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REUSE OF ALUM SLUDGE FOR REDUCING FLOCCULANT ADDITION IN WATER TREATMENT PLANTS

The recycling of water treatment residues (WTR) inside drinking water treatment plants (DWTP) seems to be a good option for reusing this type of waste, as well as for reducing the costs with its disposal off and with the acquisition of treatment chemicals. Therefore, a WTR was reused for auxiliary of the coagulation-flocculation processes for reducing the use of aluminium sulfate (coagulant) in a DWTP. Three series of experiments have been conducted involving three water samples with different turbidity and colour, different WTR samples with different total suspended solids (TSS) concentrations and different aluminium sulfate concentrations. The results showed that WTR can efficiently be used for the removal of turbidity between 21 NTU and 218 NTU and colour between 194 HU and 1509 HU for TSS concentration between 1635 mg/dm³ and 5420 mg/dm³, with better results in the range between 1635 mg/dm³ and 2678 mg/dm³. For higher TSS concentrations, the removal of both parameters decrease because there are excess of organics released to water, which demands the use of more coagulant.

1. INTRODUCTION

The treatment of raw water for human consumption or industry requires the use of different technologies in order to produce drinking water according to international standards. During water treatment, several reagents (e.g., coagulants, flocculants, softeners, pH buffers, disinfectants and polyelectrolytes) are used for removing suspended solids, dissolved solids, colloids (mostly organic matter) and microorganisms (e.g., plankton and pathogens). When coagulants such as aluminium salts ($Al_2(SO_4)_3 \cdot nH_2O$) and ferric salts ($FeCl_3$ or $Fe_2(SO_4)_3$) are added to raw water there occurs complexation

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between hydrolytic (aluminium and ferric) species (namely in the form of hydroxides) with colloidal particles of the water (e.g., humic and fulvic acids), microorganisms (e.g., bacteria, protozoa and algae), and inorganic substances (e.g., fine soil particles). Additionally, pH buffers, softening reagents (e.g., CaO and Ca(OH)₂ and Na₂CO₃), long-chain organic polymers and activated carbon can also be added for pH adjustment, water softening, increasing the efficiency of the flocculation and filtration processes and removing emergent pollutants, respectively [1].

The addition of such reagents to water treatment generate subproducts at clarifiers (settled products), filters (backwash streams), ion exchange devices (concentrate stream) and membranes (retentate stream), which are called water treatment sludge (WTS) or water treatment residuals (WTR). WTR are primarily amorphous materials, which normally contain Si, Ca, Al, P, Mg, K, Fe and Ti as the most abundant elements and pH between 6 and 8 [2, 3], being Si and Al or Fe the dominant elements depending on the coagulant used [1]. The solids content of the raw WTR ranges 1–2 wt. % and of dewatered sludge ranges 15–82 wt. % [4, 5]. According to Reali [6], WTR are mainly produced at clarifiers and filters, representing 0.1–1.5 wt. % of the total volume of water to be treated.

The raw water characteristics have high influence on the WTR characteristics and may impact disposal alternatives, namely if presents excessively high or low pH, high concentration of total suspended solids (TSS), total dissolved solids (TDS), organic matter (as chemical oxygen demand (COD) or total organic carbon (TOC)), heavy metals (e.g., As, Cd, Cr, Pb and Zn), competing ions (e.g., fluoride, sodium, sulfate and chloride) and emergent pollutants (e.g., endocrine-disrupting compounds, pharmaceutically active compounds, trihalomethanes and phenols) and pathogens [5, 7, 8]. The optimum pH for alum coagulation is from 6 to 8. The effectiveness of coagulation-flocculation processes is normally measured by parameters such as residual turbidity, residual colour or organics removal because they are directly related to the solid–liquid separation and water quality [3].

The operational procedures at the drinking water treatment plants (DWTP) also have impact on WTR characteristics, namely the medium velocity gradient (G_m and G_f for the coagulation and flocculation phases, respectively), mixing time (T_m), flocculation mixing time (T_f) and sedimentation time (T_s). The typical operational values are: 10–1000 s⁻¹ (G_m and G_f), 5 to 60 min (T_m and T_f) and 5 to 120 min (T_s) [4, 6, 9]. jar test is a useful tool for simulating the coagulation, flocculation and sedimentation processes that enhance the removal of suspended colloids and organic matter, which lead to turbidity, odour and taste problems.

