Chinhoyi University of Technology

ASSESSING THE EFFICACY OF *ELEPHANTORRHIZA GOETZEI* (NDORANI) SEED EXTRACTS FOR HOUSEHOLD WATER TREATMENT IN ZIMBABWE.

Thesis submitted in fulfillment of the requirements for the Degree of Master of Philosophy

By

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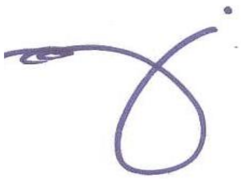

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Dedication

This thesis is dedicated to my loving wife Rutendo Mudinzwa, my supportive parents Mr and Mrs Diver and my caring siblings Charmaine and Shingirai Diver.

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First and foremost, I would like to give all the glory, honour and praise unto Jehovah God for continually showering His grace upon me throughout my study period, for without his grace I would not have made it this far.

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Abstract

Poor access to clean and safe water is one of the major problems being encountered the world over. There has been considerable attention in recent times towards the use of natural coagulants. The study seeks to investigate the efficacy of *Elephantorrhiza goetzei (E.goetzei)* seed extracts as natural coagulants for synthetic water treatment.

The methodology consisted of the proximate analysis to investigate the active ingredient(s) responsible for coagulation. Standard methods were used for experiments in this study. Coagulant dosages ranging from 0 to 300 mg/L with a rapid mixing speed of 120 rpm for 1-minute, slow mixing speed of 30 rpm for 15 minutes and settling time of 15 minutes were used for the jar test. IBM SPSS Version 20 was used for data analysis.

The results obtained from the proximate analysis of *E. goetzei* seed extracts show that values of 5.25%, 21.40%, 8.23%, 32.99%, 2.20%, and 29.93% were obtained for moisture, crude protein, crude fibre, fat, ash, and carbohydrate content, respectively. Moreover, seed extracts of *E.goetzei* achieved removal efficiencies up to 94.8%, 50.1%, 90.0%, and 53.9% for turbidity, fluoride, iron and manganese in water, respectively. The COD increased from 55.3 mg/L to 419.3mg/L as the coagulant dosage increased from 0 to 100 mg/L. The total nitrogen and phosphorus obtained in the sludge at 100 mg/L were 0.343 µg/kg and 0.194 µg/kg. Coagulant dosage had a significant effect ($p < 0.05$) on pH, TDS, turbidity, fluoride, iron, manganese and COD of the treated water and on the nitrogen and phosphorus in the sludge.

In conclusion, the study proved that *E.goetzei* seed extracts contain proteins, carbohydrates, fat, and fibre as potential active coagulation ingredients. *E.goetzei* seed extracts coagulant has potential to achieve the desired World Health Organisation (WHO) drinking water standard at higher dosages for fluoride, iron, and manganese. It can also be concluded that there is a consistent pattern of higher COD levels in the treated water as the dosage of coagulant increases. The significantly lower nitrogen and phosphorus content in the sludge following water treatment, in comparison to compost and organic fertilizer, restricts the potential use of sludge as a fertilizer.

As a recommendation, further work needs to be done on the use of *E.goetzei* seed extracts for raw water sources within households. The impact of operational parameters such as pH on treatment efficiency should be investigated. Finding solutions to address the increase in residual organic matter with higher coagulant dosages is also crucial for improving water quality.

Chapter 1: Introduction

Background

Poor access to adequate, clean, and safe water is one of the greatest world problems that people encounter (Pandey *et al*., 2021). In the year 2020, about 122 million people were getting their water directly from surface water sources (UNICEF/WHO, 2021). In 2022, about 2 billion people did not have safely managed drinking water services (World Bank, 2022).

Water can become contaminated by either natural or anthropogenic sources of pollutants (Ojha, 2020). Many anthropogenic activities taking place in the environment have increased the contamination of the available water resources (Fernando *et al*., 2018). Moreover, an increase in population in urbanized areas escalates environmental pressures from industrial practices and waste disposal habits (Boelee *et al*., 2019). When groundwater infiltrates through the pores of rocks and various soils, it can contaminate the water through natural processes by mixing with various natural substances including iron, and nitrate. Consequently, high concentrations or undesirable quantities of the naturally-occurring compounds such as arsenic, and fluoride can then contaminate the groundwater, the consumption of which could cause health problems such as dental and skeletal fluorosis (Sharmila *et al*., 2019 ; Ali & Ahmad, 2020).

In both developed and developing countries, the challenges of contamination of water are mainly because of chemical exposure which includes but is not limited to agricultural sources (Sharma & Bhattacharya, 2017). Contaminants such as insecticides, herbicides, pesticides, chemical fertilizers, heavy metals (such as mercury and lead), and hazardous chemicals mainly emanate from agricultural and industrial activities (Fawell *et al*., 2001; Ashbolt, 2004; Bekele & Yegrem, 2021). Disposal of batteries, diesel fuel, and household chemical wastes such as oils, medicines, antiseptics, and paint solvents can also result in groundwater contamination and severe health challenges (Anwar & Yaqout, 2003; Kass *et al*., 2005; Neksumi *et al*., 2022). Furthermore, the release of chemicals found in wastewater can result in the pollution of groundwater. Riverbeds and wetlands are also at risk due to rising sediment impoundments and the presence of persistent and hazardous contaminants. Such pollution can persist even after the sources have stopped producing the pollutants (Edokpayi *et al.*, 2017).

Globally, at least 2 billion people use a drinking water source with faecal contamination, causing about 485,000 diarrheal deaths each year (WHO, 2019). According to

WHO/UNICEF(2021), about 385.5 million people who were still lacking even a basic drinking water service in 2020 resided in the sub-Saharan African region. Within sub-Saharan Africa, people in rural areas of Zimbabwe drink water from various sources ranging from piped water within a dwelling, public taps, boreholes, protected and unprotected wells/springs, rivers and dams with 25.9% of households in rural areas using non-improved sources of water such as unprotected wells or springs, rivers, dams and rainwater (ZIMSTAT, 2022).

The pollution of drinking water can result in waterborne diseases such as typhoid, cholera, diarrhoea, dysentery, hepatitis, and many protozoa and helminths infections which can result in high mortalities and morbidities (Aryal *et al*., 2012; Pal *et al*., 2018). Consumption of water that has pathogens or elements that could be toxic can result in health impacts that range from discomfort to death (UNEP, 2016). Millions of people die because of water-related diseases such as cholera, diarrhoea, malaria and dengue fever (Ahmed *et al*., 2020). Diarrheal infections which are largely caused by using unsafe drinking water are responsible for about 2.2 million deaths annually in developing and undeveloped countries (George *et al*., 2022). In areas such as Gokwe, Midlands Province, Zimbabwe, and Gwanda, Matabeleland South Zimbabwe, problems such as dental fluorosis, have been reported (Magalela, 1998; Maya *et al.*, 2020).

Raw water often requires treatment to make it potable. If the water originates from ground sources via mountain springs, the water quality will usually be good and, in most cases, only disinfection is sufficient (Dursun & Sarcan, 2022). However, the suspended and colloidal particles are of greatest significance if the water is from a surface water source such as a river, lake, or dam. Various techniques to remove colloidal particles include the introduction of coagulants and the use of membranes (Owodunni *et al*., 2023).

Due to potential disturbances with purification processes downstream and adverse impacts on consumer acceptance, effective turbidity reduction is one of the main objectives in the treatment of water for drinking purposes (Hussain *et al*., 2019; Mohamed *et al*., 2022). Turbidity removal in water treatment has two purposes. Firstly, removal of turbidity results in the removal of some microbes. Secondly, turbidity removal reduces the levels of organic matter and other particles, which increases the effectiveness of the downstream treatment processes (Soros *et al*., 2019). Since heavy metals can be detrimental and dangerous to consumers' health, their removal is also very important. For example, excessive quantities of copper can be both toxic and carcinogenic (Kahlson & Dixon, 2022).

Turbidity in water can be reduced or removed using several standard techniques, including precipitation, adsorption, flotation, ion exchange, membrane filtration, and biological and electrolytic processes (Rajoria *et al*., 2022). The process of turbidity removal in the form of suspended and colloidal particles necessitate the use of a coagulant during the coagulation-flocculation stage of water treatment operations. Coagulation is the clustering process that occurs under intense mixing, whilst flocculation is the settling process that occurs under gentle mixing (Alazaiza *et al*., 2022). The coagulation process is widely utilised in water and wastewater treatment since it is efficient for removing suspended solids, turbidity, organic matter, oil, and colour (Aziz *et al*., 2018). Coagulation takes place through the addition of a coagulant, which causes colloidal particles to form bigger flocs that are able to settle. Coagulation and flocculation are related processes. Colloidal particles typically have a negative charge. Thus, coagulation is a chemical process that involves neutralizing the colloids in water, whilst flocculation is a physical process that involves the formation of flocs from particles that would have been neutralised during the coagulation process (Bahrodin *et al*., 2021). Coagulants and flocculants come in various forms and are used in conventional water treatment processes. Coagulants can be grouped into inorganic coagulants, synthetic organic polymers and natural coagulants (Halder, 2021).

Natural coagulants are polyelectrolytes, which can be anionic, cationic, or neutral polymers (Yin, 2010). According to Afangideh (2023), the utilization of natural materials of plant origin to treat turbid raw waters is not a new concept. Natural organic polymers have been used as coagulants and coagulant aids for water with high turbidity in China, India, and Africa for over 2,000 years (Asrafuzzaman *et al*, 2011). In recent years there has been significant interest in the utilization of natural coagulants (Mumbi *et al.*, 2018; Unnisa and Bi, 2018; Alenazi *et al.*, 2020; Amran *et al.*, 2022). Some of the natural coagulants which have been investigated include *Moringa oleifera* seeds (Okuda *et al.*, 1999; Saulawa *et al*., 2011; Hussian *et al*., 2012; Pandey *et al*., 2020; Nhut *et al.*, 2021; Tong *et al*., 2021), Cactus (Choudhary *et al*, 2018), Nirmali seeds (Alenazi *et al*., 2020), *Azadirachta indica* (Ahmad *et al.*, 2021) *Carica papaya* seeds (Amran *et al*., 2021) , *Aloe vera* (Benalia *et al*., 2021), *Tulsi (Ocimum Sanctum)* (Bayu *et al*., 2022), *Cassia fistula* seeds (Dunoyer *et al*., 2022),and Walnut seeds (Zedan *et al*., 2022).

Natural coagulants are biodegradable, thereby making them a potentially sustainable option for water treatment (Agarwal & Saini, 2022). They have a strong ability to sustain the pH of the treated water while being safe and economical. In contrast to chemical coagulants, natural coagulants do not increase the metal load during treatment and are characterized by the production of a small amount of sludge, thereby lowering the cost of disposal significantly (Owodunni & Ismail, 2021; Alazaiza *et al*., 2022).

Large-scale water and wastewater treatment facilities have not fully adopted the use of natural coagulants, despite their numerous benefits. A few significant obstacles have been found in the commercialisation of natural coagulants that prevent them from moving from coagulation studies at the laboratory scale to widespread practical water and wastewater treatment applications. The biggest barrier is the industry's lack of confidence in using natural coagulants in treatment procedures (Karnena & Saritha, 2021). Although the usefulness and advantages of natural coagulants have been demonstrated in laboratory-scale investigations, questions remain regarding their cost-effectiveness and performance consistency in actual treatment operations (Ang & Mohammad, 2020). Another disadvantage of using plant-based coagulants in process scale-up is the rise in organic loads, especially if unprocessed extracts are utilised.

Some of the concerns that have been raised when using natural coagulants for water treatment include low solubility in water, a small working window of pH range, undesirable isoelectric point, weak surface charge, low stability (shelf life), and moderate efficiency which leads to higher working dosage and consequently increased cost (Lee *et al*., 2014 ; Salehizadeh *et al*., 2018). The components of natural sources could make it difficult to regulate the quality control of natural coagulants because of differences in growth conditions and species (Ang & Mohammad, 2020). Natural coagulants cannot hydrolyse in water to generate strongly charged hydroxides for the elimination of contaminants, unlike inorganic coagulants like ferric chloride and alum. However, the reactive functional groups of the natural coagulants offer a chance for modification that could help to deal with some of the previously mentioned concerns (Ghimici & Nichifor, 2018). Various compounds that could considerably improve the coagulation performance of natural coagulants can be introduced to the coagulants through interaction with the functional groups.

Although some work has been made to extend piped water supply systems to rural areas as well as peri-urban areas, the shortage of finance and affordability have been the biggest impediments (Kusena & Beckedahl, 2014). A low-cost mechanism of treating water which is easy to maintain, and monitor is necessary for rural households (Daniel *et al.*, 2018; Chu *et al*., 2019;

Hussain & Al-Fatlawi, 2020). The development of locally available natural coagulants appears very attractive to Zimbabwean conditions; principally because of their low cost and easy accessibility. Various natural coagulants have been used for the removal of contaminants in water and wastewater such as turbidity, colour, iron, manganese, lead, and copper (Jagaba *et al*., 2021; Serasinghe *et al*., 2022).

The potential of the natural plant extracts such as *Moringa oleifera* seeds , chitosan, zeolite has been investigated in Malaysia whilst *Terminalia arjuna, Artocarpus heterophyllus, Strychnos potatorum L*, and *Aloe vera* in water treatment has been investigated in Sri Lanka (Muyibi & Alfugara, 2003; Jagaba *et al*., 2021; Serasinghe *et al*., 2022). A plant which is indigenous to Zimbabwe, known as *E.goetzei* (*ndorani* in Shona) has also shown similar characteristics as that of other plants which have been used as natural coagulants such as *Moringa oleifera*. Studies have shown that both *E.goetzei and Moringa oleifera* contain similar phytochemicals such as phenolic compounds, flavonoids and tannins (Saini, 2016; Maroyi, 2017).Therefore, the *E.goetzei* plant needs to be investigated to determine its efficacy as a natural coagulant for water treatment. Currently, there does not seem to be any published studies on the use of *E.goetzei* seed extracts for water treatment.

1.2 Research problem

The current poor access to safe and reliable drinking water in rural areas of Zimbabwe has led to the inhabitants drinking polluted water which exposes them to diseases such as dental and skeletal fluorosis, cancers and heavy metal contamination-related illnesses (Mamuse & Watkins, 2016; Teta & Hikwa, 2017). There is a genuine risk that the United Nations Sustainable Development Goal number 6 (SDG6) might not be met in developing countries as happened with the Millennium Development Goals on access to safe drinking water unless there are concerted efforts to find solutions for rural areas. Over the years the potential of the seeds of *Moringa oleifera* in water treatment has been investigated (Muyibi & Alfugara, 2003; Jones, 2019). Findings have shown that the *Moringa oleifera* seeds, a natural coagulant can be a feasible option to substitute, to some extent or totally, aluminium sulphate and other chemicals used in the treatment of water (Ukpong *et al.*, 2021). In parallel studies to *Moringa oleifera*, a plant which is also indigenous to Zimbabwe, known as *E.goetzei* (*ndorani* in Shona) has also shown similar characteristics as that of the *Moringa oleifera* tree as it has medicinal properties (Maroyi, 2017). In fact, a number of promising experiments have been carried out as BSc projects and there is now a need to conclusively deal with the efficacy of *E.goetzei*, and make the product ready for use in rural areas where natural surface and ground water has typical problems that can be solved by this plant extract.

1.3 General objective

To determine the suitability of the seed extracts of *E.goetzei* for household water treatment.

1.3.1 Specific objectives

1. To determine the key ingredients in the *E.goetzei* seed extracts which are responsible for the coagulation process.
2. To determine the optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity.
3. To determine the effect of *E.goetzei* seed extracts on the removal of fluoride, manganese and iron from synthetic water.
4. To determine the levels of residual content of organic matter in synthetic water treated with *E.goetzei* seed extracts.
5. To determine the quality of the sludge produced from treating synthetic water with *E.goetzei* seed extracts, and hence its feasibility for agricultural reuse.

1.4 Hypotheses and research questions

The hypotheses and research questions used in this research are presented in the subsequent subsections.

1.4.1 Hypotheses and criteria for testing the hypotheses

The researcher proposed the null (H_0) hypothesis and the alternative hypothesis (H_1). The hypotheses are defined as shown below.

H_0: Seed extracts of *E.goetzei* are not suitable for household water treatment.

H_1: Seed extracts of *E.goetzei* are suitable for household water treatment.

1.4.2 Research questions

Listed below are the research questions which will be answered by the respective objectives of the study as they are carried out.

1. What are the key ingredients in the *E.goetzei* seed extracts which are responsible for the coagulation process?

2. What are the optimal dosages of *E.goetzei* seed extracts which are required for effectively removing turbidity?
3. What is the effect of *E.goetzei* seed extracts coagulant dosage on the removal of fluoride, manganese, and iron from synthetic water?
4. How much residual content of organic matter is present in water that has been treated with seed extracts of *E.goetzei*?
5. Is the sludge produced during the treatment of raw water with *E.goetzei s*eed extracts of suitable quality for further reuse in agriculture?

1.5 Justification of the study

Since the *E.goetzei* plant is locally abundant in rural areas such as Chegutu District, developing a coagulant using its seed extracts will be a viable solution to address the problem of poor water quality in rural areas of Zimbabwe. Furthermore, the adoption of the use of the seeds at household level provides an opportunity for urban municipalities to adopt the use of the seeds of *E.goetzei* for treatment of turbid river water. This will result in a decrease in the importing of chemical coagulants such as aluminium sulphate (alum) thus significantly reducing the cost of water treatment since a locally available product will be used as a coagulant. Moreover, if the urban municipalities adopt the use of *Elephantorrhiza goetzei* instead of alum for water treatment it will promote the health of the people since alum is reported to induce Alzheimer's disease. Moreover, this research will extend currently existing global knowledge on the use of natural coagulants for water treatment.

1.6 Thesis outline

The thesis comprises six complementary chapters which are outlined in the following format:

Chapter 1 contains the background on challenges of contamination of water, water treatment and the use of natural coagulants for water treatment. It also presents the research problem which the study seeks to address and the objectives for carrying out the study together with the corresponding research questions which were answered by the research. The scope and justification of the research were also discussed in this chapter.

Chapter 2 presents the review of related literature on challenges in household water supply, coagulation-flocculation process in water treatment and types of coagulants. The chapter also contains an analysis of natural coagulants using various criteria such as dosage, active ingredients responsible for coagulation and extraction method. Studies were also reviewed to analyse the extent to which natural coagulants have been successful in the removal of contaminants that include heavy metals and turbidity in water.

Chapter 3 presents the conceptual framework which was used to carry out the research. The experimental design was also discussed in this chapter and an outline of how data for the research was collected including clear procedures for all the experimental work was presented This chapter also outlines how data analysis was done for the study.

Chapter 4 presents the results for proximate analysis of *E.goetzei* seed extracts, determination of the optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity, effect of *E.goetzei* seed extracts coagulant dosage and initial concentration on removal efficiency of fluoride, iron, manganese, effect of *E.goetzei* seed extracts coagulant dosage on the COD of the treated water, and effect of *E.goetzei* seed extracts coagulant dosage on the nitrogen and phosphorus in the sludge obtained after water treatment.

In *Chapter 5*, the findings of the research were compared to other similar studies and an explanation of the trends in the results was also presented.

In *Chapter 6*, a conclusion was given for each objective based on the results obtained. An overall conclusion on the main hypothesis is also presented. Recommendations to map the way forward for further research were then suggested.

Chapter 2: Literature review

This chapter will begin by presenting the state of potable water supply in rural areas. The relevant literature about the coagulation-flocculation process and the mechanism by which various coagulants operate including natural coagulants will also be discussed. Studies in which various natural coagulants have been used and how the coagulants performed will also be reviewed.

2.1 Challenges in household potable water supply in rural areas

Potable water is water that is safe for drinking, food preparation, personal hygiene and washing (Bos, 2016). Achieving adequate, safe, and consistently available drinking water services in rural areas is difficult, especially in low-and middle-income countries (Miller *et al*., 2019). In 2020, the global percentage of safely managed drinking water services was lower in rural areas (60%) than in urban areas (86%) with the most significant gap in the percentage of the population with access to safely managed drinking water between urban (54%) and rural (13%) areas being found in Sub-Saharan Africa. (UNICEF/WHO, 2021).

A study was carried out by Makokove *et al.* (2022) on the effect of household practices on the deterioration of microbial quality of drinking water between source and point of use in rural communities in Murewa district, Zimbabwe. The study revealed that there was a prevalent problem of poor stored water quality, with 23.3% of the household water samples contaminated with *E. coli*. This is despite the collection of water from improved water sources such as boreholes and communal piped water schemes and the water samples collected at sources testing negative for *E. coli*. The amount of handling involved in collecting, transporting, storing, and drawing water from containers may be the reason for the contamination of stored water (Makokove *et al*., 2022).

A study was conducted in which the groundwater quality of water points used for domestic purposes in Gokwe South, Nkayi, Lupane, and Mwenezi rural districts in Zimbabwe was investigated (Hoko, 2005). These areas were used since they are some of the driest districts in Zimbabwe and are therefore dependent on groundwater for various uses. The parameters analysed were pH, temperature, dissolved oxygen (DO), turbidity and electrical conductivity (EC). The pH generally ranged from 6.5 to 8.0. DO was 0.3–5.9 mg/l while the average turbidity ranged from 10.8 to 16.5 Nephelometric Turbidity Unit (NTU) with Mwenezi having the highest average turbidity value. Generally considering average district values of turbidity, all districts did not meet the WHO guideline for drinking water, which is set at 5 NTU. Conductivity ranged from 70 to 9800 µS/cm with the lowest and highest values recorded in Gokwe and Mwenezi, respectively. In another study conducted in a region spanning across Gokwe South and Gokwe North districts by Mamuse and Watkins (2016), 28% of the 126 water sources sampled contained excessive fluoride (> 1.5 mg/L) which, in some places reached up to 11 mg/L and was sufficiently high to cause skeletal fluorosis. Concentrations of TDS, chloride, sulphate and sodium ions were found to be higher than WHO taste-based aesthetic guidelines in many water sources.

2.2 Coagulation and flocculation processes in water treatment

Coagulation and flocculation processes are regularly utilised in the treatment of drinking water (Maćczak *et al*., 2022) and industrial effluents (Teh *et al*., 2016). In an aqueous medium, the majority of solids or suspended particles are smaller and have negative charges. Consequently, the particles must group together to create larger flocs to speed up the sedimentation process. Electrostatic repulsive forces, which prevent negatively charged particles from collecting on the substance, pose a challenge to this process. As a result, it takes longer to settle. To fix this problem, the particles have to be destabilized with a coagulant (El-taweel *et al*., 2023). Therefore, coagulation happens when coagulants are added during relatively intense mixing to destabilize naturally occurring particles and macromolecules (Al-Risheq *et al.*, 2021).

The process of coagulation alone does not significantly help, it needs the process of flocculation to perform effectively (Mohd-Salleh *et al*., 2019). Therefore, flocculation which allows slow mixing is a crucial step in water treatment to achieve optimum performance. The primary objective of the flocculation stage is to detect particle collisions and promote their subsequent aggregation. (El-taweel *et al*., 2023).

2.3 Mechanisms of coagulation

In general, the coagulation process consists of four steps that are grouped according to the primary mechanism that causes aggregates to develop. These four steps include charge neutralization, the formation of inter-particle bridges, the formation of electrostatic patches, and double-layer compression (Bordi *et al.*, 2004 ; Mortadi *et al.*, 2017; Ghernaout *et al.*, 2020).

Furthermore, flocculation takes place as a result of two basic phenomena, namely; orthokinetic which is a result of fluid motion, and peri-kinetic which is caused by Brownian motion (Beljon, 2021). Figure 2.1 shows the overall process of coagulation-flocculation in water treatment. The mechanisms for the coagulation process are described in the following subsections.

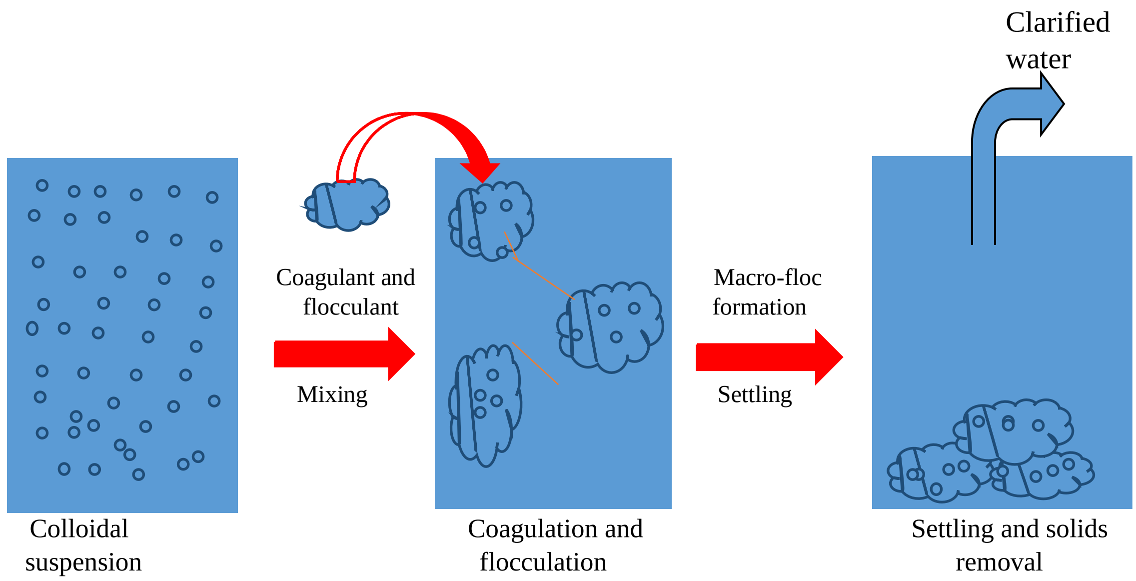

Figure 2.1 | Overview of coagulation-flocculation process, [adapted from Wilson (2014)]

2.3.1 Charge neutralization

The colloidal particle's surface, which is typically negatively charged, is coated by an oppositely charged coagulant as part of the charge neutralization mechanism, which neutralizes the colloidal particle and brings the zeta potential down to almost zero net charges (Choy *et al.*, 2015). Reduced zeta potential, which arises from reducing and neutralizing the surface charge, aids in lowering or removing the energy barrier. As a result, colloidal particles are destabilized and begin to gather together and settle down *(Alnawajha et al.*, 2022).

2.3.2 Formation of inter-particle bridges

When long-chain polymers or polyelectrolytes stretch into a solution and form threads or fibres, they bridge the gap between numerous colloidal particles. A crucial component of bridging flocculation is the molecular weight of polymeric flocculants (Wei *et al.*, 2018; Mohd-Salleh *et al.*, 2019). Polymers that possess a low degree of cationic charge and a high molecular weight should mainly work through the bridging mechanism, since the polymers with large molecular weights have long-chained structures which significantly increase the number of vacant adsorption sites. (Alnawajha *et al*., 2022).

2.3.3 The formation of electrostatic patches

Charged polymers or colloids bind to an oppositely charged particle in this process. Through areas of opposite charges, particles are drawn to one another, thereby coagulating the suspension (Agunbiade *et al*., 2016). For the charge neutralization procedure, a patch-wise medium which is referred to as an electrostatic patch mechanism is used. The surface of the colloids will patch with different cationic species, resulting in particle surfaces with positive and negative charges. Surfaces of colloids with mixed charges will have fewer repelling forces and more van der Waals forces between the particles (Chong *et al*., 2021).

2.3.4 Double-layer compression

The negatively charged colloidal particles draw ions of opposite charge to form a dense layer adjacent to the particle which is known as the Stern layer (Gołębiowski & Buszewski, 2023). The Stern layer's repulsive force and the dynamic equilibrium between the excess positive ions drawn in by the negatively charged core colloids cause the diffuse layer to form. The double layer is a term used to describe such two layers in the colloid particle interfacial region (Zhao, 2020). The double-layer will be squeezed as soon as a coagulant (positively charged) is added to a colloidal solution due to electrostatic interaction between the ions and colloids. Double-layer compression is a crucial destabilizing mechanism in naturally occurring aquatic systems, even though it does not control the colloid destabilization process in water treatment (Ghernaout *et al*., 2020).

Both the formation of inter-particle bridges and charge neutralisation, among the four coagulation mechanisms, are regarded to be the most possible coagulation mechanisms of plant-based coagulants (Bolto & Gregory, 2007; Yin, 2010).

2.4 Classification of coagulants

Different types of coagulants can be used for treating water (Tetteh *et al*., 2017). These coagulants can be chemical, non-chemical, synthetic, or natural coagulants. Each type of coagulant possesses its unique properties with ions that will be positively charged and these will entrap the negative charge of the organic matter in the water that causes turbidity (Tetteh

& Rathilal, 2020). An overview of inorganic coagulants and coagulant aids, synthetic organic polymers, and natural coagulants, is provided in this section.

