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Evaluation of the reuse potential of water from decentralized constructed wetland-based wastewater treatment plants in Aljezur, Portugal

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Mestrado em Ecohidrologia Aplicada

Dissertation made under the supervision of supervisors:
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Work Authorship Declaration

I declare to be the author of this work, which is unique and unprecedented. Authors and works consulted are properly cited in the text and are included in the listing of references.

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Resumo

A escassez de água é um problema global cada vez mais urgente, exacerbado pelas mudanças climáticas, crescimento populacional e diversas atividades económicas, como a agricultura e o turismo. Este estudo pretende avaliar o potencial de reutilização de efluentes de Estações de Tratamento de Águas Residuais baseadas em Zonas Húmidas Construídas (CW-WWTPs) em dois locais na região do Algarve, Carrapateira e Maria Vinagre, que são vulneráveis a precipitações irregulares e aumento da procura de água. Para potencializar a eficiência desses sistemas, este trabalho propõe duas experiências que integram uma abordagem híbrida.

A caracterização da amostra de água bruta indicou que os efluentes de Carrapateira e Maria Vinagre não cumprem os requisitos do Decreto-Lei Português nº 119/2019, que regula a produção e uso de águas residuais tratadas para fins de reutilização. A concentração de *Escherichia coli* foi o parâmetro de qualidade da água que excedeu o limite legal para todas as categorias de reutilização de água.

Os tratamentos testados incluem um processo de coagulação, floculação e sedimentação (C/F/S) e um filtro de Carvão Ativado à Base de Casca de Coco (CSAC). Como alternativa natural aos coagulantes químicos convencionais, o Tanfloc foi escolhido, destacando-se pelo seu custo-benefício, sustentabilidade ambiental e segurança humana. Os resultados demonstram melhorias significativas na qualidade da água, tornando o efluente reutilizável em várias categorias de irrigação, como C, D e E, e todos os usos urbanos. O Tanfloc e o CSAC mostraram-se particularmente eficazes na redução de *E. coli*, alcançando remoções de 90 a 99%. Para a turvação, o segundo parâmetro mais limitante, as remoções variaram entre 84 e 89% para o Tanfloc e entre 30 e 60% para o filtro CSAC.

Com base nesses resultados, o estudo defende uma abordagem de tratamento duplo que seja eficiente em termos de custo e escalável. Isso se alinha bem com as restrições de recursos típicas de WWTPs descentralizadas e tem implicações políticas substanciais, especialmente no contexto do Decreto-Lei nº 119/2019 de Portugal. A eficácia do Tanfloc e do CSAC poderia servir como um catalisador para mudanças regulatórias, incentivando a adoção mais ampla dessas soluções híbridas. Olhando para o futuro, há necessidade de pesquisas adicionais para ajustar as dosagens de Tanfloc e explorar as limitações da capacidade adsortiva do CSAC.

Este trabalho contribui significativamente para a gestão sustentável da água e oferece soluções para pequenas comunidades remotas que enfrentam escassez de água, já que integra soluções inovadoras e naturais no tratamento de águas residuais e que permitem a reutilização da água.

Palavras-chave: Reutilização de água, zonas húmidas construídas, coagulação, floculação, sedimentação, carbono ativado, tratamento de águas residuais, gestão sustentável da água, Tanfloc, Algarve, baseado na natureza, eco-hidrologia.

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Abstract

Water scarcity is an increasingly urgent global issue, exacerbated by climate change, population growth, and various economic activities such as agriculture and tourism. The study aims to tackle these challenges by evaluating the water reuse potential of effluents from decentralized Constructed Wetland-based Wastewater Treatment Plants (CW-WWTPs) in two locations within the Algarve region in Portugal, Carrapateira and Maria Vinagre, which are vulnerable to irregular rainfall and rising water demand. To boost the efficiency of these systems, this research proposes two experiments integrating a hybrid approach.

The characterization of the raw water sample revealed that both Carrapateira and Maria Vinagre effluents are not able to be reused as per the Portugese Decree-Law No. 119/2019, which regulates the production and use of treated wastewater for reuse purposes. *Escherichia coli* concentration was the mandatory water quality parameter which exceeded the legal threshold for all water reuse categories, therefore not allowing the Carrapateira and Maria Vinagre any reuse potential.

The treatments tested include a coagulation, flocculation, and sedimentation (C/F/S) process and a Coconut Shell-Based Activated Carbon (CSAC) filter. As a natural alternative to conventional chemical coagulants, Tanfloc was chosen due to its environmentally friendly nature and safety for human exposure. Tanfloc optimal dosage was determined upon the highest turbidity removal rates, being 40 mg/L and 50 mg/L for Carrapateira and Maria Vinagre respectively. The results show significant improvements in water quality, where Tanfloc and CSAC were particularly effective in reducing *E. coli*, achieving removal rates of 96 to 99% and 90 to 94% respectively. Turbidity removal was achieved at 84% to 89%, and 30 to 60% for Tanfloc and the CSAC filter respectively. Also, measurements and analysis of total organic carbon, pH, conductivity, biological oxygen demand, total suspended solids, Ammonium, total nitrogen, and total phosphorus were performed.

The results for the mandatory water quality parameters (*E.coli*, turbidity, BOD_5, total suspended solids) after the C/F/S and CSAC treatments revealed that the treated effluent from both Carrapateira and Maria Vinagre can be reused in various categories such as C, D, and E, categories associated with irrigation with restricted access only for agricultural crops where the consumable part is not in direct contact with water, and all urban uses.

Building on these findings, the study advocates for a complementary dual-treatment approach that is realistic, affordable, and scalable. This aligns well with the resource constraints typical of decentralized WWTPs and has substantial policy implications, especially in the context of Portugal's Decree-Law No. 119/2019. The effectiveness of Tanfloc and CSAC could catalyze regulatory change, encouraging broader adoption of these hybrid solutions. Moving forward, additional research is needed is a need for additional research to fine-tune Tanfloc dosages and explore the limitations of CSAC's adsorptive capacity.

This research contributes significantly to the broader discourse on sustainable water management and offers actionable insights for small, remote communities facing water scarcity. It opens new avenues for the integration of innovative, eco-friendly solutions in wastewater treatment and water reuse, thereby promoting sustainable water resource management at both regional and potentially global scales.

Keywords: Water reuse, constructed wetlands, coagulation, flocculation, sedimentation, activated carbon, wastewater treatment, sustainable water management, Tanfloc, Algarve, nature-based, ecohydrology.

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List of acronyms and abbreviations

AC	Activated Carbon
BOD	Biological Oxygen Demand
C/F/S	Coagulation, Flocculation and Sedimentation
CFU	Colony-forming units
COD	Chemical Oxygen Demand
CSAC	Coconut shell-based Activated Carbon
CW	Constructed Wetland
HDPE	High-density polyethylene
HSSF	Horizontal subsurface flow
IUCN	International Union for Conservation of Nature
LAQ	Laboratório de Análises Químicas
MV	Maria Vinagre
NbS	Nature-based Solutions
NTU	Nephelometric Turbidity Units
PAC	Poly-aluminum chloride
TOC	Total Organic Carbon
TSS	Total suspended solids
WWTP	Wastewater treatment plant

1. Introduction

1.1 Water scarcity, the need for reuse and Nature-based solutions

Water scarcity, an escalating global concern, results from the convergence of surging population growth and resource-intensive economic activities. This issue gains critical urgency as the availability of freshwater becomes increasingly limited. The impact of climate change, further exacerbating the decline in terrestrial water storage, compounds this challenge, amplifying water scarcity and potentially disrupting societal activities (Jingyu et al., 2020; United Nations, 2023; Visser-Quinn et al., 2021).

In response to the strain on conventional water sources, the concept of water reuse emerges as a compelling strategy (Shoushtarian & Negahban-Azar, 2020; Miller, 2006). This approach is particularly significant considering that approximately 71.7% of the annual withdrawal of about 4250 km^3 from natural water bodies is designated for agricultural use (FAO, 2021). Recycled water, notably for applications like irrigation, holds promise to alleviate pressure on freshwater supplies (Bauer, 2020; Hardy et al., 2015).

Yet, water reuse presents intricate challenges. The interplay of environmental, economic, and social factors shapes the feasibility of reuse initiatives (Boretti & Rosa, 2019; Jury & Vaux, 2007; Sharma et al., 2021). Amid these complexities, economic and technical considerations loom, underscoring the need for cost-effective technologies with broad policy and public support (Gonzalez-Flo et al., 2023).

Nature-based solutions (NbS) emerge as a compelling approach to address water scarcity sustainably. Among them, constructed wetlands (CWs) stand out as a promising avenue for tackling water reuse challenges. CWs mimic natural processes to effectively treat wastewater using a combination of physical, chemical, and biological mechanisms (Masoud et al., 2022; Zhi & Ji, 2012). With adaptability, flexible design, and controlled water flow, CWs promise sustainable water management solutions (Ferreira et al., 2023; Hadis et al., 2020).

1.2 Background and context of the case study: Aljezur, Portugal.

The south of Portugal, the Algarve, is characterized by an irregular precipitation regime varying from intermittent floods in winter, to inter-annual extreme periods of drought (Fragoso & Tildes Gomes, 2008; Nunes et al., 2006). This region has continuously experienced water scarcity scenarios due to the impacts of climate change, increased irrigation and tourism water demand, and inadequate water management practices (Soares & Lima, 2022). Under this scenario, the urban water management company *Águas do Algarve, S.A.* has been addressing the need to find alternative sources or reuse of water to supply for domestic, agricultural, and recreational purposes (Do Ó, 2007).

This clear need to find new sources of water is aligned with the development of the law decree Nº 119/2019, Portuguese legislation that establishes the legal framework for the "*production of water for reuse, obtained from the treatment of wastewater, as well as its use*" (Decreto-Lei n.º 119/2019, 2019). Before the existence of this law, there were already some initiatives in the Algarve for water reuse in, for example, golf courses, citrus agriculture and even to support specific ecosystems (Rebelo et al., 2020). Today, the law clearly establishes which effluents are allowed to be reused for specific non-potable uses.

One of the effluents that can be legally reused is treated wastewater from domestic use, which can be found in conventional wastewater treatment plants (WWTP). These treatment plants vary in their capacity to meet a given demand, the technology they use, their operation and maintenance costs, and whether they are connected or not to a broader water distribution and sanitation network. In the Algarve, some of these WWTP are sceptic systems that consist of an initial sceptic tank and a further treatment in constructed wetlands (CW). The CWs contain vegetation, substrates and microorganisms with the objective of recreating and mimicking the purifying capabilities found in natural wetlands (Calheiros et al., 2011). Furthermore, these CW-based WWTPs are designed usually for small remote populations and villages that, due to high costs, do not have access to a water and sanitation network (Figueiredo, 2019; Pereira et al., 2018).

In the Algarve, WWTPs are distributed generally in coastal regions like, for example, the town of Aljezur (Pereira et al., 2018). In this town, there are at least five CW-based WWTPs that have the objective to satisfy the domestic wastewater treatment demand throughout the year, including the demand increase during the summer season (Águas do Algarve, 2019). Considering the isolated and water scarce context in which these WWTPs are located, it would be ideal to reuse the resulting effluent for local non-potable uses, however, preliminary studies have found that the water quality of the effluent is still not good enough to meet the minimum thresholds allowed by the law decree Nº 119/2019 (Lopes, 2021). Given this situation, this research explores alternatives to complement the septic system composed of septic tanks and constructed wetlands to meet a law-abiding water quality of the treated effluent to be reused.

1.3 Research question and objectives

This study presents the following research question to guide the research, as well as discussions and conclusions:

What is the potential of using a nature-based coagulant, Tanfloc, or coconut shell-based activated carbon (CSAC) to improve the quality of the effluent for non-potable reuse purposes from the CW-based WWTPs in Aljezur?

Thus, this study focuses on evaluating the potential for reusing the effluent from CW-based WWTP systems which are used as water treatment plants for small remote populations and villages in the Algarve region of Portugal. Specifically, this research explores the use of a nature-based coagulant agent, and a filter made of nature-based activated carbon to assess their effectiveness and capacity in removing contaminants from the treated effluent.

The study involves laboratory-scale experiments of coagulation / flocculation / sedimentation (C/F/S) and a filtering media as a mean to observe the quality of the effluent before and after the processes. The proposed alternative should be low cost, nature-based, sustainable, not reliant on previously installed gray infrastructure, and able to provide an effluent quality which is consistent with Portuguese legislation on the production of irrigation water for reuse.

In this regard, the study has the following specific objectives:

- To assess the quality of the effluent from two representative CW-based WWTPs in the Aljezur region and compare it with the minimum thresholds established by the law decree Nº 119/2019 for non-potable reuse,

- To evaluate the effectiveness of Tanfloc as a nature-based coagulant agent in improving the quality of the effluent,

- To investigate the capacity of a coconut shell-based activated carbon filter in improving the quality of the effluent,

- To recommend a results-based complementary process, low-cost, hybrid, sustainable, and capable of improving the current effluent for non-potable reuse to be installed in an affordable way at the current CW-based WWTPs.

2. Literature review

2.1 Water reuse: Urgent need and constrains for implementation

Water is a finite resource in growing demand. As the global population increases and resource-intensive economic grows, water scarcity intensifies and water supply quality decreases. Water scarcity is a critical global issue that affects numerous regions, with water resources and infrastructures in many countries failing to meet the accelerating demand (Figure 2.1).

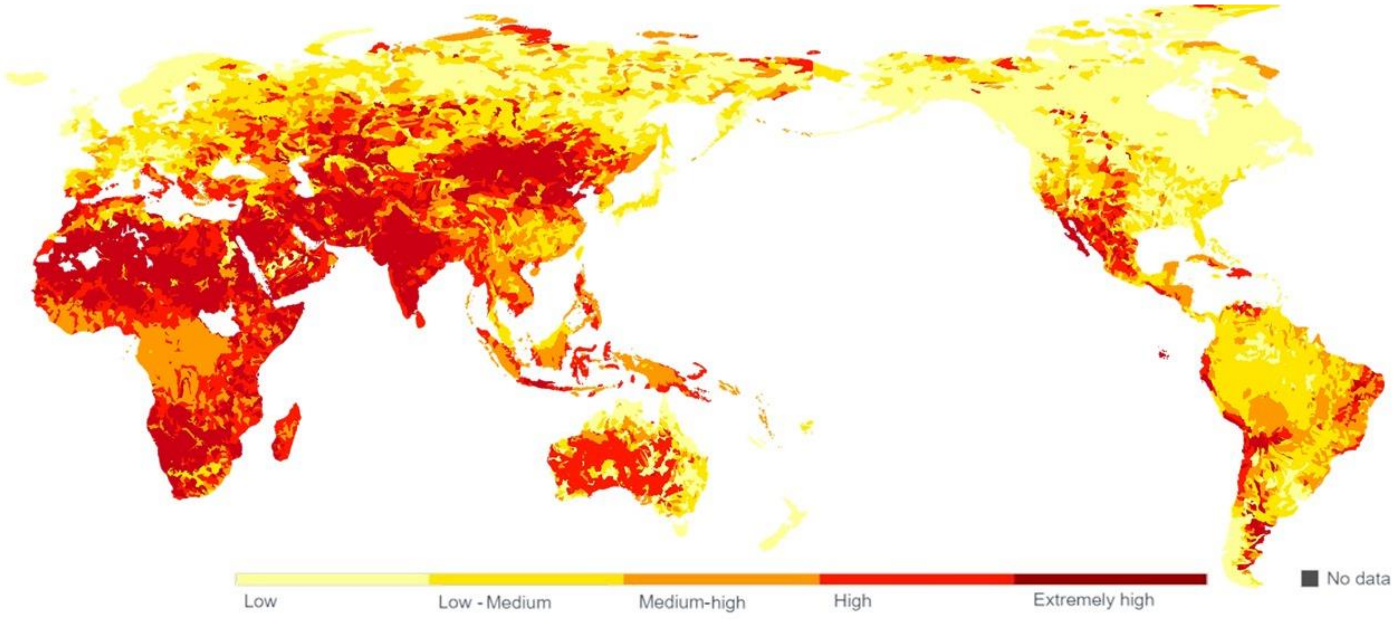

Figure 2.1 Overall worldwide water risk map (Adapted from: Hofste et al., 2019)

Moreover, the impacts of climate change are leading to a decrease in terrestrial water storage (water held in soil, snow, and ice), increasing water scarcity, which could contribute to societal activity disruption (Jingyu et al., 2020; United Nations, 2023; Visser-Quinn et al., 2021). The dwindling supply of freshwater resources has prompted the need for water reuse as a sustainable solution to alleviate the pressure on freshwater sources (Shoushtarian & Negahban-Azar, 2020; Miller, 2006).

Water reuse could be particularly advantageous for arid and rural areas which are commonly more impacted by water stress. In many regions worldwide, recycled water could be a crucial alternative water source, especially for irrigation purposes (Bauer, 2020; Hardy et al., 2015). This possibility and urgency of being an alternative source for irrigation, becomes even more important when understanding that 71.7% of the approximately 4250 km^3 annual water withdrawal from natural water bodies worldwide is attributed to agricultural use (FAO, 2021).

Under this scenario, it is essential to research and establish alternative sources for irrigation, such reclaimed water reuse schemes since they can provide reliable water amounts and contribute to relief water stress in freshwater sources. However, water reuse comes with associated constrains that must be addressed for a successful adoption. There are environmental, economic, and social factors that need to be understood and on which the potential for water reuse needs to be focused on when promoting its adoption (Boretti & Rosa, 2019; Jury & Vaux, 2007; Sharma et al., 2021). In this regard, water reuse can be useful to tackle some key challenges:

A. **Environmental Factors: Depletion and pollution freshwater resources.**
   Reuse promotes water savings and contributes to increase the availability for other purposes like drinking water and supporting ecosystems. Furthermore, it helps prevent pollution by treating and recycling wastewater.

B. **Economic Factors: Cost-effective and energy saving alternatives.**
   Reuse can be a cost-effective option where reclaimed water can be used for different non-drinking purposes (i.e., irrigation, toilet flushing, etc.). This saves money for water providers and users and reduces the need for energy-intensive processes needed to further clean water (i.e., tertiary treatments).

C. **Social Factors: Growing urban populations climate change resilience.**
   Water reuse is an effective way to meet the increasing water needs of cities while lessening the burden on freshwater sources and guaranteeing water security. It helps combat climate change by diversifying water sources and making water more accessible, especially during dry spells or when there is a shortage of freshwater.

While water reuse offers numerous benefits, constraints and challenges are very much present when trying to successfully implement it. In their work, Lee & Jepson (2020) determined that policy, economic, technical, environmental, legal and institutional bottlenecks are key factors affecting water reuse potential. These aspects have also been addressed by the European Commission (2014) through acknowledging that the costs associated with installation, maintenance, operation, and distribution, limit the expansion of water reuse projects. Also, indicating that current technology used in wastewater treatment plants is insufficient for producing potable reuse water, requiring additional infrastructure and advanced treatment methods for broadening reuse options.

Although there are several aspects to consider, it is the economic, technical, and health aspects that are the most rigid and need the most attention for the development of research. This is because water reuse initiatives are generally more constrained by economic factors and they need to be supported by inexpensive technologies in order to develop public policies and social acceptance (Gonzalez-Flo et al., 2023).

2.2 Nature-based solutions for water reuse: Constructed Wetlands

The International Union for Conservation of Nature (IUCN) defines Nature-based Solutions (NbS) "*Actions to protect, sustainably manage and restore natural or modified ecosystems that address societal challenges effectively and adaptively, simultaneously providing human well-being and biodiversity benefits*" (Cohen-Shacham et al., 2016). In other words, NbS are strategies that use nature and natural processes to address various environmental challenges and promote sustainable development, thus providing benefits for both human well-being and biodiversity. They encompass actions aimed at protecting, sustainably managing, and restoring natural or modified ecosystems, green infrastructure, sustainable land management, natural water management, coastal protection, urban greening, among others, that address different societal challenges related to water security, food security, human health, disaster reduction, among others (Cohen-Shacham et al., 2016).

The use of innovative approaches, particularly nature-based solutions like constructed wetlands, is seen as a cost-effective way to implement water reuse initiatives and overcome obstacles in proposing low-cost technologies for addressing water scarcity and promoting sustainability. When it comes to water-based issues, the concept of NbS can be specifically linked to sustainable strategies for managing water resources and improving water quality. With regards to the services that these water-related NbS provide, Taylor et al. (2018) provide the framework to understand how is it that wetlands are amongst the key ecosystems that contribute to water purification. Their mechanism relies on using microorganisms and vegetation to naturally break down waste, remove pathogens, and reduce nutrient and pollution levels (Taylor et al., 2018). In that regard, natural wetlands have been historically used for water treatment and purification, yet today they have undergone significant degradation or destruction over time resulting in more than 64% decline in their coverage around the world (Gardner et al., 2015; Vymazal, 2022). The increasing demand of water supply, treatment and reuse, coupled with the degradation of natural wetlands, have prompted, since the 1980's, the adoption of CW to mimic the purification ability that natural wetlands provide (Vymazal, 2022).