The destabilization of colloids is achieved through the addition of coagulants, leading to the formation of aquo-metal complexes, which then pass through a series of hydrolytic reactions until getting mononuclear species [3]. Those compounds are highly positively charged and react with negatively charged clay or silica particles in water, coagulating them by a purely electrostatic action. Therefore, the removal of compounds

responsible for turbidity and colour consumes high quantities of chemicals for coagulation-flocculation and the managing of WTR are difficult and costly tasks for water management authorities. A fraction of the chemicals used for coagulation-flocculation processes reaches the sludge without having reacted with the water impurities [5, 9].

For example, according to Henderson et al. [10], more than 326 000 tons per year of coagulants are used for water treatment in the UK, which has generated proportionally large amounts of WTR, around 182 000 tons of per year as dried solids [11]. For another hand, the classical final disposal options of WTR have been incineration and disposal of on landfill sites (for the dewatered sludge), and discharge into sewers systems, soils and surface waters (for the non-dewatered sludge) [3]. The last two options can have negative environmental impacts on soil and water and bringing risks for public health.

The reuse of WTR has been suggested in several guides and strategic reports of international institution such as the World Bank, United Nations and European Union [2, 12]. The European Directive 2008/98/EC [12] and the Brazilian National Policy on Solid Waste [13] pointed out for the need on reusing WTR instead of its incineration or disposal into the environment. There are thousands of drinking water treatment plants (DWTP) worldwide, which use various coagulants and produce tons of WTR per year [9], thus causing concern due to their disposal and associated costs. McCormick et al. [4] setup a study with 46 DWTP in US, most using aluminium sulfate as primary coagulant, and report that 36% of the facilities recycled WTR streams to the upstream, suggesting that the recycling backwash waters can results in cost savings associated to dispose of WTR and reduced cost for purchasing treatment chemicals.

Several studies have already presented results of WTR reused for polishing wastewater treatment in order to remove residual loads of organics, nitrogen, phosphorous or heavy metals [8,14], control of phosphorus in eutrophic soils [15], manufacturing of bricks and concrete [16], coagulant recovery using extraction methods or membranes [5], and agricultural applications [17].

The USEPA's Filter Backwash Recycling Rule [2] allows the recycling of WTR from filter backwash, thickener supernatant, and dewatering process liquids to a point in the source water treatment process where it will be treated by coagulation and filtration. The recirculation of WTR from filters and clarifiers to upstream coagulation tanks can lead to the decrease of reagents addition at the entrance, since there still exists a large fraction of reagents that can be used as additional coagulant to enhance pollutants removal, as observed by Guan et al. [18] in experiments with the reuse of alum sludge.

There are no many studies on the vantages of recycling backwash water for coagulant reduction, and most of the studies developed at DWTP were centred in the recovery of coagulants. Moghaddam et al. [19] tested the recirculation of ferric sludge to coagulation tank and noted an increase in the removal of a synthetic dye and a decrease in the consumption of ferric chloride as coagulant. Zhou et al. [7] have reused polyaluminium

chloride mixed sludge for evaluating the removal of initial turbidity and for analysing the flocs characteristics. According to Gottfried et al. [20], the removal of contaminants in previously treated water is more efficient than in raw water that have not been previously subjected to coagulation process. Cornwell et al. [21] have shown that the removal of *Cryptosporidium* in sedimentation tanks was improved when a filter backwash stream was recycled for flow rates from 4.3% to 20%, but turbidity levels were increased.

However, residuals recirculation in water treatment plants can also bring risks of water contamination with undesirable compounds that were previously removed (e.g., heavy metals and recalcitrant organics) [5] or were developed in the clarifiers or filters (e.g., pathogens microorganism and algae). Yang et al. [22] studied the consequences of phosphorous adsorption on WTR and concluded that the release of organics into the water phase is not significant for the pH range 6–7.2. Nevertheless, in plants using powdered activated carbon, the residuals can be easily removed by extending the contact time for more than 20 min [7]. Therefore, the suitable sludge recirculation rate should be adjusted for each plant periodically in order to allow the reuse of active reagents without worsening the water quality.

