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SLUDGE DEWATERING FROM WWTP USING NATURAL COAGULANT - MORINGA OLEIFERA EXTRACT

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ABSTRACT

The world is facing a water crisis because the population is growing, people are using more water, and more wastewater is being produced. The increase in wastewater production means that there needs to be more treatment of this waste. This treatment generates sludge, which is dangerous for the environment and for people's health. Dewatering sludge is an important part of treating it, which can be done better by treating this with coagulants. The seeds of *Moringa oleifera*, or MO, have the potential to be employed as a coagulant, making them a less expensive and environmentally friendly alternative. MO seeds were shelled, crushed, and their oil content removed before being dried at 45°C for 24 hours to extract the active coagulant. The active coagulant components were extracted by mixing the processed seed with a NaCl solution (10 % m/v), being the 1.0 M concentration found to be optimal. The extraction was done in two ways: one with ordinary agitation (MOC) and the other with ultrasound assistance (MOC-U), both of which were filtered to remove undissolved particles. The sludge was conditioned using a jar test with fixed rapid and slow mix configurations and different coagulant dosages. The dewatering trials were carried out for 24 hours in dewatering columns with varying sludge height (10, 20 and 40 cm) and a fixed 10 cm filtering medium of inert material. TS, volume of filtered removed, COD, pH, and turbidity were the parameters analyzed. For all experiments, the MOC coagulant increased the sludge's dewatering capacities, however the MOC-U coagulant had most of the outcomes below the control experiment results. The MOC coagulant produced the most satisfactory results in studies with an initial sludge height of 10 cm. The best TS and filtrated removal values were 18.6% and 87 mL, respectively. COD values have been demonstrated to increase as coagulant dosage is increased. In addition, the initial sludge height has been shown to have a negative impact on the sludge's dewatering abilities. To optimize the extraction process employing ultrasound technology, more research is needed. Further testing in the conditioning phase is also required to enhance the sludge dewatering process.

Keywords: *Moringa oleifera*; natural coagulant; sludge dewatering; WWTP sludge treatment

RESUMO

O mundo enfrenta uma crise de água porque a população continua a crescer, as pessoas estão a utilizar mais água, e a produzir mais água residual. O aumento da produção de águas residuais torna necessário haver mais tratamento destes resíduos. Este tratamento gera lamas, que são perigosas para o ambiente e para a saúde das pessoas. A desidratação das lamas são uma parte importante do seu tratamento, o que pode ser melhorado tratando-as com coagulantes. As sementes de *Moringa oleifera*, ou MO, possuem potencial para serem empregadas como coagulante, tornando-as uma alternativa menos custosa e ecologicamente correta. As sementes de MO foram descascadas, trituradas, e o seu teor de óleo foi removido antes de serem secas a 45°C durante 24 horas para extrair o coagulante activo. Os componentes do coagulante activo foram extraídos misturando a semente processada com uma solução de NaCl (10 % m/v), sendo a concentração de 1.0 M óptima. A extração foi feita de duas maneiras: uma com agitação normal (MOC) e outra com assistência de ultra-sons (MOC-U), ambas filtradas para remover partículas não dissolvidas. A lama foi condicionada utilizando um teste em frasco com configurações de mistura rápida e lenta fixas e diferentes dosagens de coagulante. Os ensaios de desidratação foram realizados durante 24 horas em colunas de desidratação com altura variável da lama (10, 20 e 40 cm) e um meio filtrante fixo de 10 cm de material inerte. TS, volume de filtrado removido, COD, pH e turbidez foram os parâmetros analisados. Para todas as experiências, o coagulante MOC aumentou a capacidade de desidratação da lama, contudo o coagulante MOC-U teve a maioria dos resultados abaixo dos resultados da experiência de controlo. O coagulante MOC produziu os resultados mais satisfatórios em estudos com uma altura inicial de lodo de 10 cm. Os melhores valores de TS e de remoção de filtrado foram de 18.6% e 87 mL, respectivamente. Foi demonstrado que os valores de COD aumentam à medida que a dosagem do coagulante é aumentada. Além disso, foi demonstrado que a altura inicial da lama tem um impacto negativo nas capacidades de desidratação da lama. Para optimizar o processo de extração empregando tecnologia de ultra-sons, é necessário realizar nova investigações. São também necessários mais testes na fase de condicionamento para melhorar o processo de desidratação das lamas.

Palavras-chave: *Moringa oleifera*; coagulante natural; desidratação de lamas; tratamento de lama de ETAR

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1. INTRODUCTION

Water is one of the most important resources for the humankind, being present in myriad ways on a daily basis. It is an important driver of health, food security, life quality, and economic development (Somlyody and Varis, 2006).

That being stated, water is a resource that must be preserved and deserves special attention as the world faces a water crisis since the beginning of this century (Corcoran et al., 2010). Continuous world population growth, together with the increase in living standards, fast industrialization, urbanization, and increase in food production, can be pointed out as some of the main causes of this crisis, plus the poor water management in past years makes the situation worse (Corcoran et al., 2010; Wada and Bierkens, 2014). In a period of 50 years, water consumption from households quintupled from 60 to 280×10^3 m^3 per year, and it is expected that by the end of this century it will increase to 600×10^3 m^3 per year (Wada and Bierkens, 2014). Following the rising living standards of the world's growing population, there has been an increase in water demand by industries, particularly the energy production and manufacturing sectors (UNESCO,2017). Thermal electricity generation, and manufacturing are projected to have an increase in water demand of 140% and 400%, respectively, between 2000 and 2050 (OECD, 2012).

Since almost 100 years ago, the world's water consumption has grown twice as fast as its population. This isn't sustainable, especially in areas where people are moving into cities quickly (UNDP, 2006; Corcoran et al., 2010). By 2050, 70 % of the world’s population is expected to be living in urban areas, resulting in increasing water scarcity problems (ONU news, 2019). In 2007, more than 2.8 billion people were living in conditions of water scarcity. This number is expected to increase to 3 billion by 2025 (A comprehensive assessment of water management in agriculture, 2007; Corcoran et al., 2010).

This increase in water consumption increases the production of wastewater (Corcoran et al., 2010; UNESCO, 2017). If poorly managed, it causes the discharge of untreated or partially treated wastewater, damaging human and environmental health. Hence, there is a necessity to increase treatment schemes for this waste (Corcoran et al., 2010; Sancho et al., 2015; UNESCO, 2017). It is estimated that industries discharge 300 to 400 million tons of untreated wastewater per year. High-income countries treat up to 70% of their wastewater, whereas low-income countries treat only 8% (Sato et al., 2013). Developing countries' discharge of untreated sewage into nature can be up to 80 to 90% (UN WATER, 2011). It is estimated that 80% of all the wastewater made in the world is dumped without being treated properly (UNESCO, 2017).

Sludge is one by-product of wastewater treatment and has been gaining attention because of its potential to cause secondary pollution in the environment (Andreoli et al., 2007). Sludge production has been growing worldwide because of the growing demand for wastewater treatment. Sludge management has a great impact in the cost of wastewater treatment operations, being able to account for 60% of total costs (Andreoli et al., 2007; Cieslik et al., 2014).

Because sludge is extremely putrescible and abundant in nutrients and organic matter, it has the potential to be used as a soil conditioner. Contaminants, heavy metals, organic contaminants, and pathogens are also present (Andreoli et al., 2007; Yu et al., 2013; UNESCO, 2017). The pollutants found in sludge are determined by the properties of the wastewater treated and the treatment method used. As a result, their incorrect release into nature poses a threat to the environment and human health (Andreoli et al., 2007; Yu et al., 2013; UNESCO, 2017). Sludge must be treated before it is disposed or reused to reduce the risk of it spoiling, the amount of pathogens present, and the amount of organic and inorganic pollutants present (Andreoli et al., 2007; Cieslik et al., 2014).

The dewatering process is one of the most significant processes in the sludge treatment process (Tat et al., 2012). It reduces volume, makes it more controllable, boosts heat capacity, reduces leachate output, and has a significant impact on sludge treatment costs overall (Andreoli et al., 2007; Tat et al., 2012).

Coagulants can be used to condition sludge prior to dewatering to make the process more efficient. Organic compounds and polymers, as well as inorganic coagulants such as alum and ferric salts, are frequently utilized (Ghebremichael et al., 2004; Andreoli et al., 2007; Tat et al., 2012). The cost of producing these compounds, on the other hand, is considerable, raising the expense of sludge treatment (Tat et al., 2012). Previous research suggest that the final treated sludge may harm the environment and human health (Kaggwa et al., 2001; Chang, L et al., 2001; Santos-Sotero et al., 2007; Tat et al., 2012).

In search of a less expensive and more environmentally friendly conditioner, researchers investigated the use of naturally occurring coagulants such as *Moringa oleifera* seeds (MO) (Ademiluyi, 1988; Okuda et al., 1999; Muyibi et al., 2001; Okuda et al., 2001; Ghebremichael and Hultman, 2004; Tat et al., 2010).

Using MO as a raw material to produce a conditioner (MOC) produced good results comparable to conventional conditioners. Even though it does not achieve the same results as them, MOC usage should be deemed suitable when considering production costs, sludge quality, and volume (Ademiluyi, 1988; Ghebremichael and Hultman, 2004; Ghebremicheal et al., 2005). Using the MO coagulant results in biodegradable sludge, which avoids public health and environmental issues (Sutherland et al., 1990; Suarez et al., 2002; Ghebremicheal et al., 2004; Ghebremicheal et al., 2005; Tat et al., 2010).

2. OBJECTIVES

The primary goal of this study is to look into alternative method to extract compounds with coagulant properties from moringa seeds using ultrasound technology and varying the concentration of salt solution. Check to see if they could be used instead of standard inorganic conditioners to dewater WWTP sludge.

3. BIBLIOGRAPHIC REVIEW

3.1 Wastewater

Wastewater is a mix of one or more of the following: water from households, water from businesses and institutions, storm water, urban runoff, and runoff from agriculture, horticulture, and aquaculture (Corcoran et al., 2010; UNESCO, 2017). Being only 1% of solids and 99% water, possibly being contaminated with different constituents, such as pathogens, organic compounds, synthetic chemicals, nutrients, organic matter and heavy metals (Corcoran et al., 2010; Drechsel, 2010; UNESCO, 2017). Its amount varies depending on the source that generates it (Raschid-Sally and Jayakody, 2008; Corcoran et al., 2010; Drechsel, 2010; UNESCO, 2017).

3.2 Domestic wastewater

Can be defined as being water used by a community (Sperling, 2007; UNESCO, 2017). The water has in its composition different substances that were introduced during its usage, as human body wastes, sullage, laundry, food preparation and cleaning of kitchenware (Mara, 2003; Sperling, 2007; UNESCO, 2017). Introducing a whole variety of chemicals, turning it hard to precisely determine the composition of domestic wastewater. The characterization is made by segregating the contaminants present in groups, suspended solids, biodegradable organic matter, nutrients, pathogenic organisms, and in recently was noticed the presence of emerging pollutants (Mara, 2003; Sperling, 2007; UNESCO, 2017).

3.3 Industrial wastewater

It can contain, to different degrees, the same pollutants mentioned in domestic wastewater (Corcoran et al., 2010; UNESCO, 2017). It is complex to generalize the characteristics of industrial wastewater since they vary from time to time and from industrial segment to industrial segment (Sperling, 2007; UNESCO, 2017). Contaminants like heavy metals, toxic compounds, and toxic organic compounds, are more likely to be present in industrial than in domestic wastewater (Ranade and Bhandari, 2014; UNESCO, 2017). Furthermore, industrial waste can have high temperatures, alkalinity, acidity, and be contaminated with biological material, all of which are harmful to the environment if not properly treated (Sperling, 2007; Corcoran et al., 2010; Ranade and Bhandari, 2014; UNESCO, 2017).

3.4 Wastewater treatment's significance

Inadequate discharge of wastewater originating from domestic activities and especially industrial production in the environment, generates physical, chemical, and biological pollution, which is harmful to both humans and the environment (Corcoran et al., 2010; Ranade and Bhandari, 2014; UNESCO, 2017).

The impact on human health depends on which group is exposed, how they are exposed, the location, and the type of wastewater (Corcoran et al., 2010; Sancho et al., 2015; UNESCO, 2017). For instance, there are countries that rely on the use of non-treated wastewater for irrigation in their agricultural activities (Buechler, 2005; Sancho et al., 2015). In those scenarios, the rural workers, their family members, consumers of the products raised using this kind of water, and people living near the area, are more vulnerable (Buechler, 2005). Direct contact with wastewater and consumption, increases the chances of worm infection and bacterial infections (Corcoran et al., 2010; Sancho et al., 2015; UNESCO, 2017). In China, exposure to a mixture of domestic and industrial effluents is associated with an increase of 36% in cases of liver enlargement (hepatomegaly) and 100% in cases of cancer and congenital malformations (Carr et al., 2004; Buechler, 2005; Sancho et al., 2015; UNESCO, 2017). Studies conducted on emerging pollutants have shown them to be potential causers of endocrine disruption, cancerous tumors, and the development of bacterial pathogen resistance, including multi-drug resistance (Poongothai et al., 2007; UNESCO, 2017).

The presence of untreated wastewater causes the degradation of coastal ecosystems and the services they provide, like food security, shoreline protection, tourism, and carbon sequestration (Nellemann et al., 2009).

It is also one of the main causes of eutrophication, as it increases the amount of nutrients, mainly phosphorus and nitrogen, in natural waterways (Corcoran et al., 2010; UNESCO, 2017). The eutrophication process decreases the amount of oxygen available in water, leveraging the reduction in biodiversity, being one of the causes of growing de-oxygenated dead zones in the seas (Corcorant et al., 2010). It is estimated that 245.000 km² of marine ecosystems are affected, bringing problems to fishing livelihoods and the food chain (Corcorant et al., 2010).

In addition to eutrophication, the presence of non-treated wastewater creates a condition that potentializes the bloom of toxic algae, like blue-green algae, that release toxins that humans and fauna are susceptible to (Boatman et al., 1999; Carr and Neary, 2008; Corcoran et al., 2010; Palaniappan, 2010; UNESCO, 2017).

Another impact that untreated wastewater may cause in the environment is the rise in temperature in surface waters, more specifically the temperature in surface waters resulting from the cooling process (Corcoran et al., 2010; Palaniappan, 2010). The receiving waterway can have its temperature raised by tens of degrees, altering its physical structure (Carr and Neary, 2008). Keeping the temperature constant as long as the discharge is being made independently of the season, which is harmful to species, as some of them depend on water temperature fluctuations that occur seasonally to reproduce and to grow (Hauer and Lamberti, 1996; Carr and Neary, 2008). Higher water temperatures diminish the dissolved oxygen concentrations. The two factors added together are enough to terminate species in that locality (Hauer and Lamberti, 1996; Carr and Neary, 2008). Table 1 compiles the industry segment, its effluent content and the quantity of contaminants found in each effluent.

Table 1. Industrial effluents - potential pollutants from different studies.

