Tertiary phosphorus removal to extremely low levels by coagulation-flocculation and cloth-filtration

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ABSTRACT

Higher standards in the European Water Framework Directive and national directive demand advanced wastewater treatment for removal of nutrients and organic micropollutants before the discharge into water bodies. Systematic investigations regarding relative dosage and filtration processes for removal of flocculated solids are currently lacking. In this study, the performance of technologies for advanced removal of total phosphorus down to $<100\,\mu g/L$ with pile cloth-filtration (CF) and membrane filtration was verified and synergy effects for the removal of other contaminants were identified. The results show that an over-stoichiometric addition of coagulants of >5 mol Me^{3+}/mol sRP was necessary to achieve soluble reactive phosphorus (sRP) concentrations of $<50\,\mu g/L$ in the effluent. After the coupled process of tertiary phosphorus removal and solids removal, the soluble non-reactive phosphorus (sNRP) concentration regulates the lowest total phosphorus effluent concentration. sNRP is also partially, but not completely, removed by the use of coagulants. CF has proven to be an alternative technology for the removal of phosphorus and total suspended solids below the detection limit.

Key words | cloth-filtration, coagulation-flocculation, micro-/ultrafiltration, phosphorus, secondary effluent, tertiary phosphorus removal

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HIGHLIGHTS

- Cloth-filtration with different pile fabric, microfiltration and ultrafiltration were operated for around 2 years.
- Iron-II-chloride and aluminium-III-chloride were evaluated for phosphorus removal. An efficiency difference of the coagulants was not found.
- β-value was mainly driven by sRP effluent and influent concentrations.
- sRP effluent concentrations of $\ll 50\,\mu g/L$ were observed for cloth-filtration and membrane filtration.
- sRP $\ll 50\,\mu g/L$ needed a β-value >5 mol Me^{3+}/mol sRP.

INTRODUCTION

The European Water Framework Directive requires the reduction of the phosphorus concentration in the effluent of conventional wastewater treatment plants (WWTP) to prevent eutrophication of the receiving water bodies. Eutrophication is mainly caused by soluble reactive phosphorus (sRP), which can be assimilated by microorganisms, algae, and plants. Many WWTPs face increasingly stringent standards to achieve a limit value of $100\,\mu g/L$ or less.

However, Correll (1998) reported that for most water bodies, concentrations of $100\,\mu g/L$ are unacceptable, whereas concentrations of $20\,\mu g/L$ still turn out to be problematic. In accordance, Sundermann et al. (2015) showed that the critical concentration for dissolved inorganic phosphorus at which rapid alterations in species frequency and abundance occur is $20\,\mu g/L$. The increasing load of organic micropollutants and pathogens in the aquatic environment

causes new challenges for the WWTPs. In a conventional WWTP, these pollutants are not adequately removed and therefore enter the aquatic environment (Schwartz *et al.* 2003; Jekel *et al.* 2015).

Post-filtration processes in combination with coagulants provide a solution to ensure compliance with low phosphorus concentrations and future requirements. To accomplish concentrations of 50 µg/L of sRP, coagulants must be added in a relative dosage (β-value in mol metal added per moles sRP removed) of more than 10, see Table I in the supplementary material. Benisch *et al.* (2007) achieved phosphorus concentrations ranging from 15–35 µg/L with filtration technologies. Gnirss & Dittrich (2000) demonstrated the feasibility of reaching total phosphorus (TP) effluent concentrations of approximately 30 µg/L with microfiltration (MF) systems. Low-pressure MF and ultrafiltration (UF) membranes are commonly applied to improve the quality of biologically treated wastewater with the objective to reduce pathogens (Tchobanoglous *et al.* 2014). Compared to membrane-based filtration processes, the energy consumption of cloth filtration (CF) is considerably lower. Low Total Suspended Solid (TSS) concentrations in the effluent are necessary to achieve low (total) phosphorus concentrations. A reduction of 1 g TSS decreases the particulate phosphorus by approximately 0.02–0.05 g. Many studies are available on the separation of solids or/and removal of phosphorus with CF, see Table II in the supplementary material. In the past, needle felt – a randomly woven fibre fleece consisting of a multitude of small fibres – has been used as filter material (Loy 1993; Grabbe *et al.* 1998), which was successively replaced by pile fabrics (Grabbe & Seyfried 2002). In filtration applications with pile fabrics, there is no defined pore size; however, the length, density and stiffness of the pile fibres will define the performance. Grabbe *et al.* (1998) found that the TSS effluent concentration with normal pile fabric could be reduced up to 3.7 mg/L and even to 2.7 mg/L with fine pile fabric. Since then, new pile fabrics have been used, which show an improved removal of suspended solids. The objective of this research was to assess the technical and economic feasibility of CF, MF and UF for tertiary phosphorus removal. A thorough comparison between CF, MF and UF (in case disinfection is not required) and their benefits and drawbacks was conducted. For this purpose, we ran coagulation batch and full-scale experiments to evaluate the removal efficiency of different coagulants. Additionally, β-value of different coagulants for a target phosphorus concentration of <50 µg/L and systematic research of the benefits for tertiary filtration, including

coagulation-flocculation, were studied. This study provides a further understanding on reaching sRP concentrations <50 µg/L with different coagulants.

MATERIALS AND METHODS

Treatment plant design and operation

Secondary effluent (SE) from a conventional WWTP in the south of Hesse (Germany) was used as influent of the pilot plants. The treatment processes in the WWTP included a conventional activated sludge process for biological nitrogen and chemical phosphorus removal. The average water quality of the secondary effluent during the investigation period is shown in Table III in the supplementary material. The sludge is anaerobically digested and the reject water is returned to the treatment plant without any further treatment. The tertiary WWTP consisted of a pre-treatment stage for secondary phosphorus and suspended solids removal as well as a battery of activated carbon filters for effluent polishing. For tertiary treatment, CF (Mecana Umwelttechnik GmbH, Switzerland) and membrane filtration (MF and UF, Pall Corporation, Germany) in combination with a simultaneous addition of coagulants were used (Figure I in the supplementary material). Table 1 shows characteristics and operating data of the filtration pilot plants.