Since then, the use of innovative approaches, particularly CWs, are seen as a cost-effective way to implement water reuse initiatives and overcome obstacles in proposing low-cost technologies for addressing water scarcity and promoting sustainability (Capodaglio et al., 2021; Gonzalez-Flo et al., 2023). Constructed wetlands are one of the most grounded and tested proposals of NbS for water management and treatment (Ferreira et al., 2023; Haddis et al., 2020). They are man-made systems which, in comparison of natural wetlands, offer advantages such as flexibility in design and the ability to control water flow and treatment time (Zhi & Ji, 2012). CWs use various mechanisms to treat wastewater, including biological, physical, and chemical processes that happen when water, substrate, plants, plant debris, and microorganisms are combined together (Masoud et al., 2022). Through processes of sedimentation, filtration, biological degradation, plant uptake to export through biomass harvesting, adsorption-precipitation reactions, ammonification, nitrification, and denitrification, CWs can remove concentrations of suspended solids, organic matter, nitrogen, phosphorus, pathogens, and heavy metals (Masoud et al., 2022).

Currently, CWs are used and recommended as an alternative in contexts where secondary treatment is not feasible (i.e., decentralized water treatment plants) or where there is a risk of sewer overflows, basically because of their easy implementation, simple operation, and low cost (Babatunde, 2022; Gomes Colares & Sandri, 2013; Masi et al., 2023). The removal of pollutants in CWs is achieved through plants that use a range of physical (i.e., sedimentation, filtration, absorption, precipitation, etc.), chemical (i.e., chemical adsorption, complexation, etc.) and biological (i.e., microbial interactions, plant extraction, etc.) mechanisms, as shown in Figure 2.2, to perform the process of phytoremediation, a process that mimics the natural ability in natural wetlands to purify water (Chagas et al., 2011; Tuladhar et al., 2008).

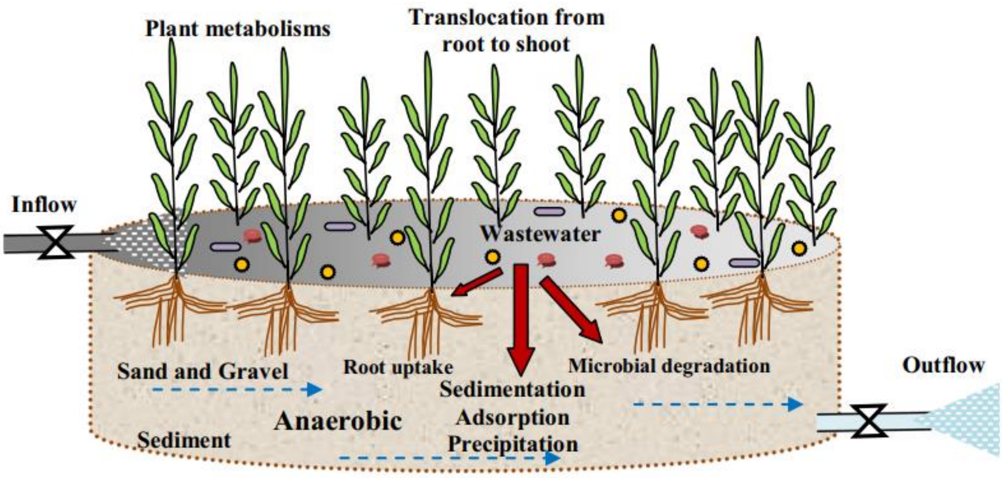

Figure 2.2 Pollutant removal mechanisms from a horizontal subsurface flow CW
(Taken from: Ilyas & Van Hullebusch, 2020. pp. 14348)

Figure 2.3 shows some of the different designs and configurations that CWs can have, from subsurface or surface flows to hybrid systems, each designed to treat wastewater and manage water quality through different processes and configurations. Nevertheless, there are other ways of categorizing CWs depending, for example, on the hydraulic grade line or the type of macrophyte present in the treatment bed (Patro et al., 2023).

The use of CWs provides a less expensive and more sustainable alternative to water regulation compared to conventional water treatment plants (Masoud et al., 2022). Furthermore, the use of CWs as a NbS is crucial to provide sustainable solutions for the improvement of water quality and to increase the resiliency to climate change effects (Taylor et al., 2018).

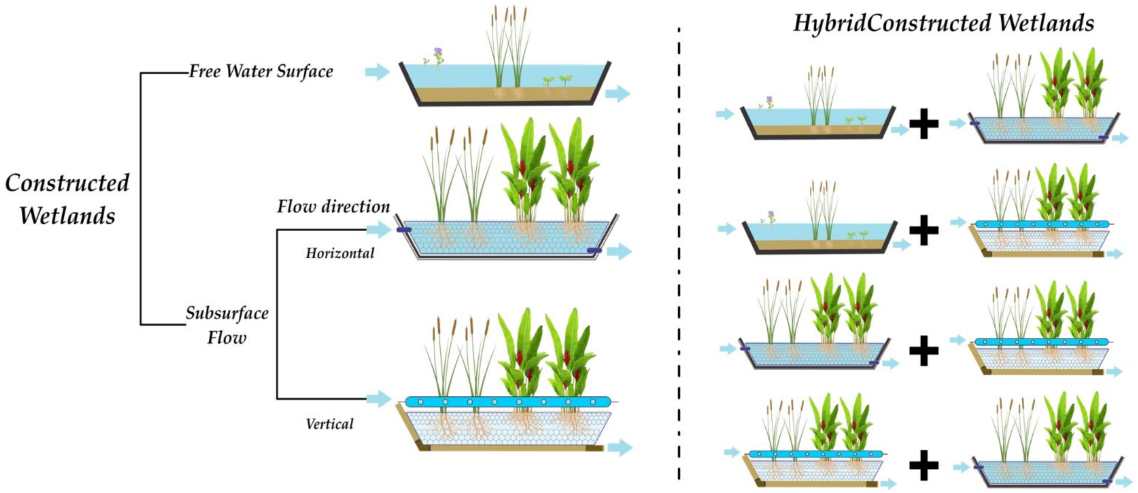

Figure 2.3 Simplistic scheme of different types and configuration of CWs
(Taken from: Marín-Muñiz et al., 2023. pp. 3).

2.3 Constructed wetland performance and effluent's quality

Independently of the inflow water quality, the critical factors determining the performance of a CW are physical design, hydraulic loading rate, local temperature, sunlight exposure, macrophyte community, substrates, and the hydraulic retention time (Hassan et al., 2021; Jingyu et al., 2020; Maiga et al., 2019; Mao & Huang, 2019). The deliberate modulation of these factors in a precise manner facilitates the enhancement of water quality metrics encompassing biological oxygen demand (BOD), turbidity, *Escherichia coli* concentration, ammonium levels, total nitrogen content, total phosphorus content, and total suspended solids (TSS) concentration within a designated wastewater sample (Abi Saab et al., 2018; Biswal & Balasubramanian, 2022).

For example, Song et al. (2019) found that the presence of *Oenanthe javanica*, a native plant from China, can help remove total nitrogen during colder seasons. Tee et al. (2012) showed that the design and hydraulic retention time (HRT) of a filter can affect how well it removes ammonia nitrogren (NH_4^+-N), with removal rates ranging from 55% to 99%. Hassan et al. (2021) reviewed studies on BOD and COD levels and found that they can be reduced by up to 77% and 78%, respectively. Waly et al. (2022) found that for any CW, regardless of wastewater source, filtering media type, or filter size, the contaminant-dependent removal efficiency values fluctuate between 53% and 88%, while the overall treatment efficiency ranges from 67% to 75%.

Despite all the above, CWs are systems that do not always operate in a consistent and sustained manner. This situation makes it more difficult for their respective effluent to comply with strict regulations on the reuse of treated water for, for example, agriculture irrigation (Šereš et al., 2021). Long-term performance and aging result in the accumulation of organic matter, clogging of the substrate, limitations in pollutant removal, changes in vegetation dynamics, and reduced hydraulic conductivity, which are some of the factors that can affect long-term performance (Lopes, 2021; Nuamah et al., 2020). Furthermore, CWs have shown to have seasonal variations that affect the performance during the winter season (Ji et al., 2020; Yan & Xu, 2014). In such cases, additional treatment steps or alternative technologies may be necessary.

2.4 Alternatives to improve the CW’s effluent quality for reuse

In general, CWs are nature-based methods whose design and implementation tend to result in a rigid infrastructure that demands a large amount of area for their construction and are significantly temperature-dependent (Nuamah et al., 2020). For domestic wastewater treatment, commonly, CWs are used as a component of a broader septic system which consists of two main parts: a septic tank and a treatment stage that is typically a subsurface soil absorption system, such as a drain field, constructed wetland, filter, duck pond, or aerobic treatment (von Sperling et al., 2019). Although these system are regarded as cost-effective to treat domestic effluents, Lopes (2021) and Gonzalez-Flo et al. (2023) work showed that for its potential reuse, the effluent quality was still not adequate according to the local water reuse regulations and laws.

Given this scenario, which evidences the limitations of a CWs to produce a reusable effluent, a potential solution is to include an artificial process for a hybrid system that allows to improve the quality for its compliance with regulation. In this regard, researchers have tested methods such as pre-treating receiving waters with aluminum- and iron-based coagulants, micro-electrolysis, activated carbon, among other methods (Bachand et al., 2019; Cui et al., 2022; Hansen et al., 2018; Mosquera-Romero et al., 2023; She et al., 2023).

2.4.1 Coagulation and Flocculation

In particular, Gonzalez-Flo et al. (2023) in their work hinted about the relevance on trying to reduce dissolved organic matter as this might be the cause of high turbidity levels found in their results which led to a non-compliance for reuse. Dissolved organic matter forming colloids in water are difficult to be reduced by ordinary filtration processes and might require a process such as coagulation-flocculation-sedimentation (C/F/S) in order to allow precipitation and posterior removal of organic matter (Scholz, 2016). This process is traditionally used for removing turbidity in drinking water but also proven effective in eliminating various contaminants like metals, toxic organic matter, viruses, and radionuclides that can be adsorbed by colloids (L. K. Wang et al., 2005). Coagulation consists of a rapid mixing to dissolve and disperse chemical coagulants into the water for treatment, whereas flocculation is a gradual mixing process wherein destabilized particles aggregate to form sedimentation-capable clusters (see Figure 2.4) (Letterman & American Water Works Association, 1999).

The fundamental processes underlying particle destabilization that can transpire during coagulation encompass double layer compression, adsorption-mediated charge neutralization, sweep coagulation, and interparticle bridging (Shammas, 2005). The specific process during C/F/S water treatment depends on its turbidity and alkalinity, with rapid destabilization reactions (rapid mixing) occurring within 0.1 to 1 seconds, while slower aggregation (slow mixing) takes place between 3 and 17 seconds (Scholz, 2016). The term "sweep coagulation" pertains to the elimination of substances through the creation of a solid precipitate, like a mantle, and is a process dependent on coagulant dosage and concentration of the particle to be removed (Scholz, 2016).

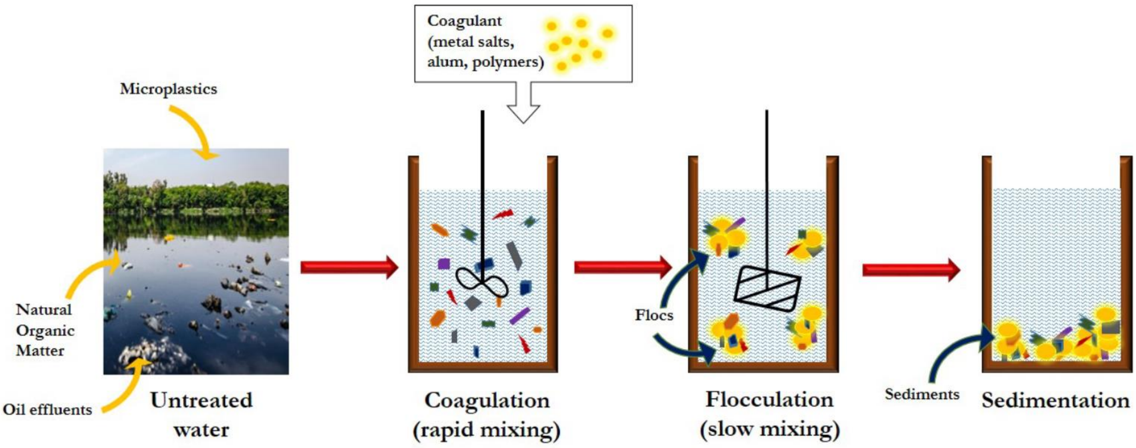

Figure 2.4. Schematic representation of pollutant removal by coagulation, flocculation, and sedimentation processes (Taken from: Pandey et al., 2022. pp. 14)

C/F/S fundamentally works as the negatively charged particles normally found in water are balanced out by the attraction of positively charged reagents such as aluminum species found in coagulants (Scholz, 2016). This interplay between opposing charges in the particles and the adsorbed species eventually corresponds and results in coagulation (Shammas, 2005). The primary coagulants can be categorized into four groups: chemical, non-chemical, synthetic materials, and natural coagulants, with notably, iron salts, aluminum compounds and poly-aluminum chloride (PAC), being the most traditional inorganic coagulants (Nayeri & Mousavi, 2022). Traditional coagulants like aluminum and iron salts are efficient but come with several drawbacks, encompassing potential health risks, and environmental apprehensions (Iwuozor et al., 2023). To avoid damage to the environment, while achieving effluent quality improvement, some researchers have opted for nature-based coagulant agents, such as for example tannin-based ones (Koul et al., 2022; Teixeira et al., 2022).

C/F/S has been previously tested as additional processes within water treatment systems containing CWs (Brunsch et al., 2019; Rahmadyanti et al., 2021). For instance, Figure 2.5 shows the field study set up done by Hansen et al. (2018) in which they used aluminum and iron based coagulants as a pre-treatment before a CW. The image shows two graphs in which the improvement in effluent quality can be clearly observed, however, it also shows how the coagulant is transported beyond this stage, which could pose a risk of contamination.

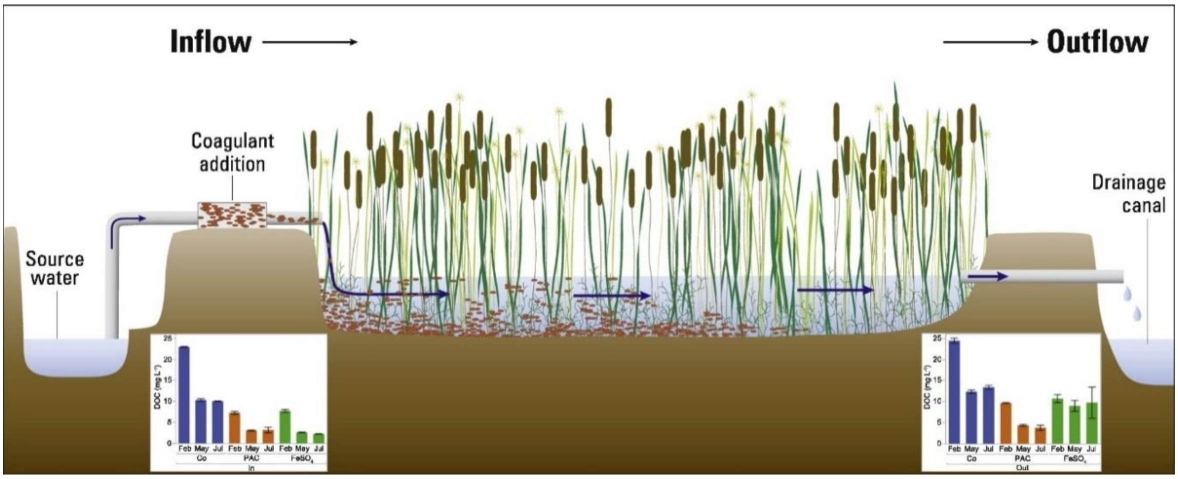

Figure 2.5. Schematic view of the study proposed by Hansen et al., (2018)
(Taken from: Hansen et al., 2018. pp. 605)

The C/F/S benefits, including cost-effectiveness, pollution removal efficiency, and ease of use, are conditional upon experimental variables like pH, coagulant-flocculant dosage, stirring speed, and settling time for its success (Shammas, 2005).

2.4.2 Carbon adsorption and activated carbon

Adsorption is one of the most convenient and versatile water treatment to effectively remove a wide range of pollutants, including organic, inorganic, and biological contaminants, offering potential for source reduction and water reclamation (Ali, 2012). It is a surface phenomenon where pollutants are physically attached to a solid surface through physical forces or weak chemical bonds. One of the most common adsorbents is activated carbon, which is regarded as having superior wastewater treatment capabilities through efficient adsorption, fast kinetics, and simple design (Tan et al., 2008). Activated carbons (ACs) are generally generated through a dual process involving carbonization and activation, where the initial stage establishes primary pore structure and carbon content while the subsequent activation stage, achievable through physical or chemical means, enhances the pore structure, with chemical activation offering advantages like higher yield compared to physical activation (Melliti et al., 2023).

Initially, ACs were being developed from costly processes using coal or lignocellulosic materials that undergo a significant energy consuming and environmentally harmful process (Jones et al., 2021). Today, the demand for ACs, along with stricter environmental regulations, has prompted interest in creating it from more cost-efficient and sustainable sources (Jones et al., 2021; Shukla et al., 2020). This scenario favors the adoption for renewable or recyclable waste as raw materials because of their widespread availability, low cost, ease of sourcing, and positive environmental impact. In this regard, some researchers have developed and tested coconut shell-based activated carbon (CSAC) to observe its potential to clean water (Azabache Liza Yrwin F. et al., 2022; Budianto et al., 2021; Chali & Yakub, 2013; Sujiono et al., 2022; Torres-Lozada et al., 2018). As presented in the work of Sbardella et al. (2018), CSAC can be used effectively as a post treatment to remove pollutants from effluents coming from a previous water treatment process.

2.5 Improving the effluent’s quality in decentralized Constructed Wetlands based WWTP in Aljezur – Case Study

The concept of decentralized WWTPs involves the collection, treatment, disposal, and/or reuse of water in close proximity to its point of generation (Bakir, 2001; Crites & Tchobanoglous, 1998; Estévez et al., 2022). The significance of this approach lies on the fact that it has been regarded as sustainable, cost-effective, and highly suitable for villages with low population density in comparison to conventional water treatment plants for remote human settlements, such as in the case of Aljezur (Hophmayer-Tokich, 2006; Roefs et al., 2017).

The current study has been designed based on the exploratory research approach. Therefore, it aims to observe and analyze results that could indicate potential processes that can complement the constructed wetland based WWTPs in Aljezur. The case study used was defined upon literature and data review, and with the selection of two WWTP as the subject of research. These treatment plants, Carrapateira and Maria Vinagre, were selected because it has been inferred from the literature review that they are the best and worst performing, respectively.

Given the constrains to change any stage in the current WWTPs, the case study considered as feasible alternatives to test a potential additional process of C/F/S or a CSAC filter to observe any potential promising results in the treatment of CW’s effluent. Moreover, to keep the overall treatment as sustainable as possible, a natural coagulant was applied for C/F/S process. The coagulant of choice was one that follows the nature-based principles, as it is a tannin-based coagulant agent called Tanfloc and produced by the company TANAC. Tanfloc is a low molecular weight organic-cationic polymer that functions as a coagulant, flocculant, and coagulation aid for water treatment, exhibiting versatility across applications as evidenced by preliminary tests (jar-tests) (TANAC S.A., 2021). For the filter, coconut shell based activated carbon (CSAC) was used. CSAC has been used before as a substrate to remove pollutants in CWs and it has had positive results (James & Yadav, 2021). However, the potential as a medium filter to improve the water quality of the effluent of a CW has not been found in literature yet.

The aim is that this ecofriendly solution removes the remaining organic matter, solids, turbidity, and *E. coli* from the effluent and, potentially, achieve the quality that would allow its potential irrigation to reuse according to the legislation.

