Applying coagulation, flocculation and discfiltration in tertiary treatment

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Applying coagulation, flocculation and discfiltration in tertiary treatment

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To my family, Ylva and Ella.

Preface

This work is the result of an industrial PhD project between Hydrotech AB, Veolia Water Solutions and Technologies, and Water and Environmental Engineering, Department of Chemical Engineering, Lund University.

The work has been conducted within the framework of VA-teknik Södra with financial support from Hydrotech AB, Veolia Water Solutions and Technologies and Svenskt Vatten.

The study focused on advanced tertiary treatment of municipal wastewater for extended phosphorus and suspended solids removal, combining metal salt aided coagulation, polymer enhanced flocculation and discfiltration.

Summary

Reducing eutrophication in our surface waters caused by nutrient overload is of importance in order to ensure an environment in ecological balance for future generations. Municipal wastewater treatment plants are the main point source of nutrients emissions. Effluent water can be treated by introducing a final advanced treatment step to existing wastewater treatment plants. The present thesis concerns addition of coagulation, polymer aided flocculation and discfiltration using a 10 µm media in tertiary treatment to reduce total phosphorous concentrations in municipal wastewater treatment plant effluents to < 0.1mg/l. Experiments were conducted in laboratory scale, using modified jar test and test tube filtration methodology, and in pilot scale. The pilot experiments were conducted as a two year study at Ruhleben WWTP in Berlin, Germany (OXERAM project), followed by two months at Lundåkraverket WWTP, Landskrona, Sweden, and three weeks at Sjölunda WWTP, Malmö, Sweden. Various chemicals and combination of chemicals were tested at various doses. Furthermore diverse process conditions regarding hydraulic retention time and mixing intensity were applied to study the impact and to optimize chemical utilization, effluent water quality and discfilter performance.

The preferred chemicals were polyaluminumchloride (PACl) as coagulant and as flocculant aid, polyacrylamide based cationic synthetic powder polymer of high molecular weight and medium to high charge density. Both iron chloride ($FeCl_3$) and anionic powder polymers of polyacrylamide was producing an effluent similar in quality but filtration rate was reduced. Residual iron (Fe^{3+}) in the effluent and polymer dosing was higher when dosing anionic polymer.

The secondary effluents were on average containing 0.3 mg/l total phosphorus of which about 0.1 mg/l was identified as orthophosphate, 10 mg/l of suspended solids and 30-50 mg COD/l. Treating these effluents, the required dose to obtain an effluent containing < 0.1 mg/l total phosphorus was on average about 1.9 mg Al^{3+}/l and 0.6 mg/l synthetic cationic polymer as active material for an optimized coagulation/flocculation process. Using iron instead of aluminum as a coagulant, a dose of about 5 mgFe^{3+}/l and an increased polymer dosing to about 1-1.5 mg/l was required.

Experiments were conducted on wastewater treatment plants with various process configurations including plants with activated sludge, BIO-DENIPHO® and plants with activated sludge and biofilm systems. The

required dose to achieve < 0.1 mg/l total phosphorous in the effluent was on a molar basis identified to be similar for the three pilot plant experiments. A molar ratio of 5-7 moleAl^{3+}/mole influent total phosphorus was shown to be applicable and 0.07-0.1 mg polymer / mg influent suspended solids was to be applied. These findings are of interest if load proportional dosing relying on online measurement of influent phosphorus and suspended solids is to be considered.

Mixing intensity in the coagulation and flocculating stage was shown to influence the discfiltration process in achieving optimum chemical utilization, effluent water quality and discfilter performance.

Mixing intensity, defined as the mean velocity gradient G, of up to 270 s^{-1} in the coagulation stage and 150 s^{-1} in the flocculation stage was found to be applicable and an improvement in chemical utilization and increased filtration rates without loss in effluent water quality was observed. This is higher than normally recommended for other processes such as sedimentation or dissolved air flotation and a tradeoff between decreased chemical usage, improved filtration rate and an increased energy demand for mixing has to be considered.

Furthermore for improved performance, a useful mixing intensity was identified to be G=150-250 s^{-1} for coagulation and G=120-170 s^{-1} for flocculation and to be combined with a hydraulic retention time around 2-3 minutes in coagulation and 6-8 minutes in the flocculation. Moreover, it is also argued that a hydraulic retention time of 1.5 minutes in the coagulation and 4 minutes in flocculation could be applied when designing for peak flow conditions and this would not have an impact on overall performance to obtain an average TP of < 0.1 mg/l.

It was observed that the chemicals and dosages was affecting the performance of the pilot in a similar way as observed in the laboratory experiments regarding effluent water quality and filtration rates, thus the laboratory jar and test tube filtration experiment were qualitatively estimating performance of a pilot plant and therefore it can reduce time for optimization and the duration of pilot experiments would be shortened. It was also shown that the measuring of total phosphorus with the cuvette method relying on the formation of molybdenum blue was comparable with the more advanced ICP-OES (inductively coupled plasma-optical emission spectrometry) method.

Furthermore, the general improvement in effluent water quality from the coagulation, flocculation and discfiltration process was shown to give secondary benefits by improving the ozone utilization in reducing micro pollutants in secondary effluents. Applying coagulation, flocculation and discfiltration prior to ozonation the applicable ozone dose could be reduced by about 1.5 mg/l for the same micro pollutant reduction.

About the papers

This thesis comprises of the following original papers which will be referred to in the text by their Roman numerals I-IV.

Paper I	Langer, M.; Miehe, U.; Väänänen, J.; Stueber, J.; Bourdon, C.; Lesjean, B. 2011. *Advanced phosphorus removal with microsieves in tertiary treatment: an alternative to membrane filtration?* Conference presentation, 6th Specialist Conference on Membrane Technology for Water and Wastewater Treatment, Aachen, Germany.
Paper II	Langer, M.; Boulestreau, M.; Miehe, U.; Väänänen, J.; Bourdon, C.; Lesjean, B. 2012. *Advanced phosphorus removal via microsieve filtration in tertiary treatment: Performance and operation*. Conference presentation; Advances in Particle Separation – Science, Technologies, Practice, Berlin, Germany.
Paper III	Väänänen J.; Langer M.; Cimbritz, M. 2013. *Phosphorus removal with discfilters in tertiary treatment: Dosing Requirements*. Conference presentation, NORDIWA 2013, The Nordic Wastewater Conference, Malmö, Sweden.
Paper IV	Väänänen, J.; Nilsson, F.; la Cour Jansen, J.; Hörsing, M.; Hagman, M.; Jönsson K. *Discfiltration and ozonation for reduction of nutrients and organic micro-pollutants from wastewater – a pilot study.* Water Practice and Technology **2014**, 9(4).

My contribution to the papers

I contributed to the following in the papers.

Paper I	I planned the experiment together with the OXERAM team, helped conducting the experiments, performed data analysis and contributed in writing the paper.
Paper II	I planned the experiment together with the OXERAM team, helped conducting the experiments, assisted in laboratory result analysis and contributed in writing the paper.
Paper III	I planned the experiments together with my co-authors. I performed the experiments at Lundåkraverket WWTP, Landskrona and Sjölunda WWTP, Malmö, Sweden and was assisting in the experiments at Berlin WWTP. I analyzed the results and wrote the paper.
Paper IV	I planned the experiments together with my supervisors and co-authors. I conducted the experiments and part of the laboratory work in cooperation with Filip Nilsson. I wrote the paper, receiving comments from my co-authors and supervisors. The analysis of pharmaceuticals was performed by the Department of Chemistry of UMU.

Abbreviations

Amu	atomic mass units
ASTM	American Society for Testing and Materials
BAP	biologically available phosphorus
BW time(%)	discfilter backwash time in percent of total time of operation
COD	chemical oxygen demand
DLVO	Derjaguin and Landau, Verwey and Overbeek theory
G	mean velocity gradient
G*t	Camp number
HRT	hydraulic retention time
ICP-OES	inductively coupled plasma- optical emission spectrometry
NTU	nephelometric turbidity unit
PACl	polyaluminumchloride
SS	suspended solids
SUP	soluble unreactive phosphorus
TN	total nitrogen
TP	total phosphorus
WWTP	waste water treatment plant

Content

1. Introduction

Phosphorus is the 11th most abundant element on earth (Morton & Edwards, 2005) and exist primary as inorganic phosphorus in phosphate rocks (mostly as calcium phosphate) but is through biochemical processes converted to organically bound phosphorus (Liu, 2008). Data from Centra Europee d´Etudes des Polyphosphate published by Liu (2008) states, that the overall chemical phosphorus production from phosphate rock is used as fertilizers (80%) detergents (10%), animal feeds (5%) and in special applications such as metal surface treatment, corrosion inhibition, water treatment and ceramic production (3%). Many of these fractions are ending up in various waste streams. The main two contributors of the total input of phosphorus to for example, the Baltic Sea are found to be from wastewater treatment plants (50%) and diffuse sources (44%) (HELCOM, 2004).

Phosphorus in abundance is the major contributor to eutrophication seen in our surface waters (Selig *et al*., 2002; Boström *et al*., 1988), the Baltic Sea is one example but eutrophication and algae blooms can be found globally. Existing processes in modern advanced wastewater treatment plants including biological and chemical phosphorus removal and final particle separation are removing phosphorus to a large extent. For exampel Ryaverket WWTP in Gothenburg and Öresundsverket WWTP in Helsingborg, both located on the Swedish west/southwest coast are regularly reporting total phosphorus levels between 0.2-0.3 mg/l in their effluent (Öresundsverket, Miljörapport 2011; Miljörapport Ryaverket, 2012). Despite the extended phosphorus removal accomplished by modern waste water treatment plants, the remaining phosphorous is shown to be available for algae growth. Ekholm and Krogeus (1998) compared the bioavailability of the remaining 0.18-1.9 mg/l of total phosphorus in effluent from five Finnish municipal waste water treatment plants with and without nitrification, showing that about 50% (0-67%) of the total phosphorus was considered bio available. Similar results are reported by Gerdes and Kunst (1998) for plants located in the Lower Saxony – Area, Germany. Thus further phosphorus reduction is needed, and a tertiary treatment step would accomplish this.

