

Article

Onsite Wastewater Treatment Upgrade for Water Reuse in Cooling Towers and Toilets

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Abstract: The increasing population size and housing density are responsible for greater consumption of water resources, causing drinking water shortages in many regions. To reduce water consumption, it is essential to perform wastewater treatment, particularly in onsite non-potable water systems (ONWS). This article discusses the performance of a wastewater treatment system in a shopping mall in Brazil (City of Guarulhos, São Paulo State, Brazil), using data collected over 3 years (2015–2018) that resulted in water reuse ranging from 12 to 42 m³ per day. The strategy used for this wastewater treatment and further reuse in cooling towers and toilets initially included nine steps; after adjustments, an additional step (tertiary decanter) was added. All steps were named as follows: (1) railing; (2) fats boxes; (3) aerobic reactors with selector tank; (4) denitrification; (5) flocculation; (6) secondary decanter; (7) ultrafiltration; (8) disinfection; (9) filtration by zeolites; and (10) tertiary decanter. Based on using FeCl₃ as a flocculant followed by filtration by zeolites (SFM) for ion adsorption and removing above 99% of the biological oxygen demand (BOD₅), generating a final BOD₅ of <2.0 mg/L, total dissolved solids of 130 to 594 mg/L, pH ranging from 6.75 to 7.79, and remaining pathogen-free. This treatment demonstrated the feasibility of reusing water in air conditioning cooling towers and toilets, generating up to 797 m³/month of treated water for reuse with savings of up to 27% in drinking water consumption at the mall.

Keywords: wastewater reuse; onsite non-potable water systems; cooling tower; ultrafiltration; flocculation-adsorptions; commercial-scale reuse



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1. Introduction

Pollution and water shortage are some of the most serious problems worldwide. In 2010, the United Nations General Assembly recognized the human right to water and sanitation; everyone has the right to affordable, safe water for personal and domestic use. Although data from 2017 showed that 71% of the world's population (5.3 billion people) had access to quality drinking water, approximately 2 billion people still use water unfit for human consumption, often contaminated by domestic sewage [1,2]. This finding suggests that the effort to develop new strategies to produce drinking water should be addressed to reduce the social inequality caused by the lack of access to this resource.

Studies have been developed to improve our capacity to remove organic matter and metal ions from wastewater using physical, chemical, and biological approaches to generate scalable production of clean water for different purposes [3–5]. These strategies have been focused on the elimination of unnecessary treatment and onsite production to save water and reduce costs and energy demand for the production of reusable water [6]. Nitrogen

compounds, such as nitrites and nitrates, and phosphates are released in water and wastewater as a result of organic matter degradation and agriculture management. According to the World Health Organization, the number of phosphates and nitrates in water bodies should be below 5 and 50 mg/L, respectively [7], otherwise these salts could result in the eutrophication of water bodies with harmful algae growing [8]. Different strategies, such as adsorption techniques, catalytic reduction, chemical precipitation, electrocoagulation, ion exchange, membrane filters, and reverse osmosis, have been used to remove phosphates and nitrate/nitrite/ammonium from wastewater [9]. Moreover, biological denitrification, such as the “deammonification” process, is carried out by aerobic ammonia-oxidizing bacteria, which degrade ammonia to nitrite and then to nitrate [6] and anoxic denitrifiers [10] have been applied to removing nitrogen in the gas form (N_2) from wastewater. Zeolites have been used in wastewater treatment for cations removing or increasing biological processes such as anaerobic digestion, nitrification, denitrification, and composting [11]. In addition, different types and modified zeolites with changed surface chemistry and pore structure have been evaluated as adsorbents in wastewater treatment [12,13]. The comparison between synthetic zeolites (Zeolite 1–6), natural (clinoptilolite), and engineered zeolite (Zeolite-N) showed differences in mechanical strength, resistance to attrition, and compression, suggesting that future studies should focus on the development of a media with more spherical shape with less sharp edges [12].

In another study, [14] removed efficiently multiple metal ions (K^+ , Mg^{2+} , Ca^{2+} , Ni^{2+} , Co^{2+} , Cu^{2+} , Pb^{2+} , Cd^{2+} , Cr^{2+} , Fe^{3+} , and Al^{3+}) from water, using a Ti–C redox interfaces. In another study, Mahmoud et al. [15] used mechanochemical synthesized graphite oxides as adsorbents to remove methylene blue, acid orange 7, and bisphenol A from contaminated water, and reached at least 93.8% removal in 30 min, suggesting that the development and use of adsorbents for wastewater treatment could be an important strategy to remove these pollutants.

Brazil possesses 12% of all freshwater reserves on the planet; nevertheless, it is also not free from water shortages in several regions. For example, in 2014–2016, the metropolitan region of São Paulo experienced the worst water crisis in its history. This area had to deal with water transport from distant hydrographic basins to meet the needs of 20 million inhabitants [16]. This crisis demonstrated the necessity to reduce freshwater consumption based on the treatment and reuse of wastewater.

The population-related increase in water consumption forced communities to create rational use programs to reduce water consumption and encourage reuse. One compelling approach is implementing safe and effective wastewater reuse projects called onsite non-potable water systems (ONWS) [17]. However, wastewater treatment plants (WWTPs) with reuse are challenging to implement on a commercial scale because there is no control over sewage generation, and there is substantial variation during the day in terms of quantity and quality. This variation depends on the day, the number of customers, the type of discard, and event seasonality [18], complicating establishing a structure that treats this sewage while maintaining water quality and quantity. The consequence of this is that the WWTP is required to treat sewage flow and organic loads (BOD_5) that can reach up to ten times the expected volume and organic matter concentration. In such cases, to be used in cooling towers, quantities of treated effluents must be below limits to avoid creating incrustations or corrosion in the equipment [19,20]. To reuse treated water in air conditioning cooling towers and toilets, it is crucial to monitor quality, to preserve health, equipment, and sanitary devices. Sanitary devices must receive disinfected reuse water free of pathogenic microorganisms and have low levels of coloring to avoid staining porcelain toilets. For reuse in heat exchangers, the primary concern is the transmission of microorganisms such as *Legionella* through the water [21]. Considering that, depending on the season, treated raw sewage varies in terms of physicochemical and biological properties and the amount of some left-over should be controlled to avoid damage to equipment. This variation depends on the habits and customs of the population, and the location of the generation sources that should be treated [19,20].

High consumption levels of foods rich in trans fats cause greater disposal of oils and greases in the sewer networks that need to be retained by devices such as fat boxes [22]. The type of products used to clean kitchens can affect the quality of effluent treated at the WWTP. The constant blockages in the sewage pipes and the increase in the organic load (BOD5) affect the regularity in the production of quality water for reuse [19]. This problem must also be solved during wastewater treatment.

