Effluent filtration for more than particle removal

Viviane Miska-Markusch

Effluent filtration for more than particle removal

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This book is dedicated to my grandfather Emmchen.

Contents

3. Description of filtration tests 45

Notation

List of symbols

α	coefficient in filtration theory	[sec^{-1}]
β	power law coefficient	[-]
λ	filtration coefficient	[m^{-1}]
ν	kinematic viscosity	[m$^{2} \cdot$ s^{-1}]
ρ_d	density of flocs	[kg $\cdot$ m^{-3}]
ρ_f	mass density of the filtration material	[kg $\cdot$ m^{-3}]
ρ_w	mass density of the water	[kg $\cdot$ m^{-3}]
σ	accumulated solids	[g $\cdot$ m^{-3}]
σ_v	volume concentration in pores	[m$^{3} \cdot$ m^{-3}]
τ	shear force	[kg $\cdot$ m^{-2}]

Re	Reynolds number	[-]
u	pore velocity	[m $\cdot$ s^{-1}]
c	content of suspended and colloidal solids	[g $\cdot$ m^{-3}]
y	depth of the filter bed	[m]
v	filtration rate	[m $\cdot$ s^{-1}]
p	porosity	[%]
k1, k2	constants	[-]
d	grain size	[m]
I	Resistance gradient	[-]

List of indices

<0.45 µm	0.45 µm filtered
0	Initial
A	Air
diss	dissolved
e	Expanded
F	Factor converting turbidity to suspended solids concentration
max	Maximum
p	particle size
part	particulate
part+diss	particulate plus dissolved
tot	total
unfil.	unfiltered
W	Water

List of abbreviations

AC	After Coagulation
AF	After Flocculation
Al	Aluminium concentration
BO	Beverwijk en Omstreken
BOD	Biochemical oxygen demand
bsCOD	Rapidly biodegradable organic matter
BZ	Beverwijk-Zaanstreek
CH_3OH	Methanol
COD	Chemical oxygen demand
CPV	Cumulative Particle Volume
dil	diluted
DO	Dissolved oxygen
DOC	Dissolved organic carbon
DWF	Dry weather flow
EDC	Endocrine disrupting compounds
EF	WWTP-effluent
Fe	Ferric concentration
$FeCl_3$	Ferric chloride
FHI	Fraunhoffer Institute
FI	Dual media filtrate
FI_ac	Filtrate activated carbon
FI_buffer	Filtrate buffer tank
FI_fs	Filtrate fine sand
G	G-value or energy input
MeP	Metal/Total Phosphorus ratio
MePO	Metal/Orthophosphate ratio
MTR	Maximum Tolerable Risk; concentrations given in Dutch directive Vierde Nota Waterhuishouding
NH_4-N	Ammonium nitrogen
N_{Kj}, TKN	Kjeldahl nitrogen
NO_2-N	Nitrite nitrogen
NO_3-N	Nitrate nitrogen
N_{tot}	Total nitrogen
NTU	Nephelometric Turbidity Units
NW4	Vierde Nota Waterhuishouding, Dutch directive
O_2	Oxygen
P	Phosphorus
PACl	Poly aluminium chloride; also PAC, PAX 11/14
PAH	Polycyclic aromatic hydrocarbon
PE	Population equivalent

PO_4-P	Orthophosphate
PSD	Particle size distribution
P_{tot}	Total Phosphorus
PVD	Particle volume distribution
RWF	Rain weather flow
SS	Suspended solids
stdev	Standard deviation
T	Turbidity
TPC	Total Particle Counts
TPS	Total Particle Surface
TPV	Total Particle Volume
TSS	Total Suspended Solids
undil	undiluted
UV	Ultra violet
WFD	Water Framework Directive, European
WWTP	Wastewater Treatment Plant

1

Introduction

1.1 Actual quality of WWTP effluent

1.1.1 Wastewater treatment: conventional and advanced

Wastewater can be defined as any water that has been adversely affected in quality by human influences. This liquid waste is removed through a sewer system from residences, institutions, and commercial or industrial establishments, and/or agriculture and eventually combined with groundwater, surface water, and storm water. The sewer system as infrastructure includes pipes, pumps, screens and channels, and transports the wastewater from its origin to the point of treatment.

Wastewater in the Netherlands has already been treated for decades. From 1900 to 1970 wastewater treatment aimed at the removal of colloidal, suspended, floatable matter, and pathogenic organisms as well as the treatment of biodegradable organics. Since the implementation of the act ‘Pollution of Surface Waters’ Act in 1970 many wastewater treatment plants were designed and built for the removal of oxygen consuming substances. The issue of ‘eutrophication’ of surface waters got special attention in the last 15 years in the Netherlands which led to the implementation of extended nutrient removal techniques (Van der Graaf, 2001).

Nowadays in the Netherlands, almost all wastewater (>98%) is treated in wastewater treatment plants WWTP (CBS, 2006). The total load of 26,213,000 population equivalents (PE's) and 1,853,577,000 m^3 wastewater is treated in 366 wastewater treatment plants (CBS, 2008). These plants include full mechanical and biological treatment, receive municipal and industrial wastewater and also have to handle rainwater from combined sewer systems.

Wastewater treatment aims for the progressive removal of wastewater constituents. Unit operations and processes provide different levels of treatment and are summarised in Table 1. 1.

Table 1. 1: Levels of wastewater treatment adopted from Metcalf and Eddy (2003)

Treatment level	Description	Process
Preliminary	Removal of rags, sticks, floatables, grit, and grease	Screening and grit removal
Primary	Removal of a portion of the suspended solids and organic matter	Sedimentation
Advanced primary	Enhanced removal of suspended solids and organic matter	Chemical addition or filtration
Secondary	Removal of biodegradable organic matter, suspended solids and nutrients (nitrogen, phosphorus)	Activated sludge, trickling filters, disinfection
Tertiary	Removal of residual suspended solids, eventually further removal of nutrients	Granular medium filtration or micro screening, disinfection, further nutrient removal

In accordance with the implementation of the European Directive for Urban Wastewater treatment 91/271/EEG (European Commission, 1991) 85% of the existing available treatment capacity includes phosphorus removal processes and approx. 60% is equipped with facilities for (biological) nitrogen removal. In 2005 almost half of the phosphorus removal capacity included biological phosphorus removal in most cases with additional chemical phosphate precipitation. As such the typical modern activated sludge treatment plant consists of anaerobic and anoxic zones for biological phosphorus and nitrogen removal.

The quality of modern WWTP effluent after biological treatment with extended nutrient removal is described in Table 1. 2.

Table 1. 2: Average quality of WWTP effluent in the Netherlands in 2006, CBS (2008)

Parameter	Unit	Average concentration
Chemical Oxygen Demand (COD)	mgO_2/L	42
Biochemical Oxygen Demand (BOD)	mgO_2/L	4
Total nitrogen (N_{tot})	mg/L	9

Parameter	Unit	Average concentration
Nitrate nitrogen (NO_3-N)	mg/L	7
Ammonium nitrogen (NH_4-N)	mg/L	2
Total phosphorus (P_{tot})	mg/L	1.8
Suspended solids (SS)	mg/L	11

The average quality of WWTP effluent in the Netherlands displayed in Table 1. 2 is well below the limits described in the national regulations for discharge of municipal wastewater (Ministry of Internal Affairs, 2008). The established concentrations within these regulations are described in Table 1. 3.

Table 1. 3: Current discharge limits for municipal wastewater treatment plants in the Netherlands (Ministry of Internal Affairs, 2008); pe: population equivalent

Parameters	Limit concentration	Remarks
BOD (without nitrification)	20 mgO_2/L	
COD	125 mgO_2/L	
SS	30 mgSS/L	
P_{tot}	1 mgP/L	> 100,000 pe
	2 mgP/L	2,000 – 100,000 pe
N_{tot}	10 mg/L	>20,000 pe
	15 mg/L	2,000 – 20,000 pe

1.1.2 *Quality parameters: regulated and non-regulated*

The pollution of surface water bodies can be attributed to different sources, such as discharges, atmospheric deposition, wash- and run-offs. The database of the Dutch emission registration (www.emissieregistratie.nl, 2006) contains data on the contribution of WWTP discharges to the total polluting load of surface waters. The contributions for significant parameters including arsenic, several heavy metals, nutrients, polycyclic aromatic hydrocarbons (PAH's) and one pesticide, are displayed in Table 1. 4 which compares the years 1990 and 2004.

Table 1. 4: Contribution of WWTP's to the polluting load of surface water bodies in the Netherlands, a comparison 1990 and 2004 in % (CBS, 2006)

	Contribution in 1990	Contribution in 2004
Arsenic	26	32
Cadmium	15	28
Chromium	37	25
Copper	18	10
Mercury	25	31

	Contribution in 1990	Contribution in 2004
Lead	13	5.8
Nickel	28	21
Zinc	18	15
Total phosphorus (P_{tot})	28	34
Total nitrogen (N_{tot})	32	24
Fluoranthene	0.87	4.1
Benzo(a)pyrene	1,1	6
Isoproturone	19	18

CBS (2006) reported that the concentrations in the WWTP effluent of almost all compounds have been significantly reduced from 1990 to 2004. Table 1. 4 indicates that the wastewater treatment plants still have a major contribution to pollution of surface waters. Around 20-30% of the pollution by heavy metals, nutrients and other micro pollutants originates at a WWTP's.

The general situation of pollution throughput Europe initiated additional measures to be taken by the European Union member states. In 2000 the European Water Framework Directive (WFD) was presented aiming to ensure a good quality of surface water and groundwater in Europe by the year 2015. The restrictions set by the Directive, especially for substances defined as priority or priority hazardous substances, will have a significant influence on the future of waste water treatment plants. These substances which have not been regulated or monitored before will become regulated in future like other conventional constituents, as chemical and biochemical demand, oxygen, nutrients and suspended solids.

1.2 Prospective for requirements of the effluent quality

1.2.1 Legislative changes

The conditions of surface water bodies and the contribution of wastewater treatment plant discharges to their pollution have led to actions on national and international (European) level. The Ministry of Transport and Water Management introduced in 1998 the fourth Memorandum on Water Management, in Dutch 'Vierde Nota Waterhuishouding' (NW4), presenting the water policy for the Netherlands until 2006 (Beesen, 1998). The Dutch directive NW4 defines the standards for the water quality and discharge points, and it gives new limits for concentrations of substances. This is translated to Maximum Tolerable Risk (MTR) concentrations as well as target values for nutrients. The MTR concentrations are related to the minimum water quality to be achieved by the end of 2015. The MTR values are preliminary and it is not known yet which final values will become effective. The target values are set as a standard to prevent negative ecological effects on a longer time scale.

In 2000 the European Union introduced the Water Framework Directive (WFD) 2000/60/EC. The WFD focuses on catchment areas, also known as river basin districts, rather than on individual countries. River basin districts can extend across country borders. In the Netherlands there are seven river basin districts. The European directive aims at a good ecological and biological state for all surface waters, coastal waters, transitional waters and groundwater in Europe by 2015. Therefore a list of 22 priority substances and their maximum allowable concentrations, FHI-values proposed by the Fraunhofer Institute (Beek and Oudendijk, 2003), are defined in the Annex of the WFD by the European Commission which can differ from the national restrictions as those presented in NW4.

Consequently, to meet the 2015 deadline, water authorities in all river basin districts in Europe are obliged to proceed with a coherent program of measures by 2009. A trans-boundary management plan must be developed if a river basin district includes more than one member state. The Netherlands are involved in four trans-boundary management plans for the river basin districts Rhine, Meuse, Scheldt and Ems.

Pending the decision on the future valid effluent requirements has been taken, the values identified in the NW4, the MTR values, remain the key priority together with the European guidelines for Swimming Water Quality and Dangerous Substances.

1.2.2 New quality parameters

A list of relevant and possibly relevant substances for Dutch wastewater treatment plants has been composed by STOWA (2005)[1]. The terms ‘relevant’ or ‘possibly relevant’ refers to the measured presence of the addressed compounds in Dutch wastewater treatment plant effluent after effluent screenings and their definition within the Water Framework Directive (WFD). This list includes substances like nutrients, microorganisms, organic pollutants such as polycyclic aromatic hydrocarbons (PAH’s), pesticides and heavy metals. Endocrine disrupting compounds (EDC’s) like hormones

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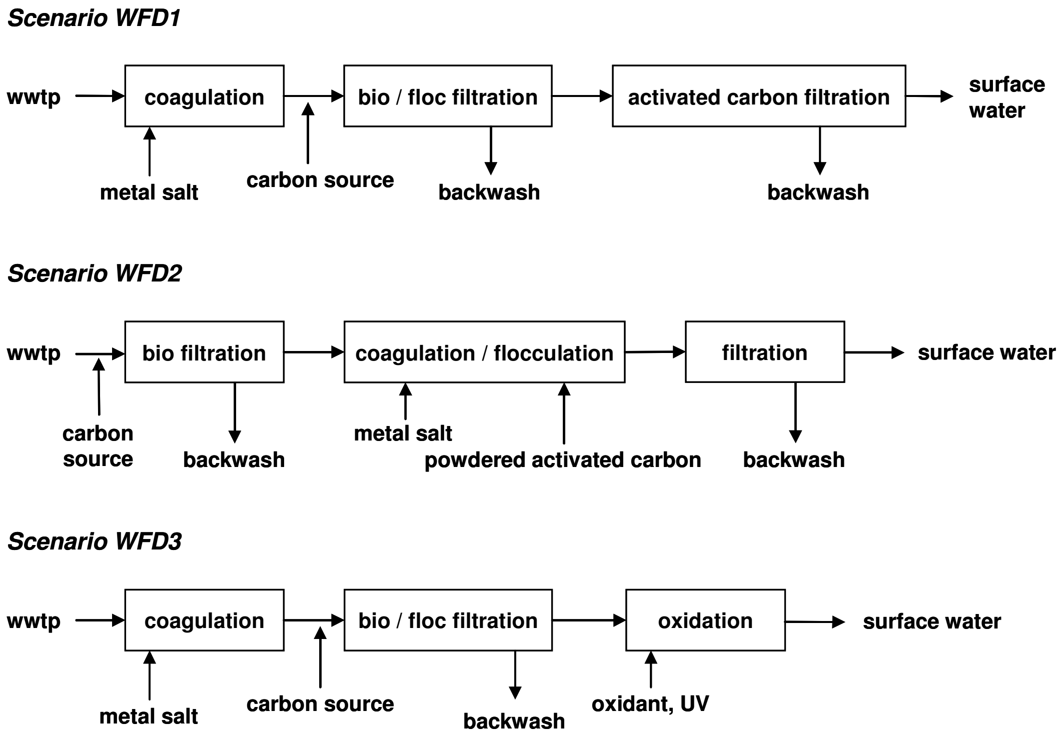
[1] STOWA (Dutch acronym for the Foundation for Applied Water Research) was founded in 1971. The foundation coordinates and commissions research on the behalf of a large number of local water administrations. The bodies which contribute to the STOWA are the 26 Water Boards, the provinces and the Ministry of Transport, Public Works and Water Management.

and medicinal substances are not yet mentioned within the WFD. The detailed list of substances can be found in the Appendix A.

Regarding the complete removal of all WWTP relevant substances, including EDC's, STOWA (2005) selected three treatment scenarios, shown in Figure 1. 1. In these scenarios, the advanced removal of suspended solids nutrients, dissolved organic macromolecules and metals is combined with adsorptive or oxidative techniques for the removal of organic pollutants and pesticides.



Scenario WFD1

Scenario WFD2

Scenario WFD3

Figure 1. 1: Treatment scenarios selected to achieve WFD effluent quality; adopted from STOWA (2005)

Filtration, before or after completed flocculation period, and with or without biological activity, is the core treatment process of the three selected treatment schemes delivering a final effluent quality accordingly to the Water Framework Directive (STOWA, 2005). With respect to the concentrations of nutrients, MTR values will be valid. This means that treatment plants discharging into a sensitive area need to improve the effluent quality to a very low nutrient level, namely 2.2 mgN_{tot}/L and 0.15 mgP_{tot}/L.

This consequently led to a search for viable and economically feasible techniques by research institutions, Water Boards and companies in order to meet those requirements in the Netherlands.

1.3 Application of conventional filtration

1.3.1 Review on conventional filtration

Slow or rapid depth filtration is, besides surface and membrane filtration, one possible unit operation for the removal of suspended material. Rapid media filters have a smaller footprint compared to slow media filters, especially regarding the filtering surface. This advantage strongly favours the applicability of rapid filters for wastewater treatment plants.

Metcalf and Eddy (2003) attributes depth filtration most commonly to the

- supplemental removal of suspended and colloidal solids, including particulate BOD, protozoan cysts and oocysts from wastewater treatment plant effluents,
- pre-treatment before membrane filtration, and
- filtration of chemical precipitated phosphate in a single or two-stage filtration setting.

Rapid granular medium filtration is the most common classification and various technologies can be compared within this filter classification. In terms of design or operation the main distinctions are (Metcalf and Eddy, 2003):

- type or implementation of filter e.g. as open or closed filter,
- operation: continuously or discontinuously with intermittent filtration and backwash cycle,
- filter bed; consisting of one or more layers with the choice of materials (e.g. sand, anthracite) and layer or bed height
- fluid flow: downward or upward.

However, the two most commonly applied filtration systems for the removal of nutrients are deep bed filters operated with upflow filtration in a continuous mode and downflow multimedia gravity filtration in a discontinuous mode (STOWA, 2006).

The international community, especially Switzerland and Germany, has developed their experience with filtration through approximately 200 installations. The United States of America have a few hundred filters for effluent treatment. Filtration is also applied at several wastewater treatment plants in Scandinavia and England. Most installations in England and the United States are designed for suspended solids removal. Germany and Switzerland utilise filters for suspended solids and phosphorus removal. Denitrifying filters applied in Germany, Switzerland and Scandinavia are mainly continuous filters with substrate addition (STOWA, 2006).

The applicability of discontinuous filtration for effluent polishing applied for advanced nutrient removal, combined or as a single process for phosphorus or nitrogen removal, is however relatively new. Investigations for denitrifying discontinuous filtration first took place in the eighties and nineties. Çeçen and Gönenç (1994), Eichinger (1994) and Yun *et al.* (1997), investigated the applicability of discontinuous filters for nitrogen removal for tertiary wastewater treatment. Koch and Siegrist (1997) evaluated rates of denitrification of 1.0 $kgN/(m^3_{bedvolume}/d)$ for denitrification with methanol in full scale deep bed filters during a winter campaign at a location in Switzerland. Operational problems influencing the stability of the process like biofilm loss and methanol breakthroughs were observed. The adjustments of methanol dosing and optimisation of the backwash procedure and frequency could improve the process stability and its performance.

Until now there is no extended experience on the combination of nutrient removal in one discontinuous filtration unit. Jonsson, L *et al.* (1997) described a pilot scale investigation comparing continuous and discontinuous filtration in the Stockholm area aiming for 15 mgN_{tot}/L and 0.3 mgP_{tot}/L in the final effluent. The filter bed consisted of an extended clay layer of 1.2 m above and sand layer of 0.3 m below. Nitrogen removal was between 0 and 4.5 $gN/(m^3_{bedvolume}/d)$ increasing with filter run time (maximal 24 hours). Clogging due to entrapment of gas entrapment, such as nitrogen and carbon dioxide, developing during denitrification has been reported (Boller *et al.*, 1997 and Jonsson *et al.*, 1997). The problem was resolved by an adjusted backwash procedure intermittent with a ‘filter bumping’. Thereby an extra water flush through the filter bed enabled the release of entrapped gas.

The presented experiences suggest that a combination of nutrient removal processes in one filtration system is possible. The reported operational problems on the other hand indicate the necessity of process optimisation to achieve a stable and controllable process.

1.3.2 Status in the Netherlands

The situation around media filtration in the Netherlands changed drastically over the last years. While sand or dual media filtration was not commonly applied in the Netherlands in 2001 (Van der Graaf, 2001), nowadays approximately 16 full scale installations (STOWA, 2006) have been built or are in design. The Dutch experience with continuous filtration is with 12 filters more established than with discontinuous filtration.

Table 1. 5 shows the wastewater treatment plants in the Netherlands where discontinuous media filtration is applied in pilot and full scale (STOWA, 2006). Pilot plant tests at 8 WWTP's in The Netherlands are mentioned by STOWA (2006).

Table 1. 5: Advanced treatment with full scale discontinuous media filtration in the Netherlands (STOWA, 2006)

WWTP	First year of operation	Flow [m^3/h]
Nijmegen	2006	1,400
Steenwijk	2006	600
De Bilt	2008	200
Ootmarsum	2008 [1)]	250

[1)] : recently in operation

According to Huisman (1996) two major differences of discontinuous operation are pointed out towards continuous operation, which may be important when very low phosphorus concentrations of 0.15 mgP_{tot}/L in the final effluent are aimed. These differences are:

- undisturbed filtration within filter run time resulting in a balanced filtration effect and more effective usage of the accumulation capacity,
- extra filtration effect of earlier captured suspended solids ('ripening').

Regarding the traditional purpose of filtration discontinuous operation is more promising versus continuous operation, especially when aiming a very high removal of small particles, flocs and substances which may be related to filterable suspended matter.

The range of applications of media filtration, in pilot or full scale, in operation or construction, indicates the importance of this treatment step for further effluent polishing in the Netherlands.

1.3.3 Possibility for further nutrient removal

Reviewing the applications of media filtration in the Netherlands showed:

- six full scale units (in operation or construction) and five pilot scale installations for advanced phosphorus removal with discontinuous filtration;
- ten full scale installations (in operation or construction) and three pilot scale units for advanced nitrogen removal with continuous filtration.

It shows that recent developments in future legislations encourage the application for advanced nutrient removal. Currently, advanced filtration only aims for the removal of either phosphorus or nitrogen. The combination of both would lead to a compact filtration step resulting in a small footprint, on either the constructional and operational level. The applicability of this is shown elsewhere in Europe but investigations or prefunded experiences with modern Dutch wastewater treatment plant effluent are currently still unknown.

Until now combined nutrient removal has only been tested in continuous filtration units at pilot scale in various locations in the Netherlands. Theoretically also discontinuous filtration seems suitable for

- excellent removal of particles involving particulate phosphorus removal to very low levels,
- the combination of denitrification and possible removal of other substances.

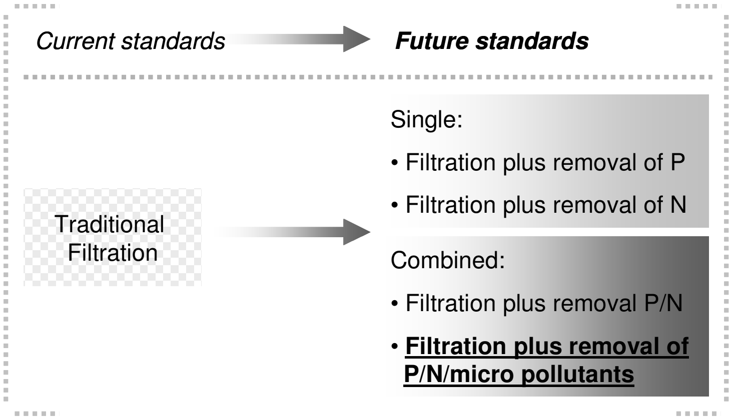


Figure 1. 2: Development of the application of filtration (P; phosphorus; N: Nitrogen)

1.4 Monitoring with particle analysis

1.4.1 Principles

During operation of a rapid discontinuous gravity filter, impurities brought up by the raw water are deposited in the pores of the filter bed. Analyses of total suspended solids (TSS) in the filtrate can indicate filter efficiency. However, the determination of total suspended TSS in the filtrate is very troublesome due to the low suspended solids concentration (often less than 5 mgSS/L). To overcome this issue monitoring of the filtration process generally occurs with online turbidity measurements which may be directly or indirectly related to the suspended solids concentration.

The parameter turbidity is a quantitative measure of remaining suspended and colloidal solids and is generally used to indicate malfunctions within the filtration process.

Turbidity is typically determined using the 90º scattered light principle in compliance with EN ISO 7027 (ISO, 1999). This analysis technique means that this parameter represents an overall level of the analyzed sample without providing any information regarding the mass distributions of particles.

In contrast, particle analysis on the other hand, can measure particles in low turbidity samples and can provide particle mass distributions qualitatively. There are two principles of particle analysis, (1) with light extinction (light blocking) and (2) light scattering (Hargersheimer and Lewis, 1998). The principle utilised in this thesis is light extinction. Light extinction devices measure the change of light intensity caused by a particle, in the micron range, as it passes through a light beam. The light intensity is transformed into a voltage pulse that can be related to particle size. The result is a report of the number of particles according to size through data acquisition, as particle size distribution (PSD) (Lawler, 1997). With the assumption of spherical particles in the sample, a particle volume distribution (PVD) can be calculated based on the PSD. Changes in distribution within the process or removal efficiencies of particle numbers or volumes etc. can be calculated and used for process evaluation.

1.4.2 Current application in wastewater treatment

Nowadays particle analyses in water and wastewater treatment are mainly used for assessment of malfunction in filtration, especially membrane, systems. In the last decade extensive research was conducted to evaluate the additional value of particle analyses for the monitoring of wastewater treatment systems.

Tiehm *et al.* (1999) described particle analyses of primary and secondary effluents at four wastewater treatment plants as a tool to develop a site specific fingerprint under dry weather conditions. The study suggested using the characteristic patterns to improve advanced treatment by deep bed filtration.

Using particle analysis to evaluate particle filtration, and therefore the filtration performance has been investigated. Furthermore, a number of researchers have investigated the relation between particles and other measured parameters, predominately hygienic parameters like bacteria or helminth eggs counts.

For example, Kobler and Boller (1997) compared the results of the particle analysis conducted for different tertiary wastewater filtration systems (deep bed, cloth drum, continuous up-flow and cell filters) to other quality parameters which are related to small particles such as microorganism and heavy metal concentrations. A correlation between the removal of small size particles (< 8 µm) and bacteria counts as well as copper was found. Chavez *et al.* (2004) studied the correlation between the number or volume of particles and the concentration of fecal coliforms, salmonella and helminth eggs in jar tests using coagulation-flocculation for raw and chemically treated wastewater.

Kaminski *et al.* (1997) concluded that particle analysis could help to detect particle detachment or particle breakthrough. The study indicates the particle range 5-10 µm developed in the initialization stage as difficult for filtration, visualises the improvement of particle filtration due to addition of chemical aids, and relates the filtration performance with the linear regression of the power law function and the parameter β.

1.4.3 Opportunities for applications in wastewater treatment

The proper utilisation of particle analysis as a monitoring tool is crucial, especially when dealing with samples of different particle concentration levels within the filtration process. Dilution might be important and the dilution factor has to be chosen with regard to the particle concentration limits defined for the used particle counter. Furthermore, appropriate parameters to be calculated via particle analysis, like particle volume or average size, should be chosen for the evaluation and interpretation of the filtration process.

To evaluate filtration performance the calculation of the total particle volume (cumulative particle volume) removal can be suitable. This cumulative parameter can indicate the efficiency of suspended solids removal.

The correlation with parameters that are generally monitored during filtration, such as turbidity, total suspended solids or possibly even phosphorus, could provide a qualitative indication for the concentration of suspended solids and particulate phosphorus. This relationship could be of use for the optimisation of filtration aiming at very low phosphorus filtrate concentration of 0.15 mgP_{tot}/L.

Furthermore, this tool could be used for the evaluation of coagulation and flocculation processes and could assist in the comparison of the applicability of different filtration systems to secondary wastewater treatment effluent treatment.

1.5 About this dissertation

1.5.1 Research objectives and approach

The following main research objectives were formulated prior to the execution of the filtration tests aiming to show the applicability of the multimedia discontinuous filtration as a process for the combined removal of suspended solids and nutrients (phosphorus and nitrogen):

- Applicability of discontinuous dual media filtration for combined nutrient removal of modern Dutch wastewater treatment plant effluent;
- Extent of simultaneous removal of phosphorus and nitrogen with enhancement of coagulant and carbon source;
- Operational conditions for simultaneous nutrient removal compared to suspended solids filtration and flocculating filtration;
- Floc formation during the whole process with particle analyses: coagulation, flocculation and filtration;
- The additional value of particle analysis within the filtration process;
- Possibility of additional removal of other substances which are listed as priority or priority hazardous substances in the Annex of the Water Framework Directive;
- Comparison with continuous filtration regarding their performance.

The term flocculating filtration is used throughout this thesis and is equal to the term contact filtration which can be found in primary literature. It describes the combination of in-line coagulant dosing, mixing and direct filtration with floc forming in the water volume of the supernatant and within filter bed. This configuration is different to floc filtration where flocs are formed in a separate tank prior to the filtration step.

Between January and June 2004 the first filtration tests were executed on the WWTP Utrecht by installing a pilot scale dual media discontinuous filter. The investigations for advanced nutrient removal with conventional filtration have been conducted in cooperation with Witteveen+Bos Consulting Engineers, and the Water Board Hoogheemraadschap de Stichtse Rijnlanden.

Additional lab scale filtration took place at the WWTP Beverwijk in the period of July to October 2004. These investigations were conducted in cooperation with the ‘Afvalwaterketenbedrijf’, a division of the Water Board Hoogheemraadschap Hollands Noorderkwartier. These tests aimed to confirm and extend the outcomes of the pilot scale investigations executed earlier at the WWTP Utrecht.

Based on the experiences and results at WWTP Utrecht and WWTP Beverwijk further pilot scale investigations were undertaken at the WWTP Horstermeer between March 2005 and April 2007 in cooperation with Witteveen+Bos and Waternet (the municipal water cycle company of Amsterdam).

1.5.2 Outline

In Chapter 2 the filtration process including the mathematical theories behind filtration, the spectrum of particles, filter resistance and backwash is described. Chapter 3 gives a comprehensive summary of the filtration tests including the three investigated research installations. This chapter includes:

- the relevant wastewater treatment plant characteristics and effluent qualities,
- the results of the preliminary jar tests as starting points for the filtration tests, and
- the description and scheduling of the performed lab and pilot scale tests on three research locations.

Chapter 4 presents the sampling and analytical methods used for evaluation of the filtration performance.

The results for the filtration tests applied for combined suspended solids and phosphorus removal are described in Chapter 5. These results serve as a reference for the evaluation of filtration results with denitrifying flocculating filtration described in Chapter 6. Additional outcomes like the profile measurements, results of fractionations of inorganic phosphorus and turbidity, evaluation of effluent improvement by three layer filtration, and the additional removal of heavy metals and other substances finalise the description of the results as the core of this thesis in Chapter 7. The evaluation of the results over all filtration experiments, including the relationships to the previous chapters and the published literature, is provided in Chapter 8.

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2

State of the art of filtration

2.1 Introduction

The particles in secondary effluent present as both colloidal and suspended solids after final sedimentation are predominantly composed of organic macromolecules including humic substances, proteins, viruses, bacteria and algae (Adin, 1998). The residual dissolved constituents are (Metcalf and Eddy, 2003):

- Dissolved organic matter (Total organic carbon, volatile organic compounds)
- Dissolved inorganic matter (Ammonia, nitrate, phosphorus, total dissolved solids)

The separation between dissolved and particulate particle fractions can differ in published literature however the predominant separation diameter is 0.45 µm. This distinction is applied with respect to biological wastewater characterization for the IAWQ activated sludge model (Van Nieuwenhuijzen, 2002; Henze, 1992; STOWA, 1996). Applied advanced particle removal with granular medium filtration is known for separation of remaining solids larger than 0.45 µm.

This chapter gives an overview of wastewater compounds and their particle sizes which may be removable with filtration. The following paragraphs describe the development from traditional filtration theory, valid for discrete particles, towards present and future practical application. The filtration theory reflects the mathematical theories for filtration, filter resistance, backwash, and backwash hydraulics. It is followed by a description of the practical value of small scale column tests. Additional processes like chemical precipitation, coagulation, flocculation and denitrification simultaneously to filtration indicate the value of filtration exceeding just suspended solids removal. Furthermore, the possible simultaneous removal of heavy metals and other substances targeted in future regulations is theoretically evaluated. The characterization and behaviour of these substances in wastewater determine significantly their eventual filterability or possible removal with other chemical or biological processes applied within the filtration process. This chapter closes with a theoretical evaluation of the combined processes.

2.2 Filtration spectrum and particles

Particles are defined based on their size as dissolved, colloidal and suspended matter. Physical separation of certain particles is achieved through filtration and dependent on the chosen pore size of the filtering media. With respect to the filtration spectrum, shown in Figure 2. 1, granular medium filtration aims at the separation of suspended matter from the water phase in the micron size range of >1.0-100 µm.

The specification of wastewater constituents filterable with granular medium filtration is useful to define substances which can be possibly removed. Their occurrence and behaviour in wastewater, as well as the characteristics of the constituents can indicate possible removal mechanisms.

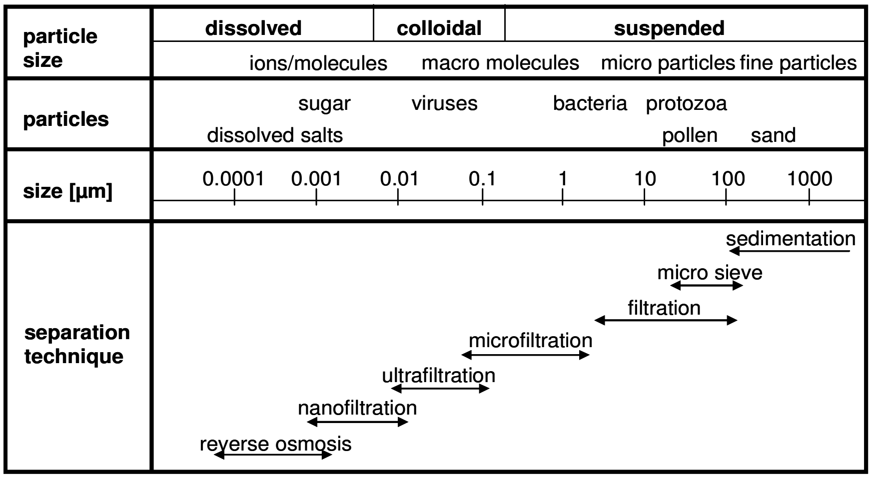

Figure 2. 1: Filtration spectrum adopted from STOWA (2006)

Particles in wastewater effluent are mostly colloidal and negatively charged. Typical floc sizes are in the range of 10 – 70 µm with floc densities of 1.015 – 1.034 g/cm^3 (Andreadakis, 1993). Adin and Elimelech (1989) and Adin et al. (1989) stated that most particles have electrostatic charges in the range of -10 mV to -18 mV, and tend to be stable in water. Analysis of activated sludge effluent particles showed that the surface of effluent particles is dominated by silica, chloride, and calcium. Table 2. 1 summarizes some examples for particles in wastewater.

Table 2. 1: Classification of particles (Metcalf and Eddy, 2003)

Particle size [mm]	Classification	Examples
10^{-1} - 10	Coarse dispersion (visible to naked eye)	Gravel, coarse sand, mineral substances, precipitated and flocculated particles, silt, macroplankton
10^{-4} - 10^{-2}	Fine particulate dispersion (visible under optical microscope)	Mineral substances, precipitated and flocculated particles, silt, bacteria, plankton, and other organisms
10^{-4} - 10^{-5}	Colloidal dispersion (sub microscopic)	Mineral substances, hydrolysed and precipitated products, macromolecules, biopolymers, viruses
< 10^{-6}	Solution	Inorganic simple and complex ions, molecules and polymeric species, polyelectrolytes, organic molecules (also nutrients), undissociated solutes

Constituents are either already in a filterable size (>1 µm) or a pre-treatment step of the wastewater prior to filtration is needed to convert particles into filterable size e.g. by chemical precipitation and coagulation. These chemical processes are enhanced by metal salt addition resulting in destabilisation of particles and change of charge towards negative and neutral charges.

2.3 Traditional filtration theory

2.3.1 Introduction

In general, filtration is a process where water flows through a permeable layer, a membrane, filter paper, a sieve, or a porous medium. In water treatment, granular filtration is a process where water flows through granular material, often sand, where suspended solids like sand, clay, iron and aluminum flocs are retained, substances are biochemically decomposed and pathogenic microorganisms (bacteria, viruses, protozoa) are removed. The suspended solids slowly reduce the pore volume, which results in an increase in hydraulic resistance. The suspended solids are removed by periodical backwash or cleaning of the filter bed. This prevents the resistance exceeding its limit or the breakthrough of suspended solids.