Industry	**Content of the effluent**	**Effluent contaminant**
Dairy	Dissolved sugars. Proteins. Fats. Aditives. Biochemical Oxygen Demand (BOD). Chemical Oxygen Demand (COD). Suspended solids. Nitrogen. Phosphorus;	> 200000 mg/L (for Bactofugate and cream-separators racking waters).
Pulp and Paper	Sodium hydroxide. Carbonates. Sulfides. Bisulfides. Chlorinated compounds (e.g, chlorine, chlorine dioxides, chlorinated carbons). Calcium oxides. Colored compounds. Adsorbable organic halogens (AOX). BOD. COD. Suspended solids.	Fe: 75.3 mg/L. Zn: 53.4 mg/L. Cu: 8.01 mg/L. Cd: 6.37 mg/L. Mg: 18.6 mg/L. Ni: 8.26 mg/L. Chloride: 79.1 mg/L. Chlorophenols: 781 mg/L. COD: 26974 mg/L. BOD: 8964 mg/L.
Iron and Steel	Colling water. Ammonia. Cyanide. Gasification products (e.g, benzene, naphtalene, anthracene, polycyclic aromatic hydrocarbons). Hydraulic oils. Tallows. Particulate solids. Acidic rinse. Hydrochloric and sulfuric acid.	COD: 4620 mg/L. BOD at 27°C: 2123 mg/L. Chloride as Cl-: 600 mg/L. Sulphide as SO-4: 320 mg/L. Oil and grease: 6 mg/L.
Tanneries	Organics. Heavy metals. Ammoniacal nitrogen. Acids. Salts. Sulfides. Suspended solids. Dyes. Fats. Oils.	Cr: 6770 mg/L. Cd: 2.214 mg/L. Pb: 32 mg/L. Cu: 2.58 mg/L. Zn: 8.69 mg/L. Fe: 109 mg/L. Mn: 17.2 mg/L. BOD: 462 mg/L.

Table 1. Industrial effluents - potential pollutants from different studies (cont.).

Industry	Content of the effluent	Effluent contaminant
Mining Industry	Heavy metals. Rock particles. Particulate hematite. Slime of fine particles. Hydroxide. Carbonates. Cyanide. Sulfur. Oxides (e.g, cadmium oxide, calcium oxide, zinc oxide, sodium oxide, barium oxide, etc.). Acidic, or alkaline drainage.	As: 7.2 mg/L. Fe: 3.8 mg/L. Cu: 7.8 mg/L. Pb: 0.2 mg/L. Zn: 0.1 mg/L. Cyanide: 66.3 mg/L. COD: 1240 mg/L. BOD5: 4.3 mg/L.
Energy	Conataminated groudwater as drilling for gas and oil. Discharge of heated water. Oil. Acid. soda sludge. hydrogen sulfide. Lead sludge. Hydrocarbons. Spent filter clay. Etylene Glycol. 1,4-dioxane.	Al: 83 mg/L. As: 151 mg/L. Ba: 1740 mg/L. B: 56 mg/L. Cd: 1.21 mg/L. Ca: 51300 mg/L. Cr: 0.03 mg/L. Cu: 5 mg/L. COD: 120000 mg/L (natural gas production). COD: 1220 mg/L (oilfield produced water). Bromide: 1149 mg/L. Chloride: 190000 mg/L (natural gas production). Total oil: 565 mg/L. Chloride: 3990 mg/L. Sulfite: 10 mg/L. Ammoniacal nitrogen: 300 mg/L. Phenols: 23 mg/L (oilfeld produced water).

Table 1. Industrial effluents - potential pollutants from different studies (cont.).

Industry	**Content of the effluent**	**Effluent contaminant**
Food	BOD. Suspended solids. Particulate matter. Dissolved organics. Surfactants. Antibiotics. Hormones. Pesticides. Insecticides. Plant organic material. Salt. Flavorings. Coloring material. Acids or alkali. Fat.	BOD: 523.5 mg/L. COD: 11900 mg/L. Total suspended solids (TSS): 265 mg/L. Total dissolved solids (TDS): 399.4 mg/L (ice cream production). Cr: 0.04 mg/L. Cu: 0.25 mg/L. Fe: 14.2 mg/L. Zn: 0.09 mg/L. Cd: 0.87 mg/L. Mn: 0.17 mg/L. Pb: 0.65 mg/L. K: 9.79 mg/L.

Source: Adapted from Andreoli et al., 2007; Palaniappan et al., 2010; Acheampong et al., 2013; Krishnaveni et al., 2013; Qasim and Mane, 2013; Ranade and Bhandari, 2014; Kabir et alk., 2017; UNESCO, 2017; Kasmi Mariam., 2018; Ahsan et al., 2019; Sharma et al., 2020.

3.5 Wastewater treatment

The treatment of wastewater should not be the same in all countries (Mara, 2003). The nature and extent of it are determined by the country's level of industrialization and the purpose of the effluent after treatment. Treatment differs depending on the country's stage of industrial development. For example, in developed countries, efficiency, dependability, and land needs are prioritized, but in poor countries, reliability, sludge production, operational and construction costs, sustainability, and treatment simplicity are top priorities (Mara, 2003)

There are two types of wastewater treatment systems: centralized systems and decentralized systems (Sancho et al., 2015; UNESCO, 2017). The last option reduces investment costs, but it is not always capable of providing treated effluent of the same quality as a centralized system; however, this does not mean that centralized systems are always the best option (Cocoran et al., 2010; UNESCO, 2017).

There are three stages of treatment: preliminary, primary, secondary, and tertiary treatment (Figure 1). Each one is responsible for removing different pollutants in wastewater (Sperling, 2007).

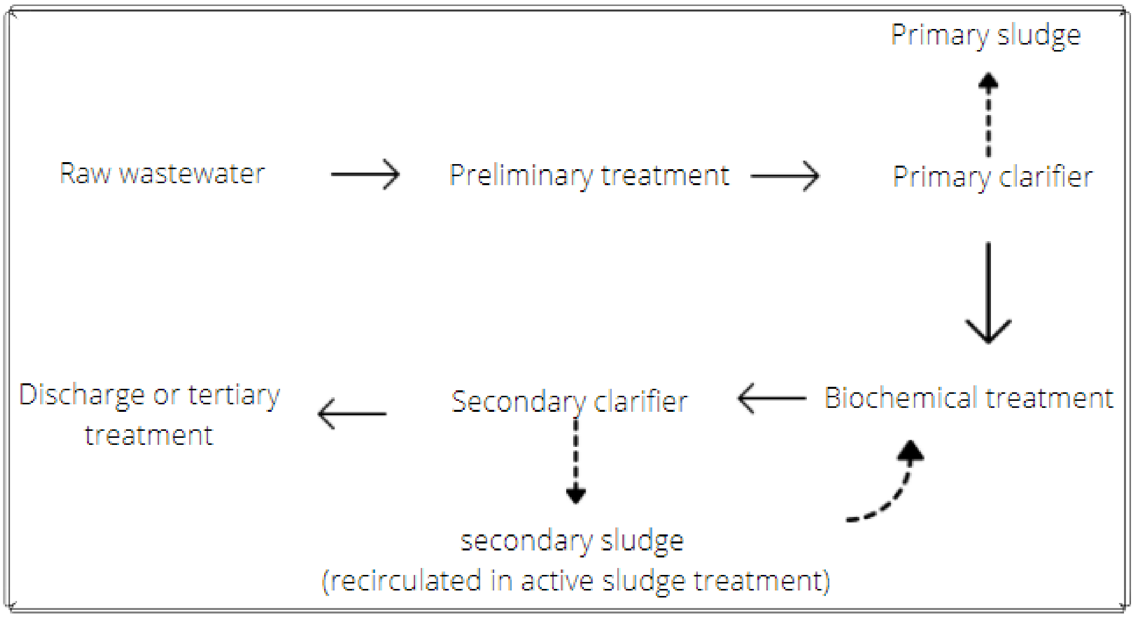

Figure 1. Simplified wastewater treatment scheme.

3.5.1 Preliminary treatment

Physical removal of suspended solids of larger dimensions, grit removal with racks or screens and sedimentation of inorganic particles in grit chambers (Mara, 2003; Sperling, 2007; World Water Assessment Programme, 2009).

3.5.2 Primary treatment

It aims to remove partially suspended solids that passed from the preliminary treatment by sedimentation, being mostly organic matter. It also aims to remove floating material like grease and oil. It is able to remove around 20–30% of the BOD and reduce 50–60% of suspended solids, also forming in the bottom of the primary sludge due to the sedimentation of the suspended solids (Sperling, 2007; Allaoui et al., 2015).

3.5.3 Secondary treatment

The main objective is to remove the organic matter that is dissolved or in suspension by speeding up the natural way that microorganisms break down organic matter (Sperling, 2007). At this stage, CO2, water, and energy are released, and biomass is produced, which will be the cause of the formation of secondary sludge (Sperling, 2007). Under controlled conditions, the secondary treatment can decompose pollutants, removing about 75% of the ammonium and 85% of the suspended solids (Sperling, 2007; World Water Assessment Programme, 2009; Allaoui et al., 2015). Figure 2 shows how a microorganism's metabolism works in a simplified way.

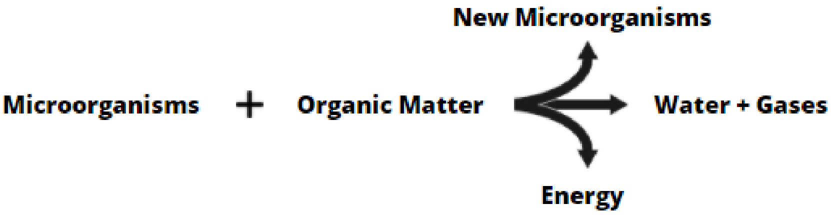

Figure 2. Simplified scheme of microorganism’s metabolism.

3.5.4 Tertiary treatment

It's a more advanced treatment that focuses on disinfecting, removing specific contaminants, or more effectively removing nutrients like phosphorus and nitrogen (Sperling, 2007; Allaoui et al., 2015). It removes almost 99 percent of contaminants, but it is more expensive than other treatments because it requires a high level of technical knowledge, skilled operators, and a consistent supply of energy, chemicals, and equipment (Sperling, 2007; World Water Assessment Programme, 2009; Ranade and Bhandari, 2014). Using UV radiation in combination with other disinfectants to inactivate more types of microorganisms is an example of tertiary treatment (Sperling, 2007; Allaoui et al., 2015).

3.6 Sludge production and characteristics

Sludge is a by-product of wastewater treatment that can be divided into three types: primary, secondary, and chemical sludge. If there is a physical-chemical stage in the treatment process, the last one is produced (Andreoli et al., 2007). Around 95% of the sludge is water, and it also has contaminants. Their quantity varies according to the quality of the treated wastewater as well as the treatment method employed (Sperling, 2007; Andreoli et al., 2007; UNESCO, 2017).

3.6.1 Primary sludge

Their production is determined by the effectiveness of the primary treatment (Andreoli et al., 2007; Sperling, 2007). It is primarily composed of settleable solids from raw wastewater, with a high ratio of suspended and dissolved organic matter, and it is likely to have a strong odor, particularly if it is exposed to high temperatures for an extended period (Andreoli et al., 2007; Sperling, 2007; Tyagi and Lo, 2013).

3.6.2 Secondary Sludge

Sludge is generated in all treatment processes involving biological processes, being mainly composed of biomass resulting from the microorganism’s metabolism, acting to remove the organic matter in the wastewater (Andreoli et al., 2007; Sperling, 2007).It is classified as organic matter (carbon, oxygen, nitrogen, hydrogen, phosphorus, and sulfur) and inorganic matter (minerals such as quartz, calcite, and microline, which are primarily composed of metals and heavy metals) (Andreoli et al., 2007; Sperling, 2007; Tyagi and Lo, 2013). To avoid bad odors caused by anaerobic decomposition of organic matter in the sludge, secondary sludge must be digested (Andreoli et al., 2007; Sperling, 2007; Tyagi and Lo, 2013).

3.6.3 Chemical sludge

Both biological and chemical sludge can be produced at the same plant. This sludge is the result of chemical precipitation with metallic salts or lime added (Chang, G et al., 2001; Andreoli et al., 2007). The sludge has fewer problems with odor production because the decomposition rate of chemical sludge is slower than that of primary sludge, and the composition of the sludge is determined by the wastewater composition and chemicals and treatments used (Chang, G et al., 2001; Sperling, 2007).

3.6.4 Contaminants in the sludge

The amount of contaminants in the sludge is directly related to the raw characteristics of the wastewater, and they are classified into three major groups: heavy metals, organic compounds, and pathogens (Andreoli et al., 2007). The first two are primarily derived from industrial processes, while the third is primarily derived from human sources, directly reflecting population health, or from animal sources (Andreoli et al., 2007; Sperling, 2007). In addition to these common contaminants, detection technologies have advanced in recent years. As a result, emerging organic contaminants such as pharmaceuticals, drugs, synthetic phenolic compounds, perfluorinated compounds, benzotriazoles, and siloxanes were noticed in the sludge composition. (Andreoli et al., 2007; Thomaidi et al., 2016).

There are other factors that are related to how the contaminants are distributed in the sludge (Andreoli et al., 2007). The concentration and toxicity of metals vary depending on external factors such as pH and the presence of other compounds. For example, the presence of cyanide increases the toxic effect of copper (Andreoli et al., 2007). The number of pathogens depends on factors such as the socio-economic level of the population, sanitation conditions, geographic region, presence of agro-industries, and type of sludge treatment (Andreoli et al., 2007; Sperling., 2007). Table 2 shows pollutants that were found in sludge.

There are limits to the concentration for most of these contaminants to prevent damages in human and environment health. In Portugal there is the decree-law number 276/2009, establishing limits for contaminants present in sludge to be reused as soil conditioners (Table 3).

Table 2. Pollutants in a WWTP sludge and concentrations.

Heavy metals	Ag. As. Au. B. Ba. Cd. Cu. Co. Cr. Cs. Hg. La. Mn. Mo. Ni. Pb. Sb. Se. Sn. U. V. W. Zn. Zr.	Pb: 80.4 mg/kg DM. Cd:10.8 mg/kg DM. Cu: 255 mg/kg DM. Hg: 2.35 mg/kg DM. Ni: 42 mg/kg DM. Cr:144 mg/kg DM. Zn: 162 mg/kg DM.
Organic contaminants	Cyanide. Phenol. Tetrachloroetylene. Chloroform. bis-2-ethyl-hexyl phthalate. 2,4-dimethyl phenol. Naphthalene. butylbenzylphtalate; acrolein. xylene; cresol. Acetophenone. metyl-sobutyl-acetone. Diphenylamine. Anilin. Ethyl acetate.	Phthalates: 125 mg/kg DM. Polychlorinated alkanes (PCAs): 36 mg/kg DM. Polychlorinated biphenyls (PCBs): 0.02 mg/kg DM. Polychlorinated dioxins/furans (PCDD/Fs): 0.82x10^{-3} mg/kg DM. (Olofsson et al., 2012). (PAHs): 33 mg/kg DM (Cai et al., 2007).

Table 2. Pollutants in a WWTP sludge and concentrations (cont.).