Membrane-ultrafiltration, sand or discfiltration are methods that can be used in a tertiary treatment step. Furthermore, this treatment step is likely to include chemical phosphorus precipitation. Discfiltration in combination with coagulation/flocculation is potentially a cost and space effective technology in treating secondary wastewater effluents to reduce phosphorus, but also additional beneficial side effects such as reducing particles, COD and improvement in light transmission can be expected. Therefore it is of importance to investigate the combination of coagulation, flocculation and discfiltration for an extended period of time at various locations to evaluate overall performance and robustness of the technology.

1.1. Hypothesis and objectives

The main hypothesis of this thesis is as follows:

It is possible to reliably achieve a total phosphorus level of <0.1 mg/l in the effluent of wastewater treatment plants with coagulation, flocculation and discfiltration in tertiary treatment. The factors, chemical type and dose, hydraulic retention time and intensity of mixing are of importance for the treatment result. Additional benefits are also obtained.

The following questions needed to be answered.

- What are the type and dose of chemicals needed?
- What are the appropriate hydraulic retention times for the coagulation and flocculation stage?
- How does the mixing intensity in the coagulation and flocculation stage relate to the treatment results, what is the appropriate mixing intensity in term of mean velocity gradient G?
- What additional benefits are there?
- Can laboratory tests with modified jar tests including test tube filtration give similar results, qualitatively or quantitatively as pilot plant experiments and if so can laboratory test replace pilot tests?

The broad objective of this thesis was to study phosphorus removal applying coagulation/flocculation and discfiltration with a 10 µm woven media on secondary municipal waste water effluents. The type of chemicals and dosages and hydraulic conditions was varied in the pilot plants and in modified jar tests to study how phosphorus removal was effected. Secondary benefits such as reduction of particles, turbidity and chemical oxygen demand and to some extent, organic micro pollutant removal were also within the scope of this thesis. The characteristic of the phosphorous remaining after coagulation, flocculation and discfiltration is fractionated and evaluated in relation to what can be found in the literature. Furthermore, effort to identify specific phosphorus compounds in secondary effluents and their origin has been made from reviewing various publications. The quantity of these compounds is related to the amount of phosphorus that is defined as untreatable soluble unreactive phosphorus. An alternative path for removal of the untreatable fraction of phosphorus found in secondary effluents by removal from its source is discussed.

1.2. Thesis outline

The present work is based on four papers that are appended to the thesis and additional results are included for the overall evaluation of the hypothesis.

Fundamentals of the characteristics of municipal wastewater and treatment technologies with emphasis on phosphorous content and removal potential, with focus on coagulation, flocculation and particle separation principles are presented in Chapter 2 and followed by a brief presentation of the methodologies employed (Chapter 3). The relevant results obtained are presented and discussed in the various sections in Chapter 4. 4.1 Type of chemicals and dosing requirements, 4.2 Hydraulic retention time in coagulation and flocculation stage, 4.3 impact of mixing intensities in the coagulation and flocculation stage, 4.4 Operational experiences, 4.5 Jar test and test tube filtration methodology compared to pilot plant results, 4.6 Analytical precision of the rapid cuvette tests, 4.7 Phosphorus fractionation of the effluent, 4.8 Additional benefits by applying coagulation/flocculation and discfiltration. The main conclusions are presented in Chapter 5 and what appears to be of future research needs related to this work is presented in Chapter 6.

2. On phosphorus removal in wastewater treatment

The following chapter will give a broad introduction of the characteristics of untreated and treated municipal wastewater and municipal wastewater treatment methods. Definitions of particulate and dissolved matter in wastewater are introduced. Available separation methods and technologies are briefly reviewed and phosphorus and phosphorus fractions related to particle size in secondary effluents are described. Thereafter follows parts on coagulation/flocculation theory, phosphorus containing compounds in secondary effluents and finally, theory on discfilter operation principles.

2.1. General municipal waste water treatment methods and water qualities

Modern wastewater treatment includes advanced biological and chemical treatment in several treatments steps. Normally a treatment train consists of mechanical treatment to remove larger objects; primary particle separation, chemical/biological treatment for nutrient reduction and final treatment in a secondary particle separation stage. Table 1 show the average composition of untreated and treated municipal wastewater.

Table 1 General municipal wastewater quality (Henze *et al*., 2002; Gillberg *et al*., 2003).

	Total phosphorus (TP)	**Ortho-phosphate (Ortho-P)**	**Suspended solids (SS)**	**pH**	**Chemical oxygen demand (COD)**	**Biochemical oxygen demand (BOD_5)**	**Total nitrogen (TN)**
	(mg/l)	(mg/l)	(mg/l)		(mgO_2/l)	(mgO_2/l)	(mg/l)
Influent	4-14	3-10	120-450	7-8	210-740	100-350	20-80
Effluent	0.3-1.5	0.1-1	5-30	7-8	30-70	10-20	5-15

The typically removal rates of total phosphorus (TP), nitrogen (TN), suspended solids (SS) and chemical oxygen demand (COD) are about 70-95% for a normal wastewater treatment plant with primary clarification, biological carbon, nitrogen and phosphorus removal, including chemical precipitation and final clarification.

2.2. Particles and waste water

The components in the waste water that consists of other than water molecules can be classified with regards to the character they possess and thus are identified as "dissolved" or particulate matter. Classification of the sewage in wastewater treatment according to particle size is often done according to Table 2. In practice membrane filtration is used to separate the dissolved fraction from the colloidal, supra colloidal and settleable fraction that is often attributed to the particulate fraction of the sewage. Normally membranes with a poresize of 0.45-1.6 µm are used for fractionation of the two.

Table 2 Classification of matter in sewage (Levine *et al*., 1991).

Designation	**Dissolved**	**Colloidal**	**Supra colloidal**	**Settleable**
Particle size (µm)	< 0.01	0.01-1	1-100	>100

Discfiltration that is included in the concept of microscreens, sedimentation or sandfiltration are separating particles down to approximately 10-30 µm. For removing colloidal particulate matter (< 10 µm), agglomeration is necessary prior to this type of separation processes.

Excluding the water molecules, examples of composition of components within the sewage and applicable removal processes for the like are displayed in Figure 1.

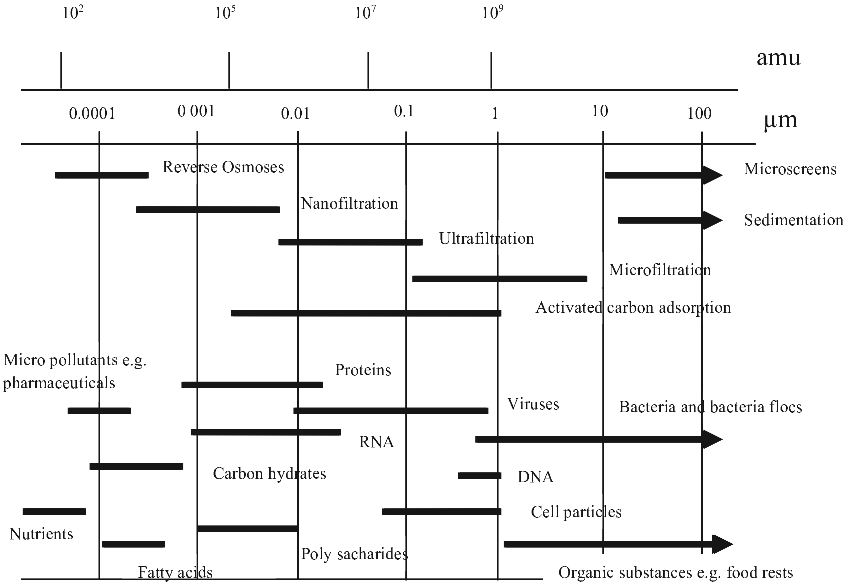

Figure 1 Composition of municipal waste water and removal technologies per approximate particle size (adapted from Nieuwenhuijzen, 2011; Metcalf and Eddy, 1998).

2.3. Phosphorus in secondary effluents

Phosphorus in secondary/final effluent from municipal wastewater treatment plants in the US and Europe generally have 20-80% of the phosphorus (TP) in a particulate range < 0.1 µm to < 0.01µm (Levine *et al.*, 1985; Tiehm *et al.*, 1999; Zheng *et al.*, 2012). In Table 3, final effluent TP concentrations from various investigations are shown.

Table 3 Total phosphorus concentrations and fractions in final effluents from a number of treatment plants.

