3	0.468±0.0	5.2±0.2
30	7	100.00±0.0	98.10±1.4	0.080±0.0	6.1±0.2
30	8	100.00±0.0	97.80±0.2	0.081±0.1	6.5±0.1
30	9	95.12±0.9	92.03±3.4	0.239±0.1	6.8±0.2

Table 16: The effect of initial pH values in the removal of Fe and turbidity from the Kou River water using jar-test with 20 mg/L alum dose

Alum dose (mg/L)	pH	Turbidity removal percentage	Fe removal Percentage	Residual Al	Final pH
20	5	91.65±3.7	86.81±1.6	0.500±0.1	4.9±0.0
20	6	98.66±1.1	95.60±1.9	0.272±0.0	5.7±0.1
20	7	85.37±3.0	80.22±4	0.417±0.0	6.4±0.1
20	8	64.39±4.6	61.26±3.3	0.544±0.0	6.8±0.3
20	9	49.15±1.9	42.86±1.8	0.354±0.1	7.2±0.1

At pH 5 and 6, 20 mg/L alum dose performed similarly with dose of 30 mg/L with elevated residual aluminum, i.e. 0.5 and 0.27 mg/L respectively (Table 16). Turbidity and Fe removal efficiency dropped continuously when pH was raised beyond 6 up to 9 indicating that variation in pH has not improved the performance of the 20 mg/L dosing. The similar performance of the two doses (20 mg/L and 30 mg/L) at pH 5 and 6 (Fig. 7) with elevated residual aluminum could be indicative of the independent effect of low pH on the removal of Fe and turbidity. The effect of pH on Fe and turbidity removal without addition of alum was presented in the later section. Figure 7 shows the effect pH on the removal of Fe and turbidity using 20 mg/L and 30 mg/L alum doses.

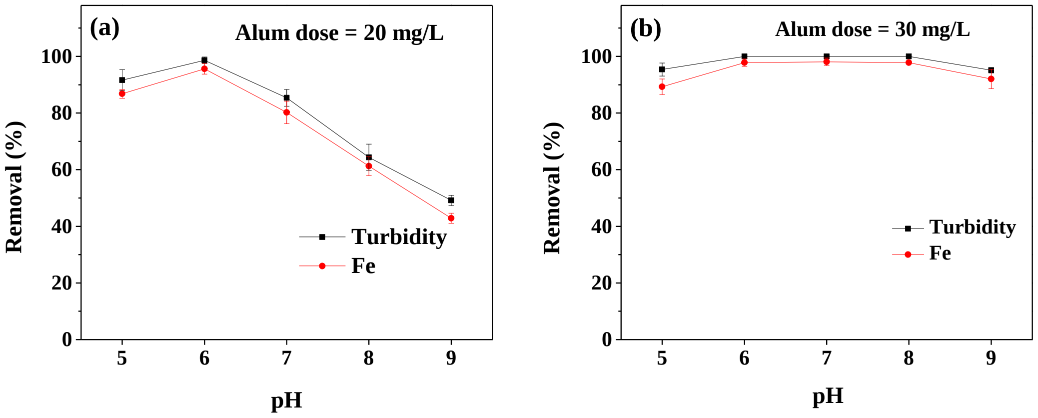


Figure 7: Determination of the optimal initial pH for the removal of Fe and turbidity from the Kou River water using jar-test (a) with 20 mg/L alum dose and (b) with 30 mg/L alum dose

Overall, pH of 5, 6 and 9 were considered unsuitable due to resulting in high residual aluminum. At 6 < pH < 9, dose of 30 mg/L had a higher performance efficiency at wider pH range than 20 mg/L alum. Therefore, dose of 30 mg/L and pH approximately between 7 and 8 (for maintaining minimum residual Al) were considered optimum working conditions. This result contradicted with another study which suggested that alum forms floc in water only at pH between 6.8-7.2 (Krupińska, 2020). This could indicate that the optimum pH range for effective alum performance is dependent on the raw water characteristics.

(iii) Effect of Rapid-Slow Mixing Rate (Experiment 3)

The working conditions to determine the optimum mixing rate were derived from experiment 1 & 2. Alum dose of 30 mg/L was applied while the natural pH of the raw water (pH = 7.7) was maintained as it was in the optimum pH range. The rapid-slow mixing time was kept similar as the above experiments (5 min -30 min). The effects of four separate rapid and slow mixing speeds followed by 15 minutes settling time were studied and presented in Fig. 8 and Fig. 9.