The dosage of coagulants was selected based on primary jar tests (see supplementary material). For the addition of the specific dosing volumes, the coagulant was diluted in a ratio of 1:10, with effluent of the MF/UF for experiments. Iron-III-chloride ($FeCl_3$) and aluminium-III-chloride ($AlCl_3$) were used as coagulants for the full-scale experiments. For each batch, the pH value as well as the iron and aluminium concentrations were analysed. The characteristics of the coagulants are presented in Table IV in the supplementary material. The coagulant was dosed just before the feed tank using the inline mixing device with a hydraulic retention time (HRT) of approximately 5 s. During the tests on High Rate (HR) flocculation, the feed tank was bypassed so that the filtration was fed directly after the addition of the coagulant (<1 min). The secondary effluent (Q = 30–35 m³/h) passed a coagulation dosing station and reached a feed tank (V = 15 m³) for flocculation. From this tank, feed water was pumped to each filtration plant. The online sRP signal (Phosphax SC, Hach Lange) of the WWTP effluent was used for the load proportional coagulant dosing. The HRT for floc formation in the feed tank was

Table 1 │ Characteristics and operation data of the filtrations pilot plants

Characteristic		Cloth-filtration		Microfiltration	Ultrafiltration
		OptiFiber® Microfiber PES-14[d,f,g]	OptiFiber® Ultrafiber UFH-12[f,h]	MF	UF
Operation period	[mm/yy]	03/17–03/18	03/18–12/18	03/17–10/17	10/17–03/18
Operational time	[d]	412	273	231	157
Type		Drum filter		Hollow fibre modules	
Strainer	[μm]	–	2,500	300	300
Nominal pore size	[μm]	–	–	0.1	0.01
Molecular weight cut-off	[kDa]	–	–	–	150
Filtration area	[m²/module]	4	4	50	60
Fiber surface	[m²/m²]	∼ 200	∼ 1,250	–	–
Number of modules	[-]	1	1	5	4
Volume flow	[m³/h]	∼ 0–20	∼ 10–30	∼ 12	∼ 12/15
Flux	[L/(m²·h)]	∼ ,500–5,000	∼ 2,500–7,500	∼ 48	∼ 50–62.5
Filter velocity	[m/h]	∼ (1.2) 2.5–5.0	∼ (2.2) 2.5–7.0	∼ 0.048	∼ 0.05–0.0625
Filter material		Polyester[e]	Polyester (80%), polyamide (20%)[e]	Polyvinylidene fluoride	
Backwash (medium)		No external backwash water		Air and permeate	
Backwash interval	[min]	∼ (5) 15–120[c]		15/30/45/60[i]	15/30/45[i]
Cleaning interval	[d]	–		∼ (7) 10[a] ∼14[b]	∼ (7) 12[a] ∼16[b]
Cleaning chemicals		–		NaOH, NaOCl, citric acid	
Cleaning duration	[min]	Backwash during operation (∼23 s)		1. 60 (alkaline/ oxidative: pH ∼ 12; T ∼ 40 °C) 2. 30/60 (filtration phase) 3. 60 (acid: pH ∼ 2; T ∼ 30 °C)	

[a]Operation with coagulation agent.
[b]Operation without coagulation agent.
[c]Depending on the solids surface load and flocculation time.
[d]Study on a modification of PES-14 (PES-14-DW) in the period between 02/18 and 03/18.
[e]Pile fabrics; back weave 54% polyester and 46% polypropylene.
[f]Use of two different suction beams: operation of PES-14 and UFH-12 with standard model and UFH-12 with modification of the suction beam from 05/18.
[g]Diameter approximately 7.4 μm; total thickness with pile fabric 14 mm ±5%.
[h]Diameter approximately 4.6 μm polyester and approximately 2.5 μm polyamide; total thickness with pile fabric 12 mm ±5%.
[i]Duration of the air-permeate flushing about 160 s.

approximately 20–30 min and the tank was equipped with a gear stirrer (Turbo-Misch- und Rühranlagen GmbH & Co. KG) as well as an inclined blade stirrer (d = 900 mm). The energy density required for the floc formation was ≤60 W/m³ and prevented sedimentation processes.

In the CF, wastewater flowed through the cloth media into the filter drum, at which time solids are retained on the cloth and afterwards removed with backwash water. Backwash intervals were approximately 120 min (or less, depending on TSS influent concentration). More details and information (see Table 1) about the principal function

of the CF can be found in Grabbe & Seyfried (2002) and Grabbe (1998). The MF unit was operated for a period of 7 months and the UF unit afterwards for a period of 5 months. Both systems, with a polyvinylidene fluoride membrane were operated in dead-end mode. Unlike the CF, filtrate was not produced during the backwashing of the membrane systems (UF and MF). After 30 or 45 min the filtration stopped and the membrane was backwashed by reverse flow (40 s), air scrubbing and reverse filtration (60 s) and forward flush (60 s) for approximately 160 s in total. Different cleaning procedures were tested during this

research, the characteristics and operation data of the filtrations plants are listed in Table 1. After 10–16 d of operation, the membrane modules of both UF and MF were cleaned with NaOCl, NaOH (at 42 °C) and citric acid (at 30 °C) in two steps for a period of 120 min.

Sampling and chemical analyses

For the assessment of the filters, 24-h flow-proportional mixed samples (24-h-MS) were taken daily by automatic samplers at the sample points. Influent samples were taken before coagulant dosing, and effluent samples were taken after each filtration unit. The 24-h-MS were collected in 5 L glass bottles and stored adequately (dark storage at 4 °C). For the analyses of dissolved parameters, the samples were filtered through 0.45 μm cellulose mixed ester membrane filter (GE). The samples were analysed for TP, soluble TP (sTP), soluble reactive phosphorus (sRP), chemical oxygen demand (COD) and soluble COD (sCOD) by Hach cuvette tests (LCK 349 and LCK 1414) using a Hach photometer (DR 5000). The spectral absorption coefficient (UV) at 254 nm, 436 nm, 525 nm and 620 nm was determined with a UV-VIS spectrophotometer (DR 5000 Hach Lange; 50 mm QS cuvette, Hellma Analytics).

Additionally, the samples were analysed for sRP according to the molybdenum blue method with ascorbic acid (see DIN-EN-ISO (2004)) by cuvette with a path length of 50 mm using a Hach photometer (DR 5000) at a wavelength of 880 nm. A calibration line (with 13 points) in the range 5–400 μg/L was used for the determination. For quality control purposes, a double measurement of each sample, the measurement of a phosphorus standard solution (1,000 μg/L) and a blank value was performed for each batch. Further information can be found in the supplementary material. The acid capacity was determined according to DIN (2005). TSS results were based on the use of cellulose mixed ester membrane filters (pore size 0.45 μm, GE) according to DIN (1987). Turbidity measurements of 24-h-MS were performed with Hach Model 2100 Q IS turbidimeter. Different online sensors were used for process monitoring in addition to laboratory analyses. In the flocculation tank, the pH and temperature were measured (Orbisint CPS11D, Endress + Hauser AG) as well as turbidity (SOLITAX, Hach Lange). In the CF, turbidity (Turbimax CUS51D and CUS52D, Endress + Hauser AG and sRP concentration (Liquiline System CA80PH, Endress + Hauser AG) were continuously monitored online. Ultraturb sc Turbidimeter (Hach) was used to measure influent and effluent turbidity of MF and UF.