2.4.1 Inorganic coagulants and coagulant aids

Conventional chemicals which are utilised for coagulation are mainly aluminium or iron-based salts. Metal-based coagulants are most widely utilised because of their advantages which include the fact that they are low-cost and widely available (Dursun & Sarcan, 2022) and can achieve removal efficiencies as much as 98.8%, 99.7%, and for removal of turbidity, total suspended solids respectively and reduction percentage of 92.3 % for COD, (Al-Wasify *et al*., 2023; Kurniawan *et al.*, 2023). The use of these metal-based coagulants for the purification of water is not without drawbacks, though. They may change the pH of the water since they deplete alkalinity, thereby resulting in lime or soda ash being added to maintain the desired pH levels (Iwuozor, 2019). Additionally, the use of metal-based coagulants causes the production of large amounts of sludge as well as the presence of residue metals in the sludge (de Souza *et al*., 2016; Freitas *et al*., 2018). The benefits and drawbacks of some inorganic coagulants for water treatment are listed in Table 2.1.

Table 2.1 | Benefits and drawbacks of inorganic coagulants. Adapted from Thakur *et al*. (2021)

Coagulant	Benefits	Drawbacks	References
Aluminium sulphate (Alum) $Al_2(SO_4)_3{\cdot}18H_2O$	• Low sludge volume index	• Limited pH effective range • Introduces dissolved aluminium salt residues to the water	Zand & Hoveidi, 2015; Neukirchen & Ries, 2020; Thakur *et al.*, 2021
Sodium aluminate $Na_2Al_2O_4$	• Efficient in hard waters • Mostly, low concentrations are needed.	• High cost • Ineffective for the treatment of soft water	Tripathy *et al.*, 2019; Krupinska, 2020
Poly-aluminium chloride (PAC) $Al_{13}(OH)_{20}(SO)_4Cl_{15}$	• Rapid aggregation velocity • Bigger and heavier flocs than conventional Al(III) salts • Less dosage is required than conventional Al(III) salts • The pH of the treated water is unaffected.	• It has considerably lower molecular weight and size than organic polymers, and its stability to withstand subsequent hydrolysis is also significantly lower than for organic polymer flocculants	Gao *et al.*,2002; Abdo *et al.*, 2020
Ferric sulphate $Fe_2(SO_4)_3$	• Less sensitive to overdosage than aluminium-based coagulants	• Very caustic • It causes rust-coloured stains that are visible and related to chemical spills and leaks. • Depletes alkalinity	Hassan, 2005; Sadeghi, *et al.*, 2017; Bahrodin *et al.*, 2021
Ferric chloride $FeCl_3{\cdot}6H_2O$	• Effective in a broad range of pH between 4 and 11	• Salts are added as dissolved solids to the water. • Doubles the amount of alkalinity that alum consumes	Bouchareb *et al.*, 2020; Bouchareb *et al.*, 2021

Ferrous sulphate $FeSO_4 \cdot 7H_2O$	• It is less sensitive to pH than lime.	• Salts are added as dissolved solids to the water. • Normally, alkalinity must be added.	Marey, 2019
Lime $Ca(OH)_2$	• Do not add dissolved salts to the water	• pH-dependent • Produces vast quantities of sludge	Kumar *et al.*, 2017; Thakur *et al.*, 2021

The settling properties of flocs produced by coagulants are improved by coagulant aids. A coagulant aid that has previously been utilised is activated silica. Activated silica was utilised as a coagulant aid together with Poly Aluminium Chloride (PAC) for enhanced coagulation of raw water from Yellow River Water (YW) and South-to-North Water Diversion (SNW) in Zhengzhou, China, with turbidity removal efficiencies of up to 96.5% obtained at PAC dosage of 35mg/L and activated silica dosage of 7 mg/L for source water YW and SNW blended in the ratio of 2:1 (Lu *et al*., 2018). Other aids that have also been used are sodium alginates and some soluble starch products such as tapioca and sago which have been used for the removal of turbidity in river water achieving turbidity removal efficiencies of 93.7% at a dosage of 1000 mg/L and 96.4% at an optimum dosage of 2000 mg/L for tapioca and sago starch, respectively (Zainol *et al*., 2020). The fact that these compounds were well-known and had previously been employed in the food business gave them the benefit of being accepted as safe for water purification. Polyelectrolytes have also been utilised and consist of many synthetic products, particularly long-chain organic chemicals which can either be cationic, anionic, or non-ionic (Jabin & Kapoor, 2020).

2.4.2 Synthetic organic polymers

Synthetic organic polymers have been utilised for decades for the treatment of water and wastewater as flocculants and also as chemical aids for sludge dewatering (Heiderscheidt *et al.*, 2016). These high molecular weight polymers which are commonly referred to as polyelectrolytes are synthetic compounds that tend to strongly adsorb the particles on the surface in an aqueous suspension (Koshani *et al*., 2020). Utilising organic polymers in the process of treating drinking water can increase the settling rate, reduce costs and improve the water quality (Iwuozor, 2019).

Some of the advantages of using organic polymers are that they require lower dosages than metal salt coagulants and that they produce less sludge with better dewatering characteristics (Dayarathne *et al*., 2021). It has been observed that floc re-growth after breakage caused by initial turbulent mixing is almost fully reversible with cationic polymers while mostly irreversible when metal salt coagulants are used (Zhu & Dou, 2021). Moreover, the introduction of organic coagulants does not consume alkalinity; therefore there will be no need for any pH adjustments of the treated water (Naceradska *et al*., 2019) This was demonstrated by Sellami *et al.* (2014) in a study in which there was a slight change in pH from 4.94 to 4.56 when synthetic organic polymer polyacrylamide was used as a flocculant for the treatment of wastewater from the food industry. According to Bradney *et al.* (2019), there have been some concerns regarding the toxicity of synthetic polymers and their impacts on human health and also on the aquatic ecosystem which is a disadvantage of using synthetic organic polymers for water treatment. However, the toxicity of these products has mostly been linked to residual concentrations of non-reacted monomers and other contaminants that might be present in the treated water and are strictly limited by legislation worldwide (Heiderscheidt *et al.*, 2016).

2.4.3 Natural polymers

Natural polymers are readily available and environmentally friendly hydrophilic compounds that are also nontoxic to people (Tetteh & Rathilal, 2020). Many naturally occurring polymers possess intrinsic cationic properties which can be modified to give a cationic polyelectrolyte to be used for solid-liquid separations as flocculants (Sillanpää *et al.*, 2018). Examples of natural polymers are chitosan, starch, and cellulose derivates such as carboxymethyl cellulose (CMC), methylcellulose (MC), and chitin (Benabid & Zouai, 2016). Chitosan, like most natural polymers, is non-toxic which makes it generally accepted on a health basis (Dibyangana *et al*., 2015).

Plants are a primary source of natural polymers. Examples of plant-based coagulants are Nirmali seeds, *Moringa oleifera* seeds, Tannin, Eggplant seeds, and Radish seeds(Ndabigengesere & Narasiah, 1998; Chandrasekhar, 2005; Kango *et al*., 2013). The main merits of utilising natural plant-based coagulants as point-of-use water treatment mechanisms are that they are cost-effective, unlikely to produce treated water that has pH which is outside the WHO recommended range of 6.5 to 8.5, and are highly biodegradable (Patchaiyappan *et al*., 2020).

These benefits are particularly enhanced if the plant from which the coagulant is produced is naturally located nearby in a rural area.

The mechanisms of coagulation-flocculation when using natural coagulants are strongly linked to the compounds contained in the natural coagulants (Oladoja, 2015; Hasan *et al*., 2021; Igwegbe *et al*., 2021). The characteristics of proteins, polysaccharides, carbonyl, carboxyl, and hydroxyl groups were reported by Fard *et al.* (2021), and they correlate to the bridging mechanism used by the natural coagulant *Lallemantia royleana* in coagulation processes. Amines, carboxyl groups, and hydroxyl groups were stated in the study by Igwegbe *et al.* (2021), and these were also linked to the charge neutralization mechanism by the natural coagulant, *Picralima nitida*. Furthermore, in the study by Magalhães *et al.* (2021), it was noted that amides, amines, carbonyl, and methylene groups are related to the charge neutralization and patch flocculation mechanisms during coagulation-flocculation processes utilising *Moringa oleifera*.

Natural coagulants have the disadvantage of being unable to handle huge pH variations. Most coagulants work effectively within a specified pH range, but their efficacy is greatly affected outside of this range (Gautam & Saini, 2020). Another disadvantage of employing natural coagulants is that they sometimes require higher dosages than synthetic coagulants (Aliyu *et al*., 2015). The application of natural coagulants such as *Moringa oleifera* seeds in water treatment contributes to the surge of organic matter in treated water. This increase of organic matter in treated water is a disadvantage of using natural coagulants in water treatment because organic matter contributes to the development of carcinogenic trihalomethanes when water is disinfected using chlorine (Camacho *et al*., 2017).

2.5 Description of *E.goetzei* and current uses

E.goetzei, which is commonly known as *Goetze's Elephantorrhiza* is a small and deciduous tree or shrub which grows in countries like Botswana, Malawi, Mozambique, Namibia, South Africa, Tanzania, Zambia and Zimbabwe (Maroyi, 2017). *E.goetzei* is also known by the names *Elephantorrhiza elongata* Burtt Davy, *Elephantorrhiza rubescens* Gibbs and *Piptadenia goetzei* Harms. The generic name '*Elephantorrhiza*' means 'elephant root' because of the large underground stem which is common to some members of this genus. Its English common name, narrow-pod elephant root is about the pods which are long and flat sometimes measuring up to

30cm long and up to 4 cm broad and also large roots up to 8 metres long. In Ndebele, *E.goetzei* is known as *ililamba* or *intolwane* and in Shona is known as chiurayi, mugudzuru, muzezepasi and ndorani (Maroyi, 2011; Ngarivhume *et al*., 2015; Hyde *et al*., 2017).

E.goetzei is a plant which is multi-stemmed and has brown or reddish bark and smooth dark twigs which have conspicuous lenticels. The leaves are finely divided and have many small, narrow leaflets with clusters of small, cream-coloured flowers which are produced along the bottom half of the aerial stem (Coates, 2002).

Research has shown that the extracts from the root bark of *E.goetzei* have antimicrobial properties (Moyo *et al*., 1999). *E.goetzei* is mainly utilised to treat pain and sores, sexually transmitted infections, gastrointestinal disorders, infections and genitourinary system disorders (Maroyi, 2017). The powder from the roots of *E.goetzei* is taken orally in porridge as a treatment for abdominal pains in Malawi, Mozambique and Zimbabwe. Root infusion of *E.goetzei* is also taken orally as an anthelmintic and treatment for depressed fontanelle in Zimbabwe (Sanghi *et al*., 2002). Furthermore, root decoction of *E.goetzei* is also taken orally as a treatment for bilharzia in Malawi and Zimbabwe (Shopo *et al*., 2022). Moreover, in Zimbabwe, the root infusion of *E.goetzei* is used as a treatment for heart pains and painful uterus to increase blood in the body. These reported properties of this plant have motivated the researcher to do further research on this plant.

2.6 Characterization of natural plant extracts according to the removal of key contaminants

The findings of investigations on the removal of turbidity, pathogens, and heavy metals from the water will be presented in this section.

2.6.1 Turbidity

To lower the turbidity of synthetic water using a standard jar test apparatus, Asrafuzzaman *et al.* (2011) evaluated the seeds of *Moringa oleifera, Cicer arietinum,* and *Dolichos lablab* for usage as locally accessible natural coagulants in Bangladesh. After introducing water-soluble extracts to the water, turbidity removal efficiencies obtained were 94.1%, 95.9%, and 88.9%, respectively. Seeds of *Cicer arietinum* treated the water to a final turbidity of 3.9 NTU which met WHO standards for turbidity in water as it was less than 5NTU whilst seeds of *Moringa* *oleifera* and *Dolichos lablab* achieved a final turbidity of 5.9 and 11.1 NTU. The natural coagulants in the study worked better with high turbidity water (90–120 NTU) in comparison to medium (40–50 NTU) or low turbidity water (25–35 NTU). These classifications of turbidity are according to Asrafuzzaman *et al*. (2011) and may differ from one study to another. Figure 2.2 shows the seeds of *Moringa oleifera, Cicer arietinum,* and *Dolichos lablab* which can be utilised in water treatment.

(a) *(b)* *(c)*

Figure 2.2 | Seeds of (a) *Moringa oleifera (Paisley Farm* & *Crafts, 2022)*, (b) *Cicer arietinum* (Sergeev, 2015) ,and (c) *Dolichos lablab* (Exporters India, 2022)

The turbidity reduction efficiency obtained by Asrafuzzaman *et al.* (2011) when *Dolichos lablab* was used for synthetic turbid water treatment at initial turbidity of 35 NTU was higher (60.9%) than that which was obtained by Antov *et al*. (2018) in which a turbidity removal efficiency of 23.0% was achieved when common oak acorn was used to treat synthetic turbid water at initial turbidity of 35 NTU. In another study which was done in Malaysia, natural coagulants *Azadirachta indica, Stenochlaena palustris, Dicranopteris linearis, Syzygium polyanthum, Manihot esculenta, Piper sarmentosum, and Melastoma malabathricum* achieved turbidity removal efficiencies ranging from 11.1% to 29.4% at an initial turbidity of 500± 50 NTU (Ahmad *et al*., 2022). The turbidity removal efficiencies obtained by Ahmad *et al*. (2022) were lesser than the 90.0% and 56.9% obtained for Holm oak acorn and Cactus juice respectively at a similar initial turbidity of 541±50 NTU (Adachi *et al*., 2023). In a separate study, a comparison of turbidity removal efficiencies for corn and potato starches in wastewater treatment was carried out at an initial turbidity of 121.6 NTU (Posada-Velez *et al*., 2023). A final turbidity of 3.0 NTU was achieved for corn starch and 3.3 NTU for potato starch . The final turbidity of the water after using both starches complied with the WHO guideline value of 5 NTU.

Another study by Ali *et al.*, (2010) produced higher removal efficiencies (96.2%) at initial turbidity of 34-36 NTU than the one obtained by Antov *et al.* (2018) for common oak acorn (23.0%) at initial turbidity of 35 NTU. A dosage of 0.4 mg/L resulted in a final turbidity of 1.32 NTU, which is under the advised WHO guideline value of 5 NTU, and a turbidity removal effectiveness of 96.2%.

2.6.2 Pathogens

In a study conducted by Okunlola *et al*. (2020), *Magnifera indica* seeds with a dosage of 500 mg/L were used to remove 100.0% of both total coliform and faecal coliform in stream water. Similarly, high removal efficiencies of total coliform (99.6%) and *Escherichia coli* (99.7%) were achieved when *Moringa oleifera* seeds with a dosage of 600 mg/L were used to treat domestic wastewater in Brazil (Andrade *et al*., 2021). In a separate study by Amran *et al*. (2021), *Carica papaya* seeds were used in Malaysia at a dosage of 196 mg/L to treat river water. Removal efficiencies of 57.6% and 62.1% were achieved for total coliform and *Escherichia coli,* respectively. These studies demonstrate that lower coagulant dosages achieved relatively lower pathogen removal efficiency whilst higher coagulant dosages also resulted in higher removal efficiency for pathogens.

Asrafuzzaman *et al.* (2011) also carried out a study in which they used seeds from the plants *Moringa oleifera, Cicer arietinum,* and *Dolichos lablab* to reduce the total coliform in turbid water by 89.0–96.0%. Total coliform in water was reduced by *Moringa oleifera* seeds from 1.35×10^3 cfu/100 mL to 5.4×10^1 cfu /100 mL. The total coliform was also reduced to 1.0×10^2 cfu/100 mL and 1.1×10^2 cfu/100 mL by *Cicer arietinum* and *Dolichos lablab*, respectively from 1.05×10^3 cfu/100 mL. The water samples that were treated using the three natural coagulants did not meet the WHO drinking water quality standards of 0 cfu/100 mL for total coliform in water and will therefore require further disinfection. Furthermore, a study by Ramavandi (2014) which was conducted in Iran also showed that faecal streptococcus was removed to almost 0 cfu/100 mL from turbid water using a coagulant extracted from *Plantago ovata*. However, the faecal coliform was not reduced to 0 cfu/100 mL and did not meet WHO drinking water quality standards (Ramavandi, 2014).

Tubers of *Maerua subcordata* and seeds of *Moringa stenopetala,* which are shown in Figure 2.3 were used in a study in which coagulation and microbial reduction experiments were done on surface river waters in Ethiopia (Megersa *et al*., 2016). Both plants managed to reduce microbial load (total coliform*, E. coli,* and heterotrophic bacteria). The five rivers that were sampled contained *E. coli* in the range of 153 to 166 cfu/100 mL. After being treated with the natural coagulants, four of these water samples had 0 cfu/100 mL, which complies with WHO drinking water quality requirements. However, one sample did not meet the WHO water quality standards as the final *E. coli* load after treatment with *Maerua subcordata* and *Moringa stenopetala* was 2 and 1 cfu/100 mL, respectively. Therefore, in this case, residual disinfection is very important to obtain zero colony-forming units in treated water (Pritchard *et al*., 2009). A greater percentage of total coliform removal (100%) was noticed for the lowest turbidity value of 20 NTU than 98.4% for the highest turbidity level of 195 NTU which was used in the study by Megersa *et al* (2016). This higher percentage of microbial load removal in lower turbidity water than in higher turbidity water may be due to the increase in particles suspended in the high-turbidity water that protect the bacteria from the action of the natural coagulants (Megersa *et al*., 2016). Sharma & Bhattacharya (2017) suggest that the presence of clay, silts or sand, or organic, algae, and leaf particles causes turbidity which may shield bacteria, thus preventing disinfection chemicals such as chlorine from invading and destroying cells.

(a) *(b)* *(c)*

Figure 2.3 | *Seeds of* (a) *Moringa stenopetala* (Moringa Matters, 2022) , (b) *Plantago ovata* (Botany Today, 2018) *and* (c) *Tubers of Maerua subcordata* (Gerald and Buff, 2002)

The antimicrobial effect of plant coagulants may be accredited to both flocculation (Nwaiwu & Lingmu, 2011) and bactericidal action (Oluduro *et al*., 2010). The presence of alkaloids and tannins in plant species could contribute to antimicrobial activities (Ejelonu *et al*., 2010). Bacterial re-growth was raised as a huge issue of concern with natural coagulants and can be made better by purification of the active agent, thus purification further removes additional nutrients and organic content (Ghebremichael, 2007).

2.6.3 Heavy metals

In a study done in Malaysia by Shan *et al*. (2017), 99.9%, 98.5%, 100.0% and 78.1% of copper, cadmium, iron and lead respectively were removed from the water samples collected from "The Sungai baluk" river when the seeds of *Moringa oleifera* were used as a natural coagulant for removal of heavy metals. Results from this study as shown in Table 2.2 indicate that increasing *Moringa oleifera* seed concentration resulted in high removal efficiency of heavy metals from the "Sungai baluk" river samples (Shan *et al*., 2017). Concerning the level of lead in the treated water, the result which was obtained by Shan *et al*. (2017), did not meet the drinking water standards of WHO (2006). The level of lead was lowered to 0.537 mg/L. This amount of lead was above the allowable limit, which is less than 0.05 mg/L. Similarly, river water was also used to determine the effectiveness of using *Moringa oleifera* seeds for water treatment (Subramanium *et al*., 2011). Removal efficiencies of 50%, 90%, 80%, 60% and 50% were achieved for zinc, copper, lead, cadmium, and chromium respectively. In another study, banana pith coagulant removed 59.72% of manganese in river water (Kakoi *et al*., 2016). The water samples from the rivers were generally collected as grab samples which were then used for further analysis.

Table 2.2 | Removal of heavy metals in water using natural coagulants

Natural coagulant	Metal removed	Final Concentration (mg/L)	Level of removal (%)	WHO standards(mg/L)	Complied with WHO standards	Reference
Neem wood powder	Copper	0.5	95	2	Yes	Arshad & Imran, 2020
	Zinc	0.4	96	3-5	Yes	
	Manganese	0.6	94	<0.1	No	
	Lead	2.8	72	0.01	No	
	Chromium	2.5	75	0.05	No	
	Arsenic	0.75	25	0.01	No	
Pine cone powder	Copper	0.1	99	2	Yes	
	Zinc	0.3	97	3-5	Yes	
	Manganese	0.4	96	<0.1	No	
	Lead	1.2	88	0.01	No	
	Chromium	1.4	86	0.05	No	
	Arsenic	0.75	25	0.01	No	
***Carica papaya* leaf powder**	Lead	10.2	89.8	0.01	No	Seetha *et al.*, 2012
Opuntia ficus indica	Chromium	3.34	66.6	0.05	No	Mane *et al.*, 2011
	Nickel	1.16	88.4	0.02	No	
Moringa oleifera	Copper	0.001	99.9	2.00	Yes	Shan *et al.*, 2017
	Cadmium	0.029	98.5	0.003	No	
	Iron	0	100.0	No health-based guideline	Yes	
	Lead	0.537	71.0	0.01	No	
Dolichos lablab	Iron	<0.05	97.17	No health-based guideline	Yes	Kabir *et al.*, 2019

In a study by Skotta *et al.* (2023), *Lepidium sativum* seeds were used for removing copper and zinc in laboratory synthetic water samples. Removal efficiencies as high as 87.0% and 87.5% were achieved for copper and zinc, respectively. In another study in which synthetic contaminated water was used, the percentage removal of lead, cadmium and iron using *Moringa oleifera* seeds was 86.2%, 47.7% and 80.0%, respectively (Sajidu *et al.*, 2005). Synthetic contaminated water was also used to determine the efficiency of using *Moringa oleifera* seed for treating water contaminated with lead, cadmium and copper (Nwagbara *et al.*, 2022), with removal efficiencies of 90.0%, 81.8% and 70.0% being achieved for lead ,cadmium and copper, respectively.

Industrial wastewater was utilised to determine the efficiency of *Cassia fistula* seeds in removing zinc and nickel (Nguyen *et al*., 2023). Removal efficiencies of 40.1% and 34.6% were achieved for zinc and nickel, respectively. Similarly, industrial wastewater was also used to investigate the effectiveness of using pine cone powder for the removal of manganese, iron, zinc, aluminium and nickel. Removal efficiencies up to 86.8%, 94.0%, 89.7%, 73.7% and 86.7% were achieved for manganese, iron, zinc, aluminium and nickel respectively. *Moringa oleifera* seed extracts, chitosan and zeolite also achieved manganese removal efficiencies as high as 91.3%, 91.7% and 90.5%, respectively for treating industrial wastewater (Jagaba *et al.*, 2020). In separate studies, pine cone powder and rosehip seed powder were used to remove manganese in industrial wastewater and achieved manganese removal efficiency of 86.8% and 85.7%, respectively (Abujazar *et al*., 2022; Shadi *et al*., 2022).

Water containing copper, zinc, and iron was treated to meet WHO water quality standards among the heavy metals examined in this section, but water which had manganese, chromium, lead, arsenic, and cadmium did not meet WHO water quality standards after treatment with various natural coagulants like neem wood, pine cones, and *Moringa oleifera* seeds. Therefore, water which is contaminated with manganese, chromium, lead, arsenic, and cadmium would therefore require additional treatment to comply with WHO water quality standards.

2.7 Criteria for analysis of natural coagulants

Natural coagulants that are derived from plants were analysed by using environmental and technical criteria which include dosage, active ingredients, extraction method, sludge produced, toxicity, technical complexity, availability, and environmental impact as suggested by Omran *et al*. (2021).

2.7.1 Dosage

To achieve satisfactory water quality, it is crucial to ensure proper and effective control of coagulation, which serves as the initial stage in conventional treatment. One of the key considerations in determining optimal conditions for the coagulation-flocculation process is the dosage of the coagulant (Saritha *et al*., 2017). The doses of coagulants have a significant impact on the coagulation and flocculation processes, and it is crucial to consider this parameter for optimization. Insufficient dosage can result in inadequate flocculation and the inability to achieve water quality targets (Mukheled, 2012). On the other hand, excessive amounts of coagulant increase treatment costs and are not economically favourable to use. Overdosing can also cause charge reversal and the subsequent re-stabilization of destabilized particles, especially when the mechanism of turbidity removal is associated with adsorption and charge neutralization (Megersa *et al*., 2016). Moreover, excessive dosing during the coagulation-flocculation process can lead to the generation of excessive sludge, leading to additional costs for sludge treatment. Furthermore, according to Wang *et al.* (2021), overdosing can lower the pH of the treated water, thereby increasing the potential for corrosion within the water distribution system.

Achieving the desired water quality in the treatment process requires determining the optimal dosage of coagulant, which is the minimum amount needed. The control of coagulant dosage is influenced by various factors, including the quality of the raw water, such as its colloidal concentration, pH, alkalinity, hardness, temperature, Natural Organic Matter (NOM) concentration, and ionic strength (Sheng *et al*., 2023). The suitability of the coagulant dosage is typically evaluated by testing the quality of the treated water. Despite its importance, controlling coagulant dosage in large-scale treatment plants faces challenges that can impede overall treatment efficiency.

To remove turbidity, dragon fruit foliage was employed as a natural coagulant. The turbid water was prepared in the laboratory with an initial turbidity of 55 NTU. The optimum coagulant dosage was found to be 250 mg/L with final turbidity of 9 NTU and will therefore require further treatment to achieve the desired WHO standard of 5 NTU in drinking water (Sciban *et al*., 2005; WHO, 2017a; Rayudu *et al*., 2022). The results from the study by Sciban *et al.* (2005) showed that seeds of common bean *Phaseolus vulgaris* and indigobush *Amorpha fruticosa* at a dose of 5 mg/L were best used for treating water with a turbidity of 17.5 NTU, which was the lowest turbidity in the study. The extracts of the seeds of black locust *Robinia pseudoacacia* had coagulation activity (greater than 50%) in water with a turbidity of 70 NTU in a dose of 30 mg/L and for lesser turbidity, lower dosages have a better performance. The optimum coagulant dosage obtained by Taiwo *et al*. (2020) when treating water with turbidity less than 50 NTU using *Moringa oleifera* seeds was 60 mg/L. The turbidity of the water dropped significantly ($p<0.05$) at this dosage, from 41.19 ± 1.5 NTU to 23.51 ± 1.03 NTU.

Seven plant species that are native to Malaysia were investigated for their potential to treat the turbidity of water. The plants are *Azadirachta indica, Stenochlaena palustris, Dicranopteris linearis, Syzygium polyanthum, Manihot esculenta, Piper sarmentosum, and Melastoma malabathricum* (Ahmad *et al*., 2022). The optimum coagulant dosage for *Melastoma malabathricum* was 0.1g/L with turbidity removal of 12.2%. As for *Azadirachta indica, Stenochlaena palustris, Syzygium polyanthum,* and *Dicranopteris linearis,* the optimum coagulant dose was 10 000 mg/L with the removal efficiency being 26.9%, 24.9%, 24.9% and 17.5% respectively. *Piper sarmentosum* and *Manihot esculenta* achieved optimum coagulant dosages of 5000 mg/L with removal efficiencies of 24.2% and 22.0% respectively. The low turbidity removal efficiencies can be attributed to the water extraction method that was used therefore the ingredients that are responsible for coagulation were not adequately extracted (Ahmad *et al*., 2022).

Moringa oleifera leaf extracts were used in a study by Pandey *et al.* (2020) for the removal of turbidity, fluoride, and *E. coli* in water. The initial values of turbidity, fluoride, and *E. coli* in water were 15.6 NTU, 2.3 mg/L, and 315 cfu/100 mL respectively. After treatment at a dosage of 150 mg/L, *Moringa oleifera* leaves reduced the turbidity, fluoride, and *E. coli* to 6.1 NTU, 1.1 mg/L, and 41 cfu/100 mL respectively. Turbidity and *E.coli* did not meet WHO standards of 5 NTU and 0 cfu/100 mL respectively whilst fluoride met the standard of 1.5 mg/L fluorides in drinking water (WHO, 2017a).

While dosage control is relatively straightforward when the raw water quality remains stable, this is often not the case, particularly for water sourced from rivers. The dosing of coagulant is closely tied to the chemical and physical characteristics of the raw water. Consequently, the fluctuations in raw water quality due to diurnal patterns, seasonal changes, and storm events present significant difficulties in dosage control (Delpla *et al*., 2023).