Rapid Mixing Rate

The initial (flash) mixing rates were varied between 50 rpm, 75 rpm, 100 rpm and 150 rpm to see the effect on flocculation. The best floc formation efficiency was obtained at rapid mixing rates of 100 rpm (more than 97%) and 150 rpm (more than 98%) Fe and turbidity removal efficiency. The performance dropped with reduction in mixing rates, i.e. at 75 rpm, about 86% Fe and 94% turbidity was removed while at 50 rpm, only 55% turbidity and 79% The Fe was removed. This indicated that slowing down the initial mixing intensity results in failure to sufficiently distribute the chemical along particles to facilitate full destabilization. Therefore, initial mixing rates with 100 rpm and 150 rpm were optimum stirring conditions which scored higher removal performances. This finding was in agreement with other studies which stated that the higher the rapid mixing speed, the better the floc formation performance (Taşdemir, 2012).

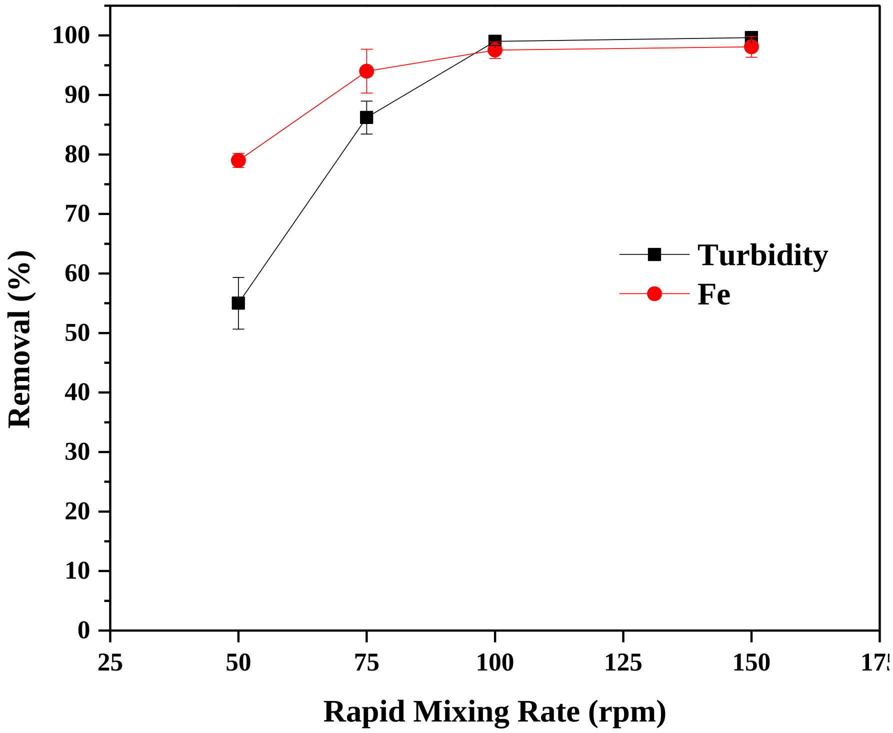


Figure 8: **Determination of the optimum rapid-mixing rate for Fe and turbidity removal from the Kou River water using jar-test**

Slow Mixing Rate

The effect of slow mixing rate on floc formation was studied using four different mixing rates; 10 rpm, 20 rpm, 30 rpm and 40 rpm after the water was initially mixed rapidly at 150 rpm. No significant variation was observed where more than 95% efficiency was achieved among all mixing rates. The least removal efficiency (95% turbidity and 97.7% Fe) was observed at 40 rpm whereas 100% turbidity and 98.8% Fe removal was achieved at 20 rpm making it the optimum slow mixing rate (Fig. 9). In research conducted in 2012, a slow mixing rate at 40 rpm for 15 minutes has been reported to perform well in removing turbidity (Taşdemir, 2012). This contradicted with the finding of the present study which indicated that after the particles destabilization stage, the mixing rate for settleable floc formation should be kept at slow rate as elevated mixing speed results in re-stabilization of particles and unsettleable small flocs formation.