Filtration can also include chemical and biological reactions. This is mainly of importance for the advanced treatment for the removal of dissolved phosphorus (orthophosphate), metals and nitrogen.

The most common application of filtration in surface water and wastewater treatment is rapid media filtration in continuous or discontinuous operation.

Rapid media filtration consists of a bed (range of height 0.6-2.0 m) with a coarse granular medium and supernatant water. The filtration velocities, normally between 5 and 20 $m^3/(m^2.h)$, are controlled by varying the water level above the filter bed (inlet-controlled) or by operating a valve at the outlet pipe (outlet-controlled). Figure 2. 2 shows a cross-section of an open gravity discontinuously operated multimedia filter.

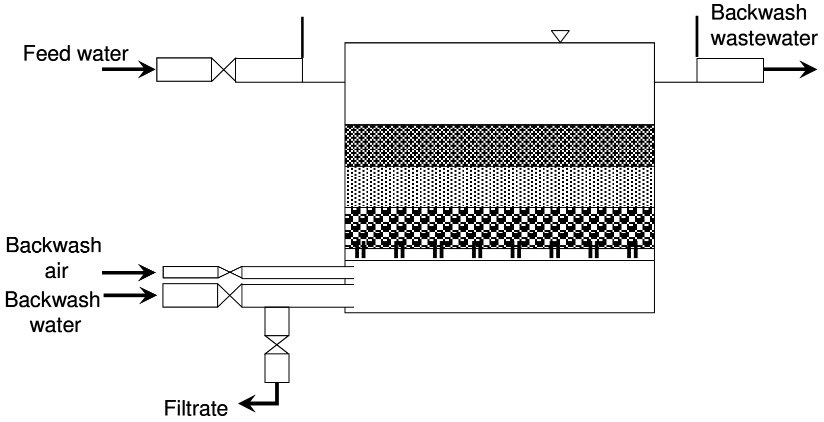

Figure 2. 2: Scheme of discontinuous open gravity media filtration

If, due to clogging, the maximum resistance is reached the filter bed must be cleaned by backwashing. The backwash occurs with water as well as with air backflush. During backwash the filter bed expands, and the accumulated suspended solids are removed. The backwash water is drained through a central trough to a waste receptacle. Depending on the application of filtration the backwash frequency can vary between 6 hours to >24 hours.

2.3.2 Filtration mechanisms

When water flows through the filter bed, suspended and colloidal particles are retained by the filter material. Among many other authors Van der Graaf (1996), Huisman (1996), and Metcalf and Eddy (2003) summarised several filtration mechanisms believed to be responsible for the removal of discrete suspended matter present in the filter feed water.

The mechanisms *inertial impaction* or *sedimentation* (Figure 2. 3) work on large, heavy particles suspended in the flow stream. These particles are heavier than the surrounding fluid. As the fluid changes direction to enter the pore space, the particle continues in a straight line and collides with the media fibers where it is trapped by inertial forces.

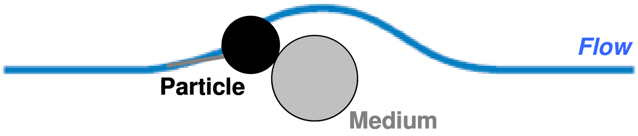

Figure 2. 3: Inertial impaction or sedimentation

With rapid filtration this mechanism does not occur often, because the larger particles (e.g. iron or aluminum flocs) are already removed in the preceding floc removal process (sedimentation or flotation). If smaller filter material is used, the pores are also smaller and the screening process results in the so-called cake filtration. The cake will also retain small particles, and treatment occurs mainly in the top layer of the filter. The disadvantage of cake filtration is the rapid clogging of the filter bed or inefficient use of the accumulation capacity of the filter bed.

During rapid filtration the removal of suspended and colloidal particles usually occurs inside the filter bed. Accumulation of particles therefore occurs over the entire height of the filter bed.

Straining or *sieving* (Figure 2. 4) has been identified as another major removal mechanism. Straining occurs when the size of a discrete particle is larger than the pore size of the filter grains.

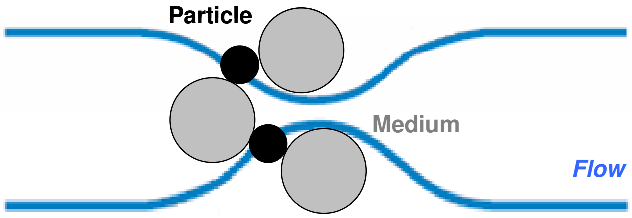

Figure 2. 4: Sieving or straining

In the case of high concentrations of large particles in the feed water of a granular media filter this mechanism can lead to cake layer formation in the upper part of the filter bed. This eventually results in unwanted clogging of the filter bed leaving most of the filter accumulation capacity unused and requiring backwash at an early stage of the filter run.

Even if other mechanisms seem to have a small contribution in the overall particle mass removal compared to the straining effect they should also be considered. The mechanism of *diffusion* or *adhesion*, displayed in Figure 2. 5:, involves the smallest particles and Brownian motion. Small particles are not held in place by the viscous fluid and diffuse within the flow stream. As the particles pass through the flow stream, they collide with the granular medium and are captured.

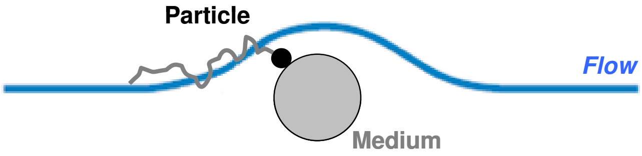

Figure 2. 5: Diffusion or adhesion

Interception (Figure 2. 6:) occurs when a particle which is following a flow streamline appears within a particle radius of a filter grain. The particle touches the grain and is captured and therefore removed from the flow. For a given particle size, there are certain streamlines which will move close enough to a filter grain or fibre so that the particle will be collected. Streamlines further than one particle radius away from a filter grain or fibre will not contribute to the interception mechanism.

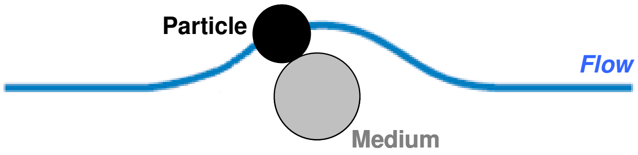

Figure 2. 6: Interception

In addition to the mechanisms discussed above, also processes like chemical/physical adsorption and biological activity can indirectly contribute to particle removal.

When the suspended and colloidal particles collide with the filter grains, attachment can take place. There are two types of forces that result in attraction or repulsion of the particles. The first, Van der Waals forces, ensures that two particles are attracted. The second, electrostatic forces, can have an attracting or repulsing effect depending on the charge of the particles. In general the filter material (e.g. sand) and the suspended and colloidal particles have a negative charge and repulsion takes place (Metcalf and Eddy, 2003).

Attachment of the particles depends on the magnitude of both opposing forces. If the particles are destabilized by the addition of trivalent iron or aluminum salts (through dosing of metals salts as coagulant), attachment will be easier than without destabilization.

Particle breakthrough, the direct passage of particles from the influent, or detachment, the break-off of earlier captured particles due to shear forces, are the reverse of the above mentioned mechanisms.

2.3.3 Filtration theories

Usually, the time frame for preliminary filtration experiments on a small scale before filtration in practice is limited. Due to the varying feed water quality during a year the results of preliminary tests conducted in a short period would also not be representative. The filtration theory makes it possible to quantitatively predict the effects of changes in design parameters, based on the results of a reduced number of filtration experiments.

During the filtration process suspended and colloidal solids accumulate on the grains. Consequently, the concentration of suspended and colloidal solids decreases with the increasing filter bed depth. In addition, the pore volume will be reduced in time due to the accumulation of suspended and colloidal solids, and the effective grain size of the filter material will increase. With a constant filtration rate (superficial velocity), the pore velocity will increase as the pore volume decreases.

The theory of filtration removing discrete macroscopic particles can be described simplified and described with a suspended solids (SS) mass balance of a filtration element (Huisman, 1996, and Lawler and Benjamin, 2008). This balance compares the concentration of SS entering an element equal to the concentration leaving the element plus storage and decomposition and can be formulated to:

$$\frac{\partial c}{\partial t} = -u \cdot \frac{\partial c}{\partial y} - \lambda \cdot u \cdot c \tag{2-1}$$

together with the mass balance:

$$\frac{\partial \sigma}{\partial t} = -v \cdot \frac{\partial c}{\partial y} \tag{2-2}$$

in which:

c = content of suspended and colloidal solids [g/m^3]

y = depth of the filter bed [m]

v = filtration rate [m/s]

p = porosity [%]

u = pore velocity (=v/p) [m/s]

λ = filtration coefficient [m^{-1}]

σ = accumulated solids [g/m^3]

In the stationary situation the following is valid:

$$\frac{\partial c}{\partial t} = 0 \tag{2-3}$$

Therefore the kinetics equation is transformed into:

$$\frac{\partial c}{\partial y} = - \lambda \cdot c \tag{2-4}$$

To solve the system of equations the value of the filtration coefficient λ must be known. However, λ depends on different factors, such as the filtration velocity, viscosity, grain size, quality of the raw water, and the clogging of the bed. After start-up of the filtration process, the filtration coefficient will initially increase because of better attachment characteristics on the pre-filtered material. Due to pore clogging, the pore velocity increases and a decreasing amount of solids will accumulate, expressed by a lower filtration coefficient λ. When the solids are retained in the top layer of the filter bed, lower layers will take over until the filter is saturated and suspended matter breaks through. The clean bed filtration coefficient λ_0 and the relationship between λ and σ have to be determined in practice (through column experiments). More than 50 researchers have found empirical relationships. Often applied relationships are those of Lerk and Maroudas.

Lerk:

$$\lambda_0 = \frac{k_1}{\upsilon \cdot \nu \cdot d^3} \tag{2-5}$$

Maroudas:

$$\lambda = \lambda_0 \cdot (1 - k_2 \frac{\sigma}{\sigma_d \cdot p_0}) \tag{2-6}$$

in which:

d = grain size [m]

p_0 = initial porosity [%]

k_1, k_2 = constants [-]

ν = kinematic viscosity [m^2/s]

The reduction in the pore space σ_v, however, is determined by the volumetric concentration of the accumulated solids σ with the density ρ_d:

$$\frac{\sigma}{\rho_d} = \sigma_v \qquad (2\text{-}7)$$

in which:

ρ_d = density of the flocs [kg/m^3]

σ_v = volume concentration in pores [m^3/m^3]

The values of the coefficients k_1 and k_2 have to be determined experimentally, which leads to the problem of relating filtration theory to filtration practice; these coefficient may be different for filtration tests conducted at different locations with non-equal feed water conditions.

With the boundary conditions $y = 0$, $c = c_o$ and the initial condition $t = 0$, $\sigma_v = 0$, the replacement of k_2 with $1/n$ and

$$\alpha = \frac{v \cdot c_o \cdot \lambda_o}{n \cdot \rho_d \cdot p_o} \qquad (2\text{-}8)$$

the solution for the theory of Maroudas becomes:

- general solution:

$$c = c_o \cdot \frac{e^{\alpha \cdot t}}{e^{\lambda_o \cdot t} + e^{\alpha \cdot t} - 1} \qquad (2\text{-}9)$$

- effluent quality ($y = L$):

$$c_e = c_o \cdot \frac{e^{\alpha \cdot t}}{e^{\lambda_o \cdot t} + e^{\alpha \cdot t} - 1} \qquad (2\text{-}10)$$

- and:

$$\sigma_v = n \cdot p_o \cdot \frac{e^{\alpha \cdot t}}{e^{\lambda_o \cdot t} + e^{\alpha \cdot t} - 1} \qquad (2\text{-}11)$$

in which:

α = coefficient in filtration theory [sec^{-1}]

Lawler and Benjamin (2008) refer to the model of Iwasaki (1935) for the general mathematical description of particle removal in slow sand filters. His empirical observations also consider the improvement of particles capture due to build-up of earlier captured particles (e.g. ripening).

Iwasaki proposed a mathematical description of the filtration coefficient increasing linearly to the specific deposit:

$$\lambda = \lambda_0 + a\,\sigma \tag{2-12}$$

In which λ_0 is the initial filtration coefficient and *a* the ripening coefficient, with the dimension of the length squared. Although Iwasaki's model was expressed in particle number, it can be expressed as particle concentration in mass per volume or particle volume per volume of suspension.

This model can still be considered appropriate for filtration where the filtration run time is determined by particle breakthrough (Lawler and Benjamin, 2008).

2.3.4 Filter bed resistance

During filtration, pore clogging increases and consequently so does the resistance in the filter bed. When the maximum accumulation capacity in the end of a filtration run is reached, the filter bed resistance reaches its maximum as well as the maximum available head loss. The filter needs to be backwashed to avoid a decrease in the filtration velocity. The maximum available head loss is determined as the difference between the supernatant water level and the head of the overflow, minus the clean bed resistance and head loss caused by filter bottoms, pipes and valves. Figure 2. 7 shows the pressure decrease along the filter bed operated with a fixed water head.

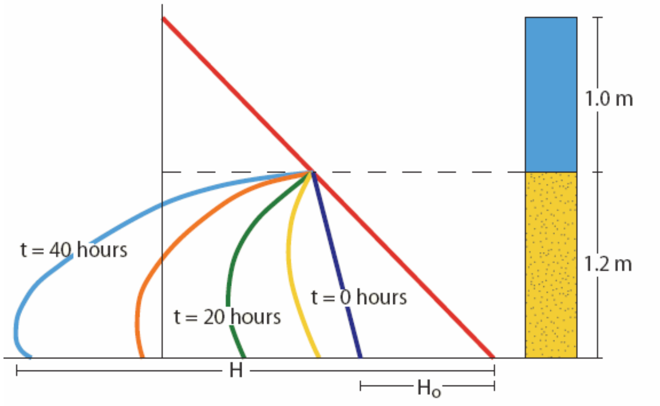

Figure 2. 7: Progress of the filter bed resistance in time (Lindquist diagram)

The clean bed resistance (H_0) derives from the equation of a flow through a pore space and is described with the Carman-Kozeney equation:

$$I_0 = \frac{H_0}{L} = 180 \cdot \frac{\nu}{g} \cdot \frac{(1 - p_0)^2}{p_0^3} \cdot \frac{v}{d_\lambda^2} \tag{2-12}$$

in which:

I_0 = initial resistance gradient [-]

This linear relationship between velocity and resistance is only valid when:

$$Re = \frac{1}{p_0} \cdot \frac{v \cdot v_0}{\nu} < 5 \tag{2-13}$$

When clogging increases in time, the formula for resistance changes to:

$$I = I_0 \cdot \left(\frac{p_0}{p_0 - \sigma_v} \right)^2 \tag{2-14}$$

in which:

I = resistance gradient [-]

If the solids accumulation in the pores σ_v along the height is known, the resistance gradient I can be calculated over the height of the filter bed. Integration of the gradient allows the total resistance over the filter bed to be calculated. It should be noted that it is difficult to examine σ_v accurately because the volume density of the accumulated solids must be known. Therefore, filtration theory may be difficult to relate to actual filtration practice.

However, as presented in Figure 2. 7 the largest resistance is built up in the upper parts of the filtration layers, where theoretically most of the solids accumulate. In time the resistance in the upper layers increases. A pressure drop in the filter bed below atmospheric (negative) pressure must be avoided. When the pressure drops below atmospheric pressure, dissolved gases will come out of solution and the released gas bubbles can clog the filter bed. A negative pressure can be avoided by the operation with a high water head above the filter bed and/or shortening of the filter run time. This is often achieved by an increase of the height of the outflow weir (Huisman, 1996).

2.3.5 *Backwash theory*

After a certain run time the solids accumulation reaches the maximum capacity of the filter bed. The porosity has decreased from p_0 to p, which results in a higher resistance and/or a poor effluent quality. Rapid filters are therefore cleaned by backwashing with filtrate.

During backwashing the water flows in the reversed direction than during filtration. The water flow results in filter bed expansion, scouring of the filter grains and abrasion of the accumulated solids from the filter material, as well as transport of the solids towards the backwash water outlet. The scouring or shear force of the water during backwash is equal to the mass of grains under water:

$$\tau \cdot \pi \cdot d^2 = \frac{\pi}{6} \cdot d^3 \cdot (\rho_f - \rho_w) \rightarrow \tau = \frac{d}{6} \cdot (\rho_f - \rho_w) \tag{2-15}$$

in which:

τ = shear force [kg/m^2]

ρ_f = mass density of the filter material [kg/m^3]

ρ_w = mass density of the water [kg/m^3]

In filtration practice, a combination of water and air is often applied creating a more turbulent backwash which facilitates a better removal of the deposits (or foulants) from the pores. Unfortunately, there is no theoretical approach for backwash with water and air.

2.3.6 *Hydraulics of backwash*

The filter bed needs to be expanded to create abrasion of the filter grains during backwash:

$$E = \frac{L_e - L_0}{L_0} \tag{2-16}$$

in which:

E = bed expansion [-]

L_0 = initial filter bed height [m]

L_e = expanded filter bed height [m]

The applied bed expansion depends on the diameter of the filter material. Usually a bed expansion of 15 to 20% is used for filter material with a diameter of 0.8 mm and 10% for grain diameter of 1.2 mm (Huisman, 1996).

During backwashing of a well-designed filter, the amount of filter material remains constant without loss of filter material. When the initial porosity (p_0), the height of the filter bed during filtration, and the height during backwashing are known, the porosity during expansion can be calculated:

$$(1 - p_0) \cdot L_0 = (1 - p_e) \cdot L \rightarrow p_e = \frac{p_0 + E}{1 + E} \tag{2-17}$$

in which:

p_e = porosity of expanded bed

Because the water flow during backwashing is no longer laminar, but situated in the transition zone between laminar and turbulent ($5<Re<100$), the Carman-Kozeney equation is not valid. An empirical equation for the resistance during backwashing has been derived from experiments (Huisman, 1996):

$$H = 130 \cdot \frac{\nu^{0.8}}{g} \cdot \frac{(1 - p_e)^{1.8}}{p_e^{3}} \cdot \frac{v^{1.2}}{d^{1.8}} \cdot L_e \tag{2-18}$$

This empirical equation is valid when the upward flow rate becomes so high that the bed fluidizes and the grains do not support each other anymore. Fluidization occurs when the resistance is equal to the mass of the filter bed under water:

$$\rho_w \cdot g \cdot H_{max} = (1 - p) \cdot L \cdot (\rho_f - \rho_w) \cdot g \tag{2-19}$$

$$H_{max} = (1 - p) \cdot L \cdot \left(\frac{\rho_f - \rho_w}{\rho_w}\right) \tag{2-20}$$

When sand is used as filter material (ρ_f = 2,600 kg/m^3 and $p=0.4$), the maximum resistance is almost equal to the bed height. The substitution of the values in the equation results in: H>>L.

The backwash rate (v) needs to achieve a certain expansion (E) and a resulting porosity (p_e) can be calculated by substitution of the equations given before:

$$(1 - p) \cdot L \cdot \left(\frac{\rho_f - \rho_w}{\rho_w}\right) = 130 \cdot \frac{\nu^{0.8}}{g} \cdot \frac{(1 - p_e)^{1.8}}{p_e^{3}} \tag{2-21}$$

From this equation the backwash rate can be calculated to (Huisman, 1996):

$$v^{1.2} = \frac{g}{130 \cdot \nu^{0.8}} \cdot \frac{\rho_f - \rho_w}{\rho_w} \cdot \frac{p_e^{3}}{(1 - p_e)^{0.8}} \cdot d^{1.8} \tag{2-22}$$

In drinking water and wastewater treatment the presence of organic pollutants can lead to adhesive binding of the filter grains which may result in the formation of ‘mud-balls’. A strong backwash causing abrasion of the filter grains maybe required in order to remove the foulants sufficiently. In current filtration practice this is often achieved by applying an air scour in a single backwash phase or combined with a water backflush. Following main backwash phases are suggested by Barjenbruch (1997) in accordance with ATV (1995):

1. Air scour: 2-5 min with a rate of v_A = 70-100 m/h
2. Combined air scour and water backflush: 1.5-3.0 min with v_A = 60-100 m/h (air) and v_W = 15-20 m/h (water)
3. Water backflush: 3-8 min with $v_W \leq$ 100 m/h

The first backwash phase breaks the filter bed while the second phase leads to mixing of the filtration material under high turbulence. The third phase aims at the removal of enclosed air bubbles and foulants with final stratification of the filtration material.

Table 2. 2 shows the backwash rates in practice for different filter materials. The filtration rates can vary with variable bed expansions and water temperatures.

Table 2. 2: Backwash rates for different filter materials (STOWA, 2006)

Type of media	Grain size [mm]	Backwash rate for sufficient bed expansion [m/h]*
Anthracite	d < 1	40
	d = 1.5 – 2.5	55
	d > 2.5	90
Hydro anthracite	d = 1.5 - 4.0	50
Quartz sand	d = 0.7 – 1.25	55
	d = 1.0 – 1.5	75
	d = 1.5 – 2.0	90
	d > 2.0	130
Basalt	d = 1.0	110

Type of media	Grain size [mm]	Backwash rate for sufficient bed expansion [m/h]*
Garnet sand	d = 0.2 – 0.6	90

* based on filtration practice and is depended on the installation

2.3.7 Filtration theory and the new strategy

The theoretical approaches described before only aim at the filtration for suspended solids removal. Advanced filtration applied for simultaneous nutrient removal from WWTP effluent implies a less value of these theories. This new application involves not only physical processes like filtration of discrete suspended solids, but also chemical processes like coagulation/flocculation and biological denitrification. Mutual effects of these processes during multi purpose filtration are likely to occur.

The variation of feed water quality may also require further process optimisation beyond the known standard strategies for backwash and modelling of the filtration process. Furthermore, the monitoring of quality parameters used for the evaluation and optimisation of the process efficiency and final effluent quality are hardly known yet. New quality parameters have to be developed considering also the wide range of particles and their properties present in WWTP effluent.

Consequently the current theories and mathematical modelling are not safe or accurate enough to apply their results for real scale filter design (Boller, 2009) or when filtration aims at multi purposes. They may be only used to verify results obtained during practical tests without replacing them. Standard filtration strategy, like development of filter bed resistance, backwash procedure and frequency, can only be used as starting point for further investigations.

2.4 Filtration practice, column tests

Experiences of other treatment plants which deal with similar water can help to determine the dimensions of a filter. Further information can be obtained with filtration tests on smaller scale, such as laboratory or pilot scale tests. In the test setup, the optimal combination of the following design parameters could be investigated:

- filter bed depth,
- grain diameter of filter material,
- filtration velocity,
- height of the supernatant water (head loss).

The optimal combination leads to a filter that is cost efficient and satisfying the required effluent quality resulting in a reasonable filter run time. Additionally, during the filter run the suspended solids should be distributed over the filter bed height to avoid cake filtration. The filter surface area should also be as small as possible to reduce investment costs. Consequently, the filtration velocity must be high. The higher the filtration velocity, the sooner the effluent quality will deteriorate during the filter run. This can be compensated by increasing the filter bed height, or by choosing filter material with a smaller grain size. A higher filter bed, however, means a higher filter construction and therefore, higher construction costs. Filter material with a smaller grain size will clog faster, shorter filter runs will occur and operational costs will be increased.

In general it is assumed that the effluent quality has to remain under a required discharge water quality. The filter run time during which the effluent quality satisfies the discharge limit is called T_q. Normally filters are backwashed after run time T_r, when a predetermined maximum resistance is reached. This is to prevent the water quality from decreasing before the maximum resistance is reached. The filter design should fulfil the condition: $T_r<T_q$. The above mentioned design parameters and operational values of filtration performance, the time and cumulative volume (throughput), directly determine the values of both T_q and T_r. Table 2. 3 summarises the effects of increasing independent variables on the two backwash criteria.

Table 2. 3 Summary of effects of independent variables on length of filter run (Lawler and Benjamin, 2008)

Independent variable	Head loss		Effluent Quality	
	Time	Cumulative Volume	Time	Cumulative volume
Depth ↑	↓	↓	↑	↑
Media Size ↑	↑	↑	↓	↓
Velocity ↑	↓	↔	↓↓	↓
Influent Conc. ↑	↓	↓	↓	↓

A down arrow in the head loss or effluent quality columns indicates the decrease in the time (or cumulative volume) with an increase of the independent variable when a criterion for backwash, either excessive head loss or poor effluent quality, is reached (Lawler and Benjamin, 2008).

In practice some restrictions are given to this optimization process. For example, safety margins are introduced to maintain the quality of the final effluent. One example of filtration practice is the adjustment of backwash frequency in drinking water treatment based on the total suspended solids load of 4 $\text{kgSS/m}^3_{\text{bedvolume}}$ (Huisman, 1996). Depended on the filter installation this can be far below the optimal capacity but on the other hand always deliver a filtrate quality in compliance with the requirements.

However, most of the time filter installations are operated below their capacity and often far from the optimal situation.

2.5 Flocculating filtration

Besides the filtration of suspended solids, dissolved phosphate can be precipitated by the inline dosing of metal salts and removed in following filtration (flocculating filtration). Alum, hydrated aluminium sulphate, ferric chloride and ferrous sulphate are widely used to precipitate phosphates as metal phosphates. The basic reactions for the metals salts are:

$$Al^{3+} + H_nPO_4^{3-n} \leftrightarrow AlPO_4 + nH^+$$

$$Fe^{3+} + H_nPO_4^{3-n} \leftrightarrow FePO_4 + nH^+$$

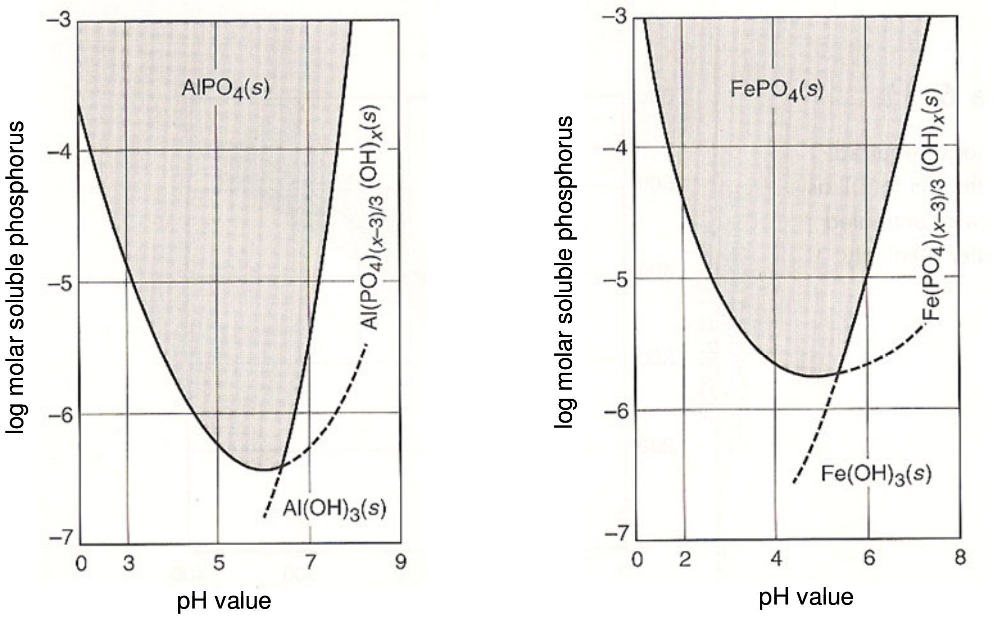

Figure 2. 8: Concentrations of aluminium and ferric phosphate in equilibrium with soluble phosphate (Metcalf and Eddy, 2003)

Similar to aluminium, ferric ions form ferric phosphate precipitates. They react slowly with the natural alkalinity.

This reaction is relatively simple, but many competing reactions have to be considered regarding their associated equilibrium constants and the effects of alkalinity, pH, trace elements in wastewater. The dosage rate required is a function of the phosphorus removal required; the precipitation efficiency decreases with decreasing concentration of phosphorus. The dosing of metal salts is proportional to the dissolved phosphate (orthophosphate) in the filter feed water and expressed in this thesis as MePO (metal-orthophosphate) ratio in mol/mol. Typical MePO ratios vary between 0.5 and 5 mol/mol (STOWA, 2006).

Besides precipitation of phosphates the dosing of trivalent metal salts also initiates coagulation, described as destabilisation of colloidal matter by decreasing the electrostatic repulsion between particles. This destabilisation takes less than a couple of seconds and therefore, mixing has to be effective to emphasise the reaction. The repulsing forces are neutralised and the attracting forces between colloids (Van der Waals forces) are enhanced. The following flocculation process results in the formation of larger compounds. The flocculation of particles in a liquid depends on collisions between particles, due by their relative motion. This relative motion may be caused by Brownian movement, by fluid movement giving rise to velocity gradients, or by particle motion due to an external force (e.g. gravity).

The most important mechanisms of coagulation for (waste-)water treatment are (1) adsorptive coagulation through neutralisation and (2) precipitation coagulation, also know as sweep or entrapment coagulation. Figure 2. 9: shows the coagulation mechanisms at various pH values and quantity of dosed coagulant containing iron.

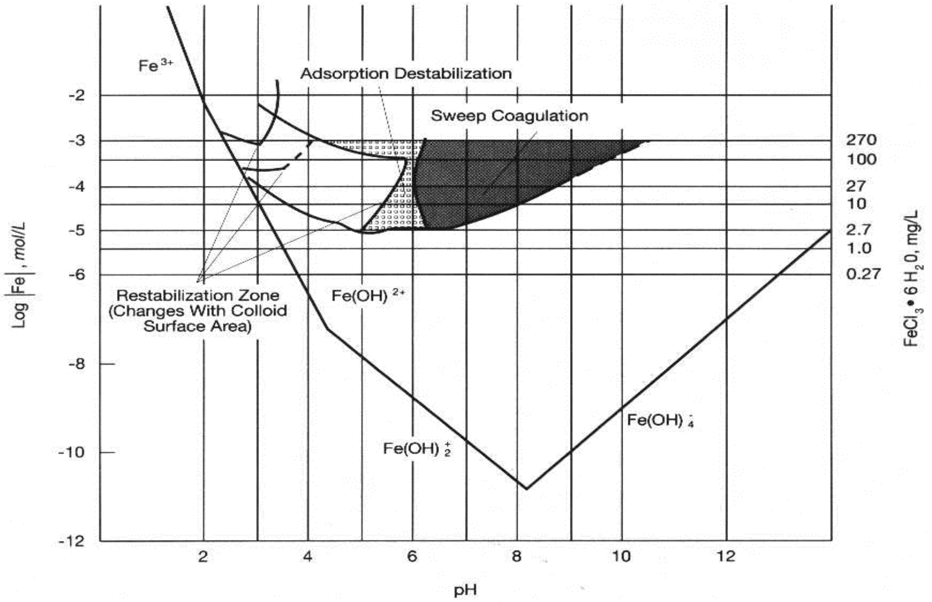

Figure 2. 9: Hydrolysed products of ferric chloride (Metcalf and Eddy, 2003)

An example for adsorptive coagulation is the adsorption of particles or other ions by positive charged ions. This can occur through hydrolysed products of the coagulant, e.g. $Al(OH)^{2+}$ and $Al(OH)_2^+$ (Figure 2. 10).

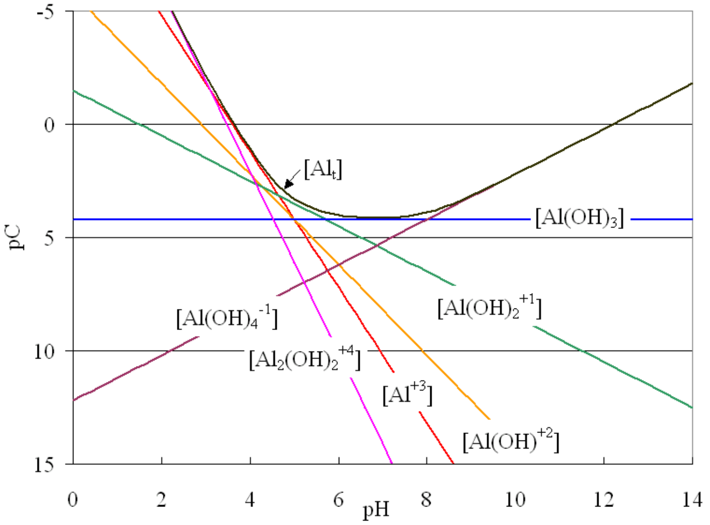

Figure 2. 10: Solubility of aluminium (adopted from Metcalf and Eddy, 2003)

These hydrolysed products are mainly present in the pH range of 5-7. The most important issues for adsorptive coagulation are:

- effective initial mixing is necessary (recommended is a mixing velocity gradient G ± 300 1/s),
- high efficiency at high concentrations of organic matter,
- chance of restabilisation caused by overdosing of metal,
- reaction within one second.

Precipitation coagulation describes the mechanism of enclosure of colloids in hydroxide flocs with the following characteristics:

- requires a higher quantity of coagulant than adsorptive coagulation,
- the hydroxide floc is uncharged,
- main coagulation mechanisms in water with low suspended solids content (<10 mg/l),
- no possible restabilisation.

It shows that the pH and the suspended solids concentration determine the efficiency of both coagulation mechanisms. Optimal removal of suspended solids and phosphorus from wastewater treatment plant effluent (in the Netherlands: pH = 6.5-7.5, SS ~ 10 mg/L) require an optimal coagulant dosage and mixing to ensure the efficiency of coagulation.

2.6 Denitrifying filtration

Wastewater treatment plant effluent still contains several microorganisms including heterotrophic organisms are able to denitrify and remove nitrate nitrogen. Enhanced conditions, high concentration of biodegradable organic matter and low concentration of dissolved oxygen, can result in the formation of a biofilm coating the filter material.

Biological denitrification occurs when dissolved oxygen (DO), the most favourable electron acceptor is depleted or present in limited concentrations. Heterotrophic bacteria then use nitrate and nitrite for degradation of rapidly (dissolved) biodegradable organic matter (bsCOD), in anoxic conditions.

The nitrate reduction can be inhibited by DO. In activated sludge flocs and biofilms, denitrification can proceed in the presence of low DO concentration. Skerman and MacRae (1957) and Terai and Mori (1975) reported inhibition for a *Pseudomonas* culture at a dissolved oxygen concentration > 0.2 mg/L. Similar results for activated-sludge treated domestic wastewater have been presented by Dawson and Murphy (1972). Nelson and Knowles (1978) reported that denitrification ceased in a highly dispersed growth at a DO concentration of 0.13 mg/L.

However, in the biofilm the DO concentration will be reduced rapidly by biological activity. The DO concentration will be almost zero when the biofilm is thick enough.

Most of bsCOD like end-products, such as volatile fatty acids, present in the influent wastewater have already been consumed during conventional activated sludge treatment. To initiate and stimulate biological growth in a subsequent biological treatment process an external carbon source with bsCOD character should be added to the wastewater. During the filtration tests described in this thesis methanol is used for this purpose as the electron donor. Due to the presence of bsCOD in dissolved form it can be easily assimilated by the biomass. The heterotrophic denitrification reaction for methanol is shown below:

$$5CH_3OH + 6NO_3^- \rightarrow 3N_2 + 5CO_2 + 7H_2O + 6OH^-$$

Commonly methanol is dosed in a ratio CH_3OH/NO_3-N of 3 to 4 g/g, used as a typical design criterion for down flow denitrification filters. Additionally the oxygen concentration has to be considered in the necessary amount of methanol to be dosed encountering its side-reaction. The reaction of methanol with oxygen is as follows:

$$2CH_3OH + 3O_2 \rightarrow 2CO_2 + 4\ H_2O$$

The theoretically calculated quantity of necessary additional carbon source expressed as COD_{CT} in mg/L, considering concentrations of nitrate, nitrite and oxygen can be calculated using the following equation:

$$COD_{CT} = 2.86 * NO_3\text{-}N\ [mg/L] + 1.71 * NO_2\text{-}N\ [mg/L] + O_2\ [mg/L]$$

It should be noted, that the concentration for nitrate, nitrite and oxygen are variable and their variations should be considered for a continuous dosing of carbon source.