Location	**Final effluent Tot-P** (mg/l)	**TP < 0.1 µm** (%)	**Reference**	
Las Vegas Nevada WWTP	0.70	83	Primary clarifier, trickling filter, chemical addition, secondary clarifier.	Levine *et al.*, 1985
	0.74	72		
	0.38	39		
	2.66	7		
Ahrensburg, Germany	0.3	32	No primary clarifier, line 1: activated sludge with bio-p, nitrification/denitrification, line 2 activated sludge < 4 days, intermediate clarifier, trickling filter, flotation $Al_2(SO_4)_2$ or $Fe_2(SO_4)_3$.	Tiehm *et al.*, 1999
Stellingen Germany	1.7	46	Primary clarifier, activated sludge with simultaneous P – precipitation with $FeSO_4$, secondary clarifier.	Tiehm *et al.*, 1999
Seevetal Germany	0.8	42	Primary clarifier, activated sludge with bio-P, nitrification/denitrification and simultaneous P -precipitation with $NaAl(OH)_4$, trickling filter, secondary clarifier.	Tiehm *et al.*, 1999
Bad Bramstedt Germany	1.0	19	Primary clarifier, activated sludge with bio-P, nitrification/denitrification and simultaneous P -precipitation with $FeClSO_4$, secondary clarifier.	Tiehm *et al.*, 1999

* < 0.01 µm

To separate phosphorus without chemical precipitation there is a possibility to use nanofiltration. Commercial membranes with pore sizes of $9 \cdot 10^4$ and $27 \cdot 10^4$ amu have shown to have a phosphate rejection of 62-97% including all protonated forms using synthetic water in the mixture of 1mM NaH_2PO_4, 1mM NaCl (Hong *et al.*, 2009).

Thus despite the high separation efficiency using nanofiltration the economical aspects for such an installation would be considerable. In order to obtain reasonable investment and operational costs, aggregation (coagulation/flocculation) prior to particle separation is probably the only alternative for very low phosphorus levels in secondary WWTP effluents.

2.4. Identified phosphorus compounds in WWTP effluents

Another approach to phosphorus removal would be to eliminate the phosphorus from its source. An example is the successful elimination of phosphorus from detergents. Specific phosphorus containing compounds in secondary effluents have been studied and compounds originating from micropollutant flame retardants, plasticizers and insecticides have been identified. Meyer *et al.* (2004) investigated the occurrence of organophosphates in waste water treatment plants effluents in North Rhine Westphalia studying the occurrence of chlorinated organophosphates, TCPP (tris-(2-cloroisopropylphosphate), TCEP (tris-(2-chloroetyl)-0phosphate, and TDCP tris-(1, 3-dichloroisopropyl)-phosphate and the nonchlorinated derivatives, TPP (triphenylphosphate) and tributylphosphate (iso and n-iso; TiBP TnBP). Calculating the phosphorus content in these compounds, the maximum contribution to the total phosphorus in the effluent was to be about < 1µg/l. Other studies have reported phosphorus in the form of TBP (tributyl phosphate), TCEP (tris-(chloroetyl)-phosphat and TBEP (tris-(butoxyethyl)-phosphate ranging from 500-30000 ng/l (Fries *et al.*, 2001), equalling to about 4 µg/l of phosphorous.

2.5. Coagulation and Flocculation

Due to the fact that most of the remaining phosphorus in secondary effluents is found in particles < 0.1 µm, the phosphorus containing particles/colloidal matter in the wastewater is to be stimulated to form larger aggregates i.e. to coagulate and flocculate for efficient separation in the discfiltration stage. The definition coagulation/flocculation is as follows (Bratby, 2006):

- Coagulation
  "Is the process whereby destabilisation of a given suspension or solution is effected. The function of coagulation is to overcome the factors that promotes stability of a given system"

- Flocculation
  "Is the process whereby destabilised particles or particles formed as a result of destabilisation, are induced to come together, make contact and thereby form large(r) agglomerates"

2.5.1. Destabilization mechanisms

Essentially from: Bache and Gregory, (2007), Bratby, (2006)

The destabilisation mechanisms in waste water are complex processes among others due to the complexity of the water matrix, thus the electrical double layer theory (DLVO) (Venvey & Overbeek, 1948) is used to describe the conceptual mechanisms of destabilisation.

The DLVO theory states that on assumed identical particles, the counterions are attracted to the surface and subsequently co ions are repelled from the surface and concentrated in the diffuse layer surrounding the particle (Figure 2). As a consequence, ions that are closest to the particle surface (within the Stern layer) are moving along with the particle in the suspension.

Particles that are approaching each other will have overlapping electric double layers thus particles will repell each other which will maintain stability of the system and a suspension is said to be in a its stable state when the electrical repulsive forces between the particles are dominating.

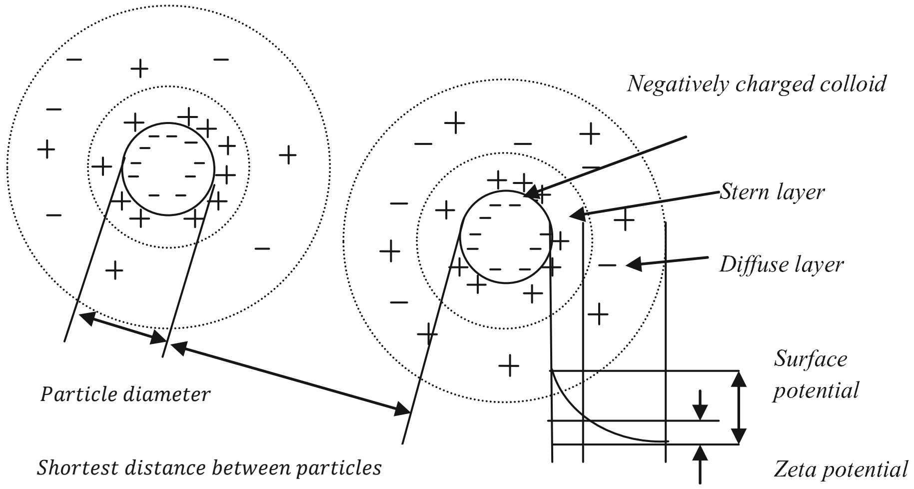

Figure 2 Conceptual scheme of electrical double layer on a negatively charged colloid/particle (adapted from Bache and Gregory, 2007)

The primary task in coagulation is to lower the repulsive forces to allow the particles to come in contact with each other and coalesce, thus achieve destabilisation of the suspension. Particles in municipal waste water are generally negatively charged at the typical pH conditions prevailing (Schrader *et al.*, 2005). On organic particulate matter these negative charges are a result of functional groups on the particle surface such as -OH, -COOH or -OPO_2H_2 groups. The charge can thus be altered by adjusting pH, and are to be positive or negative at low or high pH respectively. On solid surfaces, particle charge can also arise due to lattice imperfection in the outer atom layer. For example, on clay particles the negative surface charges can arise from atom substitution and are independent on the surrounding pH.

For organic particles in wastewater at a typical pH of 7-8 for secondary effluents, adsorption and charge neutralisation mechanisms (ACN) are thought to dominate. The positively charged metal ions such as Al^{3+} or Fe^{3+} can be adsorbed specifically, influencing the surface charge to lower the repulsive forces between the particles.

Zeta potential measurements can be conducted to measure the repulsive forces in the wastewater and the results can be used to estimate the amount of metal ions needed to minimise these forces to achieve destabilisation of the system whereby the particles can coalesce. Schrader *et al.* (2005) investigated the zeta potential of secondary municipal waste water with various Al^{3+} coagulant dosing conditions, observing a zeta potential of -12 increasing to -11 mV at a coagulant dose of 20 mg Al^{3+}/l, concluding that at normal waste water of around pH 7 there was no remarkable change in the zeta potential for the coagulant dose investigated. Zhu *et al.* (2008) have reported an average zeta potential of about -7.5 mV at pH 7.5 in secondary effluent from another treatment plant. For a dosing of 2-4 mg Al^{3+}/l and pH of 7, charge reversal is not likely to occur.

Another proposed destabilisation mechanism is charge reversal that is thought to take place if excess amount of metal ions are applied and electrostatic bridging is suggested to be the destabilisation mechanism. Precipitated products such as metal hydroxides that posses a net positive charge (*i.e.* Al_{13}^{7+}) can act as electrostatic bridges interacting with the net negative charge on the surface of the particles (Figure 3). The electrostatic bridging mechanism is thought to be dominating when dosing of metal is extensive, thus at a point when the zeta potential of the system has shifted from negative to positive value whereby charge reversal has taken place.

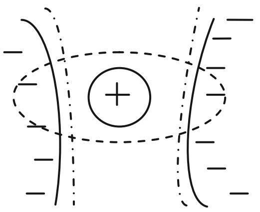

Figure 3 Schematic picture of electrostatic bridge interaction.

Growth of Al/Fe hydroxides is rapid due to their low solubility at pH around 7 (Duan *et al*., 2003). Amorphous precipitates will form enabling enmeshment of particulate matter (Neiuwenhuijzen *et al*., 2011) thus the destabilisation can arise from enmeshment/sweep coagulation mechanisms described by Packham (1965). In this context sweep coagulation also means sweep flocculation as the two processes are difficult to separate in time.

Destabilisation can also be induced from polymer bridging when polymers are used as primary coagulants and destabilization mechanism is said to rely on bridging mechanisms. Part of the polymer chain will adsorb onto the particle and some of the chain will thus tangle in the bulk liquid and randomly adsorb onto other particles, binding them together. Polymers can also destabilise particles by electrostatic patch interactions. The whole polymer is then attached to a specific area on the particle surface altering the local surface charge. Zones with opposite charge will attract each other possibly creating strong particle interaction holding the particles together. It would not be unlikely that also enmeshment/ sweep coagulation mechanisms are present and play an important role in the polymer enhanced coagulation/flocculation process investigated here.