Data processing

The laboratory and process parameters were analysed using conventional statistical methods, clustering analysis and Random Forest (RF), to identify the factors influencing the β-value. The basic idea behind RF is to combine hundreds to thousands of decision trees into a model. The outcome of the RF would be the averaged results of the decision trees that composed it. In this work, RF was built to study the interactions between operational and process variables (input parameters) to the β-value (output parameter). Two case-scenarios were evaluated, first, the interactions between influent variables and the β-value was studied and in the second case-scenario, the interactions between influent and effluent parameters with the β-value were evaluated. Feature importance is a measure extracted from the RF that allows ranking and evaluation of the importance of each variable in the model (Breiman 2001). As part of the data analysis process, k-means clustering was evaluated under the hypothesis that the data would group according to the type of coagulant used in the operation (Hartigan & Wong 1979). The pre-processing step before the application of clustering and RF was the selection of a representative dataset from the total raw data. For the influent, concentration measurements below the limit of quantification (LOQ) were not taken into account for analysis. In contrast, concentrations in the effluent below the LOQ were replaced with the LOQ value. This approach prevented the overestimation of the removal performance of the filters. Afterwards, the dataset was scaled and the data analysed. The data analysis was performed in R version 3.6.2, the *randomForest* library was used to build the RF model (Liaw & Wiener 2002). The variables were ordered hierarchically according to the feature importance measure by RF and the corresponding influencing factors identified.

RESULTS AND DISCUSSION

Tertiary phosphorus removal and behaviour of the soluble phosphorus fractions

Figure 1 shows the concentration of sRP in the influent and the effluent of the flocculation stage as well as the β-value, considering the coagulant used. sRP effluent concentrations ranged between < LOQ (5 μg/L) and 262 μg/L, resulting in β-values of up to 20 mol Me^{3+}/mol sRP. The sRP effluent concentration obtained agreed with the results of Benisch
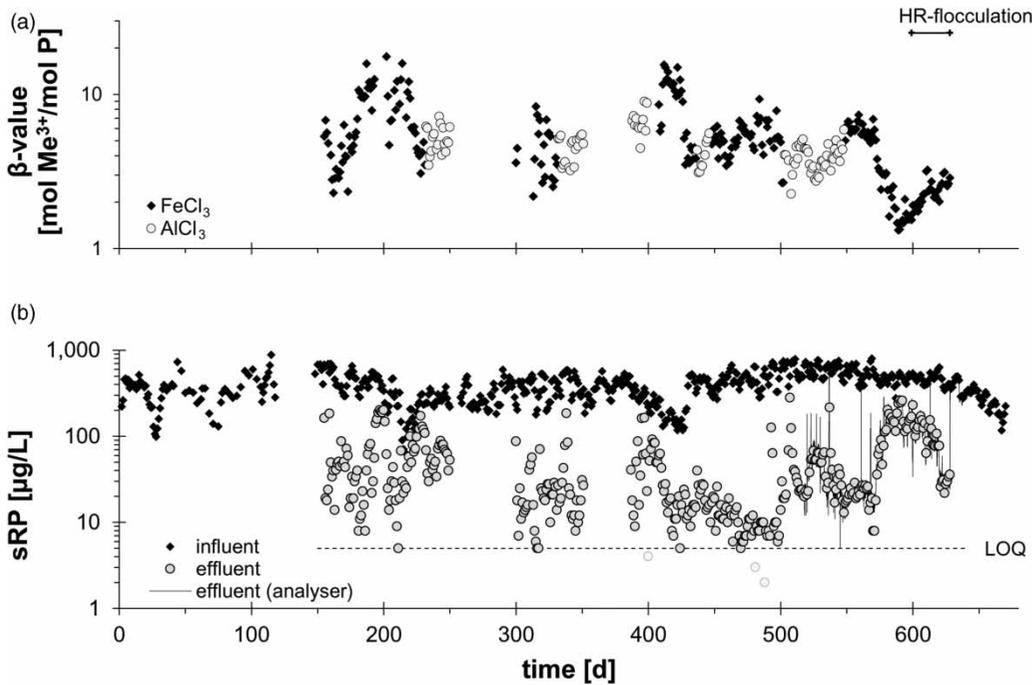
Figure 1 │ (a) Relative dosage (β-value) of coagulants ($FeCl_3$, $AlCl_3$) and (b) Influent and effluent soluble reactive phosphorus concentration (sRP) of the flocculation stage and sRP effluent concentration measured by an online analyser (CA80PH, E + H).

et al. (2011), deBarbadillo *et al.* (2010) and Dittrich *et al.* (1996). However, we observed β-values <10 mol Me^{3+}/mol sRP for removal of sRP, see Table I in the supplementary material. Figure 1(b) also shows no significant difference between the laboratory measurements and the online analysers (using the molybdenum blue method), which is applied for sRP concentrations >30 μg/L.

Figure 2 shows the concentration of sTP and soluble non-reactive phosphorus (sNRP) in the influent and effluent of the

tertiary phosphorus removal process. The results show that an efficient removal of sNRP is not achieved by the coagulation, also reported by Bratby (2016), which limits the minimum sTP concentration obtained. Due to the over-stoichiometric dosage of coagulants and the resulting coagulation and/or adsorption processes, a non-specific removal of sNRP was found. Scherrenberg *et al.* (2008) and Scherrenberg *et al.* (2011) showed that sNRP removal is associated with the increase of pNRP due to the coagulation. Stevens & Stewart
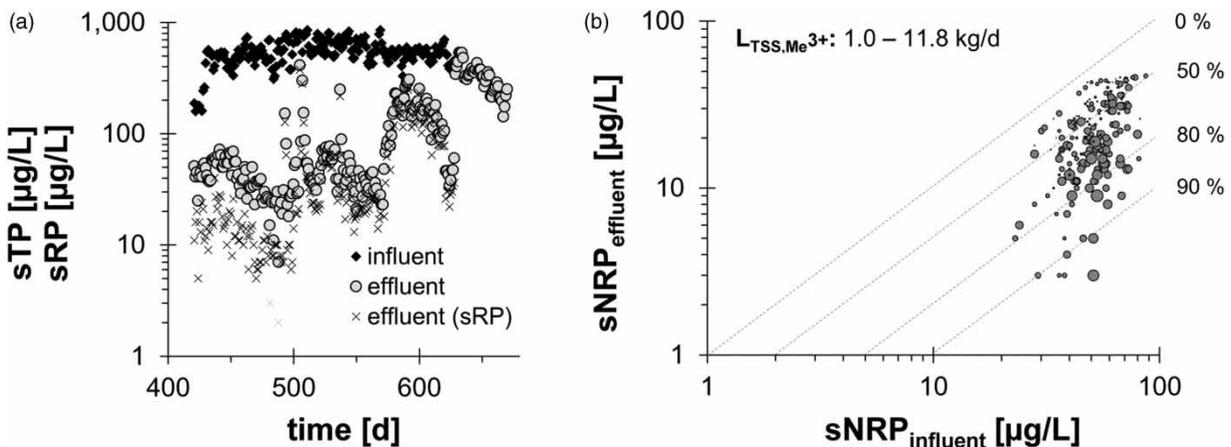
Figure 2 │ (a) Soluble total phosphorus (sTP) and soluble reactive phosphorus (sRP) over the operational time as well as (b) soluble non-reactive phosphorus (sNRP) in the influent and effluent of the flocculation stage in relation to the produced sludge during the sRP removal ($L_{TSS,Me3+}$).