Biological oxidation of certain organic substances occurs when using nitrate and nitrite as electron acceptor instead of oxygen. The nitrate reduction reactions include reduction steps from nitrate to nitrite, to nitric oxide, to nitrous oxide, and to nitrogen gas:

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

$$2\ NO_3^- + 2H^+ \rightarrow N_2\uparrow + H_2O + 5O$$

2.7 Co-removal of other substances

2.7.1 Heavy metals

Future regulations of the European Commission do not focus solely on nutrients. Several substances, described in Chapter 1, will be the focus of attention. Depending on the chemical properties of other substances and their appearance in wastewater a partial indirect or direct removal with the processes applied before and during the filtration process is assumed.

For example, heavy metals appear in wastewater through various diffuse sources and small industrial discharges and are present in dissolved and particulate form in WWTP influent. A major part will already be removed due to mechanical and biological treatment at the WWTP. However their removal is often incomplete. A possible removal technique applied for residual dissolved metals is the precipitation with hydroxide or sulphide. The particles can then be removed with following filtration. These precipitation processes are pH dependent with optimal pH ranges between 8 and 11 (Tünay and Kabdaşli, 1994; Metcalf and Eddy, 2003). Large amounts of acids or bases might be required to achieve appropriate pH conditions. On the other hand co-precipitation with dosed metals salts during flocculating filtration without any necessary pH corrections is also considered to contribute to heavy metal removal.

Furthermore, heavy metals are often associated to possibly filterable organic material which appears as weak acidic compounds. Removal mechanisms are biosorption (adsorption to microorganisms or biomass) and formation of complexes with dissolved organic material. Wang *et al.* (1998) concluded that heavy metals rather form complexes with sludge flocs at neutral or low pH, like in WWTP effluent, than with dissolved organic carbon (DOC). Also the removal of certain metals due to biological up-take through accumulating microorganisms, reported by e.g. Wang *et al.* (2003) and Hussein *et al.* (2005), should be included to the possible removal mechanisms.

2.7.2 Other substances

The chemical characteristics of substances like e.g. polycyclic aromatic hydrocarbons (PAH's), pesticides and medicinal substances also indicate possible direct or indirect removal. As described previously for heavy metals, indirect removal can occur through biosorption to filterable material or biodegradation of the denitrifying biomass. A high solubility of the substances is required for the biological transformation. Generally, aqueous solubility decreases with increasing molecular weight of the substance. For example, several PAH's consist of a low molecular weight and are therefore considered to be biodegradable. The rate of the biodegradation of PAH's appears to be limited by their dissolution rates and aqueous solubility (Brubaker *et al.*, 1992).

Biosorption onto sludge on the other hand is investigated by Ternes and Joss (2006) for several antibiotics, hormones, drugs and fragrances. Removal in media filtration after biological treatment is reported.

A partial removal of natural polymers during flocculating filtration implies the possible removal of PAH's also through sorption to natural polymers like polysaccharides, starch and the derivative cyclodextrin, typically present in wastewater (Crini, 2005).

2.8 Combined processes

The goal of the research presented in this thesis is the combination of suspended solids, phosphorus and nitrate removal and, if possible, the additional removal of other substances. The different processes like denitrification, adsorption, precipitation, coagulation and flocculation combined with filtration lead to a rather complex treatment step. The combination of processes might enhance one or the other and can additionally lead to removal of other substances beyond nutrients and suspended solids.

The filtration mechanisms pointed out in paragraph 2.3 as being responsible for particle removal need to be extended in regard to the additional processes being involved during filtration. Flocculation in the filter bed, ripening and biological activity due to denitrification enhance the before mentioned mechanisms and should therefore not be neglected.

Many researchers investigated and developed filtration models in order to understand the filtration behaviour and maybe even predict the filtration results. Lawler and Benjamin (2008) reviewed mainly three models used for the description of the geometry of the media grains, the spaces between them, and the relationship between the differential size and the whole filter bed. These geometric models consider the three mechanisms of long-range transport of particles to a media grain interception, sedimentation and Brownian motion assuming deep bed filtration in drinking water treatment after preceding treatment including alum flocculation and sedimentation. The model comparison showed significant differences in the removal of small particles which are targeted in the destabilisation process previous to filtration. Poor particle removal is expected when the destabilisation is poor. Therefore, the filtration results of particles clearly depend on proper surface chemistry and less on the straining mechanism.

In the existing models particles in the filter influent are assumed to be discrete with unchanged distribution during the filtration process. This may not hold true especially for flocculating filtration using various coagulants. Van der Graaf (2001) described the shrinking of flocs after dosing of ferric chloride which is attributed to a possible dewatering effect on organic material. The following flocculation process and a shift of the particle distribution towards larger particle sizes is likely to happen. In the filter bed the distributions can change again due to adsorption, precipitation, coagulation and flocculation. Less stable flocs can possibly break-off due to shear forces within the filter bed. However, the particle distribution in the filter feed water for flocculating filtration undergoes several changes and a prediction of the particle removal by modelling with idealised particles is very difficult. Experimental tests are still unavoidable.

This thesis focuses therefore on the filtration tests during multi-purpose filtration and the mutual relation of measured parameters deriving from the adjustments of the operational modes. The main goal of the presented tests is the optimal performance of the main removal processes resulting in a high suspended solids, phosphorus and nitrogen removal. This consequently results in the investigation of the most optimal filter operation.

Filter performance for solids removal is described by two dependent variables, the head loss and filtrate quality. These variables are again dependent from the filter bed depth, the media size, the filtration velocity and influent concentrations. And these variables can differ from pure suspended solids removal when additional removal of phosphorus and nitrogen is applied.

The dosing of metal salts increases the load of suspended solids for filtration. Van der Graaf *et al.* (2001) described an increase of suspended solids with 1.9 mgSS/L and 3.0 mgSS/L for a dosing of 1mgFe/L or 1mgAl/L, respectively. Therefore, coagulant dosing affects the filter operation, especially the length of the filter run time significantly. Biofilm growth due to denitrification can lead to narrowing of the pore space and can affect the filter accumulation capacity. On the other hand the biofilm can introduce other mechanisms or enhance the removal of particles and other substances. Nitrogen and carbon dioxide gas developing during denitrification can result in clogging of the filter bed when not released. Furthermore, the slimy biofilm coating the filter material enhances 'mud-ball' formation. The biofilm coated filter grains and unwanted 'mud-ball' might require a different backwash regime due to increased bed expansion.

However, a requirement set for the filtration tests was a negligible disturbing influence of denitrification on the suspended solids and phosphorus removal. The utilisation of particle analyses and measurements of particle distributions are meant to support results gained with other measurements and deliver a better insight view of particle behaviour and removal.

Even if the filtration mechanisms and their dynamics are not yet fully understood, the combination with biological processes, such as denitrification, in a single reactor may be a practical alternative compared to post-treatment with single purpose reactors. Optimal removal of suspended solids and phosphorus should not be affected when applying simultaneous denitrification.

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3

Description of filtration tests

3.1 Introduction

Filtration tests were conducted at three wastewater treatment plants (WWTP's) in The Netherlands with the aim of applying conventional filtration for simultaneous removal of suspended solids, phosphorus and nitrogen. The feasibility of nutrient removal down to MTR levels with dual media filtration was the focus of the investigations.

Between January and June 2004 the first filtration tests were conducted at the WWTP Utrecht using a pilot scale dual media discontinuous filter installation. Initial results were obtained for simultaneous nutrient removal with single flocculating filtration. Additionally, two activated carbon filters were applied in a second investigation partly in series or in parallel with dual media filtration. The investigations for advanced nutrient removal with conventional filtration were conducted in cooperation with Witteveen+Bos Consulting Engineers, and the Water Board Hoogheemraadschap de Stichtse Rijnlanden. In addition to the process monitoring with conventional analyses, several particle analyses were conducted to investigate its additional value.

These investigations continued with laboratory scale filtration tests using two dual media filters at the WWTP Beverwijk. These investigations, conducted in cooperation with the Water Board Hoogheemraadschap Hollands Noorderkwartier, and aimed at confirming and expanding the outcomes of the pilot scale investigations performed earlier on the WWTP Utrecht. These investigations also aimed to utilize particle analyses as monitoring tool for filtration performance in relation to other parameters. The filtration tests were performed with single flocculating filtration as well as filtration for simultaneous nutrient removal.

Based on the experiences and results at WWTP Utrecht and WWTP Beverwijk the pilot investigations were conducted at the WWTP Horstermeer, supported by Witteveen+Bos and Waternet, the municipal water chain company of Amsterdam. The focus for dual media filtration at WWTP Horstermeer was to achieve the optimum simultaneous nutrient removal. Also continuous and activated carbon filtration were investigated in order to asses their performance on particle and simultaneous nutrient removal.

This chapter describes the design and operation of the three WWTP's and the produced effluent quality. Next, the results of preliminary investigations with jar tests are evaluated. Based on the quality of the effluents, jar tests were performed in order to indicate the appropriate dose rate and type of coagulant in a preliminarily stage. The jar and filtration tests were either conducted with the dosing of iron(III)chloride (or ferric chloride, $FeCl_3$) or poly aluminium chloride products like PACl, PAX 11 or PAX 14. The product specifications for the relevant coagulants can be found in the Appendix B.2 preceding the filtration tests. This chapter concludes with the setup of the pilot and laboratory scale tests. All these items are presented for each research location in the following order: WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer.

The detailed operational parameters for the lab and pilot scale locations are presented in Appendix B.1.

3.2 Filtration tests at WWTP Utrecht

3.2.1 Design and operation of the WWTP

The WWTP Utrecht, constructed in 1991, is a municipal wastewater treatment plant with a two-stage activated sludge system. The design capacity is 400,000 PE's (population equivalents) and it has a hydraulic capacity of 15,000 m^3/h (HDSR, 2003).

The first stage of the treatment plant is high loaded and consists of two parallel aeration tanks which are followed by nine intermediate sedimentation tanks. The effluent from the first stage is supplied to the three parallel treatment trains of the low loaded second stage. Every treatment train consists of one aeration tank divided into four compartments and a final sedimentation tank (STOWA, 1997). For simultaneous phosphorus removal ferric chloride is dosed in the biological treatment step.

The scheme of the treatment plant is shown in Figure 3. 1.

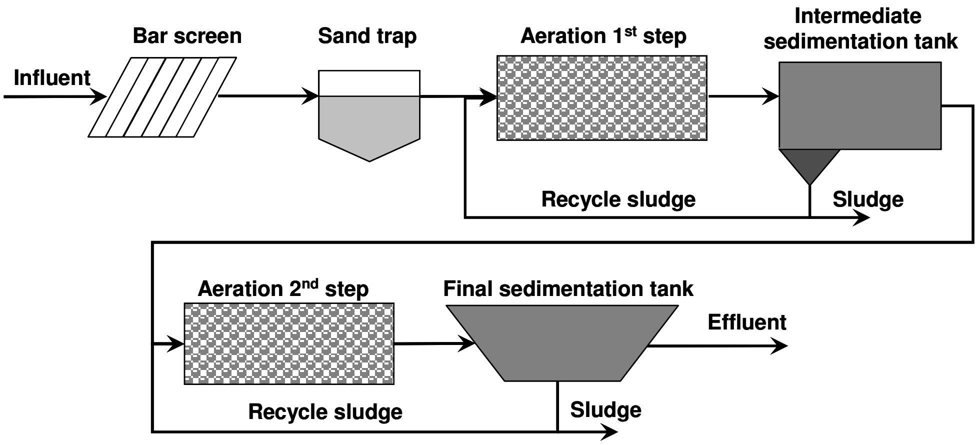

Figure 3. 1: Treatment scheme of the WWTP Utrecht

The final effluent of the WWTP Utrecht is discharged into the river Vecht, part of the basin district of the river Rhine.

In the future increasingly stringent discharge criteria will be required predominantly as a result of the Water Framework Directive. The neighbouring Water Board Amstel, Gooi and Vecht (AGV) requested investigations for reaching the discharge limits for phosphorus and nitrogen on MTR level before the deadline in 2010. As such the pilot investigations were started in January 2004 (HDSR, 2003) at the WWTP Utrecht.

3.2.2 *Effluent quality*

The average effluent quality of the WWTP Utrecht in 2003 is shown in Table 3. 1. These concentrations served as a starting point for pilot scale tests for single phosphorus and simultaneous nutrient removal (0.3 – 2.6 mg/L of total phosphorus and 3.6 – 20 mg/L of total nitrogen).

Table 3. 1: Effluent quality of the WWTP Utrecht in 2003 (Oene, 2004)

Parameter	Unit	Concentrations		
		Min	Max	Average
COD	mg O_2/L	2.2	57.0	32.0
BOD	mg O_2/L	0.5	6.4	3.0
N_{Kj}	mg/L	1.3	14.0	4.3
NO_x	mg/L	2.2	14.0	6.0
N_{tot}	mg/L	3.6	20.0	10.2
P_{tot}	mg/L	0.3	2.6	1.0
SS	mg/L	1.0	21.0	6.0
Cl^-	mg/L	65.0	150.0	92.0

Parameter	Unit	Concentrations		
		Min	Max	Average
SO_4^{2-}	mg/L	8.2	99.0	69.0

3.2.3 Preliminary investigations

3.2.3.1 Introduction

Jar tests were conducted to determine proper coagulant type and starting dose concentration. The jar tests apparatus consists of six 2 litre jars with stirrers. The samples of the WWTP effluent were conditioned with coagulant simultaneously in all six jars. The stirrers can be adjusted for equal stirring velocities for all six beakers. The mixing conditions for the performed tests were chosen in relation to the process conditions for the investigated pilot installation. Following the mixing time for coagulation (20 sec) and flocculation phase (2 min) a standard sedimentation time of 30 min was applied. During these three stages floc formation is observed visually. Depending on the size and form, flocs either settle or stay in suspension. Flocs with the ability to settle are considered easily filterable with a granular medium filter. However flocs in suspension can also be filtered with media filtration to a certain extent. Therefore, the evaluation of jar tests is not strictly representative for the filtration results of suspended matter.

Finally, the supernatant was also siphoned and analysed for turbidity, phosphorus (phosphate and total phosphorus for unfiltered samples and samples filtered with a 0.45 µm cellulose acetate membrane filter) and residual metal-concentration (Al^{3+} or Fe^{3+}). The concentrations of orthophosphate ($PO_4\text{-}P_{<0.45\mu m}$) compared to the unfiltered (total) phosphate analyses ($PO_4\text{-}P_{unfilt.}$) can be used to indicate the efficiency of precipitation. Accordingly the orthophosphate concentration is aimed at levels as low as possible. Furthermore care was taken to ensure that the dosed metal salt appears predominantly in particulate form in order to avoid an overdosing and ensures proper reaction.

Different types of coagulants are available. Ferric chloride ($FeCl_3$), aluminium sulphate (alum) and poly aluminium chloride (PAX or PACl), are commonly used and applied in the presented investigations. The advantages of poly aluminium chloride are a smaller decrease in pH following dosing, and a higher shear stability of flocs compared to aluminium sulphate. In contrast, ferric chloride has been indicated as coagulant that leads to the formation of weak and fragile flocs, which can break off due to shear forces occurring in filtration processes (Oschwald, 2000). Furthermore, poly aluminium chloride has a lower sensitivity to low temperatures and it can be utilised in a broader pH range, pH of 6 – 9, in lower concentrations (Metcalf and Eddy, 2003). The optimal pH range for ferric chloride is between 7 and 8.

3.2.3.2 Results of jar tests with dosing of ferric chloride

The samples of WWTP effluent for this test were collected 41 hours before conducting the jar tests. During the jar tests the pH of the samples averaged 7.01, the temperature 19.7 ºC, the turbidity 3.5 NTU and

- Phosphate (PO_4-$P_{unfilt.}$) ~ 0.7 mg/L,
- Orthophosphate (PO_4-$P_{< 0.45 \mu m}$) ~ 0.51 mg/L,
- Total phosphorus ($P_{tot\ unfilt.}$) ~ 0.85 mg/L, and
- Iron ($Fe_{unfilt.}$) ~ 0.24 mgFe/L.

For coagulation, flocculation and precipitation ferric chloride was used.

The dosing of ferric chloride was varied for the jar tests between 0.5 and 2.0 mgFe/l resulting in ratios of molFe per molPO_4-$P_{<0.45 \mu m}$ of 0.5 – 2.2 (Table 3. 2). These ratios were chosen between the theoretical minimal molar ratio of metal to phosphorus 1:1 and the most common applied ratio of 3:1mol/mol (Metcalf and Eddy, 2003). The results of the jar tests without dosing can be used as a reference to indicate removal potential without chemical enhancement; see Table 3. 2 for the results.

Table 3. 2: Results of the jar test with dosing of ferric chloride for preliminary investigation of filtration tests at WWPT Utrecht

Jar no		1	2	3	4	5	6	7
Parameter	unit							
Dosing Fe	mgFe/L	0.0	0.5	0.7	1.0	1.3	1.5	2.0
Turbidity	NTU	2.7	2.7	2.5	2.6	2.6	3.1	3.5
$Fe_{unfilt.}$	mgFe/L	0.18	0.74	0.87	1.23	1.33	1.51	1.35
$Fe_{< 0.45 \mu m}$	mgFe/L	0.00	0.06	0.08	0.08	0.07	0.07	0.08
PO_4-$P_{unfilt.}$	mgPO_4-P/L	0.63	0.62	0.62	0.62	0.56	0.55	0.48
PO_4-$P_{< 0.45 \mu m}$	mgPO_4-P/L	0.59	0.50	0.43	0.34	0.27	0.24	0.20
$P_{tot\ unfilt.}$	mgPO_4-P/L	0.80	0.80	0.80	0.80	0.80	0.70	0.50
$P_{tot\ <0.45 \mu m}$	mgPO_4-P/L	0.70	0.60	0.60	0.40	0.40	0.30	0.25

The floc size visibly increased with increasing amount of dosed coagulant. For analytical evaluation of the coagulation process the metal and phosphate content were measured on both, the raw an the filtered sample (0.45 µm). The phosphorus concentration is decreasing with increasing concentration of dosed metal salt.

The highest removal of phosphorus corresponded to a dosing rate of 2.0 mg Fe^{3+}/L which resulted in 0.25 mgPO_4-P/L, in the 0.45 µm filtered sample of Jar no 7. However with this dosage the filtered sample still showed a rather high concentration of orthophosphate and total phosphorus, indicating insufficient precipitation.

The high values for particulate phosphorus indicate the poor settling properties of the flocs. This indicates that iron is effective in precipitating phosphate but does not result in proper settling of the flocs. The dosing concentrations for further filtration tests should be in the MePO ratio above 2.2 mol/mol.

3.2.3.3 Results of jar tests with dosing of poly aluminium chloride

For the jar tests with poly aluminium chloride (PACl) the same sample WWTP effluent as in the previous jar tests was used, measuring an average pH of 6.89, a temperature of 19.2 °C and similar phosphorus concentrations.

During the jar tests poly aluminium chloride was dosed in concentrations 0.3 – 1.3 mgAl/l resulting in ratios of molAl per molPO$_4$-P$_{<0.45\ \mu m}$ of 0.7-2.9. Despite the lower molecular weight of Al^{3+} compared to Fe^{3+} often similar dosing ratios for efficient phosphorus removal are often suggested. In contrast, Jiang and Graham (1998) suggested a ratio of Al/P of 1.4/1.0 mol/mol for efficient phosphorus removal. The results of the jar without dosing can be used as a reference test to indicate potential removal without chemical enhancement.

Visual observation during the jar tests showed increased floc formation but the flocs generally appeared to be smaller than with ferric chloride. Results of the jar tests with poly aluminium chloride are presented in Table 3. 3.

Table 3. 3: Results of the jar tests with dosing of PACl for preliminary investigation of filtration tests at WWPT Utrecht

Jar no Parameter	unit	1	2	3	4	5	6
Dosing Al	mgAl/L	0.0	0.3	0.5	0.7	1.0	1.3
Al$_{unfilt.}$	mgAl/L	0.05	0.22	0.24	0.32	0.36	0.32
Al$_{< 0.45\ \mu m}$	mgAl/L	0.05	0.07	0.05	0.05	0.06	0.05
PO$_4$-P$_{unfilt.}$	mgPO$_4$-P/L	0.66	0.62	0.60	0.57	0.51	0.45
PO$_4$-P$_{< 0.45\ \mu m}$	mgPO$_4$-P/L	0.58	0.48	0.41	0.35	0.3	0.23
$P_{tot\ unfilt.}$	mgPO$_4$-P/L	0.90	0.80	0.70	0.70	0.60	0.50
$P_{tot\ <0.45\ \mu m}$	mgPO$_4$-P/L	0.70	0.50	0.50	0.40	0.40	0.30

Results show some flocs are removed by settling leading to decreasing concentrations of total phosphate with increasing dosages. Removal increases even more if the samples are filtered. As with ferric chloride, the highest dosage leads to the highest removal, even if the result for PO$_4$-P$_{<0.45\ \mu m}$ is still unsatisfactory.

Regarding the overall phosphorus removal, ferric chloride with 2.0 mgFe/L and poly aluminium chloride with 1.3 mgAl/L show comparable results. Considering the average pH of 7.0 of the WWTP effluent, the efficiency of poly aluminium chloride within the optimal pH range and ferric chloride at its lower limit of the optimal pH region is similar.

The visual observation of floc sizes and the removal of phosphorus by settling can only be used as an indicator for filterability of phosphorus particulates. More representative results for phosphorus removal could be obtained with filtration tests involving filtration mechanisms which positively affect the overall phosphorus removal and can not be imitated with jar tests.

3.2.4 Setup of pilot scale filtration tests

3.2.4.1 Introduction

The effluent quality and the jar tests were used as the starting point for the pilot tests at WWTP Utrecht. The initial investigations pointed towards the feasibility of further nutrient removal in a discontinuous filter as post-treatment for secondary WWTP-effluent. Suspended solids, phosphorus and nitrogen removal may be combined in one filtration unit applying denitrifying flocculating filtration.

3.2.4.2 Pilot scale installation

Figure 3. 2 shows parts of the configuration of the pilot installation including the “conventional” filtration techniques.

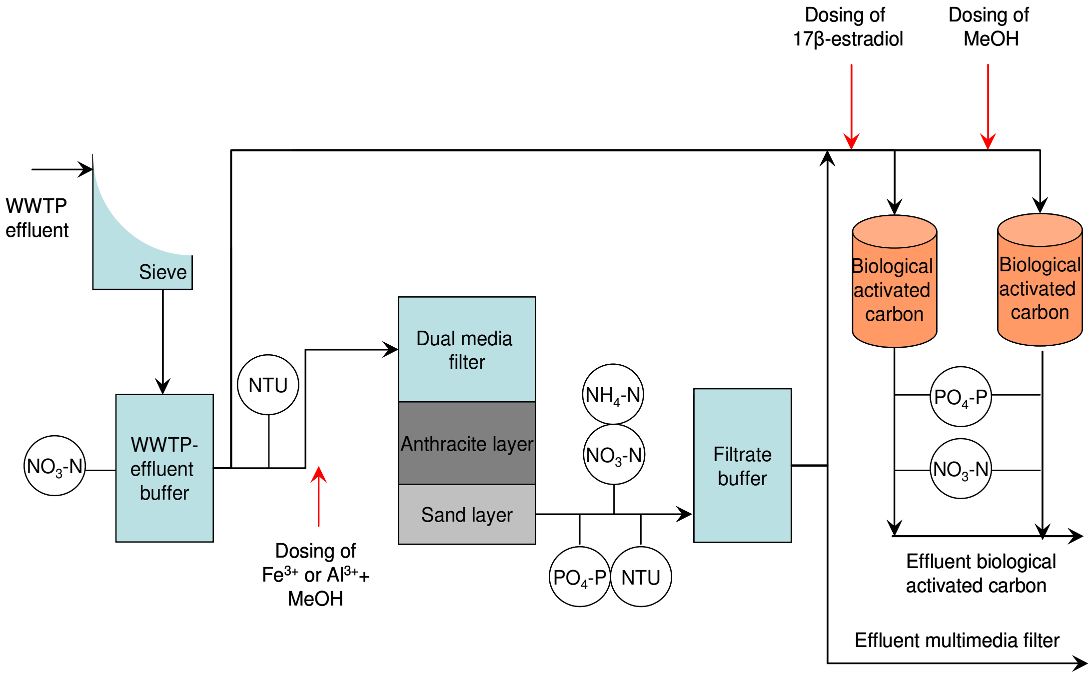

Figure 3. 2: Schematic overview of pilot installations (Te Poele, et al., 2004)

The pilot plant basically consisted of a dual media filter and two biological activated carbon filters with measuring points for online analysers and dosing points for chemicals. The WWTP effluent was collected in the WWTP effluent buffer, after passing a sieve (450 µm). The filtrate of the dual media filter was collected in the filtrate buffer. It was possible to distribute water to the other installations from both of these buffer tanks.

Turbidity was analysed online in the process influent and in the filtrate of the dual media filter. Other online measurements were:

- Nitrate in the effluent buffer (in a later period also phosphate analyser),
- Nitrate, ammonium and phosphate in the filtrate buffer, and
- Nitrate and phosphate in the filtrate of the biological activated carbon filters.

In addition to this, several parameters were measured manually. Cuvette analyses were performed for nitrogen and phosphorus content, chemical and biochemical oxygen demand, sulphate, heavy metals. The nitrogen and phosphorus results were compared to the online results regularly (Te Poele *et al.*, 2004). Furthermore, particle size and volume distributions were calculated from results obtained with a particle counter.

3.2.4.3 Dual media filter

For the pilot scale investigation a down flow discontinuous filtration installation (inner diameter 80 cm, lower layer: quartz sand Ø 1.5-2.25 mm, 40 cm height; upper layer:

anthracite Ø 2.0-4.0 mm, 80 cm height) was used. The filter installation was preceded by a constant dosing of poly aluminium chloride (PACl, dosing of 1.0-2.0 mg Al/L) or ferric chloride ($FeCl_3$, dosing of 1.0-3.0 mg Fe/L). In case of denitrifying filtration methanol was used as an external carbon source with a dosing ratio of 4.2 grams methanol per gram nitrate nitrogen (gCH_3OH/gNO_3-N). This ratio includes an overdose of 40% to remove free oxygen present in the WWTP effluent. During simultaneous denitrifying flocculating filtration a constant flow rate of 5 m^3/h (filtration rate 6.3 m/h; bed contact time 13 min) was applied.

For the experiments with single flocculating filtration the installation was operated with filtration rates of 6.3, 10.0, 12.5 and 17.0 m/h (Te Poele *et al.*, 2004).

Before the online phosphate analyser was installed, manual measurements of orthophosphate (PO_4-$P_{<0.45\mu m}$) were performed in the effluent buffer on samples filtered over a 0.45 µm cellulose acetate filter.

3.2.4.4 Scheduling of filtration tests

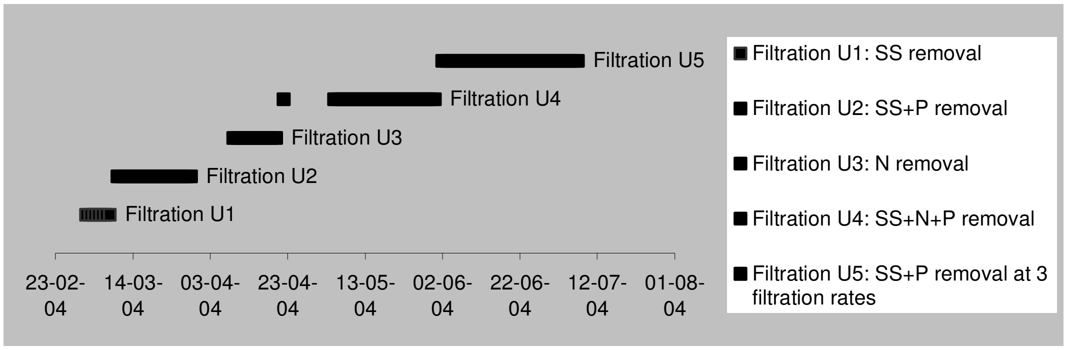

Figure 3. 3: Time schedule of filtration tests at WWTP Utrecht; SS: suspended solids, P: phosphorus, N: Nitrogen

From the detailed operational description of filtration tests conducted at the WWTP Utrecht (Appendix B.1), a general schedule was defined including five different conditions for filtration:

- Filtration U1: Filtration of suspended solids; operation: 24 h filter run time, 10 m/h filtration rate,
- Filtration U2: Flocculating filtration for combined removal of suspended solids and phosphorus; operation: 16 h filter run time, 10 m/h filtration rate, 0.5-1.2 $mgFe^{3+}/L$ or 1.0 $mgAl^{3+}/L$,

- Filtration U3: Denitrifying filtration for removal of suspended solids and nitrogen; operation: 8-16 h filter run time, 6.3 m/h filtration rate, 4.8-4.2 $kgCH_3OH/kgNO_3$-N,

- Filtration U4: Filtration for simultaneous removal of suspended solids, phosphorus and nitrogen; operation: 8 h filter run time, 6.3 m/h filtration rate, 4.2 $kgCH_3OH/kgNO_3$-N and 1.0 $mgFe^{3+}/L$ or 1.0-2.0 $mgAl^{3+}/L$,

- Filtration U5: Flocculating filtration for combined removal of suspended solids and phosphorus; operation: 4-8 h filter run time, 10.0/12.5/17.0 m/h filtration rate, 2.0 mg Al^{3+}/L.

3.3 Filtration tests at WWTP Beverwijk

3.3.1 Design and operation of the WWTP

The influent of the wastewater treatment plant Beverwijk-Zaanstreek (BZ) consists of wastewater from the surrounding districts Zaandam West, Assendelft, Zaandijk, Koog aan de Zaan, Westzaan, Wormerveer, Wormer, Oost-Knollendam, De Woude, Marker-Binnen, Castricum, and the industrial district of Beverwijk. Furthermore, excess wastewater from the WWTP Beverwijk en Omstreken (BO) up to the maximum discharge of 2,600 m^3/h and condensate plus cooling water (ca. 500m^3/h) from the neighboured sludge drying plant is discharged and treated at the WWTP BZ. The designed capacity is 152,000 PE with a hydraulic load of 3,900 m^3/h (DWF) and 8,000 m^3/h (RWF) and a total daily flow of 39,000 m^3/d.

The wastewater is treated in six parallel treatment trains each consisting of a bar screen, a primary sedimentation tank, a selector, an aeration tank and a final sedimentation tank. Conditioning with iron salt in combination with polyelectrolyte precedes the primary sedimentation. This ensures pre-precipitation and a low food to micro organism ratio for the biological treatment (HHNK, 1998). The scheme of treatment is shown in Figure 3. 4.

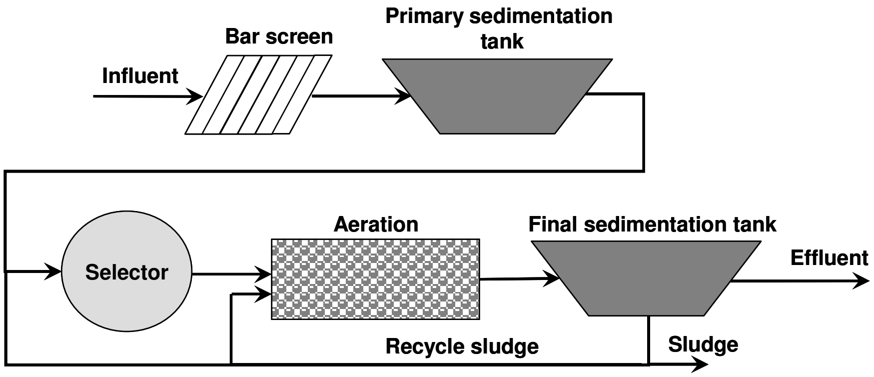

Figure 3. 4: Treatment scheme of the WWTP Beverwijk-Zaanstreek

The final effluent is discharged into the "De Pijp", an open connection with the North Sea canal.

As a result of more stringent discharge limits expected as part of the implementation of the Water Framework Directive in 2015, the Water Board Hollands Noorderkwartier agreed that filtration experiments could be executed on a laboratory scale at WWTP BZ.

3.3.2 *Effluent quality*

Table 3. 4 shows the effluent quality of the WWTP BZ in the year 2004. The displayed effluent quality served as a starting point for laboratory scale filtration tests, aimed at single phosphorus or simultaneous nutrient removal to MTR quality. The total phosphorus and total nitrogen concentrations vary between 0.7 – 2.2 mgP_{tot}/L and 4.0 – 16.1 mgN_{tot}/L, respectively.

Table 3. 4: Effluent quality of the WWTP Beverwijk in 2004

Parameter	Unit	Effluent WWTP BZ		
		Min	Max	Average
COD	mgO_2/L	23	100	44.6
BOD_5^{20}	mgO_2/L	1.5	33	4.4
N_{Kj}-N	mg/L	1.6	21	3.5
NH_4-N	mg/L	0.3	2.6	1.1
NO_2-N	mg/L	0.1	0.7	0.3
NO_3-N	mg/L	1.5	11.0	7.3
N_{tot}	mg/L	4.0	16.1	11.0
PO_4-P	mg/L	0.2	1.5	0.8
P_{tot}	mg/L	0.7	2.2	1.4

Parameter	Unit	Effluent WWTP BZ		
		Min	Max	Average
Cl^-	mg/L	115	385	255.7
pH	-	6.8	7.7	7.2
SS	mg/L	9	27	16.3

3.3.3 Preliminary investigations

3.3.3.1 Introduction

Jar tests with dosed of iron chloride ($FeCl_3$) and poly aluminium chloride (PAC) were conducted as preliminary investigations for predictions on the laboratory scale filtration tests. However, with the jar test installation a minimum of 20 seconds was needed for injection of the chemicals and reduction of the stirrers speed for the following sedimentation step.

The flocculation time calculated for the lab scale installation lasted only a couple of minutes with a slow mixing speed. For that reason it was included in the standard sedimentation time of 30 min. The concentrations of the effluent samples were as following:

- Phosphate (PO_4-$P_{unfilt.}$) = 1.1 mg/L
- Orthophosphate (PO_4-$P_{<0.45\mu m}$) = 0.90 mg/L
- Iron ($Fe_{unfilt.}$) = 0.26 mg/L
- Iron ($Fe_{<0.45\mu m}$) < 0.1 mg/L (detection limit)
- Aluminium ($Al_{unfilt.}$) = 0.09 < 0.1 mg/L (detection limit)
- Turbidity = 2.55 NTU
- pH ~ 7.3

The following section presents the results for the analysis of the siphoned supernatant.

3.3.3.2 Results of jar tests with dosing of ferric chloride

The results for the jar test series with ferric chloride, the results of which are shown in Table 3. 5. Dosing concentrations between 2.0 – 5.0 mgFe/L were applied resulting in ratios molFe per molPO$_4$-$P_{<0.45\mu m}$ of 1.2 – 3.1. These ratios were used according to proposed ratios in literature for metal / orthophosphate of 3:1 (Metcalf and Eddy, 2003).

Trends of decreasing phosphorus concentrations and rising turbidity values with increased dosing concentrations were observed. The highest removal of orthophosphate (PO_4-$P_{<0.45\mu m}$) and total phosphorus ($P_{tot\ <0.45\mu m}$) corresponded to a dose of 5.0 mgFe/L.

However, a dose of 4.0 mgFe/L already lead to a concentration of 0.14 mg$P_{tot <0.45\mu m}$ /L in the filtered supernatant. It should be noted that the difference between analyses obtained from unfiltered and filtered samples represents the particulate fraction.

Visual observation indicated the formation of large aggregates with increased dose rates. Most of these flocs were suspended instead of settled. Therefore phosphorus removal down to MTR-quality $P_{tot\ unfilt..}<0.15$ mg/L in the unfiltered sample has not been reached.

For the first filtration tests with ferric chlorides on site a starting concentration of 3.0 mgFe/L was selected.