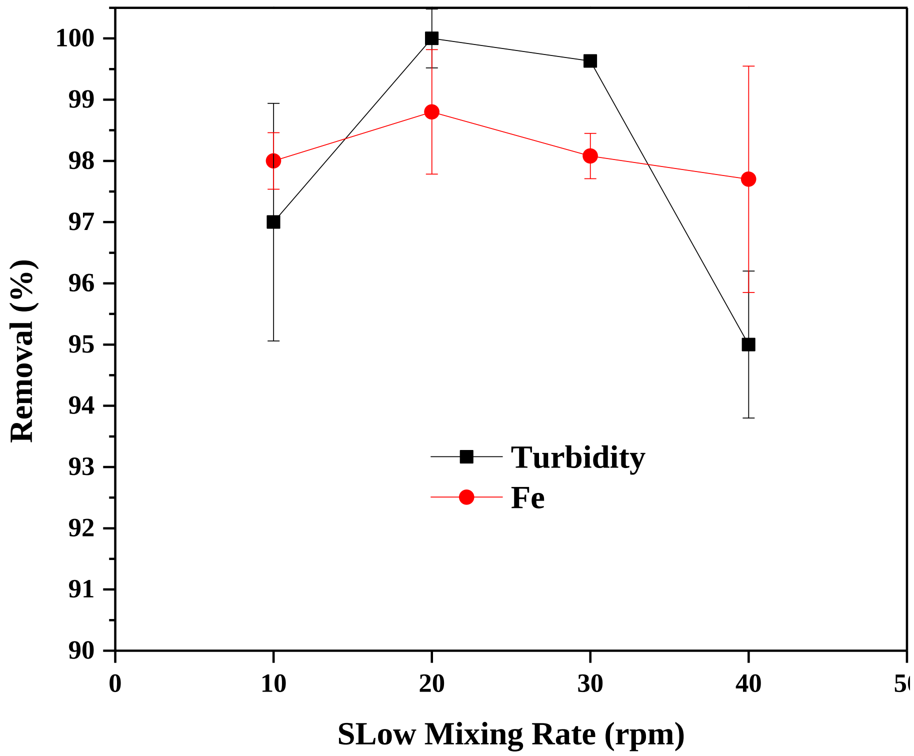


Figure 9: **Determination of the optimum slow mixing rate for Fe and turbidity removal from the Kou River water using jar-test**

Therefore, in the present study, 150 rpm - 20 rpm has been chosen as the optimum rapid - slow mixing rate for Fe and turbidity removal in the coagulation-flocculation-sedimentation water treatment process.

(iv) Effect of Rapid-Slow Mixing Time (Experiment 4)

The effect of rapid-slow mixing time with working conditions; 150 rpm-20 rpm rapid - slow mixing rates, 30 mg/L alum dose, pH of 7.7 followed by 15 minutes settling time was studied. The effectiveness of flash mixing time for 1, 3 and 5 minutes and slow mixing time for 10, 20, 30 and 40 minutes were investigated.

Rapid Mixing Time

The effect of initial mixing time on floc formation was presented in Fig. 10 for 1 min, 3 min and 5 min. As can be seen, the flocculation performance dropped slightly by about 4% Fe & 2% turbidity removal as the flash mixing time was reduced from 5 min to 1 min which could be indicative that the time provided for flash mixing should be sufficient enough to facilitate the desired collision property between particles. The difference between the 5 min and 3 min mixing times performance was negligible.

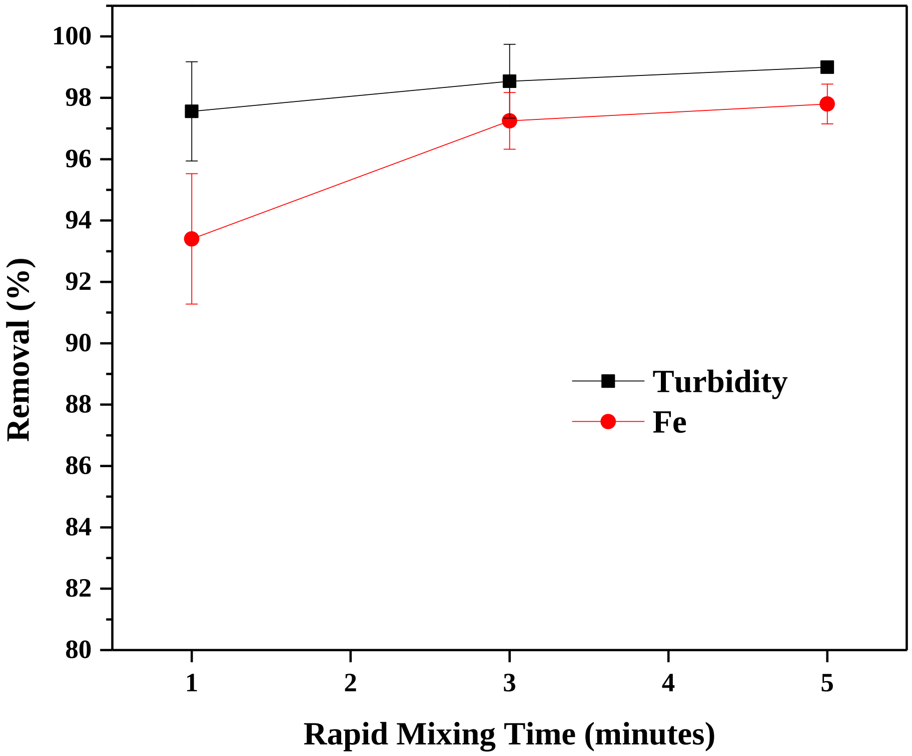


Figure 10: **Determination of the optimum rapid mixing time for Fe and turbidity removal from the Kou River water using jar-test**

Slow Mixing Time

The time provided for flocculation at slow mixing rates was varied between 10 min and 40 min to evaluate its effect on Fe and turbidity removal performance (Fig. 11). A significant increase in performance was observed as the flocculation time was raised from 10 minutes to 30 minutes which showed about 7% and 6% increment in turbidity and Fe removal respectively. The performance of the slow mixing time for 40 minutes was almost the same with that of the 30 minutes (< 1% difference). Thus, 30 minutes slow mixing time should be sufficient enough to facilitate a better flocculation. A similar result has been reported in a study where 30 minutes slow mixing resulted in great removal efficiency in the removal of turbidity using alum (Taşdemir, 2012).