2.5.1 Water stress in Aljezur and water reuse legislation

The south of Portugal, the Algarve, is characterized by an irregular precipitation regime varying from intermittent floods in winter, to inter-annual extreme periods of drought (Fragoso & Tildes Gomes, 2008; Nunes et al., 2006). This region has continuously experienced water scarcity scenarios due to the impacts of climate change, increased irrigation and tourism water demand, and inadequate water management practices (Soares & Lima, 2022). Under this scenario, local authorities have urged the need to find alternative sources or reuse of water to supply for domestic, agricultural and recreational purposes (Rebelo et al., 2020).

This clear need to find new sources of water is aligned with the development of the law decree Nº 119/2019, Portuguese legislation that establishes the legal framework for the "*production of water for reuse, obtained from the treatment of wastewater, as well as its use*" (Decreto-Lei n.o 119/2019, 2019). Before the existence of this law, there were already some initiatives in the Algarve for water reuse in, for example, golf courses, citrus agriculture and even to support specific ecosystems (Rebelo et al., 2020). Currently, the law clearly establishes which effluents are allowed to be reused for specific non-potable uses. This study focuses on the following irrigation reuse categories listed below and detailed in item 7.2 of this document.

Irrigation reuse categories:

A. Irrigation without access restriction: Urban and agricultural use which considers direct human contact with surface exposed to reused water.

B. Irrigation with restricted access: Urban and agricultural use which does not consider human contact with surface exposed to water. Also, irrigation of gardens with restricted access.

C. Irrigation with restricted access: Only agricultural use which does not consider human contact with surface of crop exposed to water, including crops for animal consumption.

D. Irrigation with restricted access: Only agricultural use which for seed production with industrial or energy use.

E. Irrigation with restricted access: Only agricultural use which for seed production, and watering areas with natural restricted use such as terraced meadows, containment areas, hedgerows.

One legally reusable effluent is domestic wastewater, commonly processed in conventional wastewater treatment plants (WWTPs) across the Algarve region. These WWTPs differ in their ability to cater to specific demands, the employed technology, operational and maintenance expenses, and their connection to a wider water distribution and sanitation network. Thus, the current research aims to assess the viability of repurposing effluents from WWTPs in Aljezur, situated within the broader Algarve area.

In Aljezur, the WWTPs are composed of a first stage septic tank and a second stage of a CWs. Septic tanks are desirable in this region because of their simple construction, uncomplicated operation and low implementation costs, characteristics that make them suitable to be implemented in areas without a public sewerage network (Franceschini et al., 2021; Gomes Colares & Sandri, 2013). The study done by Lopes (2021) reviews the performance of five different WWTPs that use horizontal subsurface flow (HSSF) CWs in Aljezur. Table 2.1 shows the different rates at which these CWs remove pollutants.

Despite the observed removal rates in the WWTPs in Aljezur, the treated effluents do not fully meet the quality standards necessary for them to be reused under the Portuguese legislation (see Table 2.1) (Lopes, 2021). Furthermore, mandatory water quality parameters such as *E. coli* have not been assessed previously for these WWTPs in Aljezur with a reuse objective. Therefore, further research and improvements are needed to ensure that the effluents coming from CWs meet the Portuguese water quality standards.

Table 2.1. Water quality parameter removal rates for CW implemented in Aljezur, Portugal (Lopes, 2021).

Parameter	Removal rate (%)	Legislation threshold[1]
Chemical Oxygen Demand (COD)	78.8 - 93.9	-
Biological Oxygen Demand (BOD_5)	89.7 - 97.0	10 – 25 mg/L
Total Suspended Solids (TSS)	64.5 - 97.9	10 – 60 mg/L
Total Nitrogen	19.8 - 48.5	15 mgN/L
Total Phosphorus	6.3 - 46.5	2 – 5 mgP/L

2.5.2 Carrapateira and Maria Vinagre WWTP

The Carrapateira WWTP, in Figure 2.6, was built to treat a maximum of 78 cubic meters of wastewater per day. It serves a population of 500 people and uses a secondary level treatment process. The effluent is discharged at coordinates 508998.11, 4114929.01, which belong to zone 29S in the Universal Transverse Mercator (UTM) system.

Figure 2.6. Google earth's view of the Carrapateira (left) and Maria Vinagre (right) WWTP constructed wetlands.

The Maria Vinagre WWTP, in Figure 2.6, is in Rogil. It was established to initially serve 500 residents as per its design and it can treat up to 100 cubic meters of wastewater per

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[1] Regime for the production of water for reuse from the wastewater treatment, as well as its use - Decreto-Lei n.º 119/2019, 2019).

day. The treated effluent is discharged at coordinates 518875.49, 4139449.68, which correspond to zone 29S in the Universal Transverse Mercator (UTM) system.

3. Methodology

3.1 Sample collection and preparation

Samples were collected at two different sampling campaigns, the first was on April 04, 2023, and the second was on May 09, 2023. To ensure a proper and safe collection, samples were collected in collaboration with *Águas do Algarve, S.A.* staff, at the exit of the treatment as shown in Figure 2.6 (red circles), using a sampler cup (Figure 3.1).

After collection, samples were transported immediately to the laboratory, using containers made of high-density polyethylene (HDPE). These were previously properly decontaminated and sterilized to guarantee a clean environment. The total volume that was sampled was 25 liters for each WWTP in each sampling campaign, thus accounting for a total of 50 liters sampled per WWTP. At the laboratory, samples were immediately properly stored and maintained at proper light and temperature conditions during the development of the work.

Figure 3.1. *Águas do Algarve, S.A.* staff using a sampler cup to collect effluent from Carrapateira (left) and Maria Vinagre (right) WWTP.

For specific tests such as biological oxygen demand (BOD), total nitrogen, total phosphorus, *E. coli* and microorganisms, 1 liter of sample was separated, from each treatment plant, to be analyzed at UAlg Laboratory of Chemical Analyses (LAQ – *Laboratório de Análises Químicas*). All the water quality parameters had at least duplicate and their standard deviation measured.

3.2 Experimental setup and procedure

3.2.1 Coagulation / Flocculation / Sedimentation (C/F/S) experiments

Conducting C/F/S tests involved employing a Jar test setup comprising four glass beakers, each with a capacity of 0.8 L. Within this setup, a Selecta Flocumatic mechanical stirrer with adjustable speed was utilized, as depicted in Figure 3.2. Placed atop each mechanical stirrer with a flat metal paddle, that was lowered to a position that reached two-thirds of the depth of the water column to ensure proper mixing (see Figure 3.2).

The employment of this apparatus facilitates the optimization of various factors such as coagulant dosage, paddle rotation speed, and operational duration for each process phase. By subjecting the samples to agitation via the rotational motion of the paddles, the coagulant reacts with the substances present in the water targeted for treatment, leading to the formation of flocs. After the agitation and mixing process, the paddles are withdrawn from the samples and allowed to settle, enabling the flocs to precipitate and consolidate, as described by Satterfield (2005).

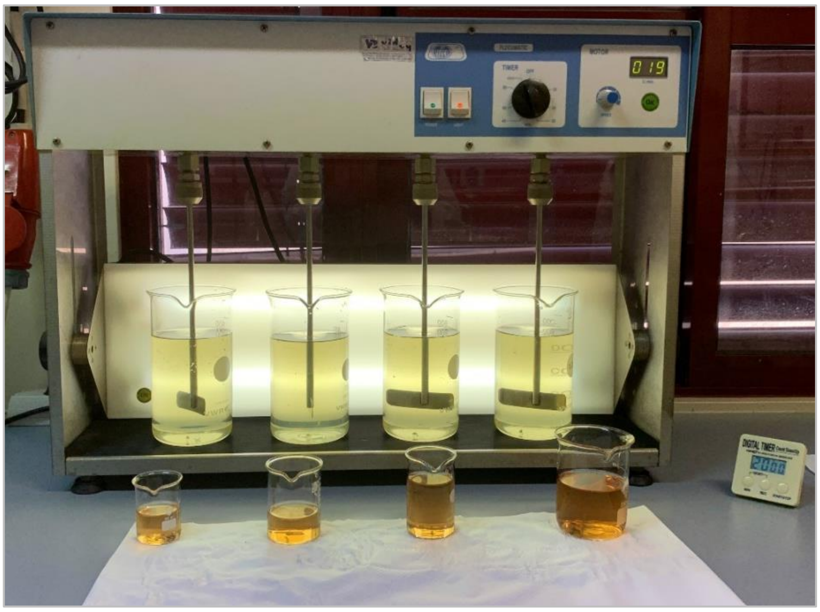

Figure 3.2. Floccumatic system prepared to perform a Jar test with 40, 50, 100 and 200 milligrams per liter of Tanfloc from left o to right.

The experimentation involving the C/F/S jar test was conducted with different concentrations of Tanfloc (1 g/L) to optimize the coagulant dose which concentrations gave the best results. Concentrations tested were 5, 15, 30, 40, 50, 100 and 200 mg/L of Tanfloc. From the Jar-test trials, the optimal coagulant dose was determined for each of the wastewaters studied based on turbidity removal. That is, the optimal dose was the one with which the lowest residual values (highest removal rates) of turbidity were obtained for each wastewater.

The parameters for conducting these jar tests (mixing speed and time) were established considering the standard operating circumstances as outlined in Letterman & AWWA (1999). The operational parameters can be observed in Table 3.1, and the whole C/F/S process took a total of 42 minutes to complete for each given test.

Table 3.1 Common operational parameters for the C/F/S phase and the settings employed for the C/F procedure in the jar tests.

Step/Parameters	Time (min)	Speed gradient G (l/s)	Agitating/Mixing speed (rpm)
Coagulation	2	743	200
Flocculation	20	24	20
Sedimentation	20	-	-

Prior to start the jar test, the water samples (800 mL) were initially placed in the beakers and stirred prior to coagulant addition for approximately 1 minute. Immediately after the coagulant was added, at the desired concentration, the C/F/S procedure started as per the reference of the work of Sousa & Ribau Teixeira (2020), which comprised an initial 2-minute coagulation step, at a velocity gradient (G) of 743 s^{-1} (200 rpm), then a 20-minute flocculation step, at G of 24 s−1 (20 rpm) and a final 20-minute step of sedimentation without any mixing.

After sedimentation (Figure 3.3), an aliquot of 100 mL of the supernatant was sampled using volumetric glass pipette (50 mL) from the mid-depth of the water column. The same C/F/S experiment was carried out with no coagulant added (0 mg/L) as control trial. The optimal coagulant dosage was determined upon the resultant turbidity removal efficiencies.

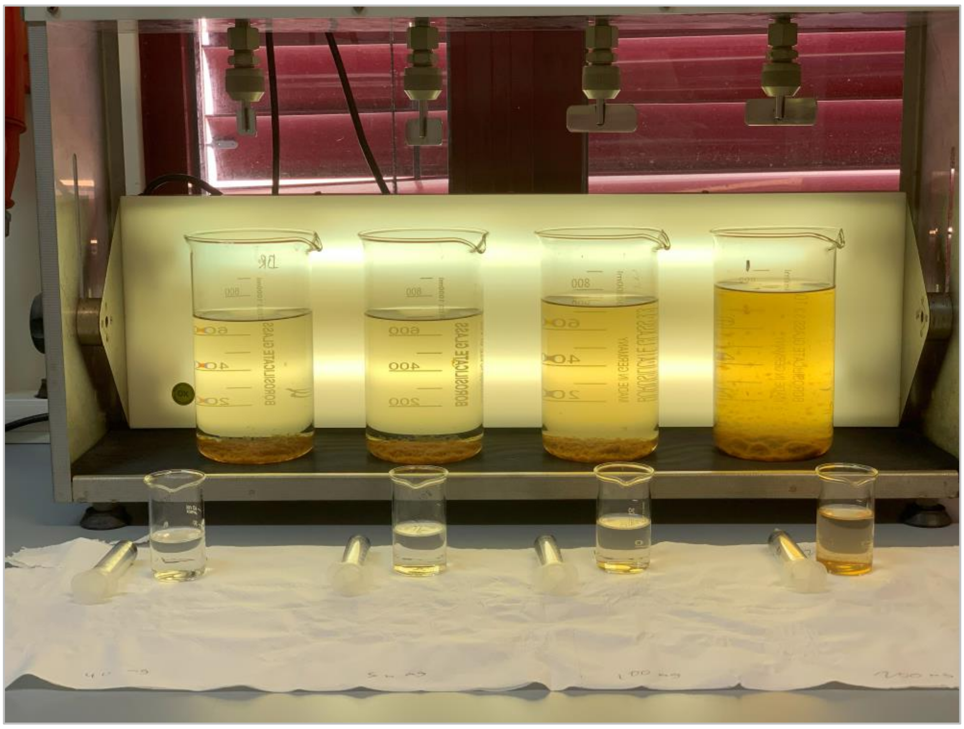

Figure 3.3. Flocumatic system after performing C/F/S process showing the sedimented flocs at the bottom.

After determining the optimal dosage, another set of C/F/S experiments were carried out so continuous measurements and analysis of turbidity (NTU), total organic carbon (TOC), pH, and conductivity (uS/cm), and, specific measurements for BOD_5, TSS, *E. coli*, NH_4^+, N_{TOTAL}, and P_{TOTAL} could be performed for the resultant treated effluents. With this information it is possible to compare the results against the law decree Nº 119/2019.

3.2.2 Carbon adsorption filter with coconut shell-based activated carbon

For the activated carbon adsorption test, coconut shell based activated carbon was used. The design of the experiment consisted of treating the effluent of both Carrapateira and Maria Vinagre WWTPs by allowing it to flow, by gravity, through three different thicknesses of the coconut-shell activated carbon (CSAC).

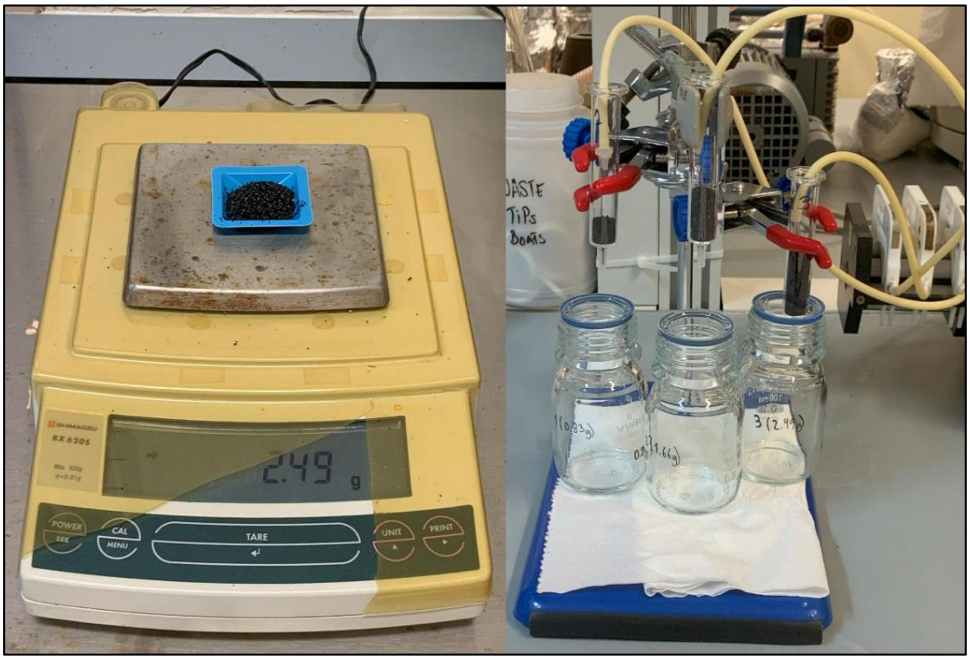

Figure 3.4. Left, the CSAC weighting process and, right, set up of experiment with glass syringes and peristaltic pump.

The characterization of the CSAC is the same as shown in the work of Ribau Teixeira et al. (2017), consisting of a particle size range between 0.25 and 0.30 mm, a surface area measured of 715.5 m²/g, a micropore area of 677.8 m²/g, and a total pore volume is 0.386 cm³/g (Figure 3.4).

The test set up included important equipment like the flow rate peristaltic dosing pump, which fed the 5 ml glass syringes continuously at a steady flow average rate of 2 ml/min of water sample. The CSAC filter experiment consisted of using three different amounts of activated carbon to form a filter through which the effluents from Carrapateira and Maria Vinagre WWTPs flowed through. Figure 3.5 shows the parameters on which each filter was set up for both samples, Maria Vinagre and Carrapateira, represented as the thickness and quantity of CSAC in the filter set up according to the size of the glass syringe. The very small size scale of the CSAC filter makes it possible to quickly see, from an experimental point of view, if a clogging of the medium occurs and allows to quickly measure its effect on the treated water without having to wait long.

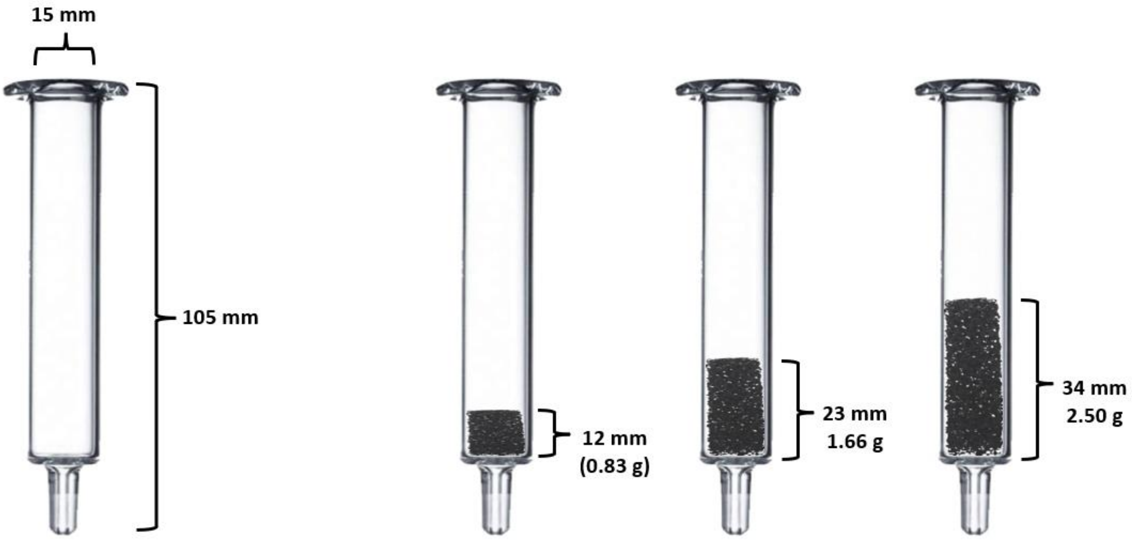

Figure 3.5. Schematic representation of the three thicknesses of CBAC filter

The CSAC-treated water was systematically collected into laboratory beakers (Figure 3.6). This facilitated ongoing monitoring of the resultant turbidity, total organic carbon (TOC), pH and conductivity levels.

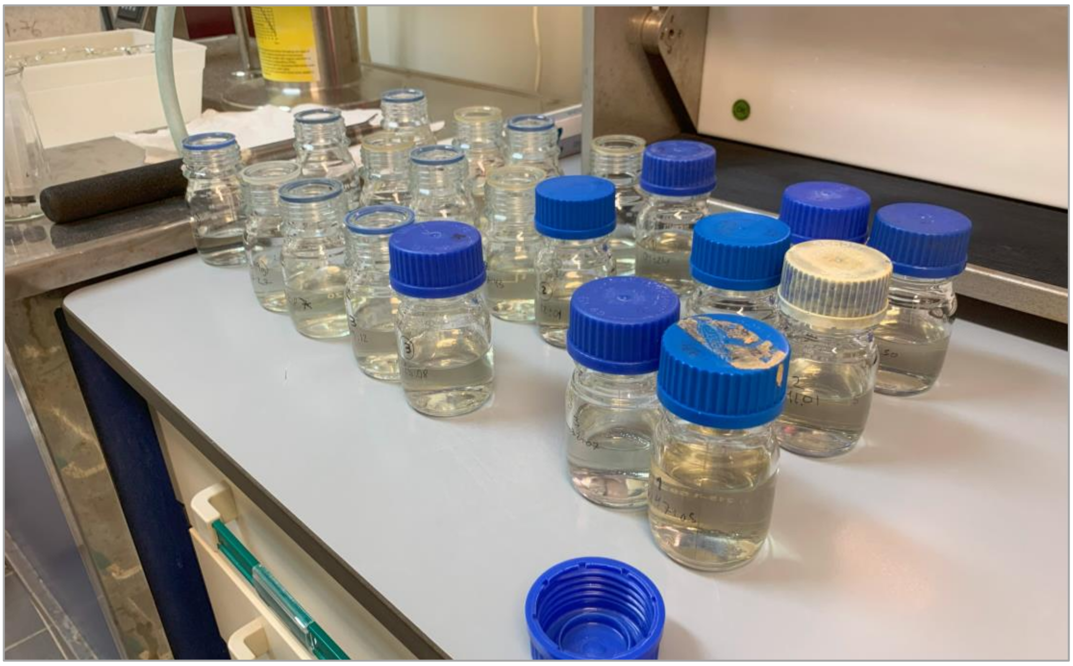

Figure 3.6. CSAC-treated water collected for further analysis.