2.7.2 Active ingredients

The active ingredients of *Moringa oleifera* seeds were discovered to be soluble cationic proteins possessing a molecular weight of about 13 kDa and isoelectric pH values of 10 and 11 (Ndabigengesere *et al*., 1995). The amino acid sequences of this protein have already been determined (Gassenschmidt *et al*., 1991,1995). Legumes mainly consist of proteins, which are stored in seed organelles to give nutrition for the growth and development of seeds (Duranti & Gius, 1997). These proteins can be categorized as storage proteins based on their solubility characteristics. There are albumins, water-soluble proteins, globulins which are soluble in dilute salt solutions, prolamins which are soluble in alcohol/water mixtures and glutens which are soluble in dilute acid or alkali (Shewry *et al*., 1995). It is well known that globulins make up the majority of the proteins in legumes. When utilised as a natural coagulant, these protein levels in legumes are crucial. It is known that peanut *(Arachis hypogaea)* seeds have arachin and coarachin which are globulin proteins (Kristianto, 2017). *Moringa oleifera* seed is a well-known natural coagulant and its activity is because it has a water-soluble protein lectin (Ferreira *et al*., 2011). The active ingredients in the *Moringa oleifera* seeds are also reported to be dimeric proteins. The protein powder is stable and soluble in water (Sapana *et al*., 2012). After lipids, the water-soluble proteins which range from 18.6 to 37.2% were reported to be the main components found in seeds of *Moringa oleifera* (Shewry *et al*., 1995; Duranti & Gius, 1997; Ferreira *et al*., 2011; Kristianto, 2017; Kumar *et al.*, 2022).

The results obtained from a study by Hussain *et al*. (2019) in which pine cone extract was used as a natural coagulant for the purification of turbid water, show that pine cone extracts consist of both cationic and anionic coagulating components. The coagulant's anionic components are in charge of removing turbidity at lower pH levels. Similar to this, cationic protein components become activated at higher pH levels. Some studies have discovered that rice hull ash has the potential to be used as a coagulant aid (Abo-El-Enein *et al*., 2011; Adams & Mulaba-Bafubiandi, 2014). This is possible because of the high content of silica in rice hulls. When the rice hull is carbonized, its ash possesses active silica that could give a stable solution with a negative surface charge which can aid aluminium or iron coagulants by forming bigger and denser floc (Abo-El-Enein *et al*., 2011). Moreover, starch can be utilised as a natural coagulant due to its low cationic polyelectrolyte properties (Oladoja, 2015). Table 2.3 shows the active ingredients from various coagulants which are responsible for the coagulation process.

Table 2.3 | Active ingredients from various plant seeds responsible for coagulation in water treatment

Plant seed	Key ingredients	Content (%)	Turbidity removal efficiency (%)	References
Picralima nitida	• Crude Fibre • Crude Fat • Crude protein • Carbohydrate	• 10.5 • 7.4 • 28.4 • 31.1	86.6	Igwegbe *et al.*, 2021
Moringa oleifera	• Protein • Crude fibre • Fat • Carbohydrate	• 47.0 • 6.8 • 25.5 • 8.3	99.3	Adesina *et al.*, 2019
Hibiscus esculentus	• Fat • Protein • Carbohydrate • Crude Fibre	• 11.0 • 23.0 • 33.0 • 13.5	94.0	Okolo *et al.*, 2021
Jatropha curcas	• Protein • Fat • Fibre • Carbohydrates	• 28.4 • 50.5 • 10.1 • 1.2	93.0	Abidin *et al.*, 2017
Phoenix dactylifera	• Fat • Protein • Fibre	• 5.4 • 4.9 • 19.3	75.0	Kuhiyop *et al.*, 2020
Magnifera indica	• Protein • Fibre • Fat	• 6.8 • 8.0 • 3.6	89.0	
Pomegranate	• Carbohydrate • Protein • Lipid • Fibre	• 73.5 • 14.7 • 15.4 • 24.6	87.2	Shabanizadeh & Taghavijeloudar, 2023
Garcinia kola	• Protein • Carbohydrate • Fat	• 11.3 • 68.3 • 3.0	81.9	Igwegbe *et al.*, 2021
Pumpkin	• Protein • Fat • Fibre • Carbohydrate	• 29.1 • 1.0 • 5.7 • 61.1	87.7	Gatew & Worku, 2023

Banana pith powder has also been utilised as a natural coagulant for treating river water and it produced commendable results for the removal of turbidity, suspended solids, and some heavy metals (Kakoi *et al.*, 2016). This was made feasible by the banana pith's numerous functional groups, which include hydroxyl, carboxylic and ketone, aldehyde, and lactone groups. Inulin content in banana pith juice was the active coagulant agent (with a high bonding capacity to form bridges of flocs) when banana pith juice was utilised to treat spent coolant wastewater

(Alwi *et al*., 2013). Papaya seeds are well known for their antimicrobial activities because it has many phytochemical contents (Peter *et al*., 2014). Approximately 10% of papaya seed protein is of the albumin type, which can also act as a coagulant agent (Kristianto, 2017). Jones & Bridgeman (2016) state that proteins are responsible for turbidity removal when okra seed is utilised as a coagulant. Recently, proteins of white mustard seed have been utilised to remove direct black 19, an anionic dye found in textile wastewater, with removal efficiencies of up to 90.9% being achieved (Tie *et al*., 2019).

The active ingredients for most natural coagulants that have been studied for water treatment in several studies are mainly proteins (Ndabigengesere *et al*., 1995; Ferreira *et al*., 2011; Ramavandi, 2014; Choudhary & Neogi, 2017; Kristianto, 2017).

2.7.3 Extraction method

Many methods have been used to extract active ingredients from natural coagulants. These include water and salt extraction, ultrasound-assisted extraction, and extraction using hexane, acetone, and ethanol. A method for extracting the active ingredient for coagulation from *Moringa oleifera* seeds was developed in which a 1.0 mol/L solution of sodium chloride and other salts including potassium nitrate, potassium chloride, and sodium nitrate were utilised and then compared with the conventional water extraction method (Okuda *et al*., 1999). With doses 7.4 times lower than those using the ordinary water extraction method, the method that employed salts for extraction showed better coagulation activity for the removal of kaolinite turbidity. The results obtained in the study by Okuda *et al*. (1999) prove that the method for using salts for extraction efficiently coagulated more than 95% of the 50 NTU initial kaolin turbidity using only 4 mL/L, while 32 mL/L of coagulant extracted using the ordinary water method removed about 78% of the same kaolin turbidity. The improvement of the efficiency of coagulation by sodium chloride is attributed to the salting-in mechanism in proteins whereby a salt increases protein-protein dissociations resulting in increasing protein solubility as the salt ionic strength increases (Okuda *et al*., 1999).

In another study, both water extraction and salt extraction using the salt sodium chloride were used to extract proteins from *Moringa oleifera* seeds (Bouchareb *et al*., 2021). The investigation proved that there was a larger amount of coagulant agent retrieved from and dissolved in sodium chloride than in distillate water.

A de-oiled peanut seed extract has been used for palm oil mill effluent treatment. The active component responsible for coagulation was extracted using three different concentrations of sodium chloride which are 0 mol/L (distilled water), 1, and 2 mol/L. The greatest total suspended solids (TSS) removal and chemical oxygen demand (COD) reduction was achieved at 2 mol/L sodium chloride extract with 94.7% and 73.5% reduction respectively. When distilled water was used for extraction of the peanut seeds a lesser removal efficiency of 90% of TSS and 66% reduction of COD were obtained (Birima *et al.*, 2015). Table 2.4 shows different extraction methods which are utilised when using natural coagulants for water treatment.

Table 2.4 | Extraction methods used when utilising natural coagulants for water treatment

Coagulant	Extraction method	References
***Moringa oleifera* seeds**	• Salt (NaCl, KNO_3, KCl, $NaNO_3$) • Water	Okuda *et al.*, 1999 Bouchareb *et al*, 2021
	• Hexane • Acetone • Ethanol	García-Fayos *et al.*, 2016 Camacho *et al.*, 2017
Peanut seeds	• Water • Salt (NaCl, KNO3, KCl, NH4Cl and NaNO3)	Birima *et al.*, 2013
	• Salt (NaCl) • Water	Birima *et al.*, 2015
***Plantago ovata* seeds**	• Salt ($FeCl_3$)	Ramavandi, 2014 Ramavandi *et al*, 2015
	• Water • Salt (NaCl) • Ammonium acetate	Dhivya *et al.*, 2017
Opuntia ficus-indica	• Ultrasound-assisted extraction (UAE) • Water	Gomes *et al.*, 2016 Adjeroud- Abdellatif *et al.*, 2020
Pine cone	• Water	Hussain *et al.*, 2019

In a study by Ramavandi *et al.* (2015), salt extraction using ferric chloride was used for the extraction of proteins from the seeds of *Plantago ovata*. Adjeroud-Abdellatif *et al.* (2020) made use of ultrasound-assisted extraction to extract proteins from the natural coagulant *Opuntia ficus-indica* by subjecting the coagulant to ultrasound waves by a sonicator probe. The most common extraction methods in several studies include water and salt extraction (Okuda *et al*., 1999; Birima *et al.,* 2015; Ramavandi *et al*., 2015; Hussain *et al*., 2019; Bouchareb *et al*., 2021). Salt extraction method produced better results than water extraction methods in several studies in which both of the methods were used (Okuda *et al*., 1999; Birima *et al*., 2015; Vishali & Karthikeyan, 2015; Benalia *et al*., 2018; Behera & Balasubramanian, 2019).

2.7.4 Sludge produced and how safe it is for reuse

Ndabigengesere *et al.* (1995) determined the efficiency of *Moringa oleifera* seeds as a natural coagulant in water treatment and compared it with alum. For a dosage of 500 mg/L of coagulants used in the study, the ratio of the volume of sludge produced in the case of alum, shelled and non-shelled *Moringa oleifera* seeds extract was 5: 2.5: 1, respectively. The problem of sludge after coagulation using *Moringa oleifera* seeds is dependent on the nature of the coagulated materials. The advantage, in this case, is that all *Moringa oleifera* by-products are organic and biodegradable. Furthermore, the sludge produced by coagulation with *Moringa oleifera* is non-toxic and is also less in volume by about four to five times than the chemical sludge which is produced via coagulation using alum. These differences in sludge volume can be attributed to the presence of aluminium hydroxide which precipitates when alum is introduced under specific conditions, trapping the colloids via the sweep coagulation process, and thereby increasing the sludge total solids and total fixed solids content from alum sludge. On the other hand, sludge from natural coagulants such as *Moringa oleifera* only emanates from the dosage and suspended particles (Vega *et al*., 2021).

In another study, the efficiency of two natural coagulants, namely calcium lactate and tannic acid was assessed and compared to conventional coagulants; namely, polyaluminium chloride and ferric chloride (Kaji *et al*., 2020). An initial turbidity of 900 NTU was used to test the four coagulants to compare the sludge produced. After 30 minutes, it was observed that the two natural coagulants created less deposited sludge than the other two chemical coagulants. It was therefore noted that the utilization of natural coagulants results in less sludge produced which is also biodegradable.

2.7.5 Toxicity

Toxicity is also another factor that can be used to assess natural coagulants. Genotoxicity by definition is the destructive effect on the genetic material of a cell thereby affecting its integrity

(Shah, 2012). Genotoxins have the potential to cause cancer, mutations, or birth defects. Cytotoxicity is also another form of toxicity which is defined as the effect of being toxic to cells caused by toxic agents and may actively progress into the death phase (Salih *et al*., 2019). The investigation of cytotoxic, genotoxic, or mutagenic effects of compounds found in plants could significantly reduce the possible risks of these agents to people's health.

An acute toxicity test by Igbokwe *et al.* (2018), proved that *Moringa oleifera* seeds are safe for consumption since an LD $_{50}$ value of 14 g/kg was obtained using 25 Swiss albino mice with weights varying from 20g to 25 g. In another study, an assessment of the genotoxicity of an aqueous seed extract of *Moringa oleifera* was carried out using three separate assay systems including the cell-free plasmid DNA, Ames, and Kado assay (Rolim *et al*., 2011). The study's findings demonstrated that the seed extract was not genotoxic in the absence of metabolic activation, and did not pose a threat to people's health. An evaluation was also carried out of the cytotoxicity of an aqueous extract of *Moringa oleifera* seeds (Araújo *et al*., 2013). After administering the extract (500 and 2,000 mg/kg) in mice for 14 days, no signs of systemic toxicity were noted and there was 100 % survival of the animals. Similarly, in another study, it was noted that *Moringa oleifera* seed cake is a non-toxic agent that is used for water treatment (Arnoldsson *et al*., 2008). Likewise, chitosan is a natural biodegradable linear copolymer that is the most abundant and non-toxic (Aider, 2010).

In another study, acute and chronic toxicity studies of aqueous extract and seed powder of *Strychnos potatorum* Linn seeds which are used for treating turbid water were done in Wistar albino mice and rats (Sanmugapriyaa & Venkataraman, 2006). The animals did not exhibit any toxic effects up to the dose of 2,000 mg/kg which was administered by mouth. In a separate study, doses as high as 2000 mg/kg of seed extracts of *Cicer arietinum* were orally given to normal rats (Doppalapudi, 2019). No mortality was observed with oral administration of all the seed extracts of Cicer *arietinum* even at a dose of 2000 mg/kg.

From the studies that were considered, it is evident that various natural coagulants such as *Moringa oleifera*, *Strychnos potatorum* Linn, and *Cicer arietinum* are non-toxic and can therefore be safely used for water treatment.

2.7.6 Technical complexity

The turbidity of surface water from rivers was reduced using a simple household technique that involved adding 0.25g of *Moringa oleifera* seed powder to 1 litre of water in a container and aggressively swirling for 20 to 25 seconds. After allowing the water to rest for an hour to allow for sedimentation, the supernatant was decanted and put through filtering using a fine mesh cotton cloth (Varkey, 2020).

According to Babu & Chaudhuri (2005), seeds of *Strychnos potatorum* and *Moringa oleifera* can be utilised easily at the household level since the implementation procedure does not require a high level of technical expertise. An uncomplicated setup for treating water at the household level would be to dose 10 L of contaminated water with *Strychnos potatorum* seed or *Moringa oleifera* seed as a coagulant (2% suspension prepared with distilled water). This is then followed by mixing via hand-stirring for a period of 2 to 3 minutes and filtering the water directly via a sand filter. The sand filter can be composed of 1,200 mm long and 150 mm internal diameter PVC pipe packed up to a depth of 500 mm using fine river sand, with 20-30 mm of pea-sized gravel at the bottom and arrangement for acquiring an initial flow rate of 3 L/min. About 2 or 3 filter runs can be done every day. The sand in the filter must then be removed, thoroughly cleaned with water, and put back in the filter when the rate of filtration has significantly decreased (Babu & Chaudhuri, 2005).

Another different way in which *Moringa oleifera* seeds can be easily used for household water treatment is firstly removing the wings from the dry seeds and grinding them to a powder (Price, 2007). The powder is then mixed with water, agitated for about five minutes, and filtered via a piece of woven fabric after approximately an hour to get treated water. Another technique is to leave a cloth with seed powder immersed in water, usually overnight, to allow contaminants to coagulate. The purified water is then decanted, leaving the coagulated particles at the bottom, and the fabric that will hold the seeds is then removed. Up to about 99% of colloidal material can be eliminated and only one seed is required per litre for slightly polluted water and two seeds for heavily polluted water (Foidl *et al*., 2001).

Burgos-vergara *et al*. (2020) explored the use of *Moringa oleifera* and *Cassia Fistula* in villages in Colombia. The seeds of *Moringa oleifera* and *Cassia Fistula* were both peeled and ground using a laboratory grinder and a special seed grinder, respectively. A fine powder of each of the seeds was stored in Ziploc bags to avoid absorption of humidity. 10 g of each powder was then added to 1 litre of raw filtered river water in two 2-litre plastic bottles. The plastic bottles were shaken rapidly (fast mixing) for 1 and 2 minutes respectively then for 5 minutes more slowly. To achieve a full dissolution of the natural compounds, the water and coagulant were mixed manually using homogenous movements from top to bottom and bottom to top. The water was left for 2 hours to allow the sedimentation process to take place.

According to Marobhe *et al.* (2007), natural coagulants can also be implemented at household level using the traditional water purification method. The method starts with the seeds being finely ground using a specialized grinding stone and stored in dry plastic bags for 1 to 2 months for daily use. To treat the 20-liter bucket of drinking water, 2 to 5 teaspoons (approximately 20 to 50 grams) of seed powder are added. The water is rapidly mixed at a speed of about 100-150 revolutions per minute (RPM) and left to swirl for approximately 5 to 10 minutes. Afterward, the formed flocs settle for 20 to 25 minutes. The clarified water is then carefully poured off and stored in a clean clay pot in a cool location (Marobhe *et al*., 2007).

In terms of the practical application of natural coagulants, the studies that were considered show that the coagulants require minimal technical expertise to implement and can therefore be easily implemented even in low-income communities.

2.7.8 Availability

Previous studies have demonstrated the availability and extraction of natural coagulants from a variety of sources, including waste biomass, natural products, and even microbes (Kanmani *et al*., 2017; Lee & Chang, 2018; Saleem & Bachmann, 2019). Thus, it can be inferred that some naturally occurring coagulants have dependable and readily accessible sources. Since plants like *Moringa oleifera* only produce mature, dried seeds twice a year, maintaining a consistent supply of raw materials that meet the minimum quality requirement is an inconvenience when considering using natural coagulants for large-scale water treatment facilities (Saleem & Bachmann, 2019).

2.7.9 Environmental Impact

Water utilities may be interested in using biodegradable natural coagulants that can reduce sludge production while also not adding to the sludge's toxicity. If the organic sludge produced by natural coagulants could be used for other tasks, such as wastewater treatment, in the construction industry for producing building materials such as cement, concrete, and mortar, for agricultural use as soil conditioners, and land-based uses, it might provide environmental sustainability (Ahmad *et al*., 2016).

Rarely is the secondary waste produced by the chemical-based extraction, purification, or synthesis of natural coagulants highlighted. Natural coagulant residues are regarded as organic materials that may interact with disinfectants which would have been produced by a subsequent disinfection process to produce carcinogenic disinfection by-products. Therefore, natural coagulants should be used cautiously if a downstream disinfection process is available (Ang & Mohammad, 2020).

2.8 Coagulation performance of conventional chemical coagulants as compared to coagulants derived from plant extracts

A comparative study was done on the effectiveness of extracts of *Cactus Opuntia* pads and *Moringa oleifera* seeds and a chemical coagulant in the treatment of raw water (Gandiwa *et al*., 2020). The results indicated that alum decreased the turbidity from 28.4 to 2.4 NTU, thus achieving removal of 91.5% at an optimum dosage of 45 mg/L. *Moringa oleifera* decreased the turbidity to 3.2 NTU thus obtaining a turbidity removal of 88.7% at an optimum dosage of 50 mg/L and *Cactus Opuntia* managed to reduce the turbidity to 3 NTU thereby decreasing the turbidity by 89.4%. This shows that alum had the highest turbidity removal efficiency. The observed minimum turbidities for each of the coagulants were less than the WHO guidelines of 5 NTU (WHO, 2017). When *Cactus opuntia* extracts were shown to be successful at removing turbidity from water, adsorption, and interparticle bridging were identified to be the main mechanisms for floc production (Nharingo *et al*, 2015; Gandiwa *et al.*, 2020). *Moringa oleifera* seed protein is also reported to have natural coagulating properties which are based on an adsorption mechanism (Nouhi *et al*., 2019).

In a separate study by Tsamo *et al.* (2021), alum had the highest turbidity removal efficiency in raw water at 72.0% compared to rice husk, cypress leaves, and eucalyptus leaves which had turbidity removal efficiencies of 53.6%, 40.6%, and 22.1%, respectively. However, the study carried out by Elsayed *et al.* (2020) demonstrated that seed extracts of Fenugreek had a turbidity removal efficiency of 98.0% in synthetic turbid water as compared to alum which had a lower turbidity removal efficiency of 85.0%. The flocculation process using Fenugreek extract could have been accomplished by adhesion mechanism, in which the proteins can adhere on the surface directly and bridge the kaolin particles, and thus encouraging their flocculation (Elsayed *et al*., 2020). The coagulation-flocculation process using rice husk could have occurred due to charge neutralization and particle's bridging mechanism (Huzir *et al*., 2019).

Moreover, another comparative study was carried out of using cactus powder and alum to analyse physiochemical parameters in synthetic water samples (Beyene *et al*., 2016) . The water samples were tested for TDS, pH, conductivity, salinity, and turbidity by making use of the jar test. The results from the study showed that the percentage reduction of turbidity from a water sample which was turbid was elevated from 23.9% to 54.0% and 28.5% to 58.2% as the dosage increased from 1000 mg/L to 7000 mg/L for cactus powder and alum, respectively (Beyene *et al*., 2016). The pH of the water was relatively unaffected when using cactus powder as compared to alum. The initial pH of the water sample was 7.81. Although the cactus powder dosages were significantly raised, the final pH values of the water sample were relatively unchanged, being 7.57 at 7000 mg/L dosage as opposed to alum, whose dosage went from 1000 mg/L to 7000 mg/L, causing the water's pH to drop to 4.18 (acidic). This decrease in pH can consume the alkalinity and an alkalizing agent may have to be introduced to maintain the hydrolysis reactions (Napacho & Manyele, 2010; Hashim *et al*., 2013). Therefore, extra costs could be incurred in maintaining the water in a neutral state.

When cactus powder and alum were used as coagulants for a water sample in the study by Beyene *et al* (2016), there was an increase in TDS, conductivity, and salinity of the water sample. Cactus powder increased the TDS, conductivity, and salinity of the water sample by 93.8%, 68.7%, and 97.1 %, respectively whilst alum increased the same parameters by 167.8%, 68.7%, and 180.0%, respectively. These results show that cactus powder was relatively better than alum in maintaining the TDS, conductivity, and salinity of the water sample (Beyene *et al*., 2016). In another study in which alginate was utilised as a natural coagulant, there was a negligible change in the pH and conductivity of the sample water (Saranya *et al*., 2022).

The initial pH of the untreated water in the study by Gandiwa *et al*. (2020), was 7.05, and when alum was introduced it decreased to 5.00 at a dosage of 60 mg/L alum. *Moringa oleifera* and *Cactus Opuntia* reduced the pH slightly to 6.4 and 6.5, respectively, when they were used at a dosage of 60 mg/L. These pH values which were obtained when *Cactus Opuntia* was used are within the WHO pH range of 6.5-8.5 for potable water (WHO, 2017). However, when *Moringa* *oleifera* was used the final pH of the water was slightly outside the WHO pH range for potable water. The conductivity of the water in the study increased by 51% as the alum dose increased from 0 to 60 mg/L. Likewise, the conductivity also increased by 38% and 36% for *Moringa oleifera* and *Cactus Opuntia,* respectively as the dosage was increased from 0 to 60 mg/L. The increase in electrical conductivity of the water as the coagulant dosage increases is because of the dissociation of the coagulant thus generating ions that increase electrical conductivity. *Moringa oleifera* and *Cactus Opuntia* have a lower capacity of dissociation in water as compared to alum and therefore the increase in conductivity is lower for these coagulants (Marobhe *et al*., 2007). In a separate study in which both natural and chemical coagulants were used for the treatment of palm oil mill effluent, it was observed that alum had the greatest dose requirement of 4000 mg/L as the optimum whilst *Moringa oleifera* had 2000 mg/L, chitosan 400 mg/L and zeolite 1000 mg/L (Jagaba *et al*., 2020).

From the studies that were analysed, it was observed that alum had generally higher turbidity removal efficiencies than natural coagulants such as cactus, rice husk, and eucalyptus leaves. However, treatment with some of the natural coagulants results in final turbidity which met WHO guidelines for drinking water quality. Natural coagulants were also generally better than alum in maintaining the physicochemical parameters of the water such as pH, TDS, conductivity, and salinity.

2.9 Potential for using natural plant extracts for water treatment

Various natural coagulants such as *Moringa oleifera, Cicer arietinum, Plantago Ovata, Maerua subcordata,* and *Carica papaya* have been investigated in various studies and have proved to be able to remove impurities such as chromium, nickel, cadmium, *Escherichia coli,* and turbidity (Megersa *et al.*, 2016; Arshad & Imran, 2020; Saranya *et al*., 2022). The successes of these studies at the lab scale call for further upscaling of the use of these coagulants at water and wastewater treatment plants. Moreover, an intriguing idea that might improve the coagulation process is the idea of multifunctionality. Coagulants that can remove both dissolved and undissolved pollutants can speed up the treatment process and provide several advantages, including cost savings and reduced environmental footprint. There is also potential to come up with hybrid coagulants by combining natural coagulants with other already existing coagulants to achieve favourable levels of removal efficiency for different contaminants.

Furthermore, since these coagulants are obtained from natural sources, the coagulants can be easily accessed (Ang & Mohammad, 2020).

2.10 Constraints of using natural plant extracts for water treatment

Plant-based coagulants have to go through some processes before they can be utilised for treatment unlike conventional chemical coagulants which are readily available for purchase and use (Kurniawan *et al*., 2020). The extraction of natural coagulants from plants depends on the characteristics of the plant in question but generally, the processes involve crushing, sieving, dialysis, washing, delipidation, removal of insoluble matter, filtration and ion exchange (Okuda *et al.*, 2001; Ali *et al.*, 2010). Therefore, this processing which is required every time they are used is a challenge since the conventional chemical coagulants that are being currently used are purchased and readily available to use. Further investigation should focus on how to retain active chemicals for long-lasting properties and turn them into powder form for simple handling and storage (Kurniawan *et al*., 2020).

Moreover, resources that are used as natural coagulants do not have to compete with other needs for which they have to be used such as basic foods. In addition, the use of these resources as coagulants has to be sustainable to avoid encountering scarcity challenges in the long-term application (Kurniawan *et al*., 2020). Therefore, considering the huge amounts of water being currently treated using conventional coagulants, it implies that a vast quantity of the plant extracts has to be exploited in the long-term application and this may not be sustainable in the long run. However, to deal with the issue of vast quantities of plant extracts being exploited, large plantations of the plants can be developed such that the plants are produced on a commercial scale and continuously to ensure viability.

Another constraint of using natural coagulants for water treatment is that in some cases, these natural coagulants have been proven to be good for high turbidity but limited efficiency has been attained for low turbidity water (Asrafuzzaman *et al*., 2011). Another challenge being faced in the use of natural plant extracts as coagulants is the lack of industrial confidence to adopt natural coagulants for water treatment due to concerns about the cost-effectiveness and performance consistency of the coagulants for large-scale water treatment (Ang & Mohammad, 2020).

In addition, most of the performance results of natural coagulants that have been reported have been limited to lab-scale studies and the move to real industrial applications has been hard to come by. The potential of natural coagulants to perform well under real application challenges such as fluctuating feed water quality, steady supply of natural coagulants with consistent quality, and storage and handling of the coagulants has not yet been established. Natural coagulants are also vulnerable to degradation where the active compounds such as proteins and polysaccharides may age during storage or be altered because of microbial attacks and external environmental factors during extraction and purification of the coagulants (Ho *et al*., 2014; Abidin *et al*., 2019). This will create a problem for the long-term storage of natural coagulants as the degradation of active compounds will affect the coagulation efficiency. Consequently, this has critical impacts on the commercialisation potential of natural coagulants. This is because the performance of the coagulants would have been affected and also due to transportation and storage problems for the coagulants (Ang & Mohammad, 2020). This can be addressed by coming up with ways to process the extracts and also adding preservatives which will ensure that the extracts can be stored for a long time without being altered or degraded.

One of the challenges that also comes with the use of natural coagulants is the release of organic load into the treated water (Sivaranjani & Rakshit, 2016). This results in the microbial decomposition of organic compounds causing colour, taste and odour. The problem is aggravated if the water is stored for long periods before it is used. In addition, further disinfection using chlorination is made impossible due to the presence of organics in the water. Furthermore, another disadvantage of using natural coagulants such as *Moringa oleifera* is that they require a longer sedimentation time than chemical coagulants and even though some of the natural coagulants have antibacterial properties that can treat *E.coli* infested water, the removal efficiency is not complete, therefore there is a risk of secondary bacterial growth (Sapana *et al*., 2012).

2.11 Opportunities and drawbacks for commercialisation of natural coagulants

Natural coagulants' ability to be commercialised successfully will always depend on whether the scale-up system can maintain comparable treatment performance at a similar or even lower cost when compared to established chemical coagulants. Given the various currency rates, inflation factors, and cost value accuracies, direct comparisons of coagulant types, processing stages, and costs across different geographic regions is a highly challenging assignment (Freitas *et al.*, 2018).

Financial large-scale use of natural coagulants is frequently a hurdle to the commercialisation of these products since potential investors are frequently wary of making investments in brand-new goods. The development of natural coagulants would be enhanced by knowledge of the risks and weaknesses involved. The results that have hitherto been published are mostly limited to the laboratory scale, which may not be sufficient for real-time applications (Bahrodin *et al.*, 2021). By putting a strong emphasis on the combination of natural and chemical coagulants, a breakthrough in this sector can start. Moreover, as a type of driving incentive to encourage water treatment operators to switch to the usage of natural coagulants, tax refunds, and subsidy programs could be made available (Choy *et al.*, 2014).