Considering the variation in the sewage volume and amount of organic matter, the purpose of this research was to implement a wastewater treatment capable of removing most of the organic and inorganic loads from sewage produced in a mall located in the city of Guarulhos, São Paulo State, Brazil, and produce reuse water on a commercial scale according to parameters established by the several environmental control bodies [23–25].

The WWTP used in the present study included activated sludge operating under a prolonged aeration modality. Nine steps were implemented: (1) railing; (2) fats boxes; (3) aerobic reactors with selector tank; (4) denitrification (by anoxia condition present in this step, the denitrifying bacteria produce N_2); (5) flocculation; (6) secondary decanter; (7) ultrafiltration; (8) disinfection; and (9) filtration by zeolites. Ultrafiltration was used as the tertiary treatment of the effluent instead of micro- or nanofiltration because the permeate is safer and less expensive [26]. This process retains suspended solids, bacteria, and most viruses, macromolecules, and proteins [27], ensuring an effluent with low turbidity free of solids and sanitized. The importance of the study is that we address several problems (disposal of fats, clogging, necessary cleaning, contaminating loads, and flow of available effluent) and provide solutions to achieve production that varied from 12 m³/day to 46 m³/day. We monitored the quality of reused water within the minimum required parameters because not many studies address or discuss these conditions.

In this context, this study aims, on a large scale, to carry out the development of a system to produce water reuse opportunities in the cooling towers and toilets for a shopping mall in Brazil. The relevance of the results is related to the adjustments made in the WWTP, considering the heterogeneity of the inlet sewage and the necessity to keep the parameters in the outlet reuse water to reduce corrosion and damage to cooling towers.

2. Materials and Methods

2.1. Implementation of a Wastewater Treatment Plant on a Commercial Scale

The WWTP in Guarulhos city, in São Paulo state, Brazil (23°26'36.039'' S and 46°32'23.571'' W) was installed underground and designed to treat up to 5.23 L.s⁻¹ in a prolonged aeration mode [18,28,29], generating 73.62 kg of sludge and 50 m³ of reuse water per day. The process consists of nine treatment stages, separated into four groups (Figure 1); in each stage, one type of pollutant is removed. The four groups were as follows: (I) pre-treatment for the grid, sandbox (for solid residues retention), and oil and grease retention boxes (for fat reduction in the sewage); (II) aerobic reactors (necessary to reduce the carbonaceous organic matter) with denitrification (to remove nitrogenous organic matter by N_2 production); (III) chemical flocculation and secondary settling for coagulant dosage (to reduce residual salts) and sludge separation (remotion of sludge and release the treated water); and (IV) disinfection (for pathogen elimination), ultrafiltration, and filtration by zeolite for the reduction of colloidal particles.

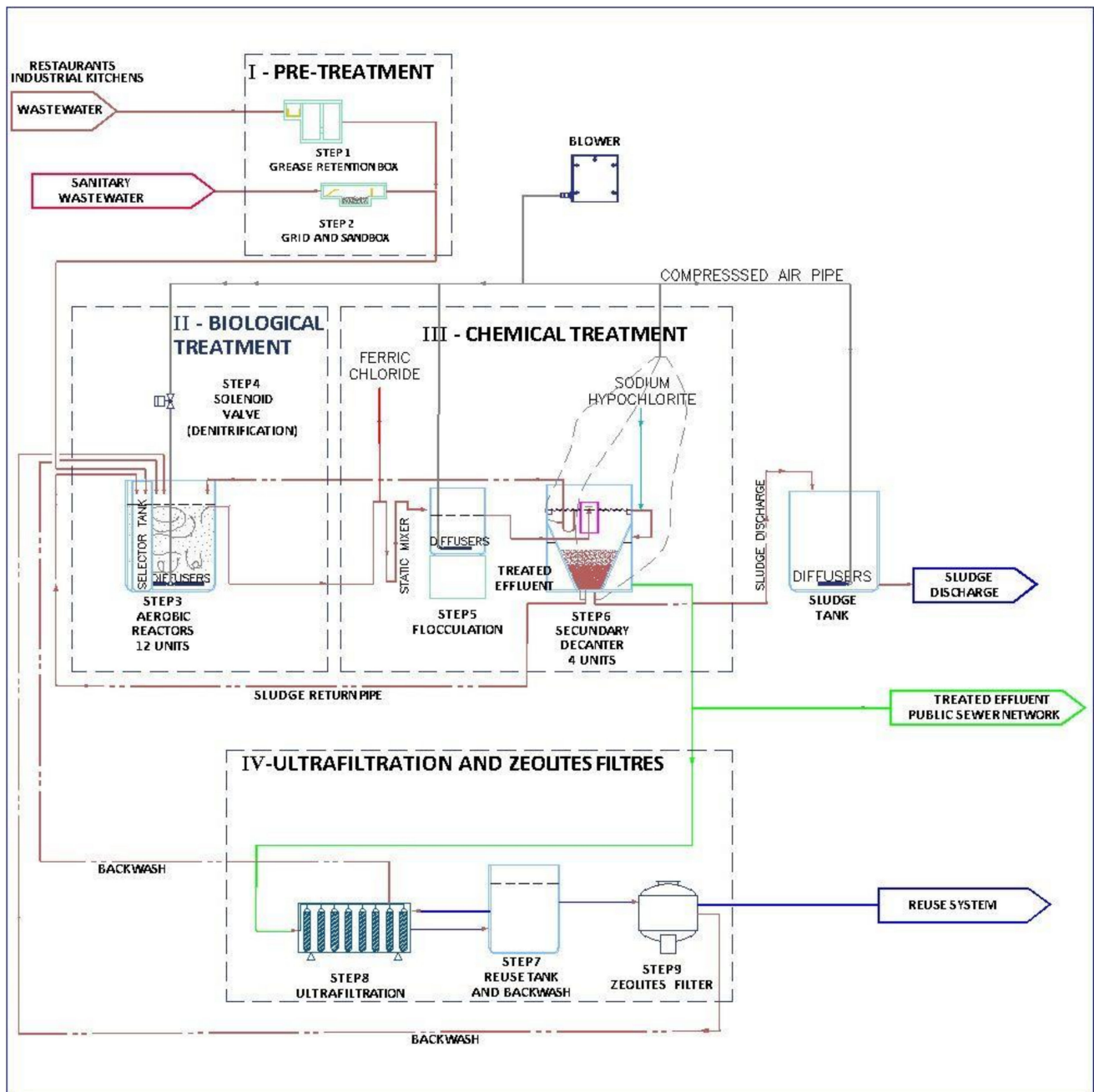


Figure 1. The WWTP with nine steps: 1-grid and sandbox for solid residues reduction; 2-oil and grease retention box to reduce the effect of these fats on the development of the microbial community; 3-aerobic reactors for carbonaceous organic matter degradation; 4-denitrification of organic matter and releasing of N_2 ; 5-flocculation of residual salts; 6-decanting for removing the sludge from the treated water, 7-disinfection to remove pathogens; 8-ultrafiltration; 9-filtration by zeolites for retention of colloidal particles.

