

Research paper

A bench-scale assessment of enhanced coagulation-filtration for greywater in decentralized system

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ABSTRACT

This study evaluates the effectiveness of an enhanced coagulation-filtration (CF) process with post-filtration for treating bathroom greywater (BGW) for non-potable reuse applications. A bench-scale experiment assessed two cost-effective inorganic coagulants, ferric chloride (FeCl_3) and hydrated lime ($\text{Ca}(\text{OH})_2$), as well as their combination, using synthetic bathroom greywater (SBGW) to determine optimal treatment conditions. The CF process, with an optimal combined concentration of FeCl_3 at 250 mgL^{-1} and $\text{Ca}(\text{OH})_2$ at 148 mgL^{-1} achieved an ideal zeta potential range of -5 mV to 5 mV for SBGW, resulting in superior impurities removal. Jar tests under varying conditions (mixing speed, contact time, and sedimentation) confirmed these findings, and optimized conditions were validated with real bathroom greywater (RBGW) samples, exhibiting slight variations due to BGW quality differences. The FeCl_3 dosage was adjusted ($220\text{--}250 \text{ mgL}^{-1}$), while the $\text{Ca}(\text{OH})_2$ dose was kept constant (148 mgL^{-1}) for all RBGWs. The tested enhanced CF process demonstrated high removal efficiencies for turbidity ($99 \pm 0.5\%$), chemical oxygen demand (COD) ($96 \pm 3.4\%$), biochemical oxygen demand (BOD_5) ($98 \pm 1.3\%$), and anionic detergents ($99 \pm 0.5\%$) values of BGW samples, as well as reducing the number of colonies for *Escherichia coli* (99%), and Coliforms/Total Count TTC (99%), meeting EU Regulation 2020/741 requirements for water reuse in irrigation. Statistical analysis supports the novelty of enhanced CF, proving its superior effectiveness compared to single ferric chloride coagulation method, making it a promising decentralized greywater treatment solution for European countries.

Abbreviations

BGW	Bathroom Greywater;
BOD_5	Biochemical Oxygen Demand;
BW	Black water;
CF	Coagulation–flocculation;
COD	Chemical Oxygen Demand;
EC	Electrical Conductivity;
GW	Greywater;
RBGW	Real Bathroom Greywater;
SBGW	Synthetic Bathroom Greywater;
SDGs	Sustainable Development Goals;
TBGW	Treated Bathroom Greywater;
TGW	Treated Greywater;
TOC	Total Organic Carbon;
TRBGW	Treated Real Bathroom Greywater;
TSBGW	Treated Synthetic Bathroom Greywater;

TURB	Turbidity;
TW	Tap water;
WW	Wastewater;
ZP	Zeta potential

1. Introduction

Water scarcity, exacerbated by climate change and urbanization, threatens global freshwater supplies, with projections indicating severe shortages for nearly half of the global population in the coming decades [1–3]. In response, water reuse has emerged as a sustainable strategy to supplement freshwater demand and maintain a reliable water supply [4–7].

Greywater (GW), the fraction of domestic wastewater (WW) separated from blackwater (BW), offers a sustainable source for non-potable reuse. GW treatment has become a key strategy to conserve water, especially in regions facing extreme water shortages, reducing

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freshwater demand and infrastructure costs, and offers a lower environmental footprint compared to alternatives such as inter-basin water transfers and desalination [7–9].

GW, which can account for up to 90% of domestic WW, becomes easier to treat once separated from BW [10–12]. Among GW sources, bathroom greywater (BGW) is less contaminated than laundry or kitchen streams, making it particularly ideal for decentralized treatment [13, 14]. GW can be treated using various methods, with biological processes often emphasized due to GW's high organic content [15]. The effectiveness of GW treatment method varies depending on their ability to remove different types of contaminants and the specific water quality standards required for reuse, which is based on the intended application [16].

Common GW treatment technologies include constructed wetlands (CW), filtration, coagulation-flocculation (CF), and membrane bioreactor (MBR) [7,17–24]. Although advanced treatments such as advanced oxidation processes, electrocoagulation and microbial fuel cell offer higher efficiency, they are expensive and require skilled operations [7,25]. CW systems require extensive land area and operate slowly, with performance declining in warmer climates [17]. In contrast, MBR systems are energy-intensive and prone to membrane fouling, which significantly reduces their performance [24]. These limitations highlight the need for a simple approach to GW treatment.

Utilizing affordable and straightforward physicochemical methods typically ensures that treated greywater meets the standards required for non-potable reuse [16]. Coagulation-flocculation (CF) paired with filtration provides a cost-effective alternative, leveraging physicochemical and biological processes to remove turbidity, organic matter, and pathogens [18,19,21]. Coagulation involves adding chemicals to destabilize suspended and dissolved solids, supported by flocculation to promote the aggregation of particles. Most suspended particles in water carry a negative charge and remain stable due to electrostatic repulsion. CF neutralizes these charges, enabling the particles to clump together and be effectively removed through sedimentation or filtration [26–28].

A wide range of coagulants have been evaluated for GW treatment, with ferric chloride showing superior turbidity removal due to its ability to form settleable flocs [14,21,29–32]. Recently, hybrid coagulants, combinations of organic, inorganic, and natural polymers, have gained attention for their enhanced performance in wastewater treatment [26, 33]. These formulations improve removal efficiency, reduce sludge production, and lower treatment costs [27,34]. For instance, combining alum and ferric chloride to achieve better removal of suspended solids compared to ferric chloride alone [10].

However, concerns over residual aluminum from alum-based coagulants, linked to potential health risks such as Alzheimer's disease, have prompted a shift toward safer alternatives [35,36]. Hydrated lime has shown promise in enhancing CF process by stabilizing pH, reducing pathogens, and improving floc structure [30,31]. When used with ferric chloride, hydrated lime introduces calcium ions (Ca^{2+}), which improve floc size and aggregation, broaden the effective pH range, and improve sedimentation performance [37].

To further optimize CF, real-time control strategies such as zeta potential (ZP) monitoring are being explored. ZP reflects particle stability and helps determine the optimal coagulant dosage. By identifying the isoelectric point (IEP), the pH at which particles carry no net charge, ZP monitoring enables precise dosing, minimizing chemical residues and reducing treatment costs [20,38,39]. CF performance is also influenced by several operational parameters, including coagulant type and dosage, temperature, pH, mixing intensity, and settling time [27,30,40]. Addressing these factors is essential for developing standardized guidelines for GW treatment.

Despite its potential, greywater reuse remains underregulated, with formal legal frameworks available in only a few countries [23]. This study aims to contribute to the development of regulatory support for GW reuse in Eastern European countries, where it is currently under-represented in both national and EU policies. It also seeks to promote

public acceptance and align with broader water reuse goals [23,41].

This study focuses on optimizing the CF process for BGW, using two affordable chemicals (ferric chloride and hydrated lime) and applying ZP monitoring to empirically delimit optimal dosages and treatment conditions. The goal is to produce treated bathroom greywater (TBGW) suitable for non-potable applications, in support of climate resilience, SDG 6 and EU water reuse regulations [42,43].

2. Materials and methods

2.1. Real bathroom greywater (RBGW) samples

RBGW samples (10 L) were collected from 50 L bathtub volumes across households in Hungary's Northern Great Plain Region over two months, expanding on Bodnar et al. [13]. Samples were collected in acid-rinsed bottles, transported to the laboratory, stored at 4°C, and analyzed within 24 h. Initial parameters of RBGWs are in Table 2.

2.2. Synthetic bathroom greywater (SBGW) samples

SBGW samples were formulated to mimic RBGW, using 0.4 gL⁻¹ shower gel, 0.1 gL⁻¹ shampoo, 0.1 gL⁻¹ corn oil, and 0.155 gL⁻¹ nutrient broth in tap water (40°C), simulating surfactant (10–20 mgL⁻¹) and organic loads resulting from hair and skin surface contamination [26,44, 45]. This approach was based on previous study where SBGW was modelled to represent a diverse range of GW from Hungarian households [21]. Homogenization was achieved via stirring at 700 rpm in a flask for 30 min. Notably, biological contaminants, such as pathogens, were not introduced into SBGW samples, addressing limitations in optimal treatment conditions under non-sterilized and ambient laboratory settings. Initial parameters are in Table 1.

2.3. Analytical and instrumental analysis

Quantitative analysis of all samples was conducted using standardized analytical methods to assess key physicochemical and microbiological parameters. pH was measured with a WTW Multi 3320 pH-meter, ZP and electrical conductivity (EC) values were determined with a Zetasizer Nano Z device (Malvern Instruments, Ltd., Malvern, UK), which also facilitated the determination of the optimal coagulant dosage for each BGW samples. Turbidity (TURB) was measured with a WTW Turb 555IR instrument (WTW GmbH). Biochemical oxygen demand (BOD₅) was determined with an OxiTop IS12 (WTW GmbH, Weilheim, Germany) following a five-day incubation period. Chemical oxygen demand (COD) was measured using the standard potassium dichromate photometric method [46] with a NanoColor Vis Spectrophotometer (Macherey-Nagel GmbH & Co. KG, Düren, Germany), after heating the samples in a thermoreactor (WTW GmbH, Weilheim, Germany) at 148°C for two hours. Total organic carbon (TOC) was analyzed with a Shimadzu TOC-V_{CPN} device (Shimadzu Europe GmbH, Duisburg,

Table 1
General Physicochemical Parameters of SBGW Samples (n = 13; 95% Confidence Interval).

Parameter	Min	Max	Mean	SD	SE	95% confidence interval
pH	7.9	8.5	8.1	0.2	0.06	7.9 - 8.2
TURB (NTU)	12.1	85.6	40.4	23.3	4.9	22.9 - 45.9
ZP (mV)	-31.0	-18.8	-26.4	3.9	1.3	(-28.4) - (-22.1)
EC (mS cm ⁻¹)	0.813	1.06	0.957	0.073	0.03	0.865 - 1.01
COD (mgL ⁻¹)	398	566	510	76	38	389 - 631
BOD ₅ (mgL ⁻¹)	130	260	170	48	16	105 - 275
TOC (mgL ⁻¹)	67.2	86.1	74.2	5.3	1.2	69.1 - 75.8
ANA (mgL ⁻¹)	12.6	17.9	14.3	3.1	1.8	6.63 - 22.0

Germany), with samples pre-filtered through a 0.45 µm membrane filter (Cellulose Nitrate Membrane Filter, Sharlau, Spain). Anionic detergent content (ANA) for GW samples, which reflects the concentration of anionic surfactants, was quantified using the two-phase standard titration method in accordance with standard [47]. Microbial analysis was performed using HygieneChek™ PLUS Dip-Slides (Romerlabs, Getzersdorf, Austria) following standard procedure [48]. Double-sided agar paddles were used to detect *Escherichia coli* (*E. coli*) and Coliforms/Total Count TTC colonies, incubated in a Memmert incubator at 35°C for 24 h (*E. coli*) and 72 h (Coliforms/Total Count TTC). The detection limit for these tests was 10² cfu mL⁻¹.

