sustainability

MDPI

Article

Modelling Treated Laundry Greywater Reuse for Irrigation Using an Affordable Treatment Method and Seed Germination Test

Camila C. A. Cardoso and Ildiko Bodnar *

Department of Environmental Engineering, University of Debrecen, H-4032 Debrecen, Hungary; ccamaracardoso@gmail.com

* Correspondence: bodnari@eng.unideb.hu; Tel.: +36-52-512-900/77828

Abstract: A potential solution in areas facing water shortages is greywater (GW) reuse. GW is produced in bathrooms, laundry rooms, and kitchens of households. With proper treatment, it can be an alternative source for the agriculture sector, which consumes approximately 70% of the world's water. This paper represents the characterization of synthetic laundry GW fraction (LGW), its treatment and modelling of treated LGW reuse for irrigation using a seed germination test. LGW's constant quality (pH = 8.0 ± 0.3, turbidity = 174 ± 73 NTU, BOD_5 = 300 ± 60 mgL^{-1}, TOC = 162 ± 40 mgL^{-1}) is suitable for testing the treatment method's efficiency. Coagulation–flocculation, applying iron(III) chloride and sand filtration as a simple treatment combination, generates good-quality irrigation water (pH = 7.27 ± 0.23, turbidity = 0.6 ± 0.4 NTU, BOD_5 = 17 ± 8 mgL^{-1}, TOC = 16 ± 6 mgL^{-1}). Seed germination tests with different waters, and elemental analysis of water, roots, and stems of the plants were done to verify the plants' quality. The sodium adsorption ratio (SAR) for the raw LGW (SAR = 4.06) was above the threshold (<3) for safe irrigation, thus it is not recommended for this purpose. Based on the elemental analysis results and SAR value of treated LGW (SAR = 2.84), it can potentially be used for irrigation purposes.

Keywords: greywater treatment; laundry fraction; germination; water reuse; water reclamation; sustainability

check for updates

Citation: Cardoso, C.C.A.; Bodnar, I. Modelling Treated Laundry Greywater Reuse for Irrigation Using an Affordable Treatment Method and Seed Germination Test. *Sustainability* **2022**, *14*, 1314. https://doi.org/10.3390/su14031314

Academic Editor: Agostina Chiavola

Received: 17 December 2021
Accepted: 20 January 2022
Published: 24 January 2022

1. Introduction

Water scarcity is a present reality in some countries, and people all over the world can encounter difficulties in access to proper quality water for drinking purposes [1]. The efficient management of water resources is necessary to ensure that people have access to safe water. The United Nations (UN) [2] has defined, as one of its Sustainable Development Goals (SDGs), the need to "Ensure availability and sustainable management of water and sanitation for all" in the SDG6, which is an initiative to encourage governments to provide good sanitation infrastructure. Alternative sources of water are important for agriculture since this sector is responsible for ca. 70% of world water consumption [3]. Greywater (GW) can account for 54–86% of the wastewater produced by a household [4], and its reuse tends to become an important tool to meet the population's need for water [5–7]. GW has different compositions according to its source origin, such as kitchen GW, bath GW, hand basin GW, and laundry GW (LGW).

Dark GW, such as laundry and kitchen sink GW, has a higher concentration of pollutants, so it is more difficult to treat than light GW [8]. Even though GW does not include a contribution from toilets, pathogens can also be present in it [9]. *E. coli* was observed in dark GW; however, it was not observed in real LGW [8]. Blackwater, on the other hand, needs more complex treatment for reuse due to its higher content of organic matter and other pollutants than greywater [8,10].

Synthetic GW can be produced with a composition similar to real GW, but with a constant quality in order to study the best treatment options [11]. When considering the laundry greywater (LGW) portion, common constituents are surfactants and bleach from the detergent and fabric softener [8]. Some other components found in the international literature include glucose, sodium acetate trihydrate, ammonium chloride, disodium hydrogen phosphate, potassium dihydrogen phosphate, magnesium sulphate, cow dung, cellulose, glycerol, etc., with the aim of simulating the suspended solids, surfactants, and chemicals released from the skin [11–13].

GW has been used for irrigation purposes even without treatment; however, it requires attention as it is considered restricted use [14,15]. Untreated GW may contain substances harmful to human health and the environment [16–18], so the challenge is to identify which compounds must be removed from wastewater [19] so that the GW can be treated to meet the quality requirements for irrigation water [6]. These harmful substances include salts, surfactants, oils, and pathogens, which pose a risk to environmental and human health [9,20,21]. Human health effects include serious irritation to the skin caused by the presence of surfactants in the LGW [22]. Rashes are common when the skin comes into direct contact with detergents, and some sensitive individuals may break out after wearing clothes that have been washed with conventional surfactant-containing detergents [23]. Respiratory problems may also result from regular exposure to these chemicals [24], as well as a disruption in endocrine function and interference with hormone balance [25,26]. Environmental effects are related to reducing the soil function and to groundwater pollution [21,27], since a high surfactant content in LGW can increase the salinity and the amount of nitrogen and phosphorus in the soil [28]. Anionic surfactants can also accumulate in the soil after irrigation with LGW [29], and can lead to increased pH and salt levels; a consequence of this is that, after the soil is saturated with greywater, it has a reduced soil hydraulic conductivity even with the use of fresh water [20,30].

There are guidelines set by different countries on agricultural wastewater reuse; some are more restrictive with a higher cost of treatment and others are less restrictive with a lower cost. However, most guidelines are general and do not require a specific type of wastewater treatment; some do not consider pH, emerging pathogens, or salinity [19]. Radingoana et al. [31] conducted an overview of GW as an alternative to increase food production and consequently reduce poverty. It was pointed out that public acceptance can be improved by promoting public awareness; this is as crucial as the technical point of view for widespread adoption [31]. As to the technical perspective, reuse should be practiced for short periods, and finding cheap solutions for treatment would be good for rural areas with low access to water, even in dry seasons [31].

GW can be treated to reach the recommended parameters using physical, chemical, or biological processes [32]. As an alternative to traditional treatment methodologies, nature-based solutions (NBS) are an effective option to treat GW with the use of constructed wetlands, green roofs, and green walls [33]. NBS solutions involve the same challenges as the traditional approach, including public acceptance, treatment efficiency, and economic value [34]. However, chemical treatment using coagulation, magnetic ion exchange resin, photocatalytic oxidation, and granular activated carbon are not as effective as using reverse osmosis and nanofiltration physical treatment, or as constructed wetlands and membrane bioreactor biological treatment [32]. Aerobic biological treatment can help with the removal of surfactants in greywater; however, high amounts of surfactants hinder biodegradation with anaerobic technology [35]. Kamińska and Marszałek were able to remove 97% of anionic surfactants from GW with a sequential biological reactor on aerobic conditions [36]. On the other hand, some surfactants have a more difficult biodegradation process, so the biological treatment of some specific surfactant-rich GW can be a challenge [5].