The start of the WWTP was considered when the organic matter was successfully degraded. For this, parameters such as pH, total suspended solids (TSS), BOD, chemical oxygen demand (COD), total phosphorus, ammonia (NH_3 and NH_4), and conductivity were monitored weekly in the sewage and the water produced according to methods described in Standard Methods for the Examination of Water and Wastewater [30] or using portable probes as described below (*Analytical Methods*). At this step, the dissolved oxygen (DO) in the aeration tanks (Figure 1—step II) was adjusted between 2 and 4 mg/L, and

300 kg (2 kg/L of volatile suspended solids) of dewatered sludge from a sewage treatment plant were inoculated in the aeration tanks [31]. Due to the excess of surfactants in the sewers, the foam production occurred and was controlled by adding 0.2 mg/L of AFE 1430 antifoam (Dow Brasil Sudoeste Ind., Hortolândia, Brazil) when necessary, reducing the sludge losses.

2.2. Experimental Procedures—JAR Tests

To determine the optimal flocculant treatment (which best removes the salts of nitrogen and phosphorus present in the effluent), 72 experiments in jar test (Jar tester—Policontrol, model Floc Control II, Diadema, Brazil) were evaluated in the laboratory. The jar test is a strategy to evaluate, in controlled conditions, the effect of different reagents in wastewater treatment. For this, we sampled 20 liters of sludge from the aeration reactors (Figure 1) and applied different flocculants and dosages (as described below) and further combined them with filter elements (zeolites and charcoal). After treatment, the effluents were evaluated according to conductivity, total dissolved solids (TDS), pH, clarification (apparent color), and total phosphorus according to the methodology described below (*Analytical Methods*).

The tested flocculants were aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$ —Êxodo Científica Ltd.a., Sumaré, Brazil) (1.25; 2.5; 5.0 and 12.5 mg/L), aluminum polychloride (PAC—Êxodo Científica Ltd.a., Sumaré, Brazil) (1.25; 2.5; and 10.0 mg/L), flocculant based on tannin (Tanfloc—produced by Tanac S.A., Montenegro, Brazil) (27.5; 41.25; 55.0; 68.75; and 82.5 mg/L) and ferric chloride (FeCl_3 —Solução Química, Guarulhos, Brazil) (13.8; 41.4; 69.0; 138.0; 207.0; and 276.0 mg/L) were used as flocculants. For filtering, we tested zeolites (ZN 0410, SMF, and ZE 0325—Watercel line, Celta Brasil Ltd.a., Cotia, Brazil) and anthracite charcoal. Considering that the conductivity is a measure of the ability of water to pass an electrical current, which correlates with the concentration of dissolved salts and other inorganic chemicals, we performed a conductivity analysis to monitor the salt remotion by the addition of different flocculants and a filter system.

2.3. Set-Up of the WWTP Operation

The WWTP was adjusted for producing high-quality wastewater for reuse. The first procedure was to add the system for filtration and regulate the electromagnetic dosing pumps (EMEC BRASIL, São Bernardo do Campo, Brazil, model VCO 0510) to inject flocculant, as defined in the jars test. The flocculant was only added when the influx of raw wastewater was highest (generally between 9 am to 11 pm).

The ultrafiltration equipment (MANN + HUMMEL, model Klar-12, Singapore) was then started up, using pre-filters that had been washed when the pressure reached 0.5 bar to avoid damage to the membranes from solid waste. After filtration, the water was decontaminated with sodium hypochlorite, using a dosing pump (EMEC BRASIL, São Bernardo do Campo, Brazil, model F12 1,5), reducing the biofilm formation after membrane backwash [26].

2.4. Monitoring of the WWTP Operation

The WWTP operation was monitored three times per week by sampling water after the ultrafiltration skid and outlet of the filter and quantifying the sludge production, electrical conductivity, and total phosphorus (in field evaluation). According to the self-monitoring plan, to comply with local legislation [24,25], further analyses were done in an accredited laboratory.

To prevent incrustations and corrosion, the physicochemical and microbiological parameters of the reuse water were adjusted according to the National Water Agency [23], the EPA [32], and the manufacturer's recommended limits for cooling tower replacement water (ALFATERM) and the air conditioning chiller (TRANE) [20]. This flocculant dosage (Table 1) was adjusted according to the maximum daily volume of treated water (m^3/day) in the WWTP. For the operation of the air conditioning chiller, it is possible to dilute the

effluent with drinking water in addition to adjusting the flow rate to be discarded in the purging of the cooling towers.

Table 1. Result of the jar tests with the evaluation of the different flocculants and zeolites and the efficiency in the removal of the SDT according to the variation of the conductivity in the coagulated and filtered effluent.

Flocculant/Coagulant	Dosage (mg/L)	Before Treatment ($\mu\text{S/cm}$)	After Jar-Test Treatment ($\mu\text{S/cm}$)	ZN 0410 ($\mu\text{S/cm}$)	Coal ($\mu\text{S/cm}$)	SFM ($\mu\text{S/cm}$)
Aluminum Sulfate $\text{Al}_2(\text{SO}_4)_3$	12.5	615	617	−10%	8%	−10%
	5.0	615	618	−10%	8%	−12%
	2.5	615	617	−8%	10%	−13%
	1.25	615	618	−10%	8%	−14%
PAC–Aluminum Polychloride	10.0	582	601	−4%	3%	−3%
	2.5	582	599	−1%	15%	−2%
	1.25	582	600	0%	15%	−2%
	82.5	582	601	−3%	65%	45%
Tannin (Tanfloc)	68.75	582	599	−2%	65%	45%
	55.0	582	602	−4%	19%	−6%
	41.25	582	595	−3%	56%	4%
	27.5	582	591	−4%	36%	−3%
Ferric Chloride (FeCl_3)	276.0	615	616	−7%	10%	−3%
	207.0	616	568	−8%	12%	−6%
	138.0	615	559	−9%	20%	−5%
	69.0	615	549	−11%	13%	−8%
	41.4	615	539	−12%	12%	−14%
	13.8	615	537	−13%	10%	−13%