All measurements were conducted in triplicate, and results are reported as mean values with errors as standard deviation (SD). SD across replicates were below 1% for pH, TURB, ZP, and EC; below 3% for TOC; and below 5% for BOD₅ and COD.

2.4. Characteristics of BGW treatment processes

Three treatment methods were applied either individually or in sequence as part of a modular greywater treatment system, (i) CF using ferric chloride and/or hydrated lime, (ii) sedimentation, (iii) sand filtration. Analytical testing was conducted after each treatment step using consistent methodologies.

Ferric chloride (FeCl₃·6H₂O; reagent grade, 99,8 m/m%; Sharlau, Spain), was prepared as a 25 gL⁻¹ stock solution. Hydrated lime (Ca(OH)₂; puriss. 95 m/m%; Sharlau, Spain) 74 gL⁻¹ (1 molL⁻¹) stock solution was utilized as an enhancing agent.

To determine the base coagulant dose, increasing concentrations of coagulant were tested in 100 mL SBGW samples with focus on ZP, turbidity and TOC removal. Samples were stirred at 100 rpm in 150 mL beakers using a magnetic stirrer. Coagulants were added during stirring and mixed for 30 s, followed by a 5-min sedimentation period. Supernatants were collected using syringes and analyzed. The initial concentrations for FeCl₃ were based on previous study [21], where 200 mgL⁻¹ and 250 mgL⁻¹ were identified as optimum for SBGW treatment, achieving effective ZP neutralization and impurity removal. Similar concentration ranges were tested for Ca(OH)₂, focusing on achieving optimum ZP of 0 ± 5 mV [39,49].

Scale-up tests were conducted using a FC-6S Jar Testing Device (VELP Scientifica, Usmate Velate (MB), Italy) according to ASTM D2035-19 [50] procedure. Parameters such as sedimentation time, slow mixing speed, coagulant type and dose were systematically evaluated. Each test followed a consistent protocol, (i) rapid mixing at 300 rpm, (ii) slow mixing to promote floc aggregation, (iii) sedimentation. Coagulants were added during the rapid mix to optimize performance [26]. The experimental setup consisted of six 800 mL beakers, each containing 500 mL of SBGW. Five beakers were treated with varying concentrations of coagulants based on preliminary testing, while the sixth beaker served as a control with no chemical addition. Ferric chloride followed by hydrated lime were added sequentially using a pipette during the rapid mixing phase (300 rpm for 90 s: 60 s before addition and 30 s after). This was followed by slow mixing at varying speeds (10, 30, or 60 rpm for 15 min) and sedimentation periods of either 30 or 60 min. Supernatant samples were collected 2 cm below the surface using a syringe for analysis. Each condition was replicated in triplicate (n = 3 per dose).

Three experimental batches were conducted, (i) evaluated optimal coagulant combinations using FeCl₃ at 190 mgL⁻¹ and 220 mgL⁻¹ paired with Ca(OH)₂ at 155.40 mgL⁻¹, 207.20 mgL⁻¹ and 251.60 mgL⁻¹, with slow mixing at 60 rpm and sedimentation time at 30 min; (ii) investigated the effect of Ca(OH)₂ dosage (74 mgL⁻¹, 118.40 mgL⁻¹, 148 mgL⁻¹, 177.60 mgL⁻¹, 220 mgL⁻¹) while maintaining FeCl₃ at 220 mgL⁻¹, with slow mixing at 60 rpm slow speed and sedimentation time at 60 min and (iii) assessed the impact of slow mixing speed (30 rpm and 10 rpm) using a fixed Ca(OH)₂ dose of 148 mgL⁻¹ and varying FeCl₃ concentrations (190 mgL⁻¹, 220 mgL⁻¹, 250 mgL⁻¹) with a sedimentation time of 60 min. These investigations altered standard reported procedure in order to

examine the effect of slower mixing speed on floc formation [31,50]. Complete raw data, including dose-response relationships for all measured parameters, are provided in Tables S9–S11. All experiments were performed at room temperature.

For larger-scale testing and pre-filtration, 5 L of SBGW was prepared in 6 L flat-bottom flasks. Coagulants were added during rapid mixing (300 rpm for 90 s), followed by slow mixing (30 rpm for 30 min) and sedimentation (60 min). RBGW samples underwent the same treatment process as 5 L SBGW samples mentioned above. Sample volume was measured using a volumetric flask and transferred to a 6 L flat bottom flask and treated accordingly. Post-sedimentation, supernatants were extracted using a peristaltic pump for subsequent filtration. Filtration was performed using quartz sand with particle sizes ranging from 0.5–1 mm. The sand had a porosity of 73% and a density of 2368 kg/m³. Based on previous studies indicating anaerobic conditions at greater media depths [19], the filter media height was selected within the typical range of 30–100 cm. Fig. 1 illustrates the structure of the applied filtration system, where the height of the sand layer was 60 cm, positioned between two 6 cm gravel layers. The grain size of the gravel was 10 mm, with porosity and density 70% and 2611 kgm⁻³, respectively.

In each case, the removal efficiency was calculated using Eq. (1).

$$E\% = \frac{C_{raw} - C_{treated}}{C_{raw}} \times 100 \quad (1)$$

where: $E\%$ is the removal efficiency, c_{raw} the raw BGW concentration and $c_{treated}$ the treated BGW concentration [43].

2.5. Statistical analysis

All statistical analysis was performed using SPSS software v22 (SPSS Statistics IBM 22, New York, USA). Descriptive statistics (minimum, maximum, mean, standard deviation), were calculated for all physico-chemical and microbiological parameters. SBGW and RBGW samples were compared using an independent samples t-test. Effect sizes were calculated to complement p-values: Cohen's d for pairwise comparisons and η^2 (Tables S1). Bootstrap resampling (B = 1,000) was used to

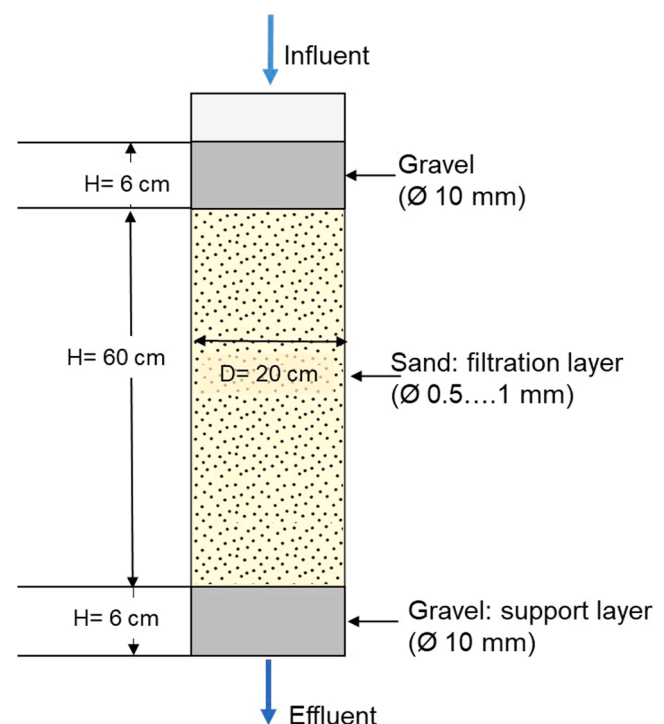


Fig. 1. Set up of filter column used as a post-filtration treatment; Filter media: sand; H=layer height; D=diameter; Ø=diameter of the particles.

compute bias-corrected and accelerated (BCa) 95% confidence intervals for mean differences and raw parameter estimates (Table S2), addressing small sample sizes (SBGW n = 13; RBGW n = 11) and potential non-normality. SBGW and RBGW samples were compared using a non-parametric (Mann–Whitney U tests) test due to the small sample size. Rank-biserial correlations were reported for non-parametric tests (Table S3 and S4). These approaches ensured robust inference despite limited sample sizes and heterogeneity. Spearman’s rank correlation was used to test the relationships between coagulant dose and water quality parameters. One way ANOVA was used to compare means of across water types, with Tukey HSD or Games–Howell post-hoc tests depending on variance homogeneity (Levene’s test, $p > 0.05$). All tests were evaluated at a 95% confidence level ($\alpha = 0.05$). Multivariate statistical analysis (CDA, Canonical Discriminant Analysis) was conducted on different physicochemical parameters (pH, TURB, ZP, EC, COD, BOD₅, TOC and ANA) of the water samples by grouping the sample type.

All analyses were interpreted alongside confidence intervals and effect sizes to emphasize practical significance. Full statistical outputs, including bootstrap intervals, effect sizes, and post-hoc comparisons, are provided in the Supplementary Material (Tables S1–S4, S12–S16).

3. Results and discussion

3.1. Relationship between RBGW and SBGW samples

The tested characteristics of both SBGW and RBGW provided valuable insights into their similarities. SBGW was applied as a substitute for RBGW for limitation of optimal treatment conditions in lab-scale tests [21]. The water quality parameters of SBGW showed well-correlated pH and turbidity values aligning with existing literature data and RBGW samples. The negative ZP values indicate the presence of suspended colloidal particles, consistent with literature [21]. The organic contents of these samples also show correlation, further supporting the classification of SBGW as LGW. In comparison, the physio-chemical characteristics of RBGW, suggest slightly more polluted samples than SBGW, though the values remain within the range of reported BGWs characteristics (e.g. pH, 6.1–8.4; TURB, 37–173 NTU; ZP, -33 mV to -12.2 mV; BOD₅, 90–375 mgL⁻¹; COD, 92–682 mgL⁻¹) [51].

Tables 1 and 2 present the water quality parameters of RBGW and SBGW, respectively, including 95% confidence intervals. Statistical comparisons (Table S1) indicated significant differences for pH ($p < 0.001$; $\eta^2 = 0.598$; Cohen’s $d = 2.5$) and EC ($p < 0.001$; $\eta^2 = 0.811$; Cohen’s $d = 4.2$), while other parameters such as TURB, ZP, COD, BOD₅, TOC, and ANA showed no significant differences ($p > 0.05$). These findings were consistent across BCa bootstrap confidence intervals

Table 2
General Physicochemical Parameters of RBGW Samples (n = 11; 95% Confidence Interval).