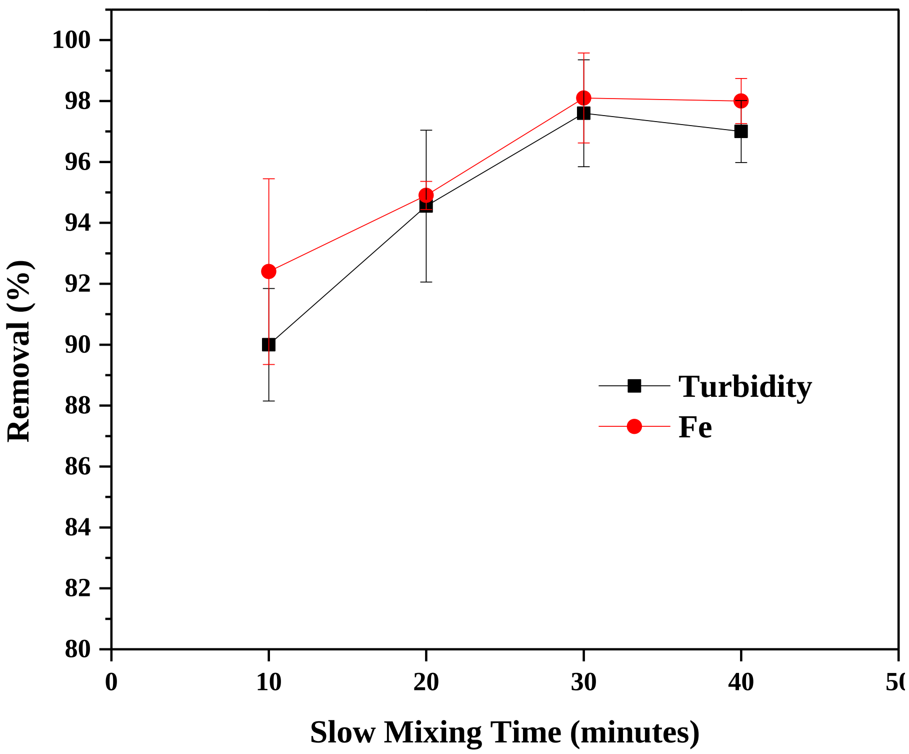


Figure 11: **Determination of the optimum slow mixing time for Fe and turbidity removal from the Kou River water using jar-test**

Therefore, a rapid-slow mixing time of 5 min – 30 min should result in better flocculation for Fe and turbidity removal from the Kou River water.

Effect of Settling Time (Experiment 5)

The effect of settling time (15 min, 30 min and 1 hr) was investigated by applying the optimized conditions from the previous experiments, i.e. alum dose of 30 mg/L, pH of 7.7, rapid-slow mixing rate at 150 rpm - 20 rpm and rapid-slow mixing time for 5 min -30 min. No variation was observed in Fe removal at all settling times (97.25%, 97.27% & 97.5%) where a slight increment (~1%) in turbidity removal at 1 hour settling time was observed as compared to the 15 minutes time (98.54%) (Fig. 12). The slight increment in performance with increasing time was due to the provision of more time for the flocs to settle. However, as the increment in removal performance of the 1 hr. settling time was not significant (only 1%), the 15 minutes settling time was considered to be sufficient and time saving. Thus, 15 minutes settling time was considered good enough to facilitate sedimentation. A similar result has been reported in a study which indicated that 15 minimum is sufficient enough for sedimentation with only a slight increment with increase in time (Taşdemir, 2012).