2.5. Analytical Methods

Throughout the WWTP operation, the physicochemical parameters of the incoming sewage and treated outgoing water were evaluated with portable tests. The analyses of the total ammoniacal nitrogen concentration (dissolved ammonia $[\text{NH}_3]$ and ammonium ion $[\text{NH}_4^+]$) were carried out using colorimetric tests (Prodimest NH_3 and NH_4 [Prodac International, Pádua, Italy]), with visual comparison using a color pattern of 0.0 to 5.0 mg/L. For nitrate (NO_3), the visual was performed using a Checker Desk with the reagent HI 38050-0 (Hanna Instruments, Nufalau, Romania) in the range of 0 to 50 mg/L. Sludge production was measured by decanting 1 L of the sludge for 30 min. Other parameters were evaluated using a portable probe: apparent color (HI 727 portable meter, Hanna Instruments, Nufalau, Romania); pH (Model Q400BC, Quimis, Diadema, Brazil); conductivity (MS Tecnopon, Piracicaba, Brazil, Model mCA-150P, calibrated in the range from 0.0 to 10,000 $\mu\text{S/cm}$); DO (oximeter Model HI 9146, Hanna Instruments, Nufalau, Romania); and total phosphorus (Model HI 706, with reagents HI706AS and HI706B-0, Hanna Instruments, Nufalau, Romania) and a laboratory accredited by the Environmental Agency of State of São Paulo (CETESB) [24,33,34]. The evaluated parameters (Table 2) were in agreement with the Environmental Agency which is responsible for the inspection of the procedures and effluent released into water bodies. Due to this inspection, all tests were performed in a laboratory accredited by the Environmental Agency of the State of São Paulo (CETESB) [24,33,34]. The BOD and COD analyses depend on the correct dilution of samples in an aqueous solution to build a calibration curve. For this, the raw sewer BOD and COD were determined, the wastewater homogenized and diluted and subsamples with known concentrations were generated. Considering that the wastewater presented an initial COD of ~1200 mg/L, dilutions in distilled water presenting COD of 1250; 1000; 750; 500; 250, and 50 mg/L were prepared and used to build the calibration curve. For BOD, appropriate dilutions were prepared to contain 500; 250; 100; 50; 25; 10, and 5 mg/L and used to prepare the calibration curve.

Table 2. Reference parameters and results obtained during monitoring of wastewater treatment.

Parameters (Max)	Unit	Limits Determined by Agencies			Analysis Date				
		ANA	EPA	Manu-Factor	1 December 2015	25 July 2016	8 November 2016	7 February 2017	2 February 2018
TDS	mg/L	500	500	2800	594	472	500	567	130
pH		6.8–7.2	6.0–9.0	6.5–8.5	7.79	6.91	7.01	6.75	6.79
Chlorides	mg/L	500	500	200	128	50.0	159	11.5	107.0
Nitrites	mg/L	-	-	-	14.6	0.040	<0.002	<0.002	<0.002
Nitrates	mg/L	-	-	-	23.5	15.5	<0.02	<0.02	1.80
Hardness	mg/L CaCO ₃	650	650	400	120.0	<0.5	92	31.0	136.0
Alkalinity	mg/L CaCO ₃	350	350	400	35.7	98.7	144.9	42.0	86.1
BOD5	mg/L	-	25	-	<2	<2	<2	<2	5.0
COD	mg/L	75	75	-	<50	<50	<50	84	<50
TSS	mg/L	100	100	20	8	10	167	33	30
Turbidity	UNT	-	50	20	2.03	6.60	0.55	8.1	1.3
Ammonia Nitrog.	mg/L	-	1.0	-	2.25	2.87	13.7	1.24	<0.1
Total Phosphorus	mg/L	-	4.0	-	19.65	5.86	6.92	5.4	1.57
Silica	mg/L SiO ₂	50	50	150	-	31.0	17.5	34.5	11.3
Aluminum	mg/L	0.1	0.1	0.1	<0.148	<0.148	<0.148	<0.148	0.41
Iron	mg/L	0.5	0.5	5.0	0.021	0.58	0.116	1.06	0.288
Calcium	mg/L	50	50	-	-	0.01	19.5	1.10	35.3
Magnesium	mg/L	30	0.5	-	-	<0.02	60.9	25.1	84.6
Manganese	mg/L	0.5	0.5	-	0.059	0.497	0.299	1.55	0.110
Bicarbonates	mg/L	24	24	-	<0.5	<0.5	2.6	<0.5	<0.5
Sulfates	mg/L	200	200	300	22	19	15	32	7
Bacteria (Col/mL)	CFU/100mL	-	-	1 × 10 ³	2.3 × 10 ⁵	1.3 × 10 ³	<1	<20	<2

3. Results

3.1. Start of WWTP

The sewage production started with the use of toilets and general cleaning. The WWTP tanks were filled with water, and the incoming air valves were adjusted. The ventilation system for the area where the WWTP was installed was activated. As a result, the concentration of DO measured in the 12 aeration tanks ranged from 2.2 to 3.4 mg/L; 300 kg of dewatered sludge was added as inoculum, and the production was evaluated. When necessary, as previously described, a defoamer was added to control foam formation and sludge loss in the aeration tanks.

Due to the high quantity of oils and greases in sewage, a strong smell was detected in the WWTP area. This problem was solved by cleaning the retaining boxes, adjusting the DO to 3 mg/L, and increasing from 1.5 to 4 times the exchanges of air volume per hour in the WWTP installation area. After these adjustments, the following input parameters were measured: BOD (around or below 400 mg/L); COD (ranging from 1000 mg/L to 1600 mg/L); total phosphorus (around or below 40 mg/L); the input ammonia (NH₃) (ranging from 40 mg/L to 180 mg/L); and the conductivity (ranging from 1100 µS/cm to 1800 µS/cm).

Due to the high sludge production, even with a low concentration of BOD₅, the frequency of sludge discharges and the flow rate of sludge return were increased, keeping the quantity of TSS in the aeration tanks below 4000 mg/L. After controlling the amount of active sludge in the reactors, the denitrification (from nitrates [NO₃] to nitrogen gas [N₂]) was adjusted. For this, the closing time of the air valves (Figure 1–step II) was changed to 20 min of anoxia every 30 min with aeration. The WWTP operation was stabilized after 30 days, generating sludge with good sedimentation, cellular aggregates, and at least 90% of the organic load (BOD) reduced.

3.2. Jar Test Experiments Results

The adequate concentration of flocculants was determined using 72 jar test experiments with 20 liters of sludge for each batch of tests, collected from the 2nd aeration tank (Figure 1). For this, aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$, 1.25, 2.5, 5.0, and 12.5 mg/L), aluminum polychloride (PAC, 1.25, 2.5, and 10.0 mg/L), flocculant based on tannin (Tanfloc (27.5, 41.25, 55.0, 68.75, and 82.5 mg/L) and ferric chloride (FeCl_3 , 13.8, 41.4, 69.0, 138.0, 207.0, and 276.0 mg/L) were used as flocculants and zeolites (ZN 0410, SMF, and ZE 0325) and anthracite coal were used as filter elements. The efficiency of salt remotion was evaluated by conductivity analysis (Table 1).

The best removal of TDS was observed with 41.4 mg/L of ferric chloride (FeCl_3) followed by filtration in SFM zeolites. For this treatment, the observed conductivity ranged from 537 to 539 $\mu\text{S}/\text{cm}$ (Table 1), corresponding to a variation of TDS from 343 to 345 mg/L, total phosphorus from 1.00 to 1.50 mg/L, nitrate less than 5.0 mg/L, and no shift in pH value. These results guided the changes made in establishing the conditions for the set-up of the WWTP operation.