The particulate phosphorus are thus said to coagulate and form larger aggregates relying on the mechanisms described above. The dissolved phosphorus fraction attributed to orthophosphate (i.e. PO_4^{3-}, HPO_4^{2-}) can be precipitated prior to coagulation and is in a simplified form brought into a solid state described by the following expression (Henze *et al*., 2002):

$$Me^{x+} + H_yPO_4{}^{z-} \rightarrow MePO_4 + yH^+$$

2.5.2. Flocculation

Initial destabilisation of the suspension is usually completed within seconds. Mononuclear alumina or iron complexes such as $AlOH^{2+}$ or $FeOH^{2+}$ are formed within nanoseconds and further adsorption to particles are also completed in the same timeframe. Moreover the double layer adjustment and Brownian diffusion collisions are all in the time range of less than 0.01 s. The slowest destabilisation mechanism are said to be from sweep coagulation (1-7 s) and particle surface charge adjustment up to (<< 1s to hours). Destabilisation occurring within the timeframe of 0.01 to 1 second seems to be most important (Amirtharajah *et al*., 1982) thus rapid dispersion of chemicals is desired. The following aggregation/flocculation rate will be governed by the collision rate, the aggregation efficiency during collisions and the rupture and breakage of the formed aggregates. The fundamental principles of flocculation kinetics are described by the following:

$$rate\ of\ flocculation = \alpha\beta(i,j)n_i n_j$$

Where

$$\alpha = collision\ efficiency$$

$$\beta = collision\ frequency\ between\ particle\ of\ size\ i\ and\ j$$

$$ni, nj = particle\ concentration\ of\ particles\ of\ size\ i\ and\ j$$

Perikinetic flocculation, are found most important for particle sizes < 1 µm (Thomas *et al.*, 1998) and the perikinetic flocculation rate is governed by collision efficiency, temperature and the particle concentration. Orthokinetic flocculation (particles > 1µm) involves mechanical stirring to create bulk fluid motion increasing the collision rate of the formed primary particles.

Camp and Stein (1943) introduced the term local root mean velocity gradient, G (s^{-1}), in the equation describing collision frequency ($\beta(i,j)$) for orthokinetic flocculation. In the Camp and Stein model, collision frequency between two spherical particles with diameter d_i and d_j are described by:

$$\beta(i,j) = \left(\frac{G}{6}(d_i + d_j)\right)^3$$

Where G was defined as

$$G = \sqrt{\frac{\varepsilon}{\nu}}$$

$$\varepsilon = local\ rate\ of\ energy\ dissipation\ (m^2/s^3)$$

$$\nu = kinematic\ viscosity\ (m^2/s)$$

According to the model, increasing collision frequency $\beta(i,j)$ by increasing G (s^{-1}) (increased mixing intensity) will render in faster aggregation. In Camp and Steins model, the rupture of formed flocs is not accounted for. However, intensive mixing will at a certain stage render in floc- breakage. Correlations between aggregation rate, maximum floc size and mixing intensity in various suspensions have been described by others (Flesch *et al.*, 1999; Gillberg *et al.*, 2003) showing that a too high mixing intensity break the formed metal hydroxide flocs but also that aggregation rate can be improved by increasing mixing intensity.

2.6. Discfilter technology

In municipal waste water treatment, commonly the microsieve technology, including disc- and drumfilters are used in tertiary treatment, primary treatment and also for stormwater treatment (Ljunggren, 2006). The discfilter technology is a continuous sieving process relying on gravity as the driving force. The membranes commonly used are of woven polyester type with a mesh size ranging from 10 to 1000 µm (Figure 4).

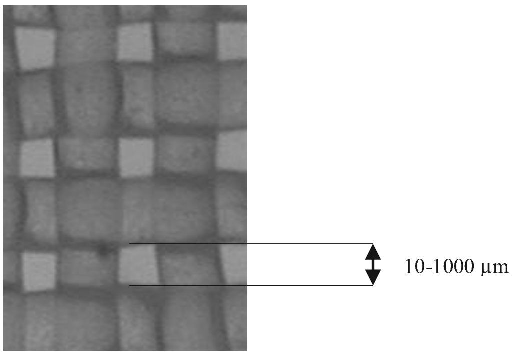

Figure 4 Micrograph of the woven polyester media used in discfilters.

In Figure 5 the Hydrotech Discfilter operation principle is shown. Water is fed into the centre of the filter and out in the filter segments. The effluent weir is placed 20-30 cm below influent water level during operation, creating the required pressure to force water through the woven media. During filtration the media is continuously being clogged causing the waterlevel to increase on the influent side of the filter. At a set level difference a backwash is initiated whereby the segment of discs starts to rotate and is backwashed, in tertiary treatment with pressurised effluent water (8 bar). During backwash solids are rinsed off, collected in a backwash gutter and by gravity drained of and clean media are being submerged in a continuous filtration cycle. The relative duration of the backwash filtration cycles in comparison to elapse time is an indicator of the relative maximum surface loading capacity available and is used to evaluate the response on the discfilter of upstream process changes. Commonly, the relative duration are expressed as a time percentage (BWtime %).

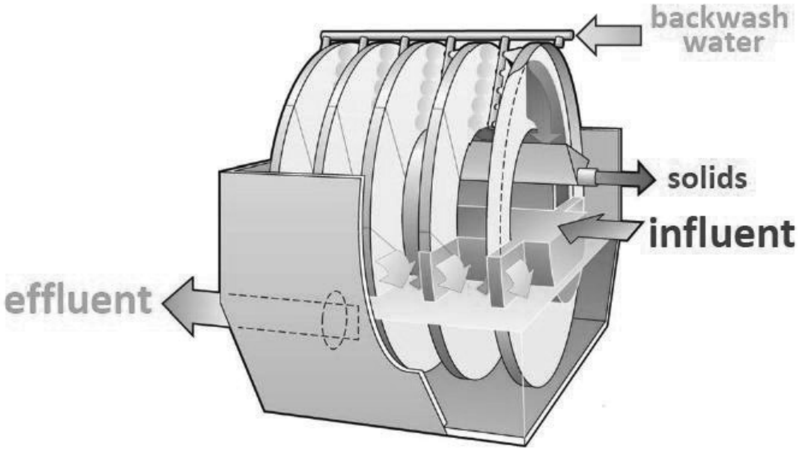

Figure 5 Hydrotech discfilter operating principle. The figure is reproduced with permission from Hydrotech AB.

3. Material and Methods

The following tasks were performed to seek answers to the research questions.

- A two (2) year pilot test running continuously at the Ruhleben WWTP in Berlin, Germany was performed within the OXERAM project.
- A two (2) month pilot tests was performed at Lundåkraverket WWTP in Landskrona, Sweden.
- A three (3) week pilot test was performed at Sjölunda WWTP, Malmö, Sweden.

Modified jar tests, including test tube filtration with 10 µm woven media were performed in conjunction to the pilot tests together with additional jar tests on other secondary effluents.

3.1. Pilot plants

In Figure 6a and b, is a schematic illustrations of the coagulation/flocculation/discfilter process with and without ozone used in the pilot tests shown. Discfilters were supplied by Hydrotech (HSF 1702/2F, HSF 1702/1F, HSF 2201/2F). The pilot plants were set up at Ruhleben WWTP in Berlin, Germany, Lundåkraverket WWTP in Landskrona, Sweden and Sjölunda WWTP in Malmö, Sweden. For the Landskrona pilot test, additionally ozone was used in combination with coagulation/flocculation and discfiltration (Figure 6b).

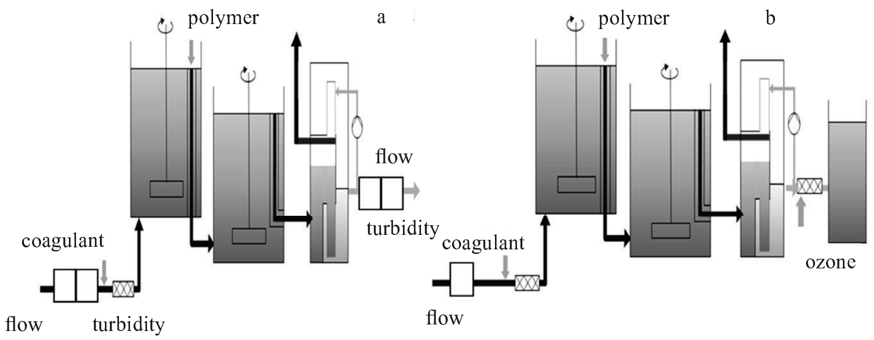

Figure 6 Schematic process configuration for a) coagulation/flocculation/discfilter and b) coagulation/flocculation/discfilter/ozone.

Process conditions were variable in the interval according to Table 4. The applied chemicals were as coagulants, polyaluminiumchloride (PACl) or iron chloride ($FeCl_3$) and as flocculants, synthetic polymers of polyacrylamide base (anionic/cationic powders and emulsions). One starch based cationic biopolymer was also used. Dispersion of chemicals was accomplished by a static mixer for the coagulant and in the overflow pipe between the coagulation and flocculation tank for the polymer.

Pictures of the pilot plants can be seen in Figure 7 to Figure 9.

Table 4 Pilot plants and process conditions.