To ensure consistent results, the filter was first stabilized by running clean water through it. This step ensured the removal of any loose carbon particles and saturated the filter. The stabilization of each filter was determined by the appearance of consistent results that not varied significantly.

3.2.3 Analytical methods

The collected samples underwent a comprehensive analysis that encompassed the continuous measurement of turbidity (NTU), concentration of total organic carbon (TOC) (mg C/L), pH and conductivity (µS/cm). The analyses were done utilizing established assessment protocols as per using Eaton & American Public Health Association (2006) as benchmarks. All materials involved in sample analysis underwent decontamination.

Turbidity quantification involved the application of the nephelometric approach in line with the standard method 2310 Turbidity (Eaton & American Public Health Association, 2006). This procedure was conducted using a HACH turbidimeter, the 2100N model. The device was equipped with a tungsten filament lamp, enabling measurements within a range of 0 to 4000 NTU. Accuracy of ±2% was maintained for readings spanning 0 to 1000 NTU. All dilutions utilized deionized water with turbidity levels lower than 0.5 NTU.

Quantification of TOC drew upon the established method: 5310 Total Organic Carbon (Eaton & American Public Health Association, 2006). To facilitate these measurements, samples underwent acidification (HCl) to pH 1 to 2 was carried out for TOC analysis. The acidification aimed to convert inorganic carbon species (carbonates, bicarbonates, and dissolved CO_2) into CO_2, allowing their removal through volatilization via aeration with synthetic gas. TOC assessment utilized a high-temperature combustion carbon analyzer, specifically the Shimazdu model TOC-5000A. This apparatus operated within a measurement range of 50 ppb to 4000 ppm, ensuring precision of 1%. During analysis, the sample underwent injection into the analyzer's furnace, where the organic carbon underwent transformation into CO_2. Comparison between the CO_2 concentration in the sample and that derived from the calibration curve facilitated quantification. Bi-monthly calibration occurred through the injection of diverse concentrations of standard solution (potassium acid phthalate) — 1000 mg C/L, 1 ppm, 2 ppm, 5 ppm, and 10 ppm.

To determine pH values, the stipulated method 4500-H+ pH was followed (Eaton & American Public Health Association, 2006). This entailed utilizing a Crison pH meter, the Basic 20+ model, at a temperature of 20°C. The pH meter's measurement range spanned from -2 to 16 for pH, and its accuracy stood at 0.01 for pH and 0.2°C for temperature.

Conductivity was assessed following the recognized procedure 2510 conductivity (Eaton & American Public Health Association, 2006). This evaluation took place at a temperature of 25°C using a Crison conductivity meter, specifically the GLP32 model. The meter exhibited a measurement range spanning from 0.01 µS/cm to 199.9 mS/cm, with an accuracy level of up to 0.5%. Calibration, performed monthly, involved two Crison conductivity standards, with values of 147 µS/cm and 1413 µS/cm, both maintained at 25°C.

Furthermore, and to comply with the Portuguese law for reuse of treated water as a reference, it was also sporadically analyzed biological oxygen demand - BOD_5 (mg/L), total suspended solids – TSS (mg/L), *Escherichia coli - E. coli* (CFU/100 mL), total nitrogen - N_{TOTAL} (mg/L), total phosphorus - P_{TOTAL} (mg/L), and ammonium - NH_4^+ (mg/L). NH_4^+-N), provided by Laboratory of Chemical Analyses from University of Algarve.

4. Results and discussions

4.1 Wastewater characterization and quality assessment

Table 4.1 shows the values for the assessed water quality parameters of the treated effluents collected from Carrapateira and Maria Vinagre WWTPs. In the following sections the Carrapateira and Maria Vinagre WWTPs will be referred to as CP and MV respectively. To assess the reusing potential of these wastewaters, the quality parameters were chosen based on legislation, and considering the needs of the Aljezur population, greater attention was directed towards irrigation reuse purposes. According to the Decree-Law No. 119/2019, turbidity, BOD_5, TSS and *E. coli* are mandatory parameters, while NH_4^+, N_{TOTAL} and P_{TOTAL} are facultative. Additionally, TOC, conductivity, and pH, which are not legislated, where also determine since they are important parameters for water treatment and were evaluated during all the experiments. Turbidity, while being of utmost importance for assessing water quality, is also a key factor for treatment performance.

At a first glance, it is important to note that the BOD_5 results fall within the operational parameters of both of these WWTPs, which are set a maximum permissible limit of 40 mg/L, as stated in the study of Lopez (2021). Furthermore, for both CP and MV, the results for BOD_5, TSS, N_{TOTAL}, and P_{TOTAL} indicate consistency with previous findings, because they fall within the range of values obtained also by Lopez (2021) when conducting the wastewater characterization for the same WWTPs.

Table 4.1 Carrapateria and Maria Vinagre wastewater characterization.

Water quality parameter	**Carrapateira (CP)**	**Maria Vinagre (MV)**
Turbidity (NTU)	5.58 ± 0.03	8.00 ± 0.05
BOD_5 (mg/L)	15	30
TSS (mg/L)	6.4	10
E. coli (CFU/100 mL)	45000	18000
NH_4^+ (mg/L)	0.83	0.50
N_{TOTAL} (mg/L)	59.67	76.19
P_{TOTAL} (mg/L)	10.02	8.81
TOC (mg C/L)	11.34 ± 0.16	13.79 ± 0.18
pH	8.05 ± 0.02	8.15 ± 0.01
Conductivity (uS/cm)	1261.00 ± 1.79	1457.33 ± 1.37

Comparing the measured values with the legislated mandatory parameters for irrigation reuse (Table 4.2), one significant finding is that both wastewaters cannot be used for none of the established reuse categories, due to the high concentration of *E. coli*, which indicate the need for further treatment to reduce microbial contamination. The maximum *E. coli.* concentration allowed for any reuse category is below or equal to 10000 colony-forming units (CFU) per 100 ml of water, however, CP and MV show values of 45000 and 18000, respectively.

Analyzing each parameter individually, it is observed that regarding turbidity most irrigation categories do not legislate for it, however, for the irrigation category A (Annex, chapter 7), which is the most demanded reuse the water quality parameter is below or equal to 5 Nephelometric Turbidity Units (NTU). In this regard, both CP and MV turbidity, with values of 5.58 ± 0.03 NTU and 8.00 ± 0.05 NTU respectively, fall above the legal threshold of ≤ 5 NTU, indicating that these effluents require further treatment to meet the category A turbidity standards, which allow reuse for agriculture intended for direct human consumption. These differences in initial turbidity levels between the two wastewater samples can influence the optimal Tanfloc dosage required for effective turbidity removal (Balbinoti et al., 2023).

Table 4.2 Water quality standards for reuse for irrigation

Water reuse categories	Turbidity (NTU)	BOD_5 (mg/L)	TSS (mg/L)	*E. coli* (CFU/100 mL)	NH_4^+ (mg/L)	N_{TOTAL} (mg/L)	P_{TOTAL} (mg/L)	pH
Water reuse categories for irrigation purposes[2]								
Irrigation A.	≤5	≤ 10	≤ 10	≤ 10	≤ 10	≤ 15	≤5	6 - 9
Irrigation B.	-	≤ 25	≤ 35	≤ 100	≤ 10			6 - 9
Irrigation C.	-			≤ 1000	≤ 10			6 - 9
Irrigation D.	-			≤ 10000	≤ 10			6 - 9
Irrigation E.	-	≤ 40	≤ 60	≤ 10000	≤ 10			6 - 9

For BOD_5, CP and MV recorded values of 15 mg/L and 30 mg/L, respectively (Table 4.1). The operational parameters for both plants are set to operate below 40 mg/L in the treated effluent for all reuse categories, and both plants are operating within this range. When compared to the thresholds set by the legislation, CP meets the criteria for all categories except Irrigation A, in contrast, MV only meets the criteria for Irrigation E.

TSS values for CP and MV were 6.4 mg/L and 10 mg/L, respectively (Table 4.1). Both values fall within the permissible limits for most reuse applications, including all categories of irrigation as per the law decree Nº 119/2019 (Table 4.2).

Total Organic Carbon (TOC) values for CP and MV were 11.34 ± 0.16 mgC/L and 13.79 ± 0.18 mg C/L, respectively. Although the law decree Nº 119/2019 does not specify a threshold for TOC, these values indicate the presence of organic matter in the effluent, which could be a concern for certain reuse applications, especially those requiring high purity water.

Conductivity values for CP and MV were 1261.00 ± 1.79 uS/cm and 1457.33 ± 1.37 uS/cm, respectively. Even if the law does not set a specific threshold for conductivity, these values provide an indication of the dissolved salts in the effluent. Higher conductivity values, like that of MV, might pose challenges for reuse applications such as irrigation, where salt accumulation in the soil can be detrimental to crops.

The values for NH_4^+ from both CP and MV are within the permissible facultative threshold limits for all reuse applications, in contrast, the values of N_{TOTAL} and P_{TOTAL} from both CP and MV fall above every legislated reuse category (Table 4.1 vs. 4.2).

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[2] Irrigation categories are described in the appendices and supplementary information section, item 7.

The pH values for CP and MV were 8.05 ± 0.02 and 8.15 ± 0.01 (Table 4.1), respectively, falling within the permissible range of between 6 and 8.5/9 for all reuse applications (Table 4.2).

Table 4.3 shows the comparison of each legislated water quality parameter against its potential reuse, as per the categories established in the legislation, for both CP and MV wastewater characterization results. From this summarized table it can be observed that *E. Coli*, as mentioned previously, is the most limiting factor for the reuse of CP and MV effluents, followed by turbidity. Furthermore, TSS, pH, and NH_4^+ parameters are the least limiting ones for reuse potential.

Table 4.3 Potential reuse for irrigation and/or urban use of Carrapateira and Maria Vinagre WWTP effluents

Water quality parameter	Carrapateira (CP)	Maria Vinagre (MV)
Mandatory water reuse thresholds		
Turbidity	Irrigation B, C, D, and E.	Irrigation B, C, D, and E.
BOD_5	Irrigation B, C, D, and E.	Irrigation E.
TSS	All categories	All categories
E. coli	None.	None.
Facultative water reuse thresholds		
NH_4^+	All categories	All categories
N_{TOTAL}	None.	None.
P_{TOTAL}	None.	None.
pH	All categories	All categories
Overall reuse potential	None.	None.

For further treatments and research parameters of turbidity and *E. coli* concentrations need improvement. It is essential to consider the specific reuse application and its requirements when evaluating the suitability of the effluent. The comparison with the legal thresholds provides a clear roadmap for potential treatment enhancements to expand the scope of reuse applications for the effluents from both plants.

4.2 C/F/S experiments using natural coagulant

4.2.1 C/F/S jar-test trials

In this section, the results obtained from the initial tests to determine the optimum concentration of Tanfloc coagulant were analyzed.

A. Turbidity

The results for both, CP and MV, wastewater treatment plants effluent's reveal that Tanfloc impacted on turbidity, with the degree of reduction varying across different concentrations. Figures 4.1 and 4.2 show the results and removal rates obtained for treating both CP and MV effluent's samples for each of the given Tanfloc tested concentrations. Also, and to visually assess the potential of the tannin-based coagulant to achieve the only legal threshold given for turbidity, a dashed black line is shown at the value of 5 NTU.

For the CP WWTP, the 0 mg/L concentration, even though it did not consider any coagulant, showed a removal of 46.93 ± 0.29 % (Figure 4.2), decreasing from an initial 5.58 ± 0.03 NTU to 2.96 ± 0.82 NTU (Figure 4.1). These values at the control concentration of 0 mg/L of Tanfloc are consistent with previous studies as Grehs et al. (2019) observed a turbidity removal of roughly 40% in their work with no addition (0 mg/L) of Tanfloc. The results showed a consecutive and consistent reduction in turbidity values from 5 mg/L (4.63 ± 0.94 NTU) until 50 mg/L (0.90 ± 0.03 NTU) of Tanfloc. After this range, turbidity values started to slowly pick up from 100 mg/L (2.98 ± 0.06 NTU) until 200 mg/L (15.35 ± 0.86 NTU) of coagulant (Figure 4.1).

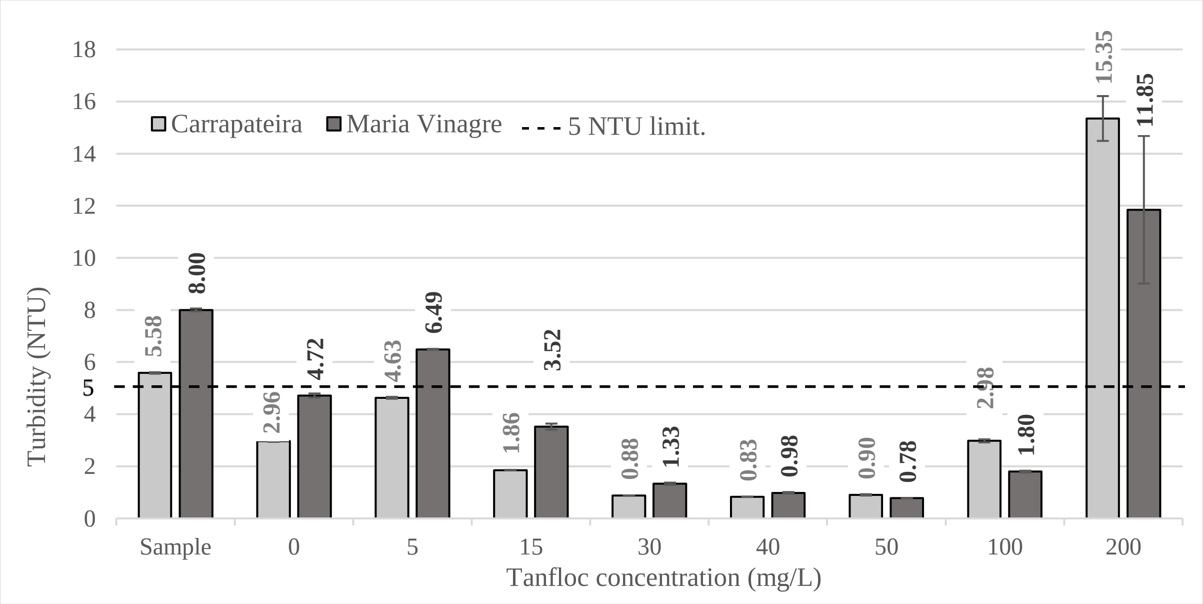

Figure 4.1 CP and MV WWTPs turbidity C/F/S results.

The most consistent turbidity reduction happened at concentrations of 30 mg/L, 40 mg/L, and 50 mg/L, with turbidity decreasing at removal rates of 84.27 ± 0.41 %, 85.14 ± 0.34 %, and 83.81 ± 1.24 %, indicating effective particle coagulation and removal. These findings are consistent with the studies by Beltrán-Heredia and Sánchez-Martín (2009) and Hameed et al. (2016), which found that their optimal Tanfloc concentration for turbidity removal was within the range of 30 to 50 mg/L, precisely 40 mg/L and 35 mg/L respectively. Additionally, Beltrán-Heredia and Sánchez-Martín (2009) calculated a removal rate of 80% at 40 mg/L of Tancfloc, very similar to the 85.14 ± 0.34 % observed for the 40 mg/L concentration in this study, demonstrating the effectiveness of this coagulant/flocculant. The concentration of 40 mg/L was the one that yielded the highest removal rate and improved the water quality, at least for turbidity thresholds, to comply for all irrigation reuse categories.

For concentrations of 100 mg/L and 200 mg/L an increase in turbidity values was observed. These values are very much in line with the Hameed et al. (2016) observations where concentrations above 70 mg/L of Tanfloc reduce the efficiency of coagulation/flocculation because of overdosing. Furthermore, Grehs et al. (2019) found that Tanfloc concentrations above 80 mg/L did not generate flocs. This situation could be explained as Balbinoti et al. (2023) states that overdosing can cause a reversal of charges, leading to particle repulsion, which in turn inhibits the formation of flocs. This last situation is clearly seen at the 200 mg/L Tanfloc concentration, where NTU values did not have any removal capacity of turbidity, in contrast it increased going from an initial 5.58 ± 0.03 to 15.35 ± 0.86 NTU.

For the Maria Vinagre WWTP, shown in Figure 4.2, the 0 mg/L Tanfloc concentration exhibited turbidity removal of 41.09 ± 3.14 %. As mentioned previously, this removal for an untreated control concentration was very similarly observed in the study of Grehs et al. (2019), being consistent with previous studies. The initial turbidity of 8.00 ± 0.05 NTU decreased to 4.72 ± 0.25 NTU, (Figure 4.1), and at 5 mg/L Tanfloc concentration, the turbidity removal was 18.93 ± 6.52 % (Figure 4.2), with turbidity decreasing from 8.00 ± 0.05 NTU to 6.49 ± 0.52 NTU (Figure 4.1).

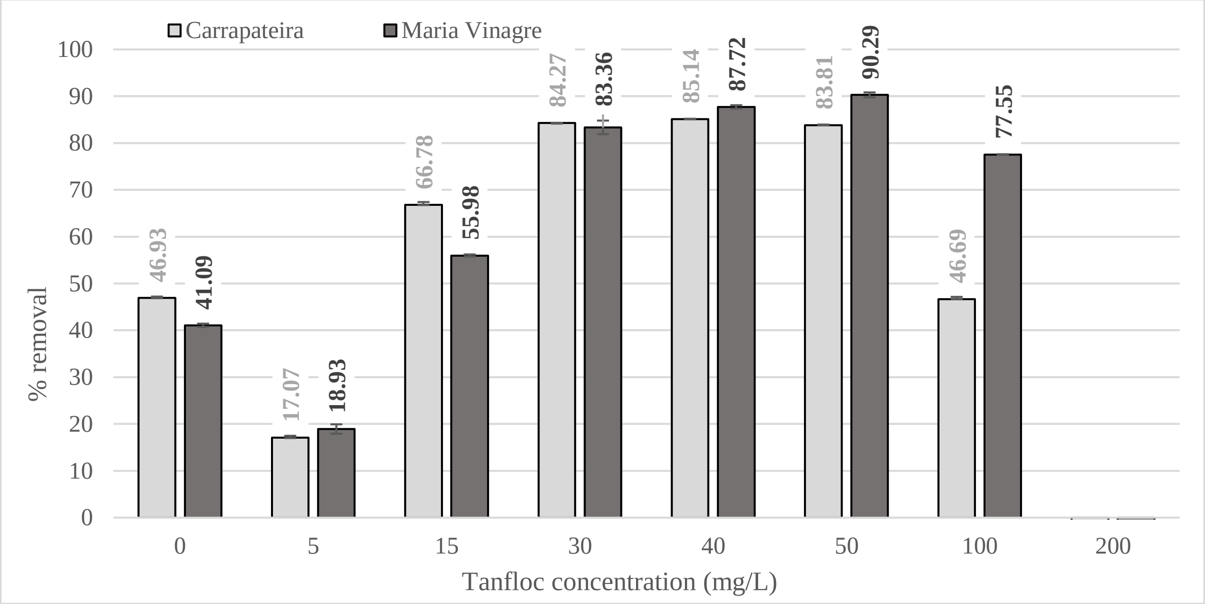

Figure 4.2 CP and MV WWTPs turbidity C/F/S removal rates

A more significant turbidity reduction was observed at concentrations of 15 mg/L (55.98 ± 1.61 %) and 30 mg/L (83.36 ± 1.86 %) (Figure 4.2). Here, turbidity values decreased from 8.00 ± 0.05 NTU to 3.52 ± 0.13 NTU, and 1.33 ± 0.15 NTU, respectively (Figure 4.1). Further improvement in turbidity removal was seen at 40 mg/L (87.72 ± 1.12 %) and 50 mg/L (90.29 ± 0.48 %, Figure 4.2) Tanfloc concentrations, where turbidity decreased to 0.98 ± 0.09 NTU and 0.78 ± 0.04 NTU, respectively (Figure 4.1). These concentrations exhibited the most consistent and efficient turbidity reduction, aligning with the optimal coagulant concentration range observed in previous studies (Beltrán-Heredia & Sánchez-Martín, 2009; Grehs et al., 2019; Hameed et al., 2016). It is worth noting that at higher Tanfloc concentrations of 100 mg/L and 200 mg/L, a reversal in the trend was observed, where turbidity values increased significantly from 0.78 ± 0.01 NTU at 50 mg/L to 1.80 ± 0.03 NTU and 11.85 ± 2.83 NTU, respectively for 100 mg/L and 200 mg/L. These findings are consistent with the notion that overdosing of Tanfloc can inhibit flocculation and even lead to particle repulsion, as discussed in Balbinoti et al. (2023), which may explain the reduction in coagulation efficiency at these concentrations.