Therefore, the recycling of WTR seems to be an alternative way for reusing this type of waste, as well as for reducing the costs with their disposal and with acquisition of treatment chemicals. The objective of this research was to study the impact of WTR recycling on the water quality in the coagulation-flocculation-sedimentation processes, using bench-scale jar test experiments for different water characteristics, WTR concentrations and coagulant concentrations. WTR were collected at the discharge point from the clarifier tank.

2. MATERIALS AND METHODS

Characterization of the residue and water samples. The WTR samples (Fig. 1a) were obtained from the clarifier of the Meia Ponte DWTP (Goiânia, Brazil) and were used in bench-scale coagulation–sedimentation experiments (Fig. 1b). The DWTP had a production capacity of 2000 dm³/s, using aluminium sulfate (Al₂(SO₄)₃·14H₂O) as the primary coagulant. WTR samples were sieved in a 5.4×5.6 mm mesh to become homogenized and their solids contents were determined as total solids (TS) and total suspended solids (TSS) for using in the experiments of point 2.2 according to standard methods [23].

An elemental analysis of the WTR was conducted though energy-dispersive X-ray spectroscopy (EDX) in a Ronthec (Germany) equipment. The particle size was measured through laser diffraction particle size analyser in a Beckman Coulter LS200 (USA) equipment.

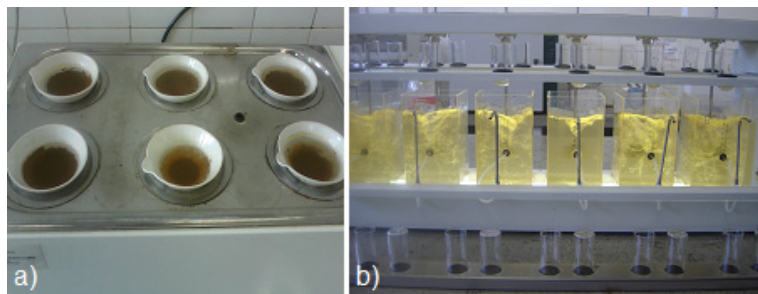


Fig. 1. Jar test experiments: a) WTR samples, b) coagulation phase

Approximately 50 dm³ of a raw water sample was collected at the entrance of the DWTP, in 3 different seasons, in order to get different turbidity and colour values according to indications of the DWTP's operators. These samples were used in 3 sets of bench-scale coagulation-sedimentation experiments. The following parameters were measured for all water samples in the experiments: pH, apparent colour, turbidity, organic matter (COD), alkalinity, TS and TSS, using standard methods setup in [23]. Turbidity was measured with a TB 1000 turbidimeter of Tecnoyon (Brazil) and for apparent colour determination it was used a DR 2800 Hach spectrophotometer (Germany). The pH was measured with a Policontrol mPA-210p meter (Brazil).

Bench-scale experiments Coagulation, flocculation and sedimentation experiments were carried out in a Nova Etica 218/LDB 06 (Brazil) standard jar test unit consisting of 6 square flasks of 2 dm³ capacity. A set of preliminary experiments were conducted for evaluating the efficiency of colour and turbidity removal for a standard coagulant concentrations for water initial turbidity below 554 NTU (i.e., 31 mg/dm³ as Al₂(SO₄)₃) (i.e., the average value used at the DWTP). Then, the following different concentrations of coagulant have been used for evaluating the best dose for turbidity and colour removal: 15.5, 18.6, 21.7, 24.8, 27.9 and 31.0 mg/dm³ as Al₂(SO₄)₃ (i.e., there were tested coagulant additions with reductions of 50%, 40%, 30%, 20%, 10% and 0% in relation to the standard concentration of 31 mg/dm³).

The volume of WTR used in the experiments was setup in 40 cm³, which is a recycling ratio of approximately 2% of the treated water volume, as suggested by Reali [6]. Zhou et al. [7] refer that the recycling ratio is responsible for the safety of reusing WTR, and the larger the recycling ratio the higher risk of water quality deterioration. To evaluate coagulation performance of the reuse process, the recycling ratio of WTR should be controlled at below 12%. Similar preliminary studies were setup by Gottfried et al. [20] for simulating the suitable residuals recycle rate to the head of DWTP.