Chemical treatment using coagulation–flocculation (CF) can be done by the addition of different chemicals such as alum, polyaluminum chloride, ferric chloride or iron(III) chloride, and ferric sulphate [37]. The use of iron(III) chloride seems to involve a higher-turbidity removal than the other three, but the removal efficiency of organic materials is similar when using these four chemicals [37]. These options are economical and have similar operating costs and disposal costs [37]. The use of coagulants enable the colloidal particles on the greywater to aggregate, followed by flocculation, which allows the aggregates to settle down [38]. The aggregation capacity can be measured using a zeta potential measurement [39]. Nyström et al. found that, for raw water, the optimum amount of coagulant could be inferred from a zeta potential between −10 mV to +5 mV, while their study found that for stormwater it can be between −16 mV and 0 mV when adding ferric chloride [38]. The filtration process is widely done using sand layer as the filter medium, due to its potential to remove colloids, suspended solids, and some pathogens [40].

The investigation of the effect of plant irrigation with raw and treated LGW can be done using seed germination tests. There are different seed germination test standards and methods, published by the Association of Official Seed Analysts (AOSA) and the International Seed Testing Association (ISTA), and also by different countries, e.g., the Canadian germination method [41] and the Hungarian Standard MSZ 22902-4 (Water toxicological tests. General specifications) [42].

The elemental analysis of the germination tests allows us to measure the element content in the irrigation water and in the roots and stems of the plants. The elemental analysis can identify the presence of heavy metals, which have the ability to biomagnify to other trophic levels and do not suffer biodegradation, thus are a threat to the environment when they are freely available [43]. They have been identified in animals and humans, causing dangerous health conditions [44]. Even though heavy metal contamination is an environmental and health threat, it is also known that some heavy metals can be favorable to plant growth in trace amounts, such as copper, iron, manganese, magnesium, molybdenum, and zinc [43,45]. When in the soil or irrigation water, the plant can uptake the heavy metals and they will be available in the plant; therefore, using nonedible plants may be useful to reduce the heavy metal concentration in the environment and concentrate on the plant. However, in the case of edible plants, the heavy metals can biomagnify [43,45]. Elements such as sodium, calcium, and magnesium can be used to calculate the sodium adsorption ratio (SAR), which indicates the salinity present in a sample and can indicate whether a water sample is suitable for irrigation [20]. Other elements that should be verified in the irrigation water are heavy metals that can be present in trace amounts, such as Al, Cd, Co, Cr, Cu, Fe, Li, Mn, Ni, Pb, and Zn; also, boron is only beneficial to plants in trace amounts [29,46,47].

In a previous study by our research group, we obtained a comprehensive picture of the quality characteristics of the fractions by analyzing real GW samples [48]. Based on several other studies [12,15,20,49,50], the use of model waters for testing the effectiveness of treatment methods is recommended. All of these provided a good basis for the creation of synthetic LGW samples with a constant composition for this study. In view of the above, the goal of this paper was to create synthetic LGW samples, to investigate the GW treatment method efficiency of combined coagulation–flocculation (CF) and filtration, and to analyze the consequences of germinating white mustard seeds with treated LGW, as well as to verify that the proposed treatment method can produce healthy plants.

2. Materials and Methods

2.1. Synthetic Laundry GW

Synthetic LGW samples were created for routine measurements and treatment. They were prepared by mixing 0.5 g concentrated washing gel, 1.0 g fabric softener, 0.2 g corn oil, and 0.3 g standard nutrient broth for every 1 L of tap water at 40 °C from Debrecen, Hungary. To establish a composition similar to real LGW samples [48], different proportions of the ingredients were mixed and analyzed for several parameters. The analysis of the water sample was conducted via a series of analytical experiments, which include the determination of Biochemical Oxygen Demand (BOD_5), Total Organic Carbon (TOC), pH, Zeta Potential (ZP), turbidity, and Anion-Active (ANA) Surfactant values. The sample homogenization of raw LGW was done using a magnetic stirrer at speed 7 for 15 min.

The treatment sequence was coagulation–flocculation, sedimentation, and filtration on a sand layer. The sample was analyzed postcoagulation and postfiltration using the same analytical methods. The steps of analysis and treatment are given later in this section.

2.2. Analytical Measurements

The BOD_5 value is a measurement to analyze the amount of organic matter in water samples. BOD_5 was measured using OxiTop IS12 measuring equipment (WTW GmbH, Weilheim, Germany) over a period of five days. The TOC value measurement was done using a Shimadzu TOC-Vcpn equipment (Shimadzu Europe GmbH, Duisburg, Germany), while the pH test was made using the WTW Multi 3320 based on a potentiometric electrode measurement. The ZP was measured using a Zetasizer Nano Z device (Malvern Instruments, Ltd., Malvern, UK). This analysis allows us to find the optimum amount of coagulant for LGW samples. The turbidity was measured with a WTW Turb 555IR equipment (WTW GmbH). The detergent concentration was determined by the methodology of the ANA Surfactants, using the so-called Two-Phase Titration Procedure (ISO 2271:1989 standard: Surface Active Agents-Detergents-Determination of Anionic-Active Matter by Manual or Mechanical Direct Two-Phase Titration Procedure [51]). The qualification tests were done for the raw, coagulated, and filtrated samples as treated LGW samples, giving the results as the average of three parallel measurements.

2.3. GW Treatment Processes

Coagulation–flocculation (CF) is a process used for the treatment of wastewater, in which the coagulation step uses a chemical called a coagulant in a rapid mix process and the flocculation step occurs without the addition of chemicals, in a slow mix process. In this study, the coagulant used was iron(III) chloride, the most common and cheapest coagulant, obtained from its hexahydrate form $FeCl_3 \cdot 6H_2O$, which was used in a 25 g L^{-1} stock solution. The preliminary coagulation tests had the objective of finding the optimum coagulant amount by using 100 mL of the LGW sample. The beaker with the LGW sample was put on a magnetic stirrer at a speed of 5. A stopwatch was turned on for 30 s when the coagulant is added to the LGW sample. To measure the ZP of the solution, a 45-mL sample was placed in a small beaker, followed by a 5-min sedimentation. With a syringe, the upper part of the sample was taken for ZP measurement. The test was repeated with an adjusted amount of coagulant until the ZP reached a value of 0 ± 5 mV; the ZP curve can be plotted to show the optimum coagulant amount. For treating a larger sample (5 L), other conditions were used for the coagulation. The CF was followed by a sedimentation step, a rapid mix (90 s), a slow mix, and settling. The flocculation occurred on a slow mix condition using the speed 1 for 15 min and the sedimentation step lasted 20 min. After the sedimentation, the supernatant was removed by a pump for analysis and for the filtration treatment. The filter medium was quartz sand (SiO_2) with a particle size of about 0.5–1 mm, so a high degree of filtration could be achieved due to the extremely small pore diameter. After the flocs settled, the supernatant was pumped into the filter. The filtered LGW was then taken for analysis.