There is also a need for synergies between the research institutions researching these natural coagulants with the industries and municipalities that treat water and wastewater so that pilot studies can be done to determine how the natural coagulants perform in a large-scale water or wastewater treatment plant and the operating conditions under which they work.

Moreover, loans can also be given to those who would be willing to grow the plants used to obtain natural coagulants such as *Moringa oleifera* and *E.goetzei* on large tracts of land. The biggest obstacle to using plant-based natural coagulants in the field of sustainability is the large-scale growing of plants to meet industrial needs. Due to any required technological adjustments, the creation of new tools and machinery, and the development of new professional requirements, there will be an instant increase in industrial costs. But over time, there will be a significant decrease in the price of the process and the pollutants it produces. It can also help with the creation of jobs because those plants will need to be grown over large tracts of land (Freitas *et al.*, 2018).

Maintaining a consistent supply of raw materials that adhere to the minimum standard of quality is another significant obstacle to commercializing natural coagulants. Only twice a year, certain plants, like *Moringa oleifera*, yield mature, dried seeds; these seeds need to be processed and stored, which adds to the expense. Moreover, producing enough natural coagulants to take the place of chemical coagulants is also a challenge when trying to commercialise the use of natural coagulants (Saleem & Bachmann, 2019).

There is potential for these natural coagulants to be commercialised and made available on the market in the same way in which chemical coagulants are readily available on the market. For this potential to be realized, investigations should be done on how to process the natural coagulants so that they are used in powder form and also find ways to make sure that there is a constant supply of the natural coagulants on the market to ensure industrial confidence in the products.

2.12 Rationale for experimental approach

This section will present the justification for the experimental approach used in the study. Firstly, grinding of the *E.goetzei* seeds to a fine powder was done so as to achieve a smaller particle size, leading to a more reliable and consistent performance of the coagulant (Azamzam *et al*., 2022) . The use of powdered plant coagulants provides a larger surface area in comparison to whole leaves or extracts. Coagulants that possess a greater surface area are more desirable as they can enhance bridging mechanisms by offering a higher density of adsorption sites (Bahrodin *et al*., 2022).The utilization of a small sieve opening yields finer particles for the *E.goetzei* seed extracts coagulant, thereby leading to an expanded surface area. This expanded surface area can contribute to enhanced water treatment performance (Desta & Bote, 2021). Applying vacuum pressure generates a greater pressure difference across the filter medium, resulting in an increased flow rate and improved filtration effectiveness (Cescon & Jiang, 2020). Synthetic water was used to determine the removal efficiencies of fluoride, iron, manganese and turbidity in water. This ensured that experiments could be repeated successfully using the same initial concentration of contaminants in the water. In a lab setting, jar experiments simulate the floc production during coagulation-flocculation processes in water treatment. In addition to being used for research, jar tests are crucial for evaluating and optimizing the operational parameters of current water treatment plants as well as for establishing the design parameters of new plants (Pivokonský *et al.*, 2022).

2.13 Research gap

Over the years the potential of the seeds of *Moringa oleifera* in water treatment has been thoroughly investigated (Muyibi and Alfugara, 2003; Jones, 2019). Findings have shown that the *Moringa oleifera* seeds can be a feasible option to substitute, to some extent or totally, aluminium sulphate and other traditional chemical coagulants used in the treatment of water. The researcher decided to investigate a plant that is also indigenous to Zimbabwe, known as *E.goetzei* which belongs to the Leguminosae-mimosoideae family. Since extensive work has been carried out on the use of other natural coagulants such as *Moringa oleifera, Cicer arietinum*, and *Cactus Opuntia* the researcher decided to work on the use of *E.goetzei* seed extracts for water treatment since research has been done on other natural coagulants and little attention has been given to *E.goetzei* seed extracts in so far as water treatment is concerned.

2.14 Chapter summary

The literature reviewed in this chapter demonstrates that plant-based extracts have been successful in the removal of contaminants such as heavy metals, turbidity, and pathogens in water (Megersa *et al*., 2016; Shan *et al*., 2017; Hussain *et al*., 2019). To achieve the best pathogen removal results for some of the coagulants, the water has to be of low turbidity. Natural extracts can be used in conjunction with conventional methods of removing heavy metals in water such as the ion exchange process and adsorption-based separation. This approach is useful when natural coagulants cannot reduce the concentration of the heavy metals to acceptable WHO standards. Moreover, the literature reviewed in this chapter showed that natural coagulants can be analysed based on the dosage required for optimum removal of contaminants, the active ingredient in the coagulant, extraction method, nature of the biomass produced, determining whether it is safe to use as a soil enhancer based on its components and toxicity. The practical application of natural coagulants in communities has to be relatively easy so that even people in marginalized areas can easily adopt the technology. The literature analysed in this chapter concludes that the availability and accessibility of plant extracts could present a solution to especially to needy communities in developing countries.

Chapter 3: Materials and methods

This chapter presents the methodological approach that was taken to carry out the study. This includes preliminary studies on the *E.goetzei* plant, the experimental design used in the study and the analytical methods employed to collect the data.

3.1 *E.goetzei* preliminary studies

Structured interviews were carried out at Walden North Farm in Chegutu District, Mashonaland West Province, Zimbabwe, to find out about how the local people were using the *E.goetzei* plant (which is shown in Fig 3.1). The interview consisted of nine questions which included, but were not limited to, the amount of rainfall the plant needs to thrive, the time it takes to develop seeds, how it is being currently used and whether other people who are not locals come to collect the seeds or any other parts of the plant. The purpose of the interviews was to assess whether the local people were aware of the *E.goetzei* plant and whether it would have other competing uses when being used for water treatment. The researcher managed to conduct a total of 12 interviews with the ages of the interviewees ranging from 37 to 78 years. Since the farm has 26 plot-holders, systematic random sampling was used to select the participants. Some of the young local people had in-depth information about the *E.goetzei* plant and were therefore considered as participants for the interviews . Out of the 12 people who were interviewed, 6 were female and 6 were male and hence the interview was gender-balanced.

Figure 3.1 | *E.goetzei* tree growing at Walden North Farm, Chegutu District, Zimbabwe

The preliminary results that were obtained when the structured interviews were carried out to gain a better understanding of the *E.goetzei* plant from the local people at Walden North Farm in Chegutu District, Mashonaland West Province, Zimbabwe, are shown in Table 3.1. The most common uses of the *E.goetzei* plant extracts were excessive flatulence treatment followed by stomach ache treatment. The local people currently use the roots of the plant for the reported medicinal purposes.

Table 3.1 | Field visit report on *E.goetzei* at Walden North Farm

Interview Number	Age	Gender	Soil	Season	Rainfall needed	Seed development (months)	Seed dispersal (months)	Plant uses	Local Abundance	Non-locals who collect the plant	Other known places of occurrence
1.	40	F	Loam	Rainy	Did not notice	4	4	Flue treatment-roots are used	Plenty	No	None
2.	76	M	Red loam	Rainy	Average	4	4	Excessive flatulence treatment	Few	No	None
3.	72	F	Black/Red/ Sandy	Rainy	Depends on soil type	4-5	4-5	Stomach ache and running stomach treatment	Average	No	Chirimhanzu
4.	69	M	Red clay/ sandy	No season	Little	3	1	Treatment for back pains	Plenty	Relatives who visit local people	Chirundas-along Chegutu-Mubaira Rd
5	60	F	Sandy	No season	Little	4	Did not notice	Treatment for back pains and excessive flatulence	Plenty	No	Msengezi
6	37	F	Red clay	Rainy	Little	4	1	Excessive flatulence treatment	Average	No	None
7	50	M	Red soil	Rainy	Excess	6	2-3	Roots for treating stomach ache	Slightly above average	No	None
8	41	F	Red soil	Rainy	Little	2-3	4-5	Treating flue and excessive flatulence	Plenty	People from Nyatsanga	None
9	49	M	Red clay	Rainy	Little	6-8	2	Excessive flatulence treatment. Makes the immune	Average	No	None

								system active.			
10	49	M	Red soil	Rainy	Excess	5	2	Roots for excessive flatulence treatment.	Average	No	Msengezi, Zvimba, Makonde
11	65	M	Red soil/ rocky areas	Rainy	Average	3	1	Roots for treating stomach problems	Plenty	No	None
12	78	F	Sandy/ black soil/ Mountainous areas	Continuous	Excess	6-7	2	Treatment for coughing, running stomach and excessive flatulence	Average	No	Tordaf Farm, Katawa Farm (Chegutu District)

3.2 Research design

The summary of activities done for the research is shown in Fig 3.2. The main areas in the research framework include the *E.goetzei* reconnaissance visit, collection and preparation of the *E.goetzei* seed extracts, the preparation of the synthetic water which was used for the experiments, and the experiments which were conducted by the researcher to determine whether the seeds of *E.goetzei* are suitable for water treatment. The experiments include those that were done to determine the active coagulation ingredients in the seed extracts (proximate analysis), determination of *E.goetzei* seed extracts optimum coagulant dosage for turbidity removal, and removal efficiencies for iron, manganese, and fluoride (coagulation-flocculation experiments), determination of organic matter in treated water (determination of COD in treated water), and determination of the quality of the sludge produced by testing the nitrogen and phosphorus content in the sludge. COD was preferred to determine organic matter in water instead of Total Organic Carbon (TOC) since it detects the presence of all types of organic matter, including both biodegradable and non-biodegradable forms. This characteristic enables COD to assess the extent of pollution in water bodies caused by organic substances and therefore provides a broader assessment of the total organic matter present in the water (Islam *et al*., 2019). Each of these components of the conceptual framework is discussed in detail in subsequent sections of this chapter.

The experimental set up which was used for the study is shown in Fig 3.3. A Completely Randomised Design (CRD) was used in this research to determine the effect of *E.goetzei* seed extracts coagulant dosage on the turbidity, fluoride, iron , manganese, nitrogen, phosphorus, COD, pH, and TDS. The treatments included varying concentrations of coagulant dosage in the range of 0 to 300 mg/L for initial turbidity ranging from 12.33 NTU to 167.67 NTU. *E.goetzei* seed extracts coagulant dosage of 0 to 100 mg/L were used for iron, fluoride and manganese with concentration of 11.16 mg/L, 5.50 mg/L , and 5.46 mg/L respectively. A total of 16 samples were tested for each of the parameters pH, TDS, and each initial turbidity. In addition, 12 samples were tested for each of the parameters iron, manganese, and fluoride whilst 6 samples were tested for each of the parameters COD, total nitrogen and total phosphorus. All experiments were done in triplicates to ensure consistency of the results.

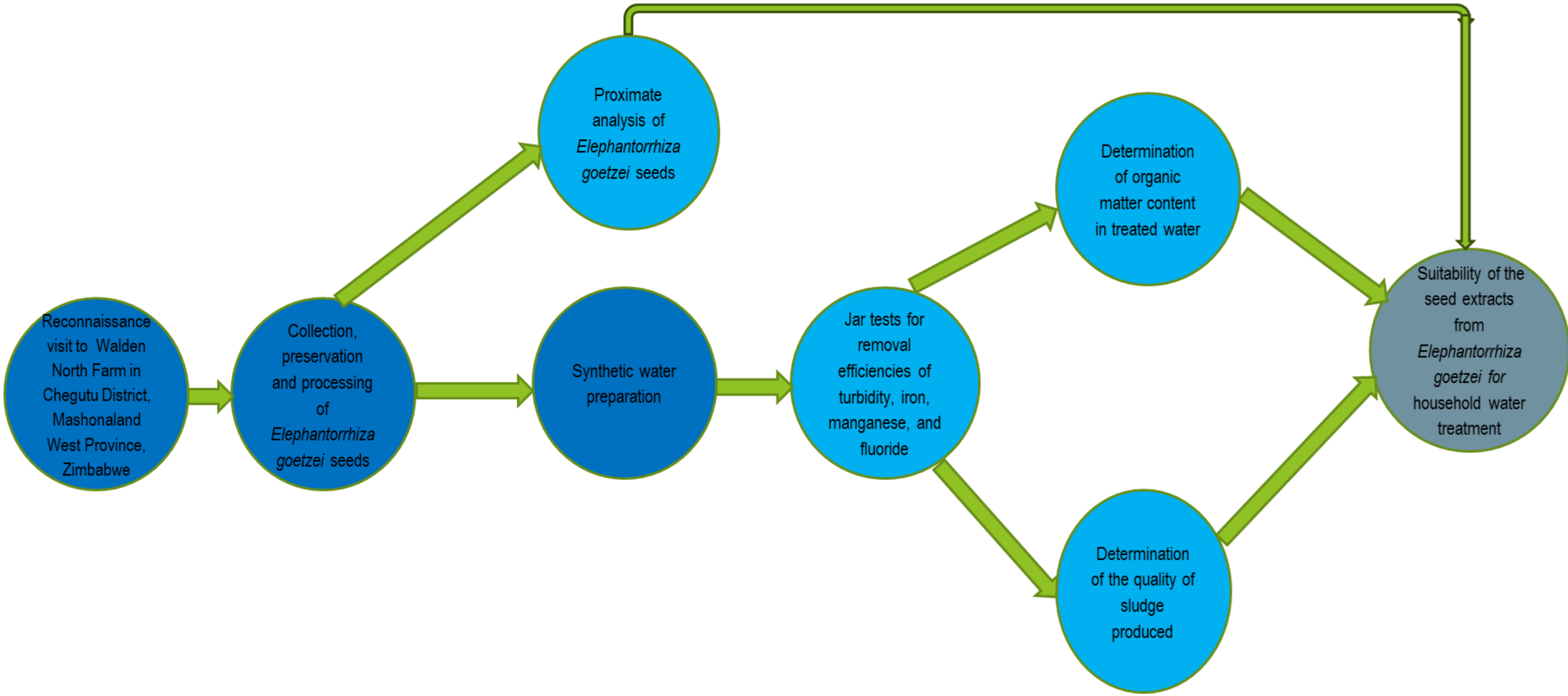

Figure 3.2 | Summary of activities done in the research

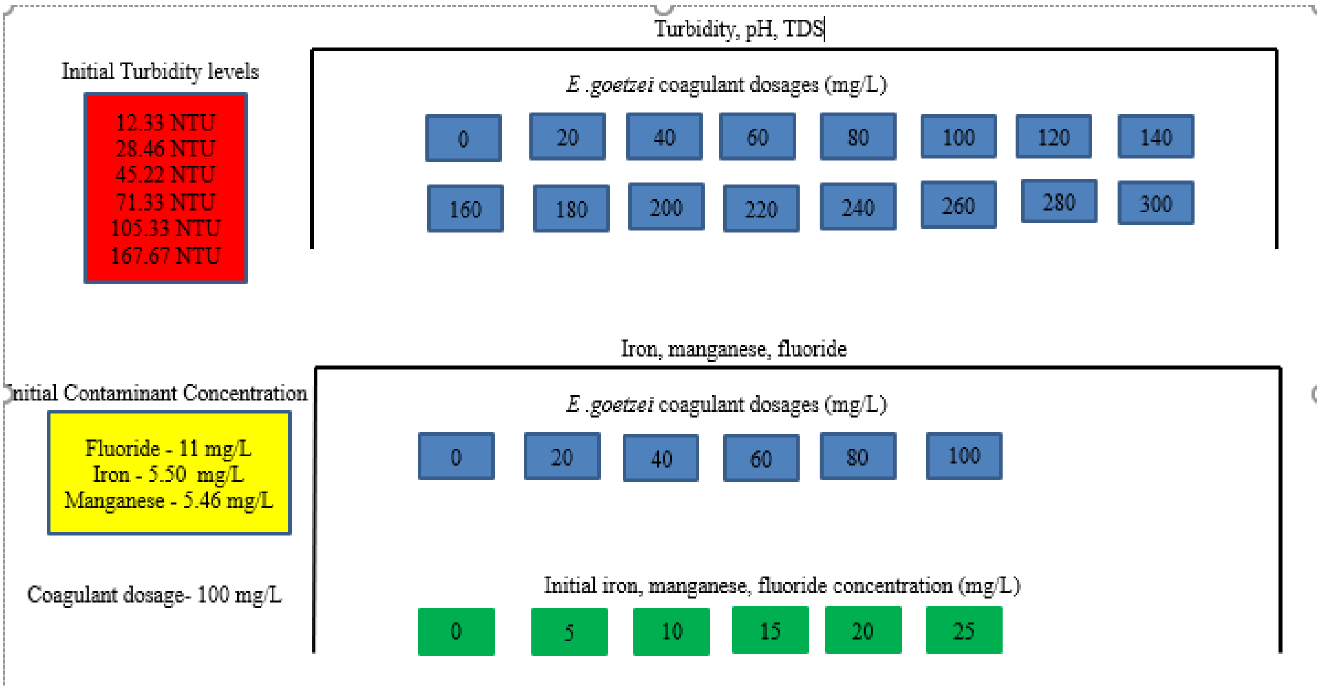

Figure 3.3 | Experimental set up for the research

3.3 *E.goetzei* seeds collection and preparation for proximate analysis

Brown *E.goetzei* seeds were collected from Walden North Farm in Chegutu District, Mashonaland West Province, Zimbabwe (S18.39239^o E030.41076^o). The researcher observed that the plants were mostly found on red clay soils in rocky and mountainous areas. The seeds were handpicked manually whilst they were still in the seed coats. The seeds damaged by insects and moisture were discarded. The seeds were then dried by laying them on a surface where sunlight can reach the seeds. The initial mass of the seeds was weighed before drying. The mass of the seeds was also measured after drying, until the mass of the seeds became constant. After drying, the seed coats were removed by manually cracking using a mortar and pestle and grinding of the seeds to a fine powder was done using an Electro Master electric grinder, model EM-BL-1058-2 IN 1-P. The ground powder was then sieved through a 250 µm sieve to obtain fine powder particles which can be used to come up with the coagulant solution.

3.4 Coagulant sample collection and preparation for coagulation-flocculation experiments

The salt extraction method adapted from (Okuda et al., 2001a) and Ali *et al*. (2010) was then used to prepare the *E.goetzei* seed extract coagulant solution. For every gram of the *E.goetzei* seed powder, 100 mL of 1 molar sodium chloride solution (prepared by adding 58.44g of sodium chloride to 1 L of distilled water) was added. The mixture was stirred using a magnetic stirrer for 30 minutes at room temperature. The suspension formed was filtered using Whatman qualitative filter paper with a pore size of 11 µm and a diameter of 125 mm on a vacuum filtration system as shown in Fig 3.4. The vacuum filtration system consists of a Bio base laboratory vacuum pump model GM-0.20 connected to a Buchner funnel with Whatman filter paper. The filtrate was used as a coagulant for subsequent coagulation-flocculation experiments.

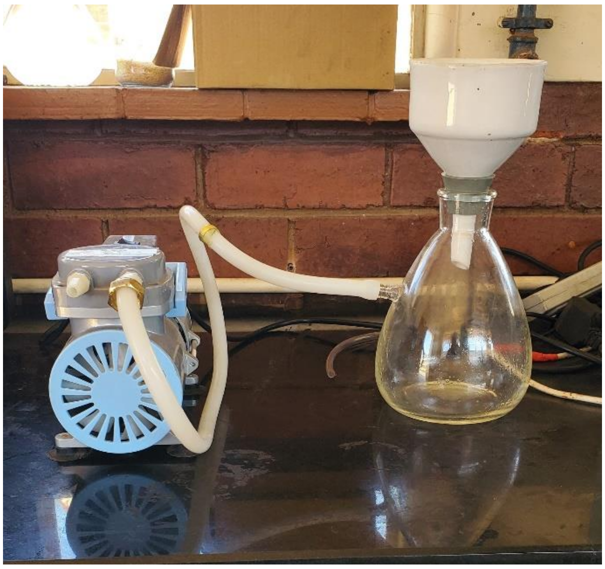

Figure 3.4 | Vacuum filtration apparatus to obtain *E.goetzei* seed extracts coagulant solution

3.5 Preparation of synthetic water samples

Synthetic water was prepared in the laboratory by spiking distilled water with known amounts of contaminants. Synthetic turbid water was prepared by adding 10 g of kaolin clay to 1 litre of tap water, stirring on a magnetic stirrer for 30 minutes for homogenous mixing and allowing to settle for 24 hours for complete hydration of the kaolin clay particles (Ghebremichael *et al*., 2005). Various turbidities were obtained by dilution using the equation $C_1V_1 = C_2V_2$. As for fluoridated water, a standard stock fluoride solution was prepared by dissolving 0.221 g anhydrous sodium fluoride in one litre of distilled water (Bayu *et al*., 2022). The fluoride concentration for this standard stock solution is 100 mg/L. Other concentrations were prepared from this stock solution by dilution. Moreover, as for iron, a 1 000 mg/L standard Fe^{2+} ferrous iron standard stock solution was prepared by dissolving 0.70 grams of ferrous ammonium sulphate hexahydrate in deionized water containing 1.0% concentrated sulfuric acid and adjusting the volume to 100 mL (Namjan *et al*., 2023). Other concentrations were then prepared by direct dilution using the equation $C_1V_1 = C_2V_2$. Manganese metal (1.0 g) was dissolved in 50.0 mL of concentrated hydrochloric acid and diluted to 1.0 litre in a volumetric flask with deionized water to obtain a 1000 mg/L standard manganese solution (Ogah *et al*., 2016). Other concentrations were then prepared by direct dilution using the equation $C_1V_1 = C_2V_2$. The initial values of the parameters for synthetic water which were tested in this study were measured in triplicates and the measurements presented as mean ±SD are shown in Table 3.2.

Table 3.2 | Characterisation of synthetic water used for coagulation-flocculation experiments

Parameter	Mean ± SD
Turbidity	12.33±0.55 (NTU)
	28.46±0.02 (NTU)
	45.22± 0.66 (NTU)
	71.33±1.15 (NTU)
	105.33±1.15 (NTU)
	167.67±3.18 (NTU)
pH	8.16±0.06
TDS	94.44±5.09 (mg/L)
Fluoride	11.16±0.01 (mg/L)
Iron	5.50±0.29 (mg/L)
Manga-nese	5.46±0.01 (mg/L)

3.6 Coagulation-flocculation experiment

The jar test was used to find the optimum concentration of the coagulant which has to be added to the synthetic water for effective treatment. The coagulation-flocculation experiment was carried out using six 500 mL beakers which had synthetic water and coagulant added to each beaker. The dosage of the *E.goetzei* seed extracts coagulant ranged from 0 to 300 mg/L (for turbidity, pH, and TDS) and 0 to 100 mg/L (for iron, fluoride ,and manganese) with the dosages being gradually increased by 20 mg/L in each beaker on the flocculator.

The experiment was done using a flocculator multiple spindle stirrer at a rapid mixing speed of 120 rpm for one minute and a slow mixing speed of 30 rpm for 15 minutes and settling was then left to take place for 15 minutes (Gidde *et al*., 2012). The supernatant from the top of each beaker was then collected to measure the turbidity, fluoride, iron and manganese after treatment. The setup for the coagulation-flocculation experiment is shown in Fig 3.5.

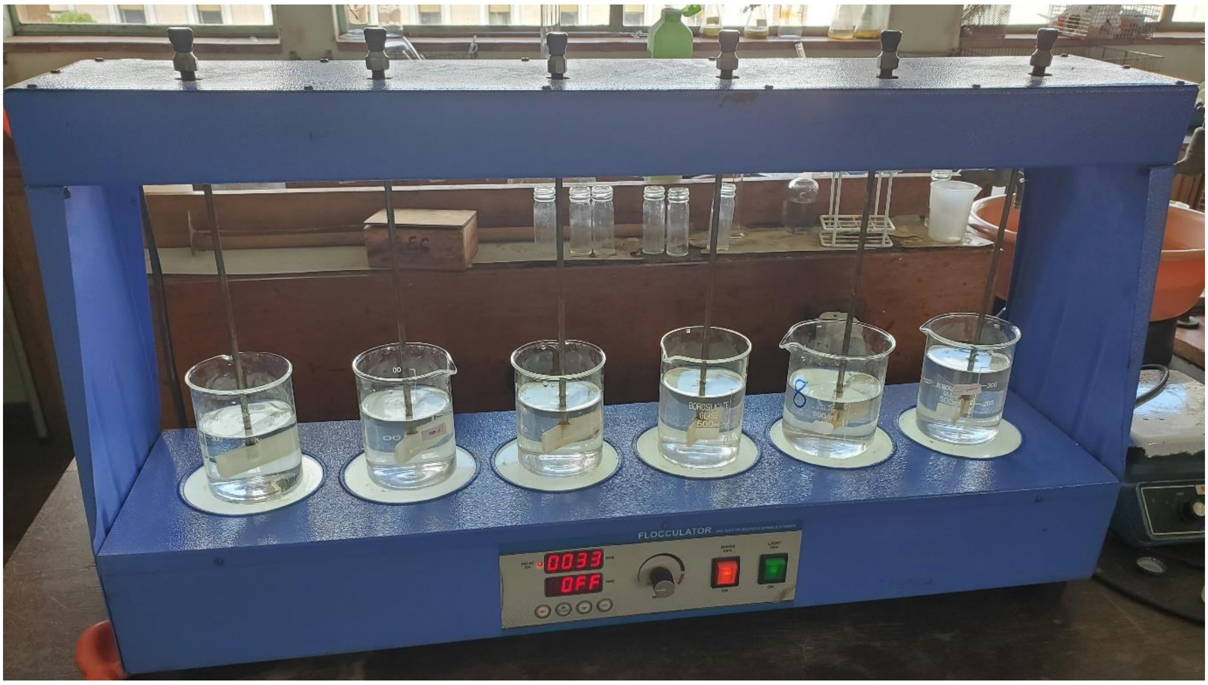

Figure 3.5 | Coagulation-flocculation experiment setup

3.7 Analytical methods

Standard methods from American Public Health Association/ American Water Works Association /Water Environment Federation , APHA/AWWA/WEF (2017) were used for the determination of various parameters including iron, manganese, and fluoride in water. Moisture content, crude protein content and fat content in the seed extracts of *E.goetzei* were also measured using standard methods from Association of Official Analytical Chemists, AOAC (2000). The experiments were done in triplicates for consistency and the results were finally averaged. The methods which were used to achieve the objectives are summarized in Table 3.3.

Table 3.3 | Methods used for research objectives

Specific objective	Method
1. **To determine the key ingredients in the *E.goetzei* seed extracts which are responsible for the coagulation process.**	**Proximate Analysis** - Ash, moisture, fats, fibre, crude protein and carbohydrates were determined using AOAC (2000) method
2. **To determine the optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity.**	**Determination of turbidity in synthetic water dosed with *E.goetzei* seed extracts solution** - Determination of turbidity 2130 B. Nephelometric Method from APHA/AWWA/WEF (2017)
3. **To determine the effect of *E.goetzei* seed extracts coagulant dosage on the removal of fluoride, manganese and iron from synthetic water.**	**Determination of fluoride in synthetic water dosed with *E.goetzei* seed extracts solution** -4500- F^- D. SPANDS Method from APHA/AWWA/WEF (2017) **Determination of iron in synthetic water dosed with *E.goetzei* seed extracts solution** - 3500- Fe B. Phenanthroline Method from APHA/AWWA/WEF (2017) **Determination of manganese in synthetic water dosed with *E.goetzei* seed extracts solution** - 3500- Mn B. Persulfate Method from APHA/AWWA/WEF (2017)
4. **To characterize and determine the levels of organic matter in water treated with *E.goetzei* seed extracts.**	**Determination of COD in treated water** - A mercury-free large-scale (10 mL) flask digestion using chromium (III) potassium sulphate and silver nitrate solutions. Determination of organic matter in treated water. Method C from Westwood (2007)
5. **To determine the quality of the sludge produced from treating raw water with *E.goetzei* seed extracts, and hence its feasibility for reuse.**	**Determination of total nitrogen** - The method adapted from EAWAG Aquatic Research (2007) was used **Determination of total phosphorus** -–Determination of phosphorus: Method 4500-P C. Vanadomolybdophosphoric Acid Colorimetric Method from APHA/AWWA/WEF (2017)

3.8 Determination of key ingredients in the *E.goetzei* seed extracts responsible for the coagulation process

3.8.1 Moisture content

The moisture content was obtained by drying 3 g of the sample in an oven for 3 hrs at 105ºC and reweighing the sample after cooling. The moisture content was then calculated using Equation 3.1.

$$\textbf{Moisture (\%)} = \frac{W1-W2}{W1} \times 100 \qquad (3.1)$$

Where:

w_1 = weight (g) of sample before drying

w_2 = weight (g) of the sample after drying

3.8.2 Ash content

Ash content was obtained by igniting the sample in a muffle furnace at 550℃ for 3 hours or more until light grey ash was obtained. The sample was then removed from the furnace to a desiccator to cool and then weighed. Ash content was then calculated using Equation 3.2.

$$\textbf{AC} = \frac{Wx - Wy}{Wz} \times 100 \qquad \textbf{(3.2)}$$

Where:

AC = Ash content

w_y = Weight of empty crucible

w_x = Weight of crucible with ash

w_z = Weight of sample

3.8.3 Crude protein content

The crude protein was determined by using the micro-Kjeldahl method according to AOAC (2000) with steps that included digestion, distillation and titration. For the digestion step, catalyst tablets (anhydrous sodium sulphate + copper sulphate) and 98% sulphuric acid were used. In the distillation unit, 15 mL of 40% NaOH were added. The ammonia was then received in a 100 mL conical flask containing 10 mL of boric acid plus 3-4 drops of methyl red indicator. The content of the flask was then titrated against 0.02N hydrochloric acid and the crude protein was calculated using Equation 3.3.

$$\textbf{CP\%} = \frac{(T.B) \times N \times 14 \times 100 \times 6.25}{WS \times 1000} \qquad \textbf{(3.3)}$$

Where:

CP = Crude protein

T = Titration reading

B = Blank titration reading

N = Normality of HCl

WS = Weight of sample

1000 = to convert to mg

6.25 = Protein factor

3.8.4 Fat content

Fat content was determined according to the method of AOAC (2000) using the Soxhlet apparatus. Extraction was carried out for 8 hours with petroleum ether, with the residual ether being dried by evaporation. The fat content is then calculated using the Equation 3.4.

$$\mathbf{FC} = \frac{Wa - Wb}{Wc} \times 100 \qquad \textbf{(3.4)}$$

Where:

FC= Fat content

W_b= Weight of extraction flask

W_a= Weight of extraction flask with fat

W_c= Weight of sample

3.8.5 Crude fibre content

Crude fibre content was obtained by treating samples successively with a boiling solution of sulphuric acid (H_2SO_4) of 0.26 Normality and potassium hydroxide (KOH) of 0.23 Normality. The samples were then heated at a temperature of 105℃ and ashed in a muffle furnace at 500℃. The crude fibre content was calculated using the Equation 3.5.

$$\mathbf{CF} = \frac{Wp - Wq}{Wr} \times 100 \qquad \textbf{(3.5)}$$

Where:

CF= Crude fibre

wp = weight of crucible with sample after acid and alkali treatment

wq = weight of crucible with sample after ashing

wr = weight of the sample.