Finally, three sets of experiments (called series of experiments) were developed, involving three raw water characteristics, different WTR concentrations (as TSS) and different coagulant concentrations of aluminium sulfate as shown in Table 1. Coagulant

concentrations were defined as reduction in coagulant addition in relation to the target concentrations of 16, 28 and 35 mg/dm³. The target coagulant concentrations were defined from the values used at the DWTP for the different raw water quality. The coagulant concentrations and the percentage reductions used in the preliminary tests and in the three series of experiments are shown in Table 1.

Table 1

Experimental conditions for the Jar-test experiments

Series	Characteristics of the raw water	Coagulant target value [mg/dm ³ as Al ₂ (SO ₄) ₃]	Coagulant dose [mg/dm ³ as Al ₂ (SO ₄) ₃]	Reduction in coagulant addition [%]	WTR addition as TSS [mg/dm ³]	Number of experiments
0 ^a	pH 7.8 turbidity 554 NTU colour 1401 HU alkalinity 38 mg CaCO ₃ /dm ³ COD 10.6 mg O ₂ /dm ³	31	15.5 18.6 21.7 24.8 27.9 31.0	50 40 30 20 10 0	0	1
1	pH 7.7±0.1 turbidity 21±2.1 NTU colour 194±39 HU alkalinity 50 mg CaCO ₃ /dm ³ COD 2 mg O ₂ /dm ³ TS 109 mg/dm ³ TSS 10 mg/dm ³ .	16	7.2 8.0 8.8 9.6 12.0 14.4 16.0	55 50 45 40 25 10 0	1635 3443 4306	96
2	pH 7.3±0.3 turbidity 95±0.1 NTU colour 1071±141 HU alkalinity 43 mg CaCO ₃ /dm ³ COD 6.5 mg O ₂ /dm ³ TS 231±44 mg/dm ³ TSS 121±10 mg/dm ³ .	28	12.6 14.0 15.4 16.8 21.0 29.5 28.0	55 50 45 40 25 10 0	2615 3642 4664 7500 8573 9670	168
3	pH 7.2±0.3 turbidity 218±27 NTU colour 1509±186 HU alkalinity 42 mg CaCO ₃ /dm ³ COD 7.7 mg O ₂ /dm ³ TS 422±46 mg/dm ³ TSS 140±23 mg/dm ³ .	35	15.8 17.5 19.3 21.0 26.3 31.5 35.0	55 50 45 40 25 10 0	2678 5420 8556 12670 14300	134

^a0 – preliminary.

Polyelectrolytes and alkalising agents were not added, because they increase the efficiency of the coagulation-flocculation processes and the specific resistance of the sludge also increases [24] which could influence the aims of the research.

The laboratory temperature was kept at 25 ± 1 °C for not influencing the results. The jar test operational parameters (G_m , G_f , T_m , T_f and T_s) were adjusted according to the characteristics of the raw water in each series of experiments, namely the values of colour and turbidity. Therefore, the operational parameters for the jar test experiments were adjusted taking in account the suggested values given in [3, 5] and also the raw water characteristics use in the three series of experiments. Operational parameters for the experiments: i) coagulation phase (rapid mixing): $G_m(1, 2, 3) = 197 \text{ s}^{-1}$, $T_m(1) = 60 \text{ s}$, $T_m(2, 3) = 20 \text{ s}$; ii) flocculation phase: $G_f(1) = 49 \text{ s}^{-1}$, $T_f(1) = 12 \text{ min}$, $G_f(2) = 30 \text{ s}^{-1}$, $T_f(2) = 6 \text{ min}$, $G_f(3) = 20 \text{ s}^{-1}$, $T_f(3) = 6 \text{ min}$; iii) sedimentation phase: $T_s(1, 2, 3) = 5 \text{ min}$ (in brackets the number of the series).

Each experiment was made in triplicate for getting average representative results. At the beginning and end of each experiment, water samples were collected for the determination of pH, apparent colour, turbidity, COD, alkalinity, TS and TSS, using standard methods [23].