2.4. Seed Germination Test

The seed germination test followed the Hungarian Standard MSZ 22902-4 (Water toxicological tests. General specifications) [42], using white mustard seeds. The analysis was done in terms of visual appearance, number of germinated seeds, as well as length, weight, and elemental analysis of the root and stem part of the plants. The developed plants were cut in order to measure the root and the stem separately.

2.5. Elemental Analysis

The elemental analysis used the inductively coupled plasma optical emission spectrometry (ICP-OES) 5110 Vertical Dual View (Agilent Technologies, Santa Clara, CA, USA) associated with the autosampler Agilent SPS4, with the help of the team at the Agilent

Partner Laboratory. The sample preparation included the wet digestion of the root and stem samples using HNO_3 and H_2O_2. After the digestion, the sample was diluted into a 25.00-mL volumetric flask with ultrapure water before being inserted into the ICP-OES equipment. The high temperature of the ICP-EOS 5110 of 10,000 K allows for measuring the elements Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Na, Ni, Pb, Sr, and Zn. The wavelength for each element is unique, thus allowing the elemental measurement using optical emission spectrometry.

The sodium adsorption ratio (SAR) was calculated using the equation below, in which $\left[Na^+\right]$, $\left[Ca^{2+}\right]$, and $\left[Mg^{2+}\right]$ are concentrations in $mmolL^{-1}$ [46]:

$$SAR = \frac{\left[Na^+\right]}{\sqrt{\frac{\left(\left[Ca^{2+}\right]+\left[Mg^{2+}\right]\right)}{2}}}.$$

3. Results and Discussion

3.1. Greywater Composition and Quality Parameters

To represent the synthetic LGW composition, the mean values for the quality parameters with respective standard deviation are given in Table 1.

Table 1. Comparison between synthetic LGW and real LGW quality parameters.

Component	**pH**	**Zeta Potential (ZP)**	**Turbidity (NTU)**	**BOD$_5$ (mgL^{-1})**	**TOC (mgL^{-1})**	**ANA Surfactants (mgL^{-1})**
Synthetic LGW	8.0 ± 0.3	-34 ± 7	174 ± 73	300 ± 60	162 ± 40	49 ± 6
Real LGW [48]	8.40 ± 1.05	-24.2 ± 8.3	219 ± 126	636 ± 336	266 ± 126 [1]	38 ± 17 [2]
Other studies [8]	5–10	-	34–510	44–3330	-	7–39 [2]

[1] DOC is comparable with TOC. [2] Surfactants measured with the MBAS method.

The synthetic LGW quality parameters are similar to those of the real LGW on all parameters [48]. Other studies did not take ZP measurement into consideration. The anionic surfactant amounts from other studies using the methylene blue active substances (MBAS) photometric method were similar to the values in the current study using the ANA surfactants measurement. The pH measurement and turbidity of the synthetic LGW were within the range found in different studies.

3.2. Coagulation Tests

The coagulation tests were able to produce a so-called ZP curve, represented in Figure 1.

The stability of colloidal systems can be inferred from the electrokinetic potential (zeta potential). The Doppler effect electrophoresis method was used to measure the zeta potential. The purpose of coagulation is to separate unstable colloids. The goal is to reach a zeta potential of 0 mV, i.e., the state in which the van der Waals forces and the chemical binding forces are predominant [48]. The marker in Figure 1 shows the range of the optimum coagulant amount, where the zeta potential value is between 0 ± 5 mV. For the synthetic LGW, we would need an amount between around 295 mgL^{-1} and 325 mgL^{-1} of coagulant in order to efficiently coagulate the sample.

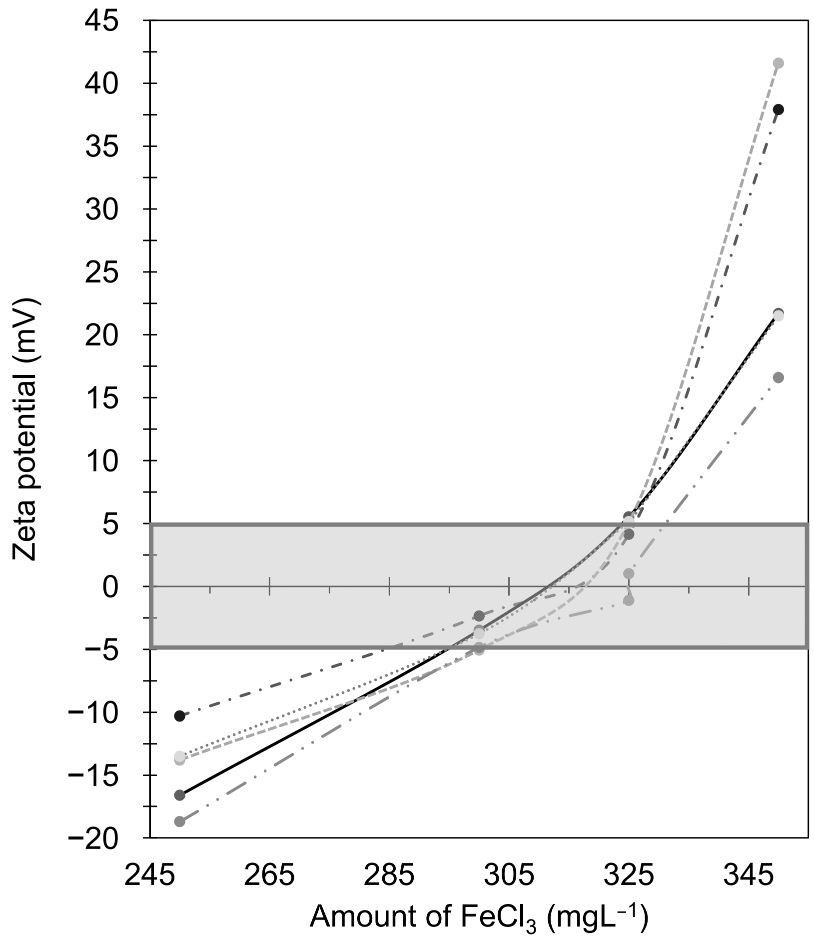

Figure 1. Zeta potential curve for the coagulation tests with 0 ± 5 mV range highlighted.

3.3. Treatment Evaluation

The goal of the treatment evaluation was to develop an affordable treatment method to generate water that has parameters close to the standards for irrigation purposes. The water quality parameters are represented in Figure 2.