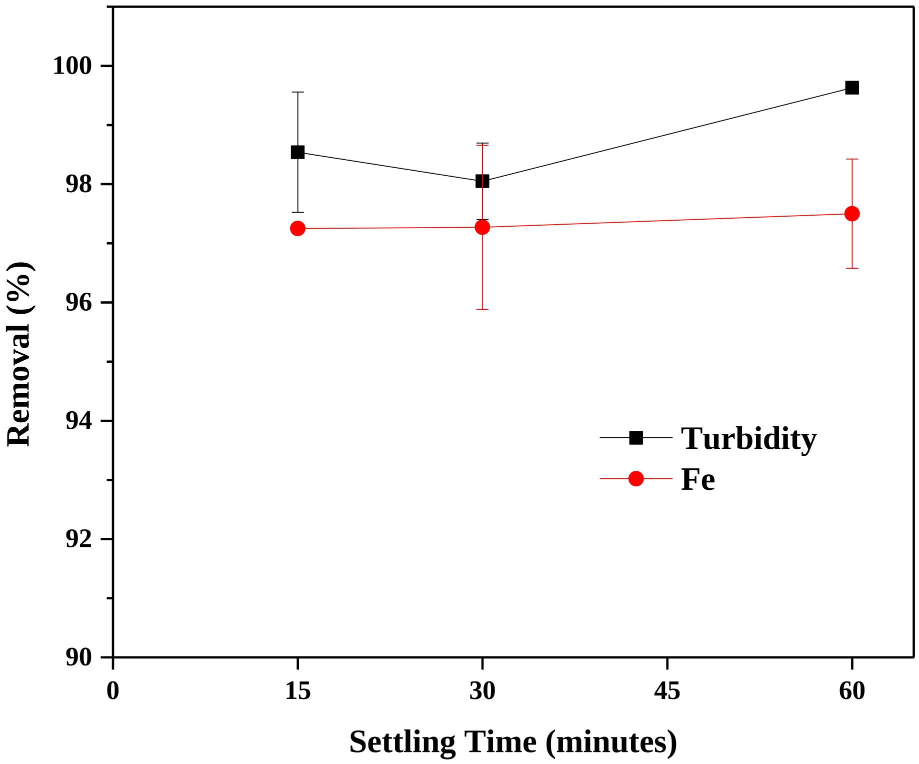


Figure 12: **Determination of the optimum settling time for Fe and turbidity removal from the Kou River water using jar-test**

Control Experiment

A control experiment was conducted to see the independent effect of mixing and pH on flocculation without any chemical addition. The pH was varied between 5 and 9 following the optimized conditions from the five experiments, i.e. 150 rpm - 20 rpm rapid-slow mixing rate, 5 min - 30 min rapid-slow mixing time and 15 min settling time. As can be seen from Fig. 13, both mixing, and pH had an effect on flocculation. Turbidity removal was in the range of 33% and 39.62% whereas that of Fe was as low as 12% to 19.31% indicating that most of the Fe precipitates and other impurities could not form large enough settleable flocs without the aid of coagulant chemical. Therefore, this experiment was evidence that the best flocculation efficiencies were obtained as a result of the addition of alum.

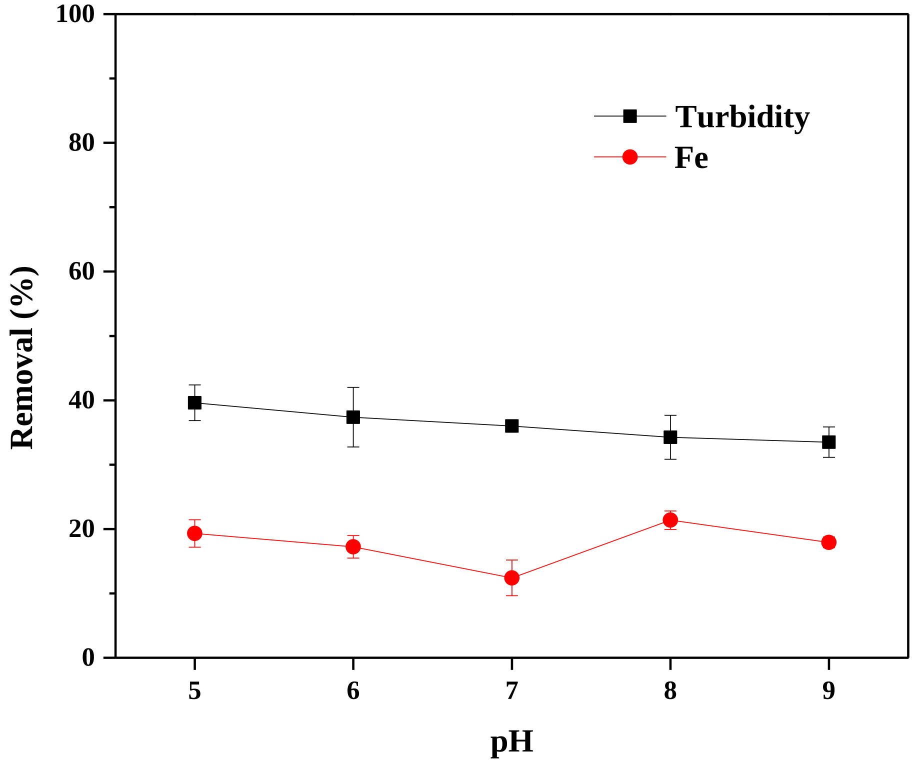


Figure 13: **Control experiment showing the effects of pH, mixing and sedimentation on the removal of Fe and turbidity from the Kou River water in the jar-test process without any addition of coagulant chemical**

Overall, the optimized conditions in the jar-test resulted in 98.85% turbidity and 96.43% Fe removal from the Kou River water with 87 NTU turbidity and 3.64 mg/L Fe initial concentration. The optimum dose and working conditions were:

- Dose of alum: 30 mg/L.
- Initial pH: approximately between 7 and 8.
- Rapid mixing at 150 rpm for 5 min.
- Slow mixing at 20 rpm for 30 min.
- Settling time:15 min.