3.3. Set-Up of the WWTP Operation

Based on the results of the jar test experiments, the parameters used to start the WWTP were adjusted. The electromagnetic metering pump was regulated to inject 41.4 mg/L of ferric chloride (FeCl_3), and this was further increased to 69.0 mg/L during the highest sewage production, allowing maintenance of the TDS concentration between 130 and 594 mg/L (Table 2).

After the ultrafiltration equipment was started, the permeate effluent showed high TSS removal (8.0 mg/L) and $\text{BOD}_5 < 2.0$ mg/L (Table 2). The operation of the equipment requires that the pre-filters be washed once the pressure reaches 0.5 bar to protect the membranes from solid waste; however, a new problem appeared. The pre-filters reached maximum pressure quickly, requiring successive stops to clean the filter bags.

Adjustments in the WWTP and Implementation of the 10th Stage

To improve the production of treated water, the site where the flocculant was added was moved to a new tertiary decanter (10th stage) which retained and removed flocculate solids (Figure 2). This procedure reduced the solids content in the pre-filters and stabilized the production of treated water with quality to be used in toilets and air conditioning heat exchangers.

3.4. Monitoring of WWTP Operation

The monitoring of the WWTP operation and the production of reused water was carried out by checking the quality of the treated effluent. Discards and cleanings were added to maintain the production. We measured the sludge production, electrical conductivity, and total phosphorus three times per week on alternate days with samples collected at the outlet of the ultrafiltration skid and the outlet of the zeolite filter. At the end of each visit made by the WWTP operator, an operation report was issued and sent electronically to the customer and the responsible technician. Every three months, the inlet and outlet effluent of the WWTP was analyzed in accredited laboratories, and a self-monitoring report was issued to meet the legal requirements of the environmental control bodies.

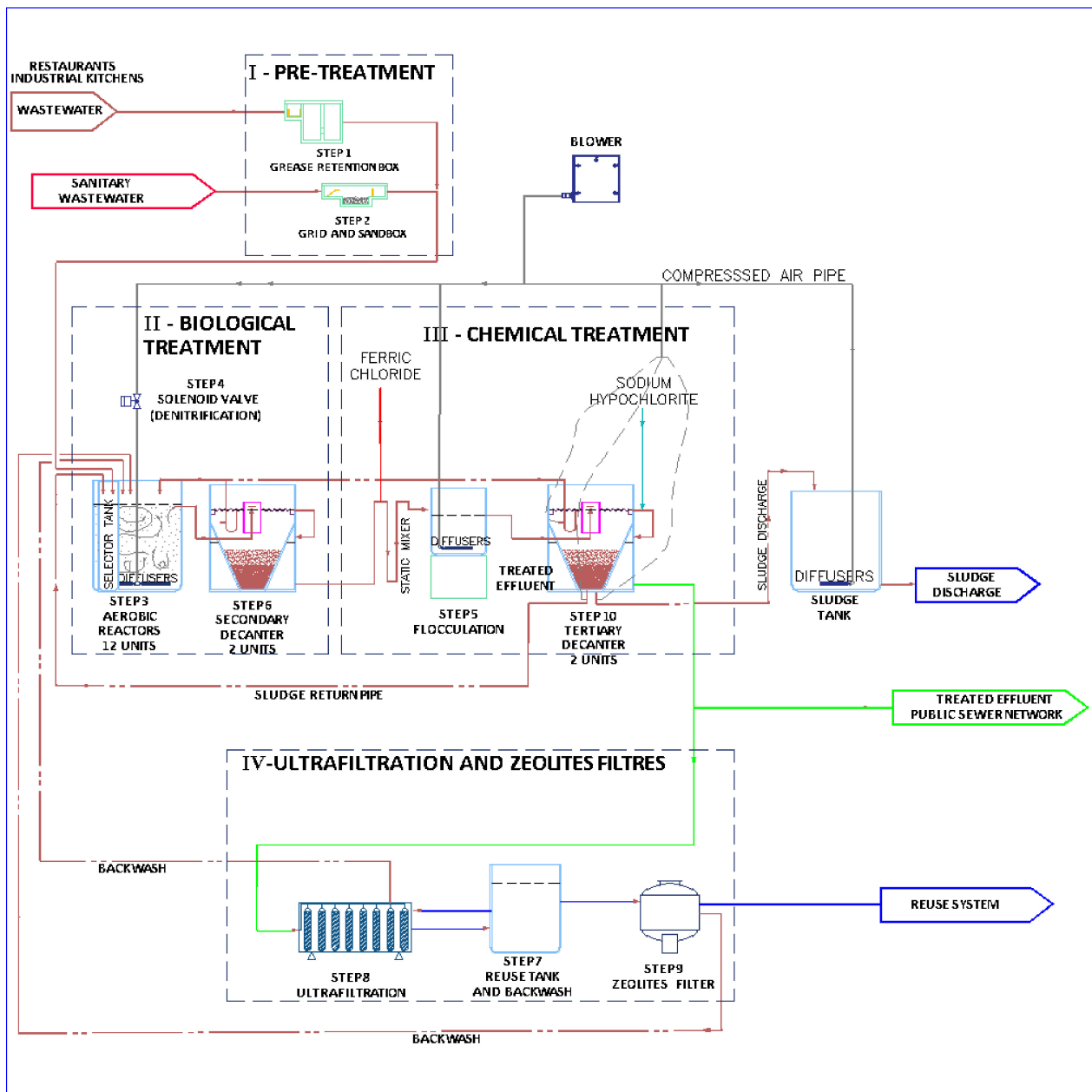


Figure 2. Schematic diagram of the wastewater treatment plant (WWTP) with 10 steps: 1-grid and sandbox; 2-oil and grease retention box; 3-aerobic reactors; 4-denitrification; 5-floculation; 6-decanting, 7-disinfection; 8-ultrafiltration; 9-filtration by zeolites and 10-tertiary decanter.

Based on our process, the water produced demonstrated the following characteristics: TDS ranged from 130 to 594 mg/L (21.2% of the maximum limit recommended by the manufacturer); the pH ranged from 6.75 to 7.79, near the levels found in the monitoring and within the established limit (Table 2); chlorides: maximum 159 mg/L, well below limits; nitrites were at 14.60 mg/L in the first analysis and then <0.002 mg/L after adjustment of the nitrification and denitrification; the same occurred with nitrates: at 23.50 mg/L and after adjustment reduced from <0.02 to 1.80 mg/L; the total hardness ranged from 31.0 to 136.0 mg/L of CaCO₃; the total alkalinity ranged from 35.7 to 144.9 mg/L of CaCO₃, both below the limits. The BOD₅ remained below the detection limit (2.00 mg/L) except for one measurement of 5.0 mg/L; COD was also below the detection limit (50 mg/L) except for one measurement of 84 mg/L (Table 2); TSS ranged from 8 to 33 mg/L, except for one measurement of 167 mg/L, above the limit (due to the excessive dosage of sodium