Parameter	Min	Max	Mean	SD	SE	95% confidence interval
pH	7.3	7.9	7.6	0.2	0.1	7.4–7.9
TURB (NTU)	10.3	161	59.0	43.0	7.8	20.1 - 57.9
ZP (mV)	-39.7	-12.3	-29.2	7.9	2.8	(-33.7) - (-20.4)
EC (mS cm ⁻¹)	0.657	0.847	0.737	0.064	0.026	0.682 - 0.807
COD (mgL ⁻¹)	241	1122	575	295	98	287 - 752
BOD ₅ (mgL ⁻¹)	85.0	205	141	32.1	13.2	108 - 170
TOC (mgL ⁻¹)	33.6	158	82.0	46.8	16.4	39.7 - 117
ANA (mgL ⁻¹)	10.8	75.3	31.1	24.1	9.5	11.1 - 56.0
E. Coli (cfu mL ⁻¹)	1.0 × 10 ²	1.0 × 10 ⁵	2.1 × 10 ⁴	4.4 × 10 ⁴	1.9 × 10 ²	-3.2 × 10 ² -7.6 × 10 ²
Coliform (cfu mL ⁻¹)	1.0 × 10 ³	1.0 × 10 ³	1.0 × 10 ³	0	-	-
Total Count TTC (cfu mL ⁻¹)	1.0 × 10 ⁴	1.0 × 10 ⁵	4.6 × 10 ⁴	4.9 × 10 ⁴	1.8 × 10 ⁴	-2.1 × 10 ⁴ - 7.8 × 10 ⁴

(Table S2) and non-parametric Mann–Whitney U tests (Table S3–S4), which confirmed robustness despite small sample sizes. Rank-biserial correlations further supported these results, with large effect sizes for pH and EC ($r > 0.80$) and negligible effects for other parameters. The significance results (Table S4) are consistent with previous statistical results, so in case of the examined RBGW and SBGW groups, the difference in pH ($p < 0.001$; $U = 3.0$; $Z = -3.978$; $r = 0.812$) and EC ($p < 0.001$; $U = 2.0$; $Z = -4.027$; $r = 0.822$) values is significant, which is both statistically and practically significant. The differences in TURB ($p = 0.284$; $U = 53.0$; $Z = -1.072$; $r = 0.219$), ZP ($p = 0.192$; $U = 49.0$; $Z = -1.304$; $r = 0.266$), COD ($p = 0.763$; $U = 30.0$; $Z = -0.302$; $r = 0.062$), BOD₅ ($p = 0.301$; $U = 31.5$; $Z = -1.035$; $r = 0.211$), TOC ($p = 0.311$; $U = 54.0$; $Z = -1.014$; $r = 0.207$) and ANA ($p = 0.212$; $U = 12.5$; $Z = -1.247$; $r = 0.255$) values were not significant ($p < 0.05$).

Based on the traditional parameters, the physicochemical characteristics of the SBGWs closely represent those of RBGW, supporting the practical usability of subsequent findings seen in Table S1–S4. RBGWs exhibited higher turbidity, varying significantly between collection points, with a range from 10.3 NTU to 161 NTU and a mean value of 59 (±43) NTU, which is consistent with previous literature [13]. The BOD₅ value was lower, averaging 141 (±32.1) mgL⁻¹ compared to 190 (±54) mgL⁻¹ for SBGWs. The ZP of RBGWs was more negative, averaging -29.2 (±7.9) mV compared to -26.4 (±3.9) mV in SBGWs. The degree of contamination in RBGW was influenced by its source, along with variability in commercially available personal care products, residents’ activity and household characteristic. A permanent composition is not expected even at the same location.

The number of E. coli, Coliform bacteria and Total Count TTC were estimated for only RBGWs with mean values of 2.1×10^4 (±4.4 × 10⁴) cfu mL⁻¹, 1.0×10^3 (±0) cfu mL⁻¹, and 4.6×10^4 (±4.9 × 10⁴) cfu mL⁻¹, respectively.

3.2. Evaluation of the coagulation treatment processes

The coagulation process is a well-established, effective and economical chemical treatment method for GW, widely employed at domestic and industrial scale. It encompasses two key processes, coagulation and flocculation, which are employed to separate dissolved and suspended particles from water. A primary objection of CF is the aggregation of colloidal particles in GW, inducing their destabilization following the addition of a suitable coagulant. These colloidal particles agglomerate during gentle mixing in the flocculation stage, forming flocs, which can grow through adsorption, bridging, electric neutralization, and net-sweeping. Optimum treatment conditions for CF processes depend on various factors including the type of coagulant, dosage, contact time, and mixing conditions, temperature and pH value [30,52].

It is well researched that conventional CF methods often struggle to achieve high removal rates for certain pollutants, such as natural organic matter (NOM) and disinfection by-products (DBPs) [31,53]. To enhance GW treatment efficiency, CF processes must be optimized to improve the removal of organic matter and turbidity by carefully adjusting coagulant dosage, pH, and treatment conditions according to the characteristics of GW [26,54]. Cations, such as Al³⁺, Fe³⁺, Ca²⁺ or Zn²⁺ content agents are used as a neutralizing coagulant to remove colloids from GW, while various polymers are usually applied as flocculants to expand the aggregate network. Enhanced CF methods can be developed by combining these well-known inorganic agents. Lime (Ca(OH)₂) is a cost-effective treatment agent, often used in combination with other inorganic salts to reduce water turbidity. It is affordable, safe, easy to use, and does not capture salts or metals, minimizing leaching while offering strong coliform reduction [55]. Due to its alkaline properties, hydrated lime is widely used for pH adjustment, water softening, and antibacterial effects [10,26]. Studies reported turbidity, color, and COD removals of 77.6%, 73.4%, and 57.0%, respectively, from palm oil mill effluent using a hybrid coagulant of copperas and Ca(OH)₂ and 75% of COD, 94.9% of total phosphorus, and 99.1% of total nitrogen from

swine industry wastewater using $\text{Ca}(\text{OH})_2$ [56,57].

3.2.1. Effect of ferric chloride on SBGW treatment

Using ferric chloride (FeCl_3) as coagulant to treat SBGW demonstrated significant improvements in the water quality. Coagulant dosage is an important factor in determining the optimum conditions for coagulation process. Intrinsically, underdosing or overdosing can lead to reduced efficiency and poor performance, making it essential to determine the ideal dosage to minimize sludge formation and reduce treatment costs [31]. In this study the optimal FeCl_3 dosage was examined by varying the coagulant dosage, while monitoring the ZP values of GW samples. Fig. 2 illustrates the ZP changes versus FeCl_3 concentration, helping to delimit optimal conditions. Results indicated that an FeCl_3 dosage of 250 mgL^{-1} was optimal for SBGWs showing strong correlation with findings from literature [22].

In this preliminary section only, some important physicochemical parameters were assessed to determine the ideal coagulant dosages (Table S5-S8), with evaluations primarily focused on turbidity and TOC removal efficiencies. pH of the GW decreased, ranging from $5.77 (\pm 0.2)$ to $6.31 (\pm 0.3)$, with increasing FeCl_3 volume corresponding to a proportional reduction in pH, due to the acidity of ferric chloride solution. Since a pH below 6 is unsuitable for reuse, the concentration was capped at $275 \text{ mg L}^{-1} \text{ FeCl}_3$, yielding a pH of $5.77 (\pm 0.2)$. All relevant data is provided in Table S2. ZP of the treated samples ranged from $-2.15 (\pm 1.1) \text{ mV}$ to $3.95 (\pm 1.36) \text{ mV}$, falling within the optimal range of $0 \pm 5 \text{ mV}$ [39] indicating the effectiveness of the dosage. While electrical conductivity (EC) increased, the change was not significant. This treatment removed up to $86\% (\pm 2\%)$ of turbidity, however, the turbidity values of TSBGW was less than the 2 NTU threshold for agricultural reuse or another non-potable reuse requirement [43,58]. TOC removal efficiency ranged from $50 (\pm 1) \%$ to $54 (\pm 1) \%$, indicating moderate effectiveness in reducing organic matter.

3.2.2. Effect of lime on SBGW treatment

Hydrated lime ($\text{Ca}(\text{OH})_2$) as coagulant greatly increased the alkalinity of the SBGW samples. With concentrations ranging from $370 - 1850 \text{ mgL}^{-1} \text{ Ca}(\text{OH})_2$, pH rose proportionally from 10.89 to 12.09, exceeding acceptable limits for non-potable reuse [43,58]. Although the ZP values achieved fell within the optimal range, the high EC raised concerns. According to Fig. 3 the optimal dosage, which achieved a ZP of 0 mV , was at 1110 mgL^{-1} lime dose. The full table of results is provided in Table S6. The turbidity also increased substantially, reaching a 1550% rise at 740 mgL^{-1} lime dose, with the highest treatment effectiveness producing a 314% turbidity increase. These findings suggest

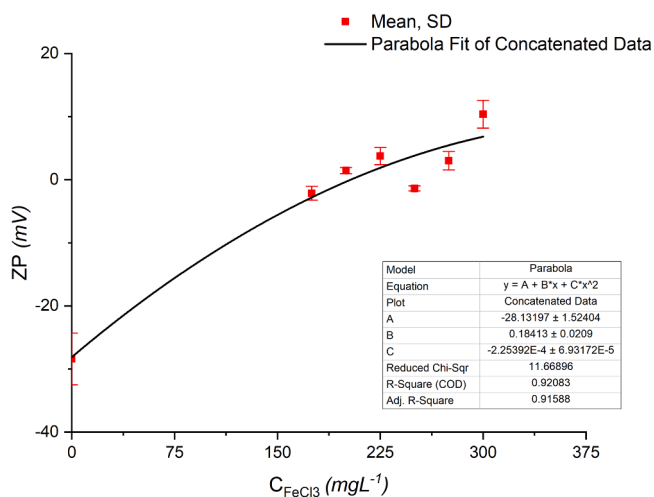


Fig. 2. ZP values as a function of FeCl_3 dosage (Table S5. The confidence level of the fitted curve is 95%).