Table 3. 5: Results of jar test series with ferric chloride for preliminary investigations for lab scale filtration tests on WWTP Beverwijk; unfilt.: unfiltered

Jar no		1	2	3	4
Parameter	unit				
Dosing Fe	mgFe/L	2.0	3.0	4.0	5.0
Turbidity	NTU	2.6	3.1	3.5	3.9
$Fe_{unfilt..}$	mgFe/L	2.45	3.64	4.71	5.29
$Fe_{< 0.45\ \mu m}$	mgFe/L	0.09	0.08	0.08	0.08
$PO_4\text{-}P_{unfilt..}$	mgPO_4-P/L	1.02	0.99	0.97	1.04
$PO_4\text{-}P_{< 0.45\ \mu m}$	mgPO_4-P/L	0.51	0.30	0.12	0.04
$P_{tot\ unfilt..}$	mgPO_4-P/L	1.09	1.10	1.10	1.12
$P_{tot < 0.45\ \mu m}$	mgPO_4-P/L	0.49	0.30	0.14	0.07

3.3.3.3 Results of jar tests with dosing of poly aluminium chloride

For the jar tests with poly aluminium chloride molar ratios of 1.3 to 3.8 (mol metal : mol orthophosphate) were used and should have resulted in dosing concentrations between 1.0-3.0 mgAl/L. However, the concentrations of aluminium measured in the unfiltered supernatant, did not match with the defined dosing, as can be seen from the results in Table 3. 6. The analyses of the concentrations in the supernatant after sedimentation showed generally higher concentrations than expected.

This difference could be attributed to one, or more of the following issues: (1) inaccuracies of dosing of small amounts using a pipette, (2) improperly cleaning of the dosing equipment, (3) higher aluminium content of the utilised product, or (4) inaccuracies while conducting the analyses. Using measured aluminium concentrations in the unfiltered sample as the actual dosing concentration leads to calculated ratios of 2.7-5.6 molAl/molPO_4-$P_{< 0.45\mu m}$. These ratios should be seen as inaccurate because the aluminium concentrations were measured in the supernatant after completed sedimentation time. The aluminium flocs which were settled are not included in the measured concentration. This becomes clear for jar no 6 where aluminium concentration is lower than in jar no 5 but has a higher removal of phosphorus.

However, increased dosing clearly leads to formation of bigger flocs. The results of the jar without dosing can present reference results. Likewise to the preliminary series with ferric chloride most of the flocs stayed in suspension instead of settling. This is confirmed by remaining high phosphorus concentrations for unfiltered samples.

The highest removal of orthophosphate and total phosphorus ($P_{tot\ <0.45\mu m}$) was measured in the jar test no 4/6 with a concentration of 0.04/0.05 mg$P_{tot\ <0.45\mu m}$/L in the filtered supernatant (Table 3. 6). The dosing of > 2.0 mgAl/L in jar no 2 already leads to a concentration of 0.16 mg$P_{tot<0.45\mu m}$ /L in the filtered supernatant.

For the initial on site filtration tests with poly aluminium chloride a starting concentration of 2.0 mgAl/L was selected.

Table 3. 6: Results of jar test series with PAC for preliminary investigations for lab scale filtration tests on WWTP Beverwijk; unfilt.: unfiltered

Jar no		1	2	3	4	5	6
Parameter	unit						
Dosing Al	mgAl/L	0	1.0	1.5	2.0	2.5	3.0
Turbidity	NTU	1.5	2.8	3.2	3.8	4.0	2.8
$Al_{unfilt.}$	mgAl/L	0.08	2.12	2.30	2.90	4.40	3.90
$Al_{< 0.45\ \mu m}$	mgAl/L	0.04	0.09	0.08	0.09	0.07	0.07
$PO_4\text{-}P_{unfilt.}$	mgPO_4-P/L	1.09	1.05	0.92	0.91	0.73	0.53
$PO_4\text{-}P_{< 0.45\ \mu m}$	mgPO_4-P/L	0.85	0.15	0.05	0.04	0.05	0.03
$P_{tot\ unfilt.}$	mgPO_4-P/L	1.13	1.10	0.98	0.95	0.82	0.55
$P_{tot < 0.45\ \mu m}$	mgPO_4-P/L	1.05	0.16	0.06	0.04	0.07	0.05

3.3.4 Setup of lab scale filtration tests

3.3.4.1 Introduction

Based on the initial experiments of simultaneous nutrient removal at WWTP Utrecht and with the intention of confirming and expanding the knowledge gained led to laboratory scale filtration experiments at WWTP Beverwijk were undertaken. The effluent quality and the results of preliminary jar tests formed the basis for these investigations.

3.3.4.2 Lab scale installation

Figure 3. 5 shows the configuration of the lab scale filtration installation. The WWTP effluent was pumped from the final clarifiers to the research installation and passed first

through a buffer tank (volume = 1 m^3) in order to achieve pre-settling of coarse material. After conditioning with coagulant and/or carbon source the effluent was pumped to the downward flow dual media filter. Pressure readings over the filter bed height were available through the installed manometer board. Backwash could be initiated manually with air, water or both.

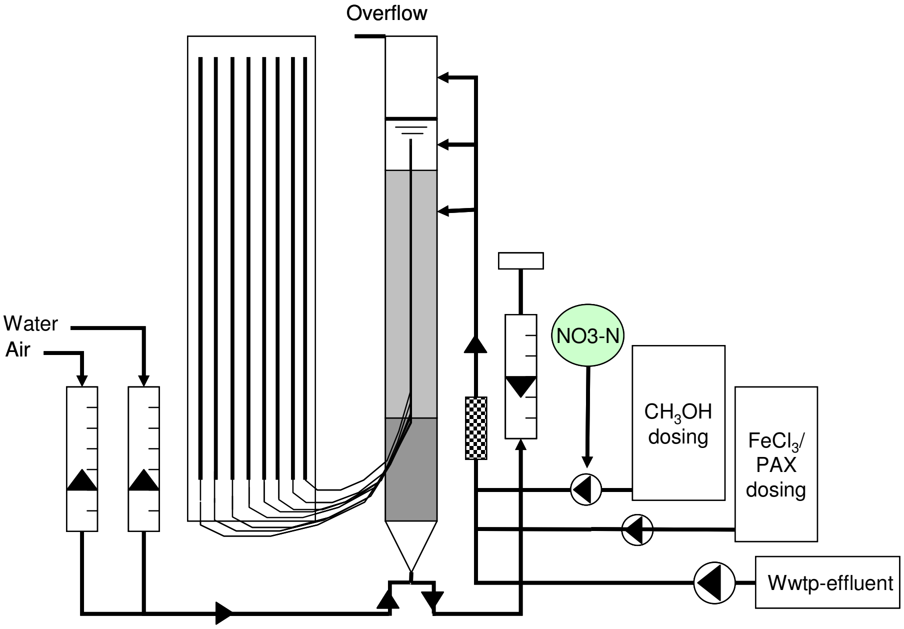

Figure 3. 5: Scheme of lab scale installation applied for filtration tests on the WWTP Beverwijk

Methanol was dosed at a rate proportional to the nitrate nitrogen concentration in WWTP effluent. During the experiments grab samples were taken from three sampling points, namely WWTP effluent (EF), flocculated WWTP effluent (AF) and process filtrate (FI). The orthophosphate content was determined for the raw samples and for samples filtered through a 0.45 μm cellulose acetate filter. The difference between the raw and filtered samples provides the concentration of particulate inorganic phosphorus.

The turbidity of these grab samples was measured with a HACH turbiditymeter. Additionally, a MetOne PCX particle counter was used to measure particle size distributions for both raw samples and samples that were diluted 200% (dilution rate 3) with demineralised water. It was necessary to analyse diluted samples because the particle concentrations of the undiluted samples exceeded the concentration limit of the particle counter (> 10,000 counts/ml).

3.3.4.3 Dual media filter

A dual media laboratory scale (inner diameter 12.9 cm; lower layer: quartz sand Ø 0.8-1.25 mm, height 40 cm; upper layer: anthracite Ø 1.6-2.5 mm, height 80 cm) filtration installation was operated at filtration rates of 10 and 20 m/h, using ferric chloride (dosing: 3-4 mgFe/L) and poly aluminium chloride (dosing: 2-3 mgAl/L) for flocculation. The filter was operated with free raising water table.

The installation was used first for blank filtration (without dosing), followed by tests with single denitrification and finally for simultaneous denitrifying flocculating filtration.

Single denitrifying or simultaneous filtration was performed with higher filtration rates of 10 m/h (131 l/h) and 20 m/h (262 l/h) with empty bed contact times of 9 and 4.5 minutes. The filtration rate was increased after the starting phase for the denitrification and operated at 6 m/h.

The methanol dose ratio was the same as for the pilot scale investigations at WWPT Utrecht (4.3 gCH_3OH/gNO_3-N). After optimal denitrification was reached the filtration rate was increased without increasing the methanol dose, resulting in a dosing rate of 3.2 gCH_3OH/gNO_3-N. In the first series of experiments, the backwash procedure was copied from the earlier research conducted at WWTP Utrecht. Optimisation of the backwash procedure occurred during the subsequent filtration tests.

3.3.4.4 Scheduling of filtration tests

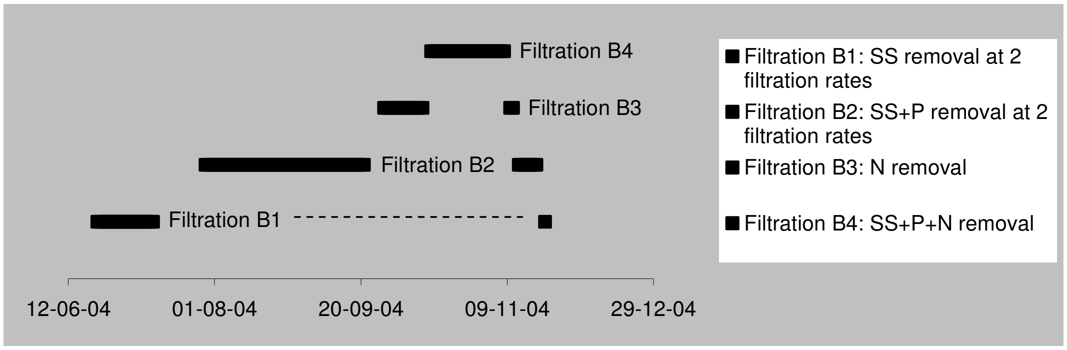

Figure 3. 6: Time schedule of filtration tests at WWTP Beverwijk; SS: suspended solids, P: phosphorus, N: Nitrogen

Detailed description of filter operation during the research period at the WWTP Beverwijk can be found in Appendix B.1. However a schematic overview is provided in Figure 3. 6. Four different operational conditions for filtration were defined with filtration run times between 7 and 24h:

- Filtration B1: Filtration of suspended solids; operation: 10 and 20 m/h filtration rate,

- Filtration B2: Flocculating filtration for combined removal of suspended solids and phosphorus; operation: 10 and 20 m/h filtration rate, 3-4 $mgFe^{3+}/L$ or 2-3 $mgAl^{3+}/L$,
- Filtration B3: Denitrifying filtration for removal of suspended solids and nitrogen; operation: 6.3 and 10 m/h filtration rate, 3.2 $kgCH_3OH/kgNO_3\text{-}N$,
- Filtration B4: Filtration for simultaneous removal of suspended solids, phosphorus and nitrogen; operation: 10 m/h filtration rate, 3.2 $kgCH_3OH/kgNO_3\text{-}N$ and 2.0-4.0 $mgFe^{3+}/L$ or 2.0-3.5 $mgAl^{3+}/L$.

3.4 Filtration tests at WWTP Horstermeer

3.4.1 Design and operation of the WWTP

The municipal wastewater treatment plant Horstermeer is in operation since 1985. It has a design capacity of 200,000 PE and a maximum hydraulic capacity of 5,000 m^3/h (daily 21,000 m^3/d). The influent consists of wastewater from the surrounding areas Naarden-Bussum, Hilversum-West and Nederhorst den Berg. After the preliminary treatment with bar screens the WWTP influent enters two primary sedimentation tanks. It is then transferred to two aeration tanks, which are divided into denitrification and nitrification zones with a sludge load of 0.07 BOD/(kgSS.d). Ferric sulphate ($FeClSO_4$) and poly aluminium chloride (PAC) are dosed in the pre-settled wastewater and in the biological treatment step, respectively. After passing through four final sedimentation tanks the effluent is discharged into the river Vecht, part of the river Rhine basin district. A scheme of the treatment processes of the WWTP Horstermeer is shown in Figure 3. 7.

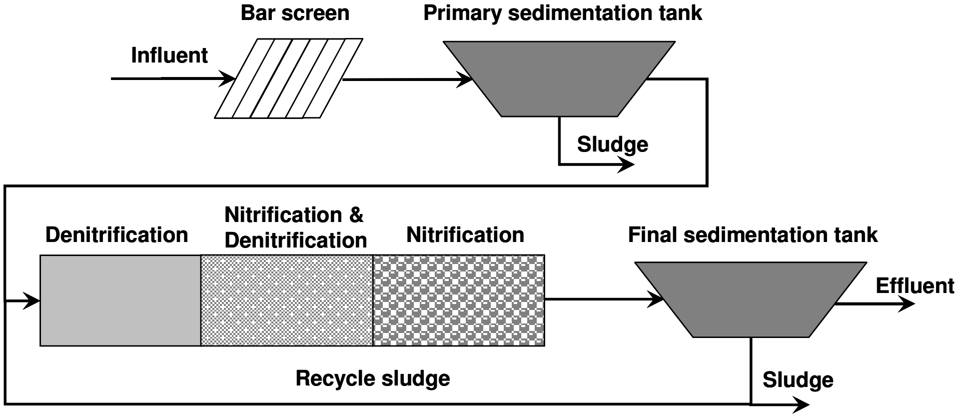

Figure 3. 7: Scheme of treatment processes on the WWTP Horstermeer

3.4.2 Effluent quality

Table 3. 7 shows the effluent quality parameters for 2004. This effluent quality served as a reference point for pilot scale investigations on the WWTP Horstermeer.

One research item of the pilot scale filtration tests at WWTP Horstermeer was to investigate the possible removal of other substances under optimal conditions for nutrient removal. For that reason Table 3. 7 also presents concentrations of polycyclic aromatic hydrocarbons (PAH's) and heavy metals besides the concentrations of "conventional" parameters. The concentrations of 0.3-2.8 mg/L total phosphorus and 9.4-33.0 mg/L total nitrogen indicate possibilities for further improvement of the final effluent quality applying advanced filtration for simultaneous nutrient removal.

Table 3. 7: Effluent quality of the WWTP Horstermeer in 2004

Parameter	Unit	Average	Min	Max	Number of measurements
Conventional parameters					
Flow	m³/d	27,500	16,500	91,500	305
COD	mg/L	33	17	55	51
BOD	mg/L	3	1	8	50
N_{tot}	mg/L	14.6	9.4	33.0	51
N_{Kj}-N	mg/L	4.1	1.2	16.2	51
NH_4-N	mg/L	2.4	0.1	12.0	51
NO_3-N	mg/L	10.2	6.1	19.3	51
NO_2-N	mg/L	0.2	0.1	0.8	51
P_{tot}	mg/L	0.9	0.3	2.8	51
PO_4-P	mg/L	0.5	0.1	2.4	51
Suspended solids	mg/L	10.1	8.0	30.9	51
PAHs					
Anthracene	µg/L		<0.005	<0.005	4
Fluoranthene	µg/L		0.01	0.03	4
Benzo (a)pyrene	µg/l		<0.005	0.0085	3
Benzo(k)-fluoranthene	µg/l		0.0059	<0.005	4
Naphthalene	µg/l		<0.039	<0.039	4
Heavy metals					
Arsenic	µg/l		<0.733	1.33	3
Cadmium	µg/l		<0.24	<0.24	3

Parameter	Unit	Average	Min	Max	Number of measurements
Chrome	µg/l		<0.7	1.27	3
Lead	µg/l		<7	<7	3
Copper	µg/l		3.5	13.6	3
Mercury (inorganic/ methyl)	µg/l		<0.01	<0.01	3
Nickel	µg/l		<3	12.6	3
Zink	µg/l		24.6	76.7	3

3.4.3 Preliminary investigations

3.4.3.1 Introduction

Jar tests were conducted using ferric chloride ($FeCl_3$) and poly aluminium chloride (PACl) as coagulant preceding the pilot filtration tests. The coagulation step in the pilot installation was only a few seconds. However, the duration of the coagulation step for the jar tests was set to 20 seconds, which is equal to the minimum duration possible on the jar test installation.

Due to the short flocculation time at the pilot installation (2 min), the separate flocculation step considered in the jar test procedure was omitted. In order to better simulate the pilot installation and improve comparability of the results the standard sedimentation time of 30 min followed directly after the coagulation time.

The concentrations of the effluent samples were as following:

- Phosphate (PO_4-$P_{unfilt.}$) = 0.50 – 0.53 mg/L
- Orthophosphate (PO_4-$P_{<0.45\mu m}$) = 0.3 – 0.4 mg/L
- Iron ($Fe_{unfilt.}$) = 0.46 – 1.25 mg/L
- Aluminium ($Al_{unfilt.}$) = 0.09 < 0.1 mg/L (detection limit)
- Turbidity = 3.5 – 5.4 NTU
- pH = 6.30 – 6.37

The following paragraphs present the analysis of the siphoned supernatant.

3.4.3.2 Results of jar tests with dosing of ferric chloride

In this jar test ferric chloride ($FeCl_3$) was dosed in concentrations 1.0-3.0 mgFe/L. This has resulted in molar ratios Fe/PO_4-$P_{<0.45\mu m}$ of 1.6-4.9 mol/mol. The majority of flocs appeared as pinpoints in suspension in all jars with a very thin layer of settlement. The analytical results show a decrease of phosphorus measurements with increasing dose. However they also show concentrations for total phosphorus lower than total phosphate (PO_4-P_{unfilt}). This can only be explained by analytical inaccuracies. Some colorimetric analyses using unfiltered samples may be influenced by suspended matter. Analyses including a digestion step (boiling at 100°C) as sample pre-treatment, for example total phosphorus analysis, will be less affected by suspended matter and as such can be considered as more reliable. This is due to the degradation of the suspended matter which reappears in solution.

However even at a dose of 3.0 mgFe/L floc formation was less than expected. This is indicated by the remaining orthophosphate (PO_4-$P_{<0.45\mu m}$) concentration of 0.14 mg/L of the filtered sample. The results are summarized in Table 3. 8.

Table 3. 8: Results of jar test series with ferric chloride ($FeCl_3$) for preliminary investigations for pilot scale filtration tests on WWTP Horstermeer (Gorter, 2005); unfilt.: unfiltered

Jar no		1	2	3	4	5	6
Parameter	unit						
Dosing Fe	mgFe/L	0	1.0	1.5	2.0	2.5	3.0
$Fe_{unfilt.}$	mgFe/L	0.39	0.76	0.95	1.15	1.34	1.6
$Fe_{< 0.45\ \mu m}$	mgFe/L	0.22	0.24	0.27	0.25	0.34	0.42
PO_4-$P_{unfilt.}$	mgPO_4-P/L	0.50	0.50	0.50	0.48	0.49	0.47
PO_4-$P_{< 0.45\ \mu m}$	mgPO_4-P/L	0.26	0.24	0.21	0.15	0.18	0.14
$P_{tot\ unfilt.}$	mgPO_4-P/L	nd	nd	nd	nd	nd	nd
$P_{tot<0.45\ \mu m}$	mgPO_4-P/L	0.38	0.30	0.32	0.28	0.27	0.22

nd: not determined

The results of phosphorus removal with jar tests can only be seen as indicative because the filtration mechanisms can not be simulated. Nevertheless, the conclusion can be drawn that the major developed pinpoint flocs will be difficult to filtration and phosphorus removal might be poor. The poor flocculating ability of ferric chloride could be caused by the low pH of the WWTP effluent and the lowering of the pH by the addition the ferric chloride solution. This may cause a pH drop below the optimal pH for reaction.

3.4.3.3 Results of jar tests with dosing of poly aluminium chloride

Table 3. 9 shows the results for the jar tests using poly aluminium chloride (PACl) as coagulant. Aluminium was dosed in similar concentrations as iron resulting in molar ratios Al/PO_4-$P_{<0.45\mu m}$ of 2.8-8.6 mol/mol. At a dose rate of 1.0-1.5 mgAl/L the orthophosphate (PO_4-$P_{<0.45\mu m}$) concentration is already below 0.1 mg/L. This indicates

that aluminium ions react more effectively that the iron ions with orthophosphate ions. However, still 0.1 mg/L of particulate phosphate (difference PO_4-$P_{unfil.}$ - PO_4-$P_{<0.45\mu m}$) remains in suspension. Larger flocs with improved settling occurred at a dose of 2.0 mgAl/L. It can be concluded that poly aluminium chloride is the favourable coagulant for this WWTP effluent which has a pH of about 6. These results led to a starting ratio of 2.8-5.7 molAl/molPO_4-$P_{<0.45\mu m}$ for the following filtration tests.

Table 3. 9: Results of jar test series with poly aluminium chloride for preliminary investigations for pilot scale filtration tests on WWTP Horstermeer (Gorter, 2005); unfilt.: unfiltered

Jar no		1	2	3	4	5	6
Parameter	unit						
Dosing Al	mgAl/L	0	1.0	1.5	2.0	2.5	3.0
$Al_{unfilt.}$	mgAl/L	0.07	0.65	0.86	0.92	1.03	0.92
$Al_{<0.45\mu m}$	mgAl/L	0.04	0.03	0.07	0.06	0.08	0.05
PO_4-$P_{unfilt.}$	mgPO_4-P/L	0.53	0.27	0.25	0.26	0.05	0.05
PO_4-$P_{<0.45\mu m}$	mgPO_4-P/L	0.40	0.08	0.06	0.04	0.03	0.01
$P_{tot\ unfilt.}$	mgPO_4-P/L	0.63	0.63	0.65	0.59	0.47	0.44
$P_{tot\ <0.45\mu m}$	mgPO_4-P/L	0.43	0.07	0.08	0.04	0.03	0.01

3.4.4 Setup of pilot scale filtration tests

3.4.4.1 Introduction

After successful simultaneous nutrient removal was achieved with dual media filtration at the WWTP Utrecht and Beverwijk, the Water Board Waternet, Water Management and Sewage Service Amsterdam, agreed on the common objective to continue the investigations. The goal was to investigate the maximum possible removal for nutrients and other substances with conventional bio-filtration. Stable operation for a longer period of time was another major research item. The pilot research started in March 2005 and predominantly involved dual media filtration. Periodically further removal by single media filtration, lab scale activated carbon and fine sand filtration was tested. The combination of pilot scale dual media followed by lab scale single media filtration provides an indication for a multi media filtration.

In a parallel research biological activated carbon filtration and continuous filtration were investigated in order to asses their abilities on removing suspended solids, nitrogen and phosphorus compared to the dual media filter installation.

Main goals of the pilot studies were

- achieve good filtration results with long term stable operation,
- investigate potential of simultaneous heavy metal removal with removal of suspended solids, phosphorus and nitrogen,
- investigate performance of simultaneous denitrifying flocculating filtration with filtration rates above 6.3 m/h, such as 10 – 12.5 m/h,
- compare of denitrifying efficiencies using methanol (and alternative carbon sources; results are not presented in this thesis),
- investigate extending the filter run time to above four hours by applying an optimised backwash regime,
- undertake an intensive study of additional value of particle characterisation and analyses.

3.4.4.2 Pilot scale installation

The pilot installation (Figure 3. 8) consisted of a dual media filter followed by two lab scale single media filter columns. Lab scale column 1 was filled with fine sand (garnet sand, 30 cm height, 0.5 – 0.8 mm grain diameter, 3,500 kg/m3 density) and column 2 with activated carbon (Norit GAC 830P, 1.2 m height, 1.6 mm grain diameter, 460 kg/m^3 density).

The dual media filter was supplied with WWTP effluent, which passed a 450 μm screen and was collected in the effluent buffer. The filtrate of the dual media filter was collected in the filtrate buffer and formed the feed water for the single media filters. Turbidity, nitrate and phosphate in the WWTP effluent were analysed online after passing the screen. The filtrate of the dual media filter was analysed online for turbidity, nitrate, phosphate and total phosphorus.

Analyses were obtained for different grab samples: WWTP effluent buffer (after passing the screen), effluent after coagulation and flocculation, filtrate dual media filter, filtrate storage tank (filtrate buffer), filtrates of lab scale filters columns consisting of a fine sand and granular activated carbon layer. In addition to conventional analyses for these samples particle measurements were also undertaken, for both undiluted (raw) and diluted samples EF, AF and FI. A dilution rate of 3, one part of the effluent sample and two parts of demineralised water was applied.

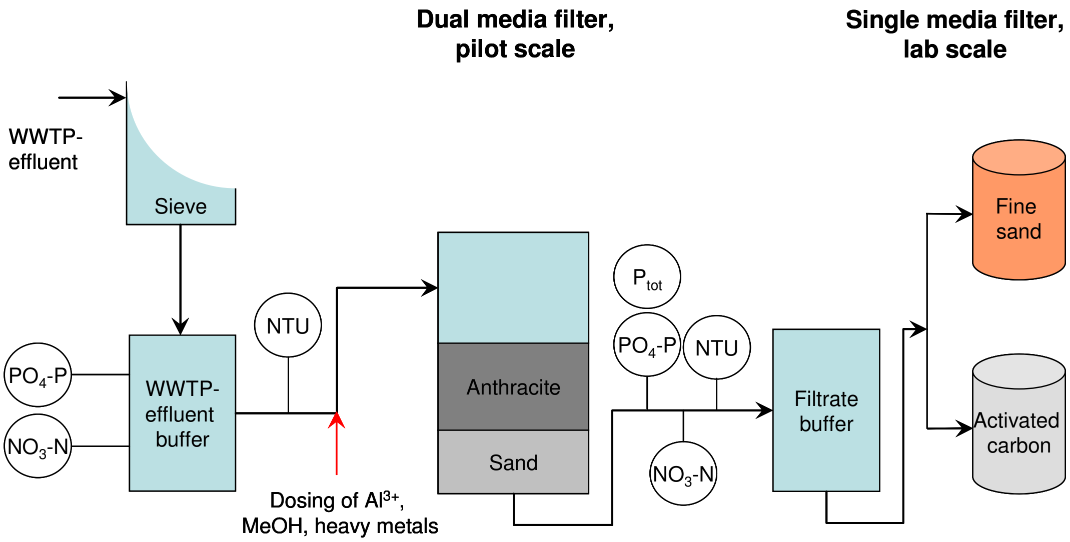

Figure 3. 8: Scheme of pilot installation for pilot investigations at WWTP Horstermeer

3.4.4.3 Dual media filter

The pilot scale installation contains two filtration layers (upper layer: anthracite, 80 cm height, 2.0 – 4.0 mm grain diameter, 1,400 kg/m^3 density; lower layer: quartz sand, 40 cm height, 0,8 – 1.25 mm grain diameter, 2,600 kg/m^3 density). In the initial stage the filter was operated at a flow rate of 8 m^3/h (filtration rate 10 m/h) resulting in filtration run times between 4 and 8 hours in the first stage. In a later stage filtration run times up to 24 h were tested and achieved.

The pilot scale installation was backwashed with air and filtrate from the filtrate buffer. For the chemical precipitation of phosphate, poly aluminium chloride (PACl) was dosed to the WWTP effluent before entering the dual media filter. Poly aluminium chloride was used as coagulant because of the low pH of the WWTP effluent. It was dosed in Me/PO_4-$P_{<0.45 \mu m}$ (mol metal per mol orthophosphate) ratios of 4.8. Higher ratios were applied for a parallel research with the same installation to investigate possible simultaneous heavy metal removal and nutrient removal.

Methanol dosage for denitrification was adjusted online and proportional to the present nitrate nitrogen concentration (usually 3 gCH_3OH/gNO_3-N) including extra methanol to remove the free oxygen concentration in the influent. Methanol dosing ratios of 3.7-5.0 gCH_3OH/gNO_3-N were used. The coagulant and methanol dosage was applied proportional to the concentration of orthophosphate and nitrate measured in the effluent buffer.

The filtrate collected in the filtrate buffer periodically provided the feed water for the lab scale installations with single-media.

3.4.4.4 Scheduling of filtration tests

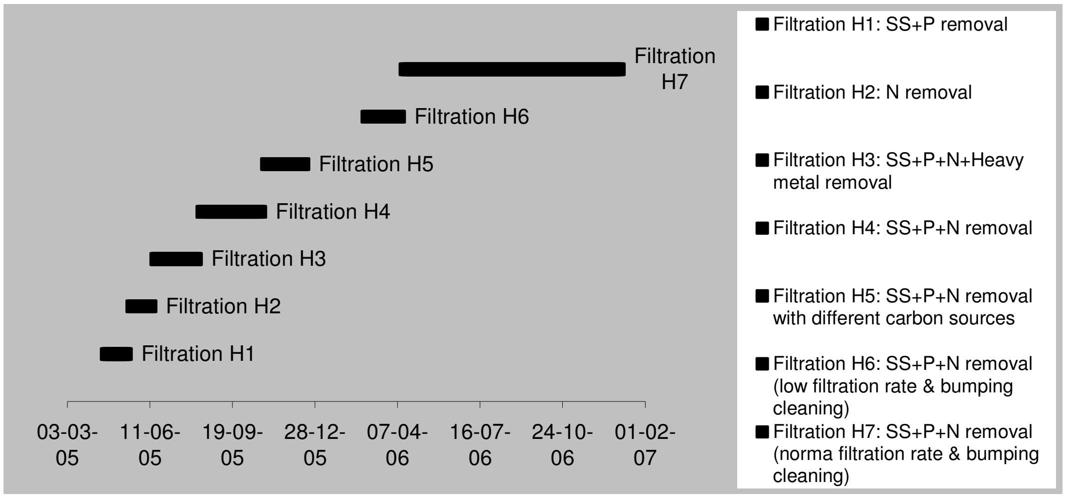

Figure 3. 9: Time schedule of filtration tests at WWTP Horstermeer; SS: suspended solids, P: phosphorus, N: Nitrogen

A general time schedule is shown in Figure 3. 9. Detailed descriptions of operational parameters during the performed filtration tests can be found in the Appendix B.1. Figure 3. 9 presents the seven major research topics during the operational period 2005-2007 for pilot tests at WWTP Horstermeer:

- Filtration H1: Flocculating filtration for suspended solids and phosphorus removal; applied: 4-8 h filter run time, 10 m/h filtration rate,
- Filtration H2: Denitrifying filtration for removal of nitrogen; applied: 6 h filter run time, 6.3 m/h filtration rate, 4.5-5.0 gCH_3OH/gNO_3-N,
- Filtration H3: Filtration for simultaneous removal of suspended solids, phosphorus, nitrogen and heavy metals; applied: 6 h filter run time, 6.3 m/h filtration rate, 4.5-5.0 gCH_3OH/gNO_3-N with several days without dosing of methanol as carbon source, poly aluminium chloride (PACl) with 0-11.7 Me/PO_4-P, addition of heavy metal solution resulting in a dosed concentration of approx. 150-200µg/L for each, copper, zinc and nickel,
- Filtration H4: Filtration for simultaneous removal of suspended solids, phosphorus, and nitrogen; applied: 4-6 h filter run time, 10 m/h filtration rate, 3.7-4.7 gCH_3OH/gNO_3-N, poly aluminium chloride (PACl) with 4.8 Me/PO_4-P; investigating the lowest possible dosing of carbon source at normal filtration rate of 10 m/h,

- Filtration H5: Filtration for simultaneous removal of suspended solids, phosphorus and nitrogen; applied: 4-6 h filter run time, 10 m/h filtration rate, 3.7-4.7 gCH_3OH/gNO_3-N, poly aluminium chloride (PACl) with 4.8 Me/PO_4-P; investigating alternative carbon sources (Carbo CT and Carbo M 70) at normal filtration rate of 10 m/h,
- Filtration H6: Filtration for simultaneous removal of suspended solids, phosphorus and nitrogen; applied: 6-12 h filter run time, 6.3 m/h filtration rate, 4.5 gCH_3OH/gNO_3-N, poly aluminium chloride (PACl) with 4.8 Me/PO_4-P; investigating extended filter run times with “bumping cleaning” every 2-3 h,
- Filtration H7: Filtration for simultaneous removal of suspended solids, phosphorus and nitrogen; applied: 10-24 h filter run time, 10 m/h filtration rate, 4.5 gCH_3OH/gNO_3-N, poly aluminium chloride (PACl) with 4.8 Me/PO_4-P; investigating extended filter run times with “bumping cleaning” every 2-3 h.

3.5 Evaluation and perspective

For all three locations, WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer, the nutrient concentrations in the effluent indicated the possibility of further improvement with advanced dual media filtration operated for simultaneous nutrient removal. Preliminary investigations with jar tests could indicate an optimal type and starting concentration of coagulants for further flocculating filtration tests. The chosen process like time and mixing energy, applied for the jar tests are in accordance with the lab and pilot scale installations.

The evaluation of efficiency of a dosed coagulant in the jar tests was initially done through visible observations and analytical measurements. The dosing of coagulant was aimed for a floc size larger than pinpoints with the ability to settle. However, for majority of the conducted jar tests the flocs remained in suspension. These results may already indicate a poor mixing during coagulation, insufficient flocculation time or insufficient amount of metal salt added but has to be confirmed by filtration tests.

Another possibility was to assess the potential for floc forming through sample filtration with a 0.45 µm polycarbonate membrane filter. Good reaction of the metal salt can be confirmed when the concentrations for the dissolved fractions (<0.45 µm) of the metal salt and orthophosphate after chemical are low compared to their concentrations before dosing.

Generally poly aluminium chloride resulted as the more optimal coagulant. This is because the pH values at the three WWTP’s around 7. Generally metal- orthophosphate ratios of 3.0 were most effective in jar tests.

It is remarked that the results for optimal metal salt concentrations obtained from jar tests only provide an indicative starting point for filtration. The influence of the filtration mechanisms and their effect on the phosphorus removal can not be simulated with jar tests and as such dosing conditions may need adjustment during the filtration tests.

The level of success when combining several removal processes depends strongly on the feed water characteristics, its temporal variations, and the ability of the process to cope with a varying hydraulic and biological load. Modern Dutch WWTP's are designed to produce an average effluent quality of nitrate < 10 mg/L, total nitrogen < 14 mg/L, orthophosphate < 1.0 mg/L, total phosphorus < 1.5 mg/L and suspended solids < 10 mg/L with some possible variability, for example due to seasonal changes. Regarding the future requirements, proposed by the Water Framework Directive, several WWTP's may reach their limit for further nutrient removal even after process integrated optimisation of the activated sludge system. Further improvements can then be achieved with additional advanced treatment, like denitrifying flocculating filtration. Interactions of the different removal processes, namely: coagulation and flocculation, precipitation, denitrification, and filtration should be investigated followed by proper operational adjustments aiming at optimal performance of possibly all processes. Future operational adjustments could be the filtration rate, type of filter material, bed height, backwash procedure and frequency, types of dosed chemicals, quantities, conditions of chemical injection. Therefore optimal operational conditions, robustness and reliability of the denitrifying flocculating filtration were one of the major investigation issues in addition to identify the process limitations.

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4

Sampling and analyses in conventional filtration

4.1 Introduction

The assessment of the different processes and removal mechanisms occurring during denitrifying flocculating filtration can be obtained by measurements at several points of the filtration installation. The processes precipitation, coagulation, flocculation, denitrification and filtration can be evaluated by comparing certain parameters of the samples before, during and after completed coagulation, denitrification and filtration. Therefore, several sampling points were introduced and applied for lab scale as well as pilot scale investigations. The measurements used for evaluation could be biological or physical and chemical parameters, like turbidity, phosphorus and nitrogen measurements, and particle analyses as an additional measurement.

Next to measurements conducted at the research locations regular analyses within the monitoring schedules at the WWTP's can be used for verification or additional information.

The sampling within the filtration process, parameters used for the evaluation, sampling and the interpretation of the conducted analyses will be described in this chapter.

4.2 Sampling within the filtration process

4.2.1 *Description of sampling points*

Several samples were taken from different sampling points at the lab and pilot scale dual media filters. The following list summarises the abbreviations used for the sampling points and gives a description of the sampling points for all three research locations:

- EF: Sample of the WWTP EFfluent and feed water for the filtration experiments taken from the final sedimentation tank or effluent buffer. The WWTP effluent applied during pilot scale filtration is pre-treated by a sieve of 450 µm aiming at the removal of remaining coarse particles and stored in a buffer tank. The feed water for lab sale investigations was buffered previous to filtration aiming at sedimentation of remaining coarse and heavy solids.

- AC: Sample After completion of the Coagulation process. This sample represents the WWTP effluent after addition of coagulant and rapid mixing in the static mixer implemented in the supply conduit. At the lab scale installation this sampling point was placed directly after the static mixer and at the pilot installation in the supply conduit before the water enters the filter.