	Discfilter type	**Filtration area according to manufacturer** (m^2)	**Flow** (m^3/h)	**Filter media** (µm)	**HRT Coagulation** (min)	**HRT Flocculation** (min)	**G Coagulation Max** (s^{-1})	**G Flocculation Max** (s^{-1})	**G dispersion of chemicals** (s^{-1})	**Coagulant dosing** (mgMe^{3+}/l)	**Polymer dosing** (mg/l)
Ruhleben WWTP	HSF 1702/2F	5.6	10-28	10	1.5-12	4-12	350*	150*	Coag. 700-1000** Flocc. 400-600**	1-5 Al^{3+}/ 2-10 Fe^{3+}	0.55-2
Lundåkra verket WWTP	HSF 1702/1F	2.8	5-20	10	2.7-10.8	2.7-10.8	120**	120**	Coag. 700-1000** Flocc. 400-600**	0-4 (Al^{3+})	0-1.5
Sjölunda WWTP	HSF 2201/2F	5.6	15-65	10	1.7-7.2	2.5-10.8	120**	120**	Coag. 700-1000** Flocc. 400-600**	1-7.7 (Al^{3+})	0.5-1.5

* Measured ** Calculated

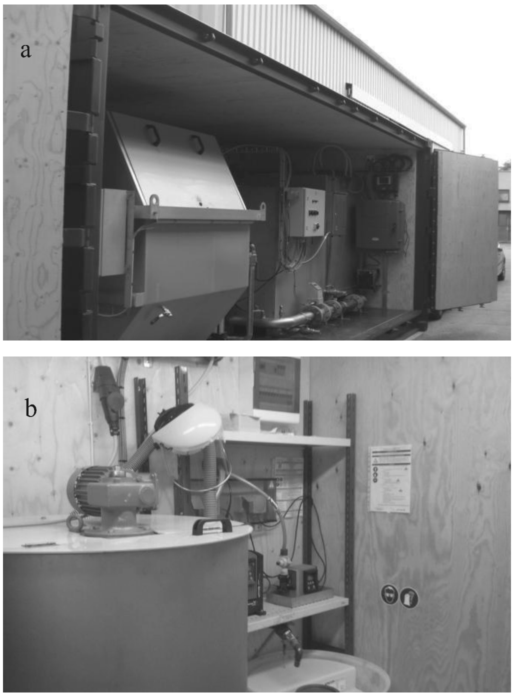

Figure 7 a,b) Pilot plant at Ruhleben WWTP, Berlin, Germany.

Figure 8 a,b) Pilot plant at Lundåkraverket WWTP, Landskrona, Sweden.

Figure 9 Pilot plant at Sjölunda WWTP, Malmö, Sweden.

3.2. Jar test and test tube filtration methodology

In wastewater treatment, jar tests are usually used for screening of appropriate chemicals (Bratby, 2006). The jar test methodology describes working procedures related to injection/dispersion of chemicals, mixing conditions and retention time (ASTM, 1995). In this study jar test methodology has been used in a modified form. The standard methodology was followed by a filtration step to simulate one filtration cycle in the discfilter.

The filtration test equipment consists of a transparent Plexiglas® cylinder with a mark placed at a distance of 200 mm from one end of the pipe, a measuring cylinder, a glass beaker with stirring device (Figure 10a and b). Furthermore test tube filter plates with a filter area of 0.0035 m^2 (Figure 10c) is mounted to the test tube and held in place with flexible pipe coupling (Figure 10d).

Figure 10 a) Transparent Plexiglas® cylinder with filter element installed, 1000 ml measuring cylinder, Kemira Flocculator 2000 b) variable speed flocculator c) cylinder filter elements d) cylinder filter element mounted on the Plexiglas® cylinder with a flexible pipe coupling.

The modified Jar test and filtration methodology was as follows:

1. A glass beaker is filled with water and the stirring devise is put into the beaker.
2. The mixer is set to a stirring speed of 400 rpm creating a mixing intensity of about $G=800\ s^{-1}$ (Cornwell *et al.*, 1983) and the coagulant is thereafter injected into the water with the aid of a pipette close to the impeller for rapid dispersion.
3. Rapid mixing is conducted for 10 seconds and thereafter slow mixing for 1-4 minutes.
4. After slow mixing of coagulant, stirring speed is increased to 400 rpm and polymer is injected close to the impeller for rapid dispersion.
5. Rapid mixing is conducted for 10 seconds followed by slow mixing for 2-4 minutes.
6. A 1000 ml measuring cylinder is used and placed under the test tube to collect the filtrate.
7. The flocculated water is poured into the test tube up to the 200 mm mark and by adding more water the differential pressure is maintained at this constant level (± 2 cm) during 10 seconds and thereafter the filtration is terminated by removing the test tube from the measuring cylinder (or vice versa if the test tube is fixated to a test rig).
   Note: Floccs accumulated on the walls of the beaker are gently brought into the suspension prior to the filtration step by tapping the walls.
8. Volume of the filtrate is recorded and additional analyses can thereafter be performed.

Stirring speed in the slow mixing stage of coagulation and flocculation are adjusted depending on the type of tests performed. In this study 50-100 rpm was used throughout the jar tests when studying mixing conditions and 50 rpm when screening for appropriate chemicals.

3.3. Pilot plant coagulation and flocculation mixing intensity measurements

During the pilot tests, the mixing intensity in the coagulation and flocculation stage was determined. The mixer motor was fixated on a friction free support bearing that was allowing the motor to rotate. Mixer power consumption was determined according to methods described by Leentvaar and Ywema, (1980). Power (P) was determined by measuring the torque with a Lutron FG 5005 torque meter. Power input was determined as torque times angular displacement according to the following.

$$P = Fl_a\omega \ \ (Nm/s)$$

$$P = mixer\ power\ consumption\ (Nm/s)$$

$$F = Force \ \ (N)$$

$$l_a = Length\ of\ lever\ arm\ (m)$$

$$\omega = 2\pi n_m$$

$$n_m = Impeller\ rotational\ speed\ (rad/s)$$

Applied mixing intensity (G) and impeller power numbers (θ) were calculated according to equation below (Bratby, 2006).

$$G = \sqrt{\frac{\theta \rho n^3 D^5}{V \mu}}$$

$$\theta = dimensionless\ powernumber\ related\ to\ impeller\ geometry$$

$$\rho = liquid\ density\ (kg/m^3)$$

$$n = mixer\ rotational\ speed\ (rps)$$

$$D = Diameter\ of\ mixer\ impeller\ (m)$$

$$V = Volume\ of\ coagulation/flocculation\, tank\ (m^3)$$

$$\mu = absolute\ viscosity\ (10^{-3}\ \ Ns/m^2\ for\ water\ at\ 20°C°)$$

Where

$$P = \theta \rho n^3 D^5 = F l_a \omega$$

3.4. Ozone

In the pilot experiments at Lundåkraverket WWTP, Landskrona, Sweden, ozone was used in combination with coagulation/flocculation and discfiltration. Ozone was produced using a Primozone GM2 ozone generator and industrial grade oxygen gas (AGA). The generation of ozone in the GM2 ozone generator is made through corona discharge. Ozone was dissolved into the water by a venture type injector and a pressurised stainless steel reaction vessel with a contact time of 2.6 minutes was used to allow ozone to react with the impurities. The experiments were conducted in cooperation with Primozone Production AB.

3.5. Analytical methods

Phosphorus was analyzed either by Hach Lange test equipment with LCK/LCS 349 cuvette tests, heat block and LZP 341 cuvetts together with a DR2800 spectrophotometer or with the ICP-OES method in accordance to DIN EN ISO 11885. Aluminum and iron was analysed with the ICP-OES method in accordance to DIN EN ISO 11885. The limits of detection for phosphorus were 5 µg/l for LCS 349 and 3 µg/l with ICP-OES method respectively. For Al of Fe the detection limit was 0.4 µmol/l and 0.5 µmol/l respectively. Transmittance and suspended solids (SS) were determined according to DIN 38404 and DIN EN 872:2005.Turbidity (Nephelometric Turbidity Units (NTU) was analysed with Hach 2100P turbidity meter. COD was analysed with Hach Lange cuvette tests, LCK 414 and Hach Lange DR2800 spectrophotometer.

4. Results and discussion

The results presented here are from the pilot experiment conducted at Ruhleben WWTP, Berlin, Germany, Lundåkraverket WWTP, Landskrona, Sweden, and Sjölunda WWTP, Malmö, Sweden and from laboratory experiments.

4.1. Type of chemicals and dosing requirements

As primary coagulants mostly iron or alumina products are used for chemical phosphorus removal at municipal WWTP. It was identified that for the application coagulation/flocculation/discfiltration in tertiary treatment, alumina (PACl) was more suitable than iron ($FeCl_3$). The main drawbacks when using iron were a reduction in filtration rate by approximately 20 % (observed as an increase in time the discfilter was in operation in backwash mode) higher residual coagulant content in the filtrate for the same dosing (mole Me^{3+}) and higher particulate fraction of TP in the range < 5 µm in the effluent (**Paper I**, **Paper II**). It can be concluded though, that both types of coagulant can be applied and depending on the type of coagulant to be used, the sizing of a discfilter installation have to be adapted. Residual Al or Fe will in some cases be decisive. Residual coagulant concentration for iron or alumina was found to be in the range of 0.01-0.04 mmolMe^{3+}/l (0.2-1 mgMe^{3+}/l) depending on coagulant/polymer dose and process conditions (**Paper I, Paper II).**

The chemical dosing requirements (10 µm pore size woven polyester media) to achieve < 0.1 mg/l TP was 1-5 mgMe^{3+}/l and about 0.6-2 mg/l cationic synthetic polymer (as active product) (**Paper III)** or about 40 mg bio-polymer/l as product. Molar ratios of about 5-7 mole Me^{3+}/ mol TP influent and a polymer dose (synthetic) of about 0.05-0.12 mg polymer/mg SS in the influent were identified to be a suitable chemical dose for the 3 pilot tests (**Paper III)**. For discfilter technology and also for other technologies used in tertiary treatment for extended phosphorus removal, a molar ratio of 1.5-2 mol Me^{3+}/ mol Ortho-P (Henze *et al.*, 2002) which would correspond to 0.75-1 molMe^{3+}/mol TP for the water matrixes used in this study would not be sufficient for an effluent with < 0.1mgTP/l (**Paper III**). It is believed that the relative high amount of coagulant on a molar basis is needed to promote sweep coagulation/enmeshment mechanisms that are thought to be the more pronounced coagulation mechanisms. The exact mechanisms for TP removal in secondary effluents are thus an open subject for further research.