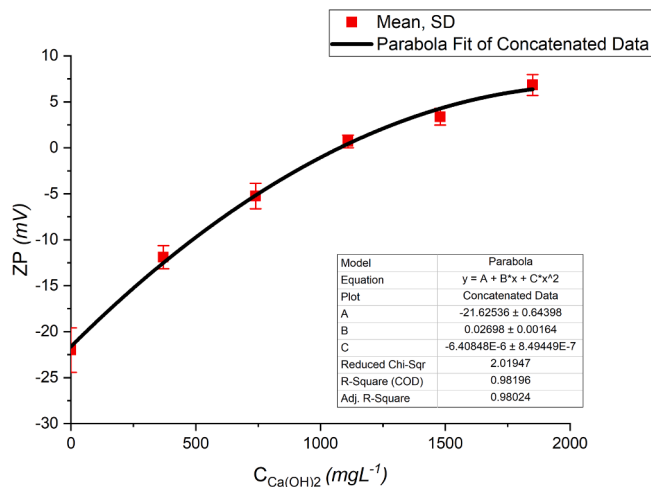


Fig. 3. ZP values as a function of $\text{Ca}(\text{OH})_2$ dosage (Table S6. The confidence level of fitted curve is 95%).

that $\text{Ca}(\text{OH})_2$ alone is not an effective coagulant, as it increased pH value, turbidity and demonstrated poor TOC removal efficiency, with a maximum reduction of only $27 (\pm 3)\%$.

3.2.3. Effect of enhanced coagulation for SBGW treatment

This approach aimed to improve the CF process using cost-efficient coagulants. $\text{Ca}(\text{OH})_2$ fixed at 1480 mgL^{-1} , paired with varying FeCl_3 concentrations for 100 mL SBGW sample, resulted in only slight alterations to SBGW characteristics compared to using $\text{Ca}(\text{OH})_2$ alone. pH remained consistently high above 12, exceeding the threshold for reuse [43,58]. According to Table S7, turbidity reduction varied, reaching up to $60 (\pm 3) \%$, though further testing is needed to establish a definite correlation between FeCl_3 dosage and turbidity removal efficiency. The ZP closest to the optimal value ($5.06 (\pm 1.4) \text{ mV}$) was achieved with a FeCl_3 dose of 200 mgL^{-1} . TOC removal efficiency improved to $47 (\pm 2) \%$ with the addition of FeCl_3 , although this also resulted in a high EC value of $5.51 (\pm 0.98) \text{ mS cm}^{-1}$ at $1480 \text{ mg L}^{-1} \text{ Ca}(\text{OH})_2$ dose.

In contrast, varying $\text{Ca}(\text{OH})_2$ concentration while maintaining a fixed FeCl_3 concentration (Table S8) yielded better results in improving SBGW characteristics. pH improved, ranging from $7.1 (\pm 0.1)$ to $8.13 (\pm 0.4)$, remaining within the reuse limit of < 9 . This combination significantly enhanced turbidity removal, achieving a $99 (\pm 1) \%$ reduction of $0.50 (\pm 0.02) \text{ NTU}$, within the acceptable range for agricultural and urban reuse [43,58]. TOC removal efficiency was slightly lower but remained comparable to that of FeCl_3 alone, with the optimal dosage being $250 \text{ mg L}^{-1} \text{ FeCl}_3$ and $185 \text{ mg L}^{-1} \text{ Ca}(\text{OH})_2$. This dosage also produced a ZP near to net zero, though slightly negative, room for further optimization. The EC at the optimal ratio was $1.25 (\pm 0.09) \text{ mS cm}^{-1}$, indicating an increase in EC due to $\text{Ca}(\text{OH})_2$ addition. Compared to alum and ferrous sulfate, ferric chloride and lime demonstrated superior turbidity removal and were more effective in eliminating colloidal suspended particles [26,59].

Enhanced CF process with FeCl_3 and $\text{Ca}(\text{OH})_2$ for GW improve coagulant effectiveness. $\text{Ca}(\text{OH})_2$ interacted with particles, enhancing floc formation and sedimentation, and stabilizing the pH. Fig. 4 represents the turbidity and TOC removal rates comparing the single treatment agent processes.

Statistical comparisons (independent samples t-test and non-parametric test; Tables S9-S11) showed that FeCl_3 combined with $\text{Ca}(\text{OH})_2$ resulted in significantly greater reductions in TURB and TOC than FeCl_3 alone ($p < 0.05$; $\eta^2 > 0.80$), confirming the enhanced performance of the tested treatment system.

Fig. 5 and Table S12 shows results of Spearman's correlation to test the relationships between coagulant doses, pH, TURB, EC, ZP, TOC and

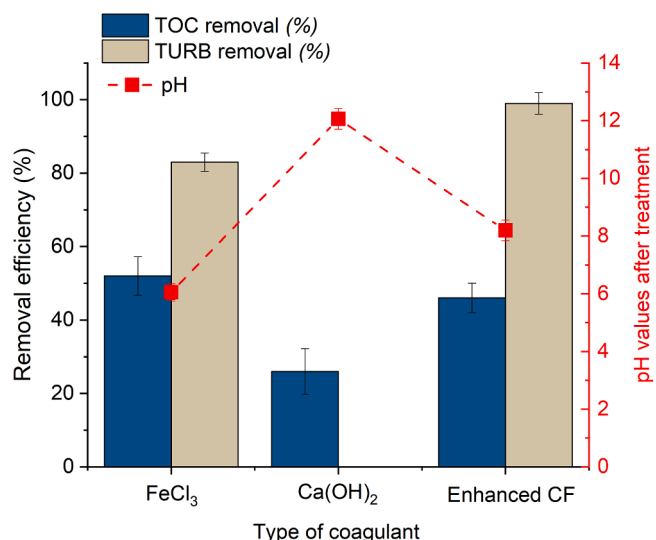


Fig 4. Comparison of pH values with turbidity and TOC removal of different treatments at optimal doses of coagulant (FeCl₃ at 250.00 mgL⁻¹; Ca(OH)₂ at 1110 mgL⁻¹; Enhanced CF: 250.00 mg L⁻¹ FeCl₃ and 148.00 mg L⁻¹ Ca(OH)₂).

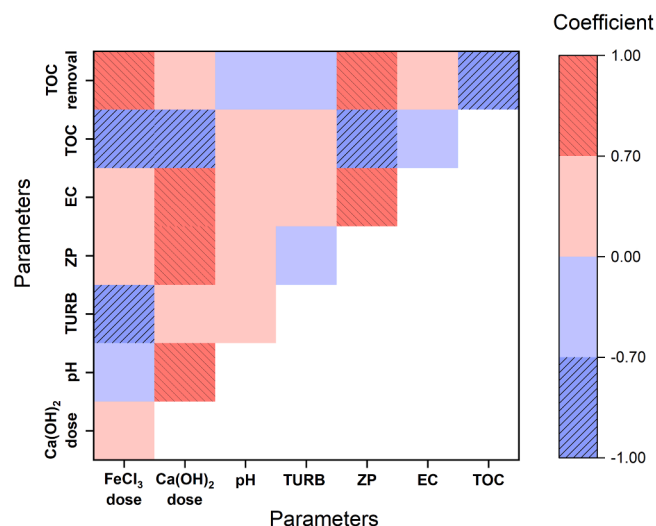


Fig 5. Heat map for Spearman's correlation coefficient between coagulant doses, pH, TURB, EC, ZP, TOC and TOC removal values.

TOC removal rates.

The results show very strong ($\rho > 0.90$) and significant ($p < 0.01$) correlations, mainly between the coagulant doses (FeCl₃ and Ca(OH)₂) and pH, EC, TOC, or TOC removal %. Negative values indicate an inverse relationship, i.e., an increase in one variable is associated with a decrease in the other parameter. In addition, a strong ($\rho > 0.70$) and significant relationship is observed between TURB-FeCl₃ dose; EC-ZP; TOC-ZP values. Therefore, an increase in ZP may result in a decrease in TOC removal % and an increase in EC values. It was shown that ZP measurement actively supports the optimization of the tested method.

3.3. Investigation of the treatment conditions

The jar test experiments revealed that the timing and sequence of coagulant addition significantly influenced floc formation. When FeCl₃ was added before Ca(OH)₂, prior to the slow mixing phase, visibly larger flocs and deposits formed, indicating enhanced coagulation. This configuration also yielded the ZP value closest to neutral, measured at -6.93 ± 0.2 mV, which is within the optimal range for floc stability and

sedimentation. Initial measurements (Table 3) showed that untreated SBGW had a strongly negative ZP (< -30 mV), necessitating careful adjustment of FeCl₃ dosage to achieve effective charge neutralization (Tables S13–S15).

Sedimentation time was another critical factor. Extending the settling period from 30 to 60 min improved treated water quality, with samples showing more favorable ZP values and reduced turbidity. Additionally, the influence of slow mixing speed was assessed by comparing 10 rpm and 30 rpm. A mixing speed of 30 rpm consistently produced better outcomes in terms of pH, turbidity, and total organic carbon (TOC) removal, while ZP remained within the optimal range, indicating stable floc formation.

The optimal coagulant mix for the scaled-up sample was 220 mgL⁻¹ FeCl₃ and 155.40 mgL⁻¹ Ca(OH)₂ for SBGWs, achieving the highest TOC removal of 41 (± 2) % after 30 min of settlement (Table S13). This resulted in pH of 7.4 (± 0.2), turbidity of 1.91 (± 0.65) NTU and ZP of $-2.78 (\pm 0.78)$ mV. However, EC remained high at 1.46 (± 0.08) mS cm⁻¹. Extending the settlement time to 60 min improved turbidity to 0.93 (± 0.09) NTU, with a ZP of $-1.82 (\pm 1.11)$ mV. EC remained relatively high at 1.37 (± 0.09) mScm⁻¹ (Table S14).

Further tests with fixed Ca(OH)₂ at 148 mg L⁻¹, and varying FeCl₃ was does (190 mgL⁻¹, 220 mgL⁻¹, 250 mg L⁻¹) showed that the best results were obtained at 220 mg L⁻¹ and 250.00 mgL⁻¹, particularly at 30 rpm slow speed (Table S15). Increasing FeCl₃ dosage slightly enhanced turbidity removal, reinforcing the importance of adjusting FeCl₃ concentration based on the initial ZP of the raw SBGW. However, while the 250 mgL⁻¹ FeCl₃ and 148 mgL⁻¹ Ca(OH)₂ showed promising results, its ZP was less optimal when mixed at slower speeds compared to 30 rpm.

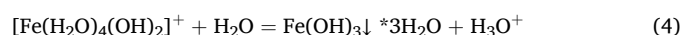
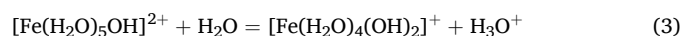
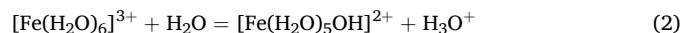
Overall, the jar test validated the optimal coagulant ratio as 250 mgL⁻¹ FeCl₃ to 148 mgL⁻¹ Ca(OH)₂, with slow mixing at 30 rpm and a sedimentation time of 60 min providing the best balance of TOC removal, turbidity reduction, and ZP stabilization.