- AF: Sample After completion of the Flocculation process. AF is therefore a follow-up sample of AC after completed flocculation, occurring in the water volume above the filter bed. This sample was taken right above the surface of the upper filtration layer. This sample was interpreted as the real feed water especially when evaluating the parameters like particle distributions and phosphorus species before filtration.

- FI: Sample of the FIltrate of the dual media filter and therefore the follow-up sample of AF after filtration. This sample taken underneath the filter directly after passing the sand layer.

Additional indices were introduced to distinguish the sampling points when applying lab and pilot scale filtration simultaneously. This was predominantly the case for the investigations at WWTP Horstermeer:

- FI_buffer: Sample of the filtrate after pilot scale dual media filtration stored in a buffer tank. The filtrate is applied for the backwash of the pilot scale filter and feed water of the lab scale filters.

- FI_ac: Sample of the FIltrate after lab scale activated carbon filtration taken directly from the filtrate discharge pipe.

- FI_fs: Sample of the FIltrate after lab scale filtration with a fine sand layer.

For profile measurements over the filter bed height samples were taken from several plug valves installed at different heights of the lab and pilot scale filter.

Figure 4. 1 shows an example of the sampling points within the pilot plant at WWTP Horstermeer.

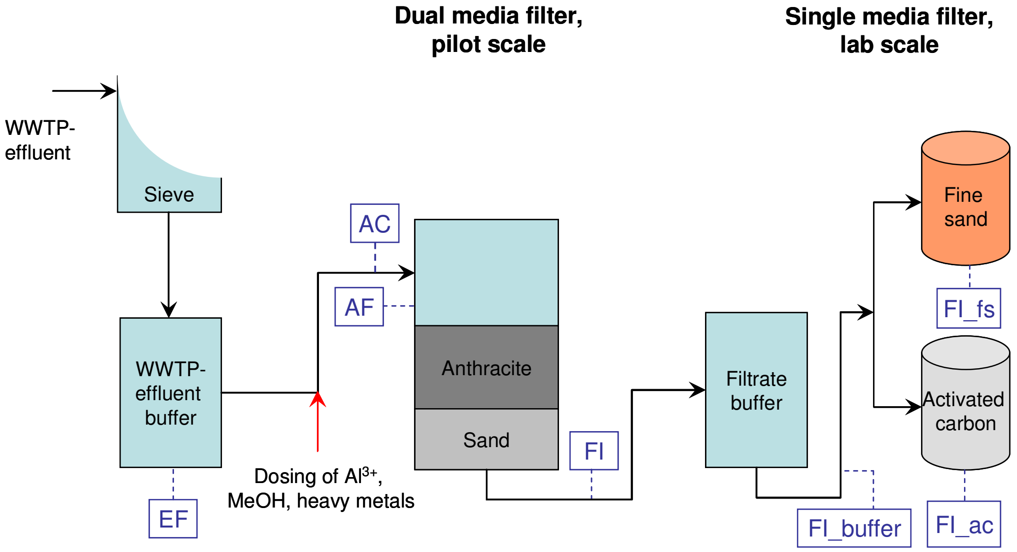

Figure 4. 1: Overview of the pilot plant set up and location of sampling points for the investigations at WWTP Horstermeer

4.2.2 Handling and pre-treatment of samples

For all manual analyses the period between sample extraction and analysis itself is limited to a very short time; maximal 30 minutes. This minimises the chance of further reaction such as sedimentation, further precipitation, coagulation and flocculation, adsorption or biological uptake which might falsify the results and their interpretation. The application of equal or at least similar handling and analytical procedures and equipment ensured the mutual comparability of measurements.

Determination of the dissolved fractions of chemical and biological parameters often requires a filtered sample. Cellulose acetate filters with a pore size of 0.45 µm were used for this purpose.

For fractionation experiments the samples were filtered with polycarbonate filters with the pore sizes of 20, 10, 5, and 2 µm. The raw (unfiltered) sample was used for every single filtration step in order to prevent excessive removal of small particles due to follow-up filtration.

In case of concentrations exceeding the measuring range of the applied analytical method sample dilution was performed with demineralised water.

To assure the accuracy of conducted particle analyses measurements were conducted with raw (undiluted) and diluted samples. The dilution was carried out with demineralised water with a dilution ratio of 3 (= 2 parts of demineralised water and 1 part sample). In order to obtain the particle number and volumes in the undiluted sample the measurements of the diluted samples were re-calculated to undiluted. The re-calculation included the dilution factor as well as the subtraction of the particle number eventually present in the dilution water.

4.3 Conventional process parameters

4.3.1 Suspended solids and turbidity

Filtration in the traditional sense is meant to remove suspended solids (SS). In WWTP effluent these solids contain mainly organic pollutants and insoluble complexes of metals. During flocculating filtration with dosing of a coagulant the suspended solids content will be increased due to the formation of metal-phosphate and metal-hydroxide precipitates; 1 mgFe/L or 1 mg Al produces 1.9 mg and 3.0 mgSS/L, (Van der Graaf *et al.*, 2001). Typical WWTP effluent contains less than 10 mgSS/L. The suspended solids concentration can be measured indirectly by the analysis of total suspended solids (TSS) according to the standard method EN ISO 11923 (ISO, 1997). During the filtration tests turbidity and particle analyses were used as indicators for the suspended solids concentration.

In general, there is a fairly good relation between turbidity and total suspended solids for the settled and filtered secondary effluent from the activated sludge process. The general form of the relation is as follows (Metcalf and Eddy, 2003):

$$\mathrm{TSS} \approx \mathrm{TSS}_f * \mathrm{T}$$

in which:

TSS = total suspended solids [mg/L]

TSS_f = factor used to convert turbidity readings to total suspended solids, in [mg/LTSS/NTU]

T = turbidity [NTU]

Metcalf and Eddy (2003) reports a value of TSS_f = 2.3 - 2.4 for settled secondary effluent and TSS_f = 1.3 - 1.6 for secondary effluent filtered with a granular medium depth filter.

For decades turbidity is used as the main water quality parameter in filtration. Generally, turbidity is defined as a measure of the light transmitting properties of water and used to indicate the quality of waste discharges and natural waters with respect to colloidal and residual suspended matter. The measurement is based on the comparison of the intensity of light scattered by a sample to the light scattered by a reference suspension under the same conditions. It is typically determined using 90º scattered light principle in compliance with EN ISO 7027 (ISO, 1999).

Within the presented investigations turbidity was applied as one indicator for the filtration efficiency regarding suspended solids removal. Usually turbidity values of < 2 NTU in the final effluent are aimed.

The measurements of turbidity were performed with Hach inline turbidity meters installed at the pilot scale dual media filter. Additional manual turbidity analyses were often performed to confirm these inline measurements. Turbidity during lab scale filtration was exclusively monitored with manual analyses using a portable Hach turbidity meter. The turbidity values were measured in Nephelometric Turbidity Units (NTU).

It should be considered that the interpretation and accuracy of the turbidity measurements could be affected by certain properties of the used samples or applied method. The results of one sample can vary depending on the used light source and the applied method of measurement (reflected versus transmitted light). Also the properties of the particles in a sample can be encountered as source for deviations. Presence of light absorbing particles, i.e. carbon fines, or particles with low light scattering ability, i.e. biological material like algaes or pathogenic micro organisms, could influence the light signal. Furthermore, the signal could be increased if a sample contains a high population of very fine particles, which have a higher light scattering ability than larger particles (Metcalf and Eddy, 2003).

The application of the same instruments and the similar method of readings ensure the qualitative comparability of measurements conducted at the different research locations.

However, neither suspended solids concentrations nor turbidity analyses can deliver information of the particle size distribution of the water samples.

4.3.2 Metal salts

During the different filtration experiments metals salts were dosed for the chemical precipitation of phosphate and coagulation and flocculation. Mainly poly aluminium chloride (PACl, PAX) and ferric chloride ($FeCl_3$) were applied. The dosing occurred either in a constant amount or with variable amount. In both cases the dosing was proportional to the orthophosphate concentration in the feed water expressed by the metal-orthophosphate ratio MePO in mol/mol. The MePO ratio was adjusted along the following guidelines:

1. The presence of ionic metal ($Al_{<0.45\mu m}$ or $Fe_{<0.45\mu m}$) and the orthophosphate concentration (PO_4-$P_{<0.45\mu m}$), determined in the 0.45 µm filtered samples, after coagulation and flocculation was maintained at very low levels (if possible under detection limit of applied method). This ensures sufficient reaction of ionic metal with orthophosphate ions and prevent overdose of the coagulant.

2. The total metal concentration ($Al_{unfilt.}$ or $Fe_{unfilt.}$) in the filtrate was maintained equal or below the concentration measured in the WWTP effluent.

The aluminium or iron content of a sample could be analysed directly with cuvette analyses supplied from by e.g. Merck and Dr. Lange.

4.3.3 Phosphorus

The fractionation of phosphorus species was applied for the evaluation of the efficiency of the precipitation, coagulation and flocculation processes and the assessment of the filtration results of the different species.

Generally phosphorus analyses at different sampling points during filtration were investigated with online analysers supplied from Hach Lange, and/or manually with Dr. Lange cuvette tests. Depending on the pre-treatment of the samples either for online or manual analyses different phosphorus species could be determined. Figure 4. 2 gives an overview of the phosphorus fractions present in water.

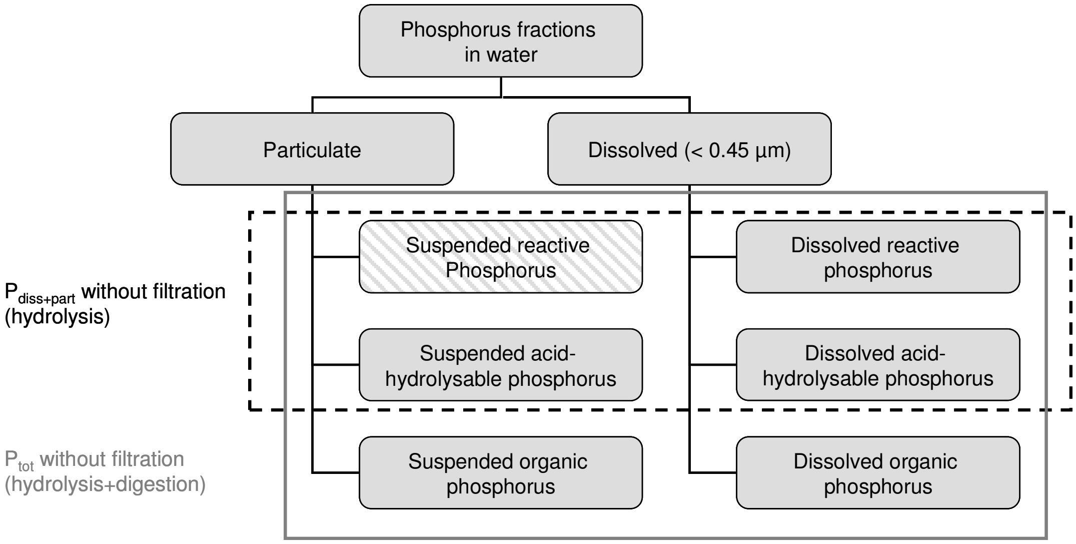

Figure 4. 2: Overview of phosphorus fractions depending on different sample pre-treatment and analysis, (Spivakov et al., 1999)

The two main fractions of phosphorus “particulate” and “dissolved” are described. The distinction between those two fractions is achieved by sample filtration over 0.45 µm. Both main fractions consist of reactive, acid-hydrolysable and organic phosphorus. The reactive and acid-hydrolysable phosphorus represent the inorganic species and are determined by sample acidification. The determination of the organic fractions requires sample digestion.

Even if this distinction is commonly known testing and interpretation of measurements often deviates from this procedure. Often in the regular monitoring on wastewater treatment plants determination of orthophosphate occurs on samples filtered over suspended solids filters. These filters do not retain all particulate inorganic phosphorus. The remaining inorganic phosphorus particles will dissolve due to acidification during sample treatment and will be measured as well as orthophosphate (PO_4-$P_{<0.45\mu m}$). Consequently the orthophosphate concentration during analyses will seem to be higher than it actually is.

In this thesis the interpretation of the phosphorus species during the different processes derived from the presented fractionation in Figure 4. 2. The detailed procedure for the measurements of the different phosphorus fractions depending on different sample pre-treatment can be found in the Appendix C.

The following inorganic phosphorus species are defined accordingly to the described procedure and are used in the following chapters of this thesis:

- P_{diss}: orthophosphate and dissolved phosphate or PO_4-$P_{<0.45\mu m}$,

- P_{part}: particulate phosphate or the difference between PO_4-$P_{unfilt.}$ and PO_4-$P_{<0.45\mu m}$,
- $P_{part+diss}$: total phosphate or P_{part} plus P_{diss}.

However, the phosphorus analyses according to this procedure ensure the determination of the phosphorus species of actual interest and leads to proper interpretation of the results during chemical and physical phosphorus removal.

For example, the inorganic particulate phosphorus (P_{part}) in contrast to the inorganic dissolved phosphorus (P_{diss}) fraction before and after coagulation and flocculation was used to determine the sufficiency of those processes. For example, a low residual concentration of orthophosphate measured may lead to two conclusions. It can either indicate the sufficiency of the applied mixing conditions during coagulation and/or confirm the appropriate type and quantity of dosed coagulant.

Also, biological phosphorus removal should be considered besides the chemical and physical phosphorus removal especially when applying biological denitrification in the filter. Under aerobic conditions free polyphosphates can be stored internally by phosphate accumulating bacteria during biological phosphorus removal applied within the activated sludge process. Also during denitrifying filtration, with anoxic conditions, phosphate uptake by present microorganisms may be expected to some extent. Phosphates present in the form of phosphate ions (PO_4^{3-}), hydrogen phosphate ions (HPO_4^{2-} and $H_2PO_4^{-}$) and phosphoric acid (H_3PO_4), are available for biological metabolism without further breakdown.

It is noted that biological phosphorus removal due to denitrifying biomass was expected to have only minor contribution to the total phosphorus removal compared to the chemical removal. Anyhow, biological removal should not be neglected when aiming at MTR-quality for total phosphorus of 0.15 mg/L.

4.3.4 Pressure readings

The accumulation of solids in the filter bed leads to increasing filter resistance. This can be monitored by pressure readings during filtration. There are two possibilities of measuring the pressure developments, (1) the increase of hydrostatic or pore pressure, or (2) the decrease of the differential pressure across the measuring element or pore. The pressure along the filter bed height, in bar or cmWC (cm water column), was observed from pressure gauges installed at the pilot scale filter and from manometers at the lab scale filters. Figure 4. 3 shows the pressure gauges combined with plug valves for sampling installed at the dual media filter at WWTP Utrecht and WWTP Horstermeer operated with a fixed water head.

Figure 4. 3: Plug valves and pressure gauges installed along the filter bed of the pilot scale dual media filter at WWTP Horstermeer

The increasing differential pressure resulting in a pressure drop, observed during pilot scale filtration, is displayed in the so-called Lindquist diagram; an example is shown in Figure 4. 4.

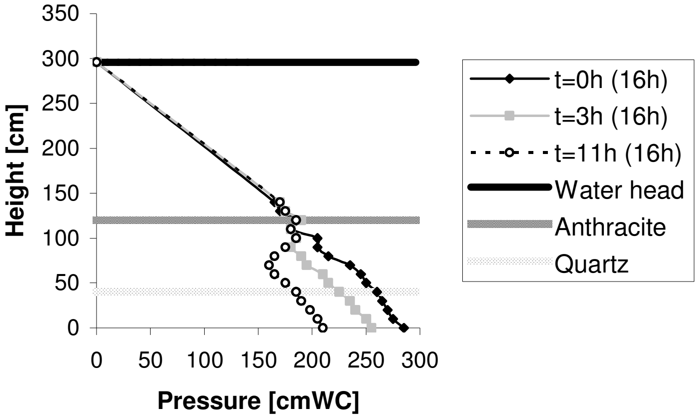

Figure 4. 4: Example of a Lindquist diagram in cmWC (cm water column) throughout the filter bed height at the pilot scale installation; t = actual run time (total run time) [h]

The pore pressure observed during lab scale filtration (Figure 4. 5) with free rising water head generally shows the pressure increase starting from the first manometer in the filter bed; approx. 2 cm below the surface of the anthracite layer. Consequently, the pressure readings are normalised to the overflow height of 2.5 m and the heights of the pressure reading points.

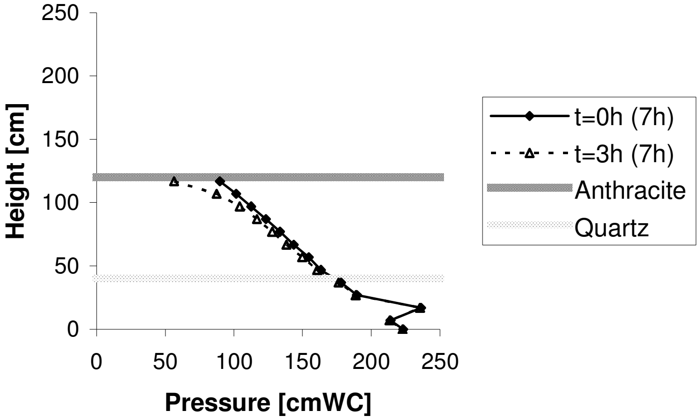

Figure 4. 5: Example of the pressure readings in cmWC (cm water column) throughout the filter bed height at the lab scale installation; t = actual run time (total run time) [h]

In general, pressure diagrams are used to identify a proper filtration process through stable and linear pressure readings. Increasing filter resistance (gradual built-up) and clogging (non-gradual built-up) is caused by accumulation of foulants in the filter bed and is discovered through pressure drops resulting in curves differing from clean bed resistance and linear development.

Not only filtered suspended matter can cause a decrease of the accumulation capacity. Also entrapped nitrogen gas (N_2) and carbon dioxide (CO_2) produced during denitrification can contribute to the clogging of the filter. This may affect the filter run time or result in adjustment of the required backwash procedure.

When the differential pressure drop exceeds a present value, the flow rate can decrease or the water head above the filter bed reaches a critical level. The filtration run must be stopped and backwash of the filter must be initiated. Even in case of flow decrease or reaching the critical water head the quality of the filtrate measured with turbidity could still be acceptable. Nevertheless, small suspended matter like phosphate flocs might break through the filter causing an increase of small particles in the filtrate. This may be detected with turbidity analyses but with a delayed response. Therefore, frequent pressure readings simultaneous to phosphorus analyses throughout the filtration run are recommended in order to identify the appropriate filtration run time and cleaning frequency.

4.3.5 DO, BOD and COD

Measurements of the Biochemical Oxygen Demand (BOD) present the relative oxygen requirements for the biochemical degradation of organic material (carbonaceous demand) in wastewaters and effluents. It is commonly determined by the difference of dissolved oxygen in a sample after 5 days at 20°C. On the other hand, the parameter COD (Chemical Oxygen Demand) indicates the oxygen demand of biodegradable pollutants plus non-biodegradable (inorganic) pollutants to be oxidised. Both parameters are expressed in mgO_2/L. However, COD is preferred for daily analysis using cuvette tests since it is inherently more reproducible, accounts for changing conditions and takes a short time to complete.

Biological denitrification occurs in the absence of dissolved oxygen (DO) and presence of sufficient rapidly biodegradable dissolved organic matter (bsCOD). This can be achieved by dosing an external carbon source. Generally, the BOD/COD ratio could be used as an indicator for the degradability of wastewaters (0: non-biodegradable; 1: easily biodegradable).

In the presented filtration tests mainly methanol was applied as carbon source due to its high content of bsCOD; with a BOD/COD ratio 0.5-0.9 (>1.2 kgCOD/L and 0.6–1.1 $kgBOD_5/L$). After start-up of the denitrification process with an increased dosing at a low filtration rate, the dosing was adjusted to the optimal dosage. During normal operation very low DO concentrations, required to avoid the deterioration of biological denitrification, were regularly observed with oxygen measurements. Furthermore, a methanol overdose resulting in high COD concentrations in the filtrate exceeding the concentrations in the WWTP effluent was prevented. This was frequently confirmed by analyses of COD.

4.3.6 Nitrogen

The nitrogen species nitrate, nitrite, ammonium and organic nitrogen together form the total nitrogen content, N_{tot}. The term organic nitrogen, N_{org}, refers to organic molecules such as proteins, amines, and amino acids which are to large extent in dissolved form and not filterable.

The organic nitrogen (N_{org}) and ammonium (NH_4-N) together present the content of Kjeldahl-nitrogen (N_{Kj} or TKN).

$$N_{tot} = \underbrace{NH_4\text{-}N + N_{org}}_{N_{Kj} \text{ or TKN}} + NO_3\text{-}N + NO_2\text{-}N$$

The concentrations of nitrate, ammonium and total nitrogen were measured regularly During the pilot investigations at WWTP Horstermeer nitrate and ammonium were analysed online (Hach-Lange) as well as manually with cuvette analyses, while total nitrogen was only determined manually.

During pilot investigations at WWTP Horstermeer the total nitrate load and conversion rate, both expressed in $kgN/(m^3_{bedvolume}.d)$, were calculated for the evaluation of the denitrification capacity.

4.4 Particle analysis as additional measurement

4.4.1 Principles

As described earlier, suspended solids analyses either directly or indirectly via turbidity measurements could not deliver information of the size of the present particles in the water sample. Particle analyses can provide this additional information. Comparisons of particle fractions with particulate phosphate (P_{part}) and turbidity may lead to extended knowledge. The surplus value of particle size analyses, in addition to conventional analyses, is one major topic of this thesis.

Basically, two different techniques are available for particle counting: light extinction (or light blocking) and light scattering (Hargersheimer and Lewis, 1995). Particle analyses presented in this thesis were conducted with the light extinction principle. Light extinction devices measure the change of light intensity caused by a particle as it passes through a light beam (Figure 4. 6). The light intensity is proportional to a voltage pulse that can be related to particle size.

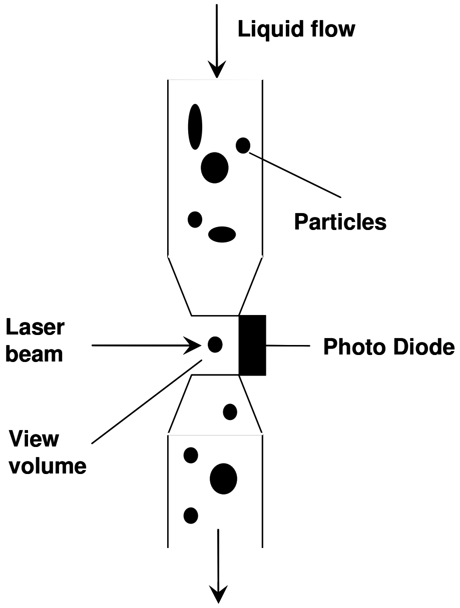

Figure 4. 6: Particle analyses with the light extinction principle

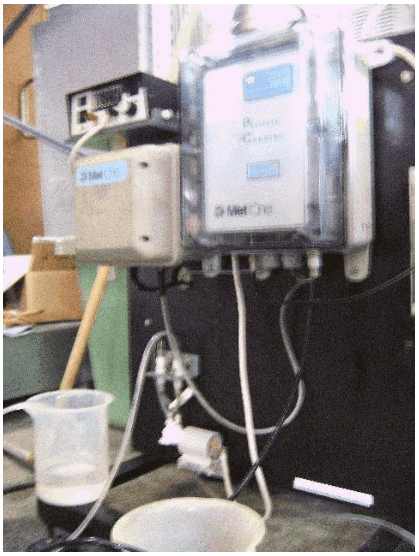

Figure 4. 7: Particle counter MetOne PCX based on the light extinction principle with a measuring range of 2-100 µm

The result is a report of the number of particles in intervals adjusted for different particle sizes as a particle size distributions (PSD). The device used, a MetOne PCX particle counter with a measuring range from 2-100 µm (Figure 4. 7), counts the particles in intervals of 0.5 µm. With the assumption of a spherical particle shape a particle volume distribution (PVD) can be calculated based on the PSD. Examples of these distributions are shown in Figure 4. 8 and Figure 4. 9.

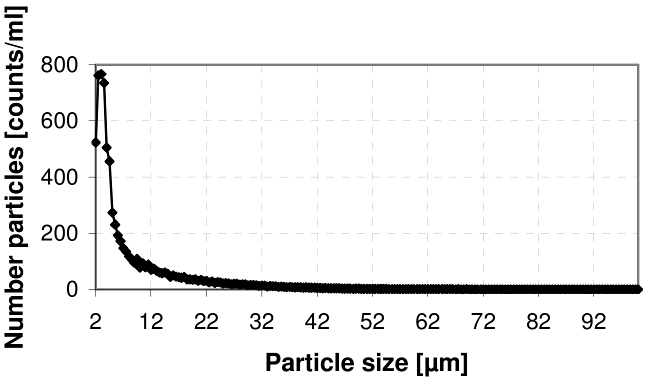

Figure 4. 8: Example of a particle size distribution (PSD)

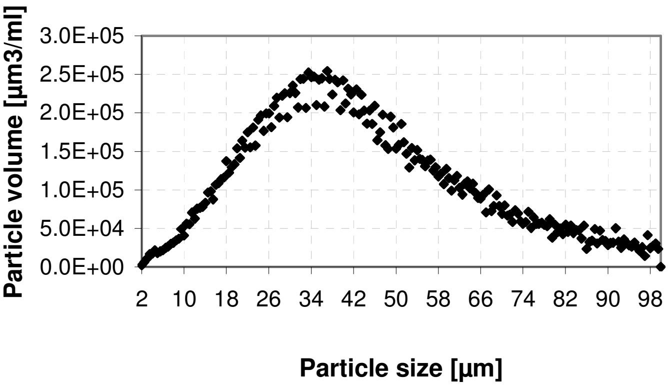

Figure 4. 9: Example of a particle volume distribution (PVD)

From a particle volume distribution (PVD) either a cumulative particle volume distribution (in $m^3_{\text{particle volume}}/m^3_{\text{sample}}$) or relative cumulative particle volume distribution (in %) versus particle size in linear or logarithmic scale can be calculated.

A cumulative particle size distribution $N(d_P)$ would always be increasing with increasing particle size, until some maximum (total number of particles in a sample) is reached.

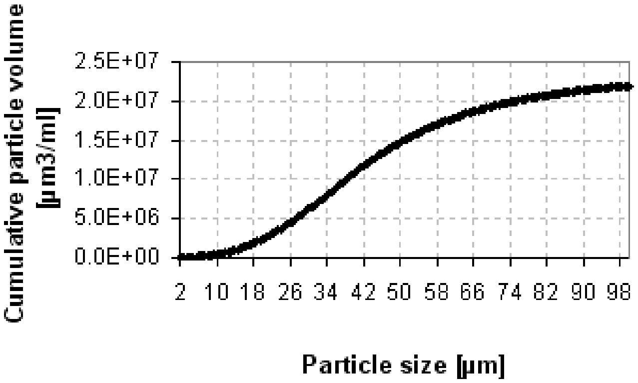

Figure 4. 10: Example of a cumulative particle volume (CPV)

The slope of such cumulative distribution ($dN(d_p)/dd_p$) is known as the particle size distribution function $n(d_p)$. The latter one can be calculated with the power law function:

$n(d_p) = Ad_p^{-\beta}$ or $\log n(d_p) = \log A - \beta \cdot \log d_p$

d_p is the nominal particle size, and A and β are constants. The power coefficient β (slope of the function) is a specific characteristic for the particle size distribution of the investigated water (Lawler, 1997; Neis and Thiem, 1997).

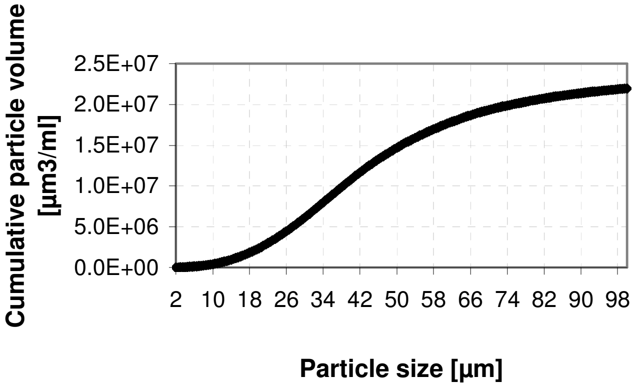

Figure 4. 11: Particle size distribution, double logarithmic

4.4.2 Current application of particle analysis

Nowadays particle analyses in water and wastewater treatment are mainly used for the assessment of malfunctioning in membrane filtration systems. In the last decade research regarding wastewater treatment was conducted to evaluate the additional value of particle counting for the monitoring of filtration performance and in relation to other measured parameters in which the relation to bacteria counts predominates.

However, the correlation with the general monitored parameter such as turbidity or particulate phosphate (P_{part}) may be an important issue for the evaluation of particle and chemical phosphorus removal processes.

The accuracy of particle counting is influenced by different factors. Besides operational inaccuracies by the user, instrumental inaccuracies may occur. Venturi effects due to narrowing within the sampling system could lead to particle break-off. Furthermore, the properties (e.g. differing from spherical shape) or concentration of particles in the sample could play a role even without exceeding the concentration limit of the analyser. In high concentrated solution, particles can be overlooked behind others while passing the measuring cell or bigger compounds could be analysed when particles appear in short distance in the measuring cell. Effluent of a wastewater treatment contains a large amount of particles of different origin and in various sizes and sample dilution may be therefore considered in order to gain more accurate countings.

In the evaluation of filtration performance the removal of the total particle volume, or cumulative particle volume, can be suitable. This value indicates the suspended solids removal efficiency indirectly through particle analyses. Also the particulate phosphate (P_{part}) removal as fraction of the suspended solids content could represent a filtration performance. A relation of these two performance indicators could be expected.

4.4.3 New applications of particle analysis

4.4.3.1 Fingerprint of WWTP effluent

Particle analysis can be used to develop a fingerprint of different samples, e.g. WWTP effluents. The signature of the distributions could lead to a qualitative interpretation regarding particle filterability and comparison of different WWTP plants. Furthermore, this fingerprint may be utilised to indicate the effectiveness of the processes like coagulation, flocculation and filtration in relation to the distribution of the original WWTP effluent.

In Figure 4. 12 the particle size distributions (PSD) for five WWTP effluents are shown. The distributions are displayed for the particles sizes 2 - 20 µm because larger particle are present in far less concentrations.

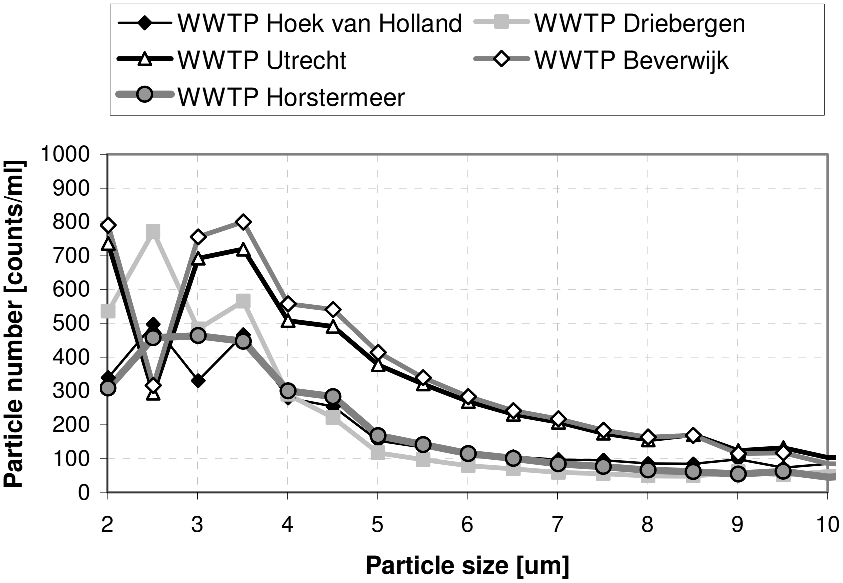

Figure 4. 12: Fingerprint of different WWTP effluents using particle size distributions

Obviously the particle size distributions of WWTP Beverwijk and Utrecht appear relatively similar. When comparing these distributions the particle concentration should be known before interpretation. The total number of particles for the effluent WWTP Hoek van Holland, Driebergen and Horstermeer is in average 4,100 counts/ml, and 6,800 counts/ml for WWTP Utrecht and Beverwijk. However, this qualitative comparison of size distributions seems weak in delivering a unique site specific fingerprint. Also other parameters were called in to emphasise a clear fingerprint. Table 4. 1 shows the square of the correlation coefficient R^2 deriving from the power law function of the double logarithmic particle size distributions and the power law coefficient β for the five effluents. The particle distributions obtained for the effluents of WWTP Hoek van Holland, Utrecht and Horstermeer result similar high values for R^2 while the values are low for the effluents of WWTP Driebergen and Beverwijk.

However, the power law coefficient β for the effluents at WWTP Beverwijk and Utrecht is higher than for the other distributions confirming the similarity of their particle size distributions, displayed in Figure 4. 12. Additional to the particle size distribution the power law coefficient delivers a good parameter of a site specific effluent.

Table 4. 1: Square of the correlation coefficients R^2 and the power law coefficient β gained from the double logarithmic particle size distributions for the five WWTP effluents

Location	R^2	Power law coefficient β
WWTP Hoek van Holland	0.97	-3.59
WWTP Driebergen	0.88	-3.37
WWTP Utrecht	0.92	-3.87
WWTP Beverwijk	0.87	-3.74
WWTP Horstermeer	0.93	-3.66

Distributions for cumulative particle volumes, even relative to the total volume account per size class, could also be determined for more specific characterisation (Figure 4. 13). All samples show a different shape of the curves, indicating increasing slopes when a high concentration of particles of a certain size is measured.

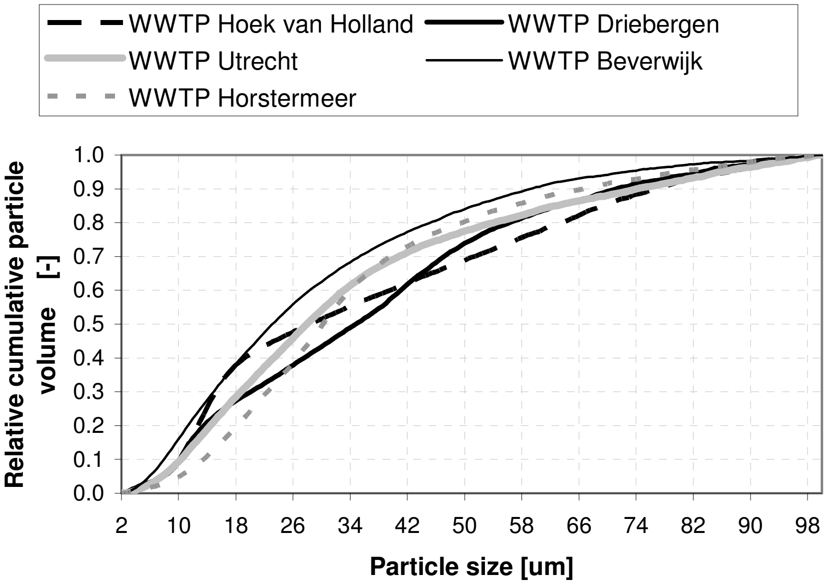

Figure 4. 13: Fingerprint of different WWTP effluents using the relative cumulative particle volume

4.4.3.2 Comparison of water samples during filtration

The efficiencies of the different processes such as coagulation, flocculation and filtration can be evaluated with particle analyses by comparing particle size or volume distributions (PSD, PVD) and the cumulative particle volume (CPV) for different sampling points. For a comparison and evaluation samples of WWTP-effluent (EF), coagulated EF (AC), flocculated EF (AF) and process filtrate (FI) may be used. In the following an example is given for the interpretation of particle analyses assessing floc growth during coagulation and flocculation.

Figure 4. 14 and Figure 4. 15 generally show an increased number and volume of particles in AC compared with the numbers and volumes in EF due to the addition of coagulant (Figure 4. 14 and Figure 4. 15).