(1982) described the increase of pNRP into colloidal NRP or sorption to colloidal substances. Figure 2(b) shows the influence of the sludge production during coagulation on the sNRP removal. A decrease of the flocculation time to <5 min (HR-flocculation) did not influence the sNRP and sRP removal. For both aluminium and iron containing coagulants (not differentiated in Figure 2), a reduction in the sNRP concentration was achieved. The average sNRP influent concentration of 51 µg/L was minimized to 22 µg/L in the effluent (Figure II in the supplementary material).

Coagulant demand and influencing factors

The degree of sRP removal is mainly controlled by three factors: dosage of coagulant, sRP influent and effluent concentrations alkalinity of the wastewater (Bratby 2016). The relationship between β-value and sRP in the effluent is illustrated in Figure 3(a). Evidently, an over-stoichiometric addition of coagulants was necessary to reach sRP effluent concentrations of less than 50 µg/L. The results confirm the results of the reviewed literature (see Table I in the supplementary material). The results of Szabó *et al.* (2008) show that in case of small dosages of coagulant, the relationship between dosage and sRP effluent concentration is close to linear; however, the specific removal of sRP decreases with increasing coagulant dosage. Figure 3 shows that, if the β-value was >3–5 mol Me^{3+}/mol sRP, sRP concentrations in the effluent of the post filtration system could be kept <50 µg/L. By returning the sludge from the

phosphorus removal to the flocculation stage, a decrease of the β-value can be expected (Wedi & Niedermeyer 1992; Maher *et al.* 2015).

The variation in the β-value is the result of the significant differences in the influent sRP concentrations and other parameters like pH (Sedlak (1991), Smith *et al.* (2008), Gu *et al.* (2007)), initial alkalinity and COD (Szabó *et al.* 2008). Optimal mixing and flocculation conditions are also essential for the adequate functioning of a subsequent coagulation (Smith *et al.* 2008). Figure 3(a) shows that significant differences in the β-value were mainly influenced by the influent concentration of sRP. For the first time, we could clearly observe the parameters influencing the β-value at full-scale. For pH, our results are consistent with Sedlak (1991) and Smith *et al.* (2008), for initial alkalinity and COD our findings are in accordance with those from Szabó *et al.* (2008). Moreover, we were not able to observe any significant influence of the applied coagulant on the sRP removal.

The specific sludge production is influenced by the β-value of coagulants, influent sRP (Veldkamp 1985) and effluent sRP concentration, see Figure 3(b). Regardless of the tested coagulants in the batch experiments, sludge production decreased for β-values between 3 and 11 mol Me^{3+}/mol sRP.

The increased sludge production at 2–3 mol Me^{3+}/mol sRP depends on the design of the jar test experiments (see supplementary material) and results from the not effective mixing of coagulants at the beginning of coagulation. The additional sludge production for tertiary sRP removal was
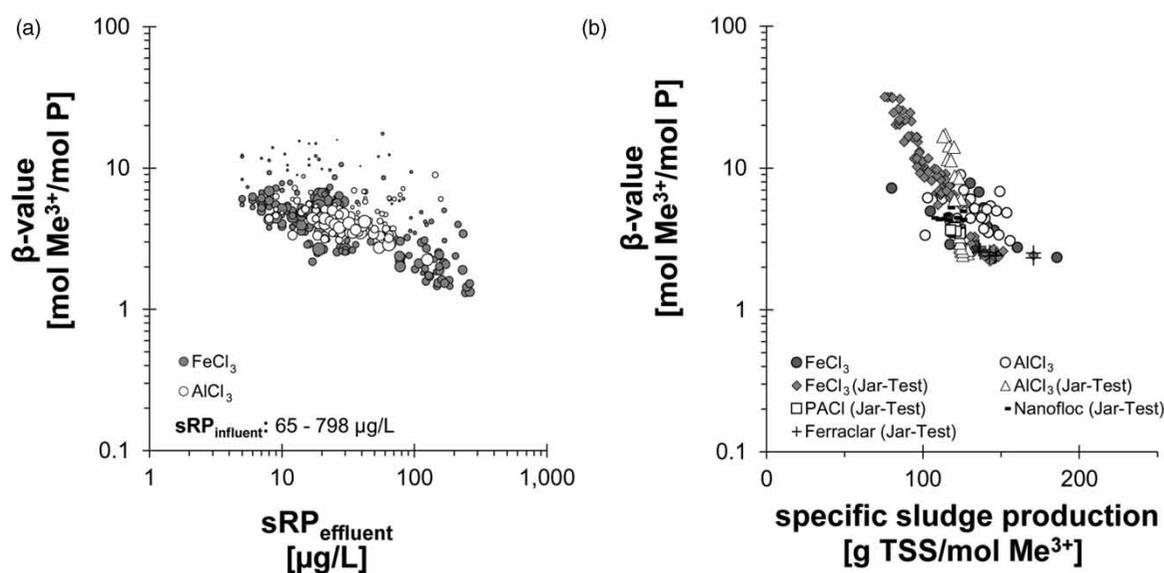
Figure 3 | Relative dosage (β-value) as a function of (a) the sRP concentration (in the influent and effluent) for FeCl$_3$ ($n = 222$) and AlCl$_3$ ($n = 108$) as well as (b) the specific sludge production due to phosphorus removal based on pilot tests and jar tests (see the supplementary material).

between 110 and 160 g TSS/mol Me^{3+} for a β-value between 2 and 11 mol Me^{3+}/mol sRP, which agrees with Wedi & Niedermeyer (1992) and the calculated sludge productions from Wedi (1995) and Veldkamp (1985). In case of low sRP concentrations in SE (<800 μg/L), higher sludge production can be expected at β-value >3 mol Me^{3+}/mol sRP.