4.3.3 Validation Using Scale-Up Pilot Plant

A scale-up pilot plant with 10 liters storage capacity was developed to validate the jar-test optimized conditions. The pilot-system was adjusted to operate with the working conditions that are derived from the jar test experiment. First, the raw water was poured into the main tank. The time required for the main tanker valve to open for drawing 10 L water to the rapid mixing tank was 7 minutes. The valve then closes, and rapid mixing motor starts rotating after 10 seconds to allow addition of alum solution manually. The rapid mixing motor rotates at 150 rpm for 5 minutes and the valve of Tank 2 opens for 7 minutes to allow water passage to the slow mixing tank (Tank 3). Then the slow mixing motor rotates at 20 rpm for 30 minutes and stops for 15 minutes to allow sedimentation. After 15 minutes, the valve of Tank 3 opens to transfer the supernatant water to the final basin where the treated water is stored. The developed scale-up pilot plant and its parts are presented in Fig. 14 and 15.

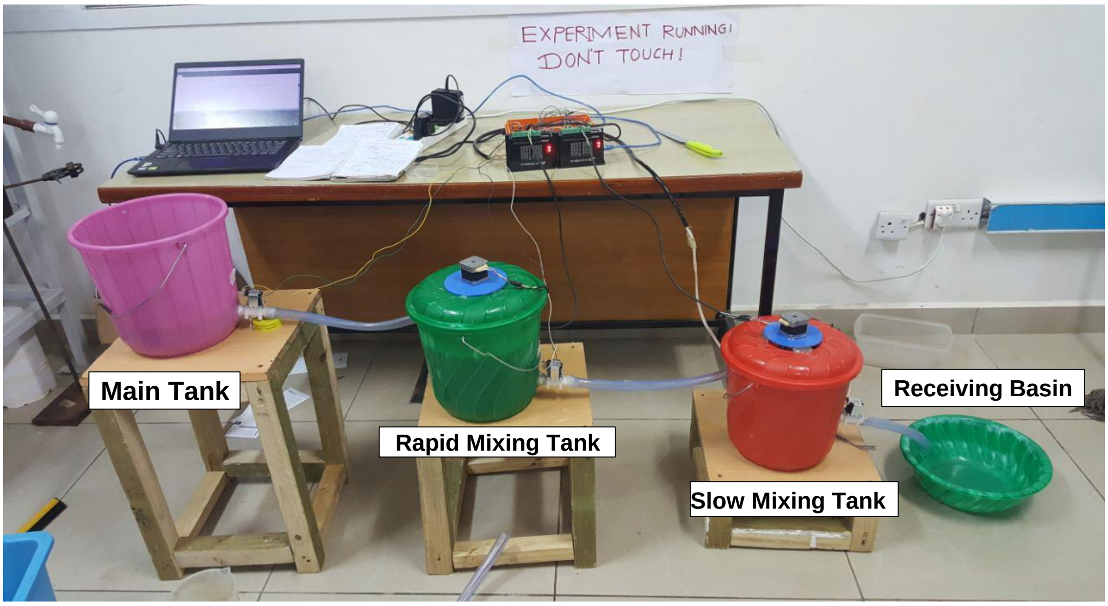


Figure 14: **Set up of the coagulation-flocculation-sedimentation laboratory-scale-up pilot plant of 10 L water treatment capacity with gravity water flow, containing main tank for raw water intake, mixing tanks, treated water reciving basin, and control box with motor drivers, controlled by arduino programing software**