Emerging organic contaminants	Caffeine. Ofloxacin. Tetracycline. Nonylphenol. nonylphenol dietoxylate. Nonylphenol. Monoethoxylate. Triclosan. 2-hydroxybenzothiazole. Decamethylcyclopentasiloxane. Dodecamethylcyclohexasiloxane. Dodecamethylpentasiloxane. Tetradecamethylhexasiloxane.	Triclosan, fluoroquinolones (FQs): 1.45 mg/kg DM. Siloxanes: 13.5 mg/kg DM.
Pathogens	Helminths. Protozoas. Fungi. Viruses. Bacteria.	Fungi: 950 CFU/g DM. Total coliforms: 66x10^3 CFU/g DM. fecal coliforms: 1200 CFU/g DM. *Shigella spp.* CFU/g DM. *Salmonella spp.* CFU/g DM.

Source: Adapted from Alloway and Jackson, 1991; Franco-Hernández et al., 2003; Andreoli et al., 2007; Governo de Portugal, 2009; Olofsson et al., 2012; Andreoli et al., 2015; Thomaidi et al., 2016.

Table 3. Portuguese limits for contaminants in sludge destined to be used as soil conditioner.

Heavy metals	Cd: 20 mg/kg DM. Cu: 1000 mg/kg DM. Ni: 300 mg/kg DM. Pb: 750 mg/kg DM. Zn: 2500 mg/kg DM. Hg: 16 mg/kg DM. Cr: 1000 mg/kg DM.
Organic contaminants	Linear alkyl benzenesulfonates (LAS): 20mg/kg DM. nonylphenols and ethoxylated nonylphenols (NPs and NPEs): 1000 mg/kg DM. Polycyclic aromatic hydrocarbon (PAHs): 300 mg/kg DM. Polychlorinated biphenyl (PCBs): 750 mg/kg DM. Polychlorinated dibenzodioxins (PCDD): 2500 mg/kg DM. Polychlorinated dibenzofurans (PCDFs): 16 mg/kg DM.
Emerging organic contaminants	Not regulamented.
Pathogens	*Escherichia coli:* <1000 cels/g fresh matter. *Salmonella spp*: absent in 50 g of the original material.

Source: Decreto-Lei n° 276/2009 de 2 de outubro.

3.7 Impacts of sludge treatments and disposal

The degree of the impact depends on the method chosen, the efficiency of it, as well as the amount of sludge, characteristics of the sludge being disposed of, duration, frequency, and extent of the disposal (Andreoli et al., 2007; Manfredi and Pant, 2013). Table 4 shows the general impacts associated with sludge disposal, which are related to climate change, ecotoxicity, and human toxicity (Andreoli et al., 2007; Manfredi and Pant, 2013).

Table 4. Disposal, treatment, and impacts of sludge in the environment.

Land applications	Risk of soil contamination by accumulating toxic elements in soil. Water contamination due to leaching and storm runoff flows. Attraction of vectors.
Land farming	Contamination of soil and water
Agricultural land applications	Contamination with toxic elements, and pathogens
landfill	Leaching of toxic liquids. Contamination of surface or groundwater; Greenhouse gas production.
Ocean disposal	Damage to benthic organisms. Introduction of bioaccumlating toxic compounds. Increase of BOD.
Incineration	Air pollution. Production of toxic ashes

Source: Adapted from Andreoli et al., 2007; Tyagi and Lo, 2013.

Before deciding on any of the strategies for sludge disposal, it is critical to consider strategies for avoiding the production of residue. One good example would be the thinking line proposed from article 4 from directive 2008/98/EC of the European Parliament and of the Council:

> Article 4 on the “waste hierarchy” states: “the following the following waste hierarchy shall apply as a priority order in waste prevention and management legislation and policy: (a) prevention; (b) preparing for re-use; (c) recycling; (d) other recovery, e.g. energy recovery; and (e) disposal.” “When applying the waste hierarchy […], Member States shall take measures to encourage the options that deliver the best overall environmental outcome. This may require specific waste streams departing from the hierarchy where this is justified by life-cycle thinking on the overall impacts of the generation and management of such waste.”

Following the waste hierarchy usually leads in taking best solution for the environment, but in certain scenarios there may be the need to deviate waste hierarchy to select the best option. Figure 3 shows a practical guidance to choose best safe option for the environment (Manfredi and Pant, 2013).

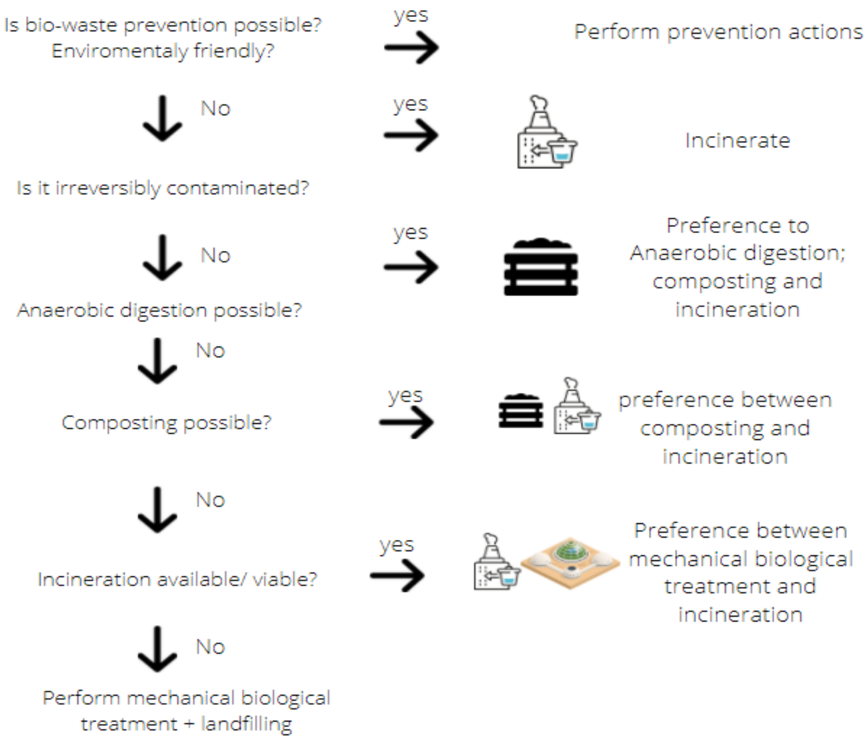

Figure 3. Flux gram showing practical guidance to safely treat sludge.

3.8 Dewatering and conditioning

Dewatering is one step prior to treat or dispose the sludge (Andreoli et al., 2007; Tat et al., 2010). Being a critical step in reducing the total volume of sludge, treatment costs, improving its handling, and lowering leachate production (Andreoli et al., 2007; Tat et al., 2010).

To aid the dewatering process, sludge undergoes a conditioning process, to increase particles size, change their distribution, surface charges and sludge particle interactions through coagulation process (Muyibi et al., 2001; Andreoli et al., 2007). Conditioning can be accomplished through the use of inorganic chemicals, organic chemicals, and thermal treatment (Chang, G et al., 2001; Muyibi et al., 2001; Andreoli et al., 2007; Tat et al., 2010).

3.9 *Moringa oleifera*

Usually used as an ornamental tree, is part of the family Moringacea, being normally found in the whole tropical belt area (Jahn, 1988; Vijay et al., 2012). Reaching

10-12 meters in height, being able to grow in regions with poor soil, and having multiple utilizations (Jahn, 1988; Vijay et al., 2012). The tree is a food source due to its nutritional values. It presents compounds with antimicrobial and fungicidal activity, is a source of valuable oil with high-oleic content, ben oil, and its seeds can be used to clarify water, therefore showing potential to be used as coagulants in water treatment (Jahn, 1988; Vijay et al., 2012; Haldar and Kosankar, 2017). Figure 4 shows the seeds of Moringa oleifera.

Figure 4. Moringa oleifera seeds used in this study.

3.10 Coagulant potential as a sludge conditioner

The active compound responsible for the MOC's coagulation capacity was identified to be a group of proteins with an unknown molecular mass. Masses have been shown to be lower than 6.5 kDa in certain studies and more than 12 kDa in others (Ghebremicheal et al., 2005; Tat et al., 2012). The proteins have similar physical properties, and studies have shown that using salt solutions to extract the active component is more effective (Okuda et al., 1999; Okuda et al., 2001; Ghebremicheal et al., 2005).

Water treatment coagulants obtained from MO produce a more biodegradable sludge, which can then be used as a biosolid in agriculture (Suarez et al., 2002; Ghebremicheal et al., 2004). MO coagulant provided a more compact sludge with high quality in solid cake concentrations and increased cake solids filterability in sludge dewatering studies (Ademiluyi, 1988). It is less harmful to the environment, resulting in a safer sludge to manage because it does not modify the medium's pH and has antibacterial capabilities that may inactivate various water-borne organisms, making the sludge safer to handle (Sutherland, 1990; Suarez et al., 2002; Ghebremicheal et al., 2004; Ghebremicheal et al., 2005).

Ademiluyi (1988), showed that MO seed powder as a coagulant produces comparable results to improving the filterability of the sludge with ferric chloride, and it was best in producing a cleaner filtrate. In a different study using the same procedure but removing the oil from the seeds, it has shown better results when using the coagulant without defatting the seeds first (Ademiluyi and Eze, 1990). Although using Moringa seed powder has some drawbacks, it significantly increases the amount of solids in the conditioned sludge (Tat et al., 2012).

To get around that problem, several studies have been conducted using the extract from active compounds responsible for coagulation in MO seeds. Studies by Okuda et al. (1999) and Okuda et al. (2000) showed that extraction of active compounds is better when using salt solutions than using distilled water.

According to Ghebremicheal et al. (2004), MO capability for conditioning the sludge using a NaCl salt solution extraction showed similar results in solids concentration compared with alum and synthetic polyelectrolyte coagulants (4.5% for the seed coagulant, 4.76% for alum, 5.95% and 6.83% for the polyelectrolytes). In the same study, the dewatering rates of the sludge were improved by the same amount between the natural conditioner and alum conditioner. In contrast to the study conducted by Ademiluyi (1988), In contrast to Ademiluyi (1988), Ghebremicheal et al. (2005) found that removing the oil did not boost the seed's capacity as a coagulant, and that it performed comparably to the unprocessed seed. Muyibi et al. (2001) observed the same tendency, but his results suggested that employing oil-extracted MO seeds produced more porous sludge flocs, increasing the sludge's filterability, and thereby trapping less water and moisture in the sludge.

3.11 Ultrasound extraction technology

Ultrasound extraction technology demands less energy, duration, and solvent to make extraction (Bindes et al., 2019). It works best because the soundwaves made by the ultrasound equipment move through the medium in cycles, expanding and contracting.

This causes changes in pressure and causes bubbles to form, which eventually pop and release high temperatures and pressures (Horzic et al., 2012). These effects can break through cell walls, letting the solvent get inside more, releasing the content inside, and improving mass transfer, which makes for better extraction (Wang and Weller, 2006; Horzic et al., 2012; Jovanoíc et al., 2017; Bindes et al., 2019).

4. MATERIALS AND METHODS

4.1 Sludge sample collection

Sludge samples were collected from the wastewater treatment plant in Bragança, Portugal (Figure 5, WWTP flowsheet), at the outlet of the sludge tank (primary and secondary sludges). The sludge was then stored in plastic gallons at room temperature until usage.

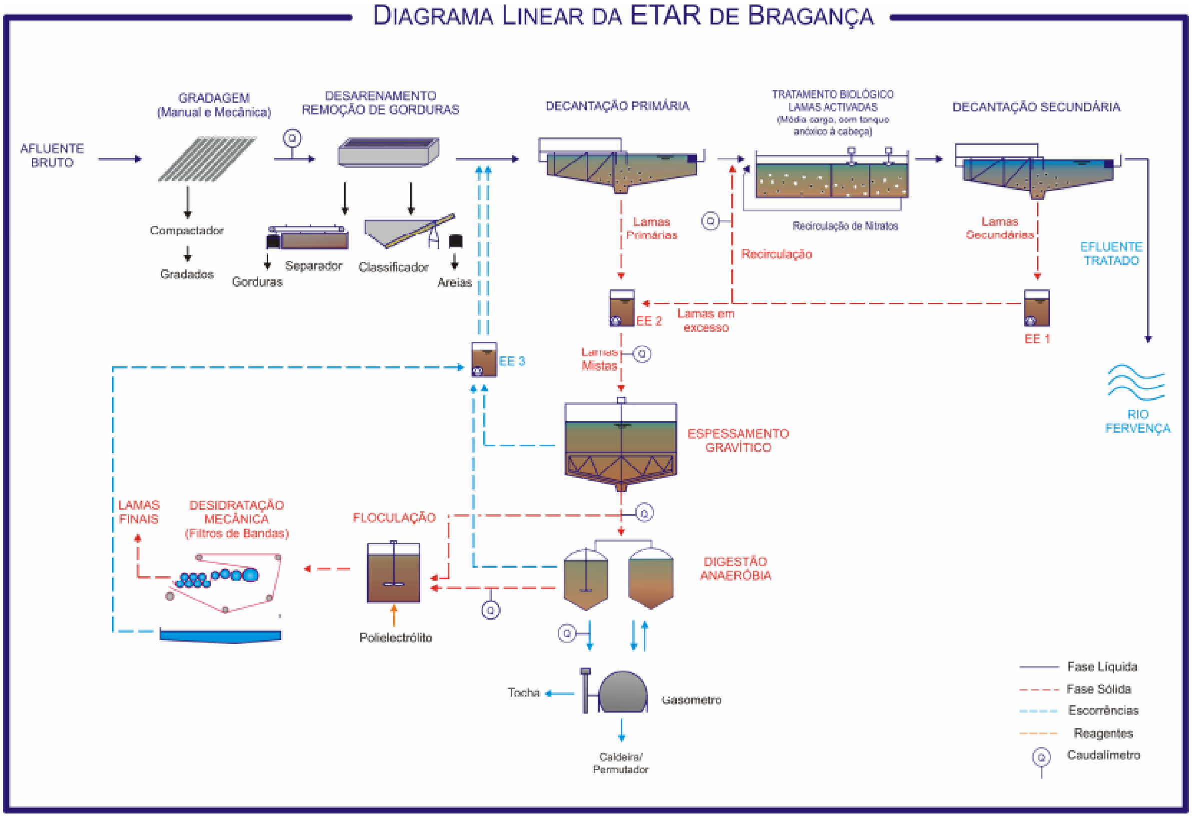

Figure 5. Bragança wastewater treatment plant flowsheet.

4.2 Moringa seeds collection

MO seeds were obtained from Universidade Tecnológica Federal do Paraná (UTFPR), Londrina, Brazil. The seeds were collected, sieved to remove impurities, and transported to the Instituto Politécnico de Bragança (IPB) laboratory, where they were kept at room temperature.

4.3 Preparation of the MO seeds

MO seeds were shelled and making sure to choose good seeds. Then, using a commercial blender, the seeds were pulverized into a fine powder, and afterwards dried at 45°C for 24 hours, according to Tat et al. (2010). The oil content was removed for 6 hours using a Soxhlet apparatus and ethanol, and then dried at 45°C for 24 hours (Ghebremicheal et al., 2005).