The volume used in the irrigation water samples' preparation was 5 L. After adding 312.5 mg $FeCl_3$ per 1 L LGW, the sample had a zeta potential of -2.3 ± 1.0 mV, which is within the optimal 0 ± 5 mV ZP range. The turbidity postcoagulation was reduced to 3.3 ± 0.8 NTU. BOD_5 was also reduced to 112 ± 10 mgL^{-1}, and the surfactants concentration went from 63 ± 42 mgL^{-1} to 7.3 ± 0.3 mgL^{-1}. It was noted during the experiments that flocculation is an important step after the coagulation; otherwise, the coagulates formed are not heavy enough to form sediment, and thus a thin layer of coagulates was observed at the top of the sample after the sedimentation. After filtration, the pH increased to 7.27 ± 0.23, the turbidity became lower than 2 NTU, as expected by most regulations, and the BOD_5 value was 17 ± 8 mgL^{-1}, which has a high standard deviation. The proposed coagulation with $FeCl_3$ reduced the quality parameter values and subsequent filtration with sand filter can generate water with quality parameter values closer to 0, as seen in Figure 2. Table 2 was created to unite the values of water quality for raw LGW, treated LGW (postfiltration), and tap water as a control.

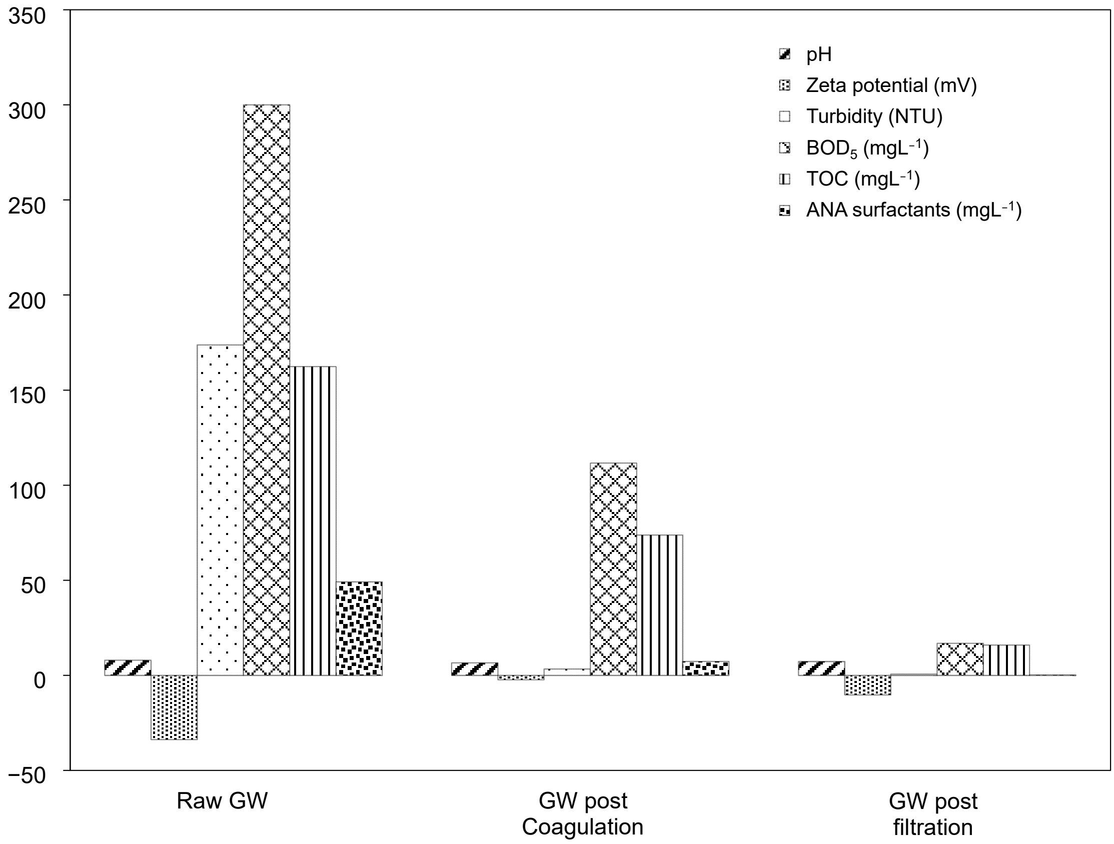

Figure 2. Illustration of changes in water quality parameters as a result of LGW treatment.

Table 2. Comparison between raw LGW, treated LGW, and tap water quality.

Type of Water	pH	Zeta Potential (mV)	Turbidity (NTU)	BOD_5 (mgL^{-1})	TOC (mgL^{-1})	ANA Surfactants (mgL^{-1})
Raw LGW	8.0 ± 0.3	−34 ±7	174 ± 73	300 ± 60	162 ± 40	63 ± 42
Treated LGW	7.3 ± 0.2	−10 ± 3	0.6 ± 0.4	17 ± 8	16 ± 6	0.2 ± 0.5
Tap water	7.8 ± 0.2	−12 ± 4	0.7 ± 0.6	3.0 ± 0.5	2.40 ± 0.02	0

The proposed treatment was able to reduce the BOD_5 values; however, it only approached but did not reach the U.S. EPA and EU standards for agricultural use in every sample, which are 10 mgL^{-1} (weekly) and 10 mgL^{-1} (90% of samples), or 20 mgL^{-1} (maximum weekly), respectively [19]. The turbidity value of treated LGW was 0.6 ± 0.4 NTU, under the U.S. EPA and EU standards of 2 NTU (24-h average) and 5 NTU (90% of the samples), or 10 NTU (maximum), respectively [19]. The treated LGW post-CF and -filtration was noted to have a slight smell of softener and was very clear, visually similar to tap water. There is no legal regulation in Hungary as to the limit for the reuse of treated greywater; therefore, our results presumably approximate the planned water quality for the implementation of irrigation goals, which we verified with germination experiments and elemental analysis.

3.4. Seed Germination Test

The seed germination tests indicated the applicability of raw or treated LGW for irrigation purposes using white mustard seeds. In our tests, irrigation with tap water was performed as a control sample. The 72-h germination test is visually represented in Figure 3.

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

Figure 3. A 72-h germination test. (**a**) Irrigation with tap water; (**b**) irrigation with raw LGW; and (**c**) irrigation with treated LGW.

The irrigation with tap water and treated LGW allowed for good development of the plant, with similar growth and properly germinated and developed roots, stems, and leaves. The irrigation with raw LGW led to a slower growth rate; there was leaf formation on only a few seeds, and the roots were not well developed. The number of germinated seeds after the 72-h germination test is given in Table 3.

Table 3. Mean number of germinated seeds after the 72-h germination test.

Number of Germinated Seeds		
Type of Irrigation Water	**Mean**	**Percentage of Germination**
Tap water	23.6 ± 1.5	94.29%
Raw LGW	22.8 ± 1.2	91.11%
Treated LGW	23.3 ± 1.3	93.33%

The percentage of germination in Table 3 shows that, even with irrigation using raw LGW, more than 90% of seeds germinated. Table 4 summarizes the mean and standard deviation of the root length and stem length.

Table 4. Root and stem length after the 72-h germination test.