3.4. Mechanistic model of lime aided ferric chloride CF process

The application of FeCl₃ and Ca(OH)₂ in BGW treatment demonstrated effective pollutant removal through coagulation, precipitation, and adsorption mechanisms. The performance of CF is governed by the kinetics of hydrolysis, metal speciation, surface charge interactions, and flocculation dynamics [30,60].

At the optimal dose, 250 mgL⁻¹ FeCl₃ (1.54 mmolL⁻¹ Fe³⁺) and 148 mgL⁻¹ Ca(OH)₂ (2 mmolL⁻¹ Ca²⁺), positively charged iron complexes formed via hydrolysis neutralized negatively charged colloids and particulate organic matter (POM), promoting the formation of larger, settleable flocs (Fig. 6).

FeCl₃ dissociates into Fe³⁺ ions, which undergo rapid hydrolysis to form intermediate species such as [Fe(H₂O)₅OH]²⁺ and [Fe(H₂O)₄(OH)₂]⁺. These species neutralize negatively charged colloids and particulate organic matter (POM) through charge neutralization and surface complexation, particularly with carboxyl and phenolic groups on natural organic matter (NOM) [30]. Eqs. (2), (3) and (4) show the hydrolysis reactions involved.



The gross reaction is shown in Eq. (5).



Hydrolysis of FeCl₃ produced amorphous Fe(OH)₃ precipitates, which acted as both coagulants and adsorbents. These precipitates effectively captured dissolved organic matter (DOM) and colloidal particles, particularly around pH 8 [31].

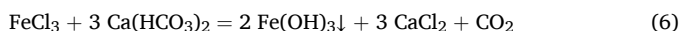
FeCl₃ hydrolysis produced H⁺ (via HCl) lowering pH value of the

Table 3
Comparison of physio-chemical characteristics of treated RBGWs with tap water and untreated samples (n=8; 95% Confidence Interval).

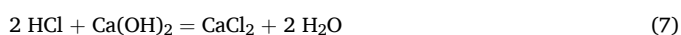
Parameter		TW	SBGW	Unfiltered SBGW	TSBGW	RBGW	TRBGW
pH	Mean	7.6	8.1	7.3	8.2	7.6	8.2
	SD	± 0.3	±0.1	±0.1	±0.1	±0.2	±0.1
	SE	± 0.02	±0.06	±0.04	±0.03	±0.08	±0.07
	CI*	7.5-7.7	7.9 - 8.2	7.1 7.4	8.1 - 8.3	7.4-7.9	8.0-8.4
TURB (NTU)	Mean	0.19	42.2	3.00	0.16	59.0	0.19
	SD	±0.03	±12.5	±1.60	±0.06	±43.0	±0.19
	SE	±0.02	±7.2	±0.94	±0.04	±10.4	±0.03
	CI*	0.11-0.26	7.35-69.6	0.81-8.88	(-0.11) - 0.29	11.4 - 69.2	0.03-0.21
ZP (mV)	Mean	-12.3	-27.6	-7.4	-7.9	-29.2	-6.3
	SD	±3.3	±2.8	±5.5	±3.5	±7.9	±1.8
	SE	±1.89	±1.62	±3.02	±2.03	±3.94	±0.72
	CI*	(-19.6) - (-3.34)	(-34.8) - (- 20.8)	(-21.5) -6.0	(-17.2) - 0.26	(-38.1) - (-16.2)	(-7.52) - (-3.52)
EC (mS cm ⁻¹)	Mean	0.830	1.02	1.39	0.830	0.740	0.720
	SD	±0.05	±0.04	±0.06	±0.02	±0.06	±0.12
	SE	±0.03	±0.02	±0.037	±0.008	±0.038	±0.032
	CI*	0.690 - 0.947	0.925 - 1.14	1.24-1.56	0.790 - 0.866	0.644-0.855	0.587 - 0.763
COD (mg L ⁻¹)	Mean	15.2	470	146	15.1	570	18.4
	SD	±1.5	±90	±1.5	±2.3	±290	±11.6
	SE	±0.9	±51	±0.88	±1.30	±125	±1.6
	CI*	11.5 - 19.1	278 - 719	141-149	10.2 - 21.4	177-872	10.7-19.6
BOD ₅ (mg L ⁻¹)	Mean	5.7	220	57	4.3	140	2.5
	SD	±0.6	±40	±5.8	±1.7	±30	±1.2
	SE	±0.3	±25	±3.33	±1	±16	±0.37
	CI*	4.2 - 7.1	102 - 318	42 - 71	0.3 -8.3	98-189	0.76-2.84
TOC (mg L ⁻¹)	Mean	3.2	73.5	41.2	2.8	82.0	4.5
	SD	±0.4	±4.9	±3.4	±0.6	±48.8	±3.4
	SE	±0.2	±2.8	±1.9	±0.35	±23.3	±0.5
	CI*	1.8 - 3.9	59.2 - 83.5	33.5 - 50.5	1.2 - 4.2	22.7 - 154	2.7 - 6.5
ANA (mg L ⁻¹)	Mean	0	12.5	6.4	0	31.1	0
	SD	±0	±0	±1.0	±0	±24.1	±0
	SE	-	-	±0.7	-	±11.5	-
	CI*	-	-	(-2.8) - 15.8	-	1.86 - 65.5	-
E.Coli (cfu mL ⁻¹)	Mean	20.0	-	-	-	2.1 × 10 ⁴	20.0
	SD	±44.7	-	-	-	± 4.4 × 10 ⁴	± 44.7
	SE	-	-	-	-	±1.9 × 10 ²	± 20
	CI*	-	-	-	-	3.2 × 10 ² - 7.6 × 10 ²	(-35.5) - 75.7
Coliform (cfu mL ⁻¹)	Mean	0	-	-	-	10 ³	2.00 × 10 ²
	SD	±0	-	-	-	±0	± 4.47 × 10 ²
	SE	-	-	-	-	-	2.00 × 10 ¹
	CI*	-	-	-	-	-	(-3.5 × 10 ²) - 7.5 × 10 ²
Total Count TTC (cfu mL ⁻¹)	Mean	-	-	-	-	4.6 × 10 ⁴	10 ³
	SD	-	-	-	-	± 4.9 × 10 ⁴	± 0
	SE	-	-	-	-	± 1.8 × 10 ⁴	-
	CI*	-	-	-	-	(-2.2 × 10 ⁴) - 7.8 × 10 ⁴	-

Note: *_ CI is 95% Confidence Interval.

treated water consuming BGW alkalinity. FeCl₃ further reacts with the bicarbonate to form Fe(OH)₃ that act as flocculants in Eq. (6).

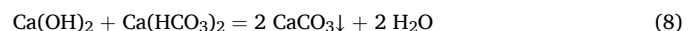


At very low pH, Fe(OH)₃ may redissolve. Addition of Ca(OH)₂ as an enhancing agent improves the Fe(OH)₃ formation by neutralization reaction seen in Eq. (7). Ca²⁺ from Ca(OH)₂ bridges anionic surfactants, raising surface potential and promoting larger flocs via co-precipitation (Ca-NOM, Ca₅(PO₄)₃OH) [52].



This study highlights that Ca(OH)₂ enhances the performance of FeCl₃ in the treatment due to their synergy, so we improved the colloidal and organic matter removal rates from BGWs. Ca(OH)₂ raised the pH and also reacted with the bicarbonate to form calcium carbonate in Eq.

(8), which provides the sludge with better structural integrity and porosity enhancing the sedimentation step.



Fe(OH)₃ precipitates more completely when Ca(OH)₂ raises the pH, and other metal ions (e.g., Pb²⁺, Cu²⁺, Zn²⁺) precipitate as hydroxides, furthermore BGW phosphate content can be precipitated as Ca₅(PO₄)₃OH (hydroxyapatite) or FePO₄.

The kinetics of particle destabilization and floc formation follow pseudo-second-order models, where aggregation rate depends on particle concentration and collision frequency. Rapid mixing increases collision rates, while slow mixing supports floc growth. Speciation studies show that Fe³⁺ primarily exists as hydrated ions in dilute solutions, but in chloride-rich environments, complexes like trans-[FeCl₂(H₂O)₄]⁺ dominate, influencing surface charge and coagulation

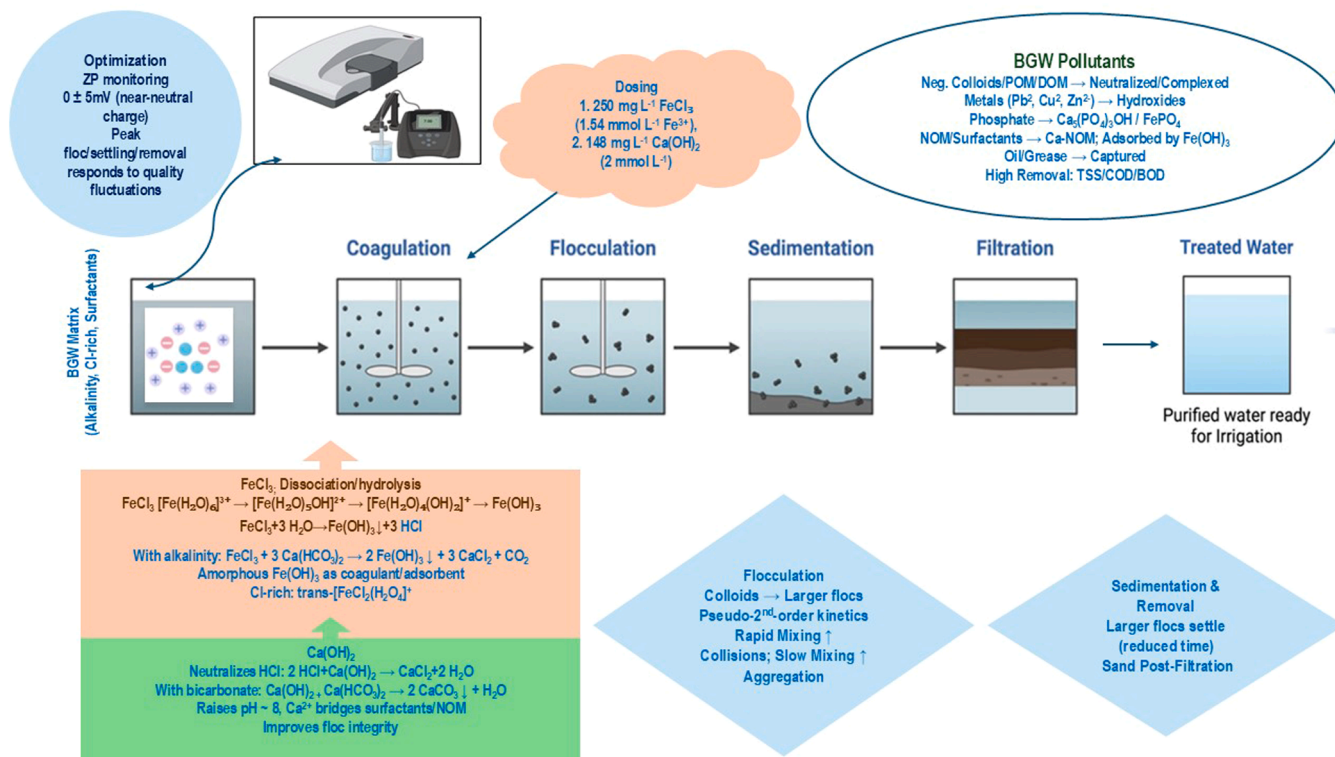


Fig 6. Schematic diagram for enhanced coagulation-flocculation mechanistic model using ZP measurement as a modern analytical solution to delimit the optimal coagulant dose.

behaviour [31,60].