The impact of different process parameters on the β-value were studied through feature selection criteria based on RF. The results show no significant difference between $AlCl_3$ and $FeCl_3$ for the sRP removal. The main influencing factors from the influent to the flocculation stage were sRP > pH > temperature, see Figure 4(a). When the parameters in the effluent were included (second case-scenario), there was a change in the order. However, sRP in the influent was still the dominant parameter. K-means clustering analysis was performed under the hypothesis that the operational data would group according to the type of coagulant used in the process (see Figure 4(c)). Indeed the optimal number of clusters resulted in two; however, after the analysis of the clusters' centers, the results showed that the data did not group following this criterion. Instead, our results suggest that there was no significant difference between $AlCl_3$ and $FeCl_3$ with regard to the β-value, which agreed with previous results. Temperature and sRP in the influent had a significant influence on the classes separation. $AlCl_3$ and $FeCl_3$ provide comparable results, though a direct comparison was not possible due to the different operating phases (see Figure 1). The studies of

Szabó et al. (2008) confirm that coagulants containing aluminium and iron are similar to pre-polymerised coagulants (such as polyaluminium chloride) and that pre-polymerised coagulants are less effective for sRP removal. A higher efficiency of $FeCl_3$ compared to $AlCl_3$ observed by Zheng et al. (2012) for the removal of sRP cannot be confirmed.

Phosphorus removal by cloth-filtration

To achieve TP concentrations of <100 μg/L, sNRP has to be removed, for which automation of dosing and process control is needed (Benisch et al. 2007; Tooker et al. 2010). The performance of the CF for TP removal is illustrated in Figure 5(a). Although less than ∼9% of the TP in SE corresponded to sNRP (51 ± 14 μg/L, $n = 201$), we were able to achieve stable effluent concentrations for TP <100 μg/L during the periods with coagulation dosage. The effluent concentration of particulate phosphorus (pTP) after CF varied according to the filter media. For PES-14, pTP concentrations of <90 μg/L and concentrations of <50 μg/L for (UFH-12), were obtained.

At the end of the test period, the flocculation stage was bypassed and the CF was fed directly without using the flocculation tank. Also with a lower flocculation time (<< 5 min), TP effluent concentrations <100 μg/L were achieved. The use of $FeCl_3$ or $AlCl_3$ on the pile fabrics at sRP effluent concentrations <50 μg/L and β-value >> 10 mol Me^{3+}/mol sRP did not cause a negative effect. The
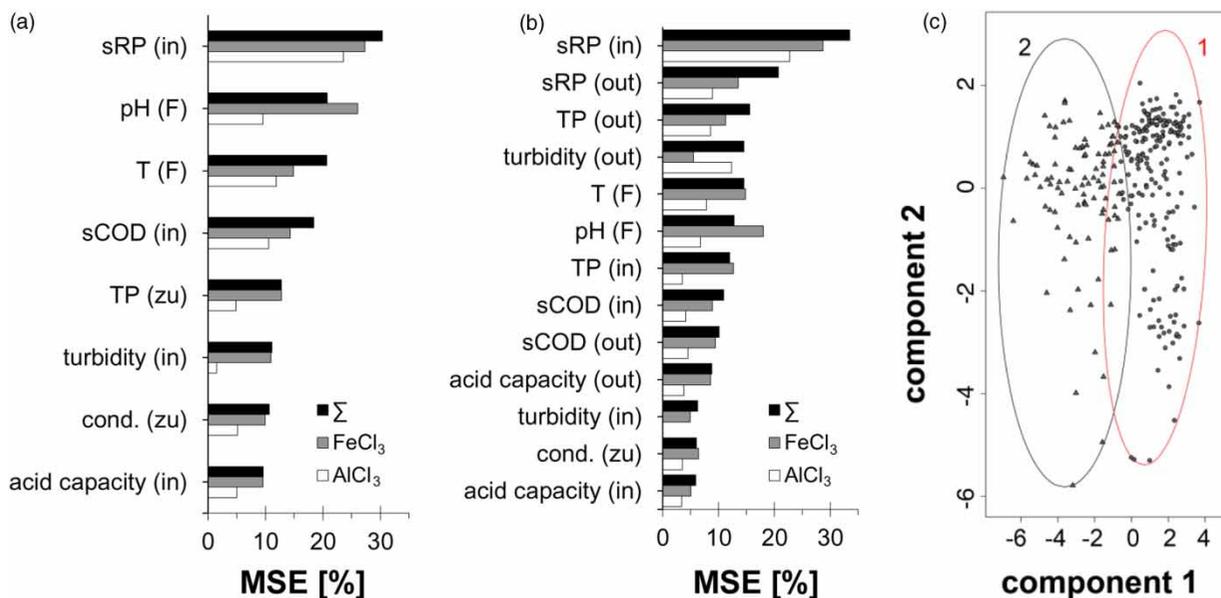
Figure 4 | Ranking of the influencing parameters for the relative dosage (β-value) in (a) effluent (out) and (b) influent (in) the flocculation stage (F) for $FeCl_3$ ($n = 222$) and $AlCl_3$ ($n = 108$) based on the mean square error (MSE) and (c) classification of the test results for the coagulants used.
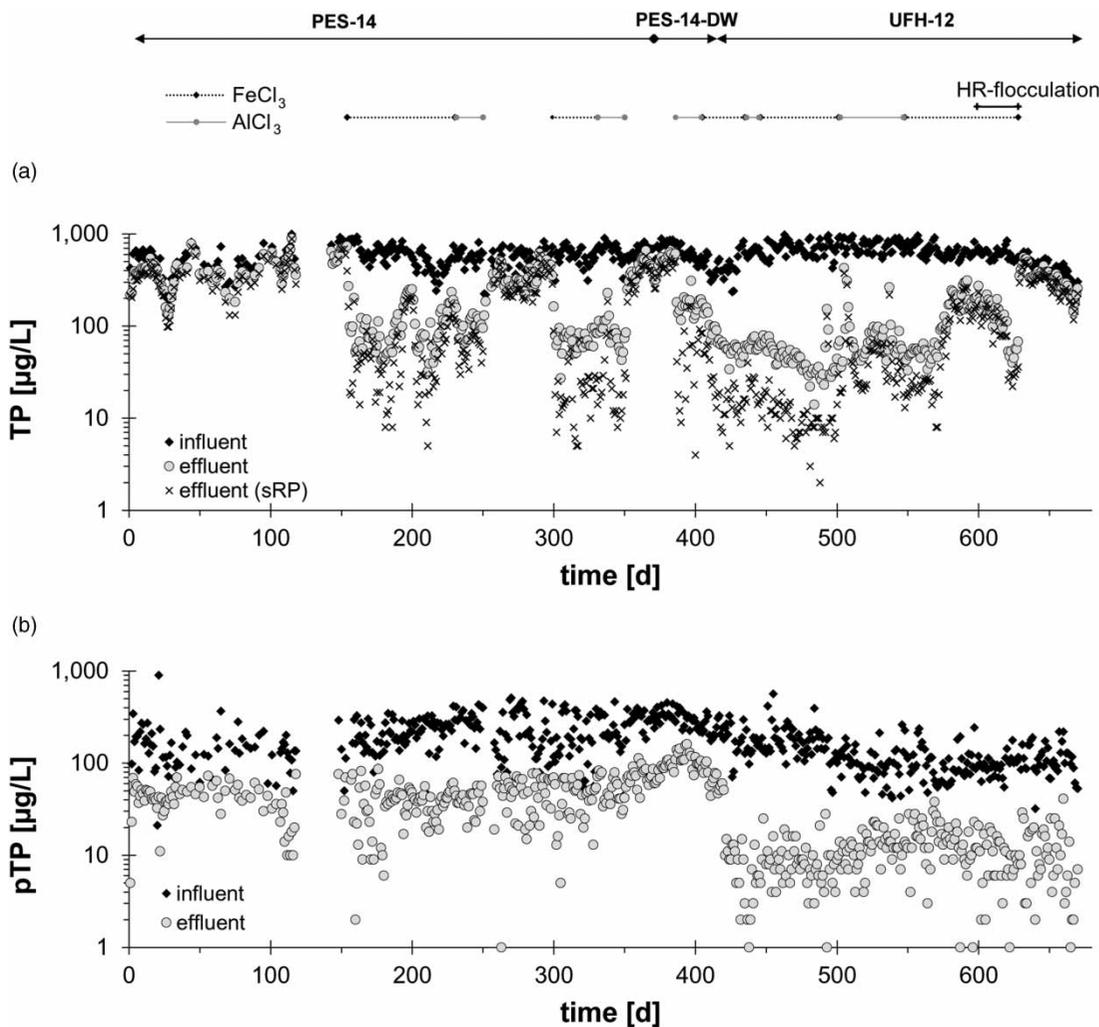
Figure 5 │ (a) total phosphorus (TP) and soluble reactive phosphorus (sRP), and (b) particulate phosphorus (pTP) based on 24-h-MS in the influent and effluent and the pile fabrics used (PES-14, PES-14-DW and UFH-12). *Note*: For PES-14 and PES-14-DW, pTP is based on the difference between TP and sRP and includes sNRP.