hypochlorite to control total coliforms); the turbidity ranged from 0.55 to 8.1 UNT, which represents only traces of turbidity; total phosphorus ranged from 19.65 to 1.57 mg/L (after adjustment of the dosage of ferric chloride), values above those recommended by the EPA and not recommended by the manufacturer; silica ranged from 11.3 to 34.5 mg/L SiO₂, also below the recommended limits; aluminum was 0.148 mg/L, except for one measurement of 0.41 mg/L, slightly above the recommended limits but without any damage to the equipment; iron ranged from 0.021 to 0.58 mg/L, below the recommended limits by the manufacturer; calcium from 0.01 to 35.3 mg/L; magnesium ranged from <0.02 to 84.6 mg/L, above the recommended limits, however, not specified by the manufacturer; manganese ranged from 0.059 to 1.55 mg/L, above the recommended limits, however, not specified by the manufacturer; bicarbonates ranged from <0.5 to 2.6 mg/L; sulfates ranged from 7 to 32 mg/L, both below recommendations; and finally, total absent coliforms ranged from 2.3×10^5 to 2 CFU/100 mL (Table 2). These findings suggest that, despite the retention of microorganisms in the ultrafiltration membranes, disinfection with sodium hypochlorite is essential.

In addition, during these 3 years, the quality of the produced water was confirmed by the absence of biocorrosion, biofouling and calcium scale in the cooling towers and reuse water plumbing.

4. Discussion

The type of project chosen for the WWTP was activated sludge, using a prolonged aeration regime, the same used in other studies [26,27,35]. In the present on-site wastewater treatment plant, the oxygen concentration in the aeration chamber was controlled and the sludge and outlet effluent quality was monitored. This control in the aeration chamber should be permanent and when necessary, the concentration should be adjusted between 1–2 mg/L to warrant the growth of microorganisms able to reduce the organic matter [36–38]. The analyses of the BOD₅ of outlet effluent of the WWTP indicated a biological sludge of good quality, stable, and with high efficiency in the removal of organic loads due also to the ultrafiltration equipment.

Experience in the operation of the WWTP has shown that it is possible to reuse effluents treated in sanitary basins, and when it is reused in air conditioning equipment, it is easier to compensate for the higher TDS content with an increased purge of the exchangers of heat than with the replacement of the filter elements and the costs with inputs and labor. The levels of TDS do not influence the sanitary flush system. The results presented demonstrated that it was possible to produce reuse water (up to 797 m³/month), which reduced drinking water consumption by up to 27%.

Monitoring of the WWTP by measuring conductivity was mediated by ultrafiltration membranes. The treated and filtered effluents in this type of equipment typically remove BOD₅, below the detection limit (<2.0 mg/L), and COD, also below the detection limit (<50 mg/L). As a result, it was decided to use the same type of control used by the pharmaceutical industry, especially for injectable liquids. The legislation of the state of São Paulo also requires, among other parameters, the monitoring of electrical conductivity [24].

Measurement of conductivity in the field proved to be as simple as measuring color and turbidity, as found in similar studies that used ultrafiltration [26,36] and in studies that included coagulation and adsorption [35,39]. However, when comparing TDS values, the ratio varied from 0.44 to 0.64, different from that described in the literature, where the TDS varied approximately 0.64 times the electrical conductivity [18]. Bearing in mind that the yellow coloration of the treated effluent may be responsible for staining toilets, this required an increase in the dosage of sodium hypochlorite and the replacement of the outlet filter elements.

Multiple metal ions (K⁺, Mg²⁺, Ca²⁺, Ni²⁺, Co²⁺, Cu²⁺, Pb²⁺, Cd²⁺, Cr²⁺, Fe³⁺, and Al³⁺) have been efficiently removed from the water, using a Ti–C redox interfaces [14]. In another study, methylene blue, acid orange 7, and bisphenol A were removed from contaminated water by the use of mechanochemical synthesized graphite oxides as adsor-

bents [15], suggesting that new strategies and adsorbents have been developed to reduce these pollutants in water for reuse.

Studies that used a flocculant such as calcium chloride (CaCl_2) at 50 mg/L [35,36] and ferric chloride (FeCl_3) at 20 mg/L, obtained an efficiency in load removal remarkably similar to that observed in the present study ($\text{BOD}_5 < 2.0$ mg/L; $\text{COD} < 50$ mg/L; turbidity up to 2.2 NTU). However, the most challenging and most important step in this experiment was not to determine the flocculant or filter element that best removes the TDS; rather, it was to produce a significant volume of reuse water (up to 42 m³/day) with the required quality for replacement in heat exchangers of air-conditioners, a finding that did not appear in previous studies [26,27,35,36]. As previously described, at the beginning of the WWTP operation, unexpected occurrences and results appeared, including excess fat discharges, lack of cleanliness of the retaining boxes, poor ventilation in the subsoil, excessive sludge production, and clogging of the pre-filters of the ultrafiltration equipment. These did not alter the quality and safety of the reuse of the treated effluent; however, they affected the flow rate of the reused water. This variation in the raw material effluent to the WWTP, (i.e., the sewage generated) is not uncommon [40].

In the present WWTP, the mall operator opted to interrupt the flocculant dosing and use the zeolites as filter elements and dosing a dye called Azunat, produced by NaturalTec (São Paulo, Brazil), at 2 liters for every 20,000 liters, which gave the reuse water a very light bluish tint. Different types of zeolites have been discovered and used as adsorbents in wastewater treatment from removing cations [13,41]. In addition to these different natural zeolites, studies have been conducted to produce synthetic zeolites with improved mechanical strength, which affect the capability to treat wastewater through ion exchange processes [12]. The authors suggest that future work should focus on the zeolites' shape, which affects the resistance to attrition and compression (reusability) and the cations removal. In the present study, we used two types of zeolites (ZN0410 and SFM) or charcoal in combination with different flocculant/coagulant concentrations. We observed that the combination of SFM and FeCl_3 (41.4 mg/L) resulted in reduced conductivity (Table 1) and produced reuse water within the parameters established by regulatory agencies. In addition, during these 3 years, biocorrosion, biofouling, and calcium scale were not observed in the water plumbing by the use of water produced in the 3-years-long large full-scale evaluation.

The appearance of coliform bacteria in the first analyses in 2.3×10^5 (NMP/100 mL) shows that, despite the ultrafiltration retaining these microorganisms, there is a need to measure and control the residual chlorine in the effluent for reuse. In the literature that used ultrafiltration equipment at the outlet of the WWTP [26,35], there is no description of this occurrence, probably due to the conditions of the WWTP and the operating time.