Overall, FeCl₃ provides a strong coagulant via hydrolysis, while Ca(OH)₂ maintains optimal pH and introduces calcium ion for floc enhancement. Together, they enable robust removal of a broad spectrum of pollutants via coagulation, precipitation and adsorption.

Zeta potential (ZP) measurements confirmed that a near-neutral charge (0 ± 5 mV) corresponded with optimal floc formation and settling efficiency. This condition accelerated sedimentation, reduced settling time, and achieved peak TURB removal efficiencies observed 0 ± 5 mV range. Real-time ZP monitoring has been proposed as a control strategy to optimize coagulant dosing and respond to fluctuations in water quality [39].

Final contaminant removal was achieved through sedimentation and post-treatment sand filtration. The combined treatment approach met reuse standards for irrigation, confirming its suitability for decentralized greywater management.

3.5. General physicochemical parameters of treated SBGW and RBGW samples

The optimized treatment process substantially improved water quality, as shown in Table 3, where treated SBGW (TSBGW) and treated RBGW (TRBGW) values closely align with tap water. 95% confidence intervals for all parameters are reported in Tables 1, 2, and 3. Statistical analysis confirmed significant differences between untreated and treated samples for TURB, COD, BOD₅, TOC, and ZP (p < 0.01), while EC changes were minor and non-significant for RBGW (Tables S16–S17). Post-hoc comparisons using Games–Howell and Tukey HSD tests (Table S17) corroborated these findings, indicating that treatment effects were consistent across both synthetic and real greywater.

Maintaining a constant FeCl₃ dose led to overdosing in some RBGW samples, evidenced by a visible yellowish coloration. The degree of overdosing correlated with the intensity of the color, emphasizing the necessity of controlling coagulant dose with ZP of GW before treatment.

Despite this, the overall treatment process was highly effective, with significant improvements observed across all tested parameters. The pH of TSBGW remained consistent, at 8.2 (±0.1) like TRBGW. Fig. 7 shows the removal efficiencies for TURB and organic matter content for both SBGW and RBGW samples. Turbidity of TSBGW was reduced by over 99 (±0.0) %, with an average value of 0.16 (±0.06) NTU, COD removal efficiency exceeded 96 (±1.0) %, with a mean COD value of 15.1 (±2.3) mgL⁻¹, within the TW range of 15.2 (±1.5) mg L⁻¹. BOD removal efficiency reached 98 (±0.4) %, with an average treated value of 4.3 (±1.7) mg L⁻¹. TOC removal efficiency was also high at 97 (±0.9) %. Anionic detergent (ANA) values were zero in both SBGW and RBGW by the applied treatment combination, with no detectable content in the treated samples, demonstrating 100 (±0.0) % removal efficiency.

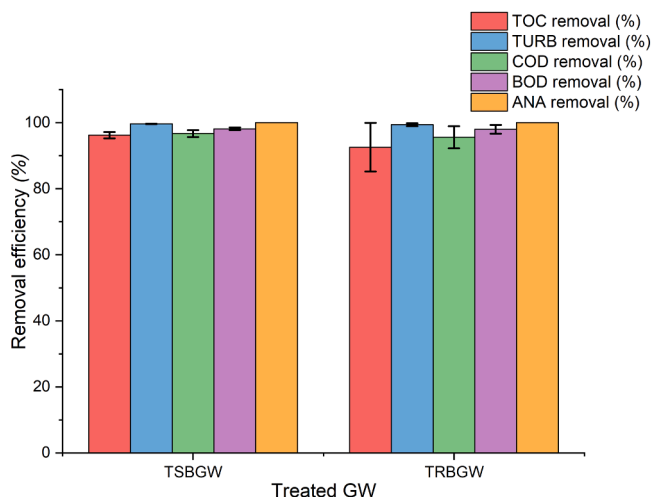


Fig 7. Removal efficiency (TURB, COD, BOD, TOC, ANA) values for TSBGW and TRBGW samples.

For the RBGW samples, additional microbiological tests were assessed the removal of *E. coli*, Coliform bacteria, and Total Count TTC, in line with the new EU regulations [43] which incorporate these parameters into general water quality standards. The removal efficiencies for microbial parameters were consistently high, at 99%. The treatment process effectively improved the microbiological quality of the RBGW, with only one sample failing to achieve the desired removal efficiency. Overall, the TRBGW met the required standards and closely matched TW quality.

TRBGW met or exceeded several key parameters outlined in EU regulations for water reuse in agriculture [43]. The pH value of 8.2 is within the acceptable range of 6 - 9, as specified for water reuse. Turbidity removal efficiency (Fig. 7) was 99 (± 0.5) %, recorded at 0.19 (± 0.19) NTU, demonstrating exceptional clarity comparable to TW, and well below the directive's threshold of < 5 NTU following secondary treatment, filtration, and disinfection, meeting reuse standards [43,58,60]. The mean COD removal efficiency reached 96 (± 3.4) %, confirming the treatment's effectiveness. The average COD value of 18.4 (± 11.6) mgL^{-1} , was within the acceptable range, comparable to TW at 15.2 (± 1.5) mgL^{-1} . However, the treatment was less effective for more contaminated RBGW samples with higher COD values, where efficiency dropped to 89%. BOD₅ removal efficiency exceeded 98 (± 1.3) %, with an average TRBGW value of 2.5 (± 1.2) mgL^{-1} , superior to TW values of 5.7 (± 0.6) mgL^{-1} , indicating excellent BOD₅ removal performance. This level of organic pollutant removal is well within the EU directive's threshold of < 10 mgL^{-1} for agricultural and urban reuse [43]. TOC level of 4.5 (± 3.4) mgL^{-1} also falls within the acceptable range for reuse range, reaching efficiency of 93 (± 7.4) %, although more polluted samples showed a reduced efficiency of 75%. Furthermore, the complete removal of ANA from the TGW highlights the thoroughness of the treatment process, meeting the directive's stringent requirement of < 1 mgL^{-1} . The number of *E. coli* colonies in the treated RBGW complied with the directive's requirement of ≤ 10 cfu 100 mL^{-1} [43], with all but one sample showing undetectable levels of *E. coli*.

The 95% confidence intervals for removal efficiencies were determined as follows: 98.9% - 99.8% for TURB, 92.8% - 98.4% for COD, 96.9% - 99.1% for BOD₅, 86.4% - 98.7 % for TOC, 92.4% - 104% for *E. Coli*, 24% - 135% for Coliform, and 87.5% - 99.7 % for TTC. For microbiological indicators, *E. coli* and TTC reductions were consistently $\geq 99\%$, with only one RBGW sample failing to meet the EU limit. These values demonstrate that, despite batch-to-batch variability, the treatment consistently achieved high performance within narrow uncertainty bounds.

Canonical Discriminant Analysis (CDA) was applied to the water samples, using various physicochemical parameters to differentiate between sample types. As indicated in the CDA biplot in Fig. 8, similarities can be seen based on the physicochemical data of TSBGW, TRBGW and TW. The first function accounts for 77.8% of the discriminating ability of the discriminating variables while the second accounts for 17.7%. The canonical correlation values are 0.987 and 0.948, respectively. The cumulative percentages are 77.8 % (CDA1) and 95.5% (CDA2).

One-way analysis of variance (ANOVA; Table S16) confirmed that all examined water quality parameters showed significant differences between the groups ($p < 0.01$). Based on the effect size (η^2), the largest differences were observed for BOD₅ ($\eta^2 = 0.925$, extremely large effect), EC ($\eta^2 = 0.890$, very large effect) and pH ($\eta^2 = 0.818$, very large effect). The lowest, but still large, effect size was observed for TURB ($\eta^2 = 0.551$). ANOVA analysis showed that there was a difference between the groups in terms of the parameters examined, but it did not indicate between which water types this difference occurred, so we used the Tukey HSD and Games-Howell post-hoc tests (Fig. 9 and Table S17) to determine this.

Significant differences ($p < 0.05$) between the water sample pairs, as follows: pH and EC for 9, TURB for 4, ZP for 8, BOD₅ for 11, COD and TOC for 10. However, the average water quality parameter values (pH, EC, TURB, ZP, EC, COD, BOD₅ and TOC) of the TSBGW and TRBGW

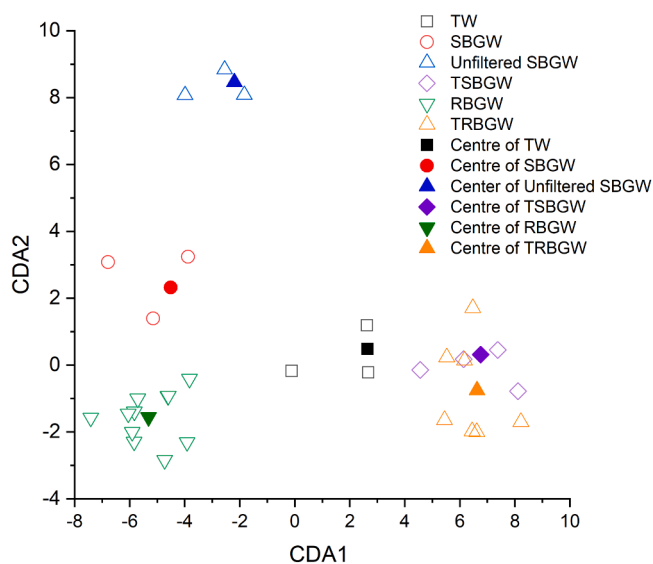


Fig 8. CDA biplot of the different water samples.

samples (Fig. 9, Pair 14) are not statistically different ($p > 0.05$). This confirms that the optimized treatment process is also suitable for real BGW samples.