3. RESULTS AND DISCUSSION

3.1. CHARACTERISTICS OF THE RESIDUE AND WATER SAMPLES

The EDX analysis on the WTR gave the following results for the elemental composition: O 67.5 wt. %, Al 13.2 wt. %, Si 10.1 wt. %, Fe 8 wt. %, K 0.4 wt. %, Ca 0.4 wt. % and Ti 0.4 wt. %. The sludge is mainly inorganic, rich in Al as result of the coagulant ($\text{Al}_2(\text{SO}_4)_3$) used for colour and turbidity removal, and also in O, Si and Fe, which are common elements in soil particles present in surface waters. The granulometric spectrum showed a particle size ranging from 2 μm to 1040 μm in diameter, with 1 wt. % corresponding to clay minerals, 25.5 wt. % to silt particles and 73.5 wt. % to sand, all constituents of soil.

The characteristics of the raw water reflect the sampling at different periods of the year. pH is similar for all samples, but significant differences were noted for the other parameters, in particular for turbidity and colour, which will be analysed in the section 3.2.

3.2. BEST WTR DOSE FOR EFFICIENT TURBIDITY AND COLOUR REMOVAL

The variations of the residual turbidity and the residual colour after the jar test experiments shown in Table 1 are presented in Figs. 2 and 3, respectively. The results of the preliminary tests showed removal efficiencies (RE) between 97.2% and 98.6% for turbidity (Fig. 2a) and between 91.9% and 95.0% for colour (Fig. 3a). Similar preliminary studies were setup by Gottfried et al. [20] for simulating the suitable residuals recycle rate to the head of DWTP.