The graph done by Hameed et al. (2016), and depicted in Figure 4.3, allows to observe the consistency of the current study’s results with their study, where they found that Tanfloc was also most efficient within similar concentrations. The results obtained from both wastewater treatment plants effluents provide valuable insights into the effectiveness of Tanfloc in achieving the legislated turbidity threshold of below or equal to 5 NTU for irrigation A reuse category. These findings are promising as they align with the legal threshold, indicating that the application of Tanfloc at these concentrations could allow these effluents to meet the most turbidity-demanding water quality standards for reuse. Conversely, the results also emphasize the importance of avoiding overdosing, as concentrations of 100 mg/L and 200 mg/L at the Carrapateira WWTP led to a significant increase in turbidity, exceeding the only legal threshold for turbidity (Table 4.2). This outcome underscores the need for careful dosage control to prevent inefficient coagulation and potential water quality issues.

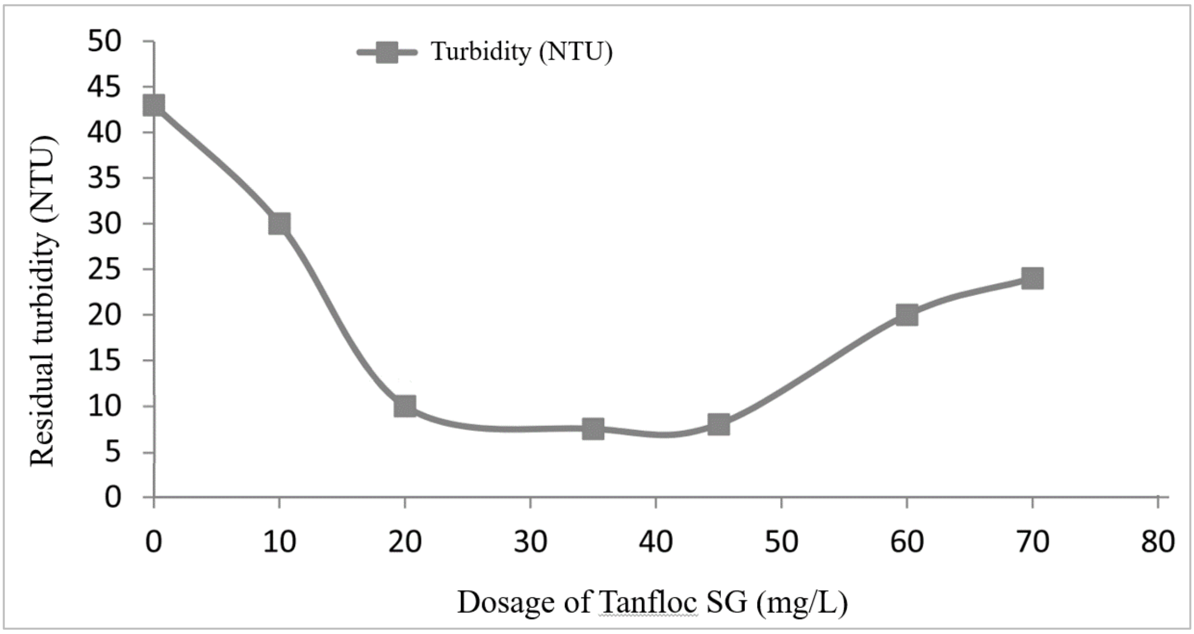

Figure 4.3 Effect of Tanfloc dosage on residual turbidity
(Adapted from: Hameed et al., 2016. pp. 502)

It is evident that Tanfloc concentrations within the range of 30 to 50 mg/L consistently achieve efficient turbidity reduction, with removal rates exceeding 80%. For the CP WWTP, the most effective concentration of Tanfloc was 40 mg/L (85.14 ± 0.06 % removal rate), while for the MV WWTP, the most effective was the concentration of 50 mg/L (90.29 ± 0.48 % removal rate).

B. Total organic carbon (TOC)

For both CP and MV WWTP effluent samples, as shown in Figure 4.4, the TOC concentrations exhibited variations relative to the water sample. For the CP WWTP, from the 0 mg/L to the 50 mg/L Tanfloc concentrations results stayed relatively stable when compared to the sample value, ranging from 9.86 ± 0.11 mg/L to 11.17 ± 1.33 mg/L. The respective removal rates for these concentrations, showed in Figure 4.5, were 13.03 ± 0.94 % and 1.48 ± 1.14 %, not showing a clear trend, therefore indicating that Tanfloc does not consistently remove TOC within this range of concentration.

In contrast, at higher Tanfloc concentration of 200 mg/L, there was a significant increase in TOC values, therefore not yieldeing any removal. These findings underscore the importance of optimizing Tanfloc dosages to avoid overdoing and for achieving optimal TOC removal. These results are consistent with the results obtained by Grehs et al. (2019), Hameed et al. (2016) and the reasoning behind Balbinoti et al. (2023), where high concentrations of nature-based coagulants, such as Tanfloc, can prevent floc formation, therefore rather increasing than decreasing TOC.

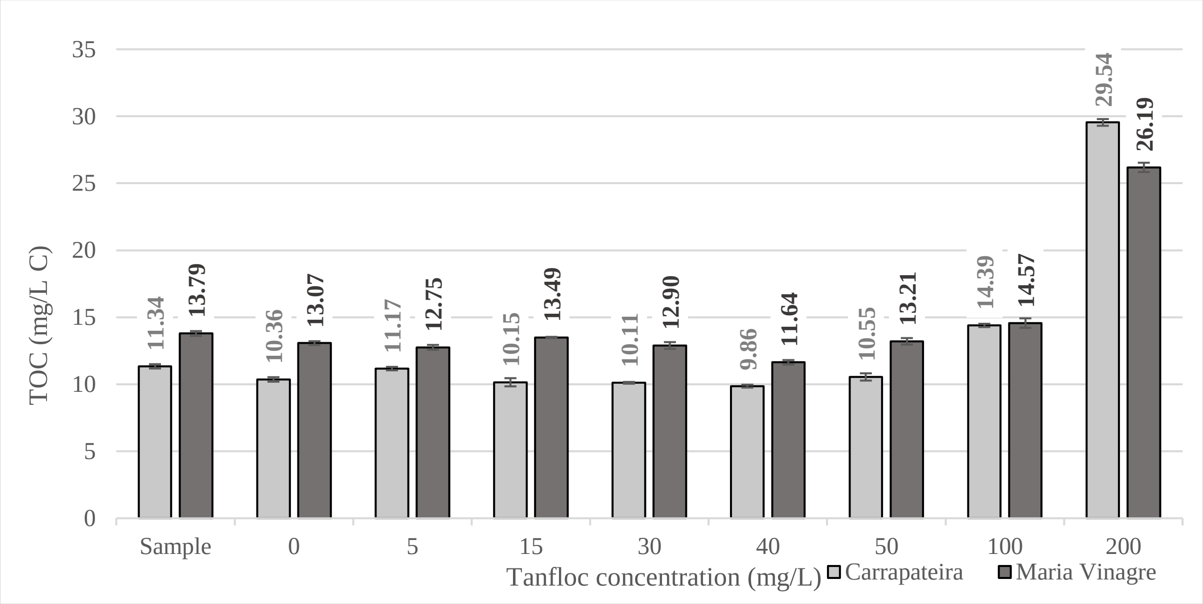

Figure 4.4 CP and MV WWTPs total organic carbon (TOC) post - C/F/S results

The lowest concentration of TOC, both for CP and MV, was observed at 40 mg/L of Tanfloc dosage, where values of 9.86 ± 0.11 mg/L and 11.64 ± 0.17 mg/L were obtained (Figure 4.4). Furthermore, the highest TOC removal rate was observed at 40 mg/L of Tanfloc concentration, with values of 13.03 ± 0.58 % and 15.57 ± 1.21 % respectively. Grehs et al. (2019) revealed a capacity of Tanfloc to remove TOC approximate of 36%. The results of the current study are below the values obtained by Grehs et al. (2019). However, it is important to note that the primary purpose of C/F/S in water treatment is to remove suspended solids and improve water clarity rather than specifically targeting TOC reduction (Shammas, 2005).

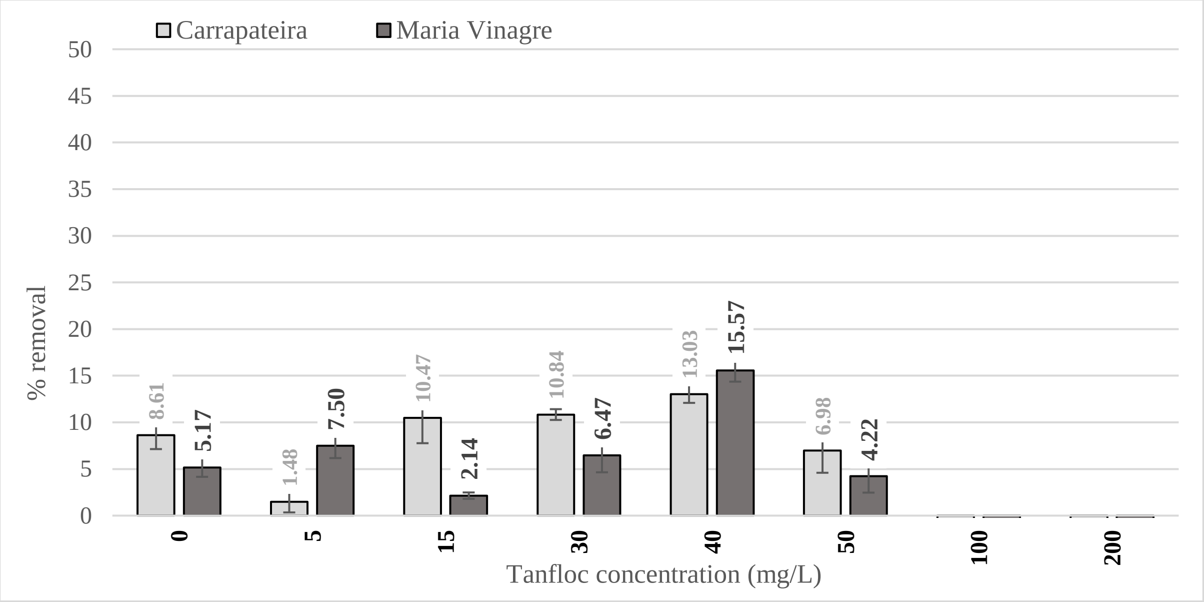

Figure 4.5 CP and MV WWTPs TOC C/F/S removal rates

Previous studies have suggested that nature-based coagulants must be considered carefully as they tend to not show significant removals of TOC, and in come cases increase the concentrations (Balbinoti et al., 2023; Beltrán-Heredia et al., 2012; Leite et al., 2019). Regarding these suggestions, the results from this current study show that Tanfloc concentrations between 0 and 50 mg/L did not increase TOC concentrations, rather, in many cases, TOC concentrations were even reduced. These findings could have implications for future research and applications in water treatment, offering a potentially more efficient and sustainable alternative.

In contrast, for both WWTPs, Tanfloc concentrations of 100 mg/L and 200 mg/L showed a significant increase in TOC levels at rates reaching 160.59 ± 2.20 % for the Carrapateira 200 mg/L concentration.

C. pH and Electrical conductivity

The results for both, Carrapateira and Maria Vinagre, wastewater treatment plants effluent’s reveal that Tanfloc had a minor effect on pH values. Figure 4.6 shows the results obtained for treating both effluent’s samples. To visually assess the potential of Tanfloc to maintain law – abiding pH values, the legal range threshold for pH (6.0 – 9.0) is shown with a black dashed – line rectangle in Figure 4.6.

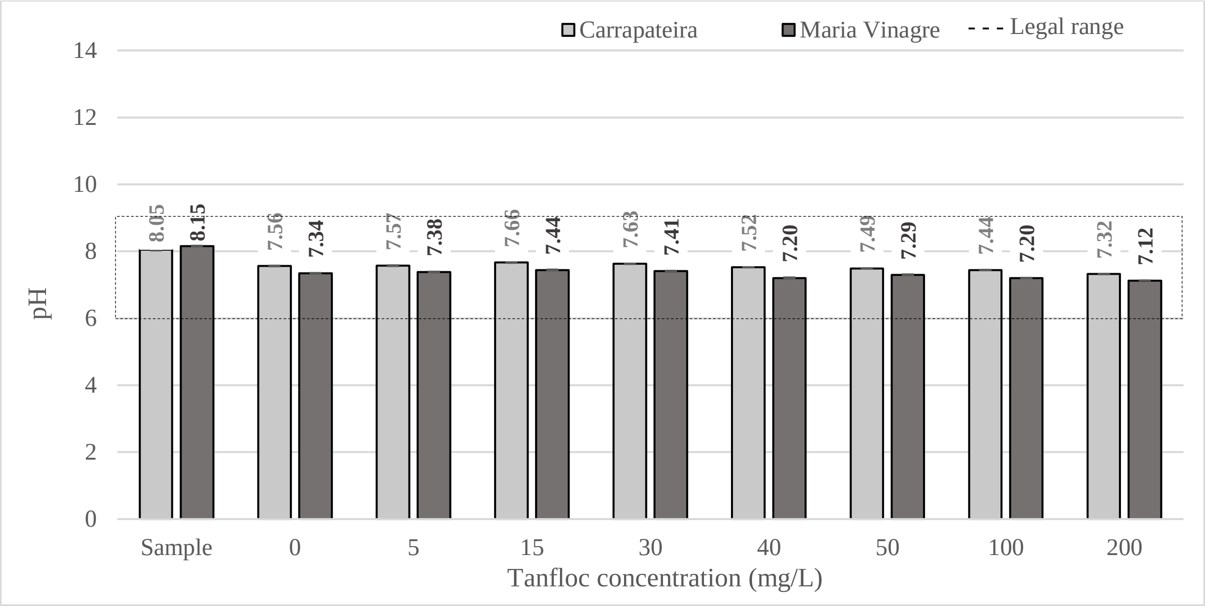

Figure 4.6 CP and MV WWTPs pH post - C/F/S results

Figure 4.6 shows that at Carrapateira, that for both water treatment plants, the obtained results revealing minimal variation in pH levels in response to varying Tanfloc concentrations are in accordance with established findings in the field of natural coagulants for water treatment. These results are consistent with Grehs et al. (2019), where it was also revealed that Tanfloc does not showed any significant effect in pH levels. This consistent pH behavior can be attributed to the inherent characteristics of Tanfloc, a nature-based coagulant, such as those derived from tannins (Balbinoti et al., 2023; Camacho et al., 2017; Leiviskä & Santos, 2023).

The observed pH stability is an advantageous feature in water treatment processes, ensuring that the pH of the treated water remains within an acceptable range while achieving effective turbidity removal through coagulation. In that regard, all the results, were within the legal threshold range (6.0 to 9.0), consequently, the present results affirm the suitability of Tanfloc as a coagulant for maintaining pH stability with the objective of reuse according to Portuguese legislation.

The examination of the impact of Tanfloc on conductivity in the effluents of both Carrapateira and Maria Vinagre are presented in Figure 4.7. It is important to note that Tanfloc concentrations had a discernible influence on conductivity values, but these changes were relatively minor. For the Carrapateira WWTP, at the baseline Tanfloc concentration of 0 mg/L, the conductivity measurement stood at 1317.67 ± 2.94 uS/cm, then, as Tanfloc concentrations increased, conductivity values exhibited subtle fluctuations. For instance, at 5 mg/L Tanfloc concentration, conductivity increased slightly to 1297.50 ± 6.89 uS/cm, indicating a minimal increase in conductivity. The subsequent concentrations, ranging from 15 to 50 mg/L, showed conductivity values oscillating within a range of 1313.17 ± 20.31 uS/cm to 1219.67 ± 53.69 uS/cm. Notably, at Tanfloc concentrations of 100 mg/L and 200 mg/L there was a trend of decrease in conductivity, with values of 1139.00 ± 79.98 uS/cm and 1149.83 ± 21.05 uS/cm, respectively. However, even at these higher concentrations, the changes in conductivity remained relatively low, suggesting that the impact of Tanfloc on conductivity is limited.

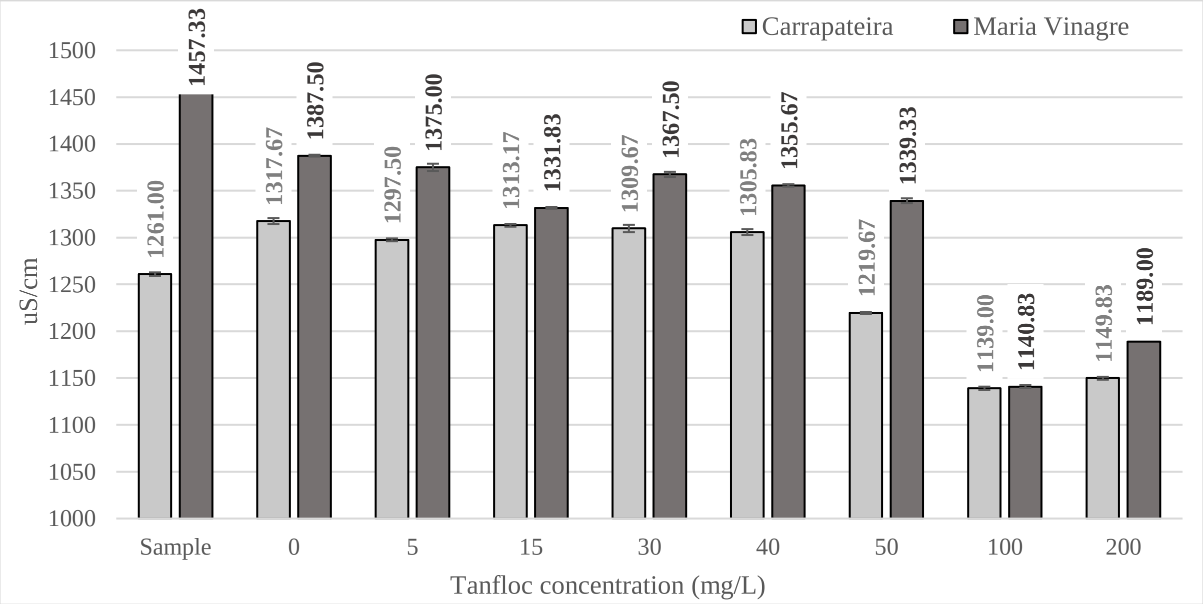

Figure 4.7 CP and MV WWTPs conductivity post - C/F/S results

In the case of Maria Vinagre, the impact of Tanfloc on conductivity was not significant, yet it showed a trend to reduce it when the concentration of Tanfloc increased. Conductivity for Maria Vinagre had a decreasing trend, because at 0 mg/L, the measurement was 1387.50 ± 3.02 uS/cm, at 5 mg/L Tanfloc concentration conductivity decreased to 1375.00 ± 4.47 uS/cm, and this trend continued as concentrations increased to 50 mg/L, where conductivity reached 1339.33 ± 2.25 uS/cm. At Tanfloc concentrations of 100 mg/L and 200 mg/L, a significant decrease in conductivity was observed compared to the initial sample, with values of 1140.83 ± 29.13 uS/cm and 1189.00 ± 28.52 uS/cm, respectively.

At both Carrapateira and Maria Vinagre wastewater treatment plants, Tanfloc concentrations of 100 mg/L and 200 mg/L resulted in slightly more significant impacts on conductivity. These results underscore the importance of considering Tanfloc concentration carefully in water treatment applications, as extremely high concentrations may lead to a substantial reduction in conductivity. Furthermore, the findings for both wastewater treatment plants reveal minor or not significant changes to electrical conductivity of water after the treatment. This slight effect is also supported by Camacho et al. (2017) work on which they noted that a nature-based coagulant, regarding of the dosage, does not significantly change the conductivity. More specifically, Leiviskä and Santos (2023) further supported these findings by highlighting that tannin-based coagulants have a low impact on conductivity of treated water. Therefore, this study's results reaffirm the notion that nature-based coagulants, such as Tanfloc, exhibit the capacity to uphold the conductivity of treated water within a restricted range with minor or insignificant variations in both Carrapateira and Maria Vinagre wastewater treatment plants.