Polymer addition was also proven to be crucial to create a sufficiently strong floc for the discfilter sieving process. In the Lundåkraverket pilot tests, PACl (Kemira kemi, PAX XL 36) was tried at a dose of 2.5 mgAl^{3+}/l without polymer addition, achieving TP removal of about 50% to be compared to filtration alone (10 µm) with a TP removal of 32%. When dosing both coagulant and polymer a TP removal of about 85-90% was achieved (**Paper III**).

The preferable polymer type was found to be linear high molecular weight, medium to high charged cationic polyacrylamide based powder polymer. Anionic synthetic polymer with similar characteristics as the cationic one was producing an effluent with similar water quality, but the main drawback was reduced filtration capacity (**Paper I**). The reason for the decrease in filtration capacity for the anionic polymer is unknown. Possible explanations are that the anionic floccs are blocking more surface area as they are adsorbed to the filter media during the sieving process. Other possibilities would be that the flocs are less effectively rinsed off during backwash. Though, anionic polymers have been tested elsewhere with satisfactory results without remarks on low filtration rates (Tooker *et al.*, 2012).

Several biopolymers of starch or chitosan base were tested in laboratory. One type was identified as promising and therefore further evaluated in pilot test. The result showed that it was possible to achieve similar filtration rates here seen as BW time% (the time the discfilter is operating in backwash mode per hour, expressed as percent of time) together with effluent quality comparable to the synthetic once in terms of TP, COD, SS and residual aluminium (Table 5).

Table 5 Comparison between cationic starch based and synthetic polymers. Influent and effluent water quality. Data is from Ruhleben pilot experiments Q=10m^3/h.

	Inf. TP (µg/l)	**Eff. TP** (µg/l)	**Inf. COD** (mg/l)	**Eff. COD** (mgO$_2$/l)	**Inf. SS** (mg/l)	**Eff. SS** (mg/l)	**Eff. Res. Al** (mg/l)	**BW time** (% per hour)
Biopolymer 2.4 mgAl^{3+}/l, 20-40 mgpol./l as product	640	56	48	32	12.3	2.5	0.330	27
Syntetic polymer 2.4 mgAl^{3+}/l, 1.5 mgpol./l	353	48	38	29	9.9	2.9	0.393	30

The price per kilo for this type of product is the main drawback and applications are thus limited.

Decreasing the chemical usage was of interest and the experience show that load proportional dosing relying on online measurement of phosphorus and turbidity can successively be applied and a reduction of chemical usage without loss in effluent water quality can be achieved (**Paper II**). Thus online monitoring of the influent water quality and adapting the chemical dosing thereafter, can be an effective tool to optimize chemical usage. Strategies that rely on online measuring of phosphorous and turbidity and adjusting the chemical dosing in relation to the input data to adapt dosing to the influent water quality would be possible. Probably the same dosing strategies can be used on numerous treatment plants without major adjustments. It was shown that to achieve an effluent with < 0.1 mg TP/l, the required molar ratio of influent phosphorus to alumina and influent suspended solids to polymer were in the same order of magnitude the same for the three pilot plant locations (**Paper III**).

4.2. Hydraulic retention time in coagulation and flocculation stage

In coagulation/flocculation and particle separation processes applied in municipal wastewater treatment, the hydraulic retention time in the coagulation and flocculation stage is often varying according to the daily flow patterns (assuming a constant tank volume). Sizing of the HRT in the plant is therefore usually done for peak flow conditions for a minimum of required retention time. In a discfilter application a variation in hydraulic retention time as a result of variations in flow affect the hydraulic loading on the discfilter. In the pilot experiment a maximum treatment capacity was obtained at flow of about 28-30 m^3/h.

The discfilter was then operating under continuous backwash without bypass. For this process settings the corresponding HRT at the maximum treatment capacity was 1.5 and 4 minutes HRT in the coagulation/flocculation stage respectively and moreover, on average a TP reduction of about 80 % was achieved and was stabel within the tested HRT and subsequent discfilter loading interval (Figure 11).

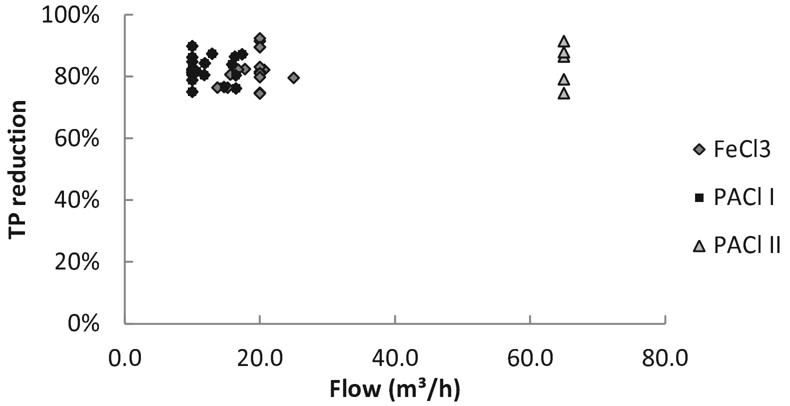

Figure 11 Total phosphorus reduction in the pilot plants for different flow conditions. Chemical dosing $FeCl_3$ 4.0 mg Fe^{3+}/l, 2.0 mg poly./l, Inf TP 0.320±0.141 mg/l, PACl I 1.9 mg/l Al^{3+}/l, 0.56 mg poly./l, Inf. TP 0.330±0.90 mg/l, PACl II 1 or 7.7 mg Al^{3+}/l, 0.5 or 1.5 mg poly./l, Inf. TP 0.42±0.12 mg/l.

It could be that the increased loading on the media (increased $m^3/m^2 \cdot h$) at increased influent flow will render in floc breakage in the filtration step that will reduce separation efficiency and a lower effluent water quality is then to be expected. Observed was that the effluent flow from the discfilter during operation was ranging between 7-28 m^3/h (**Paper II**). About 50% of the total flow is passing through the media close to a maximum filtration capacity (defined as influent flow divided by the total area of the filter media) more or less independently of the loading of the filter seen as percentage of the time the filter is in backwash operation.

A vast amount of the flocs are thus approaching the media at maximum velocity independently of the influent flow and are by that subjected to similar shear forces which could explain the stability of effluent water quality under load variations. The stability of the effluent water quality would thus imply that sufficient HRT in the upstream process are applied.

During laboratory experiments it is noticed that visible hydroxide flocs are formed during the first minute in coagulation and that flocs have a noticeable size already after 2 minutes in the flocculation stage thus it is thought that a sufficient treatment can be obtained even at lower HRT in the coagulation and flocculation stages. For a coagulant dose of 1.9 mgAl^{3+}/l and 0.6 mg/l polymer it is shown that the TP removal is > 80% at an HRT of 1 and 2 minutes in the coagulation and flocculation stage respectively. Increasing the chemical dose to 2.5 mgAl^{3+}/l and 0.7 mg/l polymer, a TP removal of > 90 was achieved (Figure 12). Increasing the HRT in the flocculation stage to 4 minutes for the lower chemical dose of 1.9 mgAl^{3+}/l and 0.6 mg/l cationic polymer, an improvement in TP removal was observed. Additionally a turbidity reduction of about 70% was observed with an average of 0.36 NTU in the effluent for an HRT of 1 + 2 minutes in the coagulation and flocculation stage compared to an average effluent turbidity of 0.31 NTU when applying 4 + 4 minutes of HRT. Thus the results are indicating that similar suspended solids reduction is obtained despite applying a shorter total HRT.

The results also show that a total retention time of about 1 minute in the coagulation stage and 4 minutes in the flocculation stage would be considered as the minimum, as TP removal was not increased despite increased HRTs in the coagulation stage. It is therefore suggested for peak flow occasions in order reduce investment costs and footprint, a reduced HRT design in coagulation and flocculation stages down to 1 + 4 minutes respectively can be applied.

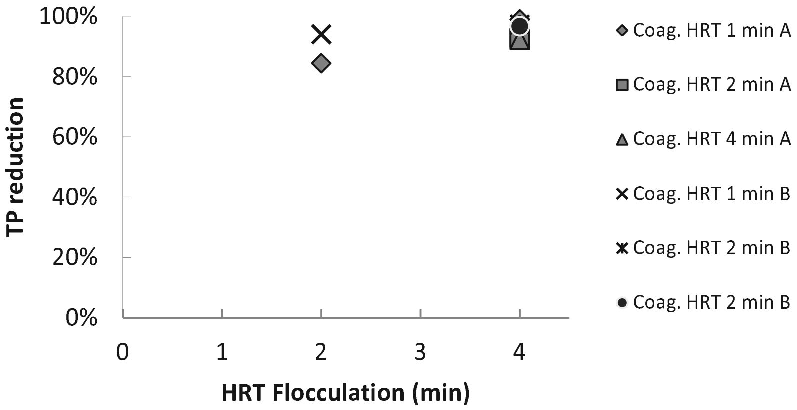

Figure 12 Jar and test tube experiments with varying retention time in the coagulation and flocculation stage. A) 1.9 mgAl^{3+}/l, 0.6 mg/l cationic polymer, influent TP 0.179 mg/l. B) 2.5 mgAl^{3+}/l, 0.7 mg/l cationic polymer influent TP 0.194 mg/l,. Coagulation/flocculation slow mixing intensity, G=130 s^{-1}. Test conducted on effluent from Källby WWTP, Lund, Sweden.