Root and Stem Length		
Type of Water	**Root Length (cm)**	**Stem Length (cm)**
Tap water	1.2 ± 0.3	1.2 ± 0.3
Raw LGW	0.5 ± 0.2	0.7 ± 0.2
Treated LGW	1.3 ± 0.4	1.3 ± 0.2

It is clear that the plants grew more intensively after the treated LGW irrigation compared with tap water irrigation, and the raw LGW did not help the plant roots develop so well, not allowing the stem to grow either. The moisture content was calculated for roots and stems to analyze the plant growth features. The moisture content of the roots after irrigation with tap water, raw LGW, and treated LGW was 94.29%, 82.50%, and 94.88%, respectively. The moisture content of the stems after irrigation with tap water, raw LGW, and treated LGW was 71.79%, 56.00%, and 72.50%, respectively. The moisture content in the roots and stem parts helps us understand that the plant has higher water uptake after irrigation with treated LGW than with tap water, which shows that the proposed treatment is promising for plant growth. On the other hand, the moisture content of stems irrigated with raw LGW is very low, due to the high surfactant content of the LGW sample, thus interfering with the water uptake. It is also possible to observe that most moisture content is present in the root part of the plants.

3.4.1. Irrigation Water Quality

The irrigation water was generated on the day of the start of the experiment (0 h). The water quality parameters were measured at time 0 h and after the 72-h germination test, in order to verify whether the water quality was maintained after 72 h. The irrigation water was kept under the same conditions of the germination, at 20 °C, in a dark room with constant aeration. These values are recorded in Table 5.

Table 5. Water quality parameters for the irrigation water.

Water Quality Parameter	Raw LGW		Post-CF [1]	Postfiltration		Tap Water as a Control Sample	
	0 h	72 h	0 h	0 h	72 h	0 h	72 h
pH	8.0 ± 0.2	7.7 ± 0.6	6.7 ± 0.1	7.4 ± 0.3	7.7 ± 0.3	7.8 ± 0.2	7.8 ± 0.3
Zeta potential (mV)	−29 ± 2	−21 ± 4	−4 ± 3	−8 ± 3	−14.0 ± 1.4	−12 ± 4	−12.6 ± 1.4
Turbidity (NTU)	177 ± 19	205 ± 30	3.9 ± 0.4	0.37 ± 0.06	1.5 ± 0.4	0.7 ± 0.6	0.5 ± 0.5
BOD_5 (mgL^{-1})	307 ± 70	280 ± 20	105 ± 9	13.2 ± 0.8	7.0 ± 1.3	3.0 ± 0.5	1.7 ± 0.8
TOC (mgL^{-1})	131 ± 4	116 ± 11	–	21 ± 7	10.5 ± 1.2	2.40 ± 0.02	2.8 ± 0.5
ANA surfactants (mgL^{-1})	46.9 ± 1.4	-	9 ± 2	0	-	0	-

[1] The CF water was measured fresh but was not used for irrigation, nor was it kept for measuring after 72 h.

The turbidity of the raw and treated LGW samples increased after 72 h, while the turbidity of the tap water decreased. However, it is possible to observe a considerable decrease in the BOD_5 for all three types of water, meaning that the raw LGW, treated LGW, and tap water microbial activity was able to decompose the organic matter that is present in the samples. The TOC value was also reduced for the raw and treated LGW; however, it increased slightly for tap water. The ANA surfactants values for tap water and treated LGW were zero at 0 h; therefore, this was not measured after 72 h. The treatment was efficient for the removal of ANA surfactants since it reduced then from 46.9 ± 1.4 mgL^{-1} in the raw sample to 9 ± 2 mgL^{-1} post-CF and 0 mgL^{-1} postfiltration.

3.4.2. Elemental Analysis

The elemental analysis of the water samples had the objective of analyzing the micro and macro elements present in the three types of irrigation water (Table 6). When the amount of an element is below the sensitivity of the ICP-OES equipment, the result is <LoD, which means that it is below the limit of detection of the equipment, but not necessarily null.

The elemental analysis in Table 6 shows that all the elements present in the tap water were also present in the LGW, indicating that these elements may come from the tap water. In fact, when analyzing Table 6, it is possible to observe that all the values that are below the LoD are for the same elements for all types of irrigation water, which include Bi, Cd, Co, Cr, Ni, and Pb; thus, the contribution from the tap water has an impact on the elemental analysis of the LGW. The values are similar for tap water and raw LGW for the macro elements Al, B, Ba, Mn, and Sr, with a slight increase in the raw LGW. The macro elements Ca, K, and Mg have the same tendency, with a small increase in the raw LGW when compared to the tap water; however, Na increased considerably from 29.0 ± 1.4 to 91 ± 2 mgL^{-1}. The metals Cu, Fe, Li, and Zn also underwent a major increase—the value is more than double in the raw LGW compared to the tap water. The applied treatment reduced some of the elements' concentrations (Al, Ba, Cu, Fe, K, Li, Mn, Na and Zn); however, some concentrations increased (Ca, Mg, and Sr) or remained constant (B).

Table 6. Elemental analysis of irrigation water.

Element	Unit	Tap Water	Raw LGW	Treated LGW
Al	(mgL^{-1})	0.020 ± 0.001	0.031 ± 0.007	0.022 ± 0.002
B	(mgL^{-1})	0.06 ± 0.01	0.08 ± 0.02	0.08 ± 0.02
Ba	(mgL^{-1})	0.071 ± 0.004	0.09 ± 0.02	0.06 ± 0.02
Bi	(mgL^{-1})	<LoD	<LoD	<LoD
Ca	(mgL^{-1})	52 ± 2	54 ± 7	69 ± 12
Cd	(mgL^{-1})	<LoD	<LoD	<LoD
Co	(mgL^{-1})	<LoD	<LoD	<LoD
Cr	(mgL^{-1})	<LoD	<LoD	<LoD
Cu	(mgL^{-1})	0.013 ± 0.005	0.0410 ± 0.004	0.0067 ± 0.0006
Fe	(mgL^{-1})	0.0043 ± 0.0006	0.024 ± 0.002	0.005 ± 0.002
K	(mgL^{-1})	3.2 ± 0.6	5.9 ± 0.3	5.2 ± 1.3
Li	(mgL^{-1})	0.009 ± 0.002	0.022 ± 0.007	0.016 ± 0.006
Mg	(mgL^{-1})	12.4 ± 0.4	13.3 ± 0.4	16 ± 2
Mn	(mgL^{-1})	0.0023 ± 0.0006	0.0047 ± 0.0006	0.003 ± 0.001
Na	(mgL^{-1})	29.0 ± 1.4	91 ± 2	71 ± 12
Ni	(mgL^{-1})	<LoD	<LoD	<LoD
Pb	(mgL^{-1})	<LoD	<LoD	<LoD
Sr	(mgL^{-1})	0.238 ± 0.007	0.25 ± 0.03	0.31 ± 0.04
Zn	(mgL^{-1})	0.14 ± 0.05	0.7 ± 0.2	0.03 ± 0.04

The analyzed waters are within the recommended levels of trace elements for irrigation water in terms of Al, Cd, Co, Cr, Cu, Fe, Li, Mn, Ni, Pb, and Zn, as well as for B [46,47]. For the SAR calculation in the irrigation water using Na, Ca, and Mg concentrations, we had results of 1.33, 4.06, and 2.84 for tap water, raw LGW, and treated LGW, respectively. The SAR value was below that recommended by FAO for irrigation (<3) in the case of tap water and treated LGW; however, the raw LGW had a higher value, so there would need to be some restrictions on its use [46]. As a consequence of the plants' uptake of water, the elements were also analyzed in the roots and stems of the plants. The results obtained are in Table 7.