With the installation of the tertiary decanter, the presence of solids in the treated effluent decreased substantially, and there was a significant increase in the flow of treated water; however, this step did not eliminate the need to clean the pre-filters. Further investment should be made to use filters with automated cleaning. The choice of monitoring the WWTP using the electrical conductivity proved to be correct. In 2017, the state regulation company established that daily electrical conductivity analyses should be carried out on reused effluents [24,25]. The project is estimated to produce up to 50 m³/day of reuse water for toilets, air conditioning towers, washing, and irrigation from treated sewage; however, the actual production varied between 12 to 42 m³/day, allowing for use in toilets and urinals only.

The nine stages initially designed were still important, that is, the grid and sandbox for holding sand and solid waste, a sandbox for retaining and cleaning the oils and greases from food production in the kitchens of the several restaurants, aerobic reactors for degrading carbonaceous and nitrogenated organic matter (BOD_{5-20}). The denitrification system for converting nitrates (NO_3) into nitrogen gas (N_2). The dosage of a disinfectant (sodium hypochlorite) to eliminate pathogenic organisms and, finally, the ultrafiltration equipment and zeolite filters to remove colloidal particles and other microorganisms. However, the addition of the tertiary decanter, and the change of the dosing point after the secondary de-

canter and before the tertiary, allowed a better removal of colloidal particles in suspension, increasing the water reuse quality.

Finally, the most challenging and essential result of the research project was not to determine the flocculant or the filtering element that best removes the TDS, but rather to provide the WWTP with the necessary steps to adapt to the specific conditions of the incoming sewage and the daily production of effluent for reuse, in addition to developing a self-monitoring plan to guide this operation.

5. Conclusions

In order to implement a wastewater treatment capable of removing most of the organic and inorganic loads from sewage produced in a mall located in the city of Guarulhos, São Paulo State, Brazil, and produce reuse water on a commercial scale according to parameters established by several environmental control bodies, changes in the WWTP were conducted allowing us to reach the main objectives. The following conclusions were made:

- (1) The inclusion of a tertiary decanter improved the quality of the water for reuse.
- (2) From the laboratory scale experiments (jar tests), the zeolite (SFM) and FeCl_3 (41.4 mg/L) were chosen for the set-up of the WWTP operation, reducing the time for adjustments in this large full-scale production of quality water for cooling towers.
- (3) FeCl_3 as a flocculant followed by filtration by zeolites (SFM) resulted in maximum removal (about 99%) of the biological oxygen demand (BOD_5).
- (4) The changes in the WWTP operation reduced consistently the TDS including chlorides and ammonia, which are described as corrosion factors in cooling systems.
- (5) The concentration of residual organic substrate, N, and P, which are associated with biofouling of cooling systems, was reduced after using the FeCl_3 as a flocculant.
- (6) The reused water produced by the WWTP presented the reduced capability to form calcium scale (calcium carbonate and calcium phosphate).
- (7) The developed treatment demonstrated the feasibility of water reused in air conditioning cooling towers and toilets, saving up to 797 m³/month (27% of consumed water in the mall).

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References

1. WHO. *Drinking—Water*; World Health Organizations: Geneva, Switzerland, 2019; Available online: <https://www.who.int/news-room/fact-sheets/detail/drinking-water> (accessed on 18 March 2022).
2. UNICEF. Global Framework for Urban Water, Sanitation and Hygiene. Urban Wash. 2019. Available online: <https://www.unicef.org/documents/global-framework-urban-water-sanitation-and-hygiene#:~:text=The%20Framework%20is%20based%20on,for%20engagement%20in%20urban%20WASH> (accessed on 26 August 2021).
3. Capodaglio, A.G. Fit-for-purpose urban wastewater reuse: Analysis of issues and available technologies for sustainable multiple barrier approaches. *Crit. Rev. Environ. Sci. Technol.* **2021**, *51*, 1619–1666. [CrossRef]
4. El-Monaem, E.M.A.; El-Latif, M.M.A.; Eltaweil, A.S.; El-Subruiti, G.M. Cobalt nanoparticles supported on reduced amine-functionalized graphene oxide for catalytic reduction of nitroanilines and organic dyes. *Nano* **2021**, *16*, 2150039. [CrossRef]
5. Hosny, M.; Fawzy, M.; Abdelfatah, A.M.; Fawzy, E.E.; Eltaweil, A.S. Comparative study on the potentialities of two halophytic species in the green synthesis of gold nanoparticles and their anticancer, antioxidant and catalytic efficiencies. *Adv. Powder Technol.* **2021**, *32*, 3220–3233. [CrossRef]
6. Capodaglio, A.G.; Hlavínek, P.; Raboni, M. Advances in wastewater nitrogen removal by biological processes: State of the art review. *Rev. Ambiente Água* **2016**, *11*, 250–267. [CrossRef]
7. WHO. *The International Statistical Classification of Diseases and Health Related Problems ICD-10*, 10th Revision ed; Tabular List; World Health Organizations: Geneva, Switzerland, 2004; Volume 1.
8. Glibert, P.M. Harmful algae at the complex nexus of eutrophication and climate change. *Harmful Algae* **2020**, *91*, 101583. [CrossRef]
9. Eltaweil, A.S.; Omer, A.M.; El-Aqapa, H.G.; Gaber, N.M.; Attia, N.F.; El-Subruiti, G.M.; Mohy-Eldin, M.S.; Abd El-Monaem, E.M. Chitosan based adsorbents for the removal of phosphate and nitrate: A critical review. *Carbohydr. Polym.* **2021**, *274*, 118671. [CrossRef]
10. Liu, T.; Jia, G.; Quan, X. Accelerated start-up and microbial community structures of simultaneous nitrification and denitrification using novel suspended carriers. *J. Chem. Technol. Biotechnol.* **2018**, *93*, 577–584. [CrossRef]
11. Montalvo, S.; Huiliñir, C.; Borja, R.; Sánchez, E.; Herrmann, C. Application of zeolites for biological treatment processes of solid wastes and wastewaters—A review. *Bioresour. Technol.* **2020**, *301*, 122808. [CrossRef]
12. Guida, S.; Potter, C.; Jefferson, B.; Soares, A. Preparation and evaluation of zeolites for ammonium removal from municipal wastewater through ion exchange process. *Sci. Rep.* **2020**, *10*, 12426. [CrossRef] [PubMed]
13. Shi, J.; Yang, Z.; Dai, H.; Lu, X.; Peng, L.; Tan, X.; Shi, L.; Fahim, R. Preparation and application of modified zeolites as adsorbents in wastewater treatment. *Water Sci. Technol.* **2018**, *2017*, 621–635. [CrossRef]
14. Huang, L.; Yan, T.; Mahmoud, A.D.; Li, S.; Zhang, J.; Shia, L.; Zhang, D. Enhanced water purification via redox interfaces created by an atomic layer deposition strategy. *Environ. Sci. Nano* **2021**, *8*, 950–959. [CrossRef]
15. Mahmoud, A.D.; Franke, M.; Stelter, M.; Braeutigam, P. Mechanochemical versus chemical routes for graphitic precursors and their performance in micropollutants removal in water. *Powder Technol.* **2020**, *366*, 629–640. [CrossRef]
16. Hespanhol, I. Um novo paradigma para a gestão de recursos hídricos. *Estud. Avançados* **2008**, *22*, 131–158. [CrossRef]
17. Rupiper, A.M.; Loge, F.J. Identifying and overcoming barriers to onsite non-potable water reuse in California from local stakeholder perspectives. *Resour. Conserv. Recycl. X* **2019**, *4*, 100018. [CrossRef]
18. Metcalf and Eddy Inc.; Tchobanoglous, G.; Stensel, H.D.; Tsuchihashi, R.; Burton, F. *Wastewater Engineering: Treatment and Resource Recovery*; Mc Graw-Hill Education: New York, NY, USA, 2014.
19. Asano, T.; Burton, F.L.; Leverenz, H.L.; Tsuchihashi, R.; Tchobanoglous, G. *Water Reuse: Issues, Technologies, and Applications*; Mc Graw-Hill: New York, NY, USA, 2007.
20. Trovat, J. Tratamento de Água Para Sistema de Resfriamento, Cursos Online. 2004. Available online: https://www.snatural.com.br/PDF_arquivos/Torre-Caldeira-Tratamento-Agua.pdf (accessed on 19 May 2016).
21. Wallin, J.; Knutsson, J.; Karpouzoglou, T. A multi-criteria analysis of Building level graywater reuse for personal hygiene. *Resour. Conserv. Recycl.* **2021**, *12*, 200054. [CrossRef]
22. GBD 2017 Diet Collaborators. Health effects of dietary risks in 195 countries, 1990–2017: A systematic analysis for the Global Burden of Disease Study 2017. *Lancet* **2019**, *393*, 1958–1972. Available online: [https://www.thelancet.com/article/S0140-6736\(19\)30041-8/fulltext](https://www.thelancet.com/article/S0140-6736(19)30041-8/fulltext) (accessed on 20 February 2020). [CrossRef]
23. ANA—Agência Nacional de Águas. *Conservação e Reuso da Água em Edificações*; FIESP—Federação das Indústrias de São Paulo in association with Ministério do Meio Ambiente: São Paulo, Brazil, 2015.
24. SES/SIMA—Resolução Conjunta nº 01 SES/SIMA DOE de 14-02-2020, Seção I, 47–48; da Secretaria Estadual da Saúde e Secretaria de Infraestrutura e Meio Ambiente—Disciplina o Reuso Direto de Água Não Potável. Governo do Estado de São Paulo: São Paulo, Brazil, 2020. Available online: http://www.mpsp.mp.br/portal/page/portal/cao_urbanismo_e_meio_ambiente/legislacao/leg_estadual/leg_est_resolucoes/Resol-cjta-SES-SIMA-01-2020_Processo-ssrh-90-2016_reuso-de-agua-nao-potavel_fins_urbano_ETE.pdf (accessed on 26 August 2021).
25. ABNT NBR 16783; Uso de Fontes Alternativas de Água não Potável em Edificações. Associação Brasileira de Normas Técnicas: Rio de Janeiro, Brazil, 2019.
26. Arévalo, J.; Ruiz, L.M.; Parada-Albarracín, J.A.; Gonzáles-Pérez, D.M.; Pérez, J.; Moreno, B.; Gómez, M.A. Wastewater reuse after treatment by MBR. Microfiltration or ultrafiltration? *Desalination* **2012**, *299*, 22–27. [CrossRef]