4.2.2 Tanfloc optimal dosage effectiveness in C/F/S experiment

After the initial C/F/S jar-test trials with Tanfloc, the optimum concentration for each WWTP was determined according to its turbidity removal rate. Thus, for the best Tanfloc concentrations, for Carrapateira 40 mg/L and for Maria Vinagre 50 mg/L a C/F/S experiment was repeated to measure a full characterization of the treated wastewater to evaluate the complying with the water reuse legislation. Table 4.4 presents a summary of the results values and removal rates of all the measurements made post - C/F/S treatment for the optimal dosages of Tanfloc. The same information is shown in Table 4.5 for the Maria Vinagre WWTP.

Table 4.4 Carrapateira Tanfloc 40 & 50 mg/L post – C/F/S results.

Water quality parameter	Carrapateira sample	Tanfloc 40 and 50 mg/L treated wastewater quality and removal rates			
		Values 40 mg/L	% removal	Values 50 mg/L	% removal
Turbidity (NTU)	5.58±0.03	0.90±0.06	84±1.11%	0.88±0.07	84±1.17%
BOD_5 (mg/L)	15	10	33%	20	0%
TSS (mg/L)	6.4	2.4	63%	1.6	75%
E. coli (CFU/100 mL)	45000	1800	96%	1000	98%
NH_4^+ (mg/L)	0.83	0.74	11%	0.63	24%
N_{TOTAL} (mg/L)	59.67	66.39	0%	74.79	0%
P_{TOTAL} (mg/L)	10.02	5.32	47%	10.46	0%
TOC (mg C/L)	11.34±0.16	10.60±0.10	-	11.11±0.45	-
pH	8.05±0.02	7.76±0.21	-	8.09±0.19	-
Conductivity (uS/cm)	1261.00±1.79	1137.33±24.22	-	1223.00±5.55	-

With this information, the following items describe and analyze the results obtained for all legislated parameters after having tested these two concentrations in both treatment plants. The results in Table 4.4, precisely for turbidity and *E. Coli*, have reduced considerably for both WWTPs.

Table 4.5 Maria Vinagre Tanfloc 40 & 50 mg/L post – C/F/S results

Water quality parameter	Maria Vinagre sample	Tanfloc 40 and 50mg/L treated wastewater quality and removal rates			
		Values 40 mg/L	% removal	Values 50 mg/L	% removal
Turbidity (NTU)	8.00±0.05	1.07±0.08	87±1.03%	0.91±0.06	89±0.76%
BOD_5 (mg/L)	30	25	17%	20	33%
TSS (mg/L)	10	2.8	72%	2.4	76%
E. coli (CFU/100 mL)	18000	700	96%	100	99%
NH_4^+ (mg/L)	0.5	0.49	2%	0.47	6%
N_{TOTAL} (mg/L)	76.19	53.78	29%	63.87	16%
P_{TOTAL} (mg/L)	8.81	5.51	37%	8.35	5%
TOC (mg C/L)	13.79±0.18	13.67±0.79	-	13.41±0.27	-
pH	8.15±0.01	8.23±0.03	-	8.24±0.06	-
Conductivity (uS/cm)	1457.33±1.37	1399.83±10.80	-	1402.83±2.48	-

A. Turbidity

For CP, the application of Tanfloct concentrations of 40 mg/L and 50 mg/L, reduced the turbidity reduced to 0.90 ± 0.06 NTU and 0.88 ± 0.07 NTU respectively, achieving removal rates of 84 ± 1.11 % and 84 ± 1.17 %. In the case of MV the same concentrations decreased turbidity to 1.07 ± 0.08 NTU and 0.91 ± 0.06 NTU with Tanfloc, achieving removal rates of 87 ± 1.03 % and 89 ± 0.76 %. Both CP and MV effluents, whether they were treated with 40 mg/L or 50 mg/L, resulted in treated water that meets the only legal turbidity threshold of ≤ 5 NTU, meaning their compliance for all irrigation water reuse categories. Furthermore, these findings are consistent with the work of Grehs et al. (2019), where they obtained turbidity removal rates of approximately 90% when using an optimal dosage of Tanfloc ranging from 50 mg/L to 80 mg/L.

B. Total organic carbon (TOC)

Total organic carbon in CP reduced slightly with Tanfloc treatment, reducing to 10.60 ± 0.10 mg/L and 11.11 ± 0.45 mg/L from an initial value 11.34 ± 0.16 mg/L. For the MV WWTP, the values obtained for 40 mg/L and 50 mg/L respectively were 13.67 ± 0.79 mg/L and 13.41 ± 0.27 mg/L from an initial value 13.79 ± 0.18 mg/L. The results for this C/F/S experiment repetition are below the values obtained during the previous C/F/S jar-test trials. However, they still yield removal of TOC giving insights into the potential of Tanfloc. For both

CP and MV WWTP effluent, the highest TOC removal was observed at 40 mg/L and at 50 mg/L of Tanfloc concentration respectively.

Similarly, as in the C/F/S jar test trials, values were consistent and not significant increase or variation of TOC was observed. This situation confirms again the innovative finding where TOC is not affected by Tanfloc concentration ranging from 0 to 50 mg/L.

C. pH and conductivity

The pH and conductivity of both CP and MV remained stable whether they were treated with 40 or 50 mg/L of Tanfloc. These results are consistent with the values obtained by Grehs et al. (2019) and Camacho et al. (2017), where it was also revealed that Tanfloc does not show any significant effect in pH and conductivity levels. Specifically for pH, which is legislated, both CP and MV treated effluents remained within the legal threshold range of 6 to 8.5/9, therefore meeting all water reuse categories pH requirements.

D. Biological oxygen demand (BOD_5)

For the Carrapateira WWTP, the initial BOD_5 was of 15 mg/L, and it reduced to 10 mg/L after treatment with 40 mg/L Tanfloc, achieving a removal rate of 33.33%. However, at 50 mg/L of Tanfloc, the BOD_5 increased again to 20 mg/L. These results are consistent with the results obtained by Beltrán-Heredia & Sánchez-Martín (2009) shown in the Figure 4.8. It can be inferred from the figure that peak BOD_5 removal is at about ≅ 40 mg/L of Tanfloc with a subsequent increase in BOD_5 around ≅ 60 mg/L. Although the effluent raw water sample characteristics are not the same, the results of these authors show that BOD_5 removal is not consistent neither proportional to Tanfloc dosages.

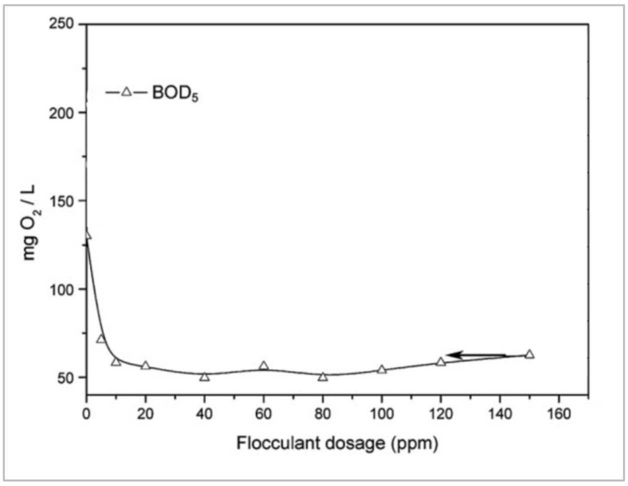

Figure 4.8 BOD_5 evolution with flocculant Tanfloc treatment
(Adapted from: Beltrán-Heredia & Sánchez-Martín, 2009. pp. 357)

In contrast with the CP results, MV's BOD_5 reduced continuously from an initial 30 mg/L to 25 and 20 mg/L, respectively for Tanfloc concentrations of 40 and 50 mg/L. These results yielded removal rates of 16.67% and 33.33% respectively. These rates are below the ones found in Beltrán-Heredia & Sánchez-Martín (2009) study (50%) and below the 60% BOD_5 removal found at Hameed et al. (2016) study using 35 mg/L of Tanfloc.

The difference in removal rates obtained in this study with those reported by Beltrán-Heredia & Sánchez-Martín (2009) and Hameed et al. (2016) is likely due to the effect of different concentrations of BOD_5 of the raw water sample. These authors had an initial BOD_5 of 130 mg/L and 100 mg/L, respectively, values much higher than the ones for CP (15 mg/L) and MV (30 mg/L).

Regarding the legislation for water reuse the Carrapateira effluent treated with 40 mg/L of Tanfloc, had a result that falls within the legal threshold of all irrigation water reuse categories (Table 4.2). For the CP at 50 mg mg/L of Tanfloc, and both results from MV, the resultant water falls within the water reuse categories of irrigation B, C, D, and E.

E. Total suspended solids (TSS)

Total suspended solids (TSS) indicate the concentration of suspended solids in water. CP's TSS reduced from 6.4 mg/L to 2.4 mg/L and 1.6 mg/L with Tanfloc, achieving removal rates of 62.5% and 75 %. MV's TSS reduced from 10 mg/L to 2.8 mg/L and 2.4 mg/L, achieving removal rates of 72 % and 76 %. Consistent with these values, but at a determined optimal dosage of 75 mg/L of Tanfloc, Leite et al. (2019) found that the tannin-based coagulant yielded a removal rate of 88.6 % for total suspended solids. Furthermore, Hameed et al. (2016) observed TSS removal rate of 63% for a dosage of 35 mg/L of Tanfloc. Both CP and MV treated effluents meet all the irrigation reuse categories.

F. *E. coli.*

E. coli concentrations were significantly reduced in CP, from an initial 45000 CFU/100mL to 1800 and 1000 CFU/100mL, and in MV, from an initial 18000 CFU/100mL to 700 and 100 CFU/100mL respectively for Tanfloc concentrations of 40 mg/L and 50 mg/L. CP achieved removal rates of 96.00% and 97.78%, while MV achieved rates of 96.11% and 99.44% for the 40 mg/L and 50 mg/L Tanfloc treatment, respectively. These high removal rates are consistent with several studies that have tested tannin-based coagulants yielding removal rates from 80 to 99% (Ibrahim et al., 2021; Sánchez-Martín et al., 2009, 2010).

As per the legislation, *E. coli* initially was the most limiting factor, preventing CP and MV effluents to be reused even in the least strict categories, however, after the 50 mg/L Tanfloc treatments, CP is now able to be used for irrigation C, D, and E reuse categories and MV for B, C, D, and E.

G. Nutrients: Total nitrogen (N_{TOTAL}) and total phosphorus (P_{TOTAL})

Total nitrogen and phosphorus levels varied with Tanfloc treatments. For N_{TOTAL} CP's values increased with the addition of 40 mg/L and 50 mg/L of Tanfloc. Previous studies have shown the problem that Tanfloc can have in increasing nitrogen content (Heiderscheidt et al., 2016; Tomasi et al., 2022). Perhaps the addition and presence of Ammonium chloride in residual Tanfloc, suggested by Hameed et al. (2016) and Tomasi et al. (2022), could be an explanation to the observed increase in N_{TOTAL} shown in the CP results.

In contrast, MV's Tanfloc-treated samples achieved removal rates of 29.37% and 16.16% respectively for the 40 mg/L and 50 mg/L dosages. Supporting these findings of N_{TOTAL} removal, Leite et al. (2019), observed up to 92.6 % removal rate of total Kjeldahl nitrogen. No studies have been found that focus particularly on analyzing the removal of total nitrogen with the use of Tanfloc. It is important to note, however, that possibly dosage, raw sample water quality, jar-test operational parameters, Tanfloc composition, sample storage, among other factors, are affecting the consistency of results.

As per the legislation, none of the treated effluents meet any of the legal threshold of ≤ 15 mg/L for all given irrigation reuse categories. However, as nutrients are a facultative in the law, these values still allow the legal reuse of the treated water.

Regarding, P_{TOTAL}, the CP WWTP achieved removal rates of 46.91% and 4.59%, while MV achieved rates of 37.48% and 5.22% for the 40 mg/L and 50 mg/L Tanfloc dosage respectively. For the 40 mg/L Tanfloc both CP and MV removal rates come close to the findings of Pereira et al. (2020) of 46.5% of P_{TOTAL} removal, however, their dosage was 212 mg/L of Tanfloc and their sample was raw dairy effluent with 56.6 mg/L of total phosphorus (5 times more than the initial CP and MV total phosphorus values). Furthermore, Leite et al. (2019) observed up to 89.9% removal from an initial 86.7 mg/L of P_{TOTAL}, and also when using 75 mg/L of Tanfloc dosage.

In both cases a significant decrease in P_{TOTAL} removal can be observed when incrementing the Tanfloc dosage to 50 mg/L. Similar as in the values of N_{TOTAL}, both CP and MV Tanfloc-treated effluents do not meet the legal threshold of ≤ 5 mg/L for all given irrigation reuse categories, yet they are still in a facultative allowance to be legally reused.

H. Ammonium (NH_4^+)

Ammonium, NH_4^+ concentrations in CP WWTP had a slight decrease from 0.83 mg/L to 0.74 mg/L and 0.63 mg/L with Tanfloc, achieving removal rates of 10.84% and 24.10% for the 40 mg/L and 50 mg/L dosage respectively. MV WWTP saw a slight reduction from the initial ammonium concentration of 0.50 mg/L to 0.49 mg/L and 0.47 mg/L, achieving removal rates of 2% and 6% for the 40 mg/L and 50 mg/L respectively. Several studies confirm the presence of ammonia chloride in Tanfloc (Dos Santos et al., 2018; Pacheco et al., 2022; Sabino et al., 2021; Sánchez-Martín et al., 2010), and for that reason the fact that it has not increased the ammonia concentration, can be argued, is an indicator that Tanfloc does not significantly alter ammonia concentration even when it is present in its composition.

It is important to mention that the water samples from the CP and MV water treatment plants already had ammonium values that met the water reuse standard, which is ≤ 5 mg/L. In that regard, the Tanfloc-treated water for both WWTPs still complies with all irrigation reuse categories as it remains within the legal threshold.

4.3 Coconut-shell activate carbon (CSAC) effectiveness in filter experiment.

For the two treatment plants, Table 4.6 shows the obtained operational parameters at the end of the experiment and for each WWTP. For CP, all three filters started the experiment, and took around 6.4 hours to be stabilized, from then on, the experiment started, and it roughly took 24 hours and a half for all three tubes to get clogged. The volume that got the filters clogged was 2.62 l, 2.63 l, and 2.71 l for filter 1, 2, and 3 respectively.

Table 4.6 Resultant operational parameters for Carrapateira and Maria Vinagre CSAC filter experiment

Operational parameters	Carrapateira (CP)		
	Filter 1	Filter 2	Filter 3
Stability time (hours)	6.44	6.47	6.47
Experiment time (hours)	24.62	24.53	24.53
Volume (l)	2.62	2.63	2.71
CSAC quantity (g)	0.83	1.66	2.5
Operational parameters	Maria Vinagre (MV)		
	Filter 1	Filter 2	Filter 3
Stability time (hours)	0.92	0.96	0.99
Experiment time (hours)	3.82	3.78	3.70
Volume (l)	1.16	1.10	0.99
CSAC quantity (g)	0.83	1.66	2.5

For MV, all three filters started the experiment, and took 0.92, 0.96, and 0.99 hours to stabilize filter 1, 2, and 3 respectively. From then on, the experiment started, and it took 3.82, 3.78, and 3.70 hours respectively for all three tubes until the experiment had to be stopped.

Table 4.7 presents a summary of the results values of all the measurements made post the CSAC filter experiment. The same information is shown in Table 4.7 for the Maria Vinagre WWTP. The average values for Turbidity, TOC, pH, and conductivity, were calculated once the filter reached stability, therefore providing steady values.

Table 4.7 Carrapateira and Maria Vinagre CSAC filter experiment results

Water quality parameter	Carrapateira sample	Coconut-shell activated carbon (CSAC) thickness		
		Filter 1	Filter 2	Filter 3
Turbidity (NTU)	5.58 ± 0.03	2.37 ± 0.66	2.26 ± 0.51	2.21 ± 0.26
BOD$_5$ (mg/L)	15	25	30	30
TSS (mg/L)	6.4	0.8	1.2	0.8
E. coli(CFU/100 mL)	45000	2700	4500	3300
NH$_4^+$ (mg/L)	0.83	1.04	1.05	0.96
N$_{TOTAL}$ (mg/L)	59.67	79.93	88.7	100.56
P$_{TOTAL}$ (mg/L)	10.02	1.9	1.93	1.95
TOC (mg C/L)	11.34 ± 0.16	10.73 ± 0.22	9.83 ± 0.36	9.17 ± 0.16
pH	8.05 ± 0.02	7.99 ± 0.50	7.98 ± 0.51	7.92 ± 0.01
Conductivity (uS/cm)	1261.00±1.79	1305.63±5.83	1308.00±1.26	1312.24±1.78
Water quality parameter	Maria Vinagre sample	Coconut-shell activated carbon (CSAC) thickness		
		Filter 1	Filter 2	Filter 3
Turbidity (NTU)	8.00 ± 0.05	4.85 ± 0.40	4.57 ± 0.14	4.35 ± 0.15
TOC (mg C/L)	13.79±0.18	11.53 ± 0.30	10.15 ± 0.20	9.42 ± 0.20

The efficiency of biologically-based activated carbon filters, such as CSAC, depends on several factors such as the quality of the water to be treated (i.e. turbidity, amount of organic matter, pH, temperature, among others) and the operating conditions, such as the contact time influenced by the filtration flow rate (Singh et al., 2020). Of the factors mentioned above, the only one that could have been manipulated is the filtration flow rate. It is important to discuss this because it is inferred that the lower the filtration flow rate, the longer the contact time and the more time the activated carbon must adsorb the contaminants. For instance, Wang et al. (2022) found that decreasing the filtration flow rate five times (from 500 ml/min to 100 ml/min) increased the penetration curve of formaldehyde adsorbed. However, the peristaltic pump did not allow a filtrate flow rate of less than 1 ml/min, and the average filtrate flow rate observed in the experiment was 2 ml/min, which is a non-significant change. Furthermore, it was observed that the different amounts of CSAC used did not really make a clear difference in relation to the results obtained.

With this information, the following items describe and analyze the results obtained for all legislated parameters after having tested these three CSAC quantities in both treatment plants. In contrast with the C/F/S experiment, Table 4.7 highlights a significant decrease in P_{TOTAL}, yet a not so significant decrease in *E. coli* and Turbidity.

4.3.1 Analysis of legislated parameters

A. Turbidity

For the examination of the Carrapateira WWTP effluent, Filter 1, Filter 2, and Filter 3 demonstrated average turbidity values of 2.37 ± 0.66 NTU, 2.26 ± 0.51 NTU, and 2.21 ± 0.26 NTU, yielding an approximate removal rate of 59%. According to the results, all 3 filters provided turbidity values that fall within the legal threshold of ≤ 5 NTU (Figure 4.9), making this CSAC-treated water with quality to meet the legal turbidity standard for every irrigation reuse category.

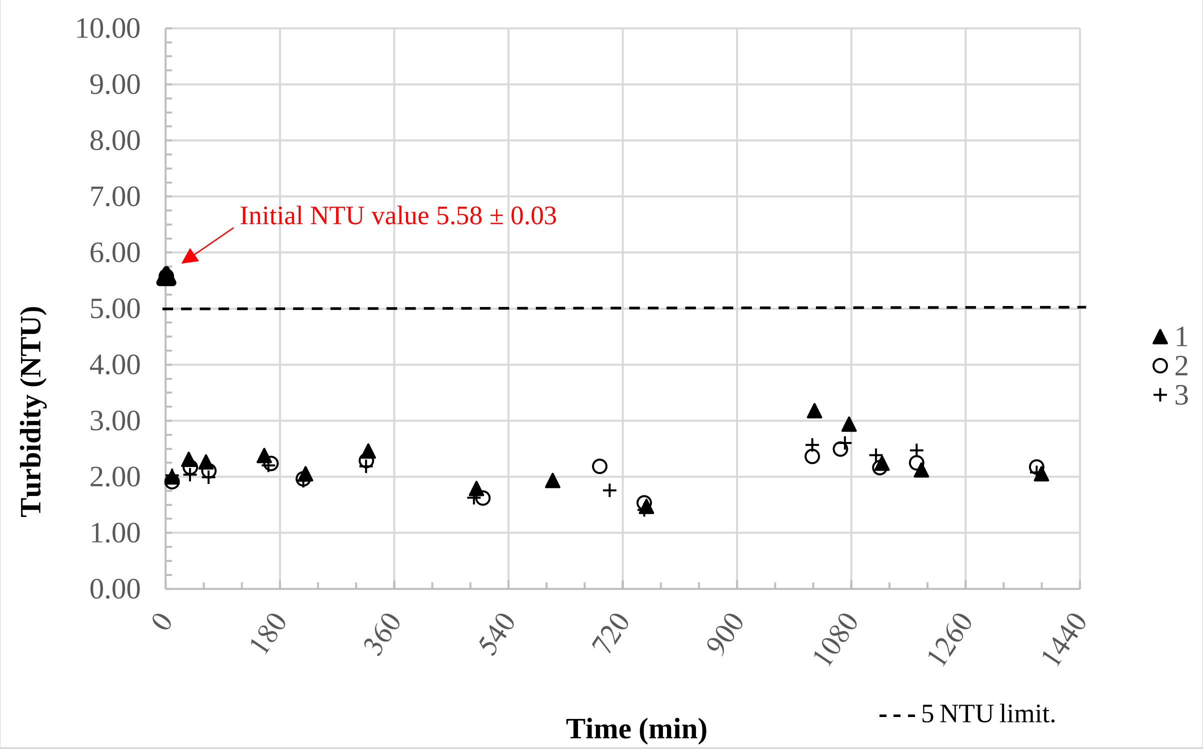

Figure 4.9 CP WWTPs turbidity results after CSAC filter experiment.