The macro elements in the roots increase after irrigation with treated LGW when compared to both irrigation with tap water and raw LGW. The amounts of Ca and Na were greater in the roots than the stems, and in the case of Mg the highest concentration was in the stem for all irrigation water types. The K concentration was greater in the roots after irrigation with tap water and treated LGW and lower in the roots after irrigation with raw LGW, when compared with the stems. As to the overall macro elements, we observed no pattern of element transport from the root to the stem and no pattern of element retention in the roots. In the case of micro elements, similar to the lack of detection in the irrigation water, the elements Bi, Cd, Co, Cr, Ni, and Pb were not detected in the roots or stems. The Fe concentration was considerably higher in the stem when compared with the roots for all irrigation water types, while the amount of Sr was greater in the roots. The three types of irrigation water led to similar amounts of Al and Mn in the roots and to similar amounts of Al, B, Ba, Cu, and Mn in the stems. The Zn concentration was greater in the roots than the stems after irrigation with tap water and raw LGW.

Table 7. Elemental analysis of root and stem parts according to the irrigation water.

Element	Irrigation with Tap Water (mg kg^{-1})		Irrigation with Raw LGW (mg kg^{-1})		Irrigation with Treated LGW (mg kg^{-1})	
	Root	Stem	Root	Stem	Root	Stem
Al	6.97 ± 2.51	1.90 ± 0.64	6.21 ± 0.51	1.85 ± 0.24	6.24 ± 0.32	1.89 ± 0.65
B	21.39 ± 3.80	6.10 ± 0.24	27.15 ± 1.17	5.45 ± 0.12	22.83 ± 0.33	6.35 ± 0.28
Ba	23.65 ± 3.39	5.50 ± 0.45	30.24 ± 2.82	5.27 ± 0.36	18.63 ± 3.71	4.10 ± 0.30
Bi	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Ca	13,065.27 ± 1400.09	4947.54 ± 446.23	12,678.76 ± 360.34	3902.36 ± 98.95	13,705.60 ± 408.80	4719.23 ± 508.50
Cd	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Co	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Cr	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Cu	11.02 ± 1.89	5.50 ± 0.33	9.88 ± 0.69	5.49 ± 0.28	7.19 ± 0.15	5.26 ± 0.15
Fe	18.45 ± 5.29	73.62 ± 7.73	20.75 ± 1.79	77.77 ± 1.38	22.86 ± 2.40	75.85 ± 1.40
K	8687.12 ± 683.84	6103.09 ± 479.24	5709.04 ± 965.45	7474.34 ± 409.10	9150.36 ± 1022.25	6537.26 ± 348.88
Li	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Mg	2450.56 ± 426.74	3158.31 ± 117.86	2050.42 ± 130.61	3034.38 ± 17.95	2537.13 ± 366.11	3192.08 ± 292.66
Mn	7.44 ± 0.61	24.85 ± 2.07	7.60 ± 0.15	23.63 ± 1.48	7.81 ± 0.41	24.59 ± 1.02
Na	3512.35 ± 648.77	1177.79 ± 381.15	4546.43 ± 983.60	1062.65 ± 406.03	5266.31 ± 1295.40	1422.08 ± 178.86
Ni	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Pb	<LoD	<LoD	<LoD	<LoD	<LoD	<LoD
Sr	56.80 ± 3.89	19.03 ± 1.02	59.50 ± 3.50	15.43 ± 1.10	54.81 ± 5.28	15.45 ± 1.66
Zn	145.03 ± 21.98	70.61 ± 3.41	113.31 ± 15.11	62.87 ± 4.07	42.88 ± 9.43	51.32 ± 2.56

Similar to the macro elements, the micro elements showed no pattern of transportation or retention when comparing the root and stem concentrations. The raw LGW led to a higher concentration of metals than tap water, which is not necessarily detrimental to plant growth, since the elements must be analyzed one by one. The recommendation for irrigation does not take into consideration the concentrations of metals present in the plant. The European Commission recommends a maximum level of lead of 0.1 mg/kg, and 0.20 mg/kg for cadmium, in fresh herbs [52], which is the case for white mustard seeds; thus, the germinated seeds are safe to be eaten in terms of lead and cadmium.

4. Conclusions

Water scarcity affects all continents of our planet, and will only increase due to fast-growing urban areas, as well as climate change and agricultural needs. Greywater, as an alternative source of water, can help address this issue, especially for agriculture, which currently consumes approximately 70% of the world's water.

Untreated GW was previously proven to adversely affect plant growth; therefore, the objective of this paper was to create synthetic laundry greywater (LGW), investigate coagulation–flocculation (CF) and filtration as a proper physicochemical treatment method, and determine the consequences of seed germination with raw and treated LGW. CF, applying iron(III) chloride and sand filtration as a simple treatment combination, was able to produce a treated LGW with quality parameters of pH = 7.27 ± 0.23, ZP = −10 ± 3 mV, turbidity = 0.6 ± 0.4 NTU, BOD_5 = 17 ± 8 mgL^{-1}, TOC = 16 ± 6 mgL^{-1} and ANA surfactants = 0.2 ± 0.5 mgL^{-1}.

The irrigation waters used in the seed germination test were raw LGW, treated LGW, and tap water as a control sample. It was shown that seeds irrigated with treated LGW had similar or greater growth than with tap water. On the other hand, irrigation with raw LGW did not lead to good growth potential due to the high amount of surfactant present in the water sample, which reduced the plant uptake of water. An elemental analysis was done for the root and stem part of the plants, as well as for the irrigation water. The three water samples had micro elements with concentrations below the maximum recommended level; however, the sodium adsorption ratio (SAR) for the raw LGW (SAR = 4.06) was above the recommended value (<3) for irrigation. The treated LGW had good values of micro elements and SAR (2.84), thus can potentially be used for plant irrigation. The analysis of the elements in the roots and stems showed that there was no pattern of element transportation from roots to the stem, as well as no pattern of element retention in the roots.

There is no legal regulation in Hungary to limit the reuse of treated greywaters, so our results presumably approximate the planned water quality for the implementation of irrigation goals.