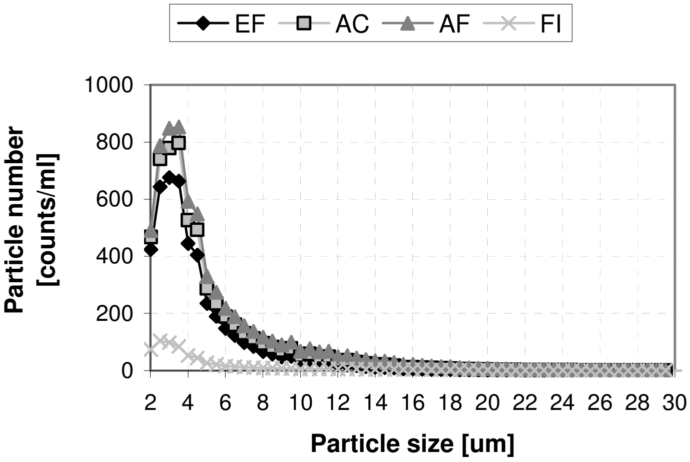

Figure 4. 14: Particle size distribution for EF, AC, AF and FI

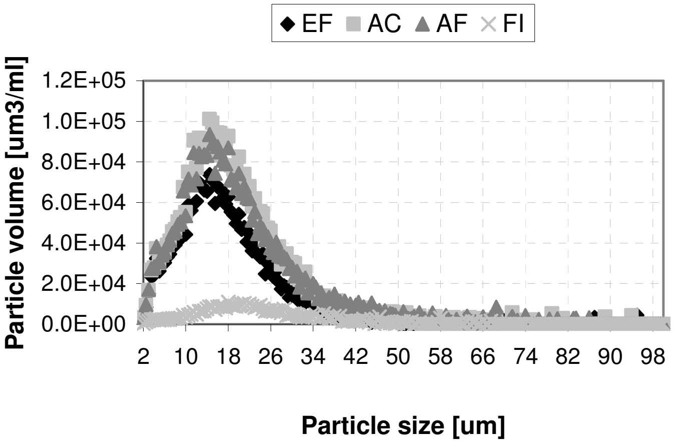

Figure 4. 15: Particle volume distribution for EF, AC, AF and FI

This increase is mainly observed for particles between 2 and 20 µm. After flocculation (AF) an expected shift from preliminary smaller particles in AC towards to larger particles is hardly visible. Only the number and volume of particles 2-20 µm increases indicating no significant floc growth. This is confirmed by the steep slope of the cumulative particle volume for this size range in Figure 4. 16. These distributions together with the filtration results of the different particle fractions can be used for the evaluation of the processes coagulation and flocculation.

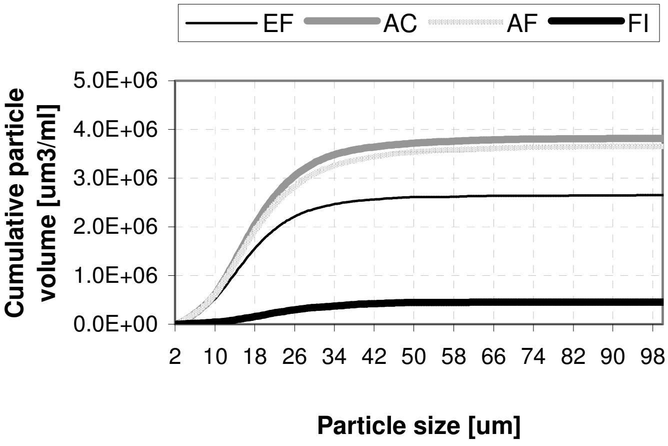

Figure 4. 16: Cumulative particle volume for EF, AC, AF and FI

The removal of the total particle counts or volume can be used as a parameter for evaluation of the total particle removal efficiency by the applied filtration process. This value can be defined as filtration efficiency or in general as a performance parameter for filtration.

4.4.4 Utilization of particle analyses

4.4.4.1 Effect of sample dilution

Particle concentration in the sample has an influence on the accuracy of the analysis itself. In case of high concentration particles can be overlooked behind others while passing the measuring cell. Or, small particles appearing close in the cell can be analysed as bigger particles. Effluent of a wastewater treatment process mostly contains a large amount of particles of different origin in various sizes and shapes. Therefore more representative particle measurements could be gained through sample dilution.

For the examination of the accuracy of counting particles, analyses of raw (undiluted) and diluted samples of the filtration process at the WWTP Beverwijk were used. Dilution was carried out with demineralised water mainly with a dilution rate of 3 (= 2 parts demineralised water and 1 part sample). The measurements of the diluted samples were used to calculate the corresponding values for the undiluted samples, including the dilution factor and subtracting of particle numbers eventually present in the dilution water.

The total particle numbers in undiluted samples versus counts measured in diluted samples (re-calculated for undiluted) over all analyses conducted at WWTP Beverwijk are shown in Figure 4. 17. A linear relation was expected and delivers $R^2 = 0.97$. Furthermore, values disperse more from linear development for total counts above 4,000 particles in the undiluted sample.

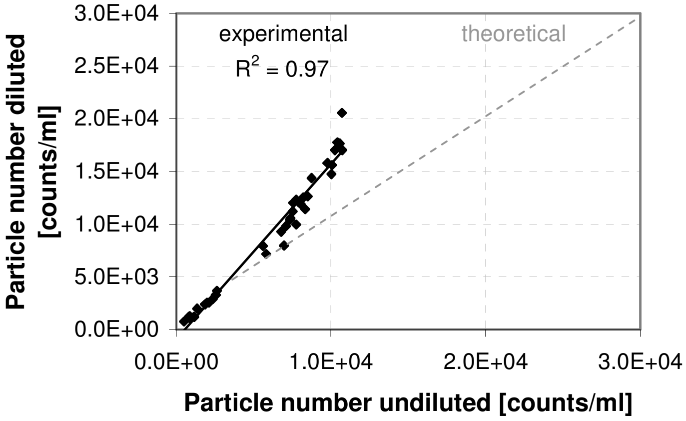

Figure 4. 17: Correlation of total particle number in undiluted versus diluted samples

It can be concluded that the dilution with demineralised water, the dilution rate and the re-calculation of diluted samples shows a good approximation of the theoretical expected relationship for particle numbers below 10,000 (1E+04) counts per ml in the undiluted samples. This corresponds with the concentration limit of the applied particle counter. The linear correlation of the particle number in the undiluted samples versus diluted samples resulted in a high square of the correlation coefficient of 0.97.

It was assumed beforehand that the sample dilution enhances the accuracy of counting small particles especially for highly concentrated samples due to minimised chance of mutual shading. A significant increase of total particle counts between 2 and 10 µm was calculated for the three sampling points EF, AF and FI. In total, 41 analyses were conducted on the undiluted samples and their dilutions. Of those the particle concentration for 9 measurements, including samples of EF and AF, exceeded the concentration limit of the particle counter (10,000 counts/ml) in the undiluted sample. For another 13 measurements, including samples EF as well as samples AF, the concentration limit was exceeded only for the diluted samples. In 19 measurements, consisting mainly of measurements in FI, the total numbers in the undiluted samples are counted below the limit. The results for different particle concentrations in the samples, partly above and below the concentration limit of the particle counter are shown in Table 4. 2.

Even if the concentration limit in the undiluted sample is not reached, sample dilution can result in a rise of max. 40% of particle counts between 2-10 µm. In the cases of measurements exceeding the concentration limit, the counts in that size range can rise by max. 56 %, indicating the reasonability of sample dilution for all sampling points. (Miska *et al.*, 2006)

Table 4. 2: Increase of number of particles 2-10 µm in recalculated diluted samples relative to undiluted samples, in %; (Miska et al., 2006)

Sample	>10,000 counts/ml in undiluted sample	<10,000 counts/ml in undiluted sample
EF	41 – 45	25 – 40
AF	36 – 56	17 – 40
FI	nd	5 – 35

However, to distinguish measurements conducted on undiluted and diluted (recalculated) samples the epithets ‘undiluted’ and ‘diluted’ will be named.

4.4.4.2 Particle number or volume as valuable parameter

For the use of particle characterisation in the evaluation of particle related processes the proper parameters should be defined. The most obvious characterisation may be with particle numbers and their removal. Nevertheless, particle volume and its removal seems a more accurate parameter. Total particle count (TPC) was plotted against the total particle volume (TPV) for the undiluted and diluted measurements of the 41 samples analysed at WWTP Beverwijk considering the total counts in the range 2-100 µm. A linear relation is expected. A linear regression over all countings is executed for each of the three samples EF, AF and FI. The measurements were selected under the conditions, that (1) a similar method and handling of sample for the particle analyses is used, and (2) only measurements of samples are compared with similar mass distribution. The latter condition was ensured by correlating analyses for each sampling point separately comparing measurements of undiluted as well as diluted samples.

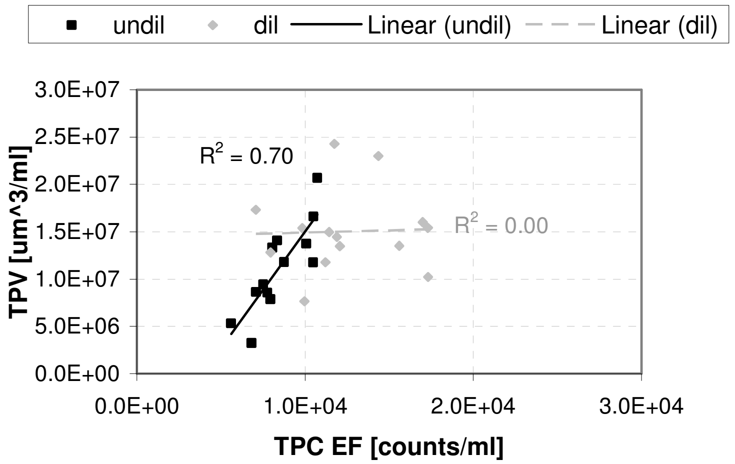

Figure 4. 18: Correlation TPC and TPV in EF (2-100 µm) for undiluted (undil) and diluted (dil) samples; (Miska *et al.*, 2006)

The linear regression of TPV versus TPC for the size range 2-100 µm for EF is displayed in Figure 4. 18. The correlation of measurements EF is higher for the undiluted than for the diluted measurements. Even if not presented here, the squares of the correlation coefficient for all analyses conducted for the samples AF and FI resulted in R^2 = 0.46/0.48 (undiluted/diluted) and R^2 = 0.75/0.78 (undiluted/diluted), respectively. Obviously, correlation increases with sample dilution for AF and FI contradictory to the analyses for EF. The highest R^2 from all values is found for FI.

However, linear regression over all measurements resulted in a poor relation.

This may indicate that an evaluation of the most appropriate parameter is difficult, either for TPC or TPV. Although Figure 4. 18 shows that several values for TPC can be related to only one specific value of TPV. The particle volume may be therefore defined as a more appropriate parameter for the assessment of particle removal processes (Miska *et al.*, 2006).

4.4.5 Relations of particle analyses versus turbidity and phosphorus

4.4.5.1 Particles versus turbidity

In other researches turbidity measurements were related generally to the total particle counts TPC. It should be considered, that the light scattering ability of a sample not only depends on the particle concentration but also on their total surface or volume. Therefore, the relations TPC and the total particle surface (TPS) versus turbidity were investigated. Figure 4. 19 and Figure 4. 20 show the linear regressions of turbidity and TPC/TPS for undiluted and diluted measurements of particles between 2 and 100 µm over all samples EF, AF and FI, representing samples of high turbidity (2.5-9.6 NTU) and low turbidity samples (0.7-3.3 NTU). The coefficient of the linear regression for TPC versus turbidity appears to be higher than for TPS versus turbidity. Anyhow, both coefficients are considered to be low.

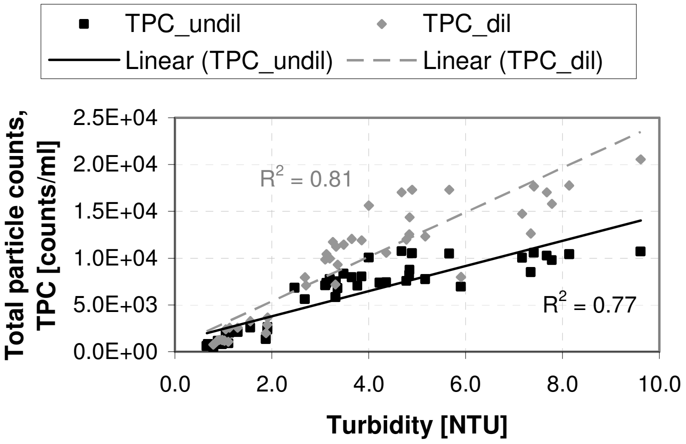

Figure 4. 19: Relation Turbidity and TPC 2-100 µm of undiluted and diluted samples

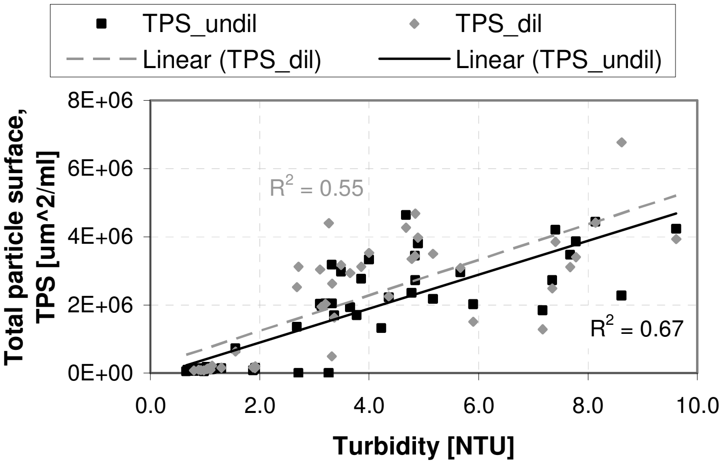

Figure 4. 20: Relation Turbidity and TPS 2-100 µm of undiluted and diluted samples

A closer look has to be given to the correlations TPC, TPS and TPV versus turbidity between the particle size range 2-100 µm and 2-10 µm for all three samples EF, AF and FI, shown in Table 4. 3. The correlation coefficients are highest for the particle size range 2-10 µm in diluted measurements. That leads to the assumption that sample dilution seems necessary and appropriate in order to receive a more representative particle counting in that lower size range. It can also be indicated that turbidity is mainly caused by particles < 10 µm. In regard to the relation of turbidity TPC gives the highest correlation followed by TPS and TPV.

Nevertheless, the relation of TPC, TPS or TPV versus turbidity are valued as weak for both undiluted and diluted samples. This could imply the necessity of additional sample analysis besides turbidity measurements. (Miska *et al.*, 2006)

Table 4. 3: Linear correlation coefficients for TPC/TPS/TPV for particles 2-100 µm and 2-10 µm versus turbidity in NTU, (Miska et al., 2006)

	R^2 for TPC versus NTU		R^2 for TPS versus NTU		R^2 for TPV versus NTU	
	undiluted	diluted	undiluted	diluted	undiluted	diluted
2-100 µm	0.76	0.81	0.67	0.55	0.61	0.37
2-10 µm	0.73	0.82	0.72	0.74	0.69	0.72

4.4.5.2 Particles versus phosphorus

In addition to the above, the relation of particulate phosphate (P_{part}) as a part of the suspended solids concentration and particle volume was investigated by comparing of their total removal. Data collected during the investigation at WWTP Beverwijk was used for this relation.

Filtration performance can be determined by (1) the cumulative particle volume removal and (2) from the particulate phosphate removal, both calculated from the difference between the concentration in AF and FI. The sample AF indicates here the flocculated WWTP-effluent that is the actual filter feed water (especially in case of coagulant-dosing) providing the actual particulate phosphate and particle volume concentration in the filter influent.

The comparison of the two performances, distinguished by different operational conditions, provides linear correlation coefficients for total particle volume TPV (2-100 µm) versus particulate phosphate removal.

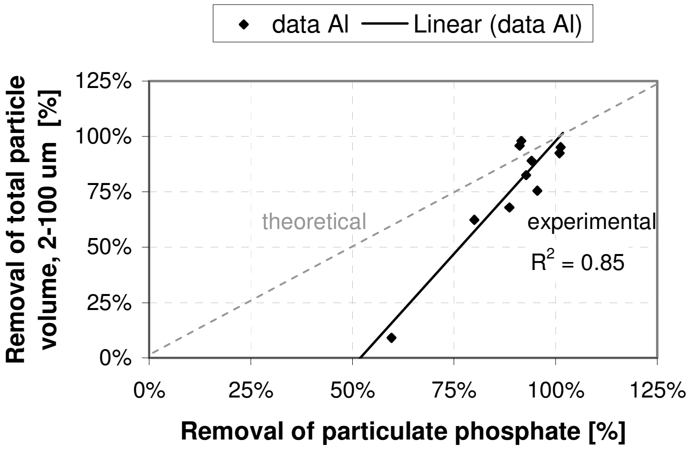

Figure 4. 21: Correlation of particulate phosphate and total particle volume 2-100 µm removal in %

The linear regression of particulate phosphate removal versus particle volume removal in percent, shown in Figure 4. 21, delivered a R^2 = 0.85 for experiments applying poly aluminium chloride as coagulant, including measurements for single flocculating and simultaneous denitrifying flocculating filtration. The correlation for all experiments during dosing of ferric chloride, single flocculating filtration and simultaneous denitrifying flocculating filtration, results in a R^2 of 0.30. The linear regression for all measurements conducted during blank and single denitrifying filtration resulted in R^2 = 0.89.

A possible explanation for the poor correlation between TPV and particulate phosphate removal for the experiments with ferric chloride might be the occurrence of floc shrinkage due to iron dosing, described by Van der Graaf *et al.* (2001), and the change in floc sizes due to mechanisms within the filtration process (Miska *et al.*, 2006). But also the formation of more small flocs below < 2 µm, not measurable with the applied particle counter, could lead to this poor correlation.

4.5 Evaluation

Several parameters, either conventional or obtained from particle analyses could be considered for the evaluation of filtration experiments applied for single flocculating filtration and denitrifying flocculating filtration. Defining sampling points valid for the research locations WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer, similar analytical methods and sample pre-treatment ensures the mutual comparability of results. Optimal process efficiencies could be achieved through proper operation and chemical dosing conditions. Evaluation requires regular monitoring assessing the processes precipitation, coagulation, flocculation, denitrification and filtration.

The evaluation of precipitation, coagulation, flocculation processes occurred with phosphorus analyses, turbidity measurements and particle characterisation. Mostly measurements for the sampling points AC or AF were compared to the reference values at EF. For optimal precipitation the concentration of orthophosphate (PO_4-$P_{<0.45\mu m}$) and dissolved metal of the added coagulant (Al^{3+} or Fe^{3+}) added, should be minimised at the entrance of the filter bed (AF). This would require a sufficient amount of metal salt dosed to form phosphate complexes avoiding the occurrence of over-dosage which results in a too high concentration of metal ions in the water phase.

Particle analyses should be conducted to observe changes in particle population during coagulation or flocculation. Analyses of EF, AC and AF would enable the monitoring of particle size or volume distributions and their mutual changes.

The fractionation of samples AF with different filter pore sizes could additionally show the distribution of particulate phosphate. The distribution and even the filterability of the fractions could be used as well as an indicator for the efficiency of coagulation, flocculation processes.

For denitrification anoxic conditions are required and the absence of free or dissolved oxygen. Dosing of an external carbon source, mainly methanol, should be sufficient for the removal of the present free oxygen and the reduction of nitrate. Refraining from an over-dosage COD concentrations in FI were maintained below the values measured in EF. The concentrations of oxygen, nitrate, ammonium and total nitrogen were measured regularly at the sampling points EF, AF and FI.

Filtration of suspended solids can be evaluated by turbidity measurements, particle analysis and phosphorus concentrations. The sampling point AF is considered as the actual sample of the process-influent for filtration, representing turbidity, particle characterisation and particulate phosphate content after the addition of coagulant. Low turbidity values < 2 NTU in FI are preferable. The removal of turbidity and particles including particulate phosphate, are examined by measurements in AF and FI.

Fractionation of total phosphate at the same sampling points can indicate the filtration results for the phosphate content and lead to steps of optimisation of precipitation, coagulation/ flocculation and filtration processes.

Several possibilities are described above applying particle analysis as additional monitoring tool for the assessment of the different processes. The relations of particle based parameters with other monitoring parameters imply its surplus value additional to conventional analyses.

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5

Filtration for the removal of suspended solids and phosphorus

5.1 Introduction

Primary filtration tests with filtration for suspended solids removal (blank filtration) and suspended solids filtration combined with chemical phosphate precipitation (flocculating filtration) were conducted only at WWTP Utrecht and Beverwijk. The application of dual media filtration for filtration of conditioned WWTP effluent and the optimal process conditions were investigated. Figure 5. 1 and Figure 5. 2 show the schedule of the filtration tests for both research locations.

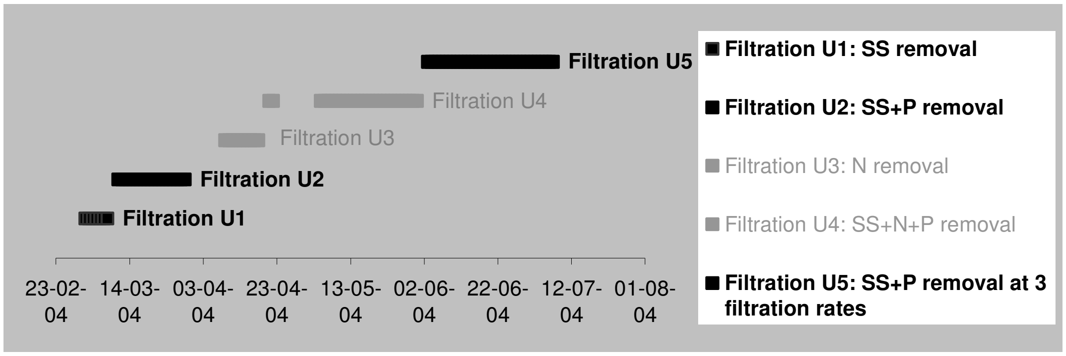

Figure 5. 1: Scheduling of filtration experiments for blank and flocculating filtration for pilot scale investigations at WWTP Utrecht

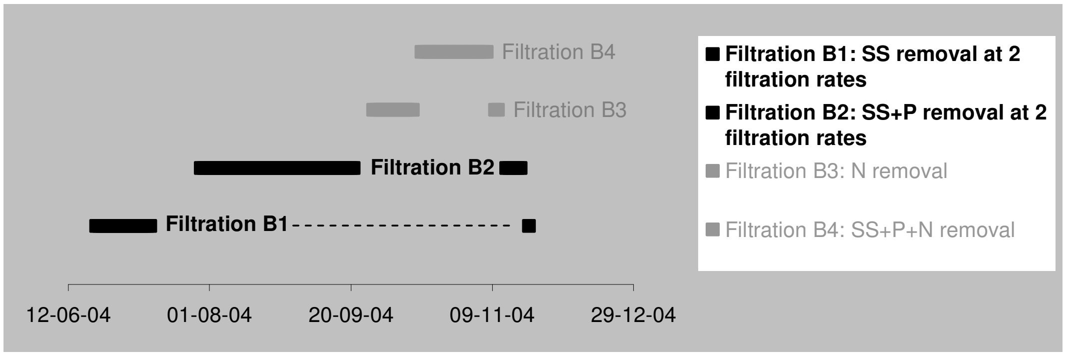

Figure 5. 2: Scheduling of filtration experiments for blank and flocculating filtration for pilot scale investigations at WWTP Beverwijk

The goal of the flocculating filtration tests was the removal of phosphorus down to MTR quality while simultaneously removing suspended solids. Together with blank filtration tests they form the reference filter performance for the investigations into simultaneous nutrient removal, which are described in chapter 6.

The following chapter describes the operational conditions for both locations like dosing conditions of coagulants, filter run time and backwash procedures, and the results including turbidity, phosphorus and particle analyses. The chapter will conclude with an evaluation of the investigations.

5.2 WWTP Utrecht

5.2.1 Dosing conditions

Preliminary jar tests with the effluent from the WWTP Utrecht indicated similar performance for ferric chloride ($FeCl_3$) and poly aluminium chloride (PACl). Metcalf and Eddy (2003) report an operating region for metal salts (referring to adsorptive coagulation): pH = 7 – 9 for iron, and pH = 5 – 7 for alum. The operating region for poly aluminium chloride is even up to a pH of 9. The average pH of the WWTP effluent varied around 7.0. That is within the optimal operational region for poly aluminium chloride and at the lower limit for iron. The most optimal coagulant was not clearly identified with jar tests, the filtration tests were therefore performed with both coagulants. The applied products and dosages of metal salts enhancing the phosphorus precipitation, coagulation and flocculation of suspended solids, were

- Poly aluminium chloride (PACl) with a constant dosage of 1-2.5 mg Al^{3+}/L, or
- Ferric chloride ($FeCl_3$) with a constant dosage of 1-3 mg Fe^{3+}/L.

The filtration results with both products were compared with the results gained from the preliminary jar tests and led to adjustment of the dosing in order to achieve the best result for phosphorus removal. The amount of coagulants dosed was adjusted manually on a daily basis, to keep a constant molar ratio with the orthophosphate ($PO_4\text{-}P_{<0.45\mu m}$) concentrations in the WWTP effluent. Daily fluctuations of the orthophosphate could have resulted in over or under dose of metal salt in the period between the adjustments.

5.2.2 Filter run time and backwash

Table 5. 1 presents the backwash procedure for the pilot scale dual media filter operated for suspended solids filtration and flocculating filtration. The backwash procedure BW1 consists of water backflush with filtrate from the filtrate buffer with varying velocities, and air backflush.

Table 5. 1: Backwash procedure BW1 for pilot scale filters at WWTP Utrecht

	Scale	Phase 1	Phase 2	Phase 3	Phase 4	Phase 5
Time	[sec]	120	60	180	60	180
Air	[-]	Off	Off	On	Off	Off
Water	[-]	On	On	On	On	On
Blower*	[Nm^3/h]	-	-	20	-	-
Velocity	[m/h]	90	10	10	30	90

*blower operation: 60 sec with 20 Hz

Figure 5. 3 shows the pressure readings of the blank filtration at the WWTP Utrecht with the applied backwash procedure BW1 (Table 5. 1), a filtration rate of 10 m/h and 24 hours filter run time. Obviously the pressure drop after 18h (24h) is not high and the pressure profile is still linear compared to the measurement in the beginning of the filter run time at 0h (24h).

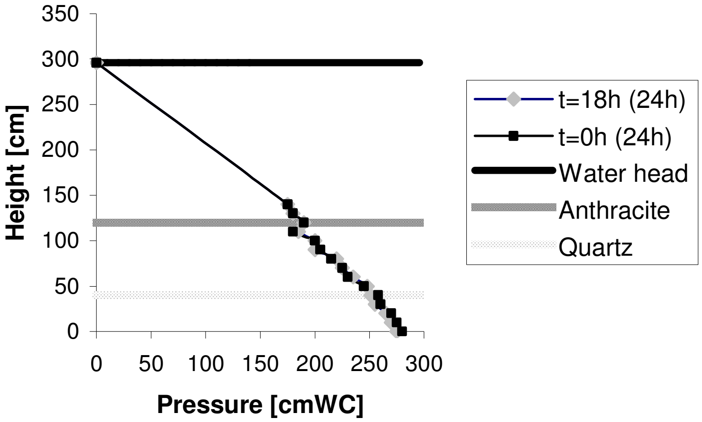

Figure 5. 3: Pressure readings at WWTP Utrecht with applied backwash procedure BW1, a filter run time of 24 hours without dosing (blank filtration) and a filtration rate of 10 m/h

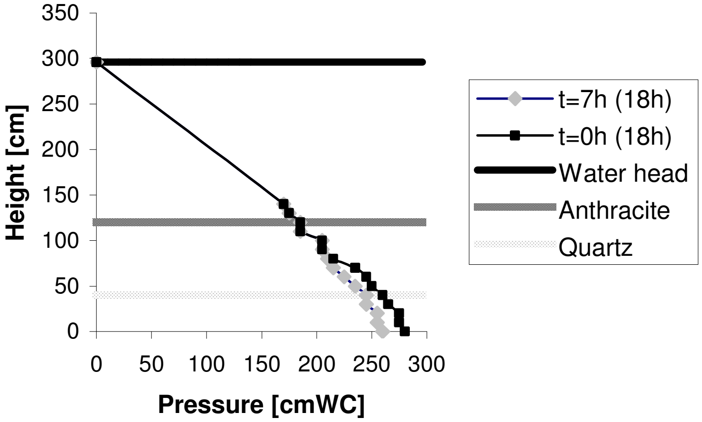

Figure 5. 4: Pressure readings at WWTP Utrecht with applied backwash procedure BW1, a filter run time of 18 hours with a constant dosing of 1.2 mgAl/L and a filtration rate of 10 m/h

The increase of suspended solids caused by the poly aluminium chloride dosing on the pressure development is shown in Figure 5. 4, with a constant dosing concentration of 1.2 mgAl/L for a filter run time of 18 hours with 10 m/h filtration rate. The pressure drops within 7 hours of the filter run but the development of the pressure can be still considered as linear.

The data curve “t=0h” indicates the first reading after completed backwash and restarted filtration run after 30 minutes. Comparison of the pressure developments “t=0” for both filtration tests indicates reversibility of the starting pressure and the sufficiency of the applied backwash procedure.

However, the optimal filtration run time for the performed flocculating filtration tests was obtained between 7 and 18 hours. Several tests with run times up to 24 hours showed premature breakthrough of suspended solids.

5.2.3 Monitoring parameters

5.2.3.1 Turbidity and phosphorus

During the pilot scale investigations at the WWTP Utrecht phosphorus removal though flocculating filtration was investigated at filtration rates of 8, 12.5 and 17 m/h. Figure 5. 5 shows the online phosphate measurements in the WWTP effluent (EF) and the filtrate (FI) during single flocculating filtration with poly aluminium chloride (PACl) and a constant dosing of 2.0 mgAl/L. The online analyses of phosphate for both sampling points were conducted on the filtered samples (<0.45µm) resulting in the presented orthophosphate concentrations (PO_4-$P_{<0.45\ \mu m}$).

The concentration of orthophosphate in the filtrate FI appears averagely below 0.2 mg/L and that almost independent from the concentration for EF and the applied filtration rate. The average value of orthophosphate in the WWTP effluent is approximately 0.5 mg/L with some peaks up to 1.0 mg/L. Due to constant amount of coagulant dosed the average metal-orthophosphate ratio of 1.5 mol/mol, the actual molar ratio varied between 0.8 to 7.5 mol/mol between the dosing adjustments.

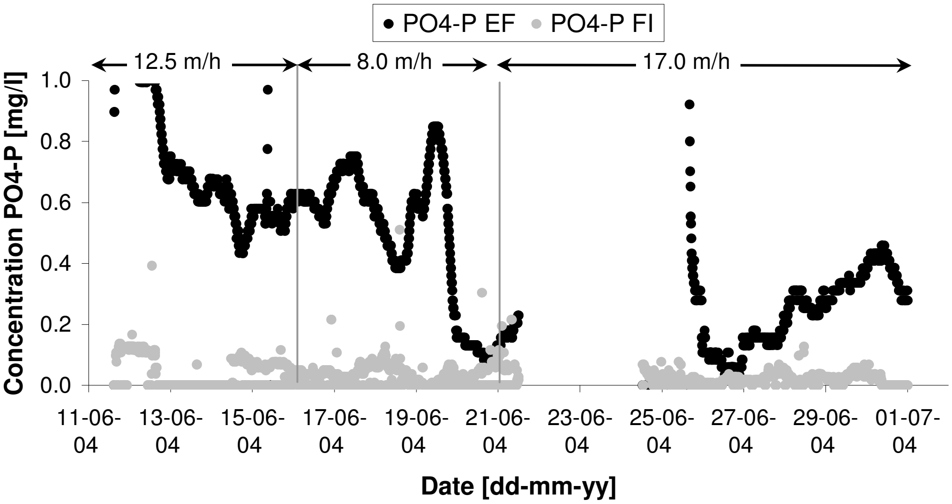

Figure 5. 5: Orthophosphate concentrations during single flocculating filtration with a pilot scale dual media filter at WWTP Utrecht applying filtration rates of 12.5, 8.0 and 17.0 m/h between 11-06-'04 and 29-06-'04

Parameter like water head above the filter bed, flow rate, temperature and turbidity for EF and FI are recorded in the data logger of the filter installation for frequent monitoring. The results for the selected period are displayed in Figure 5. 7 and Figure 5. 6.

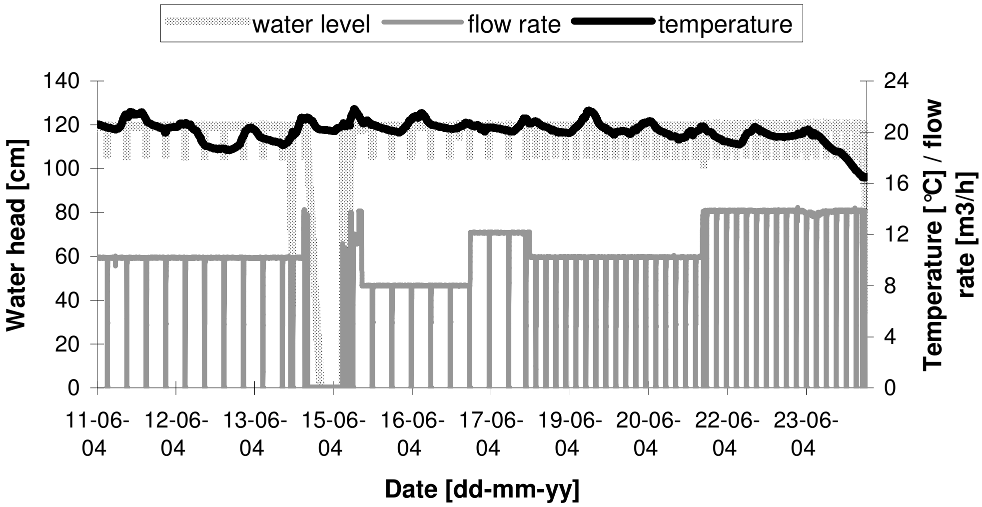

Figure 5. 6: Water head, temperature and flow rate during single flocculating filtration with a pilot scale dual media filter at WWTP Utrecht applying filtration rates of 12.5 m/h, 8.0 m/h and 17.0 m/h between 11-06-'04 and 25-06-'04

The temperature of the WWTP effluent (Figure 5. 6) varied between 16 and 22 °C.

The development of the turbidity measurements (Figure 5. 7) for EF regularly shows increasing values to above 10 NTU. Fouling of the turbidity meter due to present organic matter in the WWTP effluent, like e.g. activated sludge particles, was observed and led to increased readings. Maintenance during operation, like increased cleaning frequency and recalibration needed more attention.

However, the turbidity development in the FI during filter run time was found to be more reliable and less affected by fouling due to lower content of organic matter. The turbidity values did not vary significantly with different filtration rates applied and were almost always observed below 2 NTU.

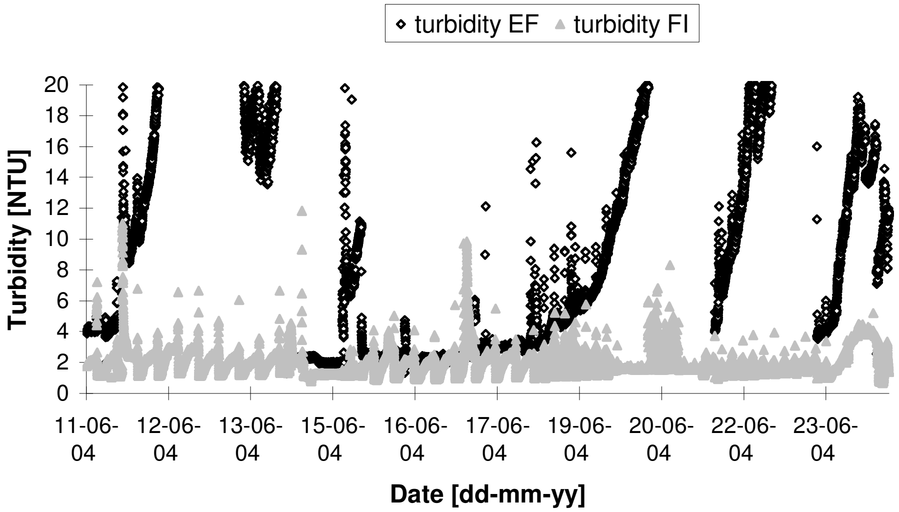

Figure 5. 7: Turbidity of EF and FI during single flocculating filtration with a pilot scale dual media filter at WWTP Utrecht applying filtration rates of 12.5 m/h, 8.0 m/h and 17.0 m/h between 11-06-'04 and 25-06-'04

Additional manual turbidity measurements, accompanied with regular sampling on the treatment plant and pilot installations, resulted in an average turbidity of 2.4 ± 1.8 NTU for the WWTP effluent and averagely 1.0 ± 0.8 NTU for the filtrate of the dual media filter.