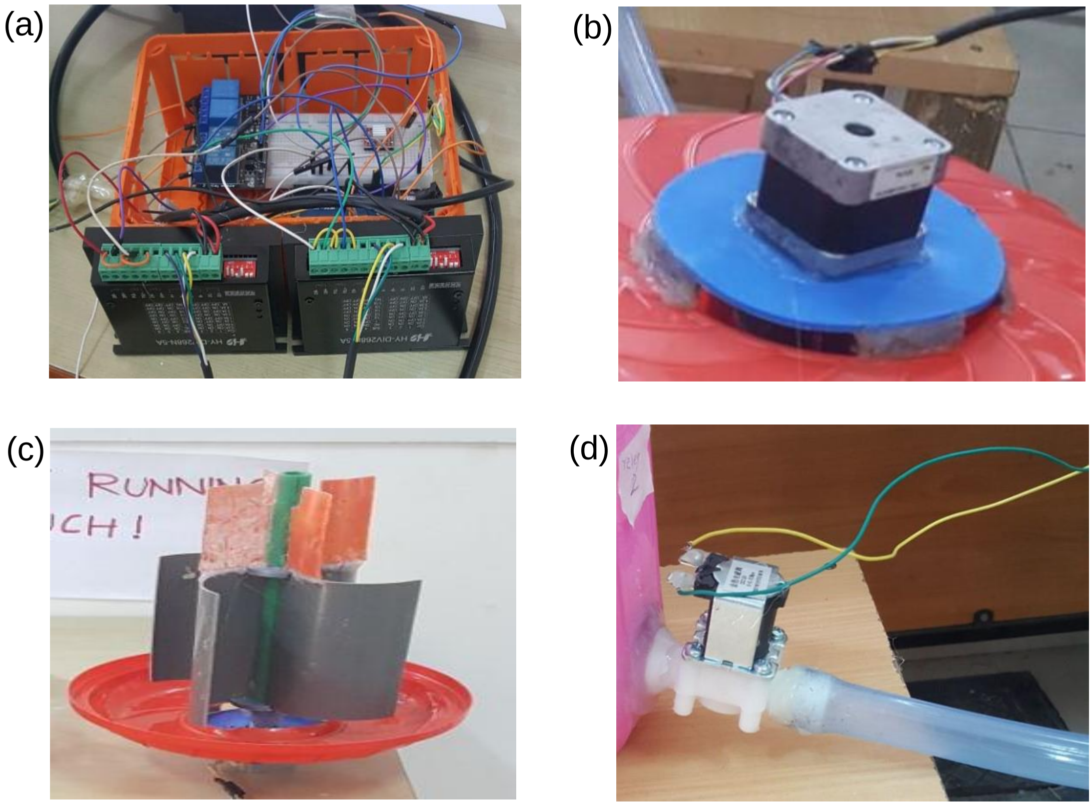


Figure 15: **The Parts of the coagulation-flocculation-sedimentation scale-up pilot plant indicating: (a) control box with drivers facilitating the mixing conditions, (b) stepper motor for rotating the shafts for mixing, (c) impellers for rotating the water in the mixing tanks and (d) solenoid valves to control water flow between tanks through plastic pipes**

Using alum dose of 30 mg/L, the Fe and turbidity removal performance of the scale-up system (94% & 87 %) reduced than that of the jar test results (96.43% & 98.85%). However, with modification in settling time, i.e., when settling time was increased from 15 minutes to 30 minutes, an increased removal performance (97.5% & 98.25%) was obtained. Thus, 30 minutes settling time was found to be optimum.

Further reduced doses of alum (10 mg/L & 20 mg/L) other than the optimum (30 mg/L) was also involved in the pilot test to observe if there was performance improvement with the smaller doses followed by the application of the optimized conditions. However, alum concentrations of 10 mg/L and 20 mg/L have not resulted in better flocculation, suggesting maintaining 30 mg/L alum dose as the optimum. Figure 16 depicts the Fe and turbidity removal performances of the three doses.

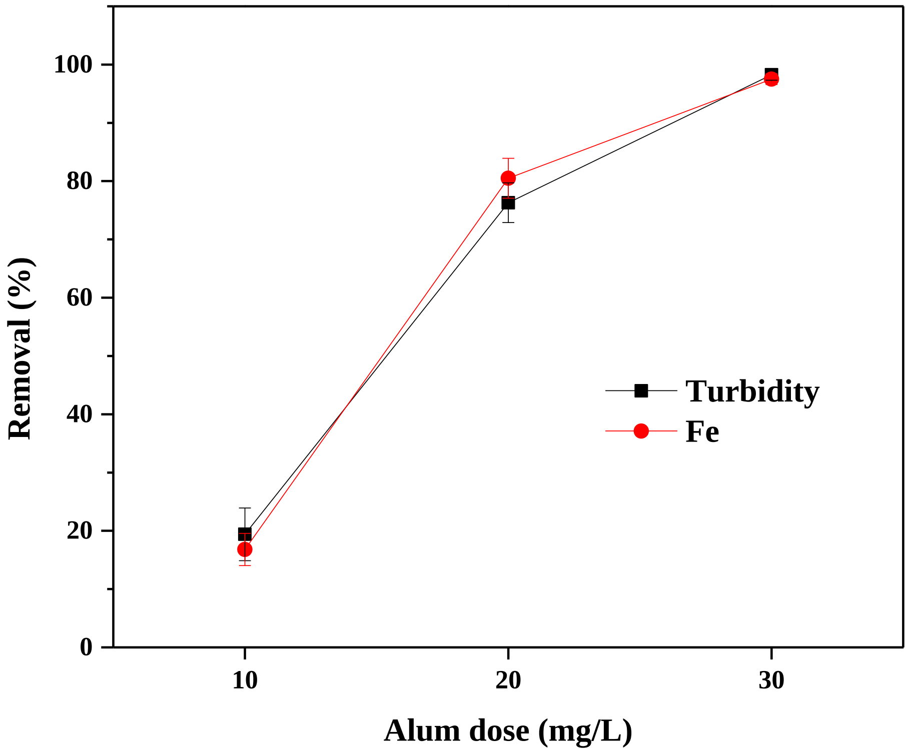


Figure 16: **Validation of the jar-test optimized coagulation-flocculation-sedimentation operating conditions and three different alum doses for the removal of Fe and turbidity from the Kou River water using the scale-up pilot plant test**

Overall, the scale-up system, with the optimum conditions along with the 30 mg/L dosing removed 98.25% turbidity and 97.5% Fe with only 0.09 mg/L residual Al which favorably validated the jar-test optimized conditions with modification in settling time to 30 minutes.