removal of particulate substances was maintained even during the phase of HR-flocculation.

Figure 6(a) and 6(b) compare the turbidity of the influent and effluent, for all the different filtration media used in the CF. The size of the points in the graph is based on filter velocity (v_F) and Solid Loading Rate (SLR) and shows the influence of those two parameters on the turbidity. Figure 6(c) and 6(d) show the pTP and TSS influent and effluent concentrations, respectively. For all the evaluated v_F and SLR (Figure III in the supplementary material), it was clear that UFH-12 provided higher removal compared to PES-14. The results show that the fibre surface had a substantial influence on the removal of particulate substances. This explains the improved removal by UFH-12, this filter media had a fibre surface 6 times larger than the surface of PES-14.

TSS after CF with PES-14 and UFH-12 were <2 mg/l over the whole operational period. The TSS-concentration in the influent had no relevant influence on the effluent. The removal of turbidity was not negatively affected by v_F and SLR. On the contrary, at higher SLR an improved removal of turbidity was observed. As a result of the accumulation of particulate substances on the filter surface, the efficiency increased over the filtration cycle, though the filtration period was shortened (Grabbe 1998). An increased SLR led to lower TSS and pTP effluent concentrations, whereas with reduced water temperature and constant influent turbidity an increase of the effluent turbidity was observed. The effect is caused by the reduced biological processes on the fibre surface, resulting in higher effluent concentrations in phases of low temperature (<12 °C). Although, pile fabric PES-14-DW had an antimicrobial
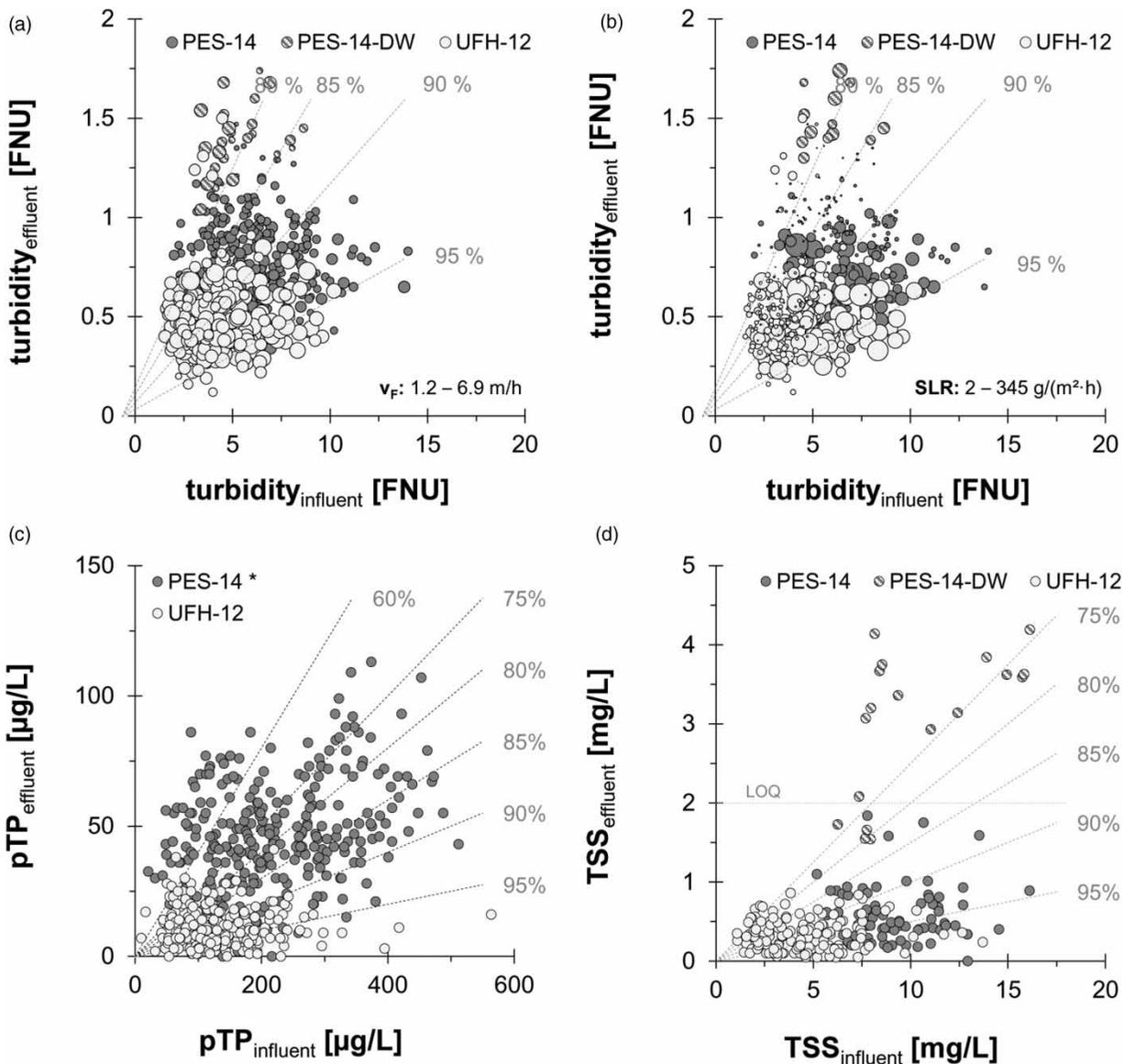
Figure 6 | Influent and effluent values with different pile fabrics (PES-14 and UFH-12) based on 24-h-MS. Turbidity values as a function of (a) filter velocity (v_F) and (b) solid loading rate (SLR). Removal of (c) particulate phosphorus (pTP) and (d) total suspended solids (TSS). *Note:* For PES-14 and PES-14-DW, pTP is based on the difference between TP and sRP and includes sNRP.