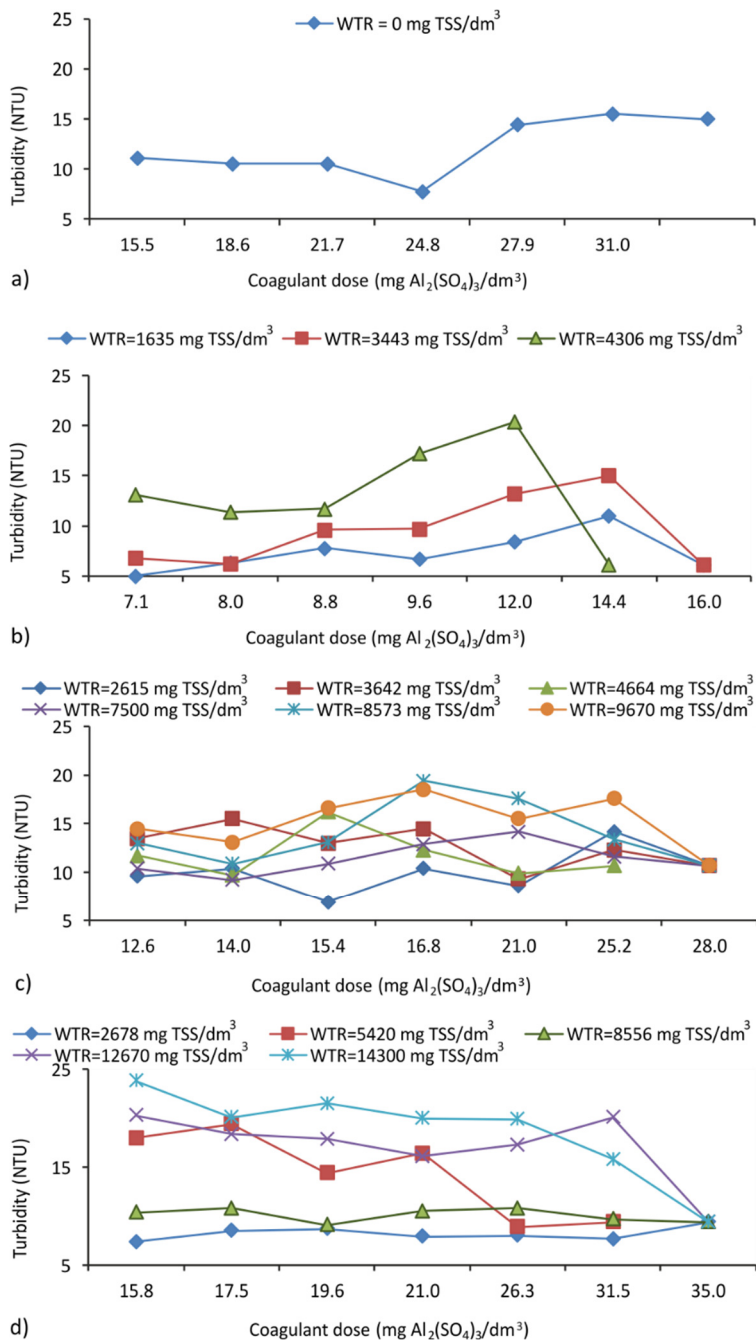


Fig. 2. Residual turbidity in relation to the coagulant concentration for the different experiments: a) preliminary series, b) series 1, c) series 2, d) series 3

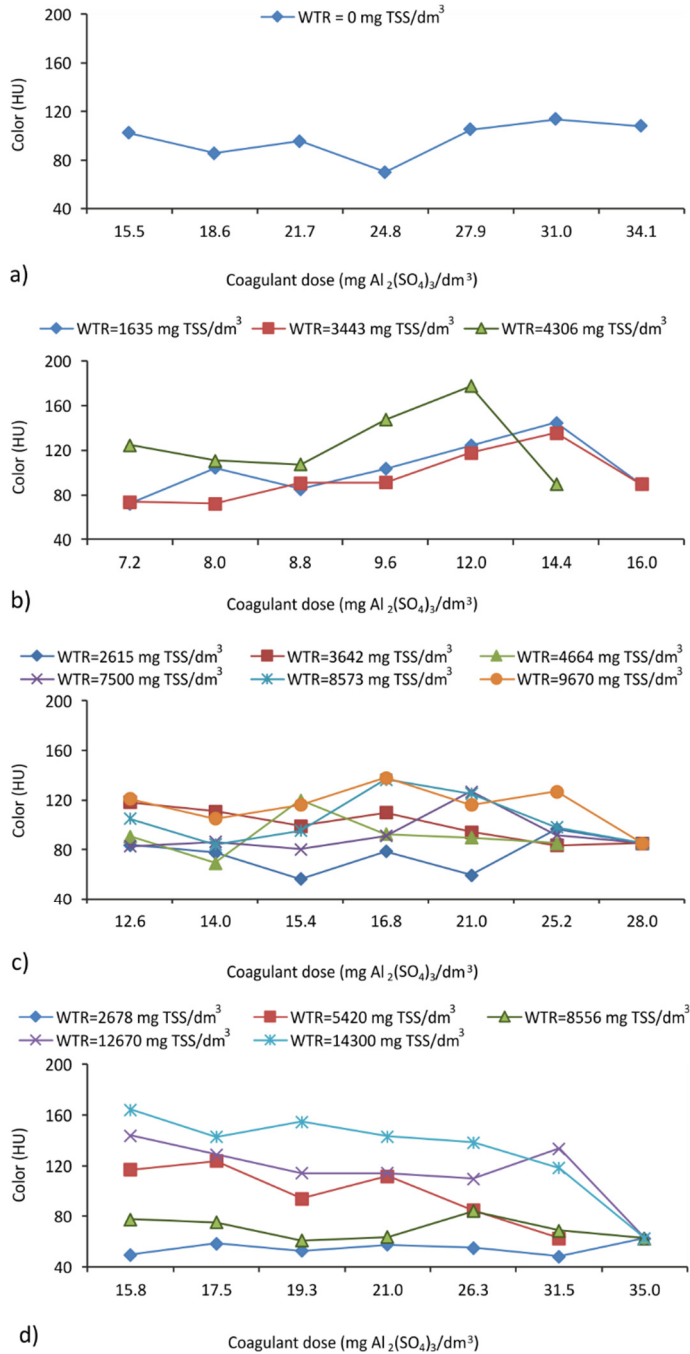


Fig. 3. Residual colour in relation to the coagulant concentration for the different experiments: a) preliminary series, b) series 1, c) series 2, d) series 3

For the low initial turbidity value (Series 1, 21 NTU) the best turbidity RE (78.9%) was observed for the WTR with low TSS concentration (1635 mg/dm^3) and 7.2 mg/dm^3 of $\text{Al}_2(\text{SO}_4)_3$ (i.e., for a coagulant reduction of 45%). The colour RE is also high for these conditions reaching 67.1%. The values at the final effluent were 72.3 HU and 5 NTU. The turbidity results are lower than the maximum desirable limit for achieving good filtration (10 NTU according to USEPA [2]).