CHAPTER FIVE

CONCLUSION AND RECOMMENDANTIONS

5.1 Conclusion

Coagulation-flocculation-sedimentation process is a commonly applied treatment technique in domestic water supplies which requires optimization for treating a particular raw water. The present study investigated the quality of Kou River water in Northern Tanzania, evaluated TMEs pollution in the sediments, and optimized a coagulation-flocculation-sedimentation water treatment process for Kou water treatment.

The degree of TMEs contamination (Ni, Fe, Mn, Co, Cr, As, Pb, Zn and Cu) in the surface water and sediment of the Kou River was evaluated. Based on NQEp and SEQ-Eau guidelines, Kou River water was not suitable for sustaining biological life in terms of Ni, Zn, Mn, Cu and Fe. Concentrations of Zn, Ni, Co, Mn, and Fe in Kou water did not meet the USEPA requirements for aquatic organisms. Apart from Fe, Co and Cu concentrations, the water was suitable for Agriculture. Overall, Kou River water failed to meet the requirements for aquatic life, biological life and agriculture with at least one metallic ion.

The overall TMEs contamination level of the Kou River sediment was moderate, with Fe, Cr, Ni and Mn being the main contaminants. In relation to the Pollution Indices, the pollution levels of Mn and Fe were in the no-to-moderate class. Nevertheless, the USEPA guideline showed a serious pollution condition with the metals. The TMEs in the sediment had no human-induced enrichment, except for Cr, which was possibly derived from anthropogenic activities. The Fe has been the only metal found to surpass all the recommended limits of water for biological life, aquatic life, and irrigation. The biologically available levels of Fe in the sediments were marginal with no potential to pose an ecological risk, hence the sediments were probably not the cause of Fe pollution in the river water. In spite of the inadequacy of Kou River water for aquatic life, biological life, and irrigation with one or more metals, the nine metals in the sediments hardly pose ecological risks. The source of high Fe in the Kou River is linked possibly to geological formation of the area where as turbidity tends to fall during dry seasons and rise during rainy seasons possibly showing the disturbance of the sediments in the water or transport of particles from the catchment with heavy rain.

The Kou River water at the Magugu water supply collection point was found to have elevated Fe concentration (3.64 mg/L) and turbidity (87 NTU) in so far as water for domestic purpose was concerned. The effect of five factors (coagulant dose, pH, mixing rate, mixing time and settling time) was investigated for optimizing a coagulation-flocculation-sedimentation process with aluminum sulfate as coagulant in treating turbidity and Fe from Kou River water. The optimal conditions obtained were 30 mg/L alum, pH of approximately 7 to 8, rapid mixing rate at 150 rpm for 5 minutes, followed by slow mixing rate of 20 rpm for 30 minutes and sedimentation time of 30 minutes. These were validated with 10-liter capacity pilot plant test which resulted in 98.25% turbidity and 97.5% Fe removal. The application of the optimal conditions in the construction and operation of the planned coagulation-flocculation-sedimentation plant is promising to effectively treat Kou River water for safe domestic water supply to the Magugu community.

5.2 Recommendations

(i) Fluctuation of turbidity over a short period of time in surface waters is a challenge in coagulation-flocculation-sedimentation process requiring continuous optimization which is time consuming. Further research on developing enhanced time-saving optimization method and developing a dose indicator technology to assign alum dose requirement based on the fluctuating characteristics of the raw waters (turbidity) is recommended.

(ii) Researches have reported that consumption of iron at higher concentration has health effects on living organism. However, toxic response coefficient for iron has not been established. There is a need to establish the toxic response coefficient for this metallic element to enable the determination of its risk to living organism using the ecological risk index.

(iii) Further research on optimization of the coagulation-flocculation-sedimentation water treatment process on a large-scale continuous system is recommended to implement the method in the real environment.

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RESEARCH OUTPUTS

(i) Publication

Gebreyohannes, N. M., Rwiza, M. J., Mahene, W. L., & Machunda, R. L. (2022). Assessment of contamination level of a Tanzanian river system with respect to trace metallic elements and their fate in the environment. *Water Supply*, 22(4), 3588-3602.

(ii) Poster Presentation