coating, as in the studies by Virgadamo Olivia *et al.* (2007), it showed reduced turbidity removal compared to PES-14. Low TP effluent concentrations resulted mainly from sTP and pTP removal. Figure 6(c) illustrates the improved retention of pTP due to the finer pile fabric. As result of removing turbidity >75%, pTP concentrations in the effluent (with UFH-12) achieved values << 30 μg/L. The average pTP concentration in the effluent was about 72% higher for PES-14 with 46 ± 21 μg/L ($n = 293$) than for UFH-12 with 13 ± 11 μg/L ($n = 279$). During the use of coagulants and filtration by CF, a TP concentration <100 μg/L was maintained regardless of the pile fabrics and the coagulants

used. The results can also be transferred to the particulate COD (pCOD), because the organic substances are almost dissolved in the effluent. The average of pCOD was 0.8 ± 0.6 mg/L (PES-14, $n = 231$) and 0.2 ± 0.3 mg/L (UFH-12, $n = 257$).

Energy demand and backwash water consumption by cloth-filtration

Operation costs are mainly comprised by the energy demand, backwash water/concentrate, cleaning chemicals of the filtration system and the dosage of coagulant. The

energy demand for backwashing depends mainly on the SLR. Figure 7 shows the energy demand in relation to the SLR for (a) PES-14 (suction beam type 1) and (c) UFH-12 (suction beam type 2). It was clear that, regardless of the pile fabrics, an increase in the energy demand occurred with increasing SLR. Although the suction beam was adjusted during the operation of UFH-12, the energy demand for similar SLR was about two times higher than for PES-14. The energy demand was <6.8 Wh/m^3 (PES-14, $n = 62$) and 11.8 Wh/m^3 (UFH-12, $n = 61$) respectively at SLR ≤ 100 g/(m^2·h) and >50 g/(m^2·h). The relationship between backwash water and SLR is shown in Figure 7 for PES-14 (b) and UFH-12 (d). The 6 times higher fibre surface area of UFH-12, resulted in higher backwash water volume at similar SLR by a factor of 2. Compared to Grabbe (2000) the backwash water quantities obtained in this work were higher; 1.6% (normal pile fabric) and 3.7% (fine pile fabric), at 5 m/h and 20 mg TSS/L. During HR-flocculation with UFH-12, the energy demand increased by a factor of 1.9 at SLR ≤ 50 g/(m^2·h). The modification of the suction

beam (type 2) resulted in an increase of the effective filtration surface due to the arrangement of the pile fabric as well as a more efficient cleaning. The modification of the suction beam resulted in a reduced energy demand by a factor of approximately 2.4 for the same SLR.

Comparison between cloth-filtration and membrane-based filtration processes

The TP and COD effluent concentrations using different filter systems with coagulation-flocculation are shown in Figure 8(a) and 8(b), respectively. The examined filter techniques performed well below sRP values of 100 μg/L. All the tested technologies achieved sRP concentration in the effluent <100 μg/L. Effluent concentration of pTP or sNRP for CF achieved values between 40 and 60 μg/L. In case of MF and UF, TP concentration corresponded approximately with concentrations of sRP, which indicated low concentrations of sNRP. By the application of membrane-based tertiary filtration, an average TP effluent concentration of
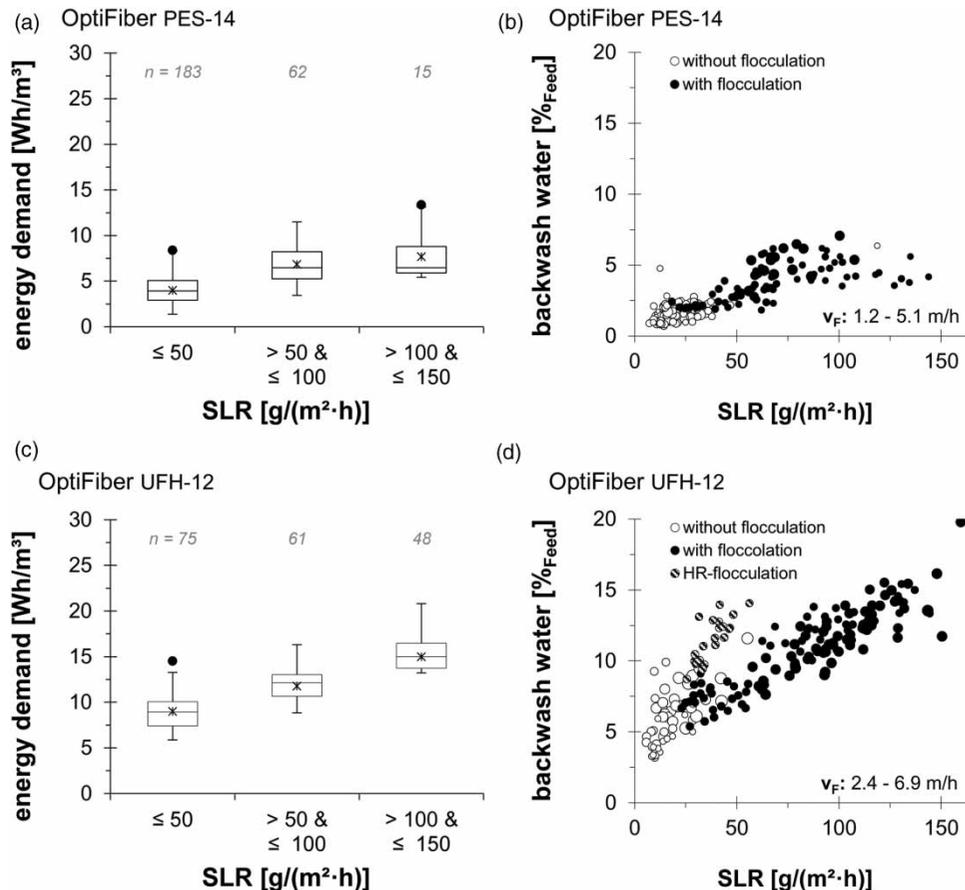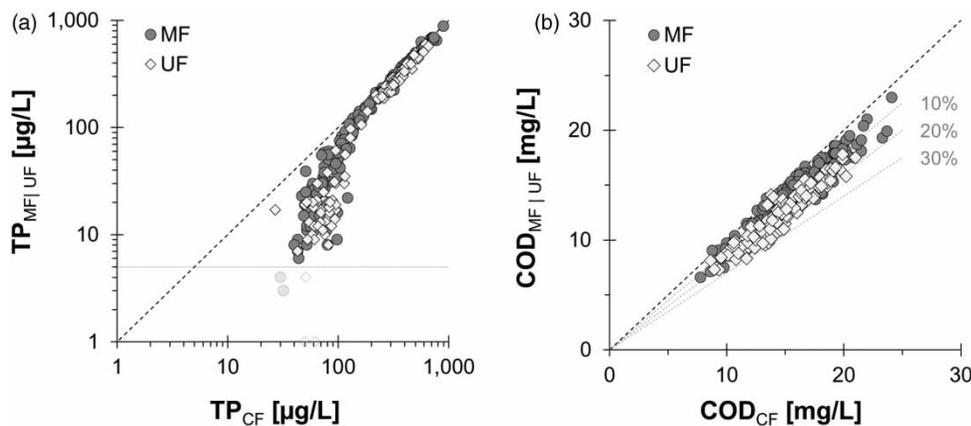