Regarding the removal of turbidity and phosphorus during flocculating filtration the observed optimal filter run time is 16h. The increase of the filter run time above 16 hours led to breakthrough of suspended solids.

5.2.3.2 Particle analyses

During this research period a few particle analyses were conducted in addition to the other monitored parameters for the investigation of a possible additional value of this technique as monitoring tool. Especially particle distribution changes during the coagulation, flocculation and filtration process were interesting for investigation. The analyses were done with the before described MetOne PCX100 particle counter, counting particles between 2 and 100 µm in intervals of 0.5 µm.

The comparison of samples, WWTP effluent after coagulation (AC) and after flocculation (AF), with the original WWTP-effluent (EF) was meant to indicate the efficiencies of these processes. The addition of coagulant resulting in an addition of suspended solids should result in an increased number of particles after coagulation. Van der Graaf *et al.* (2001) refers to 1mg Fe/L or 1mgAl/L causing an amount of 1.9 mgSS/L and 3.0 mgSS/L, respectively. Furthermore a shift towards bigger particles after flocculation is expected before the flocculated effluent enters the filter bed. In the following particle size distributions are only displayed for particles between 2-30 µm; larger particles in the filter feed water are present in less amounts and are filtered completely during normal filtration.

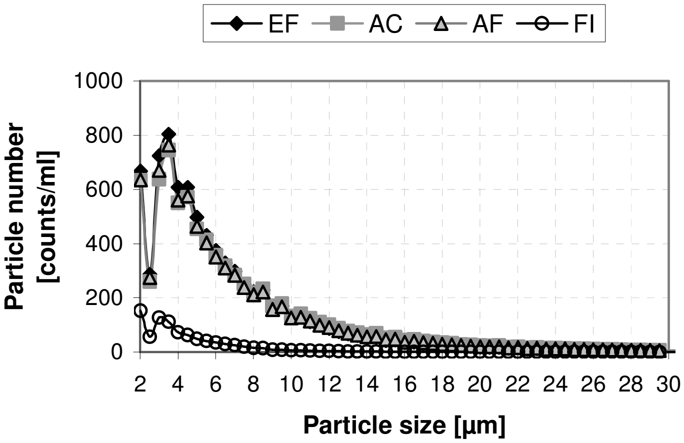

Figure 5. 8: Particle number distribution of the sampling points EF, AC, AF and FI for a constant dosing of 1-1.5 mgAl/L

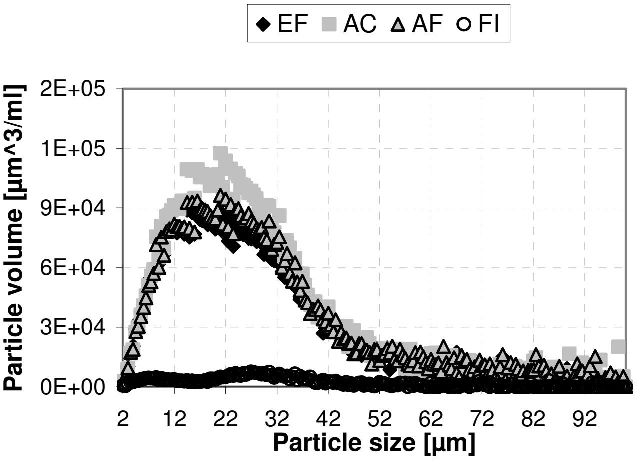

Figure 5. 9: Particle volume distribution of the sampling points EF, AC, AF and FI for a constant dosing of 1-1.5 mgAl/L

However, the few analyses at the WWTP Utrecht indicated almost no increase of particle number and volume after coagulation and flocculation compared to the WWTP effluent. This observation was made during the filter operation with a constant low dosing of 1-1.5 mgAl^{3+}/L using poly aluminium chloride as coagulant (Figure 5. 8 and Figure 5. 9). This indicates that the dosed amount of coagulant does not significantly increase the concentration of suspended solids. No shift towards bigger particles could be reported, which leads to the conclusion of a poor flocculation. Similar results were found for a dosing of 1 $mgFe^{3+}/L$ as ferric chloride. Although, the removal of turbidity and total phosphorus, and especially the phosphate particulates formed prior to precipitation, were high what indicates that the formation of small flocs did not hinder suspended solids and phosphate removal.

The comparison of particle size and volume distribution of FI with the actual filter feed water AF generally indicated the particle fraction of 2-10 µm to be the most critical fraction in the filtration process.

5.3 WWTP Beverwijk

5.3.1 Dosing conditions

The jar tests, conducted prior to the lab scale filtration tests at WWTP Beverwijk and presented in chapter 3, indicated ferric chloride as optimal coagulant for chemical phosphate precipitation. The relatively high pH of 7.5 measured in the WWTP effluent lies well within the operational region of iron salts (Metcalf and Eddy, 2003). The effluent contained averagely 0.30 mgFe/L, of which 0.15 mgFe/L in dissolved and 0.15 mgFe/L in particulate form. The total aluminium concentration was determined to 0.05 mgAl/L, with 0.03 mg/L and 0.02 mg/L present in dissolved and particulate form.

In the following filtration tests, ferric chloride ($FeCl_3$) and poly aluminium chloride (PAC) were used as coagulant added in a constant amount of 2-4 mgFe/L or 2-3 mgAl/L in order to deliver the proper choice of coagulant. The dosing amounts resulted in metal-orthophosphate ratios of 0.9-1.9 $Al/PO_4\text{-}P_{<0.45\mu m}$ and 1.5-3.1 $Fe/PO_4\text{-}P_{<0.45\mu m}$. A filtration rate of 10 m/h was applied.

5.3.2 Filter run time and backwash

The backwash procedure for the lab scale filters at WWTP Beverwijk is shown in Table 5. 2. The procedure consisted of 4 phases using air and tap water for backflush. This procedure is partly adapted from the procedure used for the prior pilot scale tests at WWTP Utrecht with some modifications considering the smaller scale of the filters.

In the first phase the filters were backflushed only with air. This aimed to break the filter bed and to separate filter grains which might be adhered by biomass (“mud balls”). The following three water backflush phases with decreasing velocities removed the foulants and caused a proper stratification of the two filter layers. The duration of the last two backflush phases was investigated by observation of the time needed to remove the foulants through the filter overflow and achieve a clean water head above the filter bed.

Table 5. 2: Backwash procedure BW1 for lab scale filters at WWTP Beverwijk

	Unit	Phase 1	Phase 2	Phase 3	Phase 4
Time	[sec]	180	300	180	180
Air	-	On	Off	Off	Off
Water	-	Off	On	On	On
Blower	[Nm^3/h]	1-2	-	-	-
Velocity	[m/h]	-	75	40	15

Due to complete manual operation of the installation the filter performance could only be observed during working hours of the treatment plant. This resulted in a filter backwash frequency of twice a day, allowing a filter run time of seven hours during day time. After the first backwash analyses of conventional parameters were performed. The filter is left after the second backwash without monitoring for 17 hours. To observe the filter performance after 17 h, first analyses during day time were conducted before the first backwash.

The pressure readings of the lab scale columns with free rising water table in Figure 5. 10 and Figure 5. 11 show almost linear development of the pressure along the filter bed height with the highest increase of pressure occurs in the uppermost 10 to 20 cm of both filtration layers. For blank filtration the highest practical filter run time without decrease of filter performance for suspended solids removal was found to be 24h. The experimentally established filter run time range for flocculating filtration was obtained between 7 and 17 h; the exact run time could not be defined due to working hours at the WWTP.

The data curves “t=1h” for both filtration tests indicate recovery of pressure after backwash resulting in similar pressure developments after 1-1.5 hours filter run time. It may be concluded that the backwash procedure was sufficient in order to reach the original clean bed resistance. Comparable effluent quality achieved during different filtration runs at WWTP Beverwijk confirmed the effectiveness of the applied backwash procedure.

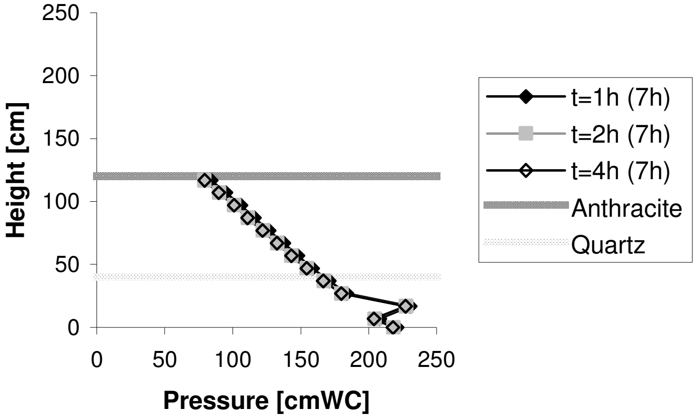

Figure 5. 10: Pressure readings (with a rising water table) at WWTP Beverwijk with a filter run time of 7 hours and a filtration rate of 10 m/h

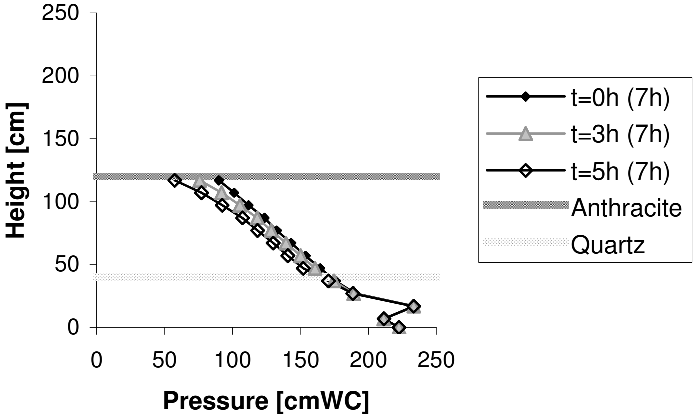

Figure 5. 11: Pressure readings (with a rising water table) at WWTP Beverwijk with a filter run time of 7 hours, with a constant dosing of 4.0 mgFe/L and a filtration rate of 10 m/h

5.3.3 Monitoring parameters

5.3.3.1 Turbidity and phosphorus

Manual turbidity measurements during lab scale filtration tests at the WWTP Beverwijk resulted in an average turbidity of 4 NTU for the WWTP effluent (EF), 7 NTU for the flocculated EF (AF) and 2 NTU for the dual media filtrate (FI). Filtrate turbidity values measured in the first hour of a new filtration run exceeded 2 NTU. This is explained by small particles breaking through the filter more easily in the beginning of filter run than in progressed filter run time, when ripening occurs. In time captured particles will result in an extra filtration effect and the filtration results are improving during filter run time.

The turbidity in FI seems to increase with increased filtration rate from 10 to 20 m/h, and especially when ferric chloride was used as coagulant (Figure 5. 12).

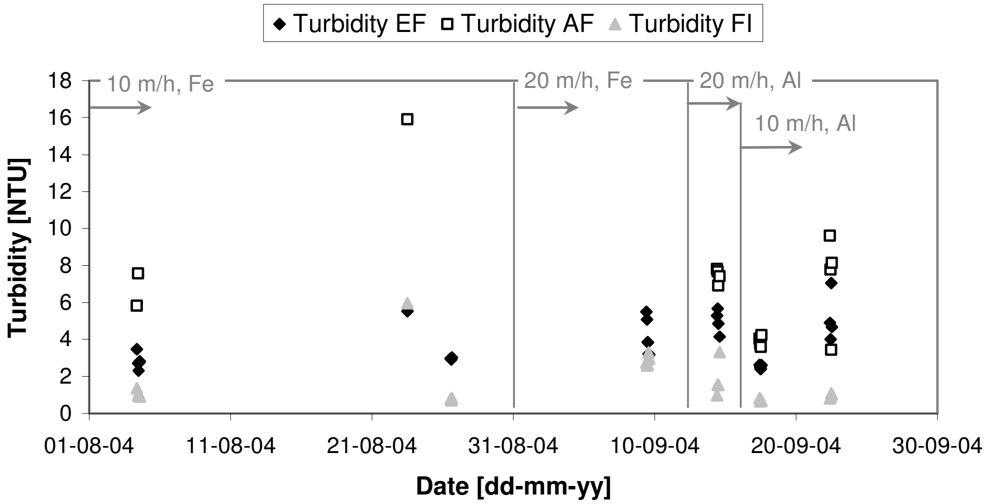

Figure 5. 12: Manual turbidity measurements during simple filtration and single flocculating filtration on lab scale at the WWTP Beverwijk between 01-08-'04 and 30-09-'04 (EF: WWTP effluent; AF: flocculated EF; FI: filtrate dual media)

The manual phosphate measurements of unfiltered samples EF and FI displayed in Figure 5. 13 show similar poor removal of total phosphate, like described before for turbidity, in the first hour after backwash. This occurred most probably due to the breakthrough of small phosphate flocs. After one hour of filtration phosphate removal improved resulting in filtrate concentrations of 0.15 - 0.25 mg/L.

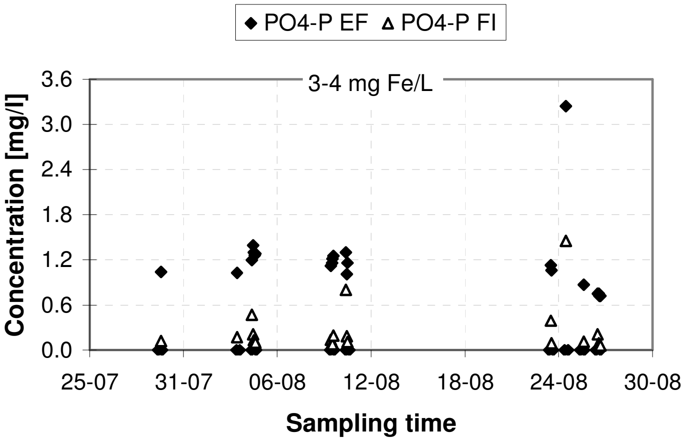

Figure 5. 13: Manual total phosphate measurements for EF and FI in lab scale installation during flocculation filtration with ferric chloride; 3-4 mgFe/L

Phosphate fractionations were performed to investigate the distribution of particulate phosphate and orthophosphate concentrations present during filtration and to evaluate the overall removal of particulate phosphate during the tests. Following measurements were conducted for the sampling points EF, AF and FI:

- Total phosphate (PO_4-P), determined for unfiltered sample to determine the particulate phosphate plus orthophosphate ($P_{part} + P_{diss}$), and
- Orthophosphate of the 0.45 µm filtered samples (P_{diss}).

The particulate phosphate fraction (P_{part}) can be then calculated as the difference of both phosphate measurements.

Figure 5. 14 shows the average concentrations of all phosphate measurements divided into particulate and dissolved fractions conducted for the sampling points EF, AF and FI. This overview includes the results, obtained during filtration tests conducted without coagulant addition (blank filtration), and with prior ferric chloride or poly aluminium chloride dosing. The average values of more than 60 measurements for each test condition are given with their standard deviations.

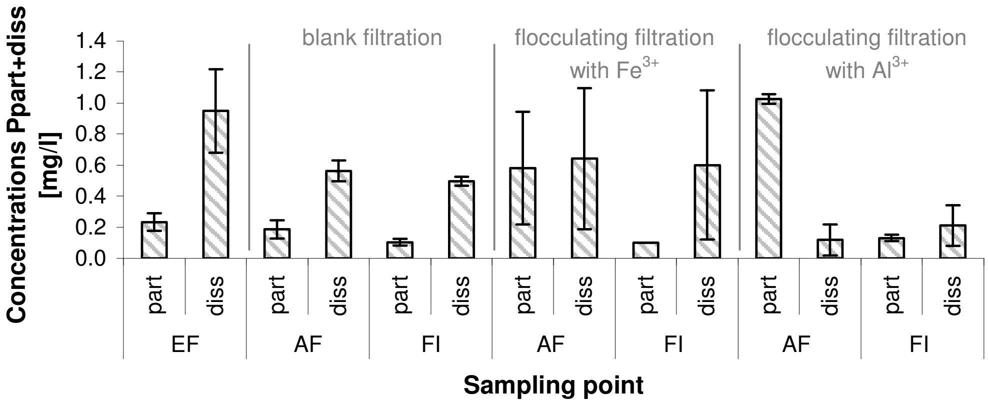

Figure 5. 14: Fractionation of average particulate and dissolved phosphate in EF, AF and FI with standard deviation (±) for blank and flocculating filtration using ferric chloride or poly aluminium chloride conducted at WWTP Beverwijk

During blank filtration, phosphate measurements (P_{part} and P_{diss}) of the EF sample differ from the measurements of the AF sample, taken at the filter directly above the anthracite layer. Flocculation of phosphate with matter able to settle between those two sampling points, adsorption to suspended matter or uptake by micro organisms could provide an explanation for this difference. However, the concentration for $P_{part+diss}$ (unfiltered PO_4-P) in AF measured for the three operating conditions were in a similar range; approximately 0.75 mg/L for blank filtration, 1.2 mg/L for flocculating filtration with ferric chloride dosage and 1.1 mg/L for flocculating filtration with poly aluminium chloride.

Generally, a maximum of 20% of $P_{part+diss}$ is removed during blank filtration. This can be mainly attributed to the filtration of P_{part}.

When comparing the measurements during flocculating filtration using ferric chloride or poly aluminium chloride, considering the concentration at AF as actual feed concentration, differences of the phosphate distributions were found. The dosing of ferric chloride seems to results in averagely less P_{part} after completed flocculation than poly aluminium chloride. This could indicate a sub-optimal initial coagulation and flocculation process. This does not correspond with the results of the jar tests (see Chapter 3), in which ferric chloride resulted in a good reaction with orthophosphate.

The standard deviations of 0.36 for P_{part} and 0.46 for P_{diss}, indicate also a high dispersion around the average values in AF when ferric chloride is applied.

The concentration of P_{diss} in FI after flocculating filtration with ferric chloride was averagly not significantly lower than the concentration in AF. Even if the incidental instability of the ferric chloride solution could be considered inhibiting the formation of metal-orthophosphate particulates, also no further coagulation and flocculation occurs within the filter bed.

On the other hand the average concentration of P_{part} is significantly lower after filtration with prior ferric chloride dosing than with poly aluminium chloride. Furthermore, the concentrations of P_{part} in FI are comparable for both coagulants.

Generally, Figure 5. 14 indicates much smaller standard deviations for the analyses of samples P_{part} and P_{diss} in samples of AF and FI during filtration with poly aluminium compared to ferric chloride. A possible explanation is a more stable filtration with less variable measurements. However, the average concentration of P_{part} is a little less for filtration with ferric chloride than with poly aluminium chloride.

Considering the removal of total phosphorus to MTR level (<0.15 mgP_{tot}/L) the organic phosphorus concentration should be encountered. Regular manual measurements indicated a maximum concentration of 0.2 mgP_{org}/L in the unfiltered sample of which 0.1 mgP_{org}/L was analysed as dissolved or particulate organic phosphorus.

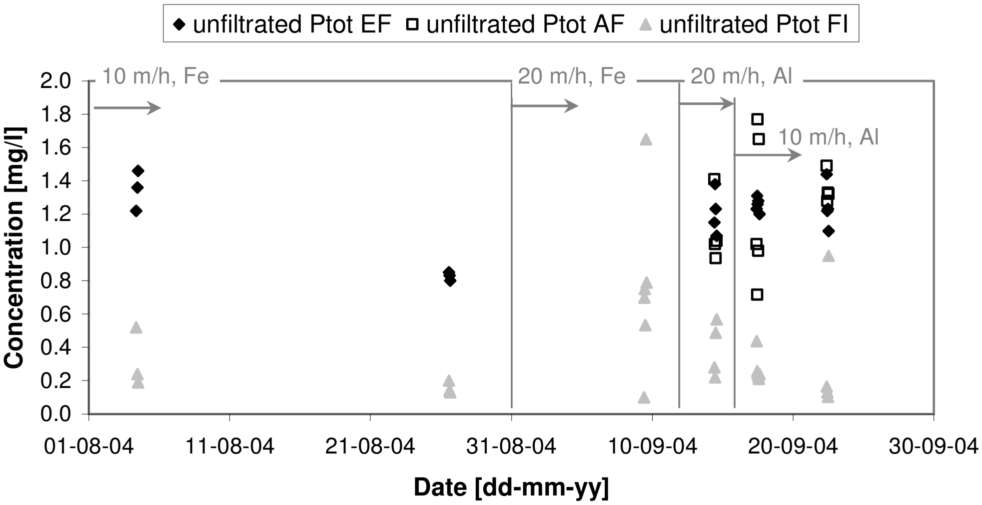

Figure 5. 15: Manual P_{tot} measurements of unfiltered samples during filtration and single flocculating filtration on lab scale at the WWTP Beverwijk between 01-08-'04 and 30-09-'04 (EF: WWTP-effluent; AF: flocculated EF; FI: filtrate dual media)

Figure 5. 15 displays the total phosphorus measurements (P_{tot}) of unfiltered samples for the sampling points EF, AF and FI. Averagely 0.4 mgP_{tot}/L was analysed in the filtrate, calculated over all experiments operated as flocculating filtration. Like described before, analyses conducted in the first hour of a new filtration run after completed backwash resulted in increased concentrations. The average total phosphorus concentration of the filtrate was calculated to 0.2 mg/L, without the results of the first hour of run time.

Generally it can be concluded that low filtrate concentrations of less than 0.2 mgP_{tot}/L can only be achieved when the concentrations of total phosphorus in the WWTP effluent do not exceed 0.8 mgP_{tot}/L and a filtration rate of 10 m/h is applied. The phosphorus removal show comparable results for either ferric chloride or poly aluminium chloride applied as coagulant. A higher filtration rate of 20 m/h led to less good phosphorus removal results.

5.3.3.2 Particle analyses

During lab scale experiments at WWTP Beverwijk the additional value of particle counting as monitoring tool was investigated more intensively than during the prior filtration tests at WWTP Utrecht.

Earlier research at the WTTP Utrecht showed problematic removal of particles in the range of 2 to 20 µm. The particles that showed to be most critical to filtration were indentified in the range of 2 and 10 µm. Therefore the results of the particle countings at WWTP Beverwijk were fractioned. Because of the effect of dilution resulting in more accurate counting of small particles, described in chapter 4, only measurements on diluted samples were considered and will be shown. The particle distributions of diluted samples were recalculated to present values for undiluted samples, while taking into account the dilution ratio and the amount of particles present in the dilution water. Still the term ‘diluted’ sample will be used for the description of the results. The detailed results of the particle analyses can be found in the Appendix D.

Figure 5. 16 displays the cumulative number of particles for the fractions 2-10 µm, 10-20 µm and 20-100 µm for samples of EF, AF and FI during blank filtration, flocculating filtration with ferric chloride and flocculating filtration with poly aluminium chloride. The figures show average values over all experiments conducted either as blank or flocculating filtration. In total 22 measurements of EF, 6 for blank filtration, 10 for flocculating filtration with ferric chloride and 8 for flocculating filtration with poly aluminium chloride, were used to calculate the average values.

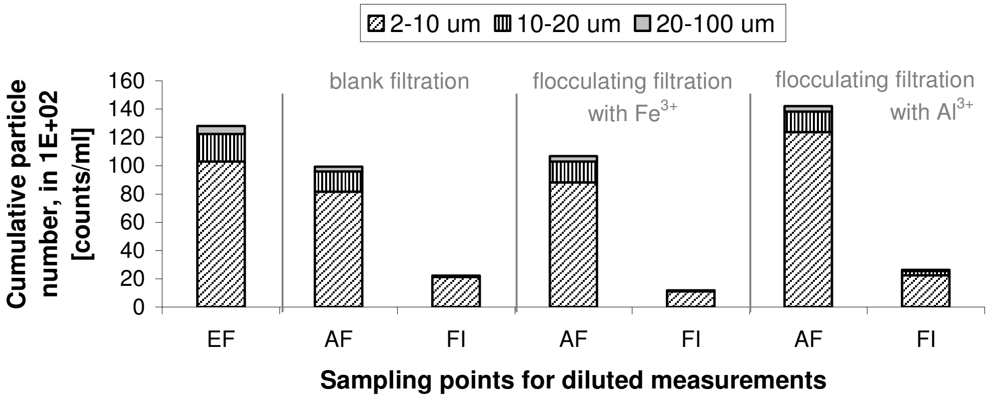

Figure 5. 16: Cumulative particle number for diluted samples EF, AF and FI conducted during blank filtration and flocculating filtration with ferric chloride ($FeCl_3$) or poly aluminium chloride (PAC) at WWTP Beverwijk from the cumulative particle number, in [1E+02 counts/ml]

A slight decrease of particle numbers of 2-100 µm can be observed comparing the sampling point EF and AF in Figure 5. 16 obtained for blank filtration and flocculating filtration with ferric chloride ($FeCl_3$). This possibly occurred due to e.g. flocculation of suspended matter and settling in the buffer tank, providing the feed water for the lab scale columns. Similar observations were made for particulate phosphate concentration (Figure 5. 14), showing decreasing concentrations between the EF and AF. On the other hand the total particle number for AF during flocculating filtration with poly aluminium chloride (PAC) increases significantly.

Even if the total particle numbers of AF for the three filtration tests seems to vary, the distribution of the three particle fractions does not change exceptionally with or without dosing of coagulant. Again, no shift towards bigger fractions is visible in the distributions AF compared EF. The number of small particles 2-10 µm, which is described earlier as problematic for filtration, appears as the biggest number fraction for all three filtration configurations at all sampling points. It can be concluded that dosing of coagulant (Fe^{3+} or Al^{3+}) does not lead to a significant decrease of the difficult filterable fraction 2-10 µm when comparing its concentration in AF for the three filtration configurations.

The comparison of the removal efficiencies of cumulative particle numbers and volumes between AF and FI, in Table 5. 3, indicates better results for flocculating filtration with ferric chloride over all three particle fractions than the results for the filtration tests using poly aluminium chloride as coagulant.

Table 5. 3: Average removal of particle fractions 2-10 µm, 10-20 µm, 20-100 µm for the blank and flocculating filtration with ferric chloride ($FeCl_3$) or poly aluminium chloride (PAC) at WWTP Beverwijk, in [%]

	Removal efficiency [%]		
	2-10 µm	10-20 µm	20-100 µm
Cumulative particle number			
Blank filtration	73.7	94.7	97.0
Flocculating filtration ($FeCl_3$)	87.3	96.2	98.3
Flocculating filtration (PAC)	81.8	79.5	76.8
Cumulative particle volume			
Blank filtration	85.6	95.1	94.7
Flocculating filtration ($FeCl_3$)	93.5	96.6	96.8
Flocculating filtration (PAC)	81.7	79.1	74.4

The conclusion that poly aluminium chloride applied as coagulant results in a decrease of the removal efficiency could be confirmed by the average diameter calculated for the same experiments, showed in Table 5. 4. Evaluating the average diameter in AF, the size slightly increases when ferric chloride was applied and significantly decreases when poly aluminium chloride was dosed compared to blank filtration. After flocculating filtration a larger diameter for FI (PAC) than for FI ($FeCl_3$) is found. This confirms the breakthrough of larger particles and the less efficiency of poly aluminium chloride.

Table 5. 4: Overview of the average diameter (± standard deviation) for the particle analyses on diluted samples analysed during blank and flocculating filtration with ferric chloride ($FeCl_3$) or poly aluminium chloride (PAC) at WWTP Beverwijk

	EF	AF	FI
Blank filtration	6.90 ± 0.70	6.40 ± 0.56	3.93 ± 0.21
Flocculating filtration ($FeCl_3$)		6.44 ± 0.33	4.17 ± 0.36
Flocculating filtration (PAC)		5.88 ± 0.50	5.18 ± 1.27

Even if the number of particles in the range 2-10 µm are counted more accurately with dilution the volume they occupy in a sample remains small compared to the total particle volume. It is still small compared to the volume of particles 10-100 µm. The particle volume distributions for all sampling points are shown in Figure 5. 17.

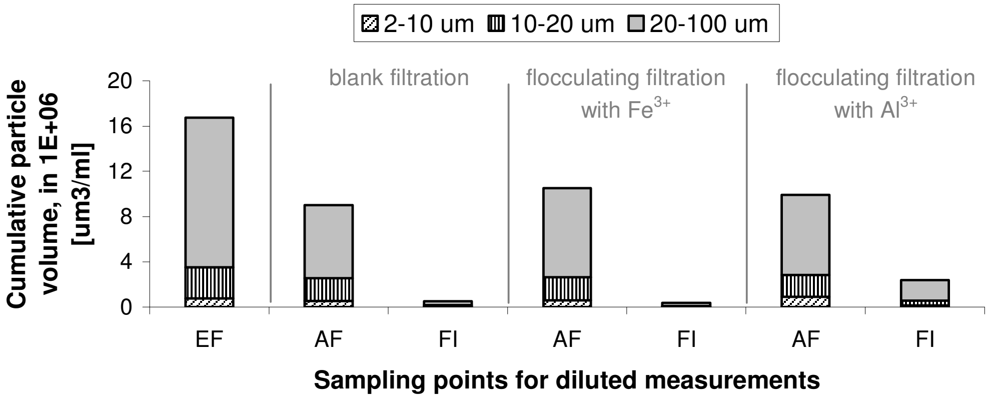

Figure 5. 17: Cumulative particle volume for diluted samples EF, AF and FI conducted during blank filtration and flocculating filtration with ferric chloride ($FeCl_3$) or poly aluminium chloride (PAC) at WWTP Beverwijk from the cumulative particle number, in [1E+06 μm^3/ml]

The total particle volume of 2-100 µm decreases significantly between the sampling points EF and AF for blank filtration and flocculating filtration with ferric chloride. The increase of particle number 2-100 µm during flocculating filtration with poly aluminium chloride, shown in Figure 5. 16, could be explained more detailed from Figure 5. 17. The number of particles 10-100 µm seems to decrease while the range 2-10 µm is represented in a much higher concentration. Settlement within the piping or degradation to smaller particles due to turbulence could be possible explanations.

Furthermore a slight increase of bigger particles (20-100 µm) is observed for AF after dosing ferric chloride compared to AF with no coagulant dosing or doing of poly aluminium chloride. It should be kept in mind that a small increase of bigger particles has a major effect on the total particle volume.

5.4 Evaluation

Filtration tests were performed as blank or flocculating filtration to provide the reference filter performance for further investigations operated for simultaneous nutrient removal. If suspended solids and phosphorus removal is kept on an optimal level simultaneous removal of other substances, like nitrogen, could be established without reverse effect to the basic removal purposes. In order to obtain reference results the suspended solids and phosphorus removal was analytically observed. One indicator is the Lindquist diagram, which visualises the pressure development over the filter bed height. Depending on the suspended solids concentration in the feed water even after coagulant addition, the pressure drop will increase and accumulation capacity of the filter will decrease with increasing filter run time. Due to coagulant dosing, the suspended solids load of the filter increases resulting in even faster decrease of the remaining accumulation capacity and filter run time. A linear curve indicates good filtration performance. If the pressure profile differs significantly from linear development the maximum load of the filter was reached or clogging on a certain level occurred. In this case the filter has to be backwashed. Most importantly the reversibility of a clean bed pressure should be maintained by comparing pressure curves under comparable filter operation and different filtration runs.

The filtration tests at WWTP Utrecht and WWTP Beverwijk provided pressure developments in progressed filter run time for both configurations, blank and flocculating filtration. The filter run time for blank and flocculating filtration resulted in 24h and 16h at WWTP Utrecht, and > 24h and 7-17h at WWTP Beverwijk, respectively. Before completed run time the pressure development can be considered as linear without major filter bed clogging. With the application of coagulant the pressure drops faster than without dosing. The starting pressure after backwash is reversible for filtration tests on both locations.

The low dosing of approx. 1-1.5 molAl/molPO_4-$P_{<0.45\mu m}$ or 2.0-3.0 molFe/molPO_4-$P_{<0.45\mu m}$ seemed efficient to remove total phosphorus to an average of 0.2 mg/L in the filtrate. These molar ratios are within the practical applied range of Me/P (metal/total phosphorus) between 1-3 mol/mol (Metcalf and Eddy, 2003). The constant dosing of coagulant resulted in periodical over or under dosing due to changing phosphorus concentrations in the feed water during filtration tests at WWTP Utrecht and Beverwijk. Less optimal results for phosphorus removal were incidentally observed. Proportional dosing to the orthophosphate concentration in the feed water is necessary and will result in a more stable removal of phosphorus; essential when phosphorus needs to be removed structurally to very low levels like the MTR level (< 0.15 mgP_{tot}/L).

The installed online turbidity analysers during initial pilot scale investigations at WWTP Utrecht showed a decreasing reliability of measurements in a relatively short time. The analysers should be maintained on a regular base to remove eventual fouling. Furthermore, verification of the online measurements with periodical manual analyses still remains necessary in order to confirm the results.

Particle analyses at WWTP Utrecht showed little removal for particles ranging from 2-20 µm. Aiming at low phosphorus concentrations and consequently implying high particle removal, extended particle analyses were conducted at WWTP Beverwijk to investigate their mutual removal results and changes in fractions during the filtration process. The analyses indicated only minor change in particle size and volume distribution when comparing blank and flocculating filtration using poly aluminium chloride or ferric chloride as coagulant. The preliminary assumption of ongoing processes in the filter bed, like further precipitation, coagulation and flocculation inside, which is expected to lead to changes in particle distributions, was not clearly confirmed. However, only the removal of the orthophosphate fraction (PO_4-$P_{<0.45\mu m}$ or P_{diss}) during flocculating filtration with poly aluminium chloride, indicated the occurrence of such ongoing processes.

Generally, the filtration results of even small particles could be evaluated as good with a removal of 50% in the size range of 2-10 µm. The removal seemed strongly to depend on the applied coagulant. For example, the particle number or volume removal in this range during flocculating filtration with dosing of poly aluminium chloride showed to be more difficult than with the dosing of ferric chloride. This observation coincides with the observation that ferric chloride leads to less particulate phosphate and less good filtration than poly aluminium chloride.

The fractionation of phosphate, in dissolved and particulate form, indicated the effect of phosphate distribution changes during the coagulation, flocculation and filtration process. Even if the dissolved phosphate fraction is partly removed during filtration the formation of particulate phosphate is very important to filtration. The concentration of particulate phosphate seemed to be in accordance with remaining small particles (2-10 µm) in the filtrate with that were identified as difficult for filtration. Further investigation may lead to further process optimisation aiming to minimise these particle fractions and improve phosphorus removal. Improved understanding of how particles especially in the range 2-10 µm are reduced, may lead to better and more stable removal of phosphorus down to MTR level.

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6

Filtration for simultaneous nutrient removal

6.1 Introduction

This chapter presents the results of simultaneous nutrient removal, combining flocculating and denitrifying filtration in one dual media filtration step. The filtration tests were performed at the three wastewater treatment plants WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer.

The initial pilot scale tests at WWTP Utrecht aimed to evaluate the applicability of dual media filtration for this multi purpose approach and were confirmed and followed up by lab scale investigations at the WWTP Beverwijk. Optimised long term pilot scale tests at WWTP Horstermeer finalised the research period. Figure 6. 1, Figure 6. 2 and Figure 6. 3 show the schedule of filtration tests at the three research locations.