Author Contributions: I.B. was responsible for the conceptualization and methodology. C.C.A.C. took part in the visualization, review, editing, and experiments validation. I.B. was responsible for the review, supervision, and project administration. All authors have read and agreed to the published version of the manuscript.

Funding: The publication is supported by the EFOP-3.6.1-16-2016-00022 project. The project is co-financed by the European Union and the European Social Fund.

Data Availability Statement: All data generated or analyzed during this study are included in this published article.

Acknowledgments: We would like to thank the Agilent Partner Laboratory and Edina Baranyai for the elemental analysis measurement.

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

References

1. UNDESA. Water and Sanitation. Available online: https://sustainabledevelopment.un.org/topics/waterandsanitation (accessed on 11 September 2020).
2. UNDP. Sustainable Development Goal 6 Targets. Available online: http://www.undp.org/content/undp/en/home/sustainable-development-goals/goal-6-clean-water-and-sanitation/targets (accessed on 11 September 2020).
3. Leonel, L.P.; Tonetti, A.L. Wastewater reuse for crop irrigation: Crop yield, soil and human health implications based on giardiasis epidemiology. *Sci. Total Environ.* **2021**, *775*, 145833. [CrossRef]
4. Roshan, A.; Kumar, M. Water end-use estimation can support the urban water crisis management: A critical review. *J. Environ. Manage.* **2020**, *268*, 110663. [CrossRef] [PubMed]
5. Mahmoudi, A.; Mousavi, S.A.; Darvishi, P. Greywater as a sustainable source for development of green roofs: Characteristics, treatment technologies, reuse, case studies and future developments. *J. Environ. Manag.* **2021**, *295*, 112991. [CrossRef] [PubMed]
6. Al-Mefleh, N.K.; Othman, Y.A.; Tadros, M.J.; Al-Assaf, A.; Talozi, S. An assessment of treated greywater reuse in irrigation on growth and protein content of Prosopis and Albizia. *Horticulturae* **2021**, *7*, 38. [CrossRef]
7. Mohamed, R.M.S.R.; Kassim, A.H.M.; Anda, M.; Dallas, S. A monitoring of environmental effects from household greywater reuse for garden irrigation. *Environ. Monit. Assess.* **2013**, *185*, 8473–8488. [CrossRef]
8. Shaikh, I.N.; Ahammed, M.M. Quantity and quality characteristics of greywater: A review. *J. Environ. Manag.* **2020**, *261*, 110266. [CrossRef]
9. Jung, J.; Fowdar, H.; Henry, R.; Deletic, A.; McCarthy, D.T. Biofilters as effective pathogen barriers for greywater reuse. *Ecol. Eng.* **2019**, *138*, 79–87. [CrossRef]
10. Subramanian, P.S.G.; Raj, A.V.; Jamwal, P.; Connelly, S.; Yeluripati, J.; Richards, S.; Ellis, R.; Rao, L. Decentralized treatment and recycling of greywater from a school in rural India. *J. Water Process Eng.* **2020**, *38*, 101695. [CrossRef]
11. Hourlier, F.; Masse, A.; Jaouen, P.; Lakel, A.; Gerente, C.; Faur, C.; Le Cloirec, P. Formulation of synthetic greywater as an evaluation tool for wastewater recycling technologies. *Environ. Technol.* **2010**, *31*, 215–223. [CrossRef]