27. Subtil, E.L.; Hespanhol, I.; Mierzwa, J.C. Biorreatores com Membranas Submersas (BRMs): Alternativa promissora para o tratamento de esgotos sanitários para reúso. *Ambiente Água* **2013**, *8*, 129–142. [[CrossRef](#)]
28. Von Sperling, M. *Princípios do Tratamento Biológico de Águas Residuárias. Lodos Ativados*; UFMG: Belo Horizonte, Brazil, 2002.
29. ABNT NBR 12209; Elaboração de Projetos Hidráulicos-Sanitários de Estações de Tratamento de Esgotos Sanitários. Associação Brasileira de Normas Técnicas: Rio de Janeiro, Brazil, 2011.
30. APHA. *Standard Methods for the Examination of Water and Wastewater*, 23rd ed.; American Public Health Association: Washington, DC, USA, 2017; 1504p.
31. Madigan, M.T.; Martinko, J.M.; Bender, K.S.; Buckley, D.H.; Stahl, D.A. *Microbiologia de Brock*; Artimed: Porto Alegre, Brazil, 2016.
32. EPA. *Guidelines for Water Reuse*; Environmental Protection Agency: Washington, DC, USA, 2012. Available online: <https://www.epa.gov/sites/production/files/2019-08/documents/2012-guidelines-water-reuse.pdf> (accessed on 2 August 2016).
33. CONAMA-Resolução 430; Dispõe Sobre Condições e Padrões de Lançamento de Efluentes. Conselho Nacional do Meio Ambiente: Brasília, 2011.
34. CONAMA-Resolução 357; Dispõe Sobre a Classificação dos Corpos de Água e Diretrizes Ambientais Para o Seu Enquadramento, Bem Como Estabelece as Condições e Padrões de Lançamento de Efluentes, e dá Outras Providências. Conselho Nacional do Meio Ambiente: Brasília, 2005.
35. Abdessemed, D.; Nezzal, G.; Ben Aim, R. Coagulation-adsorption-ultrafiltration for wastewater treatment and Reuse. *Desalination* **2000**, *131*, 307–314. [[CrossRef](#)]
36. Abdessemed, D.; Nezzal, G. Tertiary treatment of a secondary effluent by the coupling of coagulation-adsorption-ultrafiltration for reuse. *Desalination* **2005**, *175*, 135–141. [[CrossRef](#)]
37. Dymaczewski, Z. (Ed.) *A Sewage Treatment Plant Operator's Guide*; PZITS Poznań: Poznań, Poland, 2011; ISBN 978-83-89696-38-X.
38. Karczmarczyk, A.; Kowalik, W. Combination of Microscopic Tests of the Activated Sludge and Effluent Quality for More Efficient On-Site Treatment. *Water* **2022**, *14*, 489. [[CrossRef](#)]
39. Vesilind, P.A.; Morgan, S.M.; Heine, L.G. *Introdução à Engenharia Ambiental*; Cengage: São Paulo, Brazil, 2018.
40. SABESP-NTS 181; Dimensionamento do Ramal Predial de Água, Cavalete e Hidrômetro—Primeira Ligação. Companhia de Saneamento Básico do Estado de São Paulo: São Paulo, Brazil, 2017. Available online: <http://www2.sabesp.com.br/normas/nts/NTS181.pdf> (accessed on 14 April 2018).
41. Ghasemi, Z.; Sourinejad, I.; Kazemian, H.; Rohani, S. Application of zeolites in aquaculture industry: A review. *Rev. Aquacult.* **2018**, *10*, 75–95. [[CrossRef](#)]