4.4 Preparation of MO conditioner (MOC)

The powdered defatted seed was dissolved in NaCl solutions (0.5 M, 1.0 M, 1.5 M) to prepare the MOC. The power was mixed with the salt solution by dissolving 10 g of powder in 100 mL of salt solution (10% m/v). For the extraction, a magnetic stirrer was used to keep the mixture constantly agitated for 1 hour. To remove the undissolved solids from the filtrate, a Büchner funnel with qualitative filter paper, 185 mm from Filter-Lab was used. The MOC was then kept refrigerated at 4°C until it was used.

4.5 Preparing MO conditioner with ultrasound assist (MOC-U)

All stock solutions were prepared to achieve a concentration of 10%, just as in the previous preparation, but only to salt solution that exhibited better results.

The equipment used was a Q500 from QSONICA. The extraction method was based on a study conducted by Adjeroud-Abdellatif et al. (2020). Using a sonicator probe, set the equipment to an amplitude of 60% for a total duration of 10 minutes of extraction. The extraction was made in cycles of 30 seconds on and 10 seconds off to prevent overheating equipment. It used 150 mL of the 1.0 M NaCl solution with the dried material. While performing the extraction, the mixture recipient was put in an ice bath to avoid overheating the mixture. After extraction, the mixture was filtered using the same methodology as in topic 4.4. The filter, MOC-U, was then refrigerated until it was time to use it.

4.6 Sludge conditioning

The coagulant dosages used were 5 mL of coagulant per L of sludge, 10 mL/L, 25 mL/L, and 50 mL/L. It was made sure that the coagulant was at room temperature before it was added. To condition the sludge, it was mixed in the jar test apparatus, establishing a rapid mix of 150 rpm (3 min) followed by a slow mix of 20 rpm (12 min). The conditioning was based on the Standard Methods for the Examination of Water and Wastewater, 2017.

4.7 Drying column experiment

Acrylic columns of 3 cm diameter and 60 cm in height were used to dewater the sludge. Non-conditioned sludge was used as a control. In all experiments, the columns were filled with 10 cm of inert material with a granulometry of 0.85–1.18 mm (filtering medium). The heights of the sludge varied from 10 cm (200 mL), 20 cm (400 mL), and 40 cm (600 mL).

For each column, the amount of filtrate was measured with a graduated cylinder at 15, 30, 60, 300, and 1440 minutes. After each experiment, the filtrate was stored in the refrigerator at 4°C until it was time to analyze it. All experiments were done in duplicate. Figure 6 shows the apparatus used to dewater the conditioned sludge, and Figure 7 shows the end of the dewatering experiments for different sludge height experiments.

Figure 6. Experimental apparatus, drying columns.

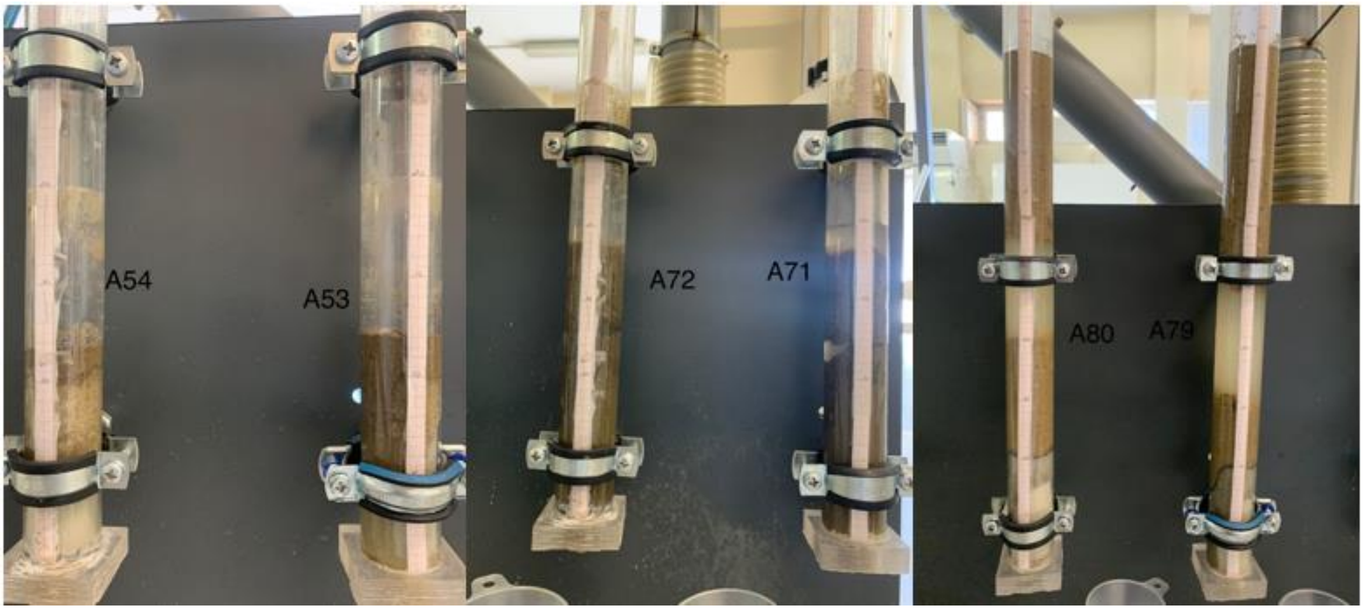

Figure 7. Dewatering experiments for different sludge heights after de dewatering process.

4.8 Analytical methods

4.8.1 Total solids (%TS)

TS content in the sludge was determined following the methodology in Standard Methods for the Examination of Water and Wastewater (2017), procedure 2540 B. The determination was made for the raw sludge, non-conditioned sludge after drying and all conditioned sludge after drying experiments, according to equation 1.

$$\%TS = (m_2 - m_0) \div (m_1 - m_2) \qquad \text{(Eq. 1)}$$

Where:
m_0 = mass of evaporating dish, dried at 105°C.
m_1 = mass of (evaporating dish dried at 105°C + sludge sample).
m_2 = mass of (evaporating dish+ sludge sample), dried at 105°C.

4.8.2 Chemical Oxygen Demand (COD)

COD tests were made by following the procedure used in Standard Methods for the Examination of Water and Wastewater (2017), procedure 5220 D, closed reflux, colorimetric method. Analyzing with a spectrophotometer at a 600 nm, COD tests were made of the raw sludge and filtrate. Calibration curve was prepared accordingly (Figure 8).

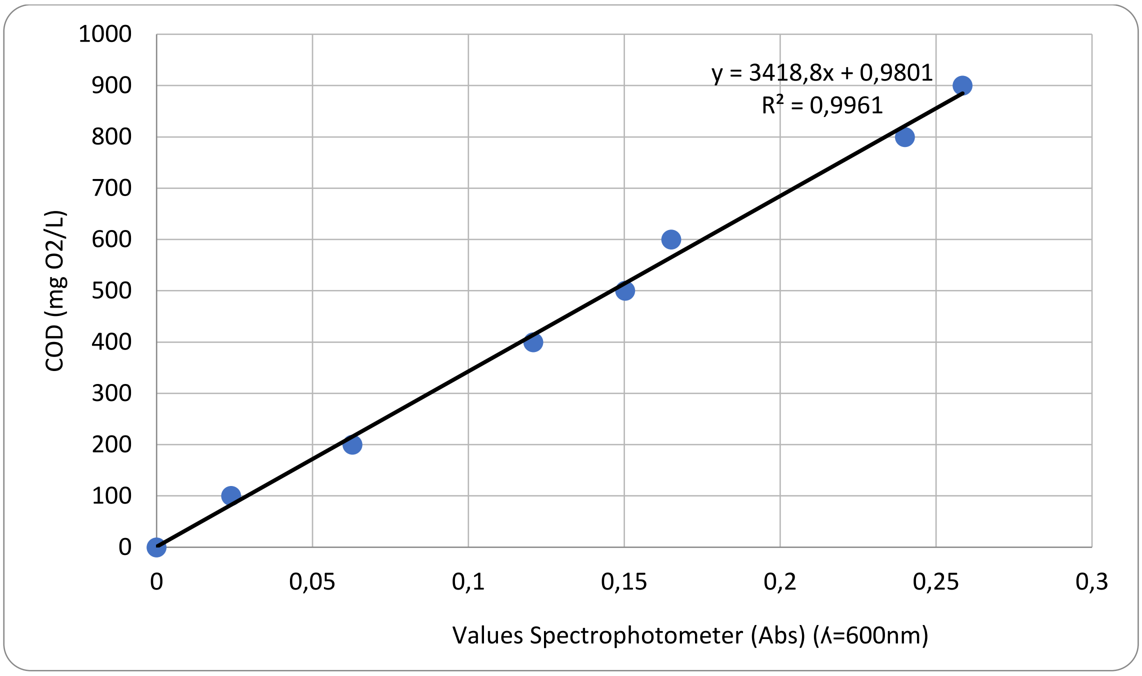

Figure 8. COD calibration curve.

4.8.3 Turbidity and pH

Turbidity was measure using a turbidimeter, MERCK Turbiquant 3000IR, and was only taken the values from the filtrate turbidity.

The pH values were taken from the raw sludge samples as a form of characterization of the sludge. pH values were also taken from the filtrate from the dewatering experiments. All measures were made using a pH meter from HANNA Instruments, model EDGE.

5. Results and Discussion

5.1 Coagulant extraction made using different NaCl solutions

The best salt concentration was determined by comparing results from dewatering experiments (Table 5). Sludge height was kept at 10 cm, varying the salt concentration. The coagulant dosages varied as described in section 4.6.

The salt solution showed a positive effect in extraction until a salt solution of 1.0 M. This effect is explained by the salting-in effect. The addition of salt into the solution increases the solubility of the protein responsible for the coagulation process. Higher salt concentrations than 1.0 M have negative effects in extracting the active coagulant agent. This can be explained with the salting-out effect, which diminishes the solubilization of the protein, due to the addition of salt beyond optimal (Abidin et al., 2013).

Table 5. Sludge dewatered using *Moringa oleifera* coagulant, with sludge height fixed at 10 cm.

Coagulant dosage (mL/L)	TS initial (%)	TS final (%)	Increase (%)	COD initial sludge (g/L)	COD final fitrate (g/L)	Total filtrate (mL)	Turbidity (NTU)
Salt solution, 0.5 M							
Control	2.19	4.49	2.3	10.7	1.94	56	371
5		3.17	0.98		1.22	60	385
10		4.46	2.3		1.46	71	350
25		2.87	0.68		2	62	166
50		4.64	2.5		2.53	80	161
Salt solution, 1.0 M							
Control	1.52	6.85	5.33	10.2	0.56	78	50
5		8.97	7.45		0.65	80	41
10		10.7	9.2		1.22	80	49
25		13	11.5		1.84	83	58
50		18.6	17.1		3.4	87	120
Salt solution, 1.5 M							
Control	1.52	6.85	5.33	10.2	0.56	78	50
5		4.44	2.92		1	70	72
10		10.5	8.94		1.48	75	64
25		12	10.4		2.09	79	101
50		15.3	13.7		2.58	78	120

The 1.0 M was optimal for extracting the coagulant from the *Moringa oleifera* seeds and dehydrate the sludge, showing, in all different coagulant doses, better liquid removal. Also, this concentration has produced a higher quality cake solid. Starting at initial TS of 1.52% and maximum TS result of 18.6%, with dosage of 50 mL/L. This salt concentration produced a filtrate with lower turbidity. This salt concentration was found to be optimal in other studies made by Okuda et al. (1999), and Bouchareb et al. (2021).

5.2 Coagulant extracted using 1.0 M NaCl solution (MOC)

5.2.1 Total solids (%TS)

Table 6, and Figure 9, shows the TS results for the different experiments. In the 10 cm sludge height experiments as there is an increase in coagulant dosage, there is also an increase in the TS values. Similar behavior was found in studies of Ghebremicheal et al., (2004). The 10 cm experiments showed a opposite behavior than the 20 and 40 cm experiments. The highest TS value was of 18.6% attained at coagulant dosage of 50 mL/L, starting in at an initial TS of 1.52%.

The 20 and 40 cm experiments produced similar results, having the maximum TS result when the coagulant dosage was at its lowest. The maximum result for 20 and 40 cm sludge height experiments were of 2.65%, and 2.50%, with coagulant dose of 5 mL/L. It is possible to say that the initial sludge volume conditions negatively the TS results.

Table 6. TS using MOC coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	TS initial (%)	TS final (%)	Increase (%)
Sludge height, 10 cm (V = 200 mL)			
Control	1.52	6.85	5.33
5		8.97	7.45
10		10.7	9.2
25		13	11.5
50		18.6	17.1
Sludge height, 20 cm (V = 400 mL)			
Control	1.52	2.22	0.7
5		2.65	1.13
10		1.8	0.28
25		1.74	0.22
50		1.58	0.06
Sludge height, 40 cm (V = 600 mL)			
Control	1.98	3.5	1.52
5		2.5	0.52
10		2.18	0.2
25		2.2	0.2
50		2.07	0.1

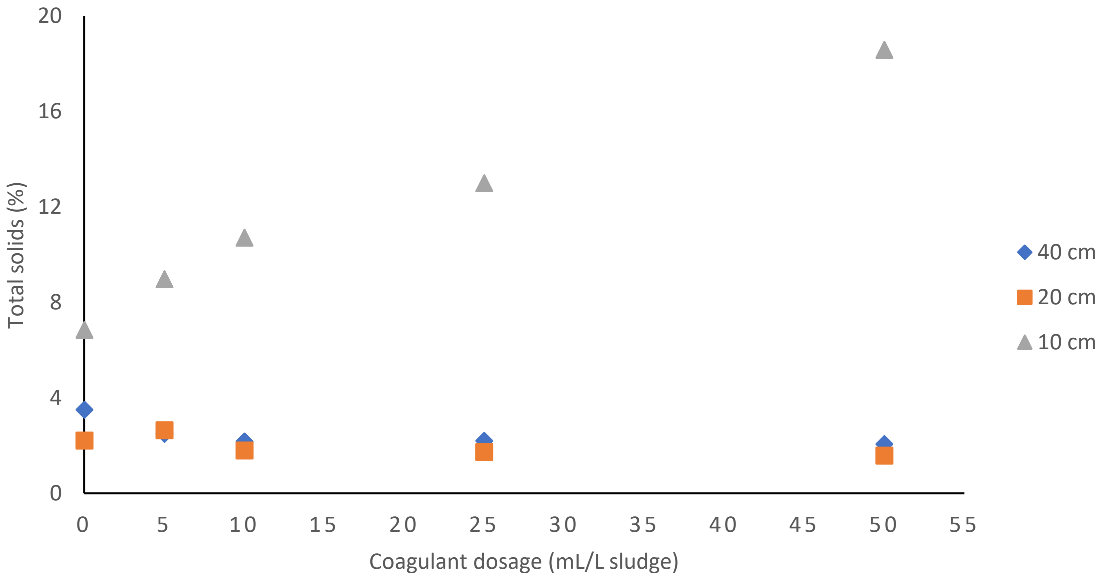

Figure 9. Sludge TS for different sludge height and coagulant dosage, using MOC coagulant (NaCl solution 1.0 M).