3.8.6 Carbohydrates content

To determine the carbohydrate content, several other components such as ash, moisture, crude protein, fat content, and crude fibre are first measured. After obtaining the values for these components, the sum of their contents is subtracted from 100% to calculate the carbohydrate content . This calculation follows Equation 3.6, which specifies the method for determining the carbohydrate content.

Carbohydrate content = 100 % - (Ash % + moisture % + CP % + FC % + CF %)

(3.6)

3.9 Determination of optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity from synthetic water

Determination of turbidity and physicochemical parameters

Turbidity was determined using the 2130 B, Nephelometric Method according to APHA/AWWA/WEF (2017). The method is based on a comparison of the intensity of light that is scattered by the sample under defined conditions with the intensity of light scattered by a standard reference suspension under the same conditions. The higher the intensity of scattered light, the greater the turbidity. The instrument that was used to measure turbidity was a Lutron turbidity meter (TU-2016). The turbidity removal efficiency was expressed using Equation 3.7

$$\textbf{Turbidity removal efficiency (\%)} = \frac{(T_i - T_r)\times 100}{T_i} \qquad (3.7)$$

Where T_i and T_r represent the initial and final turbidity respectively.

The pH, electrical conductivity (EC) and total dissolved solids (TDS) were also measured before and after coagulation-flocculation using a Milwaukee MW 801 pH/EC/TDS Smart Combined Meter.

3.10 Effect of *E.goetzei* seed extract coagulant dosage on the removal of fluoride, manganese and iron from synthetic water

3.10.1 Determination of fluoride in water

Fluoride concentration was determined using the 4500- F- D, SPANDS Method according to APHA/AWWA/WEF (2017). The SPANDS colorimetric method is based on a reaction that occurs between the fluoride ions and a zirconium-dye lake. Fluoride reacts with the dye-lake dissociating a part of it into a colourless complex anion ($ZrF^{2-}{}_6$), and the dye. As the amount of fluoride becomes greater, the colour produced gradually becomes lighter. A spectrophotometer is used to read the absorbance of the sample after setting the reference point of the photometer. The fluoride removal efficiency was calculated using Equation 3.8

$$\textbf{Fluoride removal (\%)} = \frac{Ce - Cg}{Ce} \times 100\%. \qquad (3.8)$$

Where Ce and Cg are the initial and final fluoride concentrations respectively.

3.10.2 Determination of iron in water

Iron was determined using the 3500- Fe B, phenanthroline method according to APHA/AWWA/WEF (2017). For the Phenanthroline Method, iron is brought into solution, reduced to the ferrous state through boiling with acid and hydroxylamine and treated with 1,10 phenanthroline at a pH of 3.2 -3.3. Three molecules of phenanthroline then chelate each atom of ferrous iron resulting in the formation of an orange-red complex. Photometric measurements are carried out and standards are read against water set at zero absorbance and a calibration curve is plotted including a blank.

3.10.3 Determination of manganese in water

Manganese was determined using 3500- Mn B, persulfate method according to APHA/AWWA/WEF (2017). Persulphate oxidation of soluble manganese compounds to form permanganate is done in the presence of silver nitrate. The colour that is formed is stable for at least 24 hours if excess persulphate is present and organic matter is absent. Photometric measurements are done against a distilled water blank. Manganese and iron removal efficiency was calculated using Equation 3.9.

$$\textbf{Removal efficiency (for manganese and iron)} = \frac{Ci-Cf}{Ci} \times 100\%. \qquad \textbf{(3.9)}$$

Where Ci is the initial concentration of manganese or iron in water and Cf is the final concentration of manganese or iron in water

3.11 Levels of residual content of organic matter in water treated with *E.goetzei* seed extracts

The organic matter was measured as COD. COD in water was obtained using Standard Method C (A mercury-free large scale (10 ml) flask digestion using chromium(III) potassium sulphate and silver nitrate solution) according to Westwood (2007). The COD test is not susceptible to hazardous substances that would interfere with tests that rely on biological oxidation. Samples were oxidized by a reflux process involving 5 mL potassium dichromate and 15 mL sulfuric acid silver sulphate solution. To catalyse the oxidation of some substances, such as alcohols and low molecular weight acids, 0.6 mL of silver nitrate was added to 10 mL of the sample. The addition of chromium (III), in the form of 0.40 mL chromium (III) potassium sulphate, along with an excess of silver, suppresses chloride interference because of ammonia present. The resultant mixture was refluxed for two hours, and any residual dichromate was then measured spectrophotometrically (Westwood, 2007).

3.12 Quality of sludge produced from treating synthetic water with *E.goetzei* seed extracts

3.12.1 Determination of total nitrogen

The total nitrogen was obtained using the method by the Swiss Federal Institute of Aquatic Science and Technology , EAWAG Aquatic Research (2007). Sample digestion was done using a digestion reagent which consisted of 20.1g of potassium persulphate, $K_2S_2O_8$ and 3 g of sodium hydroxide in 1 L of reagent water. After digestion, total nitrogen was measured similarly to NO_3 with pH being neutralised with 30% sodium hydroxide, before adding 1 mL of Na-salicylate. Nitrate in a strongly acidic solution with salicylic acid forms 3- and 5-nitro salicylic acid. In alkaline solutions, their concentrations were determined colorimetrically using a spectrophotometer.

3.12.2 Determination of total phosphorus

Total phosphorus was determined using the 4500-P C method which is also known as the Vanadomolybdophosphoric Acid Colorimetric Method from APHA/AWWA/WEF (2017). In a dilute orthophosphate solution, ammonium molybdate reacted under acidic conditions to form a heteropoly acid and the acid formed was molybdophosphoric acid. The intensity of the yellow colour which was measured using a spectrophotometer corresponded to the phosphate concentration. Standard phosphate solution was prepared by dissolving in distilled water 219.5 mg of anhydrous KH_2PO_4 and diluting to 1000 mL; 1.00 mL = 50.0 µg $PO_4{}^{3-}$-P.

3.13 Data analysis and presentation

The statistical analyses were carried out using IBM SPSS Statistics Version 20 and Microsoft Excel 2021. Analysis of variance (ANOVA) was utilised to analyse the data in order to determine the relationships and interactions between the variables and the corresponding responses so as to determine significant levels ($p < 0.05$). Microsoft Excel 2021 was used to construct the graphs for the effect of coagulant dosage on pH, TDS, turbidity, iron, manganese, fluoride, total nitrogen and total phosphorus and the effect of initial contaminant concentration (fluoride, iron and manganese) on the removal efficiency of the same contaminants.

Chapter 4: Results

This chapter gives an outline of the results obtained after data had been collected on the composition of the *E.goetzei* seed extracts and their use for synthetic water treatment and the composition of the sludge produced after treating synthetic water using seed extracts of *E.goetzei*. The presentation of results is arranged based on each objective.

4.1 Determination of key ingredients in the *E.goetzei* seed extracts responsible for the coagulation process

Table 4.1 shows the results which were obtained for moisture, crude protein, crude fibre, fat, ash and carbohydrate content in the seed extracts of *E.goetzei*. A total of 6 experiments were conducted and 3 replicates were done for each experiment. The results are reported as mean± Standard deviation (SD). The component with the lowest value was ash content which was 2.20±0.10 % whilst fat content had the highest value which was 32.99 ±0.74 % followed by carbohydrates which had a value of 29.93± 0.82 %. Crude protein also had a relatively high value of 21.40±0.30 %.

Table 4.1 | Proximate analysis results for *E.goetzei* seeds collected from Walden North Farm in Chegutu District, Mashonaland West Province, Zimbabwe

Parameter	Mean (%)±SD
Moisture content	5.25±0.14
Crude Protein	21.40±0.30
Crude Fibre	8.23±0.29
Fat Content	32.99±0.74
Ash content	2.20±0.10
Carbohydrates Content	29.93± 0.82

4.2 Optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity from synthetic water

The optimum coagulant dosages were 200 mg/L and 240 mg/L for initial turbidity ranging from 12.33 NTU to 45.22 NTU and 71.33 NTU to 167.67 NTU respectively. The percentage removal efficiency of turbidity using seed extracts of *E.goetzei* increased with an increase in dosage in all the turbidities which were investigated as shown in Figure 4.1. The lowest removal efficiency obtained was 34.5% at an initial turbidity of 28.46 ±0.02 NTU and a coagulant dosage of 20 mg/L. The highest removal efficiency was 94.8 % at an initial turbidity of 45.22 ± 0.66 NTU and coagulant dosage of 260 mg/L. Appendix A shows that initial turbidity had a significant effect on the turbidity removal efficiency since the value of p obtained was < 0.05.

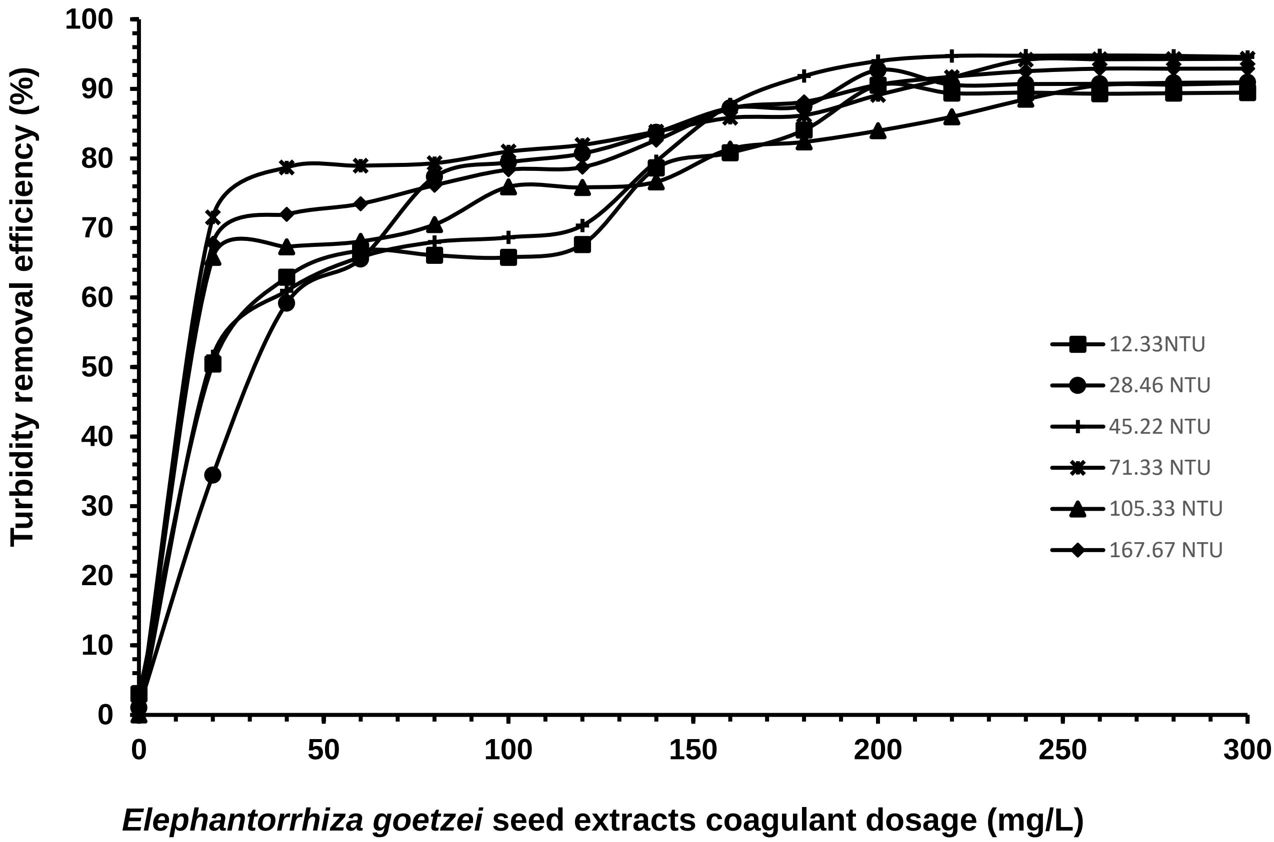

Figure 4.1 | Effect of *E.goetzei* seed extract coagulant dosage and initial turbidity on turbidity removal efficiency

As illustrated in Figures 4.2-4.4, the residual turbidity obtained in this study at the maximum coagulant dosage of 300 mg/L coagulant dosage was 1.30 NTU, 2.58 NTU, and 2.45 NTU at initial turbidities of 12.33 NTU, 28.46 NTU, and 45.22 NTU, respectively. Generally, for all the initial turbidities in the study, there was a decrease in the residual turbidity as the coagulant dosage increased from 0 mg/L to 300 mg/L. The models developed for the initial turbidities of 12.33 NTU, 28.46 NTU, and 45.22 NTU accounted for 82.4%, 85.0%, 88.8%, of the variations in the residual turbidities, respectively. The WHO standard of 5 NTU for turbidity in drinking water was met using *E.goetzei* seed extract coagulant dosages in the range of 40 to 180 mg/L .On the other hand, the regression models for initial turbidities of 71.33 NTU, 105.33 NTU, and 167.67 NTU as shown in Figures 4.5; 4.6 and 4.7 had lower R^2 values than the initial turbidities ranging from 12.33 NTU to 45.22 NTU.

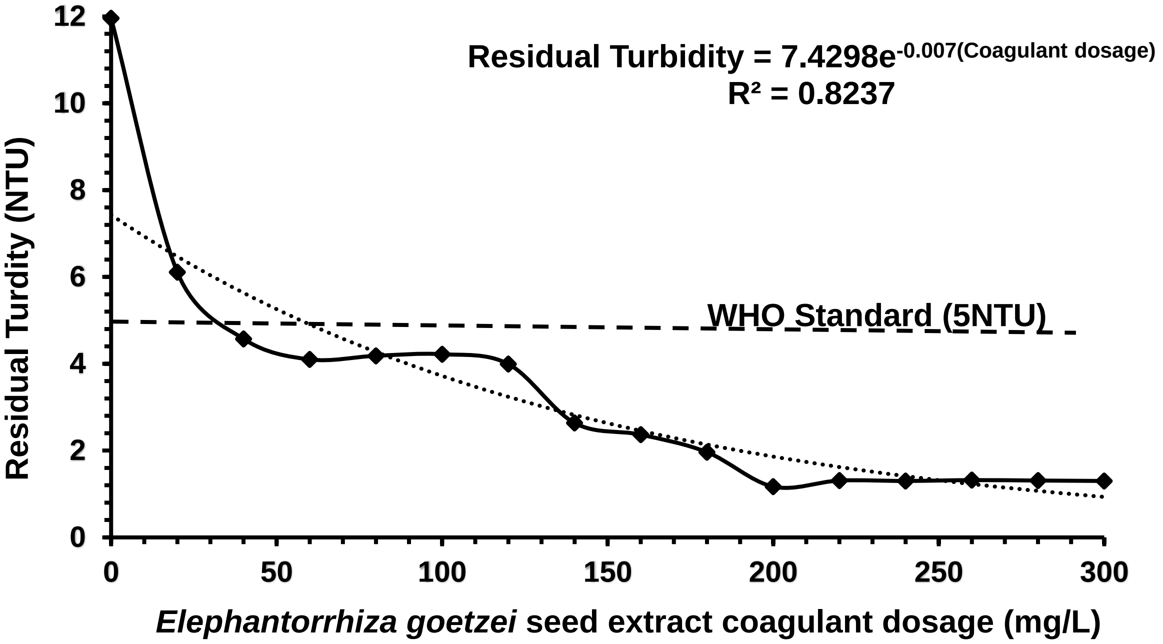

Figure 4.2 | Effect of *E.goetzei* seed extract coagulant dose on initial turbidity of 12.33 NTU

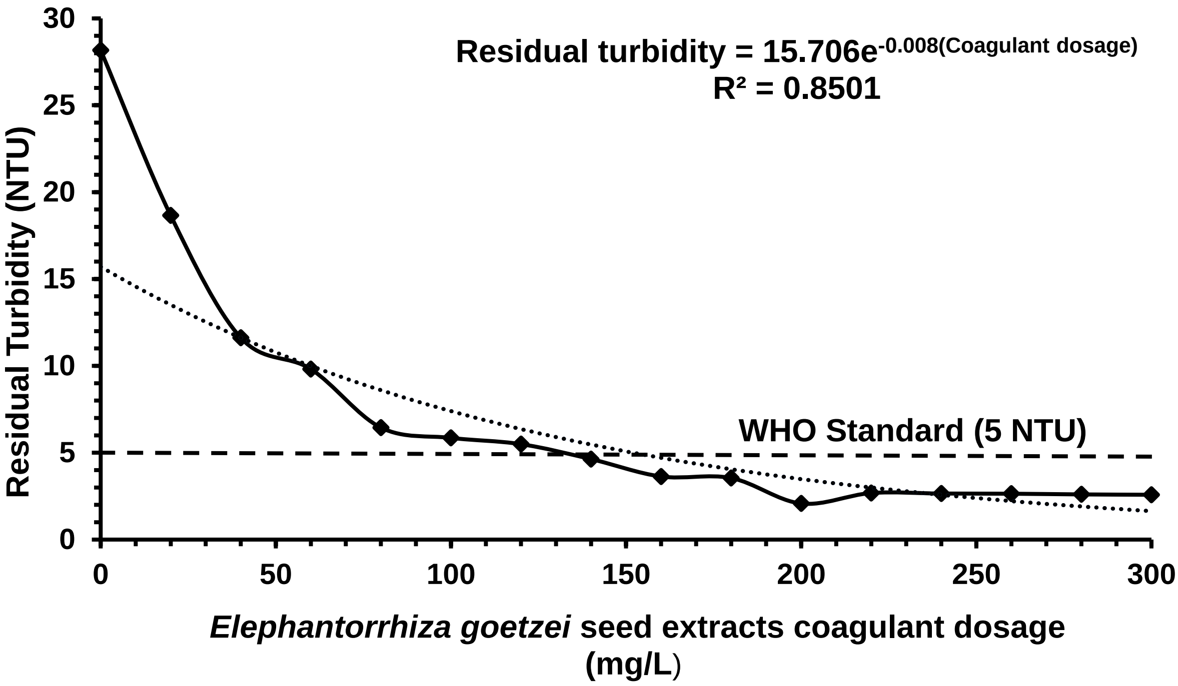

Figure 4.3 | Effect of *E.goetzei* seed extract coagulant dose on initial turbidity of 28.46 NTU

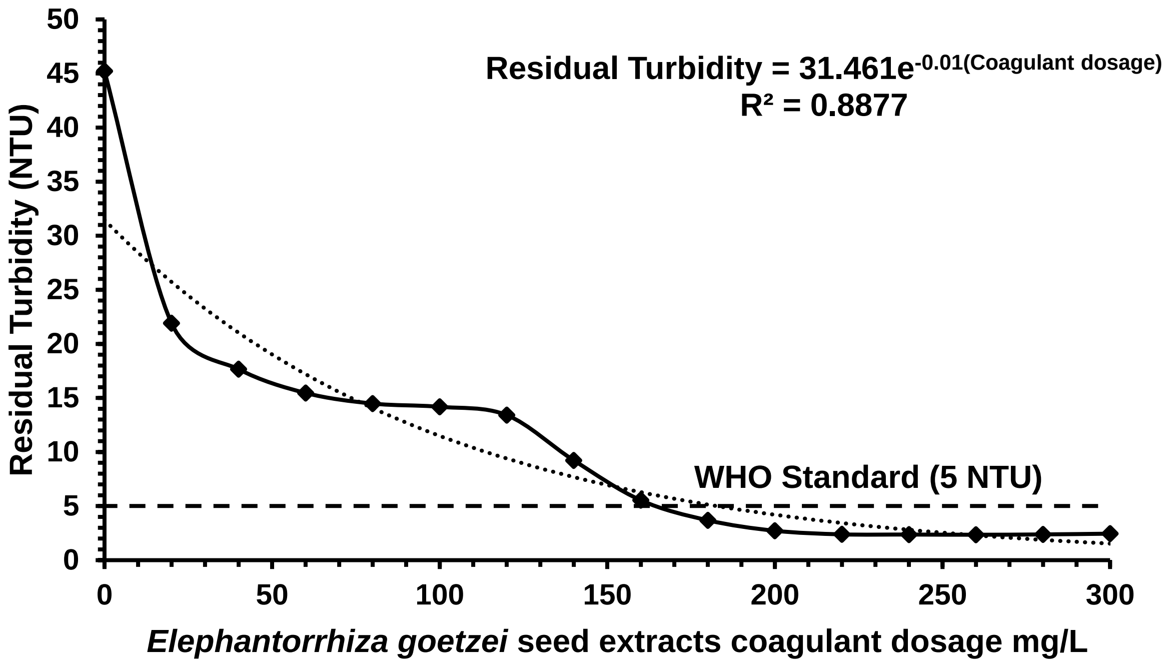

Figure 4.4 | Effect of *E.goetzei* seed extract coagulant dose on initial turbidity of 45.22 NTU

As illustrated in Figure 4.5-4.7, when the initial turbidities were 71.33 NTU, 105.33 NTU, and 167.67 NTU, the residual turbidities achieved at the maximum dosage of 300 mg/L were 4.05 NTU, 9.64 NTU, and 11.89 NTU, respectively.

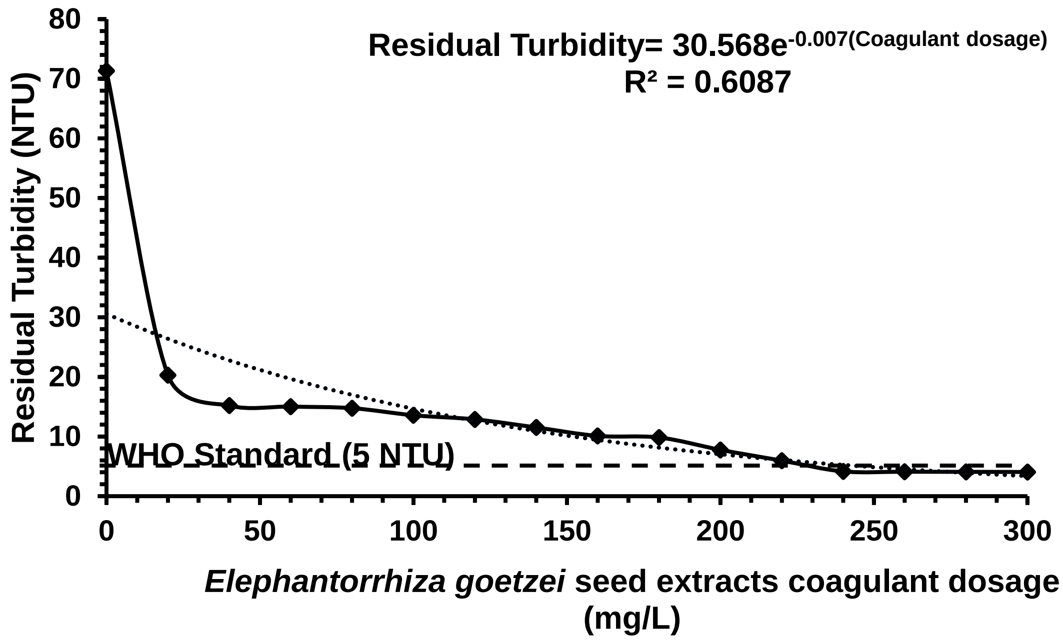

Figure 4.5 | Effect of *E.goetzei* seed extract coagulant dose on initial turbidity of 71.33 NTU

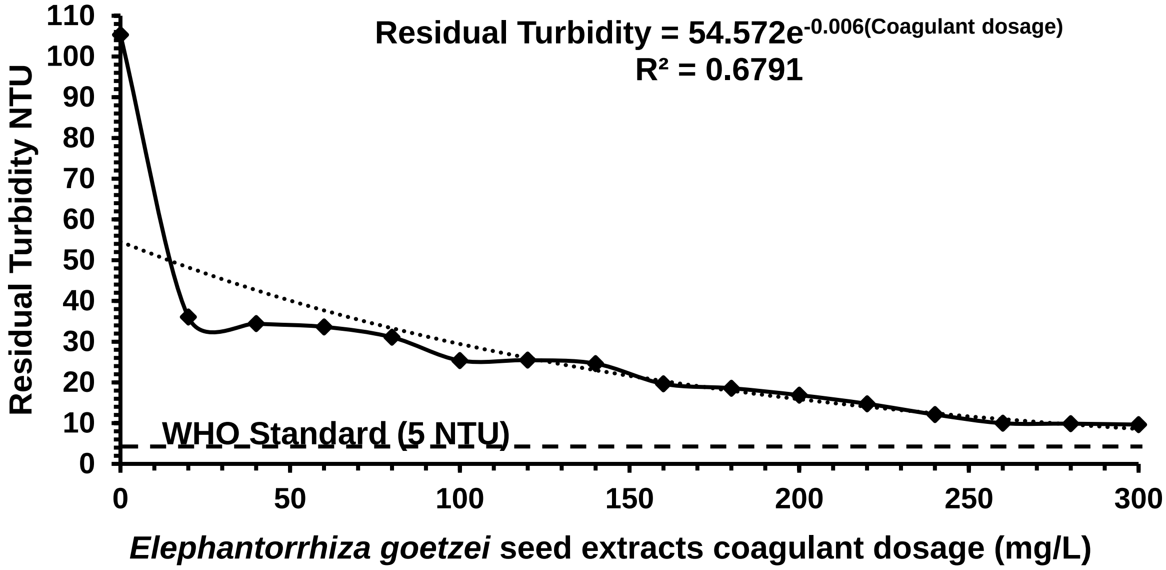

Figure 4.6 | Effect of *E.goetzei* seed extract coagulant dosage on initial turbidity of 105.33 NTU

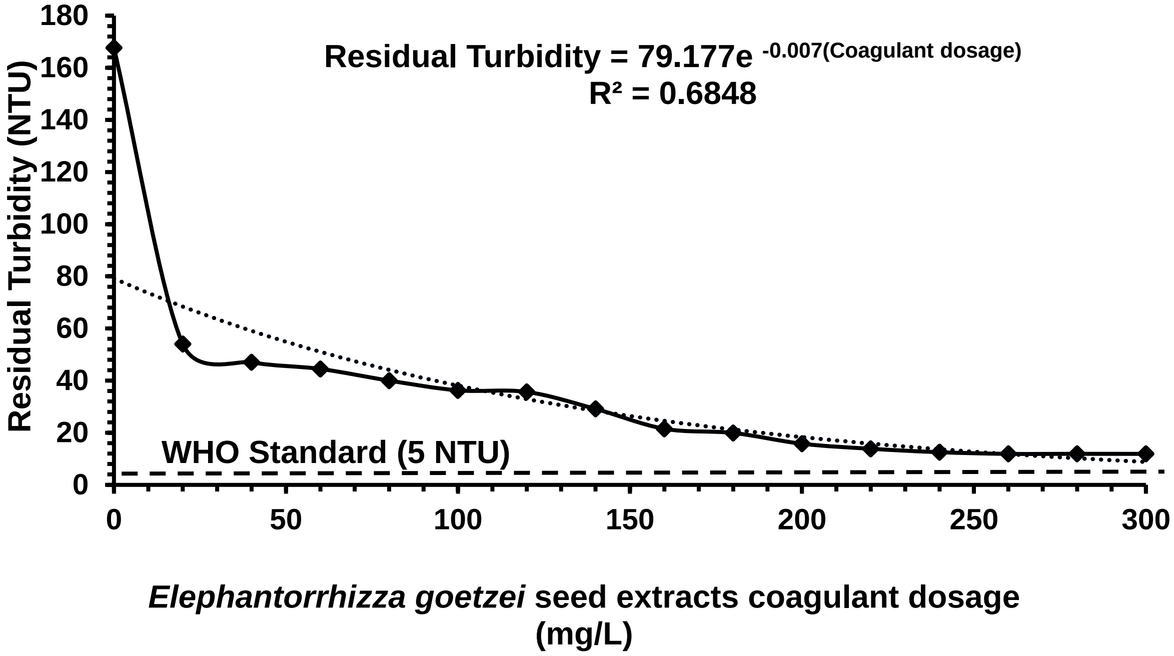

Figure 4.7 | Effect of *E.goetzei* seed extracts coagulant dosage on initial turbidity of 167.67 NTU

The R^2 values varied across different initial turbidity levels, with the highest value recorded as 0.89 at an initial turbidity of 45.22 NTU, while the lowest value was 0.61 at an initial turbidity of 71.33 NTU. The models developed for initial turbidity of 12.33 NTU (lowest turbidity) and 167.33 NTU (highest turbidity) account for 82.4% and 68.5% of the variance in residual turbidity, respectively. The ANOVA was significant for all the initial turbidities ($p < 0.05$) as shown in Appendix B . Thus, there is significant evidence to conclude that both *E.goetzei* seed extracts coagulant dosage and initial turbidity had a significant effect on the turbidity of the treated water.