4.3. Impact of mixing intensities in the coagulation and flocculation stage

Supposed an appropriate polymer is used, mixing intensity in the coagulation and flocculation stage can play an important role for chemical utilization, effluent water quality and discfilter performance and considerations related to sizing of mixers would allow for an optimal process.

In the pilot plants, the rapid mixing and dispersion of the chemicals, defined as the mean velocity gradient (G) was calculated to be about 800-1000 s^{-1} for the dispersion of the coagulant and 400-600 s^{-1} for the polymer and was similar as utilized by others (Bratby 2006; DVGW W217-218). Thus to achieve the target of an effluent containing < 0.1 mgTP/l it was considered important to have a sufficient rapid dispersion of the chemicals during the injection phase to fully disperse the chemicals within the first few seconds into the water volume.

It was identified that the filtration rate was improved by increasing the mixing intensity in the flocculation stage by up to 70% in laboratory tests by increasing the mixing intensity from 50 s^{-1} up to 250 s^{-1}. It was also improved in pilot tests, shown as a decrease in BW time% for an equivalent flow with velocity gradients from about G=40 s^{-1} to G=150 s^{-1}. The improvement was thus not as significant as in the laboratory experiments. Thus if only to considerer the gain in filtration rate and this is to be equal to the energy expense, a vast increase in mixing intensity seems unrealistic before the improvement in filtration capacity is leveled out as the energy expense is about a power of two to the increase in mixing intensity.

Nevertheless it is also indicated that increased mixing improves chemical utilization and that it is to be conducted in both the coagulation and flocculation stage to be most effective. TP, SS and COD reduction was lower and residual aluminum in the effluent was higher when applying a low mixing intensity in the coagulation stage compared to applying a higher value for a higher mixing intensity in the flocculation stage. Thus a tradeoff between decreased chemical usage and improved filtration capacity and increased energy usage for mixing has to be considered.

The energy the floc is subjected to, the length scale of energy dissipation varies with retention time it is therefore of interest to takes this into account. The mean velocity gradient was for the optimized pilot plant allowed to vary between G=160-270 s^{-1} in the coagulation stage and the mixing intensity was adjusted to around G=150 s^{-1} in the flocculation stage for improved discfilter performance and better chemical utilization. The corresponding HRT was for the coagulation and flocculation stage 1-3 minutes and 6-10 minutes respectively.

4.4. Operational experiences

Applying chemical cleaning to the filter media at appropriate intervals is a must in order to maintain a high filtration rate and reduce operational costs. (**Paper II**). The general trend during piloting was that clogging was less if operation was continuous and that chemical dosing was held within the identified optimum dosing interval and during these process conditions, on average, a linear reduction of maximum filtration capacity of about 50-60% could be observed after 5-7 weeks of continuous operation. Calculations show that it is economically wise to do a chemical cleaning approximately once every 3 to 4 weeks since the cost of increased energy demand is higher than the cost of cleaning chemicals. The chemical cleaning will also render in less backwash water.

As polymer is used, the produced flocs was from time to time displaying floating properties, it is therefore important to have a design that facilitates floating sludge to be removed from the flocculation stage. In the pilot plant this was accomplished by having the effluent for subsequent separation in the discfilter placed in the top of the flocculation tank as a surface flow arrangement.

The water consumption for backwash was on average 1-3% of the treated flow during the 2 year pilot test at Ruhleben WWTP, and the backwash sludge/water mix was containing 580-1000 mgSS/l with an SVI index << 50 ml/g (**Paper II**). Calculations showed that the Lundåkraverket and Sjölunda pilot tests also had similar sludge production and compositions. The backwash water/sludge was observed to be sedimenting leading to clogging of the backwash gutter and this resulted in accumulation of sludge within the discfilter segments that eventually was reducing the filtration capacity close to zero. The occurrence of clogging of the gutter was most significant during low hydraulic loading and at increased chemical dosing conditions (> 2.5 mgAl^{3+}/l and > 1.5 mg polymer/l). In order to deal with this, a spray nozzle was mounted inside the gutter to flush at set intervals to keep it free of accumulated sludge. Avoiding the build-up of sludge within the gutter collecting the backwash water would also be important in full scale installations for reliable operation.

4.5. Jar test and test tube filtration compared to pilot plant results

Pilot tests results were compared to jar and test tube filtration results regarding TP reduction to evaluate the possibility if the jar test and test tube filtration experiments can eliminate or reduce the need for pilot experiments. It was observed that the TP reduction was more or less in the same range (Figure 13), thus the jar test and test tube filtration methodology is to be considered a useful tool to give accurate estimates on achievable effluent water qualities and screening of chemicals and dosing intervals and mixing conditions to be applied.

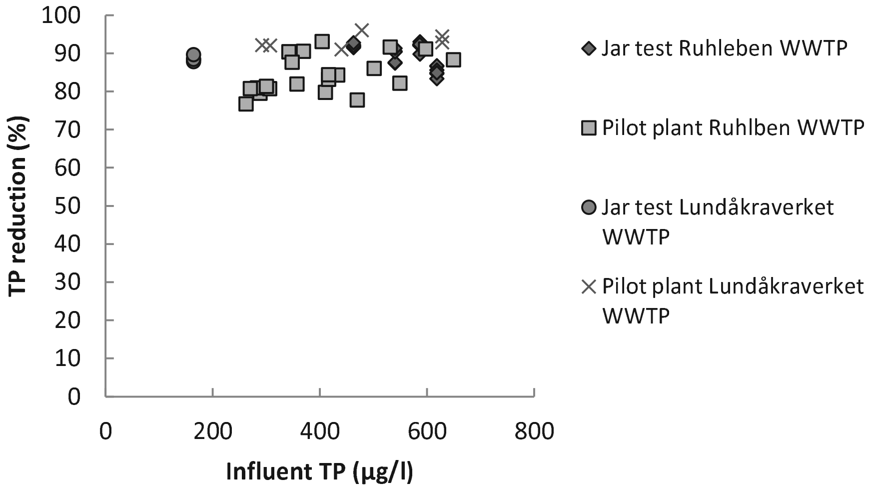

Figure 13 Jar and test tube filtration test results compared to discfilter pilot plant performance (10 µm) with a chemical dose of 4-6 mg Fe^{3+} /Al^{3+}/l and 1.5-2 mg cationic polymer/l.

Comparing the Ruhleben, Berlin pilot plant filtration rates to the obtained filtration rate in the test tube experiments, a conversion factor of approximately 0.2 was used to estimate achievable average filtration rates in the pilot from test tube experiment data. Thus for the other two pilot experiments the many changes in dosing and reoccurring restarts did not allow for stable operation and thus it was not possible to establish a realiable conversion factor for an average maximum filtration rate. Thus it was noted that the initial filtration rate with new media was comparable with the experience from the Ruhleben WWTP experiments. Noted are that the conversion factor is calculated for pilot plant peak filtration rates and on the nominal filter area of 5.6 m^2 and for the test tube filter segment with a filtration area of 0.0035 m^2. Studying the development of clogging of the media should be done in pilot scale as the development of the reversible clogging is occurring in the timeframe of 3-4 weeks or approximately 60 000 to 80 000 filtration cycles.

4.6. Analytical precision of the rapid cuvette tests

Two different methods were used for phosphorus measurements. A comparison between the colorimetric molybden blue method used in the rapid LCK/LCS 349 cuvett test from Hach Lange and the ICP-OES (inductively coupled plasma- optical emission spectrometry) according to (DIN EN ISO 11885) can be seen Table 6. In general the molybden blue method was estimating slightly lower phosphorus concentration compared to the ICP-OES method. The highest deviation was found for the chemically untreated influent water with an average of 22-36 µg/l depending if TP was analyzed for total or soluble total phosphorus. For the chemically treated effluent, the deviation between the two fractions was more or less nonexistent and was in the range from 3 to -6 µg/l. Moreover, for the soluble total phosphorus fraction in the effluent the ICP-OES method was thus measuring lower values compared to the LCS cuvette method and the reason for this is unknown. The conclusion is that when using the rapid test method, it will in most cases estimate a lower phosphorus reduction compared to the ICP-OES method. Nevertheless the deviation is not significant to influence the conclusions in this thesis. Inconsistency in reproducing results has been reported by others (Huang & Zhang, 2008) and Ormaza *et al.* (1996) showed that the decomposition methods play an important role for the precision of the analysis and that decomposition efficiency varies depending on the substance in question, thus when measuring phosphorus in waste water, inconsistency in the analysis should be expected.

Table 6 Total phosphorus measurements comparing the ICP-OES and molybden blue cuvette test method. The result is shown as the deviation in analysis results from grab samples (µg/l).

	ICP-OES - LCK influent (µg/l)	**ICP-OES – LCS effluent** (µg/l)	**ICP-OES – LCS influent 0.45 µm** (µg/l)	**ICP-OES – LCS effluent 0.45 µm** (µg/l)
TP	36±44	3±7	22±61	-6±15
n	33	33	33	17

4.7. Phosphorus fractionation of the effluent

Advanced phosphorus removal processes such as sand/dual/tri media filtration, microscreening or membrane ultrafiltration have previously been demonstrated to produce an effluent with a TP down to < 50-70 µg/l (Gu *et al.*, 2011; Tooker *et al.*, 2010; Genz *et al.*, 2011). The phosphorus content in the effluent at the Ruhleben WWTP pilot was characterised in terms of total phosphorus (TP), total phosphorus < 0.45µm, meaning the phosphorus content in the effluent after additional filtration through 0.45 µm membrane, orthophosphate and soluble unreactive phosphorus (SUP). It was identified (Table 7) that 37% was particulate (> 0.45 µm) and the remaining was defined as soluble, either as orthophosphate (15%) or as soluble unreactive phosphorus (SUP, 56%) thus similar as reported by others conducting coagulation/flocc and particle separation using either sedimentation, or membrane, sand, dual/tri media filtration on secondary effluents (Li and Brett, 2012; Zheng *et al.*, 2012; Ekholm *et al.*, 1998).