5.2.2 Total filtrate drained and drainage rate

Table 7 shows the total filtrate and drainage rate results obtained for each experiment using the MOC coagulant. In the 10 cm sludge experiments, as there is an increment in the coagulant dose, there is an increase in total filtrate drained. Following a similar behavior as the TS results. The maximum filtrate removal was of 87 mL, achieved with maximum coagulant dose of 50 mL/L.

The 20 and 40 cm experiments presented opposite behavior as the 10 cm experiments. The maximum filtrate removal from the 20 and 40 cm sludge experiments were of 114 mL and 91 mL with coagulant dosages of 5 mL/L and 0 mL/L, respectively.

The drainage rate for the 10 and 20 cm sludge experiments was improved, having all the coagulant dosages presenting higher drainage rates than the control experiment. The best drainage rate attained for 10 and 20 cm sludge experiments were of 0.06 mL/min, and 0.08 mL/min, obtained at coagulant doses of 50 mL/L and 5 mL/L, respectively.

The 40 cm sludge height experiments did not show any final drainage rates better than the control experiment, and the best result was attained at minimum coagulant dose of 5 mL/L, with a drainage rate of 0.05 mL/min.

The 10 and 20 cm sludge height experiments improved the dewatering capacities of the sludge, as shown by Ghebremichael et al. (2004). While for the 40 cm sludge height experiments did not improve the dewatering capabilities of the sludge. Using the MOC coagulant improved the drainage rate of the experiments by 1.11 times for the sludge heigh of 10 cm, and by 1.34 times for 20 cm sludge height. The improvement in drainage rates were not as performant as the improvement attained by Ghebremichael et al. (2004). The initial sludge volume conditions negatively the thickening process, since as the sludge height increases, it diminishes the total filtrate drained.

Table 7. Drainage rate and filtrate removal using the MOC coagulant, NaCl solution 1.0 M.

Coagulant dosage (mL/L)	Drainage rate for each pre-determined period (mL/min)					Total filtrate (mL)
	15 min	30 min	60 min	300 min	1440 min	
Sludge height, 10 cm (V = 200 mL)						
Control	0.73	0.55	0.38	0.16	0.05	78
5	1.1	0.65	0.49	0.19	0.06	80
10	1.03	0.72	0.47	0.17	0.06	80
25	1.2	0.8	0.52	0.19	0.06	83
50	1.63	1.02	0.65	0.26	0.06	87
Sludge height, 20 cm (V = 400 mL)						
Control	2.07	1.18	0.68	0.2	0.06	85
5	2	1.15	0.68	0.21	0.08	114
10	1.9	1.1	0.64	0.21	0.07	99
25	1.93	1.15	0.68	0.21	0.07	102
50	1.67	1.02	0.63	0.2	0.07	97
Sludge height, 40 cm (V = 600 mL)						
Control	0.93	0.65	0.41	0.13	0.06	91
5	1	0.68	0.43	0.14	0.05	72
10	0.73	0.5	0.4	0.14	0.05	65
25	0.8	0.55	0.36	0.14	0.05	66
50	0.57	0.4	0.28	0.01	0.03	49

5.2.3 Chemical Oxygen Demand (COD)

For all experiments, it is noticeable that higher coagulant dosages result in smaller COD reduction values (Table 8). *Moringa oleifera* as a coagulant tends to release organic substances in to the medium, increasing the values of COD. The same behavior was found in other studies regarding the usage of natural coagulants (Okuda et al., 2001; Sánchez-Martín et al., 2010; Gidde et al., 2012).

The maximum filtrate COD values for the 10, 20 and 40 cm sludge height experiments were of 3.4 g/L, 4.67 g/L, 6.25 mg/L, all with maximum coagulant dosage of 50 mL/L. The starting filtrate COD values were of 0.56 g/L, 1.62 g/L, 4.87 g/L, obtained in the control experiments.

The smaller COD reduction presented in experiments regarding 10 and 20 cm sludge with 50 mL/L of coagulant dosage is due to the presence of sludge in the final drained (Figure 10). The presence of sludge in the final drained caused a substantial raise in COD values, therefore producing the lowest values of COD percentage reduction. Like in topic 5.2.2, the initial sludge volume also conditions negatively COD reduction.

Table 8. COD from the initial and conditioned sludge, and filtrate using the MOC coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	COD initial (g/L)	COD final filtrate (g/L)	Reduction (%)
Sludge height, 10 cm (V = 200 mL)			
Control	10.2	0.56	95
5		0.65	94
10		1.22	88
25		1.84	82
50		3.4	67
Sludge height, 20 cm (V = 400 mL)			
Control	10.2	1.62	84
5		1.59	84
10		1.83	82
25		2.99	71
50		4.66	54
Sludge height, 40 cm (V = 600 mL)			
Control	27.4	4.87	82
5		4.93	82
10		5.87	79
25		5.87	79
50		6.25	77

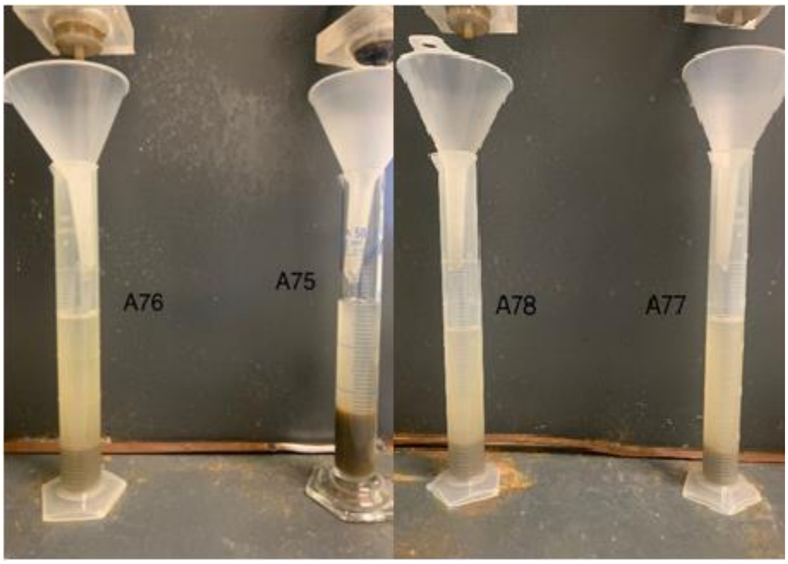

Figure 10. Sludge presence in the final filtrate in 10 and 20 cm dewatering experiments.

5.2.4 Turbidity and pH

The coagulant extracted from *Moringa oleifera* does not significantly alters the pH values of the filtrate nor the sludge (Table 9, Figure 12, Figure 13 and Figure 14), as it was mentioned in other studies, Idris et al. (2016), and Sotheeswaran et al. (2011). The biggest pH difference between initial conditioned sludge and final filtrate was of 1.17, with a coagulant dose of 10 mL/L and sludge height of 20 cm. And the biggest pH difference between the unconditioned sludge and conditioned sludge was of 0.56, when coagulant dose was at 50 mL/L and sludge height of 20 cm.

The increment in coagulant dose shows an increase in turbidity values and, likewise, the increase in sludge height causes the same effect (Table 9 and Figure 11). The maximum turbidity values from the 10, 20 and 40 cm experiments were of 120 NTU, 482 NTU, 1037 NTU, respectively, with maximum coagulant dosage of 50 mL/L.

Table 9. pH and turbidity from the initial and conditioned sludge, and the filtrate, using MOC coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	pH sludge	pH filtrate	Turbidity (NTU)
Sludge height, 10 cm (V = 200 mL)			
Control	6.16	7.56	50
5	6.45	7.55	41
10	6.53	7.32	49
25	6.37	6.83	58
50	6.42	6.49	120
Sludge height, 20 cm (V = 400 mL)			
Control	6.16	7.28	132
5	6.61	7.72	330
10	6.44	7.61	381
25	6.78	7.21	472
50	6.72	6.84	482
Sludge height, 40 cm (V = 600 mL)			
Control	5.6	4.92	581
5	5.36	4.95	465
10	4.82	5.22	635
25	5.1	5.06	703
50	5.1	5.06	1037

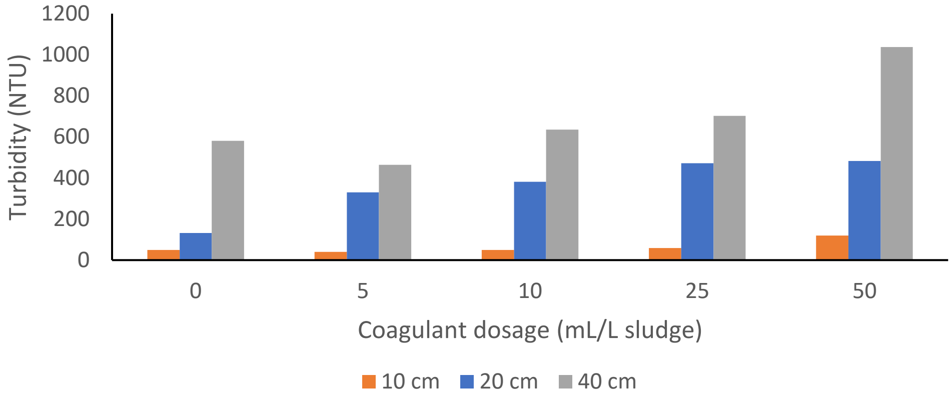

Figure 11. Turbidity values regarding the coagulant dosage, using MOC coagulant (1.0M).

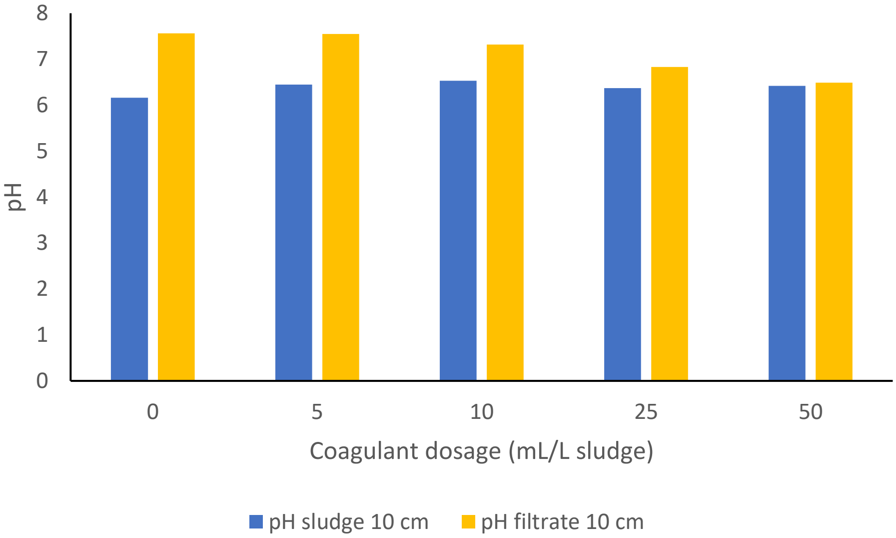

Figure 12. pH from the sludge and filtrate, for 10 cm sludge height experiments, using MOC coagulant (1.0M).

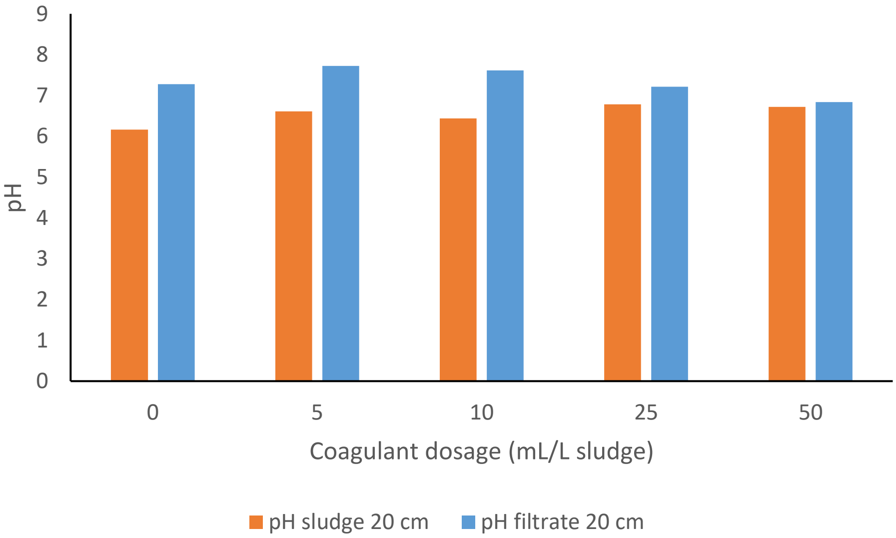

Figure 13. pH from the sludge and filtrate, for 20 cm sludge height experiments, using MOC coagulant (1.0M).

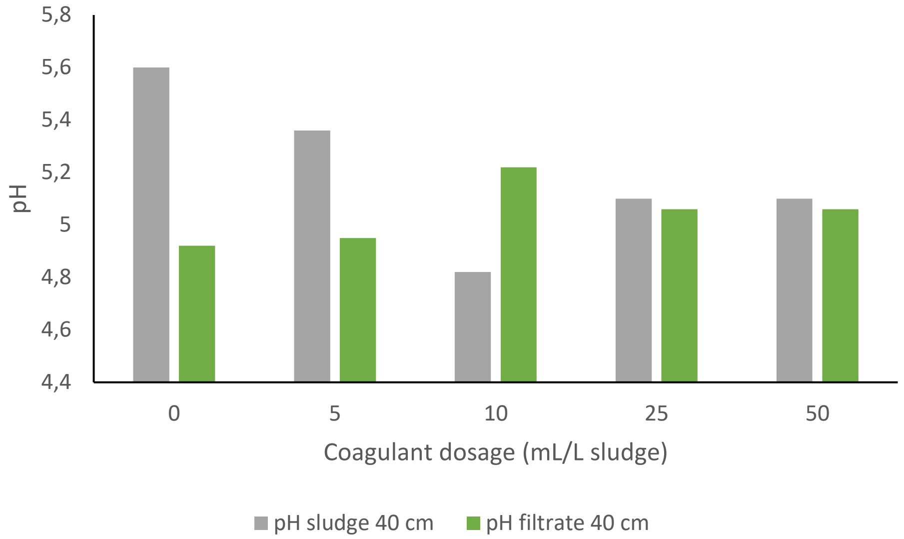

Figure 14. pH from the sludge and filtrate, for different sludge height experiments, using MOC coagulant (1.0M).

5.3 Coagulant extracted with ultrasound technology (MOC-U)

5.3.1 Total Solids (%TS)

For all experiments involving the MOC-U coagulant, the TS values stood beneath from the control experiments (Table 10 and Figure 15). The 10 cm sludge height experiments the TS amount gets higher as there is more coagulant available. Following the same behavior found in topic 5.2.1. The maximum TS value was of 2.77% with coagulant dosage of 50 mL/L, with at an initial TS value of 3.01%.