In the analysis of the effluent from the Maria Vinagre Carrapateira WWTP, it is observed average turbidity values of 4.85 ± 0.40 NTU for Filter 1, 4.57 ± 0.14 NTU for Filter 2, and 4.35 ± 0.15 NTU for Filter 3. These values correspond to removal rates of 39%, 43%, and 46%, respectively. Based on the findings, the turbidity values from all three filters were below or equal to the permissible limit of ≤ 5 NTU (Figure 4.10), indicating that they meet the legal requirements for reuse within this specified parameter.

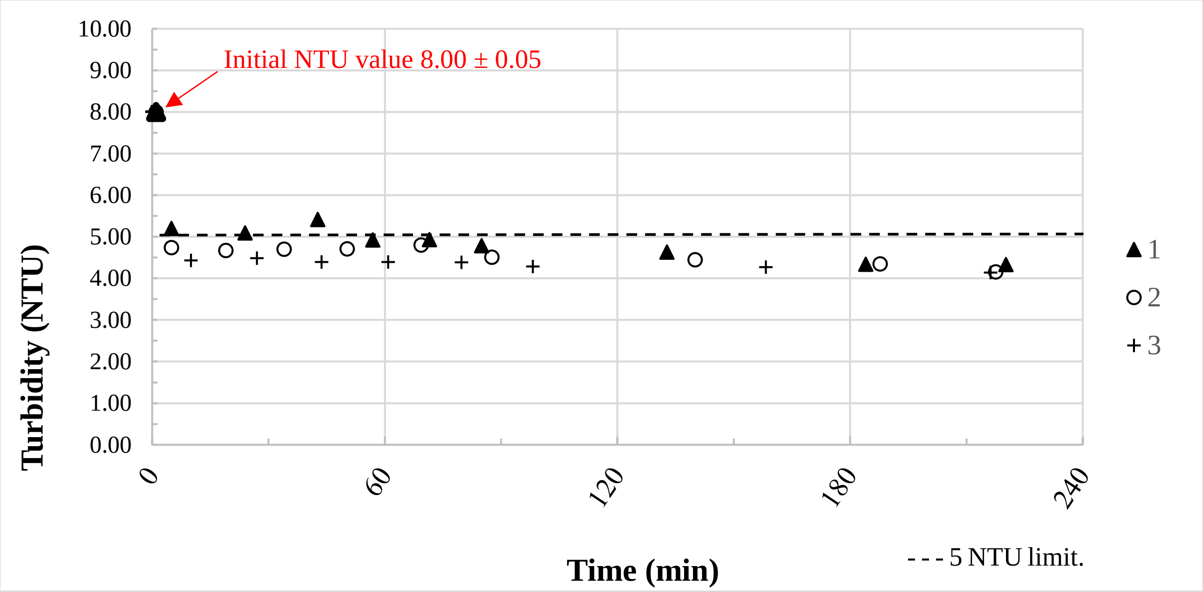

Figure 4.10 MV WWTPs turbidity results after CSAC filter experiment.

In contrast with CP, MV results in Figure 4.10 show that around the 90 minutes of elapsed experiment time mark, turbidity reached values below 5 NTU. These results might differ from the ones obtained for CP because, as stated previously, the efficiency of biologically-based activated carbon filters significantly depends on the quality of the water to be treated (Singh et al., 2020). In that regard, CP initial turbidity values were much lower (5.58 ± 0.03 NTU) to the ones in MV (8.00 ± 0.05 NTU).

For both CP and MV, the CSAC results show higher turbidity values than the ones found in the C/F/S experiments previously. It is noteworthy that all three filters, for both CP and MV, eventually reached a point where further reduction in turbidity was minimal (Figures 4.9 and 4.10). However, both WTTP treated effluents eventually reached a below 5 NTU value, therefore complying with all irrigation reuse categories for that given parameter.

B. Total organic carbon (TOC)

The TOC values for CP decreased after CSAC treatment, indicating its effectiveness in organic carbon removal. From an initial value of 11.34 ± 0.16 mg/L, TOC decreased to 10.23 ± 0.03 mg/L, 9.81 ± 0.45 mg/L, and 9.42 ± 0.16 mg/L respectively for filter 1, 2, and 3. These results represent removal rates of 10%, 13%, and 17% respectively after 480 min, indicating a trend relating more quantity of CSAC to higher removal of TOC. Figure 4.11 shows the clear tendency of decreasing values of TOC, and the stability of the results, which fluctuate between 11 and 8.5 mg/L of TOC. Similarly, as in the C/F/S experiment, CP's CSAC-treated effluent values were consistent and not significant increase or variation of TOC was observed.

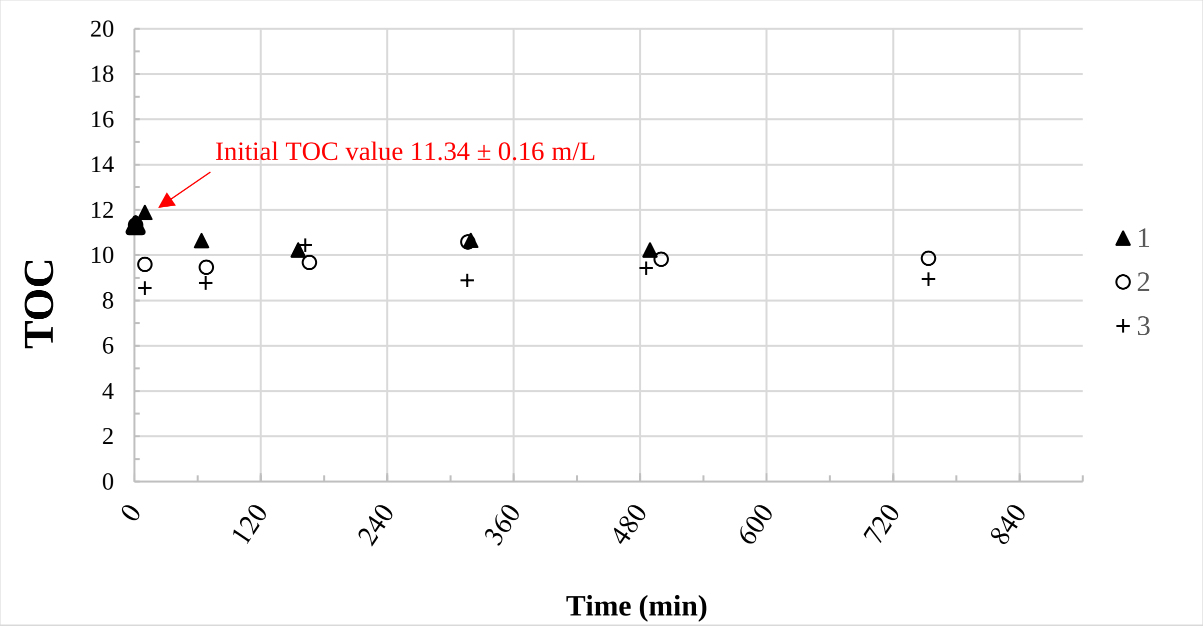

Figure 4.11 CP WWTPs TOC results after CSAC filter experiment

Following CSAC treatment, there was a decline in the TOC values for MV. Initially recorded at 13.79±0.18 mg/L, the TOC decreased to 10.24 ± 0.15 mg/L, 9.68 ± 0.17 mg/L, and 10.45 ± 0.12 mg/L for filters 1, 2, and 3, respectively, after 180 min (Figure 4.12). These reductions correspond to removal rates of 26%, 30%, and 24%. Figure 4.12 shows the tendency of decreasing values of TOC, and the stability of the results, which fluctuate between 13 and 7.5 mg/L of TOC.

When comparing the TOC evaluation, both the CP and MV cases exhibit lower values than those observed in the C/F/S experiments. This discrepancy can be attributed to the inherent differences in the removal mechanisms of the two treatment processes.

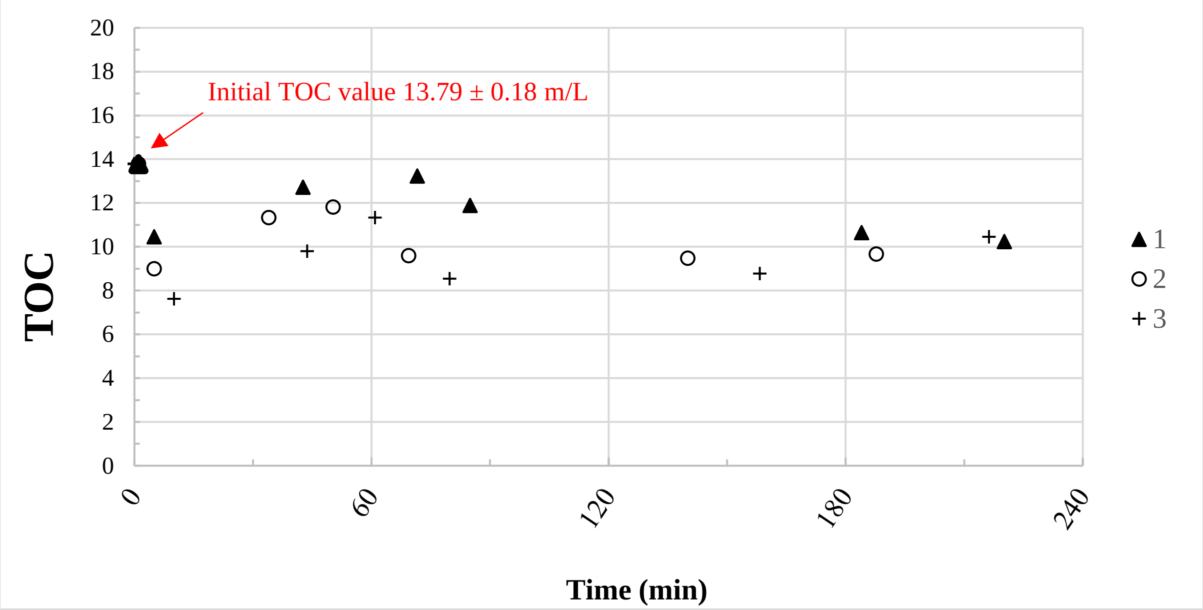

Figure 4.12 MV WWTPs TOC results after CSAC filter experiment

The TOC in water comprises both particulate organic carbon (POC) and dissolved organic carbon (DOC). While C/F/S is adept at removing POC and some high molecular weight DOC, it might not be as effective in targeting low molecular weight DOC or specific organic compounds that resist coagulation and flocculation. On the other hand, activated carbon treatment, with its adsorption mechanism, can effectively target a broader spectrum of DOC, including those compounds that might elude the C/F/S process.

This is perhaps why the TOC values in the CSAC experiment are notably lower than in the C/F/S process. The references to Chow et al. (2009) and Yapsakli & Çeçen (2010) further substantiate the adsorptive capabilities of activated carbon in removing a diverse range of organic compounds.

C. pH and conductivity

For Carrapateira wastewater treatment plant effluent, CSAC exhibited minimal impact on pH levels, as depicted in Figure 4.13, which presents the outcomes from treating the effluent samples. Importantly, these values remained within the neutral range, aligning favorably with most water reuse applications. It is worth noting that the treated effluents consistently fell within the acceptable pH range of 6-9, making them suitable for the reuse categories, including Irrigation A to E, as well as all urban uses.

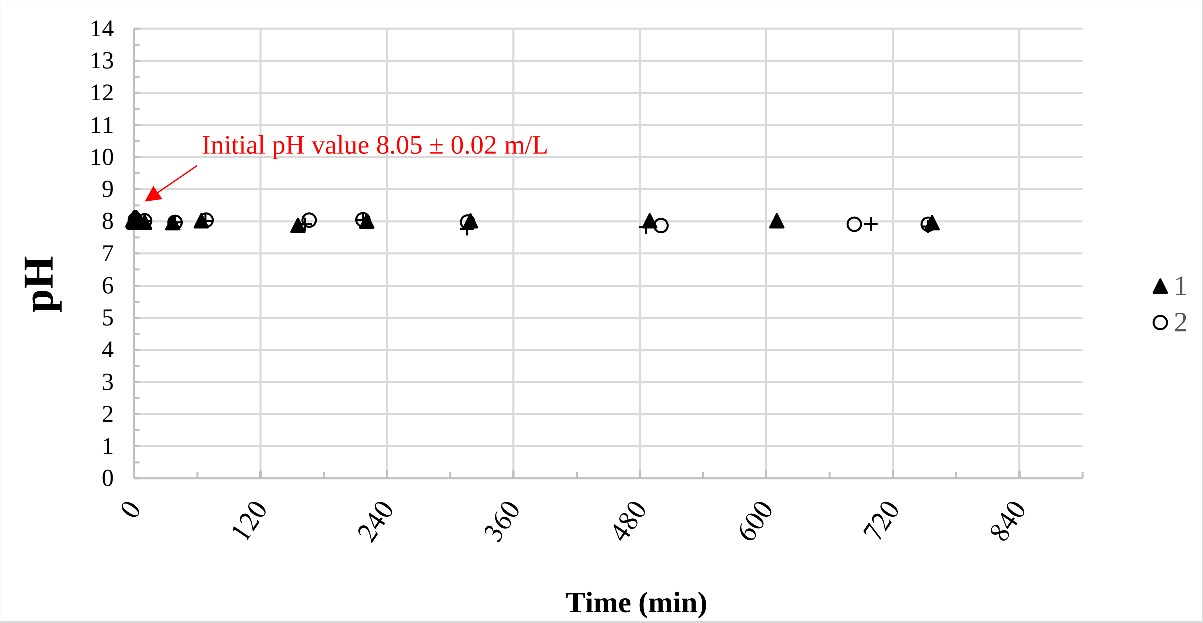

Figure 4.13 CP WWTPs pH results after CSAC filter experiment

Conductivity for CP showed a slight increase, with post-treatment with values of 1285.33±2.52, 1295.33±0.58, and 1300.00±2.00 uS/cm for filter 1, 2, and 3 respectively and observed after 750 minutes (Figure 4.14). However, these variations remain non-significant.

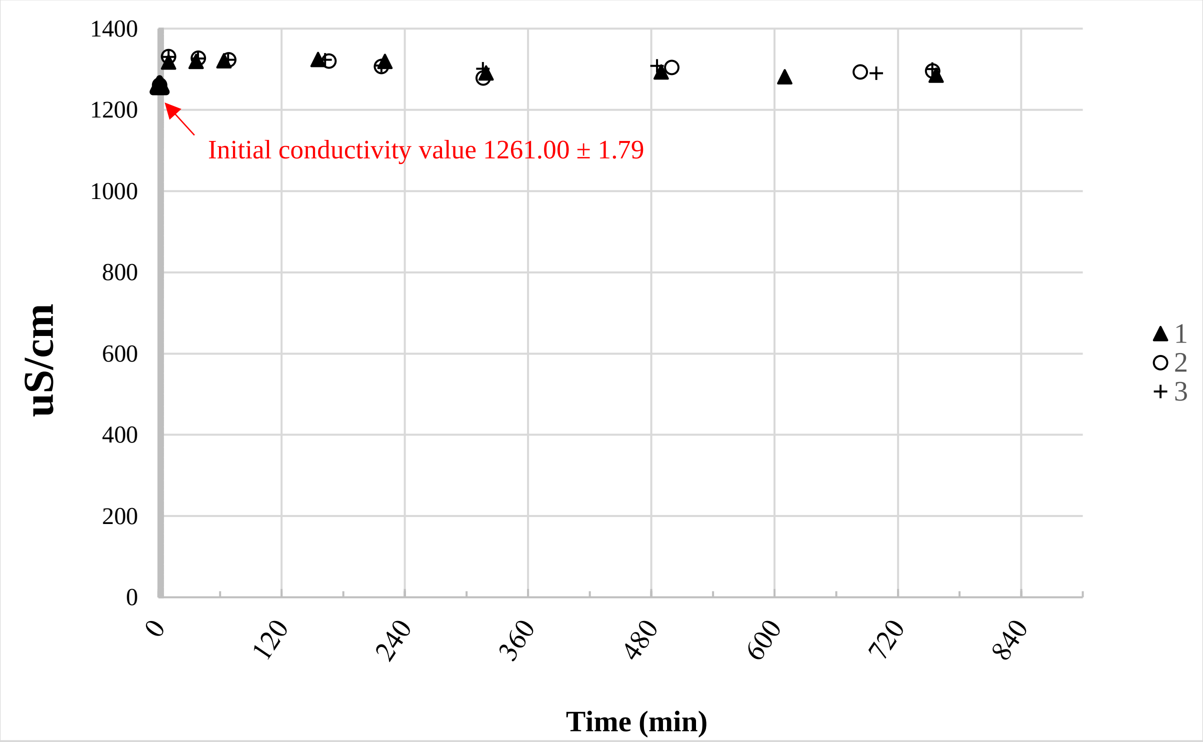

Figure 4.14 CP WWTPs conductivity results after CSAC filter experiment

D. Biological oxygen demand (BOD_5).

The decline in TOC values indicates a reduction in total organic carbon. Yet, the BOD_5 values for CP increased from an initial 15 mg/L to 25, 30, and 30 mg/L in the three filters (Table 4.7). The rise in BOD_5 values suggests, perhaps, increased microbial decomposition. The high levels of *E. coli* found in the raw sample water, might be indicative of heightened microbial activity, which could be consuming more readily biodegradable organic compounds, leading to the elevated oxygen demand. While the overall organic carbon has decreased, the active microbial environment, signaled by the *E. coli* presence, may be affecting the bioavailability of the remaining organic matter, thus influencing BOD_5 values.

By staying within the ≤ 25 mg/L of BOD_5 threshold, filter 1, for this given parameter, is allowed to be legally reused for irrigation categories B to E. For filters 2 and 3, the CSAC-treated Carrapateira effluent meets the ≤40 mg/L threshold, which suffices reuse irrigation categories E.

E. Total suspended solids (TSS).

CP's TSS reduced from 6.4 mg/L to values below 1.2 mg/L in all filters, achieving removal rates from 81% to 88% after 1450 minutes. These values are consistent with the reported in the review done by James & Yadav (2021), were they presented that activated carbon from coconut husk had achieved a decrease from an initial 375 mg/L to 60 mg/L, thus yielding a removal rate of 84%.

Th CSAC-treated effluent from CP, for all the filters containing different amounts of activated carbon, had a remarkable response to the reduction of TSS. In this given water quality parameter, the treated water meets the strictest legal threshold of ≤10 mg/L TSS and, therefore, meets the TSS requirements for all irrigation reuse categories.

F. *E. coli.*

The initial *E. coli* count for CP was 45000 CFU/100 mL, which reduced dramatically to 2700, 4500, and 3300 CFU/100 mL in the filters1, 2, and 3 respectively. While this reduction is commendable, yielding removal rates of 95%, 90%, and 93%, the values still exceed most of the *E. coli* legal thresholds for the irrigation water reuse categories. The CSAC-treated water from filters 1 to 3 only meets the requirements for categories D and E for irrigation, which present a limit of ≤ 10000 CFU/100mL. Further treatment or a combination of methods might be necessary to achieve this stringent threshold.