The turbidity removal and colour removal did not improve for either high WTR doses or higher coagulant doses. For the WTR with the increased TSS concentration of 4306 mg/dm^3 (i.e., 2.6 times higher than the low tested TSS concentration) of WTR the RE of both parameters did not show any improvement (up to 27.9% for colour and up to 37.1% for turbidity) than the RE obtained with the control experiment (with no WTR and 100% of the initial coagulant dosage). The effluent values of colour and turbidity were between 107.1 HU and 177.5 HU and between 11.4 NTU and 20.4 NTU.

Zhou et al. [7] investigated the reuse of polyaluminium chloride mixed sludge on the turbidity removal in the coagulation-flocculation processes and have defined an appropriate dosage of $60 \text{ cm}^3/\text{dm}^3$ (i.e., a recycling ratio of 12%) as effective for the removal of approximately 89% of initial turbidity for initial values below 45 NTU.

For the raw water turbidity of 95 NTU (Series 2, Fig. 2b), the best results were achieved when residue of the low concentration of TSS (2615 mg/dm^3) was added and for an initial coagulant dose of 12.6 mg/dm^3 (i.e., with 55% of reduction in the coagulant dose). For these conditions, the RE was 94.6% for colour and 92.9% for turbidity. The values of the final effluent were 56.2 HU and 6.9 NTU, which are according to the maximum desirable limit for achieving good filtration defined by [2].

For WTR with increased TSS concentrations (between 1.4 and 3.7 times the tested TSS concentration), the RE did not increase, independently of the initial coagulant concentration. For WTR with concentrations higher than 7500 mg TSS/dm^3 (i.e., more than 3 times higher the tested TSS concentration), the RE for both parameters was even worse than the values obtained for the control experiment (with no WTR and 100% of the initial coagulant dosage). In this case, the effluent values of colour and turbidity were between 80 HU and 138 HU and between 11 NTU and 19.4 NTU, respectively.

Similar results were observed for series 3 (218 NTU). The best RE for both parameters (96.9% for colour, and 96.7% for turbidity) was obtained for the WTR with low TSS concentration (2678 mg/dm^3) and coagulant dose of 19.3 mg/dm^3 (i.e., with 45% of reduction in the coagulant dose). The values at the final effluent were 49.8 HU and 7.4 NTU, which are good values for achieving afterwards filtration. For either WTR with high TSS concentrations or coagulant doses, the RE was not improved and for TSS concentrations higher than 8556 mg/dm^3 (i.e., more than 3 times higher the tested TSS concentration) the RE were even better for the experiments with no addition of residue.

This trend in worsening the RE of both parameters as the dose of WTR increases might be related to the increase of organic matter released from the alum-sludge as its dose increases. The trapped organics in the residues are released at high WTR doses

leading to removal efficiency decline. Some studies have noted the increasing in solid levels after recycling WTR, which have led to the increase in influent turbidity [25]. This might mean that, for certain doses of residue, the quantity of organics released into the water was not efficiently removed because the aluminium sulfate concentration was not sufficient to destabilize the surface of colloids. Therefore, for these doses of WTR, it would have been necessary adding more coagulant.

pH of the water samples after jar test experiments slightly decreased ranging between 7.2 and 7.6 in the three series, which means this parameter did not change too much when WTR were added. The hydrolysates distribution of aluminium hydroxide in the WTR can be influenced by low pH values (<6), as observed by Zhou et al. [7], resulting in low RE of organics and turbidity. The pH interferes with the balance between the reactions of organic functional groups with hydrogen ions and Al hydrolysis products. At low pH, hydrogen ions out-competed with metal hydrolysis products for organic ligands and the RE of organics and turbidity can decrease [26].