Both 20 and 40 cm experiments go from an increasing behavior to a decreasing behavior for TS. In the 20 cm sludge height experiments, the increasing behavior stops and reaches its maximum value at a coagulant dosage of 10 mL/L, with 6.90%. In 40 cm sludge height experiments the increasing behavior stops and reaches its maximum at a coagulant dosage of 25 mL/L, with maximum TS value of 5.38%. The initial TS for the 20 and 40 cm sludge height experiments were of 8.36% and 5.79%, respectively.

Table 10. TS using MOC-U coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	TS initial (%)	TS final (%)	Increase (%)
Sludge height, 10 cm (V = 200 mL)			
Control	1.98	3.01	1.03
5		1.4	---
10		1.59	---
25		1.85	---
50		2.77	0.79
Sludge height, 20 cm (V = 400 mL)			
Control	4.5	8.36	0.7
5		6.8	1.13
10		6.9	0.28
25		6.8	0.22
50		5.86	0.06
Sludge height, 40 cm (V = 600 mL)			
Control	4.5	5.79	1.29
5		4.39	---
10		4.5	0
25		5.38	0.88
50		4.21	---

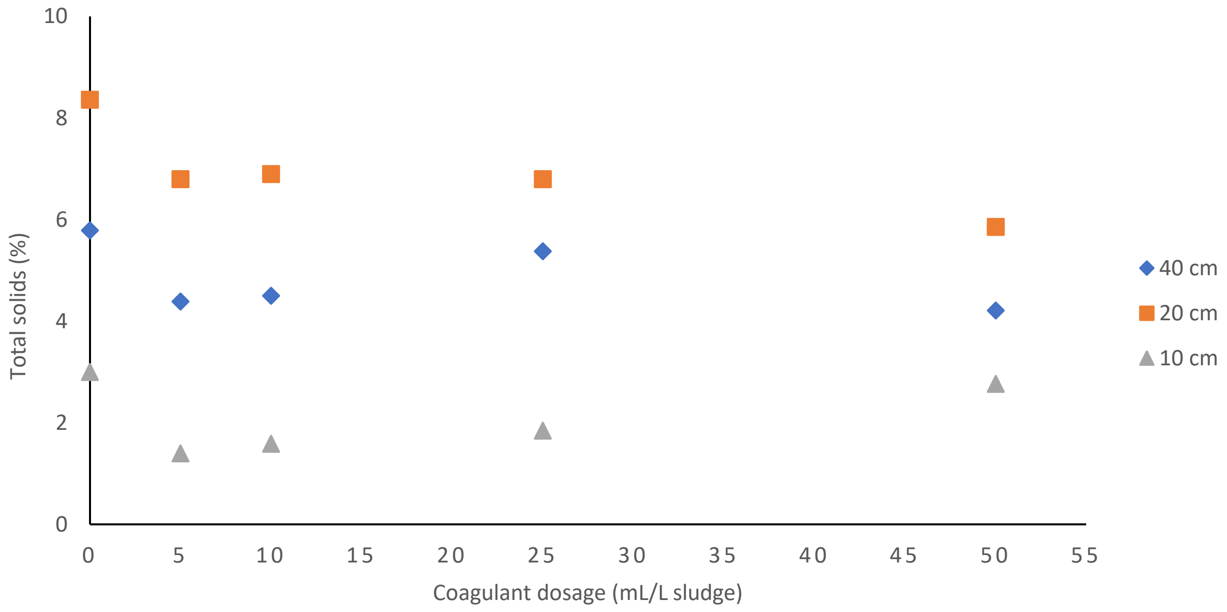

Figure 15. TS for different sludge height and coagulant dosages experiments sing the MOC-U coagulant, NaCl (1.0 M).

5.3.2 Total filtrate drained and drainage rate

The 10 and 20 cm sludge height experiments had performed worse than the control experiments. Only 40 cm experiments resulted in more filtrate removal, when compared to the control experiment (Table 11).

The 10 cm sludge height experiment showed a decreasing behavior until the coagulant dosage reaches 50 mL/L with a maximum filtrate drained value of 60 mL. Showing an opposite behavior than the previous experiments using the MOC coagulant. The 20 and 40 cm sludge heights experiments showed an increasing behavior in TS result as the coagulant dose increases, showing an opposite behavior than the results found in experiments using the MOC coagulant (topic 5.2.2).

The maximum filtrate removal for the 10, 20 and 40 cm sludge height experiments were of 63 mL, 119 mL, 123 mL, respectively. The maximum values were attained in control experiment for the 10 and 20 cm sludge height experiments and 50 mL/L for the 40 cm sludge height.

In the experiments using the MOC-U coagulant the 10 and 20 cm sludge height experiments presented all drainage rate values lower than the control experiment. Only the 40 cm sludge height experiments showed better drainage rates than the control experiment.

Both 20 and 40 cm sludge height experiments presented drainage rates results at an increasing behavior as there is more MOC-U coagulant available. The 10 cm sludge experiment presented a decreasing drainage rate behavior until the coagulant dosage reaches 50 mL/L, presenting same behavior as the liquid removal values.

The maximum drainage rates for the 10, 20 and 40 cm sludge height experiments were of 0.04 mL/min, 0.08 mL/min, 0.09 mL/min, respectively. The MOC-U coagulant did not improve the drainage capabilities from the sludge for the 10 and 20 cm experiments. Only the 40 cm experiments had the drainage properties from the sludge enhanced. Improving the dewatering by 1.57 times comparing to the control experiment.

Table 11. Drainage rate and filtrate removal using the MOC-U coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	Drainage rate for each pre-determined period (mL/min)					Total filtrate (mL)
	15 min	30 min	60 min	300 min	1440 min	
Sludge height, 10 cm (V = 200 mL)						
Control	0.8	0.53	0.35	0.12	0.04	623
5	0.53	0.38	0.26	0.11	0.04	57
10	0.57	0.38	0.26	0.09	0.04	51
25	0.53	0.4	0.27	0.1	0.04	51
50	0.77	0.48	0.34	0.11	0.04	60
Sludge height, 20 cm (V = 400 mL)						
Control	1.4	0.95	0.63	0.24	0.08	119
5	0.8	0.58	0.42	0.25	0.06	84
10	0.93	0.63	0.43	0.17	0.06	89
25	0.93	0.63	0.43	0.17	0.06	89
50	0.93	0.65	0.48	0.18	0.06	91
Sludge height, 40 cm (V = 600 mL)						
Control	0.67	0.48	0.34	0.13	0.05	78
5	1.2	0.87	0.57	0.21	0.07	104
10	1.33	0.92	0.6	0.22	0.07	107
25	1.3	0.9	0.61	0.23	0.08	113
50	1.33	0.95	0.65	0.24	0.09	123

5.3.3 Chemical Oxygen Demand (COD)

The COD results for MOC-U coagulant in the final drained were mostly higher than the previous experiments (Table 12). In addition, the MOC-U coagulant showed similar results of reduction percentage than the experiments using the MOC coagulant. Except for the experiments in which sludge had deposited into the filtrate. The initial COD values from the raw sludge used in MOC-U coagulant experiments were greater than the initial values of the sludge from the experiments using the MOC coagulant. This fact could explain the higher COD values found in the filtrated from experiments using the MOC-U coagulant.

The COD reduction results tends to decrease as the coagulant dosage increases, for the same reasons explained in topic 5.2.3 of this study. The maximum filtrate COD values for the 10, 20 and 40 cm sludge height experiments were of 8 g/L, 11.1 g/L, 13.7 g/L, with maximum coagulant dosage of 50 mL/L. The starting filtrate COD values were of 4.87 g/L, 7.76 g/L, 8.84 g/L, obtained in the control experiments.

Table 12. COD from the initial and conditioned sludge, and filtrate using the MOC-U coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	COD initial (g/L)	COD final filtrate (g/L)	Reduction (%)
Sludge height, 10 cm (V = 200 mL)			
Control	27.4	4.87	82
5		6.23	77
10		6.57	76
25		6.75	75
50		8	71
Sludge height, 20 cm (V = 400 mL)			
Control	63.7	7.76	88
5		9.24	85
10		10.5	84
25		10.5	83
50		11.1	83
Sludge height, 40 cm (V = 600 mL)			
Control	63.7	8.84	86
5		11.9	81
10		11.1	83
25		13	80
50		13.7	79

5.3.4 Turbidity and pH

The pH values from initial sludge, conditioned sludge and final filtrated tended to be constant (Table 13, Figure 17, Figure 18 and Figure 19), like in the experiments using the MOC coagulant. The biggest pH difference between the initial conditioned sludge and final filtrate was of 0.5, when the coagulant dose was at 10 mL/L and sludge height of 20 cm. And the biggest difference between pH values from the unconditioned sludge and the conditioned sludge was of 0.65, with a coagulant dose of 25 mL/L and sludge height of 10 cm.

The turbidity values from the filtrate using MOC-U are higher in comparison to MOC coagulant experiments (Table 13 and Figure 16). This could be explained because of the higher COD values obtained in the filtrate in MOC-U coagulant experiments. Since high COD values in the filtrated may be due to particles that went through the filtering medium of the columns, therefore increasing the turbidity values (Bhuptawat et al., 2007). The maximum turbidity values from the 10, 20 and 40 cm experiments were of 1037 NTU, 996 NTU, 970 NTU, with coagulant dosage of 50 mL/L, 10 mL/L, 50 mL/L, respectively.

The turbidity results from the 10 cm sludge height experiments, presents the same increasing behavior noticed in the experiments using the MOC coagulant (topic 5.2.4). The 20 cm sludge height experiments seem to present an increasing behavior until the coagulant dosage reaches 10 mL/L. The last experiments regarding the 40 cm sludge height experiments do not appear to have a recognizable behavior. In figure 13 the behaviors mentioned are more explicit.

Table 13. pH and turbidity from the initial and conditioned sludge, and the filtrate, using MOC-U coagulant, NaCl solution (1.0 M).

Coagulant dosage (mL/L)	pH sludge	pH filtrate	Turbidity (NTU)
Sludge height, 10 cm (V = 200 mL)			
Control	5.6	5.1	581
5	5.21	4.82	812
10	5.00	4.84	882
25	4.96	5.36	964
50	5.05	5.36	1037
Sludge height, 20 cm (V = 400 mL)			
Control	4.66	4.93	406
5	4.59	4.75	563
10	5.02	4.57	996
25	5.02	4.61	986
50	4.61	4.66	924
Sludge height, 40 cm (V = 600 mL)			
Control	4.66	4.71	604
5	4.64	4.56	724
10	4.59	4.65	896
25	4.88	4.64	819
50	4.84	4.64	970

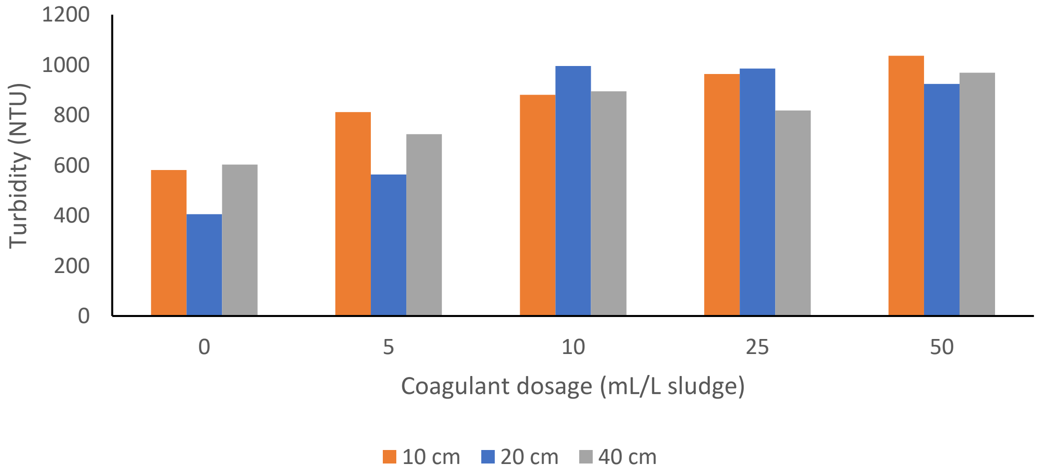

Figure 16. Turbidity values regarding the coagulant dosage, using MOC-U coagulant (1.0M).

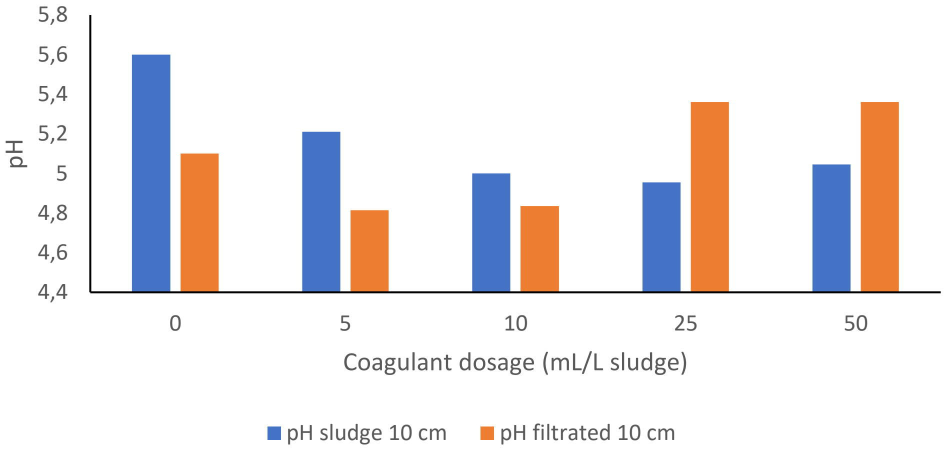

Figure 17. pH from the sludge and filtrate, for 10 cm sludge height experiments, using MOC-U coagulant (1.0M).

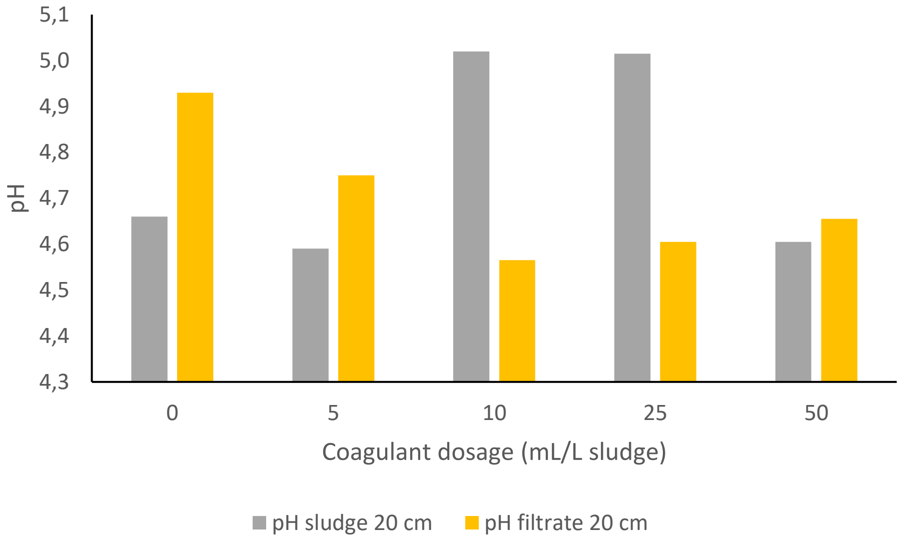

Figure 18. pH from the sludge and filtrate, for 20 cm sludge height experiments, using MOC-U coagulant (1.0M).