4.2.1 Effect of E.goetzei seed extracts coagulant dosage on pH

There was a slight change in the pH of the water when the coagulant was introduced into the water at different doses, as demonstrated in Fig 4.8, with the pH being recorded as 8.00 ±0.02 at 300 mg/L coagulant dosage.

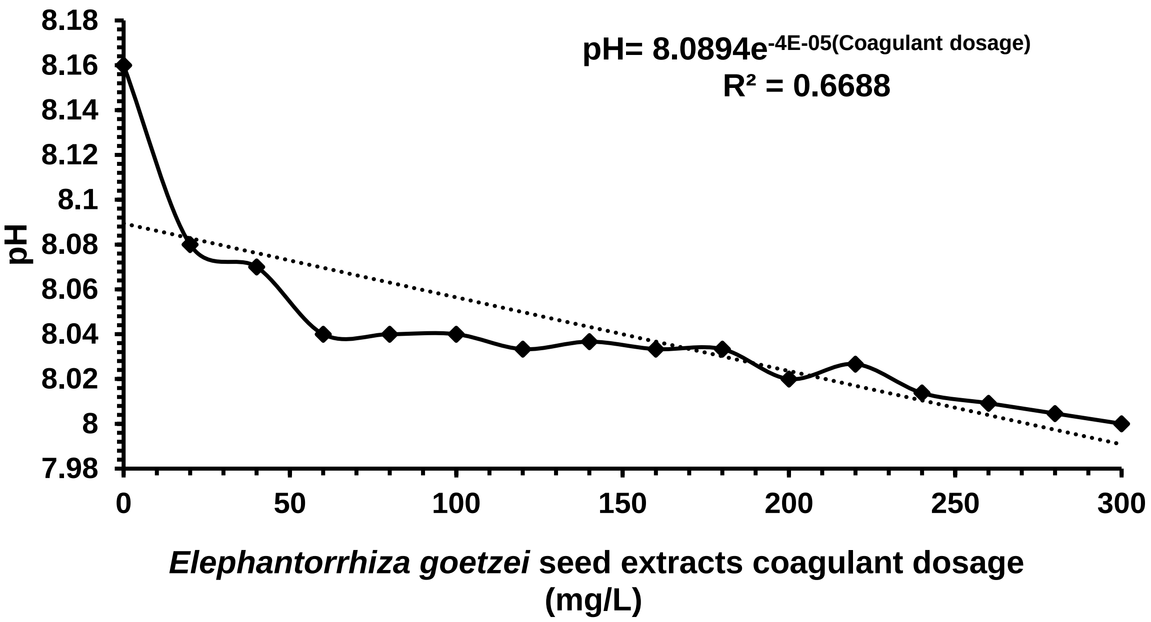

Figure 4.8 | Effect of *E.goetzei* seed extracts coagulant dosage on the pH of the treated water

4.2.2 Effect of E.goetzei seed extracts coagulant dosage on TDS

According to Figure 4.9, the TDS reached a maximum value of 1316.28 ±0.00 mg/L when the *E.goetzei* seed extracts coagulant dosage was 300 mg/L, as observed in this study. The coagulant dosage had a significant influence on both the pH and TDS levels of the treated water, as indicated by a p-value below 0.05. The relationship between TDS and coagulant dosage had a high R^2 value of 0.99.

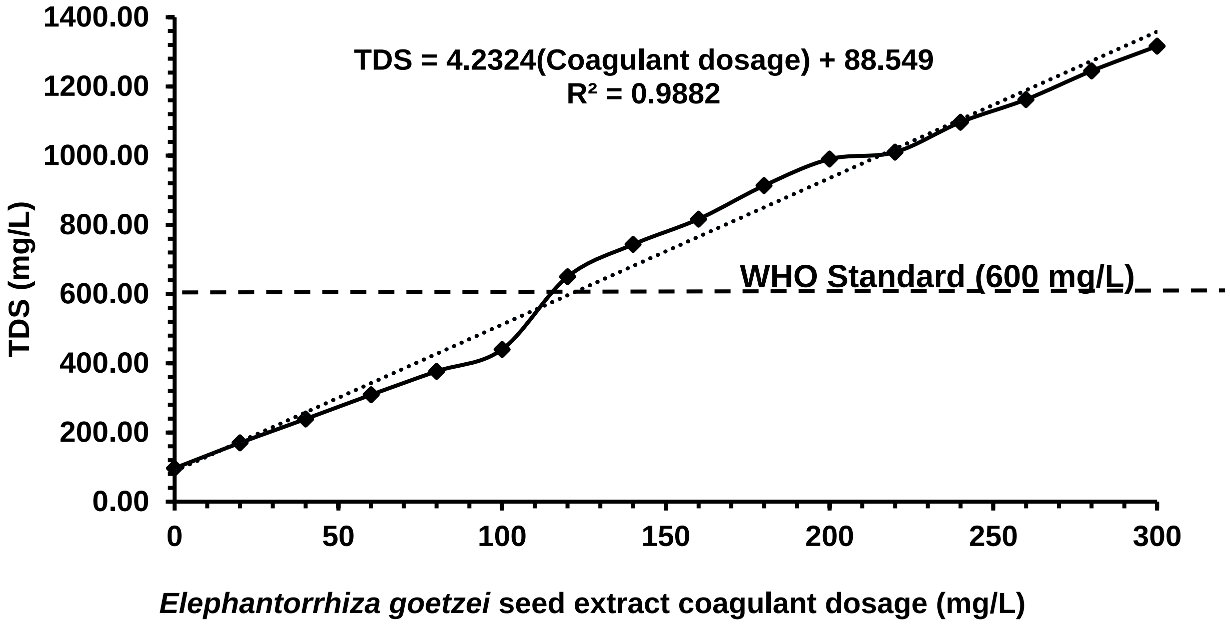

Figure 4.9 | Effect of *E.goetzei* seed extracts coagulant dosage on TDS of the treated water

4.3 Effect of *E.goetzei* seed extract coagulant dosage on the removal of fluoride, manganese and iron from synthetic water

4.3.1 Effect of E.goetzei seed extracts coagulant dosage on fluoride removal

The synthetic water used in the study had an initial fluoride concentration of 11.16 ± 0.01 mg/L. Figure 4.10 shows that the fluoride concentration reached a minimum of 5.57 mg/L when a coagulant dosage of 100 mg/L was applied. Figure 4.11 illustrates that the fluoride removal efficiency improved with increasing coagulant dosage as it increased from 0 to 50.1% as the *E.goetzei* seed extract coagulant dosage increased from 0 to 100 mg/L. Using the model , Fluoride conc= -0.0538(Coagulant dosage) + 10.836 , the WHO standard of fluoride in drinking water of 1.5 mg/L can be met at a coagulant dosage of 173.5 mg/L.

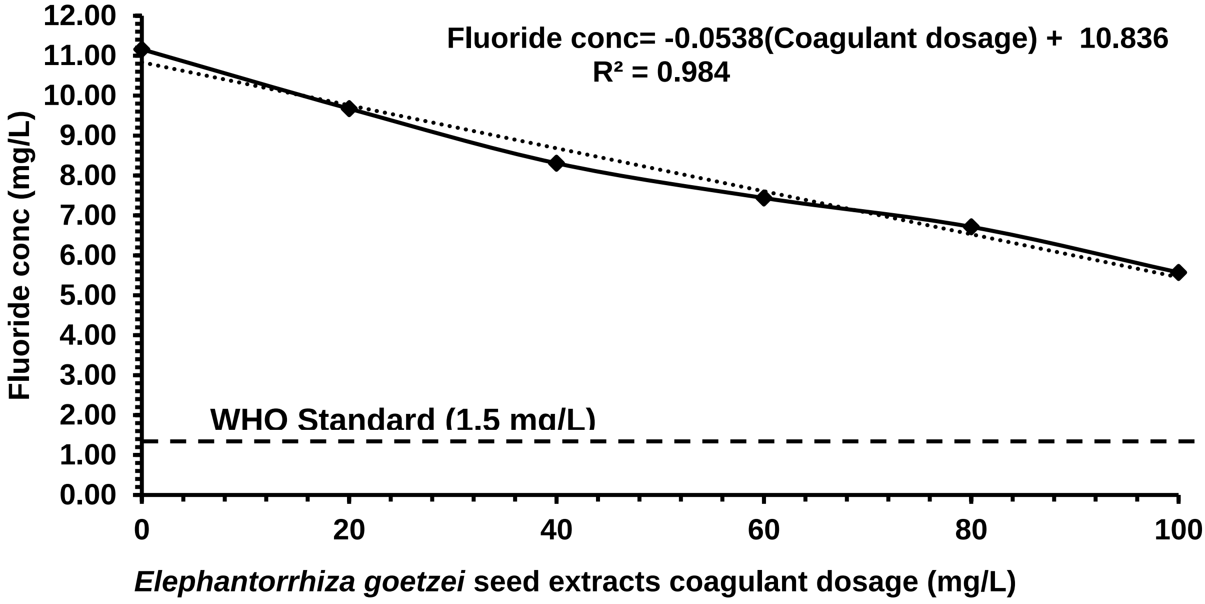

Figure 4.10 | Effect of *E.goetzei* seed extracts coagulant dosage on fluoride concentration in water

E.goetzei seed extracts coagulant dosage had a significant effect on the fluoride concentration of the treated water ($p < 0.05$). The relationship between fluoride concentrations and coagulant dosage had an R^2 value of 0.98.

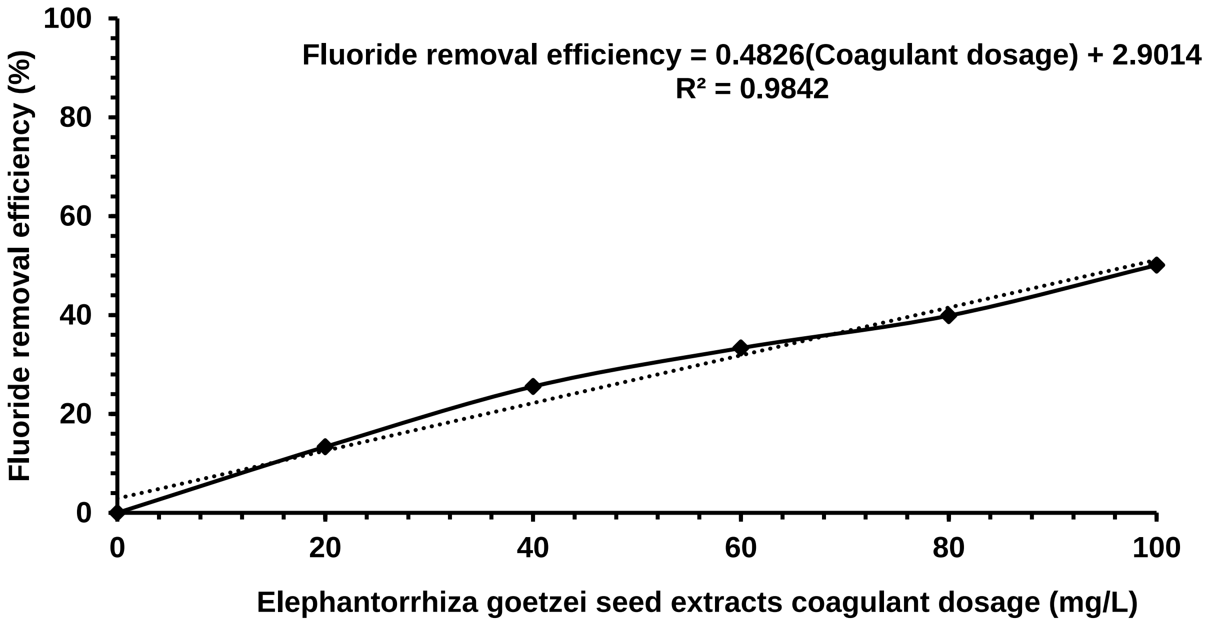

Figure 4.11 | Effect of *E.goetzei* seed extracts coagulant dosage on fluoride removal efficiency

4.3.2 Effect of initial fluoride concentration on fluoride removal efficiency

As shown in Fig 4.12, there was an increase in fluoride removal efficiency from 21.0% to 53.1% as the initial fluoride concentration increased from 5.00 mg/L to 25.00 mg/L. The relationship between initial fluoride concentration and fluoride removal efficiency had an R^2 value of 0.96.

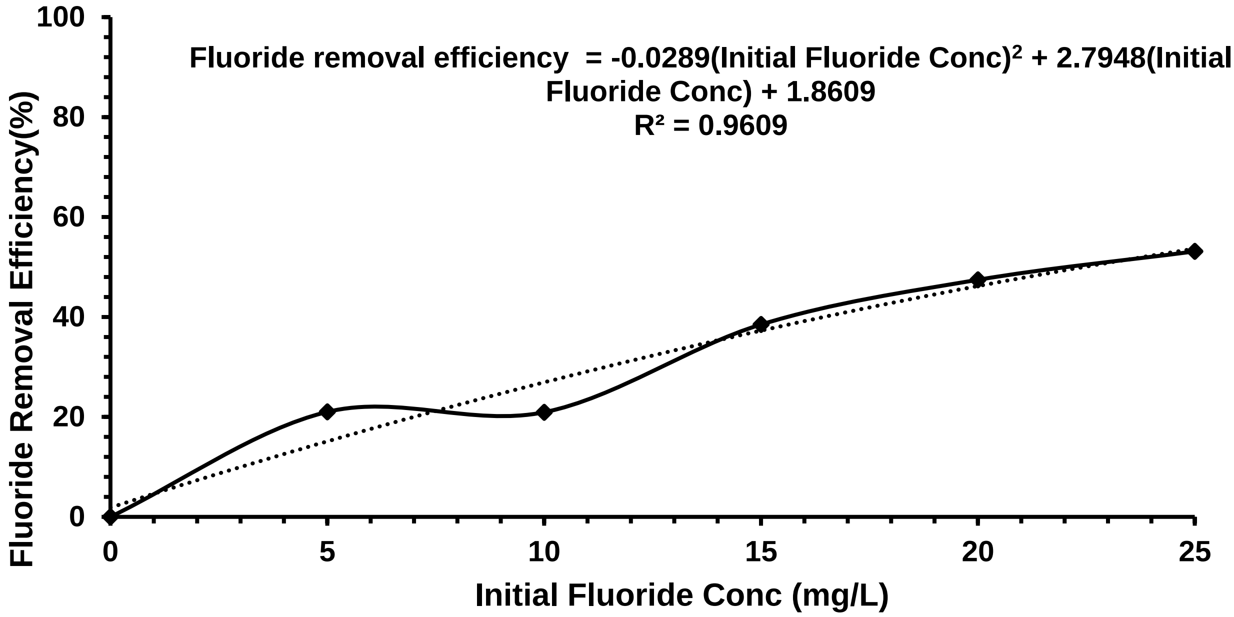

Figure 4.12 | Effect of initial fluoride concentration on fluoride removal efficiency

4.3.3 Effect of E.goetzei seed extracts coagulant dosage on iron removal

The iron concentration in the synthetic water was measured to be 5.5 ± 0.29 mg/L. Figure 4.13 indicates that at a coagulant dosage of 100 mg/L, the remaining iron concentration was found to be 0.55 ± 0.02 mg/L. The statistical analysis revealed a significant effect of the coagulant dosage on the iron concentration of the treated water, as indicated by a p-value below 0.05. Furthermore, the relationship between the iron concentration and coagulant dosage displayed a high R^2 value of 0.99.

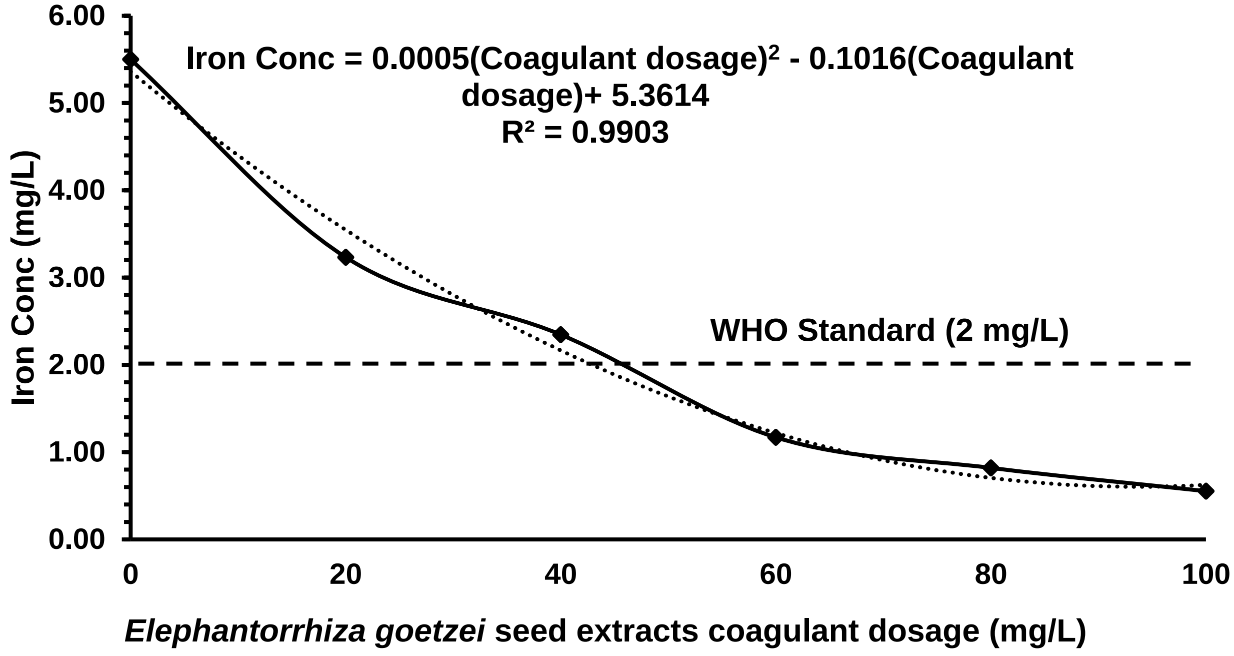

Figure 4.13 | Effect of *E.goetzei* seed extracts coagulant dosage on iron concentration in water

Figure 4.14 illustrates that there was an increase in iron removal efficiency as the coagulant dosage increased from 0 to 100 mg/L. The highest iron removal efficiency obtained from this study was 90.0% at the maximum coagulant dosage of 100 mg/L.

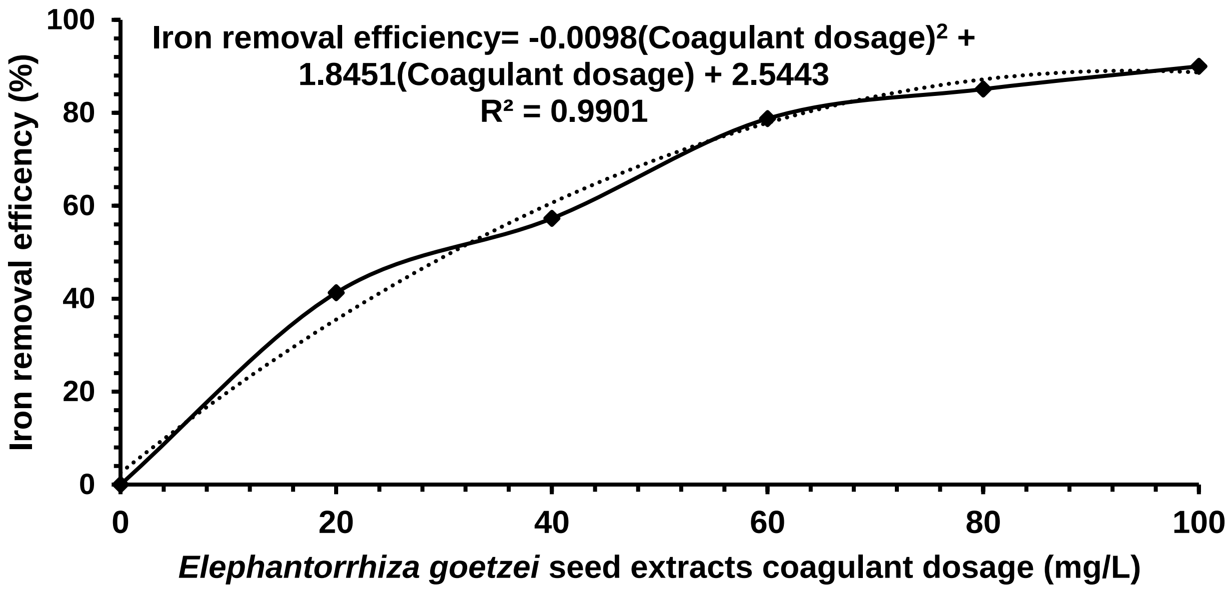

Figure 4.14 | Effect of *E.goetzei* seed extracts coagulant dosage on iron removal efficiency

4.3.4 Effect of initial iron concentration on iron removal efficiency

Generally, there was an increase in iron removal efficiency from 34.2% to 86.1% as the initial iron concentration increased from 5 mg/L to 25 mg/L as exhibited by Fig 4.15.

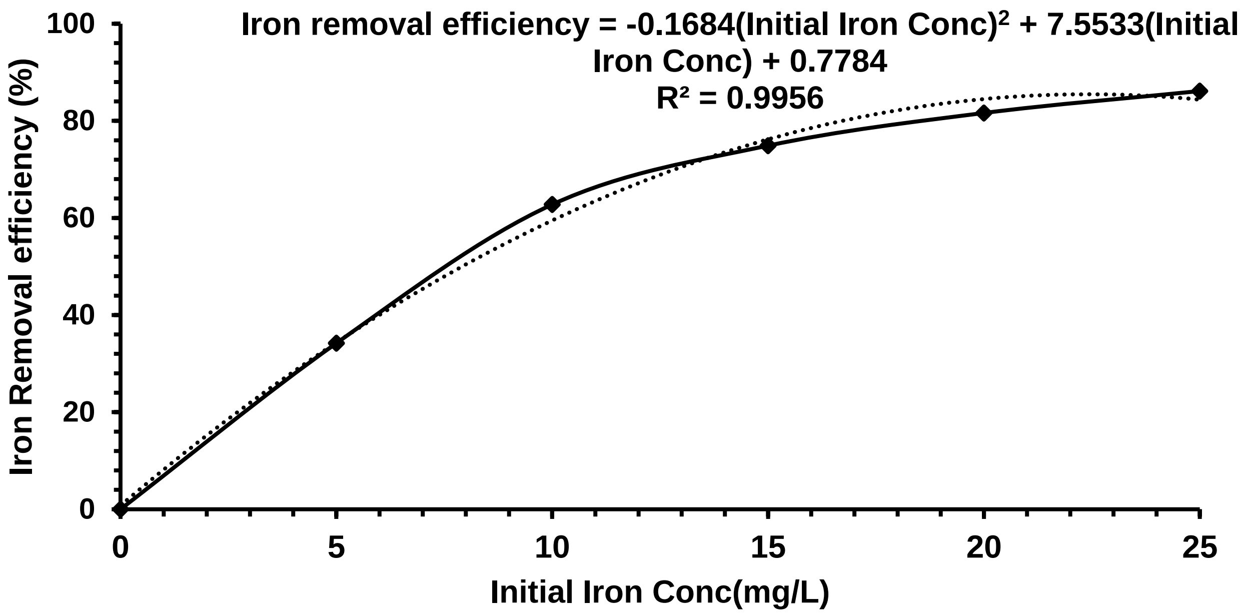

Figure 4.15 | Effect of initial iron concentration on iron removal efficiency

4.3.5 Effect of E.goetzei seed extracts coagulant dosage on manganese removal in water

Figure 4.16 shows that the residual manganese concentration at the maximum coagulant dosage of 100 mg/L was 2.52 mg/L. The R^2 value for the relationship between manganese concentration and coagulant dosage was 0.73.

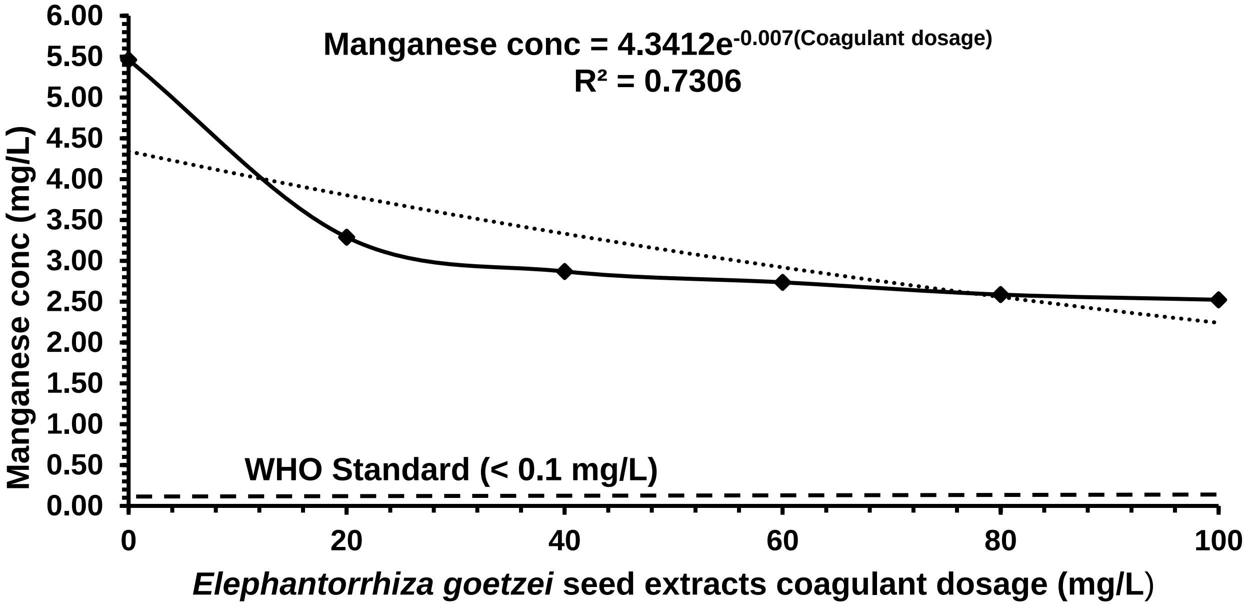

Figure 4.16 | Effect of *E.goetzei* seed extracts coagulant dosage on manganese concentration in water

Additionally, as shown in Figure 4.17, there was a noticeable improvement in manganese removal efficiency as the coagulant dosage increased. The removal efficiency increased up to 53.9% when the highest coagulant dosage of 100 mg/L was utilised.