3.6. Practical implications

The main issues with CF processes are sludge generation and management. While this paper did not evaluate sludge generation parameters, it is well established that FeCl_3 and Ca(OH)_2 produce substantial sludge composed of iron hydroxides, calcium carbonate, and entrapped organic and particulate matter [38]. These solids settle efficiently but increase in volume with higher coagulant dosing, requiring more frequent removal cycles. Sludge characterization is essential for evaluating treatment and disposal options. FeCl_3 -based sludge typically contains high concentrations of Fe_2O_3 , while lime addition contributes CaCO_3 , improving sludge structure and dewaterability [31]. Ferric sludge is known for its gelatinous nature and resistance to dewatering, but lime significantly enhances water release by reducing specific resistance and increasing compressibility. Lime dosages up to 3% of total solids have been shown to facilitate mechanical dewatering. The total sludge generated is 5-10% of treated GW volume [61].

Residual concentrations of iron and calcium in treated greywater are influenced by coagulant dosage and pH. Under optimal dosing conditions (250 mg L^{-1} FeCl_3 and 148 mg L^{-1} Ca(OH)_2), residual levels remain within acceptable limits for non-potable reuse of $\text{Fe} < 0.3$ mg L^{-1} and $\text{Ca} < 40$ mg L^{-1} [62]. However, deviations from these conditions such as overdosing can lead to elevated residuals, posing environmental risks. Continuous monitoring is therefore critical to ensure compliance with reuse standards.

From a disposal perspective, options include landfilling, incineration, and beneficial reuse [52]. Lime-treated sludge offers improved pathogen control and structural integrity, making it suitable for agricultural applications if heavy metal concentrations are within safe limits [63]. However, its alkalinity may restrict land application in sensitive soils, and some regions require regulatory testing. In line with circular economy principles, recent research has explored sustainable sludge reuse strategies, such as recovering coagulants for reuse, modifying sand filters with sludge-derived materials, and developing building materials from treated sludge. These approaches reduce environmental impact and enhance the sustainability of decentralized water treatment systems [3,64–66].

To validate lab-scale findings and ensure real-world applicability, pilot-scale systems are essential. Modular treatment units integrating

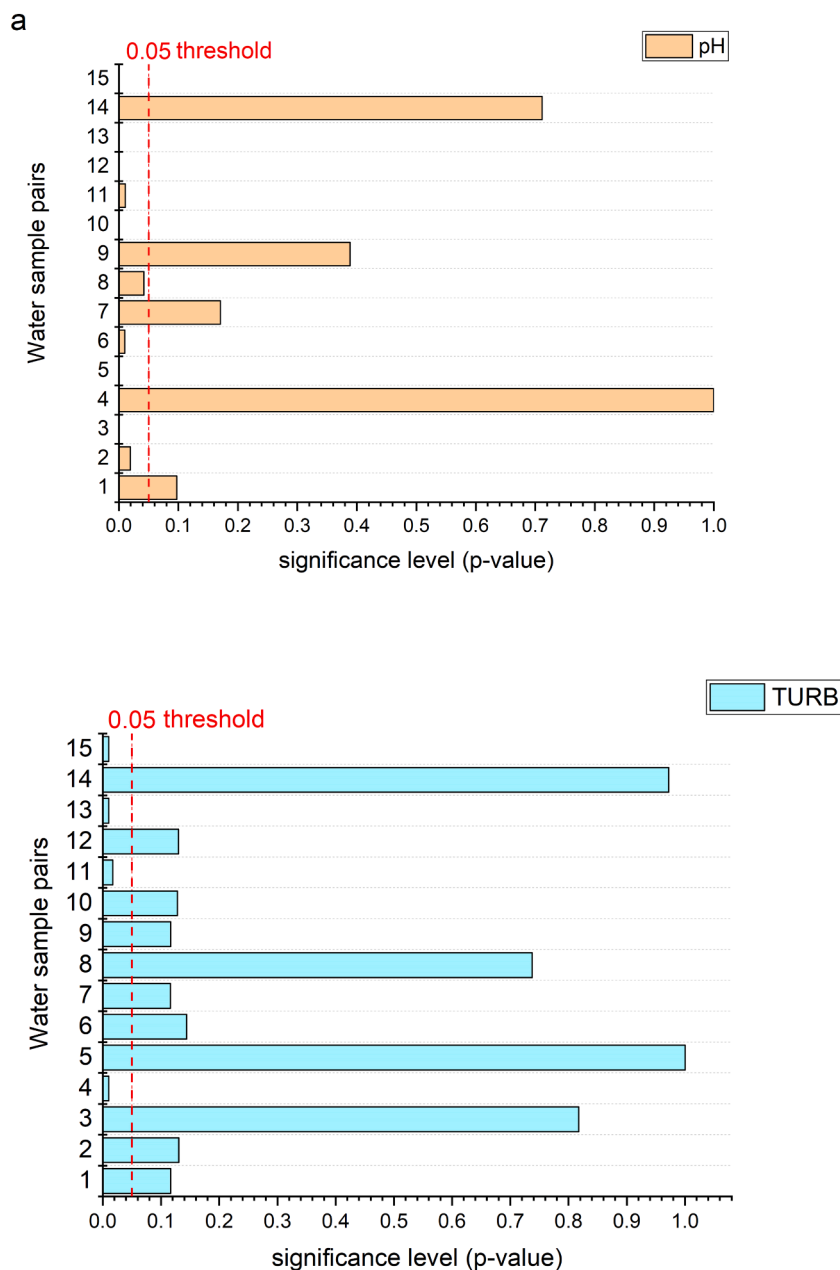


Fig 9. Comparison of water sample pairs in terms of pH (a), TURB (b), ZP (c), EC (d), COD (e), BOD₅ (f) and TOC (g) values based on the significance level (1: TW-SBGW; 2: TW- Unfiltered SBGW; 3: TW-TSBGW; 4: TW-RBGW; 5: TW-TRBGW; 6: SBGW- Unfiltered SBGW; 7: SBGW-TSBGW; 8: SBGW-RBGW; 9: SBGW-TRBGW; 10: Unfiltered SBGW –TSBGW; 11: Unfiltered SBGW –RBGW; 12: Unfiltered SBGW - TRBGW; 13: TSBGW-RBGW; 14: TSBGW-TRBGW; 15: RBGW-TRBGW).

CF, sedimentation, filtration, and UV disinfection have achieved high removal efficiencies for BODs, COD, turbidity, and pathogens. These systems support flexible operation modes, such as fill-and-draw or continuous flow, allowing adaptation to fluctuating influent characteristics. Seasonal variability in greywater composition, driven by changes in temperature, rainfall, and brewery operations, can significantly affect treatment performance. Parameters such as pH, TURB, BOD, and COD vary across seasons, influencing coagulant demand and floc formation dynamics. For example, warmer months may increase organic loading and microbial activity, requiring higher coagulant doses or modified mixing regimes. Adaptive control strategies are therefore essential to maintain consistent treatment outcomes. Online ZP analyzers and advanced control platforms have been successfully integrated into treatment facilities, enabling automated coagulant dosing based on ZP

feedback, reduced chemical consumption, improved effluent quality, extending filter run times and improved sludge dewatering [39,67,68].

Despite effective pollutant removal, post-filtration disinfection is often required to meet health standards for non-potable reuse. Greywater may still contain residual pathogens, necessitating a final disinfection step. Compliance with local standards, including microbial limits, residual disinfectant levels, and setback distances is essential for safe and legal implementation [10].

3.7. Limitations of study and further perspective

A key limitation of this study is the relatively small sample size (SBGW n = 13; RBGW n = 11), which may constrain statistical power and generalizability given the inherent variability of greywater. To

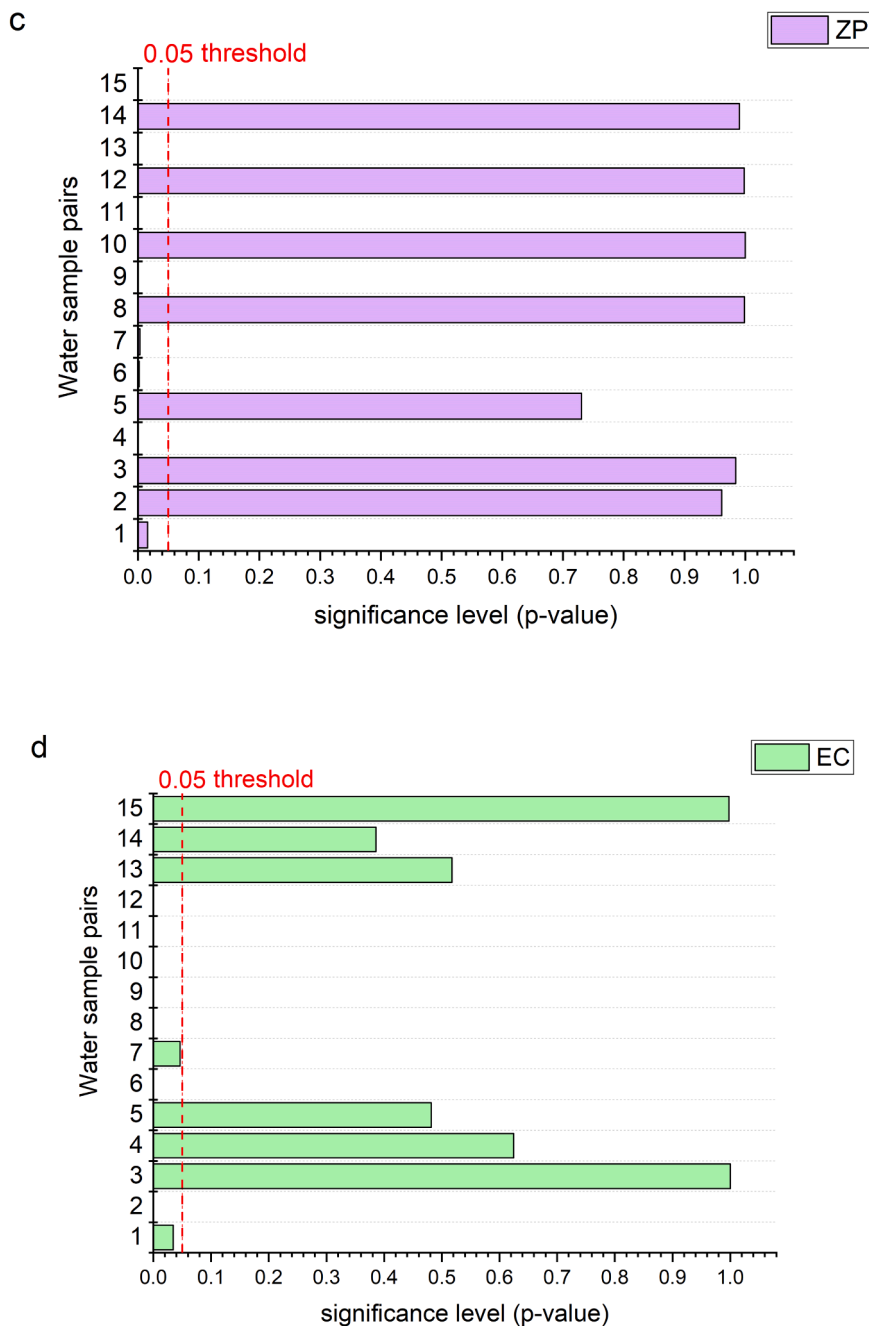


Fig 9. (continued).

mitigate this, we employed a multi-layered statistical approach: (i) replicate-level analysis with low within-condition variability ($SD < 5\%$ for key metrics), (ii) effect size reporting (Cohen’s d , η^2) to capture practical significance (Tables S1 and S13), (iii) non-parametric tests (Mann–Whitney U test) with rank-biserial correlations (Table S4) to confirm robustness under assumption violations, and (iv) BCa bootstrap confidence intervals for mean differences and raw parameter estimates (Table S2). These complementary methods, combined with ANOVA and post-hoc comparisons (Tables S9-S10; S17), provide strong evidence that the observed treatment effects are both statistically and practically significant despite sample size constraints. Future work should validate these findings under larger, more diverse datasets and continuous-flow pilot systems to enhance external validity.