12. Abed, S.N.; Scholz, M. Chemical simulation of greywater. *Environ. Technol.* **2016**, *37*, 1631–1646. [CrossRef]
13. Nazim, F. Treatment of Synthetic Greywater Using 5% and 10% Garbage Enzyme Solution. *Bonfring Int. J. Ind. Eng. Manag. Sci.* **2013**, *3*, 111–117. [CrossRef]
14. Radingoana, M.P.; Dube, T.; Mazvimavi, D. An assessment of irrigation water quality and potential of reusing greywater in home gardens in water-limited environments. *Phys. Chem. Earth Parts A B C* **2020**, *116*, 102857. [CrossRef]
15. Chan, C.M.; Mohamed, R.M.S.R. Post-filtration compressibility characteristics of peat used as greywater filter media. *Middle East J. Sci. Res.* **2013**, *17*, 647–654. [CrossRef]
16. Gorgich, M.; Mata, T.M.; Martins, A.; Caetano, N.S.; Formigo, N. Application of domestic greywater for irrigating agricultural products: A brief study. *Energy Rep.* **2020**, *6*, 811–817. [CrossRef]
17. Nolde, E. Greywater reuse systems for toilet flushing in multi-storey buildings - over ten years experience in Berlin. *Urban Water* **2000**, *1*, 275–284. [CrossRef]
18. Islam, M.O.; Khan, R.; Das, A.K.; Akhtar, M.S.; Oki, Y.; Adachi, T. Impacts of industrial effluents on plant growth and soil properties. *Soil Environ.* **2006**, *25*, 113–118.
19. Shoushtarian, F.; Negahban-Azar, M. World wide regulations and guidelines for agriculturalwater reuse: A critical review. *Water* **2020**, *12*, 971. [CrossRef]
20. Travis, M.J.; Wiel-Shafran, A.; Weisbrod, N.; Adar, E.; Gross, A. Greywater reuse for irrigation: Effect on soil properties. *Sci. Total Environ.* **2010**, *408*, 2501–2508. [CrossRef] [PubMed]
21. Abd-Elwahed, M.S. Effect of long-term wastewater irrigation on the quality of alluvial soil for agricultural sustainability. *Ann. Agric. Sci.* **2019**, *64*, 151–160. [CrossRef]
22. Gerster, F.M.; Vernez, D.; Wild, P.P.; Hopf, N.B. Hazardous substances in frequently used professional cleaning products. *Int. J. Occup. Environ. Health* **2014**, *20*, 46–60. [CrossRef] [PubMed]
23. Robinson, M.K.; Kruszewski, F.H.; Al-Atrash, J.; Blazka, M.E.; Gingell, R.; Heitfeld, F.A.; Mallon, D.; Snyder, N.K.; Swanson, J.E.; Casterton, P.L. Comparative assessment of the acute skin irritation potential of detergent formulations using a novel human 4-h patch test method. *Food Chem. Toxicol.* **2005**, *43*, 1703–1712. [CrossRef] [PubMed]
24. Crawford, C.; Zirwas, M.J. Laundry detergents and skin irritancy—A comprehensive review. *Skinmed* **2014**, *12*, 23–31. [PubMed]
25. Sosa-Ferrera, Z.; Mahugo-Santana, C.; Santana-Rodríguez, J.J. Analytical methodologies for the determination of endocrine disrupting compounds in biological and environmental samples. *Biomed Res. Int.* **2013**, *2013*, 1–23. [CrossRef] [PubMed]
26. Roy, J.R.; Chakraborty, S.; Chakraborty, T.R. Estrogen-like endocrine disrupting chemicals affecting puberty in humans—A review. *Med. Sci. Monit.* **2009**, *15*, RA137–RA145. [PubMed]
27. Siggins, A.; Burton, V.; Ross, C.; Lowe, H.; Horswell, J. Effects of long-term greywater disposal on soil: A case study. *Sci. Total Environ.* **2016**, *557–558*, 627–635. [CrossRef]
28. Pinto, U.; Maheshwari, B.L.; Grewal, H.S. Effects of greywater irrigation on plant growth, water use and soil properties. *Resour. Conserv. Recycl.* **2010**, *54*, 429–435. [CrossRef]
29. Wiel-Shafran, A.; Ronen, Z.; Weisbrod, N.; Adar, E.; Gross, A. Potential changes in soil properties following irrigation with surfactant-rich greywater. *Ecol. Eng.* **2006**, *26*, 348–354. [CrossRef]
30. Misra, R.K.; Sivongxay, A. Reuse of laundry greywater as affected by its interaction with saturated soil. *J. Hydrol.* **2009**, *366*, 55–61. [CrossRef]
31. Radingoana, M.P.; Dube, T.; Mazvimavi, D. Progress in greywater reuse for home gardening: Opportunities, perceptions and challenges. *Phys. Chem. Earth Parts A B C* **2020**, *116*, 102853. [CrossRef]
32. Yoonus, H.; Al-Ghamdi, S.G. Environmental performance of building integrated grey water reuse systems based on Life-Cycle Assessment: A systematic and bibliographic analysis. *Sci. Total Environ.* **2020**, *712*, 136535. [CrossRef] [PubMed]
33. Boano, F.; Caruso, A.; Costamagna, E.; Ridolfi, L.; Fiore, S.; Demichelis, F.; Galvão, A.; Pisoeiro, J.; Rizzo, A.; Masi, F. A review of nature-based solutions for greywater treatment: Applications, hydraulic design, and environmental benefits. *Sci. Total Environ.* **2020**, *711*, 134731. [CrossRef] [PubMed]
34. Kotsia, D.; Deligianni, A.; Fyllas, N.M.; Stasinakis, A.S.; Fountoulakis, M.S. Converting treatment wetlands into "treatment gardens": Use of ornamental plants for greywater treatment. *Sci. Total Environ.* **2020**, *744*, 140889. [CrossRef] [PubMed]
35. Khalil, M.; Liu, Y. Greywater biodegradability and biological treatment technologies: A critical review. *Int. Biodeterior. Biodegrad.* **2021**, *161*, 105211. [CrossRef]
36. Kamińska, G.; Marszałek, A. Advanced treatment of real grey water by SBR followed by ultrafiltration-performance and fouling behavior. *Water* **2020**, *12*, 154. [CrossRef]
37. Ucevli, O.; Kaya, Y. A comparative study of membrane filtration, electrocoagulation, chemical coagulation and their hybrid processes for greywater treatment. *J. Environ. Chem. Eng.* **2021**, *9*, 104946. [CrossRef]
38. Nyström, F.; Nordqvist, K.; Herrmann, I.; Hedström, A.; Viklander, M. Laboratory scale evaluation of coagulants for treatment of stormwater. *J. Water Process Eng.* **2020**, *36*, 101271. [CrossRef]
39. Li, H.; Chen, J.; Peng, C.; Min, F.; Song, S. Salt coagulation or flocculation? In situ zeta potential study on ion correlation and slime coating with the presence of clay: A case of coal slurry aggregation. *Environ. Res.* **2020**, *189*, 109875. [CrossRef]
40. Song, S.; Rong, L.; Dong, K.; Liu, X.; Le Clech, P.; Shen, Y. Particle-scale modelling of fluid velocity distribution near the particles surface in sand filtration. *Water Res.* **2020**, *177*, 115758. [CrossRef] [PubMed]

41. SGS Standard Germination Tests. Available online: https://www.sgs.com/en/agriculture-food/seed-and-crop/seed-services/standard-germination-tests (accessed on 28 April 2021).
42. Hungarian Standards Board. *MSZ 22902-4:1990*; Water Toxicological Tests. Seedling Plant Test. Hungarian Standards Board: Budapest, Hungary, 1990; p. 3.
43. Khan, A.H.A.; Kiyani, A.; Mirza, C.R.; Butt, T.A.; Barros, R.; Ali, B.; Iqbal, M.; Yousaf, S. Ornamental plants for the phytoremediation of heavy metals: Present knowledge and future perspectives. *Environ. Res.* **2021**, *195*, 110780. [CrossRef]
44. Zhou, P.; Adeel, M.; Shakoor, N.; Guo, M.; Hao, Y.; Azeem, I.; Li, M.; Liu, M.; Rui, Y. Application of nanoparticles alleviates heavy metals stress and promotes plant growth: An overview. *Nanomaterials* **2021**, *11*, 26. [CrossRef]
45. Shafique, F.; Ali, Q.; Malik, A. Effects of heavy metal toxicity on maze seedlings growth traits. *Biol. Clin. Sci. Res. J.* **2020**, *2020*. [CrossRef]
46. Ayers, R.S.; Westcot, D.W. Water quality evaluation. In *Water Quality for Agriculture*; FAO Irrigation and Drainage Paper; FAO: Rome, Italy, 1985; ISBN 92-5-102263-1.
47. Ayers, R.S.; Westcot, D.W. Miscellaneous problems. In *Water Quality for Agriculture*; FAO Irrigation and Drainage Paper; FAO: Rome, Italy, 1985; ISBN 92-5-102263-1.
48. Bodnar, I.; Szabolcsik, A.; Baranyai, E.; Uveges, A.; Boros, N. Qualitative characterization of household greywater in the northern great plain region of Hungary. *Environ. Eng. Manag. J.* **2014**, *13*, 2717–2724. [CrossRef]
49. Dalahmeh, S.S.; Pell, M.; Vinnerås, B.; Hylander, L.D.; Öborn, I.; Jönsson, H. Efficiency of bark, activated charcoal, foam and sand filters in reducing pollutants from greywater. *Water. Air. Soil Pollut.* **2012**, *223*, 3657–3671. [CrossRef]
50. Dalahmeh, S.S.; Pell, M.; Hylander, L.D.; Lalander, C.; Vinnerås, B.; Jönsson, H. Effects of changing hydraulic and organic loading rates on pollutant reduction in bark, charcoal and sand filters treating greywater. *J. Environ. Manag.* **2014**, *132*, 338–345. [CrossRef] [PubMed]
51. Iran National Standards Organization. *ISO 2271:1989*; Surface Active agents—Detergents—Determination of Anionic-Active Matter by Manual or Mechanical Direct Two-Phase Titration Procedure. Iran National Standards Organization: Tehran, Iran, 1989; p. 6.
52. Commission Regulation (EC) no. 1881/2006. Setting maximum levels for certain contaminants in foodstuffs. *Off. J. Eur. Union* **2006**, *364*, 5–24.