Depending on the aggressiveness of phosphorus removal in the treatment of the wastewater, the bioavailability varies. It has been demonstrated that without the use of chemicals in the upstream process about 79% of the phosphorus in secondary effluent is bio available thus can give rise to eutrophication and algae blooms. By applying simultaneous precipitation with the aid of alum the bioavailability of the phosphorus decreases to about 61%.After further tertiary treatment with alum and additional filtration via membrane and/or up flow/granular/sand filtration the bioavailability was lowered to 4-14% (Li and Brett, 2012) of the remaining TP in the effluent.

Table 7 Average total phosphorus distribution in effluent after coagulation/flocculation/discfiltration (μg/l).

TP (μg/l)	**TP < 0.45** (μg/l)	**Ortho-P** (μg/l)	**SUP** (μg/l)
55±23	39±17	8±5	31±12

The effluent orthophosphate concentration corresponds rather well to the 2-10 μg/l bio-available phosphorus (BAP) that was observed by Li and Brett (2012). Thus the coagulation/flocculation/discfilter process is thought to produce an effluent with a very minor available amount of phosphorus that can contribute to eutrophication.

A major part of the phosphorus (31 μg/l) in the effluent is of unknown character, usually defined as soluble unreactive phosphorus or SUP (Nanny *et al.*, 1997). The distribution of the TP was similar to other findings treating secondary effluents by applying coagulation and filtration (Li and Brett, 2012; Zheng *et al.*, 2012; Ekholm *et al.*, 1998). As the SUP is shown to be a relative large part (60%) of the remaining TP in effluents after advanced tertiary treatment, to further lower TP it would be necessary to asses this part and possibly some of the remaining SUP is of micropollutant origin and could be eliminated from the source.

As the various organophosphate compounds are vast and are used in large quantities for example as insecticides (Kiely *et al.*, 2004) it can´t be excluded that they in total can contribute to more than the identified and quantified 1-5 μg/l of micropollutant originated remaining phosphorous in secondary effluents.

4.8. Additional benefits by applying enhanced tertiary treatment

Micropollutants are increasingly being observed in waste water effluents worldwide (Gesuale *et al.*, 2010) and are recognised to disturb sensitive aquatic organisms. Ozone is known to be an effective oxidising agent in reducing micropollutants in surface and wastewater (Fontel *et al.*, 2011; Bahr *et al.*, 2007), therefore it was of interest to combine and test the two processes, coagulation/flocculation/discfiltration and ozone to both reduce mainly phosphorus (but also suspended solids and COD) and micropollutants in secondary effluent.

As ozone (O_3) is a highly reactive gas with a high oxidation potential and due to the high reactivity and short life length of the ozone molecule it has to be produced and consumed continuously as storage is not possible. In the study, ozone was applied prior or after coag./flocc. and discfiltration to compare the ozone efficiency on the removal of the studied micropollutants (**Paper IV**). The most interesting findings from this study was that ozone is more effective in reducing micropollutants if coagulation/flocculation and discfiltration is performed prior to ozonation and equal micropollutants reduction would be achievable with 1-1.5 mgO_3/l less ozone. Moreover as ozone had no effect on phosphorus, turbidity and suspended solids reduction, ozone could be applied either prior or after coagulation/flocculation/discfiltration. The general improvement in water quality (TP, SS, turbidity) is thought to be the main reasons for more efficient ozone utilisation. DOC is also known to effect ozone efficiency negatively in eliminating micropollutants (Hey, 2013), therefore another explanation could be from the absorption of micropollutants or other dissolved organic carbon compounds (DOC) on the formed flocs that are removed in the following sieving process thus resulting in a lower total DOC content in the water to be treated (Table 8).

Table 8 Influent and effluent DOC concentration from test at the Ruhleben WWTP pilot *n=17* (mg/l).

DOC	Influent	Effluent
(mg/l)	12.2±3	10.6±2

5. Conclusions

The combination of coagulation, polymer enhanced flocculation and discfiltration with a 10 μm woven polyester media treating secondary WWTP effluent was proven to be a reliable process combination to continuously produce an effluent with a water quality containing < 0.1 mgTP/l. On average, an effluent containing 0.05-0.06 mgTP/l was achieved during the pilot tests. Fractionation of the effluent revealed that about 0.016 mg/l was particulate (> 0.45 μm), 0.008 mg/l was identified as soluble reactive phosphorous or orthophosphate and the remaining 0.031 mg/l was untreatable and would belong to the soluble unreactive phosphorus pool. Furthermore, the soluble unreactive phosphorus concentration in the effluent from the coagulation/flocculation/discfiltration process was similar to what is achieved with a coagulation/ultrafiltration process. Micropollutant originated phosphorous in the form of organophosphates has been identified in secondary effluents previously at levels of about 0.001-0.004 mg/l. These compounds are contributing to unidentified remaining TP that would be part of the soluble unreactive phosphorus.

Comparing the effluent water quality regarding the eutrophication potential to similar studies including coagulation and particle separation in tertiary treatment, the water quality are indicating that the effluent from the coagulation/flocculation/discfiltration process would have a negligible contribution to the eutrophication in the receiving water bodies.

Ordinary secondary effluent containing 0.3-0.4 mgTP/l, 5-15 mg SS/l and 30-40 mgCOD/l, a coagulant dose of 1.9 mgAl^{3+}/l and 0.55 mg cationic synthetic polymer/l was sufficient to reach an effluent containing < 0.1 mg/l TP on regular basis. Iron can also be applied and the corresponding dosing is about 4-5 mgFe^{3+}/l. Synthetic polymer can be replaced with a starch based bio-polymer but the polymer requirement are increased several fold and a required dose of 20-40 mg product/l for similar effluent water quality was needed. A dosing relationship of 5-7 moleMe^{3+}/ mole influent TP and 0.07-0.1 mg polymer/mg influent SS can be used to control the dosing of chemicals in relation to the influent total phosphorus and suspended solids water quality.

The intensity of mixing in the coagulation and flocculation stage was found to be important for the performance of the discfilter regarding operation and effluent water quality. It was concluded to be higher than for sedimentation or dissolved air flotation. Mixing intensity in the coagulation stage was shown to improve effluent water quality and improve chemical utilization. A mean velocity gradient of up to G = 160-270 s^{-1} and a HRT of 2-3 minutes can be safely used. For the polymer enhanced flocculation, an increased mixing intensity improved filtration rates and a mean velocity gradient of G = 120-150 s^{-1} and allowing HRT of about 5-6 minutes can be applied. Noted is that a rapid dispersion of coagulant and polymer was applied.

Despite a variation in hydraulic loading ($m^3/m^2_{media}\cdot h$), effluent water quality was more or less constant with a TP reduction of about 80% and the constant effluent water quality was thought to be related the comparable shear forces the formed flocs are subjected to during the sieving process.

To maintain a high maximum filtration capacity chemical cleaning was necessary and it is suggested to be conducted every 3-4 weeks for the process to be operated most economically. Cleaning the media by applying hydrochloric acid followed by sodium hypochlorite there were no permanent or irreversible clogging observed during the two year pilot experiments conducted at the Ruhleben WWTP in Berlin, Germany. The filtration capacity decrease was lowest when the pilot plant was in continuous operation without changing chemicals or dosing settings. Changing chemicals and dosing was accelerating the fouling.

It was also shown that the pilot plant and a modified jar test, including a test tube filtration step produces equal effluent water quality, thus the modified jar test and test tube filtration methodology can be used in the screening of appropriate chemicals, chemical combinations, dosing interval dosing and mixing conditions to shorten optimization and save time in piloting. It also gives qualitative information on expected filtration rates. In this study it was found that a correlation factor of about 0.2 could be used to estimate the average filtration rates obtained in the pilot plant during a 4-6 week period from the filtration rate obtained in the laboratory test tube experiments. Thus it would be necessary to perform further parallel experiments due to the relatively few pilot experiments conducted, before a more general correlation on average filtration rates from laboratory and pilot experiment can be obtained.

The general improved water quality observed when applying coagulation/flocculation/discfiltration process was shown to improve ozone utilization for micro pollutant removal. The preferable process configuration was to conduct coagulation/flocculation/discfiltration prior to ozonation in eliminating/converting the investigated micro pollutants to the highest extent possible for the applied ozone dose.

6. Suggestions for further work

In this thesis secondary effluent was treated for extended phosphorus removal with the combination coagulation, flocculation and discfiltration. It was identified that on average a remaining phosphorous content of about 30 µg/l or 60% of the total phosphorous in the effluent could be assigned to be soluble unreactive phosphorus. It would be interesting to make an attempt on characterizing this fraction on a molecular level to identify specific compounds for further elimination from its source.

For the process combination coagulation/flocculation/discfiltration and ozone it would be interesting to study the development of the biological activity inside the discfilter and to specifically design for a bio film to growth on suitable carriers inside the discfilter to study the removal of ozonation byproducts.

Study the application of coagulation/flocculation and discfiltration in full scale would be of interest comparing to evaluate operational experience. At present, full scale installations are few and have recently being commissioned but as process data are accumulating this would be a very interesting study to perform. Additionally, it would be interesting to investigate the cause of the lower filtration capacity observed when using anionic polymers further and the combination of bio-coagulants and biopolymers could also be very interesting to test.

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