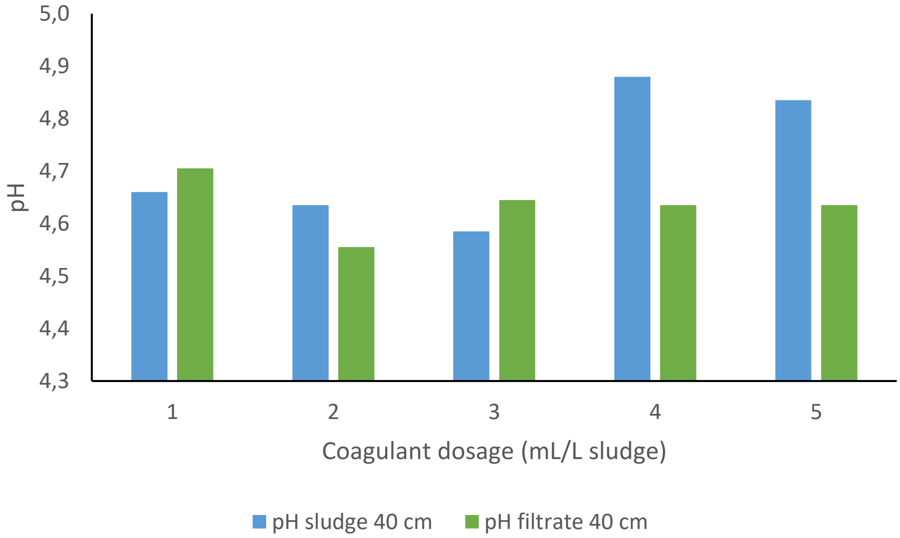

Figure 19. pH from the sludge and filtrate, for 40 cm sludge height experiments, using MOC-U coagulant (1.0M).

5.4 Comparison between both coagulants

The coagulant prepared with ultrasound technology, MOC-U, was not as performant as the coagulant extracted only with salt solution and agitation, MOC. Furthermore, most of the MOC-U results were lower than the control experiments.

Therefore, the ultrasound methodology utilized to extract the active compounds form the *Moringa oleifera* seeds was not optimal. And it ended up worsening the coagulation capabilities, and the dewatering of the sludge, since most of the liquid removal results when using the MOC-U was worse than the control experiments. The only exception was the experiments regarding a sludge height of 40 cm. Table 14 shows the best results of filtrate removal and drainage rate for each experiment.

Even though the results of the experiment using MOC-U with sludge height of 20 and 40 cm were better than the ones using the MOC, it cannot be attributed to the action of MOC-U coagulant, since the best results are attained when there is no addition of the coagulant.

Table 14. Comparison between MOC and MOC-U coagulants of the best liquid removal and drainage rates (coagulant extraction made with NaCl solution of 1,0 M).

MOC				MOC-U			
Sludge height (cm)	Coagulant dosage (mL/L)	Final drainage rate (mL/min)	Filtrate removal (mL)	Sludge height (cm)	Coagulant dosage (mL/L)	Final drainage rate (mL/min)	Filtrate removal (mL)
10	50	0.06	87	10	0	0.04	63
20	5	0.08	114	20	0	0.08	119
40	0	0.06	91	40	50	0.09	123

The coagulation capabilities of the MOC-U coagulant were not as active as the MOC coagulant. Since all TS results using the MOC-U coagulant were beneath the control experiment. Table 15 shows the TS results comparing both coagulants. Although, the TS in different scenarios, the results obtained using the natural coagulants in this study were higher than the ones obtained using an alum-based conditioner in other studies from Ghebremichael et al, (2004). In his study the optimal total solids for the alum conditioner were of 4.76%, while in this study the optimal results were of 18.6% and 6.9%, for the MOC and MOC-U coagulant. Being important to highlight the fact that in his study the dewatering time was lower than the dewatering time in the present study.

Table 15. Best TS result for both coagulants in comparison to the control experiment (experiment coagulant extraction made with NaCl solution of 1,0 M).

MOC				MOC-U			
Sludge height (cm)	Coagulant dosage (mL/L)	TS initial (%)	TS final (%)	Sludge height (cm)	Coagulant dosage (mL/L) (mL/L)	TS initial (%)	TS final (%)
10	0	1.52	6.85	10	0	1.98	3.01
10	50		18.6	10	50		2.77
20	0	1.52	2.22	20	0	4.5	8.36
20	5		2.65	20	10		6.9
40	0	1.98	3.5	40	0	4.5	5.79
40	5		2.5	40	25		5.38

Even though the MOC-U was not as performant as the MOC, it does not imply that the ultrasound technology isn't a viable technology for extracting the coagulant from the seeds. Since in this study it was only experimented extraction process using a fixed amount of time, and potency from the equipment.

Prolonged exposure to the ultrasound waves may cause the destruction of the coagulant agent, therefore losing the coagulation ability (Abidin et al., 2013, Adjeroud-abdellatif et al., 2020; Bouchareb et al., 2021). Therefore, different extracting duration and potency could possibly generate better results, as obtained in studies using other natural occurring coagulants (Opuntia ficus-indica or barbary fig, and Jatropha curcas seeds) made by Abidin et al. (2013) and Adjeroud-abdellatif et al. (2020).

Also, the duration of the conditioning process used in this study is longer than the ones compared to other works using natural coagulants (Okuda et al., 2001; Muyibi et al., 2001; Ghebremichael et al., 2004; Tat et al., 2012). Further investigation should be done to achieve better dewatering results.

The coagulation performed by the MOC appears to form lighter flocs and more porous than the flocs formed using the MOC-U. Behavior noticed in previous studies made by Muyibi et al. (2001). Figure 20, shows the formation of a two-phase system. The MOC coagulant had the solid content being on top of the liquid content, indicating lighter and more porous flocs. While the MOC-U showed opposite results, forming a two-phase system, but with the sludge being deposited on the bottom of the dewatering column.

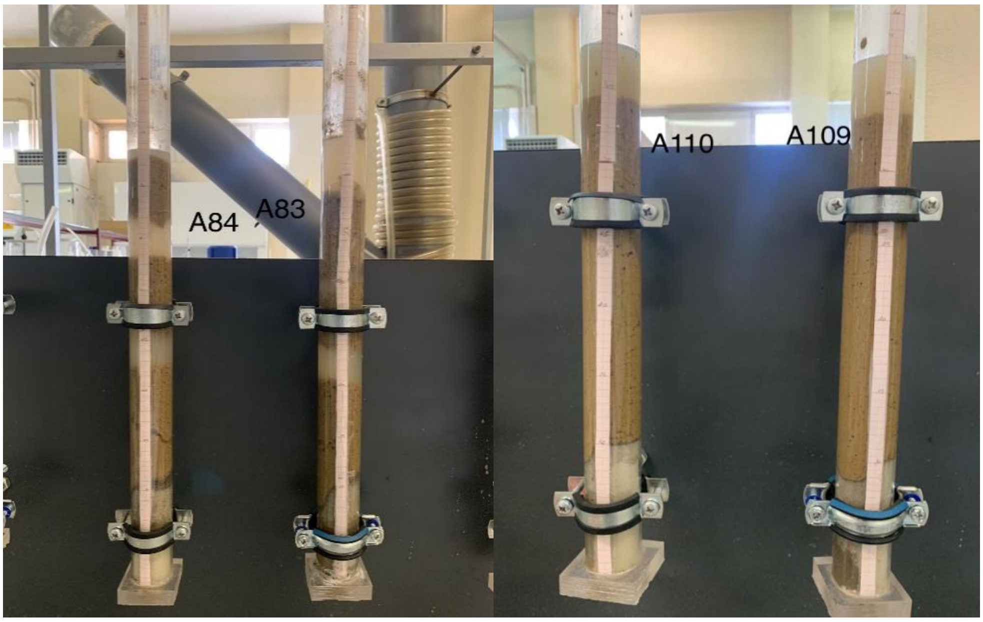

Figure 20. Conditioned sludge with MOC and conditioned sludge with MOC-U in the dewatering columns respectively.

5.5 Important considerations about the experiments

The solids cake left in the columns in the 10 cm sludge experiments using MOC coagulant were next to the filtering medium material, as it is shown in Figure 21. This hindered the sample collection to determine the TS, since it was not possible to gather much material.

Also, the sludge taken from the wastewater treatment plant of Bragança was collected in different dates, which granted sludge with different raw characteristics. One example of that is the TS determined in each experiment. The sludge used in experiments with MOC coagulant presented lower initial TS values than the sludge used in experiments using MOC-U coagulant. Table 16 shows the raw characteristics from the sludges used in the experiments.

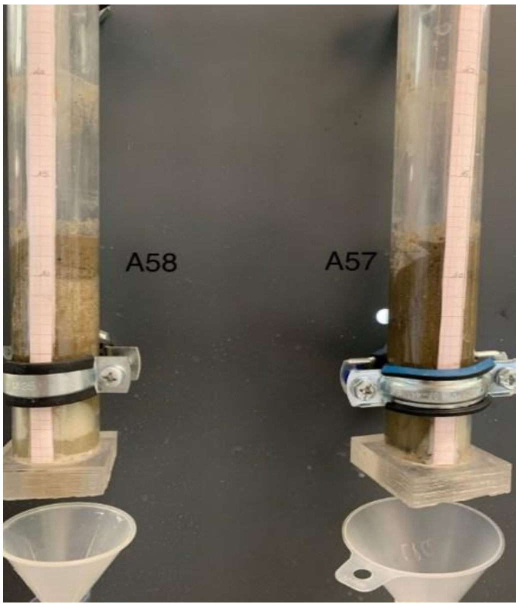

Figure 21. Cake solids left in the column for 10 cm sludge dewatering experiments with MOC coagulant.

Table 16. Raw characteristics from the sludge collected for the dewatering experiments.

SLUDGE SAMPLE	TS (%)	COD (g/L)	pH	COLLECTION DATE
6	1.52	10.4	6.53	04/11/2021
7	1.98	27.4	5.6	27/01/2022
8	4.5	63.7	4.6	03/03/2022

6. CONCLUSIONS

- The coagulant extracted from the *Moringa oleifera* seeds with salt solution, MOC, performed better than the ultrasound assisted coagulant, MOC-U. The MOC coagulant improved the dewatering capabilities of the sludge, being a more accessible, safer, and ecologic friendly option than the usual coagulants used nowadays.
- The best dewatering results were obtained using the MOC coagulant, with a sludge height of 10 cm. The best TS results was of 18.6%, starting from an initial value of 1.52%. And the best filtrated removal value was of 86.5 mL, with an initial sludge volume of 200 mL. Both achieved with maximum coagulant dosage of 50 mL/L.
- Both coagulants did not have a significant impact on sludge nor filtrated pH. Comparing the pH values from the unconditioned sludge and conditioned sludge, the biggest difference was of 0.65 and the biggest difference between conditioned sludge and filtrated was of 1.17.
- The coagulants used increased the COD values in the final filtrate as the coagulant dosage increases. Also, the initial sludge volume used have the same effect, increasing the COD as the initial sludge volume increases.
- Further investigations need to be done to optimize the extraction of the coagulant present in *Moringa oleifera* seeds. And conditioning of the sludge to optimize de dewatering capabilities of the Sludge.

7. REFERENCES

David, M. (Ed.). (2007). *A Comprehensive assessment of water management in agriculture. Water for food Water for life.* London: Earthscan, and Colombo: International Water Management Institute. http://dx.doi.org/10.1016/j.indcrop.2012.05.003

Acheampong, M. A., Paksirajan, K., Lens, P. N. L. (2013). Assessment of the effluent quality from a gold mining industry in Ghana. *Environmental Science and Pollution Research*, 20(6), 3799-3811. DOI 10.1007/s11356-012-1312-3

Ademiluyi, J. O. (1988). Sludge conditioning with moringa seed. *Enviromental International*, 14(1), 59-63.

Ademiluyi, J. O., Eze, R. M. (1990). Improving the sludge conditioning potential of moringa seed. *Environmental Management*, 14(1), 125-129.

Adjeroud-abdellatif, N., Hammoui, Y., Boudria, A., Agab, S., Choulak, F., Leclerc, J., Merzouk, B., Madani, K. (2020). Effect of a natural coagulant extract from *Opuntia ficus-indica* cladode on electrocoagulation-electroflotation water treatment process. *International Journal of Environmental Analytical Chemistry*, DOI:10.1080/03067319.2020.1804889

Ahsan, A., Satter, F., Siddique, A. B., Akbor, A., Ahmed, S., Shajahan., Khan, R. (2019). Chemical and physicochemical characterization of effluents from the tanning and textile industries in Bangladesh with multivariate statistical approach. *Environmental Monitoring and Assessment*, 191(575). https://doi.org/10.1007/s10661-019-7654-2

Ahmadun F. R., Pendashteh, A., Abdullah, L. Q., Biak, D. R. A., Madaeni, S. S., Abidin, Z. Z. (2009). Review of technologies for oil and gas produced water treatment. Journal *of Hazardous Materials*, 170(2-3), 530-551. https://doi.org/10.1016/j.jhazmat.2009.05.044

Allaoui, M., Schmitz, T., Campbell D., de la porte, C. A. (2015). Good Practices for Regulating Wastewater Treatment: Legislation, Policies and Standards. *United Nations Environment Programme*. ISBN: 978-92-807-347-4

Alloway, B. J., Jackson, A. P. (1991). The behavior of heavy metals in sewage sludge-amend soils. *The Science of the Total Environment*, 100, 151-176.

Andreoli, C. V., Sperling, M. V., Fernandes, F. (2007). *Sludge Treatment and Disposal, Biological Treatment Series*. London, IWA publishing. ISBN:1-84339-166-X; ISBN 13: 9781843391661.

Andreoli, C. V., Garbossa, L. H. P., Lupatini, G., Pegorini, E. S. (2015). Wastewater sludge management: a Brazilian approach. 117-130. Available at https://www.researchgate.net/publication/228670690

Anwar, F., Rashid, U. (2007). Physiochemical characteristics of *moringa oleifera* seeds and seed oil from a wild provenance of Pakistan. Pakistan *Journal of Botany*, 39(5), 1443-1453.

Baird, R. B., Eaton, A. D., Rice, E. W. (2017). *Standard Methods for the Examination of Water and Wastewater*. Washington DC, American Public Health Association, American Water Works Association, Water Environment Federation, ed. 23.

Bindes, M. M. M., Miria, H. M. R., Cardoso, D. C. B. (2019). Ultrasound-assisted extraction of bioactive compounds from green tea leaves and clarification with natural coagulants (chitosan and *Moringa oleifera* seeds). *Ultrasonics-Sonochemistry*, 51, 111-119.

Boatman, N., Stoate, C., Gooch, R., Carvalho, C. R., Borralho, R., Snoo, G., Eden, P. (1999). *The Environmental Impact of Arable Crop Production in the European Union: Practical Options for Improvemen*t. UK, Allerton Research and Educational Trust; Belgim, Eurinco Ltd; Portugal, ERENA; The Netherlands, CML (Centre of Environmental Science), Leiden University. Available at http://ec.europa.eu/environment/agriculture/pdf/arable.pdf.