As referred by Suman et al. [27], when destabilized particles of WTR are mixed with raw water, it increases the number of both collision sites and charge neutralization sites, resulting in the increasing of floc aggregation in the flocculation process, and decreasing of settling rate. The increasing in turbidity removal has occurred for recycling rates of WTR up to 10% and for raw water turbidity between 25 NTU and 200 NTU. Therefore, the addition of WTR can result in increasing of turbidity and colour removal, because the residual aluminium sulfate in the residue can be used for organics precipitation. For the increased doses of residue, the organics concentration increases and may be necessary adding more coagulant.

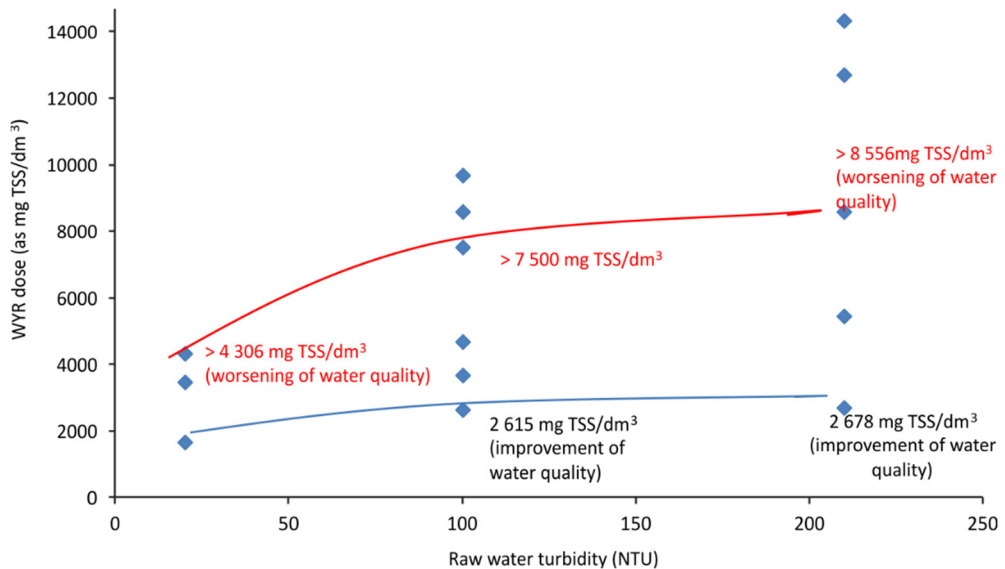


Fig. 4. Relationship between recycled WTR (as TSS) and raw water turbidity

Wu and Huang [28] have also used jar test experiments for applying WTR for the removal of low turbidity in the coagulation-flocculation process and observed an improvement in the settling characteristics of the sludge. They also observed a more compact flocs structure than the obtained with the coagulant alone, suggesting that the addition of residues lead to a better settling processes.

The results obtained in each experiments allowed a graphic relationship between the raw water turbidity and the ideal dose of WTR to be applied (Fig. 4). The blue line reflects the ideal dose of WTR to be added for an efficient removing of the turbidity. The red line shows the maximum WTR dose that can be applied until the water quality starts to decrease. It can also be observed that as the raw water turbidity increases from approximately 20 to 100 NTU, it is needed to increase the dosage of residue in approximately 60%, whilst for raw turbidity values between approximately 100 and 200 NTU the required increase in WTR is only 2.5%.

The results of this research suggest that WTR can be recycled for increasing turbidity and colour removal, but only up to 1635 mg TSS/dm³ (for raw water turbidity of 21 NTU), and up 2678 mg TSS/dm³ (for raw water turbidity between 95 NTU and 218 NTU). This practice, would decrease the needed of raw coagulant dosages.

4. CONCLUSIONS

The results of this research allow to conclude that water treatment residues (WTR) can be satisfactory applied for the removal of turbidity and colour in DWTP for raw water with turbidity between 21 and 218 NTU. The applications of residue doses between 1635 and 2678 mg TSS/dm³ has allowed a turbidity reduction from 78.9% to 96.7% and colour reduction from 67.1% to 96.9%, and an additional reduction of coagulant consumption (aluminium sulfate) between 45% and 55%. Therefore, recycling WTR to the coagulation-flocculation processes of DWTP can allow reduction of operational costs with reagents, besides allowing the reuse of residues produced in the same plant.

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