Some other limitations of this study were associated with the bench-scale experiments, the absence of intermediate-scale validation or

continuous flow experiments may not fully reflect operational conditions such as hydraulic retention time (HRT) and influent quality variations. Furthermore, the microbial analysis detection limit (10^2 cfu mL^{-1} for *E. coli* and Coliforms) may be insufficient for stringer reuse standards, potentially underestimating health risks. Other limitations include the lack of life cycle assessment to evaluate environmental impacts, such as carbon footprint and sludge production characteristics.

Several reuse applications are supported by the treatment process in terms of the tested physicochemical parameters, including meeting the stringent turbidity requirement of < 2 NTU set by USEPA and WHO [61]. However, the study did not assess other potential contaminants such as residual chemicals, total suspended solids, or emerging pollutants, which represents a limitation. Further research is needed to evaluate the system’s effectiveness in removing a broader range of pollutants before a comprehensive framework can be considered. The choice of

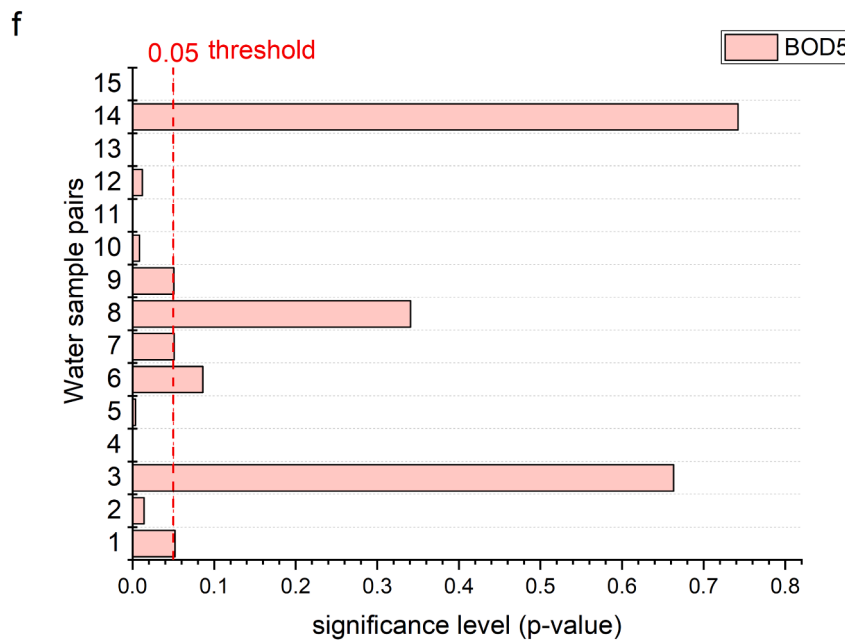
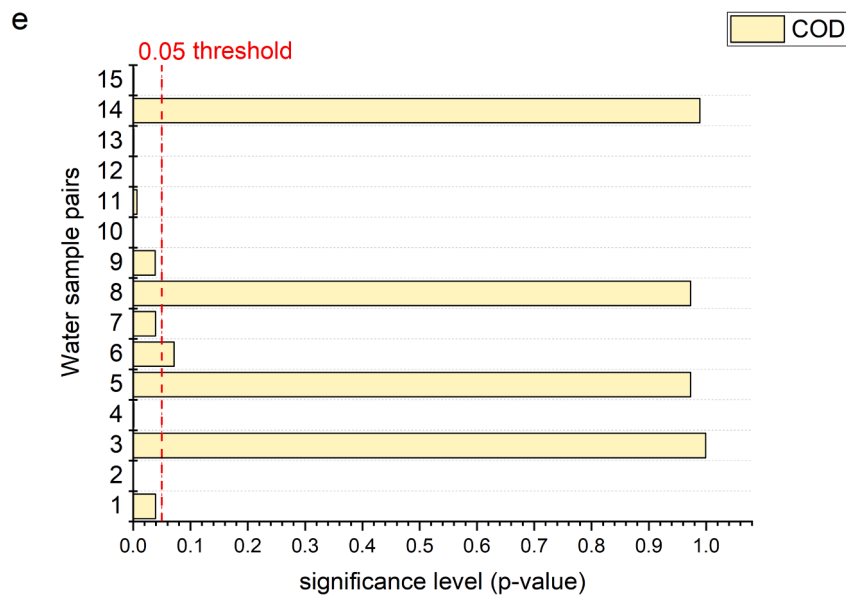


Fig 9. (continued).

reuse application should be purpose-driven, for example, if irrigation is the most relevant use based on local conditions, then the applicable regulatory standards, particularly the more stringent ones, should be prioritized.

Despite these limitations, the study provides a strong foundation for advancing decentralized greywater treatment. Future perspectives include conducting pilot-scale studies with continuous flow regimes to assess scalability and operational dynamics, incorporating dimensional analysis for process optimization. Multivariate techniques, such as response surface methodology (RSM) or artificial neural networks (ANN), could elucidate interaction effects between coagulant doses, mixing intensity, and environmental variables, enabling predictive models for real-time applications. Additionally, exploring automated dosing systems based on real-time ZP monitoring could improve process control and adaptability to influent variations. Finally, testing the enhanced CF process on mixed greywater sources and integrating

advanced filtration media could broaden its applicability, supporting urban water resilience in alignment with EU regulations. These directions will bridge the gap between bench-scale proof-of-concept and practical deployment in European countries.

Future full-scale applications should incorporate these elements to ensure regulatory compliance, operational resilience, and alignment with circular economy principles.

4. Conclusion

This study highlights enhanced coagulation-flocculation with ferric chloride and slaked lime, supplemented with sand post-filtration, as a simple and highly effective solution for GW treatment solution for European countries complying with EU Regulation 2020/741. The process yielded exceptional removal efficiencies in synthetic and real bathroom greywater, with up to 99% turbidity removal, 96% COD removal, 98%

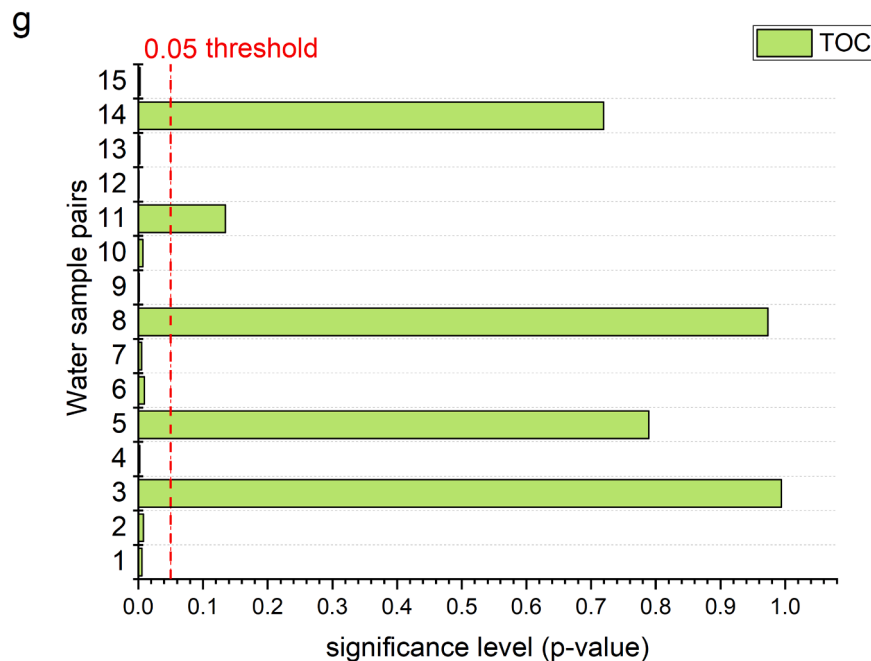


Fig 9. (continued).

BOD₅ removal, 93% TOC removal, 100% ANA removal, and coliform levels reducing to $< 10^2$ cfu mL⁻¹. Treated greywater supported all reuse methods, and if irrigation is the most important according to local conditions, then the corresponding strict regulations should be taken into account. This will be particularly important in the future in connection with climate change. Statistical analyses, including, ANOVA, and Spearman correlation, demonstrated that both FeCl₃ dosage and mixing speed significantly affected treatment performance. Zeta potential emerged as a strong predictor of turbidity removal, was utilized in optimizing coagulant dose. Mechanistic model indicated that charge neutralization and sweep flocculation were primarily driven by Fe(OH)₃ precipitation and complexation/adsorption with dissolved organic matter, enhanced by pH adjustment through Ca(OH)₂ addition. Ca(OH)₂ also acts as an anti-bacterial agent reducing the required post-treatment stages.

By addressing the limitations mentioned above, the tested enhanced coagulation-filtration process can advance sustainable water management and promote a resilient decentralized greywater treatment solution for reuse applications in the EU, reducing the strain on freshwater demand.

Data Availability Statement

All data generated or analyzed during this study are included in this published article.

CRediT authorship contribution statement

Frederick Akpomie: Writing – review & editing, Visualization, Validation. **Andrea Szabolcsik-Izbéki:** Writing – review & editing, Visualization, Validation. **Ildiko Bodnar:** Writing – review & editing, Supervision, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare no conflict of interest. The funder had no role in the design of the study; in the collection, analyzes or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.rineng.2025.107639](https://doi.org/10.1016/j.rineng.2025.107639).

Data availability

Data will be made available on request.

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