G. Nutrients: Total nitrogen (N_{TOTAL}) and total phosphorus (P_{TOTAL}).

The Carrapateira N_{TOTAL} concentrations increased after CSAC treatment. N_{TOTAL} raised from 59.67 mg/L to 79.93 mg/L, 88.70 mg/L, and to over 100 mg/L in the third filter and after 1450 minutes, therefore not achieving any removal at all. For the P_{TOTAL} concentrations decreased after CSAC treatment. P_{TOTAL} lowered from 10.02 mg/L to 1.90 mg/L, 1.93 mg/L, and to 1.91 mg/L for filters 1 to 3 respectively after 1450 minutes.

H. Ammonium (NH_4^+)

Ammonium, NH_4^+, concentrations in CP reduced from 0.83 mg/L to 1.04 mg/L, 1.05 mg/L, and 0.96 mg/L for filters 1 to 3 after 1450 minutes, not yielding any removal and rather slightly increasing ammonium concentration. In summary, the use of Coconut-shell Activated Carbon as an adsorbent has demonstrated potential in improving the quality of effluents from the Carrapateira and Maria Vinagre wastewater treatment plants. While the treated effluents met several of the thresholds established by the Portuguese legislation for non-potable reuse, certain parameters, especially *E. coli* counts, require additional attention.

4.4 Compliance Portuguese legislation and potential of water reuse

This section summarizes the results obtained for the C/F/S and CSAC experiments for the treatment of effluents from both Carrapateira and Maria Vinagre WWTPs in Table 4.8 and Table 4.9 respectively. In addition, the results are presented in such a way that they reveal in red or green whether the values obtained, for each mandatory and facultative water quality parameter, comply with the requirements of the law decree Nº 119/2019. Additionally, a colorized scheme has been added to observe if the treatment had an improvement on a given water quality parameter or if it remained the same.

Table 4.8 Summary of the Carrapateira C/F/S and CSAC results expressed in water reuse potential for irrigation categories.

Water quality parameter	Sample	C/F/S Experiments		CSAC Filter experiment		
	Carrapateira (CP)	40 mg/L of Tanfloc	50 mg/L of Tanfloc	Filter 1	Filter 2	Filter 3
Mandatory water reuse thresholds						
Turbidity (NTU)	Irrigation B, C, D, and E.	All categories	All categories	All categories	All categories	All categories
BOD5 (mg/L)	Irrigation B, C, D, and E.	All categories	Irrigation B, C, D, and E.	Irrigation B, C, D, and E.	Irrigation E.	Irrigation E.
TSS (mg/L)	All categories	All categories	All categories	All categories	All categories	All categories
E. coli (CFU/100 mL)	None.	Irrigation D, and E.	Irrigation C, D, and E.	Irrigation D, and E.	Irrigation D, and E.	Irrigation D, and E.
Facultative water reuse thresholds						
NH4+ (mg/L)	All categories	All categories	All categories	All categories	All categories	All categories
NTOTAL (mg/L)	None	None	None	None	None	None
PTOTAL (mg/L)	None	None	None	All categories	All categories	All categories
pH	All categories	All categories	All categories	All categories	All categories	All categories
Overall reuse potential	None	Irrigation D, and E.	Irrigation C, D, and E.	Irrigation D, and E.	Irrigation D, and E.	Irrigation D, and E.

Color legend:	Not reusable	Reusable	Improved with treatment	Remained the same

Table 4.9 Summary of the Maria Vinagre C/F/S and CSAC results expressed in water reuse potential for irrigation categories.

Water quality parameter	Sample	C/F/S Experiments		CSAC Filter experiment		
	Maria Vinagre (MV)	40 mg/L	50 mg/L	Filter 1	Filter 2	Filter 3
Mandatory water reuse thresholds						
Turbidity (NTU)	Irrigation B, C, D, and E.	All categories	All categories	All categories	All categories	All categories
BOD5 (mg/L)	Irrigation E.	Irrigation B, C, D, and E.	Irrigation B, C, D, and E.	-	-	-
TSS (mg/L)	All categories	All categories	All categories	-	-	-
E. coli (CFU/100 mL)	None.	Irrigation B, C, D, and E.	Irrigation B, C, D, and E.	-	-	-
Facultative water reuse thresholds						
NH4+ (mg/L)	All categories	All categories	All categories	-	-	-
NTOTAL (mg/L)	None.	None	None	-	-	-
PTOTAL (mg/L)	None.	None	None	-	-	-
pH	All categories	All categories	All categories	-	-	-
Overall reuse potential	None.	Irrigation B, C, D, and E.	Irrigation B, C, D, and E.	-	-	-

Both Table 4.8 and 4.9 are self-explanatory, however it is important to highlight that all the carried-out experiments with measured results were able to improve the water quality for each and all the mandatory water reuse thresholds except from TSS, which already met the legal threshold before the treatment. Furthermore, for the CP WWTP the reuse potential improved from not being able to be reused in any irrigation reuse category to irrigation D and E. For MV WWTP, the reuse potential improved from none to Irrigation B, C, D, and E, only for the C/F/S results. Lastly, for the CP WWTP, the CSAC had a significant impact on the facultative parameter of PTOTAL, improving it from a none reuse potential standard to an all-inclusive standard (Table 4.8).

In summary, for the overall experiments, the effluent of Carrapateira was able to reach the requirements for irrigation category C, and the effluent of Maria Vinagre, the irrigation category B using the proposed treatments. Furthermore, both WWTP's treated effluent's meet the legal threshold for all reuse categories for urban uses as described in the law decree Nº 119/2019 for non-potable reuse.

4.5 Result-based recommendation for a complementary process.

To recommend a complementary process, it is necessary to realistically consider the context in which the measure will be proposed. In this sense, the water treatment plants of Carrapateira and Maria Vinagre are decentralized plants that try to meet the demand for domestic wastewater treatment remotely. Usually, this type of WWTPs do not have extraordinary budgetary resources to redesign the infrastructure that is already installed. For this reason, this recommendation considers the implementation of an additional and subsequent process to the already installed system of constructed wetlands.

The recommendation builds upon the work of Lopes (2021), in which the author evaluated the factors that influence the performance of specifically the Carrapateira and Maria Vinagre WWTPs. In that regard, the recommended design depicted in Figure 4.15 presents the set up to improve the performance of the Aljezur CW-based WWTPs, which include Carrapateria and Maria Vinagre. The recommendation in the current study will build upon the components 5 because it is compatible with the treatments that have been experimented on in this study and they are placed after the CWs.

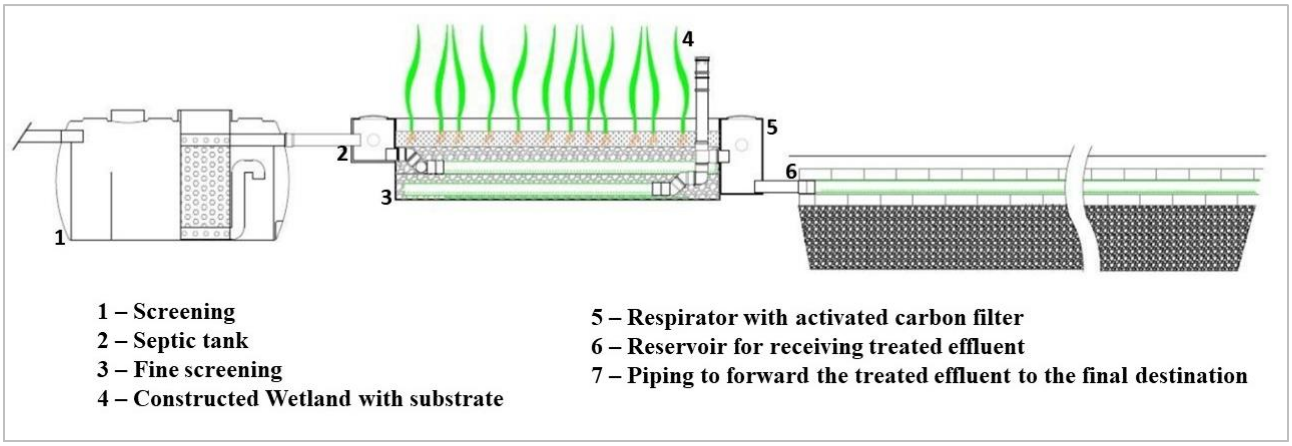

Figure 4.15 Lopes (2021) recommended set up for CW-based WWTP in Aljezur
(Taken from: Lopes, 2021. pp. 55)

It is recommended for the Carrapateira WWTP that component 5, as depicted in Figure 4.15, be an CSAC-based activated carbon filter and designed so that the entire flow of the effluent flows through. It is proposed in this way since in the first instance, the partial reduction of *E. coli* and turbidity obtained with the CSAC experiment, which are not of great magnitude, already raise the quality of the effluent so that it can be used in categories irrigation D and E, therefore providing already a reusable treated water. Furthermore, it is proposed that the entire flow be treated since the investment is probably not large, thus representing a cost-efficient and sustainable option. This is because if we observe, the CSAC experiment that was carried out, on average, had a flow rate of 2 ml/minute in which the results presented in Table 4.7 were obtained. The three filters presented results that were not very different, thus making the most cost-efficient option to emulate the content of filter 1. This filter had a CSAC experiment quantity of 0.83 grams, and the Carrapateira treatment plant, according to Lopes (2021), has an average influent flow rate of 39 m3/day. Taking the CSAC experiment flow rate and converting it to m3/day, the flow rate would be 0.00288 m3/day.

Making a preliminary proportional calculation, in a very general way, without considering external variables and converting it to cubic meters, it can be argued that the required CSAC WWTP filter quantity (kg) to emulate the results of filter 1 for the CP effluent would be that of 11.24 kg approximately, amount that realistically can be obtained and set up in a structure. Calculations:

- *CSAC experiment flow rate:*

  $2\ ml/min = 2 \times 10^{-6}\ m^3/min \times 1440\ min/day = 0.00288\ m^3/day$

- *CSAC experiment quantity:*

  $0.83\ g = 0.00083\ kg$

- *Carrapateira WWTP influent flow rate:*

  $39\ m^3/day$

- *Calculation:*

$$\frac{CSAC\ experiment\ quantity\ (kg)}{CSAC\ experiment\ flow\ rate\ (\frac{m3}{day})} = \frac{CSAC\ WWTP\ filter\ quantity\ (kg)}{WWTP\ influent\ flow\ rate\ (\frac{m3}{day})}$$

- *CSAC WWTP filter quantity:*

  $= (39\ m^3/day)\ (0.00083\ kg)\ /\ 0.00288\ m^3/day = 11.24\ kg$

Another recommendation is that the CSAC filter in component 5 (Figure 4.15) is coupled with a posterior, UV disinfection system that can further decrease *E. coli* concentrations if there is a demand for higher quality water for reuse for instance in irrigation reuse category A, B or C. This is recommended because, as the results have shown, *E. coli* is the most limiting factor, after turbidity, when trying to comply with all the irrigation reuse categories. Due to the context of the WWTPs, a small UV light system is suggested like the Altima model of the AQUADA UV series by the company Wedeco (Figure 4.16). This model is the most economical one and still it can treat from 0.73 to 13.4 m^3/hour or the equivalent to 17.52 to 321 m^3/day, which is within the influent flow of both Carrapateira and Maria Vinagre of 39 and 100 m^3/day (Lopes, 2021; WEDECO Aquada, n.d.).

Figure 4.16 AQUADA UV series by the company Wedeco (WEDECO Aquada, n.d.)

C/F/S had a significant positive effect in *E. coli* removal, this is because this process has been designed to reduce particulate matter and *E. coli* is particulate. However, installing a C/F/S system would not be cost-efficient as the infrastructure demand for C/F/S is much more than for CSAC, and both experiments obtained very similar values already. Only if there is the need for further improvement of *E. coli* values without the possibility of a UV system, a C/F/S is recommended, for both Carrapateira and Maria Vinagre WWTPs. The placement would be as in the component 6 presented by Lopes (2021) in Figure 4.15, which should be adapted in such a way that it can either have a rapid hydraulic mixing tank, a flocculation tank and a sedimentation tank as represented in Figure 4.16, or perhaps use a combined coagulation, flocculation and sedimentation unit as designed by Ismail et al. (2012).

However, in both cases, the objective is to place a system that allows the use of Tanfloc to replicate and emulate the C/F/S experiment within an infrastructure that is within a decentralized WWTP budget. In that sense, building an infrastructure that can work with the entire effluent of Carrapateira (39 m^3/day) and Maria Vinagre (100m^3/day), would be disproportionate for the purpose of these WWTPs (Lopes, 2021). It is for this reason that it is recommended that for Carrapateira, the C/F/S system only addresses a partial volume of the total effluent from both Carrapateira and Maria Vinagre, perhaps not more than 20% so the infrastructure costs are not disproportionate with the nature of a decentralized WWTP.

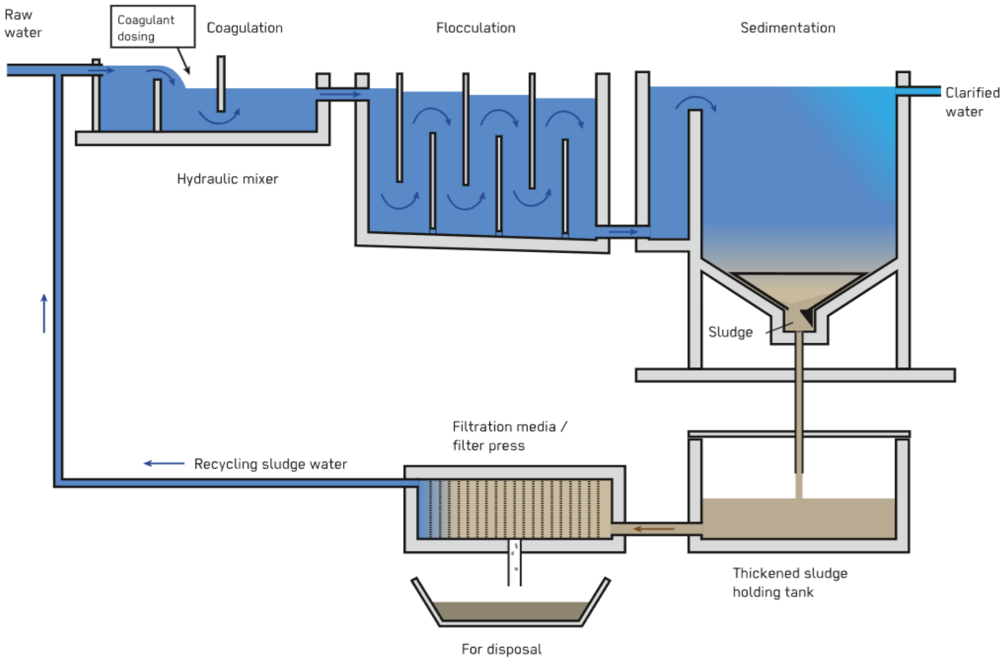

Figure 4.17 Representation of the recommended C/F/S system.
(Taken from: Coerver et al., 2021. pp. 104)

In summary, it is highly recommended to install a CSAC filter to treat the total effluent flow combined with a posterior UV disinfection system to treat the total amount of the effluent using a simile to the Altima model. A further C/F/S system would only be needed if UV is not an option and there is the demand for high quality water. For the Maria Vinagre WWTP the recommendation is the same as the results have been mostly consistent throughout both experiments.

5. Conclusion and recommendations for future research and practice

This research set out to address a pressing question: "What is the potential of using a nature-based coagulant agent, Tanfloc, or coconut shell-based activated carbon (CSAC) to improve the quality of the effluent for non-potable reuse purposes from the CW-based WWTPs in Aljezur?" The findings demonstrate that both Tanfloc and CSAC hold significant promise in enhancing the quality of wastewater effluent, thereby making it suitable for non-potable reuse in various irrigation categories.

The study conducted a comprehensive assessment of the effluent quality from two representative CW-based WWTPs in Aljezur, Carrapateira and Maria Vinagre. While the effluent from these plants largely met the criteria established by Portuguese Decree-Law No. 119/2019, it was evident that elevated levels of *E. coli* and turbidity necessitate additional treatment interventions.

The study demonstrates the effectiveness of the C/F/S process and the CSAC filter in water treatment. Both Tanfloc and CSAC significantly reduced *E. coli* concentrations, with rates up to 99% and 94%, respectively. Specifically, Tanfloc emerged as a particularly effective agent in reducing turbidity and *E. coli* concentrations, with the optimal turbidity removal achieved at 40 mg/L for Carrapateira and 50 mg/L for Maria Vinagre. Since C/F/S is not focused on nutrient removal, the efficacy of Tanfloc to remove parameters such as TOC and nutrients was not consistent. For C/F/S, TOC values did not increase as in previous studies and they rather slightly decrease, and CSAC excelled in reducing turbidity and TOC but was less effective in other areas. The treated effluents from both CP and MV met the standards for restricted reuse categories C, D, and E, emphasizing the potential of these treatments in water resource management for specific agricultural and urban applications.

Given these findings, the study advocates for a hybrid-treatment approach. For both WWTPs, the recommendation is to implement a CSAC filter followed by a UV disinfection system to treat the entire effluent flow, and, if there is the demand for irrigation reuse category A and not an option for UV disinfection, then use a C/F/S system.

The policy implications of this research are substantial, especially in the context of Decree-Law No. 119/2019. The demonstrated efficacy of Tanfloc and CSAC in meeting mandatory water reuse thresholds could serve as a catalyst for regulatory change, encouraging the broader adoption of these nature-based solutions.

Looking forward, there is a manifest need for additional research to fine-tune the dosages for Tanfloc and to delve deeper into the limitations of CSAC's adsorptive capacity. Future inquiries could also explore the long-term sustainability and economic viability of the proposed treatment methods.

In summary, this study makes a significant contribution to the broader discourse on water scarcity and sustainable management. By empirically validating the effectiveness of nature-based solutions like Tanfloc and CSAC, the research paves the way for the development of sustainable, low-cost wastewater treatment methods. This, in turn, promotes water conservation and reuse, thereby addressing some of the most pressing environmental challenges of our time.

6. References

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7. Appendices and supplementary information

7.1 Water reuse categories thresholds

Table 7.1 Overall water reuse categories per type of purpose and legal water quality parameter thresholds

Water reuse categories	Turbidity (NTU)	BOD_5 (mg/L)	TSS (mg/L)	*E. coli* (CFU/100 mL)	NH_4^+ (mg/L)	N_{TOTAL} (mg/L)	P_{TOTAL} (mg/L)	pH
Water reuse categories for irrigation purposes								
Irrigation A.	≤5	≤ 10	≤ 10	≤ 10	≤ 10	≤ 15	≤5	6-9
Irrigation B.	-	≤ 25	≤ 35	≤ 100	≤ 10			6-9
Irrigation C.	-			≤ 1000	≤ 10			6-9
Irrigation D.	-			≤ 10000	≤ 10			6-8.5/9
Irrigation E.	-	≤ 40	≤ 60	≤ 10000	≤ 10			6-9
Water reuse categories for urban uses purposes								
Support of ecosystems.	-	-	-	-	-	-	-	6-9
Recreational uses.	≤5	≤ 25	-	≤ 10	≤5	-	≤2	6-9
Washing of streets.	-		-	-	-	-	-	6-9
Combat water to fires.	≤5		-	≤ 10	-	-	-	6-9
Water for cooling.	-		-	≤ 200	≤5	-	-	6-8.5/9
Cisterns	≤5		-	≤ 10	≤ 10	-	-	6-9
Washing of vehicles.	≤5	-	-	≤ 10	-	-	-	6-9

7.2 Irrigation water reuse categories description.

Table 7.2 Overall irrigation water reuse description

Category	Description
A	Irrigation without access restriction (urban and agricultural uses): irrigation of crops consumed raw in which the consumable part is in direct contact with water; irrigation of public gardens without access restriction; irrigation of private gardens.
B	Irrigation with restricted access (urban and agricultural uses): irrigation of crops consumed raw, which grow above the ground, and in which the consumable part is not in direct contact with water; irrigation of agricultural crops intended for processing and agricultural crops not intended for human consumption, including crops intended for animal consumption (milk or meat production), except pigs; irrigation of gardens with restricted access, including leisure and sports areas (eg golf courses).
C	Irrigation with restricted access (agricultural uses): irrigation of consumed crops in raw, which grow above the ground, and in which the consumable part is not in direct contact with water; irrigation of agricultural crops intended for processing and agricultural crops not intended for human consumption, including crops intended for animal consumption (milk or meat production), except pigs.
D	Irrigation with restricted access (agricultural uses): seed production, including seeds for industrial use or energy production.
E	Irrigation with restricted access (agricultural uses): seed production; watering areas of naturally restricted use (eg, hedgerows, containment areas (terraced meadows).