Bouchareb, R., Derbal, K., Benalia, A. (2021). Optimization of active coagulant agent extraction method from Moringa oleifera seeds for municipal wastewater treatment. *Water & Science Technology*, 84(2). DOI: 10.2166/wst.2021.234

Buechler, S., Mekala, G. D., Keraita, B. (2005). Wastewater Use for Urban and Peri-urban Agriculture. *Cities Farming for the Future: Urban Agriculture for Green and Productive Cities*. 1, 473.

Bhuptawat, H., Folkard, G. K., Chaudhari, S. (2007). Innovative physico-chemical treatment of wastewater incorporating *Moringa oleifera* seed coagulant. *Journal of Hazardous Materials.* 142, 477-482. doi:10.1016/j.jhazmat.2006.08.044

Cai, Q. Y., Mo, C. H., Wu, Q. T., Zeng, Q. Y., Katsoyiannis, A. (2007). Occurrence of organic contaminants in sewage sludges from eleven wastewater treatment plants. *Chemosphere*, 68, 1751-1762. doi: 10.1016/j.chemosphere.2007.03.041

Carr, R. M., Blumenthal, U. J., Mara, D. D. (2004). *Health Guidelines for the Use of Wastewater in Agriculture: Developing Realistic Guidelines*. [S.l], CAB International.

Carr, G. M., Neary, J. P. (2008). *WATER QUALITY – for Ecosystem and Human Health*. [S.l] United Nations Environment Programme Global Environment Monitoring System, 2nd edition. Available at https://www.cbd.int/doc/health/health-waterquality-en.pdf.

Chang, L. L., Raudenbush, D. L., Dentel, S. K. (2001). Aerobic and anaerobic biodegrability of a flocculant polymer. *Water Science and Technology*, 44(2-3), 461-468.

Chang, G. R., Liu, J. C., Lee, D. J. (2001). Co-conditioning and dewatering of chemical sludge and waste activated sludge. *Water Research*, 35(3), 786-794.

Cieslik, B. M., Namiesnik, J., Konieczka, P. (2015). Review of Sewage Sludge Management: standards, regulations and analytical methods. *Journal of Cleaner Production*, 90, 1-15.

Corcoran E., Nelleman C., Baker E., Bos R., Osborn D., Savelli H. (2010). *Sick Water? The central role of wastewater management in sustainable development*. Norway, United Nations Environment Programme., UN-HABITAT., GRID-Arendal, 1-85.

Drechsel, P., Scott, C. A., Raschid-sally, L., Redwood, M., Bahri, A. (2010). *Wastewater irrigation and health: assessing and mitigating risk in low-income countries*. [S.l], International Water Management Institute (IWMI); London, UK: Earthscan; Ottawa, Canada: International Development Research Centre (IDRC), 404p.

Decreto-Lei n° 276/2009 de 2 de outubro. *Diário da República n° 192/09 – 1ª Série*. Lisboa: Ministério do Ambiente, do Ordenamento do Território e do Desenvolvimento Regional.

Franco-hernández, O., Mckelligan-gonzalez, A. N., Lopez-olguin, A. M., Espinosa-ceron, F., Escamilla-silva, E., Dendooven, L.(2003). Dynamics of carbon, nitrogen and phosphorus in soil amended with irradiated, pasteurized and limed biosolids. *Bioresource Technology*, 87, 93-102. https://doi.org/10.1016/S0960-8524(02)00188-8

Ghebremichael, K. A., Hultman, B. (2004). Alum sludge dewatering using *Moringa oleifera* as a conditioner. *Water, Air, and Soil Pollution*, 158, 153-167.

Ghebremicheal, K. A., Gunaratna, K. R., Henriksoon, H., Brumer H., Dalhammar, G. (2005). A simple purification and activity assay of the coagulant protein from *Moringa oleifera seed*. *Water Research*, 39, 2338-2344.

Gidde, M. R., Bhalerao, A. R., Malusare, C. N. (2012). Comparative study of different forms of *Moringa oleifera* extracts for turbidity removal. *International Journal of Engineering Research and Development*, 2, 14-21.

Haldar, R., Kosanar, S. (2017). *Moringa oleifera*: The Miracle Tree. *International Journal of Advance Research, Ideas and Innovations In Technology*, 3, ISSN: 2454-132X

Hauer, F. R., Laberti, G. A. (1996). *Methods in Stream Ecology*. USA, UK, Elsevier Inc., 2 ed. ISBN 13: 978-0-12-332908-0, ISBN 10:0-12-332908-6.

Horzic, D., Jambrak, A. R., Belscak-cvitanovic, A., Komes, D., Lelas, V. **(**2012). Comparison of conventional and ultrasound assisted extraction techniques of yellow tea and bioactive of composition of obtained extracts. *Food Bioprocess Technology*, 19(1), 721 – 728. DOI: 10.1007/s11947-012-0791-z

Idris, M. A., Jami, M. S., Hammed, A. M., Jamal, P. (2016). *Moringa oleifera* seed extract: A review on its environmental applications. *International Journal of Applied Environmental Sciences*, 11(6), 1469-1486.

Jahn, S. A. A. (1988) Using moringa seeds as coagulants in developing countries. *Journal of American Water Works Association*, 80, 43-50.

Jovanoíc, A. A., Dordevic, V. B., Zdunic, G. M., Pljevljakusic, D. S., Savikin, K. P., Godevac, D. M., Bugarski, B. M. (2017). Optimization of the extraction process of polyphenols from *Thymus serpyllum* L. herb using maceration, heat- and ultrasound-assisted techniques. *Separation and Purification Technology*, 179, 369-380.

Kabir, M. M., Fakhruddin, A. N. M., Chowdhury, M. A. Z., Fardous, Z., Islam, R. (2017). Characterization of tannery effluents of Hazaribagh area. *Bangladesh Pollution*, 3(3), 395-406.

Kaggwa, R. C., Mulalelo, C. I., Denny, P., Okurut, T. O. (2001). The impact of alum discharges on a natural tropical wetland in Uganda. *Water Research*, 35(3), 795-807.

Kasmi, M. (2018). Biological Process as Promoting Way for both treatment and valorization of dairy industry effluents. *Waste Biomass Valor*, 9, 195-209. DOI 10.1007/s12649-016-9795-7

Krishnaveni, R., Devi, Y. P., RAO, S. R. (2013). Bioremediation of steel industrial effluents using soil microorganisms. *International Journal of Advanced Biotechnology and Research,* 4, 914-919. ISSN 0976-2612, Online ISSN 2278–599X

Manfredi, S., Pant, R. (2013). Improving the environmental performance of bio-waste management with life cycle thinking (LCT) and life cycle assessment (LCA). [S.l], Int J Life Cycle Assess, Springer, 18, 285-291. DOI 10.1007/s11367-012-0497-5

Mara, D. (2003). *Domestic wastewater treatment in developing countries*. UK, USA, Earthscan; WWF-UK; International Institute for Environment and Development. ISBN:1-84407-019-0 paperback, ISBN:1-84407-020-4 hardback.

Muyibi, S. A., Noor, M. J. M. M., ONG, D. T., KAI, K. W. (2001). *Moringa oleifera* seeds as a flocculant in waste sludge treatment. *International Journal of Environmental Studies,* 58(2), 185-195. Available at: https://doi.org/10.1080/00207230108711326. Access on 28/09/2021.

Nellemann, C., Corcoran, E., Duarte, C. M., Valdés, L., De young, C., Fonseca, L., Grimsditch, G. (2009). *Blue Carbon. A Rapid Response Assessment*. Norway, United Nations Environment Programme; GRID-Arendal. ISBN:978-82-7701-060-1.

OECD. (2012). *OECD ENVIRONMENTAL OUTLOOK TO 2050: The Consequences of Inaction – HIGHLIGHTS*. [S.l], OECD. Available at https://www.oecd.org/g20/topics/energy-environment-green-growth/oecdenvironmentaloutlookto2050theconsequencesofinaction.htm.

OFFICIAL JOURNAL OF THE EUROPEAN UNION. 2008. Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives. Available in https://eur-lex.europa.eu/legal-content/PT/TXT/?uri=celex:31986L0278.

Okuda, T., Baes, A. U., Nishijima, W., Okada, M. **(**1999). Improvement of extraction method of coagulation active components from *moringa oleifera* seed. *Water Research*, 33, 3373-3378. https://doi.org/10.1016/S0043-1354(99)00046-9

Okuda, T., Baes, A. U., Nishijima, W., Okada, M. (2001). Coagulation mechanism of salt solution-extracted active component in *Moringa oleifera* seeds. *Water Research*, 35(3), 830-834.

Olofsson, U., Bignert, A., Haglund, P. (2012). Time-trends of metals and organic contaminants in sewage sludge. *Water Research,* 46, 4841-4851. https://doi.org/10.1016/j.watres.2012.05.048.

ONU News. (2019). *ONU prevê que cidades a abriguem 70% da população mundial até 2050*. Access on June 12. 2021. Available on: https://news.un.org/pt/story/2019/02/1660701.

Palaniappan, M., Gleick, P. H., Allen, L., Cohen, M. J., Smith, J. C., Smith, C. (2010). *Clearing the waters: A Focus on Water Quality Solutions*. UNEP, 91p. Available on https://pacinst.org/publication/clearing-the-waters-focus-on-water-quality-solutions/.

Poongothai, S., Ravikrishnan, R., Murthy, P. (2007). Endocrine disruption and perspective human health implications: A review. *The Internet Journal of Toxicology*, 4(2).

Qasim, W., Mane, A. V. (2013). Characterization and treatment of selected food industrial effluents by coagulation and adsorption techniques. *Water Resources and Industry*, 4, 1-12. http://dx.doi.org/10.1016/j.wri.2013.09.005

Ranade, V. V., Bhandari, V. M. (2014). *Industrial Wastewater Treatement, Recycling, and Reuse*. UK, USA, Elsevier Ltd. ISBN: 978-0-08-099968-5

Rashid-sally, L., Jayakody, P. (2008). Drivers and characteristics of wastewater agriculture in developing countries: Results from a global assessment. Colombo, *International Water Management Institute*, 29.

Sánchez-martín, J., GhebremichaeL, K., Beltrán-heredia, J. (2010). Comparison of single-step and two-step purified coagulants from *Moringa oleifera* seed for turbidity and DOC removal. *Bioresource Technology*, 101, 6529-6261.

Sancho, F. H., Diallo, B. L., Sagasta, M., Qadir, M. (2015). *Economic Valuation of Wastewater-The cost of action and the cost of no action*. [S.l] UNEP, 68 p.

Santos-sotero, R. B., Rocha, O., Povinelli, J. (2007). Toxicity of ferric chloride sludge to aquatic organisms. *Chemosphere*, 68, 628-636.

Sharma, P., Tripathi, S., Chandra, R. (2021). Metanogenic analysis for profiling of microbial communities and tolerance in metal-polluted and paper industry wastewater. *Bioresource Technology*, 324, 4841-4851. https://doi.org/10.1016/j.watres.2012.05.048

Somlyody, L., Varis, O. (2006). Freshwater under pressure. *International Review for Environmental Strategies*, 6(2), 181-204.

Sotheeswaran, S., Nad, V., Matakite, M., Kanayathu, K. (2011). *Moringa oleifera* and other local seeds in water purification in developing countries. *Research Journal of Chemistry and Environment*, v. 15(2). 135-138.

Sperling, M. V. (2007). *Biological Wastewater Treatment Series*. UK, IWA publishing. Wastewater Characteristics, Treatment and Disposal. v. 1. ISBN: 1 84339 161 9; ISBN 13: 9781843391616

Suarez, M., Entenza, J. M., Doerries, C., Meyer, E., Bourquin, L., Sutherand, J., Marison, I., Moreillon, P., Mermod, N. (2002). Expression of a plant-derived peptide harboning water-cleaning and antimicrobial activities. *Wiley Periodics*, 81(1), 13-20. doi: 10.1002/bit.10550

Sutherland, J. P., Folkard, G. K., Grant, W. D. (1990). Natural coagulants for appropriate water treatment: a novel approach**.** *Waterlines*, 8(4), 30-32.

Tat, W. K., Idris, A., Noor, M. J. M. M., Mohamed, T. A., Ghazali, A. H., Muyibi, A. (2010). Optimization study on sewage sludge conditioning using *Moringa oleifera* seeds. *Desalinization and Water Treatment*,16, 402-410 p. Available at: https://doi.org/10.5004/dwt.2010.1271

Thomaidi, V. S., Stasinakis, A. S., Borova, V. L., Thomaidis, N. S. (2016). Assessing the risk associated with the presence of emerging organic contaminants in sludge-amended soil: A country-level analysis. *Science of the Total Environment*, 548-549 280-288 p. http://dx.doi.org/10.1016/j.scitotenv.2016.01.043.

Tyagi, V. K., Lo, S. L. (2013). Sludge: A waste or renewable source for energy and resources recovery?. *Renewable and Sustainable Energy reviews*, 25, 708-728.

UNDP. (2006). *Human Development Report 2006. Beyond Scarcity: Power, poverty and the global water crisis*. New York, UNDP. Available at www.undp.org/content/undp/en/home/librarypage/hdr/human-development-report2006.html.

UNESCO. (2017). *Wastewater the untapped resource*, *The United Nations World Water Development Report 2017***.** Paris, UNESCO, 0-198. ISBN 978-92-3-100201-4

UNFPA. (2009). *State of the World Population 2009. Facing a changing world: women, population and climate*. UNFPA, 1-94. Available at: https://www.unfpa.org/publications/state-world-population-2009.

UN WATER. (2011). *Policy Brief – Water Quality*. UNEP. Available at https://www.unwater.org/publications/un-water-policy-brief-water-quality/.

Vijay, K. K., Rubha, M. N., Manivasagan, M., Ramesh, B. N. G., Balaji, P. (2012). *Moringa oleifera* – The nature's gift. *Universal Journal of Environmental Research and Technology*, 2, 203-209. Available at www.environmentaljournal.org.

Xu, C. C., Lancaster, J. (2012). Review: Treatment of secondary sludge for energy recovery. 187–212.

Wada, Y., Bierkens, M. F. P. (2014). Sustainability of global water use: past reconstruction and future projections. *Environmental Research*. 9(10), 1-17. 104003. Available at https://iopscience.iop.org/article/10.1088/1748-9326/9/10/104003/meta.

Wang, L., Weller, C. L. (2006). Recent advances in extraction of nutra-ceuticals from plants. *Food Science & Technology*, 17, 300-312.

World Water Assessment Programme. (2009). *The Uniterd Nations World Water Development Report 3: Water in a Changing World*. Paris, London, UNESCO, Earthscan. ISBN: 978-9-23104-095-5; ISBN: 978-1-84407-839-4; ISBN: 978-1-84407-840-0

Yu, S., Zhang, G., Li, J., Zhao, Z., Kang, X. (2013). Effect of endogenous hydrolytic enzymes pretreatment on the anaerobic digestion of sludge. *Bioresource Technology*, 146, 758-761. Available at: https://doi.org/10.1016 /j.biortech. 2013.07. 087