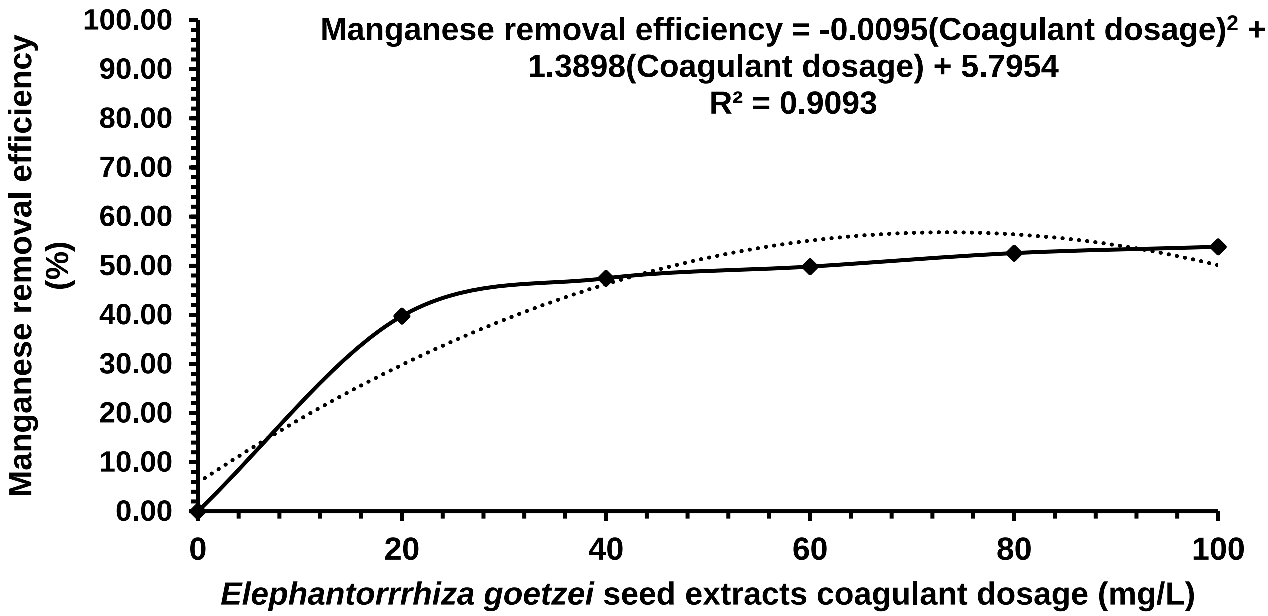

Figure 4.17 | Effect of *E.goetzei* seed extracts dosage on manganese removal efficiency

4.3.6 Effect of initial manganese concentration on manganese removal efficiency

Figure 4.18 illustrates that, as the initial manganese concentration increased from 5 mg/L to 10 mg/L there was an increase in manganese removal efficiency from 25.0% to 81.0%. The removal efficiency then decreased from 81.0% to 38.0% as the initial manganese concentration increased from 10 mg/L to 25 mg/L.

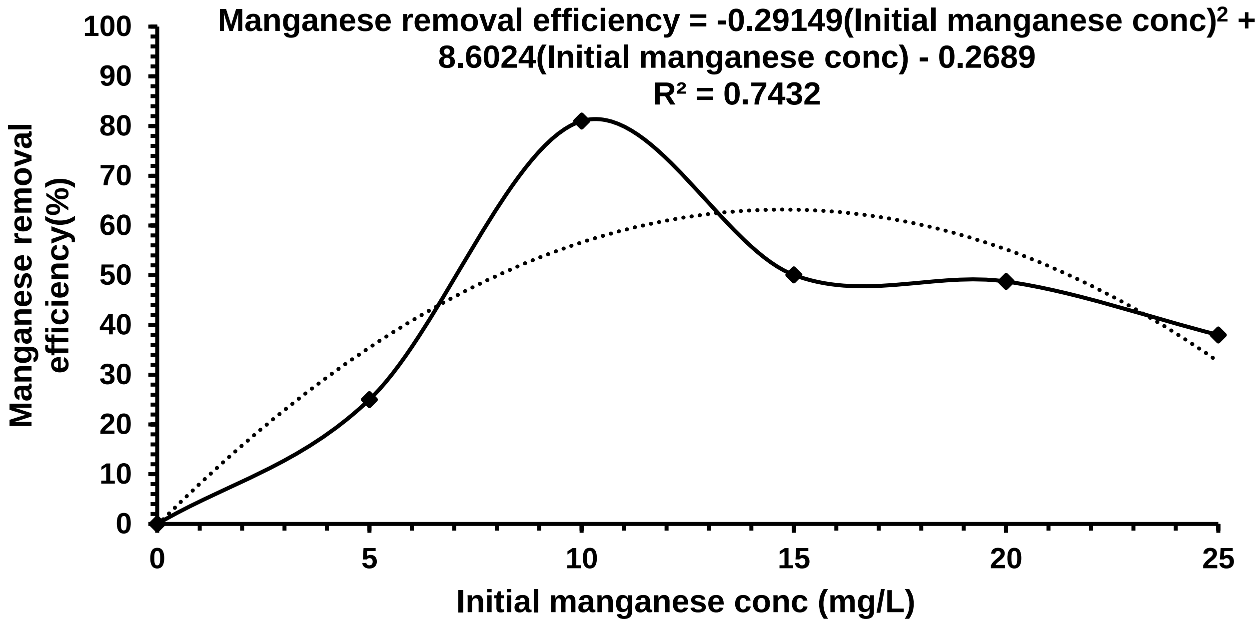

Figure 4.18 | Effect of initial manganese concentration on manganese removal efficiency

4.4 Levels of residual content of organic matter in water treated with *E.goetzei* seed extracts

The residual content of organic matter in the treated water was determined as COD. Fig 4.19 shows that there was an increase in COD from 55.33 mg/L to 419.33 mg/L as the coagulant dosage increased from 0 mg/L to 100 mg/L. Coagulant dosage had a significant effect on the COD concentration of the treated water ($p<0.05$).

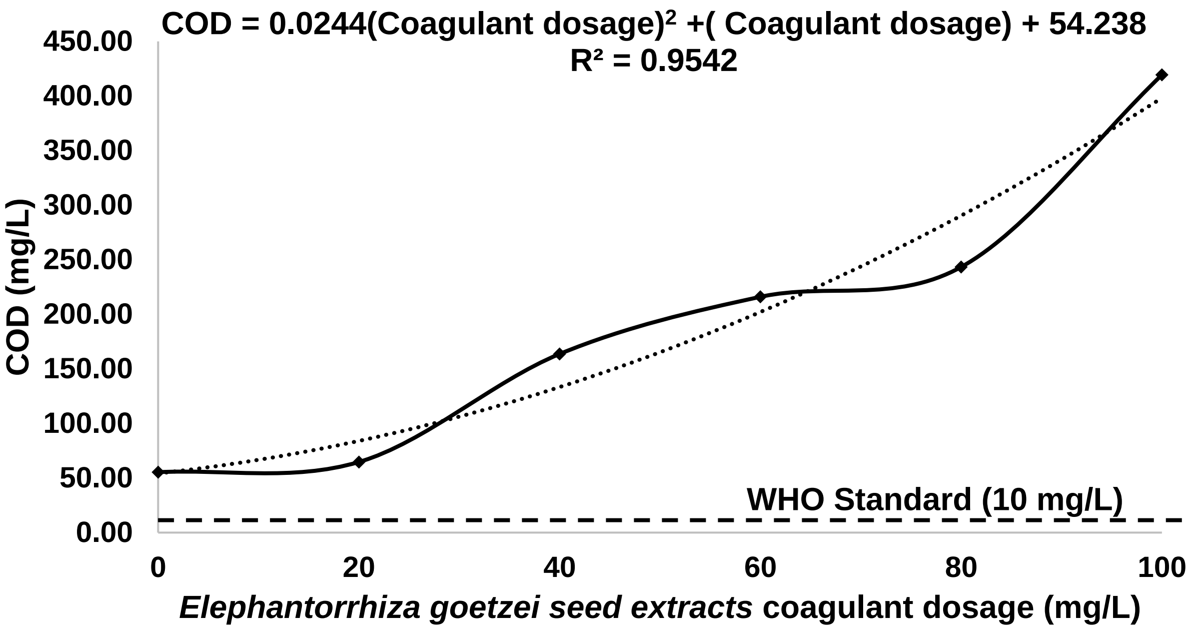

Figure 4.19 | Effect of *E.goetzei* seed extracts coagulant dosage on COD of treated water

4.5 Quality of sludge produced from treating synthetic water with *E.goetzei* seed extracts

4.5.1 Effect of E.goetzei seed extracts coagulant dosage on total nitrogen in sludge

As shown in Figure 4.20, the total nitrogen fluctuated as the coagulant dosage increased from 0 to 100 mg/L, which could be due to a phenomenon known as nitrogen rebound whereby nitrogen is re-accumulated in the sludge after a period of nitrogen reduction (Xiujie *et al.*, 2019). At the highest coagulant dosage of 100 mg/L, the total nitrogen was 0.343 µg/kg. The p value was less than 0.05, hence this indicates that coagulant dosage had a significant effect on the total nitrogen in the sludge. The R^2 value for the relationship between coagulant dosage and total nitrogen was 0.7467.

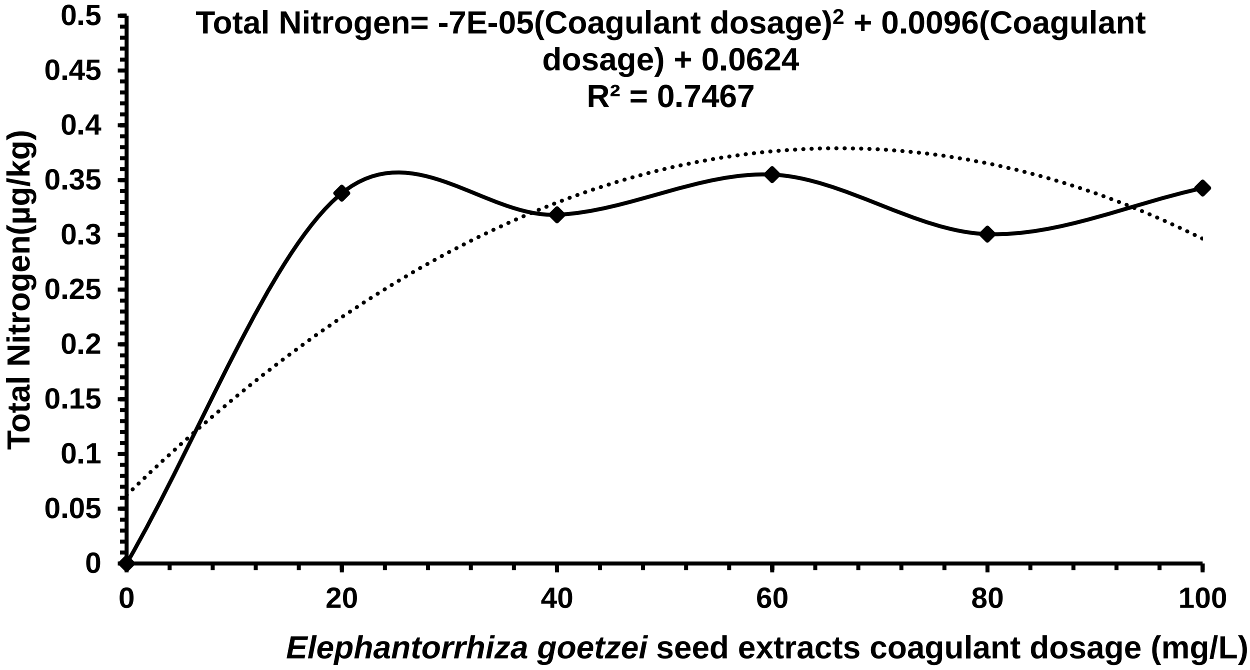

Figure 4.20 | Effect of *E.goetzei* seed extracts coagulant dosage on total nitrogen in sludge

4.5.2 Effect of E.goetzei seed extracts coagulant dosage on total phosphorus in sludge

Generally, there was an increase in the total phosphorus in the sludge as the coagulant dosage increased from 0 mg/L to 100 mg/L as depicted in Fig 4.21. At the maximum coagulant dosage used (100 mg/L), the total phosphorus was 0.194 µg/kg. Results from the ANOVA presented in Appendix C, show that *E.goetzei* seeds extracts coagulant dosage had a significant effect on the total phosphorus in sludge ($p < 0.05$).

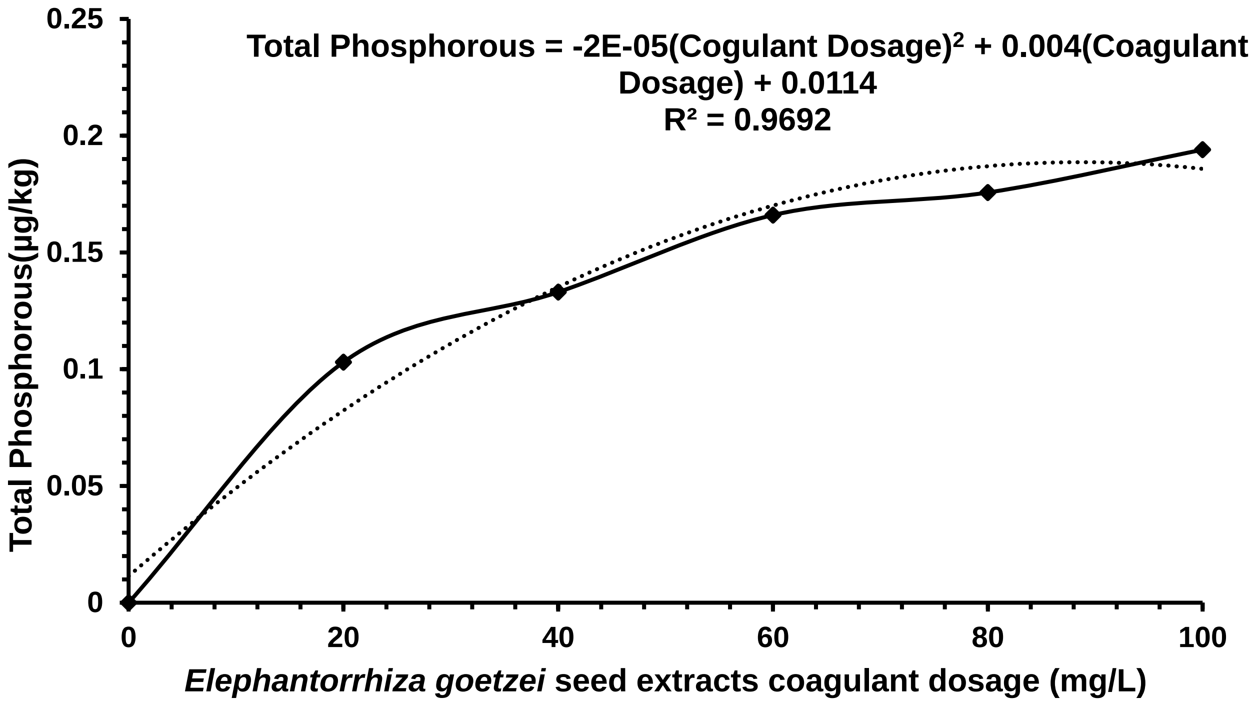

Figure 4.21| Effect of *E.goetzei* seed extracts coagulant dosage on total phosphorus in sludge

Chapter 5: Discussion

This chapter explains the results presented in the previous chapter by providing possible theories to justify the outcome obtained in Chapter 4. The results are also examined and compared with results which were obtained in other studies.

5.1 Determination of key ingredients in the *E.goetzei* seed extracts responsible for the coagulation process

The mean moisture content of the *E.goetzei* seed extracts was 5.25±0.14 %. The low moisture content of the seed extracts of *E.goetzei* is because the seed was still whole and the hard cell wall gives it protection against any penetration or accumulation of moisture. Low moisture content is beneficial since this can help to prevent the deterioration of the seeds, thereby improving the shelf life of the product (Xiao *et al*., 2011).

The value obtained for moisture content were also comparable to values which were obtained in other studies which reported moisture content of 5.35±0.20 % for *Jatropha curcas* seeds (Abidin *et al*., 2017), 4.00% for *Moringa oleifera Lam* (Abdulkadir *et al*., 2016) and 4.50 % for *Tympanotonos Fuscatus* shell (Menkiti *et al*., 2017). The moisture content of *E.goetzei* seed extracts obtained for this study was lower than the 12.58% which was reported by Obiora-Okafo *et al.* (2020) for fluted pumpkin (*Telfairia occidentalis*) seeds. This indicates that the seed extracts of *E.goetzei* might have a longer shelf life compared to the fluted pumpkin seed. This is because the moisture content of seeds, fruits and vegetables is indicative of their shelf life. The higher the moisture content, the more susceptible the seeds, fruits and vegetables will be to microbial attacks, hence reducing its shelf life (Thomas *et al*., 2018).

The crude protein content of *E.goetzei* seed extracts was 21.40±0.30%. A crude protein content of 21.40% is considered to be high and comparable to values obtained in already existing research (Nnaji *et al*., 2022; Sofiavizhimalar *et al*., 2022). The crude protein content was comparable to that obtained in separate studies for *Moringa oleifera* seeds, watermelon seeds, Mucuna seeds and *Hibiscus esculentus L.* (okra) (Magaji *et al*., 2015; Chikomo & Manyuchi, 2016; Menkiti *et al*., 2017; Okolo *et al*., 2021). Protein is a crucial component for neutralizing colloidal particles by attracting oppositely charged coagulants to the surface. This process triggers coagulation via bridging, which promotes the formation and growth of flocs (Igwegbe, *et al.*, 2021). The crude protein content of *E.goetzei* seed extracts provides active sticking sites for the coagulation to take place (Menkiti *et al*., 2017).

The carbohydrate content of the *E.goetzei* seed extracts was 29.93±0.82%. As depicted in Table 2.3, similar results were obtained for *Picralima nitida* and *Hibiscus esculentus* in separate studies (Igwegbe *et al.*, 2021; Okolo *et al.*, 2021). In contrast to the findings from the current study, higher carbohydrate content (61.07% - 68.33%) was obtained in various studies for natural coagulants such as *Pumpkin* seed and *Garnicia kola* seeds (Igwegbe *et al*., 2021; Gatew & Worku, 2023; Shabanizadeh & Taghavijeloudar, 2023). The carbohydrate molecules facilitate the formation of particle aggregates, which can contribute to the reduction of pollutants (Choy et al., 2015) . Moreover, the presence of carbohydrates implies the presence of hydrogen ions, and the bio-coagulant sample contains functional groups such as C-C and C-H bonds (Ovuoraye et al., 2022) .

The mean ash content of the seed extracts of *E.goetzei* was 2.20%. The ash content reflects the availability of carbon compounds and inorganic compounds in the form of salts and oxides. Due to its porous nature, carbon plays a very important role in the adsorption and removal of contaminants such as metals and other particles from water (Smith & Rodrigues, 2015). Based on the research by Iloamaeke & Julius (2019), it is possible that the seed extracts of *E.goetzei* can also act as coagulants in their granular or ash form.

Another constituent of particular interest is the fat content (32.99 ± 0.74 %) of *E.goetzei* seed extracts, which may contain oils that have surfactant properties. Surfactants are compounds that can lower the surface tension of water, a property that can facilitate the removal of impurities and pollutants (Adetunji & Olaniran, 2021). Therefore, the fat in *E.goetzei* seed extracts may have the potential as a natural surfactant for water treatment. Additionally, the crude fibre in *E.goetzei* seed extracts (8.23±0.29%) may also have some potential for water treatment. Fibre is known to absorb water and can be used to remove impurities and contaminants from water by acting as a filter (Zheng *et al*., 2016). Therefore, the fibre in *E.goetzei* seed extracts may have some potential as a natural filter for water treatment. The availability of fibre in the seed extract shows that the coagulant is an organic polymer with repeating small molecules that can extend as tails and loops when dispersed in water (Obiora-Okafo *et al*., 2020).

There were limitations which were noticed when the proximate analysis procedure was utilised to determine the key ingredients in the *E.goetzei* seed extracts. One of the main limitations is that it provides only a general overview of the seed's composition and does not identify individual components or compounds (Akintelu *et al*., 2020). Moreover, this procedure does not provide information on the presence of bioactive compounds such as phytochemicals, which can have important health benefits. Another limitation is faced in the determination of carbohydrate content. This is the only parameter in the Proximate Analysis which is not directly measured but is calculated by difference. Therefore, this parameter can easily accumulate all of the errors that exist in other proximate analysis components. To overcome this limitation, the experiments were done in triplicate to reduce any errors that might occur during the experimental run.

5.2 Determination of optimal dosages of *E.goetzei* seed extracts for the efficient removal of turbidity from synthetic water

The optimum coagulant dosages were 200 mg/L (for initial turbidities of 12.33 NTU, 28.46 NTU and 45.22 NTU) and 240 mg/L (for initial turbidities of 71.33 NTU,105.33 NTU and 167.67 NTU), since beyond these dosages analysed in this study, no significant reduction in turbidity was observed. Every coagulant has a unique crucial inflexion point, past which the aggregated particles would be re-dispersed once more, disrupting particle settling (Tawakkoly *et al*., 2019). Since excess coagulants no longer interact with oppositely charged colloidal particles once the majority of the colloids have been neutralised and precipitated, water turbidity may worsen if the coagulant dosage is exceeded (Tong *et al*., 2021). High turbidity removal (94.8 %) could have been obtained because of the soluble crude proteins in *E.goetzei* seed extracts (21.4 ± 0.30 %) which make the processes of coagulation and flocculation better (Vargas *et al*., 2022).

The highest removal efficiency was 94.8 % at an initial turbidity of 45.22 NTU at a coagulant dosage of 260 mg/L. The maximum removal efficiency obtained in this study is higher than the 69.4%, 81.6%, and 68.2% obtained for *Moringa oleifera, Cicer arietinum, and Dolichos lablab* respectively at a dosage of 100 mg/L and initial turbidity of 48 NTU (Asrafuzzaman *et al.*, 2011). Moreover, the maximum removal efficiency obtained when *E.goetzei* seed extracts coagulant was used for synthetic water with initial turbidity 71.33 NTU and coagulant dosage of 300 mg/L was 94.3%. Saritha *et al*.(2019) conducted a study using a similar turbidity of 70

NTU and achieved lower turbidity removal efficiencies of 92.6% and 85.6% using chitin and sago, respectively. For an initial turbidity of 105.33 NTU in the present study, *E.goetzei* seed extracts achieved a turbidity removal efficiency of 90.8 % at a coagulant dosage of 300 mg/L. This turbidity removal efficiency obtained in the present study is slightly lesser than the 95.2 % which was achieved using *Cactus opuntia* at a coagulant dosage of 50 mg/L and at a similar initial turbidity of 104 NTU (Zhang *et al*., 2006).

Avoiding an excessive or inadequate dosage that would ultimately lead to poor performance throughout the flocculation process is crucial (Remy *et al*., 2014). When *E.goetzei* seed extracts are added to turbid water, it is anticipated, as deduced from Vunain *et al.* (2019), that the protein in the seed extracts will connect with the negatively charged turbidity-causing particles such as clay and dust to create heavier flocs that can be precipitated.

The residual turbidity obtained in this study at the maximum dosage of 300 mg/L is 1.30, 2.58, 2.45, and 4.05 NTU at initial turbidities of 12.33, 28.46, 45.22, and 71.33 NTU, respectively. These complies with the WHO standards for turbidity in drinking water which stipulate that the turbidity should be kept below 5 NTU (WHO, 2017b). However, at initial turbidities of 105.33 and 167.67 NTU, the residual turbidities obtained at the maximum dosage of 300 mg/L were 9.64, and 11.89 NTU, respectively, and these did not meet WHO standards for turbidity in drinking water of 5 NTU. The models which were developed for initial turbidity of 105.33 NTU and 167.67 NTU were used to predict the final turbidity when there is further addition of the coagulant. At a dosage of 400 mg/L, the anticipated turbidity is 4.95 NTU and 4.81 NTU for initial turbidity of 105.33 NTU and 167.67 NTU respectively. Therefore, at a dosage of 400 mg/L it is anticipated the that the turbidity will satisfy WHO standards for turbidity in drinking water.

There was a slight change in the pH of the water when the coagulant was introduced into the water at different doses with the final pH being recorded as 8.00 at 300 mg/L coagulant dosage. The change in pH of the treated water could be explained by the increase in the concentration of dissolved substances due to the increase in the dosage of the coagulant (Skoronski *et al*., 2014). The slight change in pH is comparable to the change in pH of water which was treated with cactus powder in a study by Beyene *et al* (2016). The pH was slightly increased from 7.33 to 7.57 as the cactus powder dosage increased from 0.50 to 3.50 g. The pH in the current study was maintained within the pH range for drinking water of 6.5 - 8.5 (WHO, 2007). Results obtained in this study concur with the findings of Benalia *et al.* (2018), in which there was a slight change in pH with increasing coagulant dosage when acorn leaves were used to treat raw water from a water treatment plant.

Pandey *et al.* (2020), discovered that both *Moringa oleifera* leaves and alum (dosage 50 mg/L for both *Moringa oleifera* leaves and alum) managed to maintain the pH of the water close to the neutral position (7.6 for *Moringa oleifera* leaves hexane extracts and 7.0 for alum). Similarly, in the present study , *E.goetzei* seed extracts maintained the pH at an alkaline level which was between 8.16 and 8.00. In contrast to the findings obtained in the present study, a study by Vunain *et al.* (2019) demonstrated that there was an increase in pH from 4.8 to 7.1 at a *Moringa oleifera* dosage of 5 g/L. The pH then decreased to 6.2 as the coagulant dosage increased to 50 g/L. Cotton seed and Jute seed extracts decreased the pH from 7.81 to 5.13 and 6.64 respectively, therefore the pH obtained when Jute seed from 20 mL to 40 mL was used for water treatment is outside the WHO recommended range of 6.5 to 8.5 in drinking water (Lawal *et al*., 2020).

The increase in TDS with increasing *E.goetzei* seed extracts coagulant dosage is much higher (170.00 mg/L to 1316.28 mg/L) as the *E.goetzei* seed extracts coagulant dosage increased than that which was obtained by Beyene *et al*., (2016) in which the TDS value increased from 354 mg/L to 686 mg/L as the dosage of cactus powder increased. In a separate study by Vunain *et al.* (2019), there was an increase in TDS from 251.9 mg/L to 542.8 mg/L as the *Moringa oleifera* seed coagulant dosage increased from 15 g/L to 50 g/L. However, in a study by Pandey *et al.* (2020), there was a decrease in TDS from 512 ± 9.612 mg/L to 322 ± 15.34 mg/L as *Moringa oleifera* leaves' dosage increased from 50 mg/L to 150 mg/L and from 512 ± 9.612 mg/L to 226 ± 11.07 mg/L when alum with dosage of 50 mg/L was used for treatment of groundwater (Pandey, *et al*., 2020). Analysis of TDS includes mineral salts, metals, cations, and anions dissolved in water. Increasing the concentration of *E.goetzei* seed extracts coagulant in water treatment is likely to elevate the levels of anions and cations present in the water, which may be released from the coagulant solution. This provides a reasonable explanation for the observed rise in TDS in synthetic water as the dosage of *E.goetzei* seed extracts coagulant increased (Vunain *et al*., 2019). Moreover, the increase in TDS is much higher than the increase in coagulant dosage, and this could be attributed to the presence of elevated concentrations of specific dissolved salts such as chlorides, in the synthetic turbid water . These salts can have a substantial impact on TDS levels. In such cases, the coagulant's effectiveness in removing these salts may be constrained, resulting in a faster increase in TDS levels relative to the coagulant dosage (Li *et al*., 2020).

Synthetic water was used to obtain the results in the present study. However, the quality of groundwater can change over time due to factors like seasonal fluctuations, variations in recharge rates, and human activities. The fluctuations in the quality of surface water can lead to modifications in protocols for treating water to make it safe for consumption. Variations in certain physicochemical factors, such as the pH and alkalinity of the raw water, have an impact on the water treatment process (Mensah-Akutteh *et al*., 2022). Synthetic water may not accurately reflect these temporal variations. Consequently, it is crucial to account for the temporal variability in groundwater quality and evaluate how treatment processes would function under diverse conditions (Mohsine *et al*., 2023).