Figure 7 | Energy demand (a, c) and backwash water (b, d) as a function of the solids loading rate (SLR) and the pile fabric used: PES-14 (a, b) and UFH-12 (c, d) (Note: energy demand for UFH-12 with suction beam type 2).

Figure 8 | (a) total phosphorus (TP) and (b) COD effluent concentration of cloth-filtration (CF) with the pile fabric PES-14 and micro- (MF) or ultrafiltration (UF) in comparison based on 24-h-MS according to Fundneider *et al.* (2019).

60 μg/L (MF) and 30 μg/L (UF) was achieved, the TP concentration in the effluent was linked to the sNRP fraction. Differences between the CF and membrane-based post-filtration were only observed for TP concentrations >100 μg/L. In our work, we also demonstrated that pCOD can be nearly completely removed with CF. Additionally, the results showed that pCOD can be efficiently removed, similar to MF and UF, which can also remove sCOD. The mean removal for sCOD without the use of coagulants was 14% for MF and UF. The removal of sCOD can be increased up to 17 and 22%, by dosing of coagulants for MF and UF respectively, which agrees with Zheng *et al.* (2012).

The limiting factor of the COD discharge value is the proportion of sCOD compounds. More details on the removal of dissolved organic substances and the colouring depending on the dosage are presented in Figure IV in the supplementary material. The addition of coagulants ($AlCl_3$ and $FeCl_3$) within the tested dosage range (50 μmol/L and 100 μmol/L) did not achieve a significant removal (>5%) of micropollutants (Fundneider *et al.* 2019). For the retention of facultative-pathogenic bacteria and genes, CF were unsuitable (Bourgeous *et al.* 2003; Fundneider *et al.* 2019) in comparison to the membrane-based systems (Dittrich *et al.* 1996; Fundneider *et al.* 2019). The potential synergy effects of the use of a CF combined with membrane filtration included the removal of microplastic particles >90% and antibiotic resistance genes (>90%) (Fundneider *et al.* 2019). Figure V (supplementary material) shows the specific energy demand for CF, MF and UF during the investigation period of each system. The energy demand for MF and UF was on average about 0.1 $kW/m^3_{effluent}$ and corresponds to the results of Gnirss & Dittrich (2000) for a MF system (0.2 μm, hollow fibre) with an energy consumption of

0.12–0.17 $kW/m^3_{effluent}$ (depending on the backwash intervals with compressed air). In terms of MF and UF, the use of coagulants did not increase the energy consumption.

CONCLUSIONS

The process of coagulation-flocculation and filtration is essential to achieve extremely low phosphorus concentrations in the effluent of WWTPs. The main goal of tertiary filtration with coagulants is the removal of sRP and particulate substances. In this work, full-scale experiments were conducted to evaluate the capability of CF to achieve and quantify extreme low levels of phosphorus concentrations in the effluent. The benefits of this technology were also discussed. Based on the results, the following conclusions are highlighted:

- sRP effluent concentrations of << 50 μg/L were observed for CF, MF and UF. All the systems required optimal mixing as well as automated dosage of the coagulant. For achieving concentrations of sRP << 50 μg/L a β-value >5 mol Me^{3+}/mol sRP was needed.
- The sNRP concentration significantly affected the minimum sTP concentration that was achieved by flocculation filtration. The use of coagulants also led to a reduction in sNRP concentration.
- The β-value was mainly driven by sRP effluent and influent concentrations. The lower the influent concentration at constant effluent concentrations, the higher the β-value. An efficiency difference between iron and aluminium containing coagulants was not found.
- Regardless of the pile fabrics used, the CF achieved a TP concentration of << 100 μg/L when using coagulants. In

this context, sTP (mainly driven by sNRP) was >50% of TP.

- The CF achieved an effluent turbidity $<<$ 2 FNU regardless of v_F (PES-14: 1.2–5 m/h; UFH-12: 2.2–7.0 m/h), SLR (PES-14: 2–183 (345) g/(m^2·h); UFH-12: 6–160 g/(m^2·h)) and the pile fabrics used. On average, a turbidity of 0.76 ± 0.22 FNU ($n = 314$) was determined for PES-14 and 0.49 ± 0.17 FNU ($n = 275$) for UFH-12. The TSS concentration was below the LOQ of 2 mg/L, for all the cases.

- The energy demand of the CF depended on both the SLR and the pile fabric. With a SLR of ≤100 g/(m^2·h), the energy demand was <6.8 Wh/m^3 (PES-14, $n = 62$) or <11.8 Wh/m^3 (UFH-12, $n = 61$) respectively, and in comparison to MF or UF (with approximately 0.1 kWh/m^3) was significantly lower.

DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

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