5.3 Effect of *E.goetzei* seed extract coagulant dosage on the removal of fluoride, manganese and iron from synthetic water

5.3.1 Effect of E.goetzei seed extracts coagulant dosage on fluoride removal.

The highest fluoride removal efficiency obtained from this study was 50.1 % at a coagulant dosage of 100 mg/L. Higher concentrations of the *E.goetzei* seed extracts coagulant solution may be more effective at removing fluoride because there are more active sites on the adsorbent, which improves fluoride adsorption (Masquetti *et al*., 2015). The fluoride removal efficiency of 50.1% at a dosage of 100 mg/L was more than the removal efficiency obtained of 29.3 % at an *Azadirachta indica* coagulant dosage of 100 mg/L, 46.4% at a *Moringa oleifera* coagulant dosage of 100 mg/L, 14.3% at a *Moringa oleifera* coagulant dosage of 50 mg/L when the two natural coagulants were used to treat groundwater samples with initial pH (8.2 ± 0.2) which was similar to the pH of the synthetic water which was used for the current study (8.2 ±0.1) (Pandey *et al.*, 2020). However, when the leaves of Azadirachta indica and *Moringa oleifera* were combined to treat groundwater samples in the study by Pandey *et al.* (2020), a higher fluoride removal efficiency of 64.3% was achieved at a coagulant dosage of 100 mg/L of which was similar to the *E.goetzei* seed extracts coagulant dosage which was used in the present study. Therefore, the combination of these two natural coagulants has a higher fluoride removal efficiency than *E.goetzei* seed extracts. Higher removal efficiencies of 91.0% were achieved when a *Moringa oleifera* dosage of 2,500 mg/L was utilised to treat fluoridated reverse osmosis water which had an initial fluoride concentration of 10 ppm (Bazanella *et al.*, 2012). The higher removal efficiencies obtained by Bazanella *et al.* (2012) at similar initial fluoride concentrations to the one used in the present study can be attributed to the much higher concentration of *Moringa oleifera* (2,500 mg/L) which was used as compared to the 100 mg/L which was used for *E.goetzei* seed extracts.

Singh *et al*. (2020) investigated *Tamarindus indica, Moringa oleifera*, and *Tinospora cordifolia* seed powders for defluoridation of synthetic fluoridated water which had a similar initial fluoride concentration (10 ppm) to the one used in the present study. Removal efficiencies achieved were 80.0%, 70.0%, and 54.0% for *Tamarindus indica, Moringa oleifera,* and *Tinospora cordifolia,* respectively. All of the three seed powders investigated in the study by Singh *et al*. (2020) achieved removal efficiencies higher than the fluoride removal efficiency of 50.1% obtained by *E.goetzei* seed extract coagulant dosage. The ability of *E.goetzei* seed extracts to remove fluoride from water could be due to positively charged proteins in the seed extracts that interact electrostatically with the negatively charged fluoride ion to make water-soluble and slightly soluble compounds. This ion exchange procedure might have aided in the production of flocs that gravitationally settled as reported by Masquetti & Ueda, (2021) who used *Moringa oleifera* for groundwater defluoridation. The residual fluoride at 100 mg/L coagulant dosage was 5.57 mg/L and did not meet WHO standards of 1.50 mg/L for fluoride in drinking water. However, using the model in Fig 4.10 to predict the final fluoride concentration in the synthetic water for coagulant dosages which were not investigated in the study, it is anticipated that the fluoride will be approximately 1.15 mg/L at a coagulant dosage of 180 mg/L thereby meeting the WHO standards for fluoride in drinking water.

5.3.2 Effect of initial fluoride concentration on fluoride removal efficiency in synthetic water

The findings in this study show that there was an increase in fluoride removal efficiency from 21.0% to 53.1% as the initial fluoride concentration was increased from 5 mg/L to 25 mg/L. Similarly, in a study by Masquetti & Ueda (2021), there was an increase in fluoride removal efficiency from 56.7% to 72.0% with an increase in initial fluoride concentration from 3.0 ppm to 7.5 ppm at a coagulant dosage of 2 g/L when *Moringa oleifera* was used to treat fluoridated distilled water.

The findings from the current study are also in agreement with the findings of Masquetti *et al.* (2015) who found out that when *Moringa oleifera Lam* was used for treatment of fluoridated distilled water at coagulant dosage of 5 g/L, and pH 3 and 7, the removal efficiency increased from 68.7% to 72.0% and 60.0% to 71.4%, respectively, as the initial fluoride concentration was increased from 3 mg/L to 10 mg/L. However, a study by Bayu *et al.* (2022) revealed that fluoride removal efficiency decreased from 79.8% to 59.5% as the initial fluoride ion concentration was increased from 5.5 mg/L to 10 mg/L when using leaves of *Tulsi* to treat artificially fluoridated water.

Findings from the study by Agnihotri *et al.*(2013), demonstrate that there was a decrease in removal efficiency from 22.0% to 6.3 % as the initial fluoride concentration was increased from 2 mg/L to 10 mg/L at a coagulant dosage of 4 g/L when *Moringa oleifera* seed residue was used to treat synthetic fluoride contaminated water. The greater removal efficiency at lower initial concentrations of fluoride could be due to the many positively charged sites available on the coagulant to bond with negatively charged fluoride ions (Agnihotri *et al*., 2013).

5.3.3 Effect of E.goetzei seed extracts coagulant dosage on iron and manganese removal

There was an increase in iron removal efficiency as the coagulant dosage was increased from 0 to 100 mg/L. The highest iron removal efficiency obtained from this study was 90.0% at 100 mg/L. The residual iron concentration at 100 mg/L coagulant dosage was 0.55 ± 0.02 mg/L. The iron removal efficiency obtained when using *E.goetzei* seed extracts is the same as the iron removal efficiency of 90.0% which was obtained in the study by Sajidu *et al.* (2005) when *Moringa oleifera* seeds of dosages 2,500 mg/L was used to treat synthetic heavy metal contaminated water with an initial iron concentration (5.00 ppm) which was similar to the initial concentration of iron in the synthetic water sample used for the current study.

Pinecones powder achieved a removal efficiency of 94.0% at a coagulant dosage of 3,000 mg/L when the powder was used to treat industrial wastewater with an initial iron concentration of 5.30 mg/L (Shadi *et al*., 2022). This removal efficiency was slightly higher than the 90.0% which was achieved at a similar initial iron concentration for *E.goetzei* seed extracts in the present study. Similarly, Abujazar *et al.* (2022) used rosehip powder to treat industrial wastewater which had an initial iron concentration of 5.30 mg/L which was similar to the iron concentration of the synthetic water for the present study, which was 5.50 mg/L. The rosehip powder was used to achieve an iron removal efficiency of 91.7% which was almost the same as the iron removal efficiency achieved when *E.goetzei* seed extracts coagulant was used in the current study.

There was an increase in manganese removal efficiency with increasing coagulant dosage. The highest removal efficiency obtained from this study was 53.9% at a coagulant dosage of 100 mg/L. The residual manganese concentration at a coagulant dosage of 100 mg/L was 2.52± 0.02 mg/L and did not meet WHO standards for manganese in drinking water (<0.1 mg/L). Therefore, the model in Figure 4.16 was used to predict the coagulant dosage at which a manganese concentration of at least 0.1 mg/L would be achieved. Using the model in Figure 4.16, it is anticipated that the WHO standard for manganese in drinking water will be met at an *E.goetzei* seed extracts coagulant dosage of 540 mg/L.

The highest manganese removal efficiency obtained in this study at a coagulant dosage of 100 mg/L is slightly lesser than the 59.7% which was obtained when 100 mg/L of banana pith was used to remove manganese in river water (Kakoi *et al*., 2016). The initial manganese concentration used in the study by Kakoi *et al*. (2016) which was 4.32 mg/L is comparable to the initial manganese concentration of 5.46 mg/L which was used in the study for determining effect of *E.goetzei* seed extracts coagulant dosage on the manganese concentration. Pine cones powder and rosehip seed powder also achieved higher removal efficiencies (86.8% and 85.7% at dosages of 3,000 mg/L and 1,000 mg/L respectively) than *E.goetzei* seed extracts for water samples with similar initial manganese concentrations (Abujazar *et al*., 2022; Shadi *et al.*, 2022).

The process by which the elimination of iron and manganese takes place was not investigated during this research, but it might be by precipitation after oxidation. *E.goetzei* seed extracts may therefore contain oxidizing chemicals that change and cause iron and manganese to precipitate as oxides (Nkurunziza *et al*., 2009). *E.goetzei* seed extracts managed to remove iron and manganese from water and this could be due to the adsorption of these elements into the flocs and the precipitation of some of these impurities with precipitates. The increase in removal efficiency of iron and manganese with increasing coagulant dosage can be attributed to the higher dose permitting colloidal suspension destabilization and floc development (Pramanik *et al*., 2016).

5.3.4 Effect of initial manganese and iron concentration on manganese and iron removal efficiency

When the initial metal concentration was increased, the removal efficiency was also increased and after the threshold had been reached, any further increase in the metal ions concentration then resulted in lesser removal efficiency being achieved. This phenomenon could be taking place due to the saturation of the binding sites on the coagulant as more ions are added to the solution. The number of metal ions vying for available binding sites on the coagulant rises as metal ion concentration rises. As a result, binding sites soon reach saturation since the coagulant's concentration stays constant (Sharma *et al*., 2006; Rahman & Islam, 2009). The increase in electrostatic contacts (compared to covalent interactions) that involve locations with increasingly lower affinity for metal ions is responsible for the improvement of metal absorption (Obuseng *et al*., 2012).

The major limitation when determining the optimum dosage and the effect of coagulant dosage on turbidity, iron, manganese, and fluoride on removal efficiency was limited financial resources when conducting the experiments. Therefore, due to limited financial resources, the researcher could not conduct the experiments on a wider coagulant dosage range. However, the researcher managed to develop models so as to assess expected values after further dosing using *E.goetzei* seed extracts coagulant. Moreover, another limitation that was encountered when carrying out the research was the lack of field validation to facilitate the transition of the research from the laboratory to household level water treatment facilities. Use of the *E.goetzei* seed extracts coagulant on a larger scale can account for how the coagulant responds to changes in water quality and differences in operational conditions which are faced during the treatment of water at household level.

5.4 Levels of residual content of organic matter in water treated with *E.goetzei* seed extracts

The initial COD concentration in the synthetic water sample was 44.67 mg/L. The elevated level of COD in the synthetic water could be due to organic impurities in the kaolin clay (Aroke *et al*., 2013). There was an increase in COD from 44.67 to 419.33 mg/L of the water as the coagulant dosage increased from 20 to 100 mg/L. Similarly, in a study by Shan *et al.* (2017), as the dosage of *Moringa oleifera* seed increased from 10,000 mg/L to 30,000 mg/L, the COD increased from 99.50 mg/L to 164.00 mg/L in river water and from 529.50 mg/L to 987.00

mg/L in a leachate water sample. In another study by Ghebremichael (2007), the COD also increased from 10 mg/L to 90 mg/L with increasing *Moringa oleifera* seed dosage from 0 µL to 500 µL.

In agreement with the findings from the current study, Ndabigengesere and Narasiah (1998) used shelled and non-shelled seeds of *Moringa oleifera* seeds for the treatment of synthetic turbid water. As the coagulant dosage increased from 5 mL/L to 20 mL/L, there was an increase in COD from 60 mg/L to 200 mg /L and from 70 mg/L to 300 mg/L for shelled and non-shelled *Moringa oleifera* seeds, respectively. The COD increased with an increase in *E.goetzei* seed extracts coagulant dosage because of the oil which is present in the seed extracts (Shan *et al*., 2017). The aqueous extract of *E.goetzei* seed extracts also contains organic matter such as lipids and carbohydrates which elevate the organic load in the treated water. The maximum COD (419.33 mg/L) obtained in this study is much higher than the WHO standard for COD in drinking water of 10 mg/L. Therefore, to prevent the rise of organic content following coagulation with *E.goetzei* seed extracts, an alternative is to incorporate a purification step during the preparation of the *E.goetzei* seed extracts coagulant. This purification process aims to separate and purify the active agents by removing the elements within the raw seed that contribute to the increase in organic load (Vega *et al*., 2021).

The major limitation faced when determining the COD of the treated water is the interference from other compounds that might be present in the water sample. The measurement of COD in treated water can be affected by the presence of diverse organic and inorganic compounds. Compounds such as remaining disinfectants, suspended solids, and specific organic substances have the potential to cause inaccurate COD readings, resulting in both overestimations and underestimations of the actual COD levels and compromising the measurement's accuracy. This limitation was addressed by suppressing these elements such as chlorine so that they do not interfere with the COD results. Moreover, the researcher did not manage to conduct experiments on pathogen removal (a parameter linked to the COD concentration in water) using *E.goetzei* seed extracts, due to constraint in resources. Therefore, further studies need to be conducted to determine whether *E.goetzei* seed extracts can remove pathogens in water. The implication of the proposed further studies is that if further disinfection using chlorine is required, then a way of solving the problem of elevated COD in water after treatment with the *E.goetzei* seed extracts will be needed. This is because when water containing organic matter is exposed to chlorine, it generates disinfection by-products (DBPs) such as trihalomethanes

(THMs) and halo acetic acids (HAAs), which are substances that pose health risks (Mazhar *et al*., 2020).

5.5 Quality of sludge produced from treating synthetic water with *E.goetzei* seed extracts

The total nitrogen fluctuated between a slight increase and a slight decrease as the coagulant dosage increased from 0 to 100 mg/L. At the highest coagulant dosage of 100 mg/L, the total nitrogen was 0.343 µg/kg. The total nitrogen at a coagulant dosage of 100 mg/L is lower than the total nitrogen of 0.419 g/kg which was obtained in a separate study in which 25 mg/L *Moringa oleifera* seed was used to treat river water with initial turbidity up to 380 NTU (Feria-Díaz *et al*, 2016). There was an increase in the total phosphorus in the sludge from 0.073 µg/kg to 0.194 µg/kg as the coagulant dosage increased from 0 mg/L to 100 mg/L. At the highest coagulant dosage of 100 mg/L, the total phosphorus was 0.194 µg/kg. Feria-Díaz *et al*. (2016), obtained a much higher value of total phosphorus (0.002 g/kg) when *Moringa oleifera* seeds (25 mg/L coagulant dosage) were used to treat river water with initial turbidity of up to 380 NTU.

Moreover, the total nitrogen and phosphorus content obtained in this study is much lesser than the values obtained by Olivares-campos & Jáquez-balderrama (2012), for compost samples which were 22.00 g/kg for nitrogen and 1.40 g/kg for phosphorus. Furthermore, the nitrogen and phosphorus in the current study were also lesser than the values obtained for organic fertilizer which were 1.32 g/kg and 0.21 g/kg for nitrogen and phosphorus respectively (Lam & Lee, 2012). The nitrogen and phosphorus in the sludge obtained when *E.goetzei* seed extracts were used to treat water were lesser than the values obtained for municipal sewage sludge in the study by Antonkiewicz *et al.* (2020), which were 13.70 g/kg and 2.80 g/kg respectively. Higher values of 39.8 g/kg and 20.6 g/kg for nitrogen and phosphorus respectively in sewage sludge were also obtained (Pöykiö *et al*., 2019), as compared to the lower values for nitrogen and phosphorus obtained for the sludge in the present study.

The main limitation that was faced when determining total nitrogen in the sludge was the possible volatile loss of nitrogen compounds. Certain nitrogen compounds have the potential to evaporate during sample handling, digestion, or analysis, causing their loss (Sáez-Plaza *et al*., 2013). As a consequence, the measured levels of total nitrogen can be reduced, resulting in an underestimation of its concentration.

5.6 Protocol for water purification in rural households and economic assessment of ***E.goetzei*** seed extracts as a natural coagulant at the household level

The powder of the *E.goetzei* seed extracts coagulant can be used at the household level via the traditional method of using natural coagulants. The traditional method is appropriate since the seeds are readily available in rural areas such as Walden North Farm, and Chegutu District. The method explained by Marobhe *et al* (2007), was adopted in this study. The equipment and materials to be used include 2 buckets with a capacity of 20 L each, cotton cloth, and *E.goetzei* seeds. The protocol for water purification in rural households using *E.goetzei* seeds is shown in Fig 5.1. The seeds are first collected and dried in the sun for 24 hours after which the brown coat is removed and the seeds that are damaged are disposed of. The seeds are then crushed using a mortar and pestle to attain a fine homogenous powder. Sodium chloride salt (58.44 g) is then dissolved in 1 litre of water to obtain a 1 molar sodium chloride solution which will be used to prepare the coagulant solution. The *E.goetzei* solution is then prepared by adding 10 g of *E.goetzei* seed powder to 1 L of sodium chloride in a container and shaking the container for 30 minutes. The solution is then filtered using a cotton cloth to obtain the coagulant solution to be used for treating the water. The *E.goetzei* solution (480 mL) is then introduced to the coagulation -flocculation tank (20 L bucket). This dosage is based on the optimum dosage of 240 mg/L and initial turbidities of 71.33 NTU,105.33 NTU and 167.67 NTU used for this specific study. The dosage required may differ depending on the initial turbidity of the water to be treated. After introducing the *E.goetzei* coagulant solution, the water is then manually stirred rapidly (120 rpm) for one minute and slowly (30 rpm) for 15 minutes (Gidde *et al*, 2012). The formed flocs are then allowed to settle for 15 minutes. Stirring is done using readily available wooden cooking sticks. The clarified water is then decanted and filtration will be done by reusing cotton cloth available within the households. The water is then stored in a clean bucket or clay container. The rapid and slow mixing which is then followed by settling is adopted from the jar tests which are done to determine optimum doses for coagulation-flocculation in large-scale water treatment plants (Marobhe *et al*, 2007).

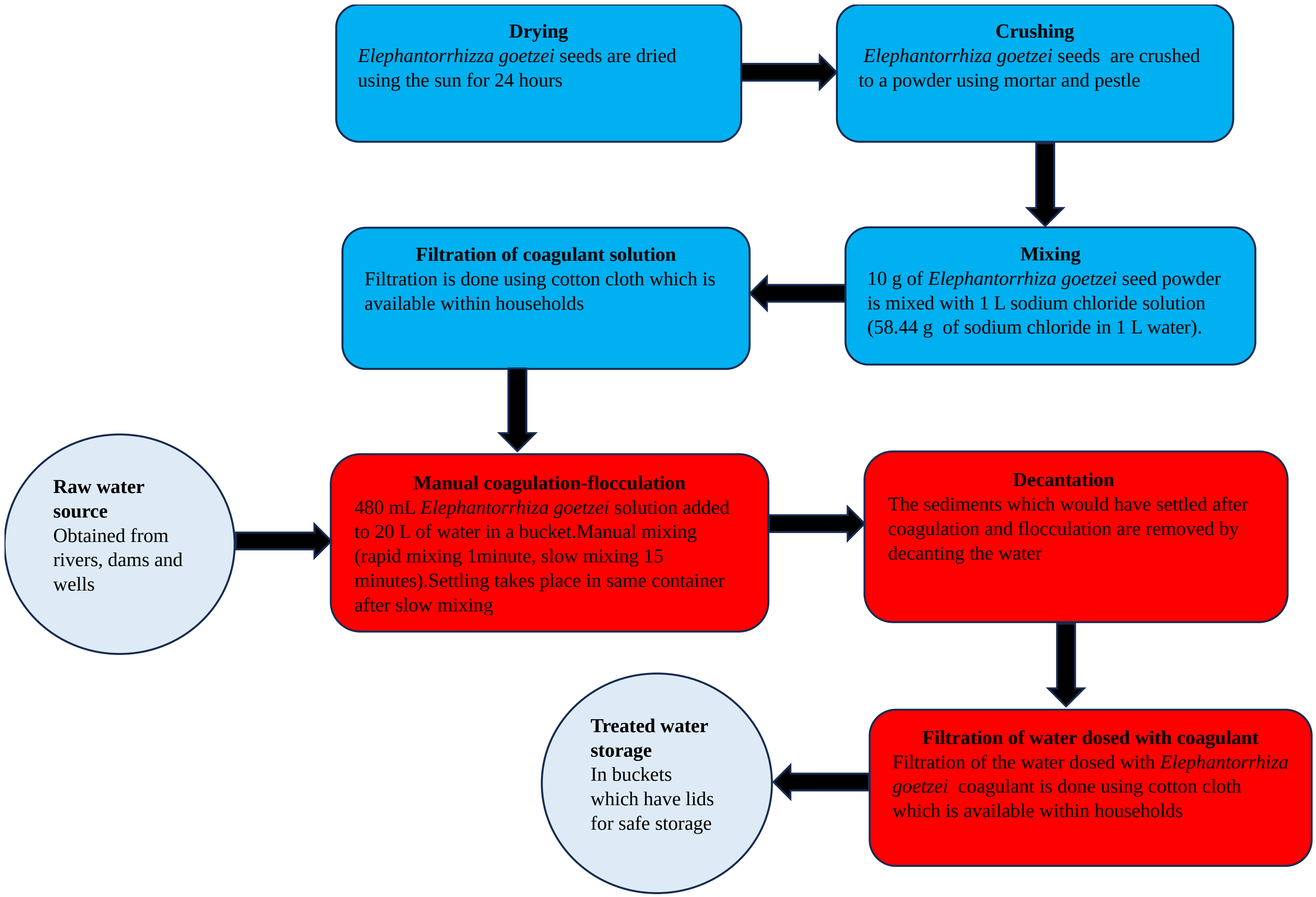

Figure 5.1 | Protocol for water purification in rural households using *E.goetzei* seed extract

Conducting an economic feasibility analysis holds significant importance as it directly impacts the potential for commercializing a product in industrial applications. The primary expense in the coagulation-flocculation process mainly consists of material costs (Igwegbe *et al.*, 2021). In the present study, the traditional method which was proposed for use in households entails that there will be limited costs incurred in collecting and using the natural coagulant since the *E.goetzei* seeds are readily available in Chegutu district and can be collected by the villagers and used free of charge. Collection of the seeds by the local people will potentially be an efficient process since the local people already have in-depth knowledge about the *E.goetzei* plant as they are already using it for treatment of health problems such as flue, stomach pains, and back pains. Moreover, the villagers will reuse cotton cloth for filtration, make use of buckets and stirrers (traditional wooden cooking spoons) that they already have, thereby further limiting material costs. The cost of salt (as obtained from OK supermarket in Zimbabwe) used for extracting the active coagulation ingredients in the seeds is \$0.012 for every 20 L bucket of water, which then translates to \$0.60 per m^3. The cost of treating water with *E.goetzei* seeds (\$0.60/m^3) using the protocol adopted in this study is slightly higher than the \$0.56 /m^3 obtained by Thirugnanasambandham & Karri (2021) for *Azadirachta indica A. Juss*. The \$0.60/ m^3 obtained for the present study is greater than the \$0.042/ m^3 cost for treating water using *Moringa stenopetala* for treatment of synthetic turbid water (Megersa et al., 2019). However, when alum was used by Thirugnanasambandham & Karri (2021), a greater cost (\$1.73/ m^3) was incurred than the cost of \$0.60/m^3 incurred for treating synthetic turbid water in the present study using *E.goetzei* seeds. Conclusively, it is economically feasible to use *E.goetzei* seed extracts for water treatment. This is because the cost of using *E.goetzei* seed extracts for water treatment was found to be lower than using alum whilst at the same time it was almost the same as other natural coagulants such as *Azadirachta indica A. Juss.*

Chapter 6: Conclusions and recommendations

This chapter presents the conclusions that were derived from the findings from each of the five objectives that were investigated in this study. Moreover, recommendations on how to improve the study and potential areas for further research are also presented.

6.1 Conclusions

The conclusions are presented for each objective of the study. The conclusion section of this thesis therefore provides a concise and conclusive summary of the study as follows:

1. Based on the findings from this study, it can be concluded that the possible active ingredients in *E.goetzei* seed extracts are proteins, carbohydrates, fat, and fibre as they all possess unique properties which enable them to remove impurities in water. These components exhibit distinct characteristics that enable them to effectively eliminate impurities in water. The properties that make them fit to be active ingredients include the ability to neutralize colloidal particles, possessing negative charges to attract metallic ions and having surfactant properties.

2. Based on the findings from this study, it can be concluded that *E.goetzei* seed extracts coagulant can be used to treat water with initial turbidity of up to 71.33 to the required WHO standard of 5 NTU in drinking water. It was also concluded that as the coagulant dosage increased, the turbidity, fluoride, iron and manganese removal efficiency also increased. Furthermore, as the coagulant dosage increased there was an increase in the TDS of the treated water whilst there was a slight change in the pH of the treated water, nonetheless , it was maintained within the WHO recommended range of 6.5-8.5. *E.goetzei* seed extracts coagulant has potential to achieve the desired WHO drinking water standard at higher dosages for fluoride and iron, and manganese. Generally, there was an increase in removal efficiencies for fluoride and iron as the initial concentration of contaminants increased in water. As for manganese, the removal efficiency began to decrease after the saturation point had been reached. Residual fluoride and manganese at the highest coagulant dosage did not meet WHO standards for fluoride and manganese in drinking water which are 1.5 mg/L and <0.1 mg/L respectively. However,

the models developed in the study show that with further coagulant dosing, the WHO standards for fluoride and manganese in drinking water can be met. The TDS value at the highest coagulant dosage was above the WHO guideline value of 600 mg/L.

3. Based on the findings from this study, it can be concluded that there is a general trend of increasing COD in the treated water as the coagulant dosage increases ($R^2 = 0.9542$). This suggests that higher dosages of the coagulant may introduce or contribute to the presence of organic compounds or substances that contribute to the COD value.

4. Based on the findings from this study , it can be concluded that the content of nitrogen and phosphorus in the sludge after water treatment is not viable for reuse applications in agriculture. It is significantly lower compared to compost and organic fertilizer. This finding hinders the use of sludge as a fertilizer, as it may not provide sufficient levels of these essential nutrients required for plant growth.

5. Findings from the study demonstrate the suitability of *E.goetzei* seed extracts for water treatment. This is because the seeds contain the active coagulation ingredients such as proteins and carbohydrates which enables the seed extracts to be used for water treatment. Furthermore, the seed extracts of *E.goetzei* are suitable for water treatment since they can remove contaminants such as iron , fluoride, manganese and turbidity in water.

6.2 Recommendations

The recommendations section of this thesis plays a significant role in offering insightful suggestions based on the research and analysis discussed in the earlier chapters. This part, which draws on the research done, tries to provide feasible recommendations that can advance the field of study and address any gaps or challenges that were identified during the research. Based on the respective objectives, the following recommendations were derived from the findings of this research:

1. More studies need to be done on the proteins which are part of the active ingredients in the seed extracts of *E.goetzei*. The proteins can be characterised to determine their molecular weight and amino acid sequences. Further experiments must also be

conducted to determine the presence of bioactive compounds such as phytochemicals in the *E.goetzei* seed extracts.

2. Further studies should be carried out using a wider coagulant dosage range of the *E.goetzei* seed extracts for removal of turbidity, iron, manganese, and fluoride to determine the dosage which is required to achieve WHO standards for drinking water quality and compare with the values obtained from the models used in this study. More studies are also required to investigate the effect of other operational parameters such as pH, mixing speed during coagulation-flocculation, settling time and coagulant extraction method on the turbidity, iron, fluoride and manganese removal efficiency. It is also important to investigate the effect of combining seed extracts of *E.goetzei* with other natural coagulants such as *Moringa oleifera*, *Cactus Opuntia* and *Azadirachta indica* on the turbidity removal efficiency of *E.goetzei* seed extracts. Further work must also be done on assessing the efficiency of the seed extracts of *E.goetzei* as a coagulant on a typical large-scale water treatment plant or at the household level in marginalised areas. Moreover, studies on the effects of temperature variations are also key to determine the thermodynamics and kinetics of the coagulant removal process.

3. There is a need to investigate ways of dealing with the issue of an increase in COD in treated water with increasing coagulant dosage to improve the overall quality of the water. This can be done by examining different methods for preparing *E.goetzei* seed extracts coagulant solutions. Different extraction techniques, concentration, and processing parameters can be explored to identify the best way to manage the issue of an increase in COD levels of treated water.

4. Further studies must be done on the sludge produced from water treatment using *E.goetzei* seed extracts to determine other parameters of the sludge such as water retention to come up with a beneficial use for the sludge. The sludge from water treatment using seed extracts of *E.goetzei* must also be tested to determine whether it can be successfully used in the revegetation of degraded land within the environment.

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Appendices

Appendix A | ANOVA results for effect of initial turbidity and *E. goetzei* seed extracts coagulant dosage on turbidity removal efficiency

Source	Type II Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	47617.791[a]	20	2380.890	107.833	.000
Intercept	557450.244	1	557450.244	25247.584	.000
Initial Turbidity	618.295	5	123.659	5.601	.000
Coagulant Dosage	46999.496	15	3133.300	141.911	.000
Error	1655.951	75	22.079		
Total	606723.987	96			
Corrected Total	49273.743	95			

Appendix B | ANOVA results for effect of initial turbidity and coagulant dosage and on residual turbidity

Source	Type II Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	36646.508[a]	20	1832.325	11.627	.000
Intercept	26692.340	1	26692.340	169.383	.000
Initial Turbidity	13764.285	5	2752.857	17.469	.000
Coagulant dosage	22882.223	15	1525.482	9.680	.000
Error	11818.957	75	157.586		
Total	75157.804	96			
Corrected Total	48465.465	95			

Appendix C | ANOVA results for effect of *E. goetzei* seeds extracts coagulant dosage on total phosphorous

	Sum of Squares	df	Mean Square	F	Sig.
Between Groups	.075	5	.015	51.869	.000
Within Groups	.003	12	.000		
Total	.079	17			

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