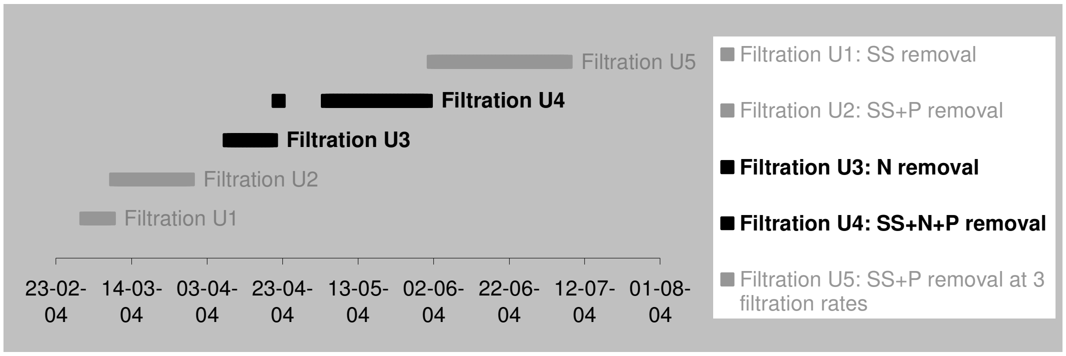

Figure 6. 1: Scheduling of filtration experiments for denitrifying flocculating filtration for pilot scale investigations at WWTP Utrecht; SS: suspended solids, N: nitrogen, P: phosphorus

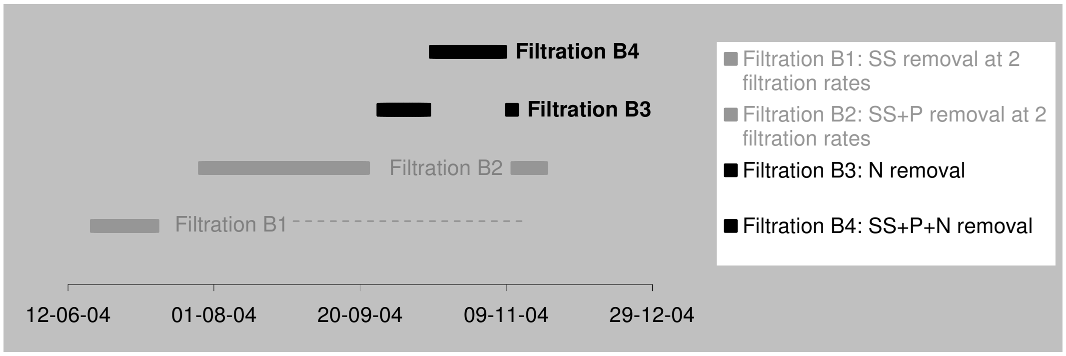

Figure 6. 2: Scheduling of filtration experiments for denitrifying flocculating filtration for lab scale investigations at WWTP Beverwijk; SS: suspended solids, N: nitrogen, P: phosphorus

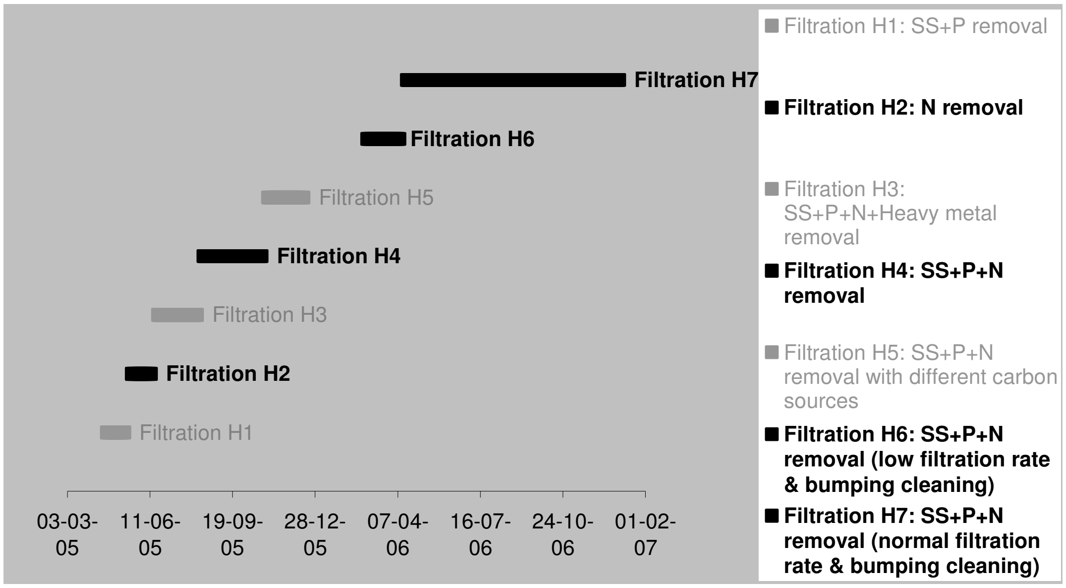

Figure 6. 3: Scheduling of filtration experiments for denitrifying flocculating filtration for pilot scale investigations at WWTP Horstermeer; SS: suspended solids, N: nitrogen, P: phosphorus

The results regarding filter operation, removal of particles, turbidity, phosphorus, nitrogen during simultaneous denitrifying flocculating filtration, will be presented in this chapter, finalised by an evaluation of the results.

6.2 WWTP Utrecht

6.2.1 Dosing conditions

For simultaneous nitrogen removal during flocculating filtration methanol was applied as a external carbon source due to the high biodegradability indicated by a BOD/COD ratio about 0.9 kg/kg. A 10% methanol solution, consisting of pure methanol (95%) and tap water, was used for these investigations.

For the start-up of the denitrification process the methanol solution was dosed in a ratio of 4.8 gCH_3OH per gNO_3-N. This ratio is adopted from literature such as described by Eichinger (1994) indicating a ratio of 3.0 gCH_3OH per gNO_3-N for denitrification in tertiary filtration. Furthermore, an overdose of 60% was applied for present free oxygen concentration (averagely 5.6 ± 0.3 mgO_2/L). The methanol dosing was adjusted online and proportional to the nitrate concentration continuously measured in the WWTP Utrecht. During the start–up period of denitrification no simultaneous dosing of coagulant for chemical phosphate precipitation was applied.

After one week of good denitrification results the dosed amount of methanol was reduced to 4.2 gCH_3OH/gNO_3-N for a filtration rate of 6.3 m/h. This ratio includes an overdose of 40% for the free oxygen present in the WWTP effluent.

Some operational problems occurred during those first filtration tests for simultaneous nutrient removal concerning mainly the methanol dosing. Disturbances caused by accumulation of air in the dosing line, clogging of dosing points, and incidental lack of methanol in stock, were notified. More frequent attention to maintenance on the dosing system was required. The periodical insufficient methanol dosing resulted in increased nitrate concentrations in the filtrate due to incomplete denitrification. Adjusting the dosing conditions to normal operation showed a recovery of denitrification in a short period of time, usually within 30 minutes.

The application of coagulant for chemical phosphate precipitation simultaneous to denitrification caused additional operational problems resulting in rapid filter clogging. These problems indicated the importance to investigate an appropriate backwash procedure and frequency to ensure the stability and reliability of the multi purpose process.

Coagulant was dosed in this period with a fixed amount of 1.0 $mgFe^{3+}/L$ or 1.0-2.0 $mgAl^{3+}/L$ manually adjusted in regard to the orthophosphate concentration in the WWTP effluent.

6.2.2 Filter run time and backwash

The simultaneous removal of nitrogen by denitrifying biomass on the filter material required optimisation of the backwash procedure. The occurrence of “mud-ball” forming was very likely to occur in case of insufficient cleaning of the filter bed which may lead to partially clogging of filter bed. Furthermore, entrapped nitrogen gas, developing by the nitrogen conversion reactions, could also cause clogging resulting in an exceeding pressure drop. Therefore, the backwash needed to be adjusted in order to ensure sufficient release of foulants, separation of adhered grains and stratification of the filter layers but preventing unfavourable removing of biomass caused by exaggerated turbulence.

During these first experiments with denitrifying filtration at a low filtration rate of 6.3 m/h the backwash procedure BW1, described in chapter 5, was replaced by the second shortened procedure BW2, shown in Table 6. 1. The air backwash phase in the sequence of the original procedure BW1 was skipped because a negative effect on the denitrification capacity by oxygen intake in the filter bed was assumed.

Table 6. 1: Backwash procedure BW2 for pilot scale filters at WWTP Utrecht

	Scale	Phase 1	Phase 2
Time	[sec]	20-100	60
Air	[-]	Off	Off
Blower	[-]	-	-
Water	[-]	On	On
Velocity	[m/h]	90	10

The backwash phase 1 of the procedure BW2 was varied between 20 and 100 sec. aiming to prevent "mud-ball" forming and proper removal of the accumulated foulants from the filter bed. Furthermore, the high water velocity in the backwash phase 1, was expected to be sufficient for the release of nitrogen gas (N_2) and carbon dioxide (CO_2) which is formed during denitrification.

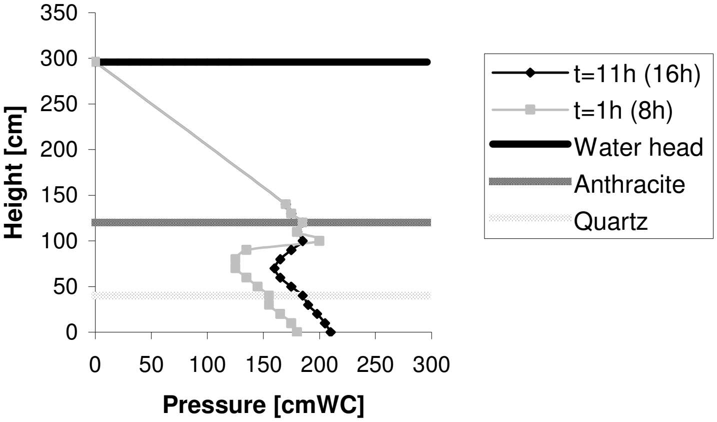

Figure 6. 4: Lindquist diagram obtained at WWTP Utrecht with applied backwash procedure U2 (phase 1: 20-60 sec) at filter run times of 16 and 8 hours with a ratio of 4.2 gCH_3OH/gNO_3-N, 1.0 mgAl/L and a flow rate of 5 m^3/h

The variation of backwash phase 1 between 20 and 60 sec. (Table 6. 1) only showed minor effect on the improvement of filter bed stratification (Figure 6. 4) and prevention of mud-ball-forming, revealing from the deformed curve "t=1h (8h)". This curve differs strongly from a linear development which is contradictory to the expectation for one hour of operation after backwash.

The increase of the duration to 100 seconds led to a significant improvement (Figure 6. 5) resulting in more linear pressure development throughout filtration run time.

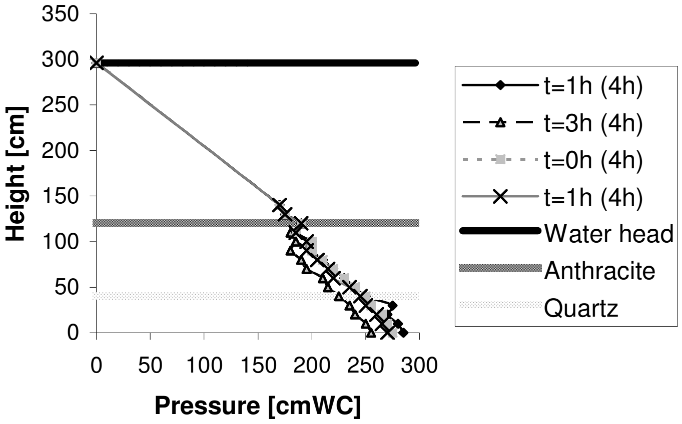

Figure 6. 5: Lindquist diagram obtained at WWTP Utrecht with backwash procedure U2 (phase 1: 100 sec) at a filter run time of 4 hours with a ratio of 4.2 gCH_3OH/gNO_3-N, 0.9 mg Al/L and a flow rate of 5 m^3/h

Only a minor pressure drop is visible in Figure 6. 5 during filtration with the applied backwash procedure BW2 (phase 1: 100 sec). To achieve a stable filtration process the run time had to be reduced to 4 hours with a filtration rate of 6.3 m/h.

With this backwash regime the denitrifying biomass was satisfactory kept in the filter, as was confirmed by the good denitrification results after three days of operation.

In the following filtration tests at WWTP Utrecht the filter run time was reached to 8 hours. But in the following days and weeks of the project these adjustments led to problems of mud-ball forming, insufficient removal of foulants and rinsing of anthracite.

Further optimisation of the backwash regime could not have been completed in the available project period at WWTP Utrecht but were resumed during the following lab scale filtration tests at WWTP Beverwijk.

6.2.3 Nutrients and other conventional parameters

Similar to the filtration tests for single flocculating filtration, described in chapter 5, poly aluminium chloride (PACl) and ferric chloride ($FeCl_3$) were used as coagulant for chemical phosphate precipitation during simultaneous nutrient removal at the WWTP Utrecht.

Figure 6. 6 displays the manual and online conducted measurements for total phosphate (PO4-Punfilt.) and nitrate (NO3-N) in the period of 19-04-'04 and 26-04-'04. In this period poly aluminium chloride was dosed.

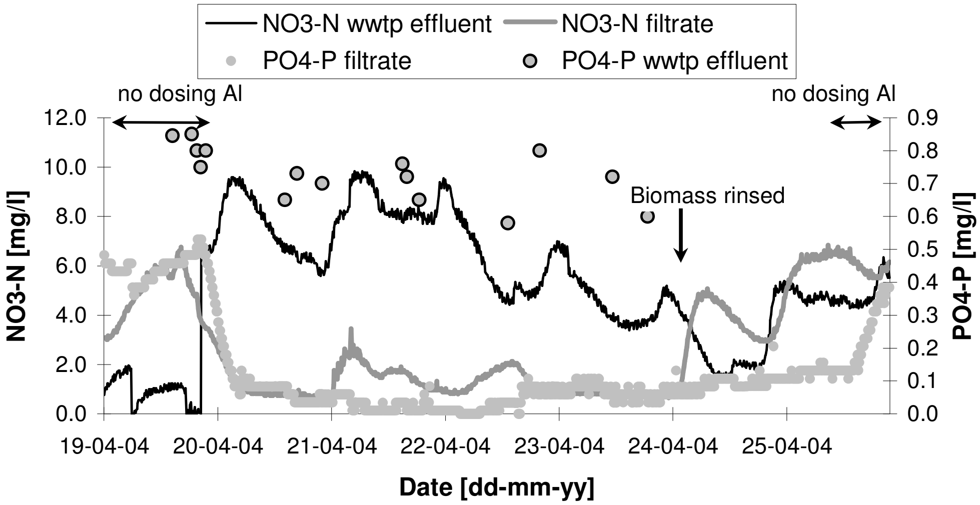

Figure 6. 6: Online and manual nitrate and total phosphate measurements at the WWTP Utrecht between 19-04-‘04 and 26-04-’04 with a filtration rate of 6.3 m/h, a filtration run time of 4 hours and the backwash procedure BW2; Al: Aluminium

The few manual analyses of total phosphate (orthophosphate plus particulate phosphate) in the WWTP effluent deliver an approximate average value of 0.7 mgPO$_4$-P/L. The trend of a good phosphorus removal was found with proper conditions of coagulant dosing. Low concentrations of 0.04 – 0.1 mgPO$_4$-P/L in the filtrate were obtained with a dosing of 1.5 mgAl/L (metal-orthophosphate Me/PO ratio of averagely 2.5), between 21-04-‘04 to 24-04-’04 with a filtration rate of 6.3 m/h.

Without presenting the results, it should be noted that similar removal was examined for an average orthophosphate concentration of 0.5 mg mgPO$_4$-P/L in the WWTP effluent and a dosing of 1.0 mgFe/L (Me/PO ratio of averagely 0.8 mol/mol) in the period 05-05-‘04 to 07-05-’04.

The calculation of the organic phosphorus in the filtrate showed a maximum concentration of 0.1 mg/L, averagely 0.06 mg/L, through the whole project period. Considering this average organic phosphorus concentration additional to the achieved low total phosphate content the total phosphorus resulted in concentrations of 0.1-0.16 mg/L. The goal of MTR quality with $P_{tot} < 0.15$ mg/L was therefore achieved. However, this result was only maintained for a very short period (less than a week). Further investigations should be conducted in order to obtain a filtrate quality reaching the MTR-values in long term filtration tests, thus providing more reliability and robustness.

The evaluation of the phosphorus assimilation by denitrifying biomass showed a maximum biological uptake of 0.2 mgPO$_4$-P/L during the whole research period. These analyses were carried out during single denitrifying filtration tests.

The results of the nitrogen removal, after one week of start-up for denitrification, showed fluctuations of nitrate (NO_3-N) removal efficiencies due to periodical failures. These failures were dedicated to lacking maintenance of the methanol dosing system and biofilm loss due to strong and frequent filter backwash, as mentioned above.

Similar to the operational disturbances strong short term variations in the feed water concentration between 2-10 mgNO_3-N/L sometimes hindered the nitrate removal performance. Daily concentration peaks were defined as problematic. Denitrifying biomass requiring an adaptation period for changing loading conditions of days or even weeks, can explain the unstable nitrate removal.

However, the achieved concentrations in the filtrate below 2 mgNO_3-N/L, shown in Figure 6. 6, indicate the possibility of nitrate removal to low levels with denitrifying flocculating filtration. Stable denitrification in a longer term may be possible in case of further optimised process conditions, e.g. for backwash, dosing etc. and less variable WWTP effluent quality.

Filtration tests with incidental low orthophosphate concentrations, below 0.1 mgPO_4-P/L, in the feed water (WWTP effluent), did not indicate any inhibition effect on the denitrifying biomass. These results are not displayed in Figure 6. 6.

During the periods of high denitrification the concentrations of COD concentrations in the filtrate did not exceed the feed water concentrations. Therefore overdosing of methanol and breakthrough could be excluded but is not finally proved with methanol analyses. On the other hand, even when high nitrate removal occurred, the free oxygen was not fully removed. An average concentration of 1.1 mgO_2/L (Table 6. 2) was measured in the filtrate of the dual media filter. This concentration is above the value 0.2 mgO_2/L, reported by Skerman and MacRae (1957), Dawson and Murphy (1972), Terai and Mori (1975), and Nelson and Knowles (1978) as limiting value for inhibition of denitrification.

Table 6. 2: Average concentration of certain parameters measured in the WWTP effluent (before methanol dosing) and filtrate of the dual media filter during denitrifying flocculating filtration at WWTP Utrecht

Parameter	Unit	WWTP effluent	Filtrate dual media filter
pH	[-]	6.9 ± 0.1	6.8 ± 0.1
Temperature	[ºC]	18.2 ± 3.2	18.0 ± 3.2
Turbidity	[NTU]	2.4 ± 1.8	1.0 ± 0.8
BOD	[mg/L]	2.7 ± 1.7	1.5 ± 0.9
COD	[mg/L]	34 ± 11	33 ± 17
O_2	[mg/L]	5.6 ± 0.3	1.1 ± 1.5
N_{kj}-N	[mg/L]	2.8 ± 0.7	1.9 ± 0.5
NH_4-N	[mg/L]	0.9 ± 0.4	0.3 ± 0.3

Parameter	Unit	WWTP effluent	Filtrate dual media filter
NO_2-N	[mg/L]	0.2 ± 0.2	0.1 ± 0.3
NO_3-N	[mg/L]	6.1 ± 4.0	1.3 ± 0.3
Arsenic	[mg/L]	< 2	< 2
Cadmium	[µg/L]	< 0.05	< 0.05
Chrome	[µg/L]	< 2	< 2
Copper	[µg/L]	3 ± 1	3 ± 1
Nickel	[µg/L]	3	3 ± 2
Lead	[µg/L]	< 1	< 1
Zink	[µg/L]	18 ± 4	20 ± 5
E-coli	[col/ml]	128 ± 95	56 ± 31

Furthermore, Table 6. 2 indicates removal of Kjeldahl nitrogen (N_{Kj}), ammonium nitrogen (NH_4-N) and e.coli. E.coli's are removed exclusively due to filtration mechanisms. The reduction of Kjeldahl nitrogen may be dedicated to the filtration of organic matter and the removal of ammonium, most probably due to simultaneous nitrification within the biofilm. Metcalf and Eddy (2003) as well as Rittman and Langeland (1985) reported denitrification in the biofilm interior and nitrification at the floc exterior under low free oxygen concentration conditions.

Figure 6. 7 presents the online measured turbidity values obtained for EF and FI.

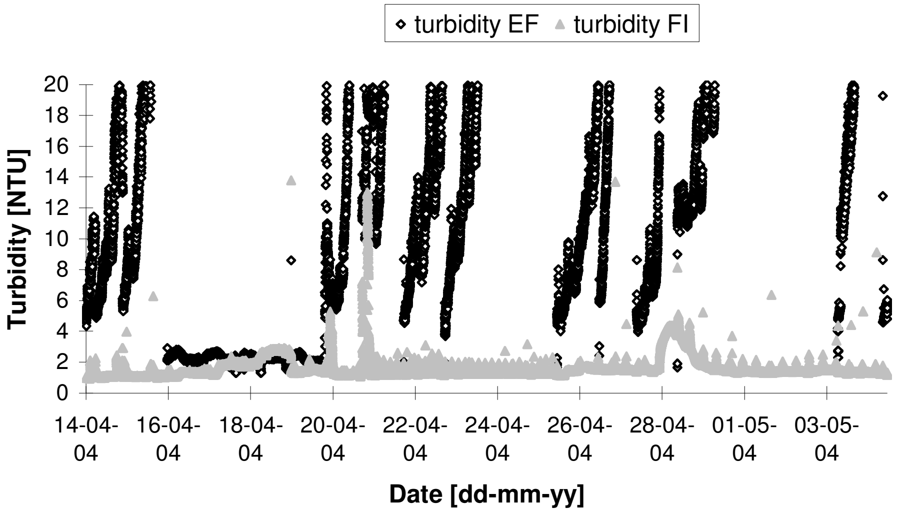

Figure 6. 7: Turbidity of WWTP effluent (EF) and filtrate (FI) during simultaneous denitrifying flocculating filtration in the period from 14-04-04 to 04-05-04

The turbidity values, shown in Figure 6. 7, led to the following conclusions regarding the online turbidity meter function:

- rapid fouling of the turbidity meter in the WWTP effluent occurred within one day of operation and after cleaning, resulting in increased values from 4 to 20 NTU. 20 NTU is the upper limit of detection range,
- filtrate turbidity delivered relative constant values of below 2 NTU. Fouling of the filtrate turbidity meter was observed but to less extent.

6.2.4 Particle analyses

A few particle analyses were conducted during the filtration tests at WWTP Utrecht. An important issue for the additional analyses was to investigate the influence of methanol dosing, the applied carbon source for denitrification, on the floc forming. The main requirement for the application of simultaneous nutrient removal in the filtration tests was the optimal removal of particles, comparable to results during single flocculating filtration, without any deterioration due to denitrifying biomass.

Figure 6. 8 shows the cumulative particle volume distributions for the effluent with methanol dosage (EF_b) and without (EF_a). In order to evaluate the comparability measurements of effluent samples were selected according to similar particle mass distributions. The distributions after addition of poly aluminium chloride are also presented in Figure 6. 8, using the indices "AF_a' " after a dosage of 1.0 mgAl/L and "AF_b' " after a dosage of 2.0 mgAl/L.

The comparison of the distributions, before and after flocculation, showed no adverse effect of methanol dosing on the floc forming, despite the different dosages of poly aluminium chloride.

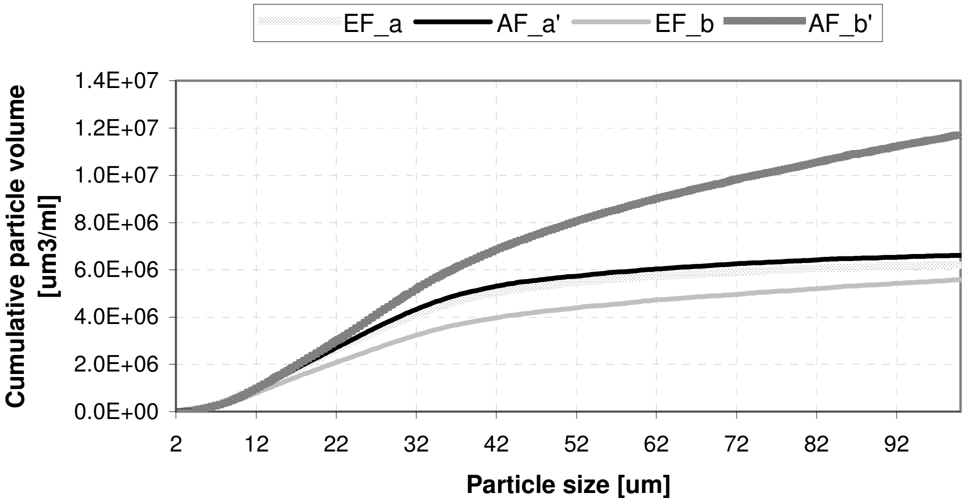

Figure 6. 8: Comparison of cumulative particle volume distributions of the WWTP effluent EF and flocculated WWTP effluent AF for single flocculating filtration and denitrifying flocculating filtration; a: without methanol dosage, b: with methanol dosage

6.3 WWTP Beverwijk

6.3.1 *Dosing conditions*

The dosing conditions for the lab scale investigations at WWTP Beverwijk were adopted from earlier investigation conducted at WWTP Utrecht. This ensured the comparability of the achieved results with previous investigations at WWTP Utrecht. Methanol was also used for these tests as carbon source for denitrification in a similar dosing ratio of 4.8 $gCH_3OH/gNO_3\text{-}N$ during the start-up of denitrification and was dosed proportionally to the online nitrate concentration in the WWTP effluent.

After one week proper denitrification with filtrate nitrate concentrations below 2.0 $mgNO_3\text{-}N/L$ was observed. From this point on the filtration rate was increased from 6.5 to 10 m/h for filtration with a more practical filtration rate. Meanwhile the dosed methanol concentration was not increased to minimize the methanol consumption. The dosing amount resulted in a methanol/nitrate ratio of 3.2 $gCH_3OH/gNO_3\text{-}N$ with an overdose of only 10% for the free oxygen (averagely 3.5 mgO_2/L) present in the WWTP effluent.

In following filtration tests with simultaneous chemical phosphorus removal mainly ferric chloride was dosed in a constant amount of 2.0-4.0 $mgFe^{3+}/L$, with manual adjustment proportional to the orthophosphate concentration ($PO_4\text{-}P_{<0.45\mu m}$) in the WWTP effluent. Regarding the variations of the orthophosphate in the WWTP effluent between 0.6-1.2 mg/L the amount of coagulant applied resulted in a metal-orthophosphate ratio of averagely 1.8. Only a few tests were conducted with poly aluminium chloride (PAC) with similar dosing ratios.

6.3.2 Filter run time and backwash

The original backwash procedure BW1, described in chapter 5, needed to be further adjusted for the operation of denitrifying filtration and denitrifying flocculating filtration. This optimisation started with the adjustment which was already undertaken during the previous pilot scale investigations at WWTP Utrecht.

Generally, the duration of all backwash phases from the original procedure BW1 were reduced to minimize the abrasion of the biomass from filter grains which may possibly resulting in biofilm loss. Compared to the procedure applied at WWTP Utrecht during denitrifying flocculating filtration the total duration for BW2 (Table 6. 3) at WWTP Beverwijk was extended to 6 minutes. An additional air backflush phase was carried out in order to achieve a better removal of foulants.

Due to the small scale of the installation compared to the pilot scale filter at WWTP Utrecht the water backflush was chosen with a higher rate in order to ensure proper filter bed cleaning.

Table 6. 3: Backwash procedure BW2 for lab scale filters at WWTP Beverwijk

	Unit	phase 1	phase 2	phase 3	phase 4
Time	[sec]	120	180	30	30
Air	-	On[1)]	Off	Off	Off
Water	-	Off	On	On	On
Velocity	[m/h]	-	~100	~50	~20

[1)] compressed air with 1-2 Nm^3/h

Figure 6. 9 and Figure 6. 10 show the pressure readings for denitrifying flocculating filtration with similar chemical dosages (4.2 $gCH_3/gNO_3\text{-}N$ and 3.0 $mgMe^{3+}/L$), a filtration rate of 10 m/h and a filter run time of maximal 7 hours.

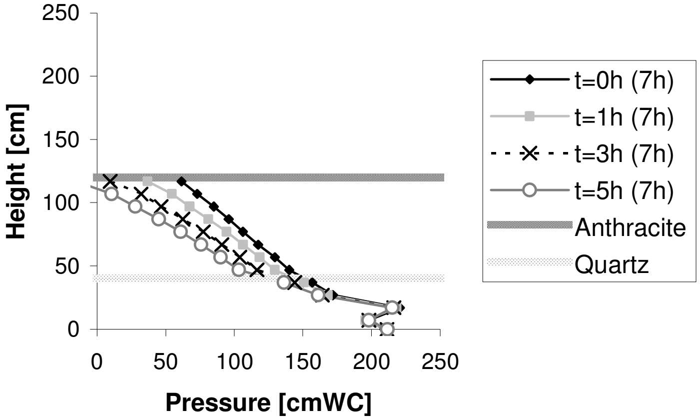

Figure 6. 9: Pressure readings (with a rising water table) obtained at WWTP Beverwijk with a filter run time of 7 hours, with a ratio of 4.2 gCH_3OH/gNO_3-N, 3.0 mgAl/L, PO_4-P_{EF}=1.6 mg/L and a filtration rate of 10 m/h

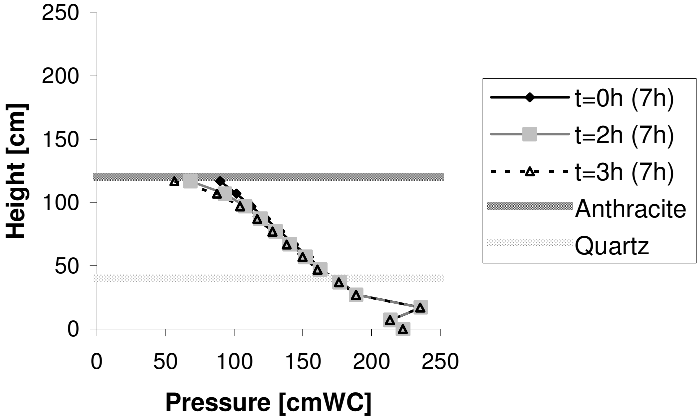

Figure 6. 10: Pressure readings (with a rising water table) obtained at WWTP Beverwijk with a filter run time of 7 hours, with a ratio of 4.2 gCH_3OH/gNO_3-N, 3.0 mgFe/L PO_4-P_{EF}=0.85 mg/L and a filtration rate of 10 m/h

In both filtration tests the curve “t=0h (7h)” shows a similar shape indicating reproducible and comparable filter bed conditions after backwash.

Filtration with poly aluminium chloride as coagulant dosed for a orthophosphate concentration of 1.6 m/L in the WWTP effluent leads to a greater pressure drop after 3 hours compared to filtration with ferric chloride with 0.85 mgPO_4-P/L. Even filter bed clogging occurred after 5 hours of operation. This development is dedicated to the higher dosing due to a higher orthophosphate concentration and the higher amount of suspended solids produced by the addition of aluminium salt (1 mgAl/L = 3.0 mgSS/L compared to

1mgFe/L = 1.9 mgSS/L suggested by Van der Graaf *et al.*, 2001). Possible inefficient filter bed cleaning may deliver an additional condition resulting in this quick end of filtration.

However, in the following filtration tests mainly ferric chloride was used as coagulant reaching 7 hours of filter run time.

6.3.3 Nutrients and other conventional parameters

The preliminary jar tests using WWTP effluent with an average pH 7.55 indicated $FeCl_3$ as a proper coagulant for lab scale filtration tests at WWTP Beverwijk. The results gained from single flocculating filtration tests, which are described in chapter 5, indicated both, iron and aluminium, with similar efficiencies regarding floc forming and phosphorus removal. A possible slight advance of iron regarding overall particle removal was evaluated. Therefore, ferric chloride was predominantly used as coagulant for the following denitrifying flocculating filtration tests.

After the start-up of denitrification methanol was dosed in the ratio of 3.2 gCH_3OH/NO_3-N at a filtration rate of 10 m/h. This resulted in almost complete nitrate removal while the free oxygen was not fully removed. It should be noted that no breakthrough of methanol was obtained during the filtration tests with optimal denitrification.

In the following, results are presented showing the nitrogen and phosphorus concentrations during single denitrifying filtration between 02-10-04 and 13-10-04 and simultaneous denitrifying flocculating filtration from 14-10-04 to 04-11-04.

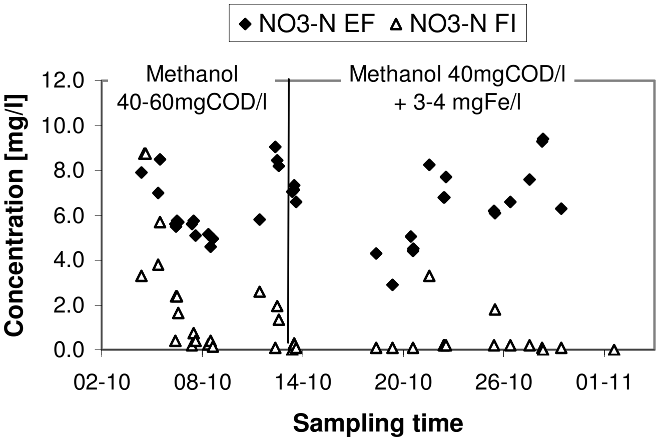

Figure 6. 11: Manual NO_3-N measurements for WWTP effluent (EF) and filtrate (FI) in lab scale installation during simultaneous denitrifying flocculating filtration at WWTP Beverwijk

A less efficient nitrogen removal performance (Figure 6. 11) in the first hour of a new filter run occurred. This may be dedicated to biomass loss and/or entering of oxygen during backwash with air inhibiting denitrification. In the second hour after backwash the nitrate concentration in the filtrate (FI) was reduced under the detection limit of the manual cuvette test of 0.5 mgNO_3-N/L.

High nitrate removal was observed during relatively constant nitrate concentration in the feed water of averagely 7.0 mgNO_3-N/L, with a considerable low methanol dosing ratio of 3.2 gCH_3OH/gNO_3-N, a filtration rate of 10 m/h and a maximum filter run time of 7 hours.

In the evaluation of the total nitrogen removal aiming to reach MTR-quality (N_{tot}<2.2 mg/L) not only nitrate removal should be considered. Regular measurements framing the monitoring program at the WWTP Beverwijk indicated periodical high Kjeldahl nitrogen concentrations with averagely 3.0 mg/L. This sum-concentration consisted of 1-3 mg/L organic nitrogen and 1-2 mg/L ammonium, caused by incomplete nitrification in the biological activated sludge treatment at WWTP Beverwijk. Consequently MTR-quality for total nitrogen in the final effluent was incidentally not achieved which can be almost exclusively addressed to non optimal wastewater treatment prior to filtration.

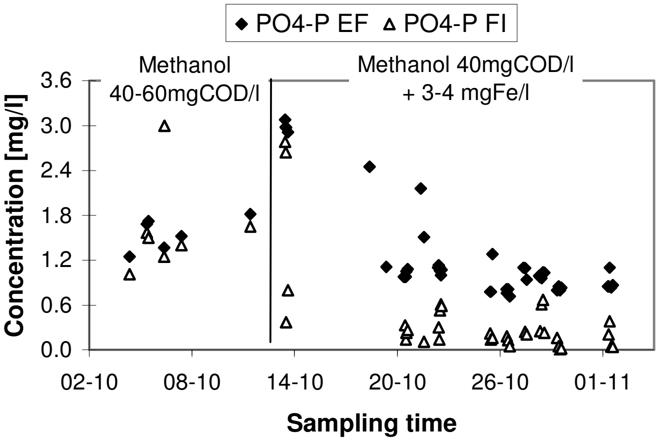

Figure 6. 12: Manual total phosphate measurements for WWTP effluent (EF) and filtrate (FI) in lab scale installation during simultaneous denitrifying flocculating filtration at WWTP Beverwijk

Figure 6. 12 shows the results for simultaneous phosphorus removal. Measurements of total phosphate (using unfiltered samples to determine P_{part}+P_{diss}) are presented, obtained for the WWTP effluent (EF) and filtrate (FI) during filtration with ferric chloride as coagulant (approx. 3-4 mgFe/L, MePO 2.1 -2.8 mol/mol).

The analyses of total phosphate resulted in average concentrations of 0.8 mg/L in EF and 0.2 mg/L in FI.

As already done during flocculating filtration tests, total phosphate analyses were fractionated indicating dissolved and particulate phosphate concentrations. These fractionations were performed during the different test conditions at the appointed sampling points of the pre-settled WWTP effluent EF, the flocculated effluent AF and the filtrate FI. The concentrations shown in Figure 6. 13 are given as average concentrations of more than 60 measurements obtained during the filtration tests operated as single denitrifying filtration and simultaneous denitrifying flocculating filtration using either ferric chloride or poly aluminium chloride as coagulant.

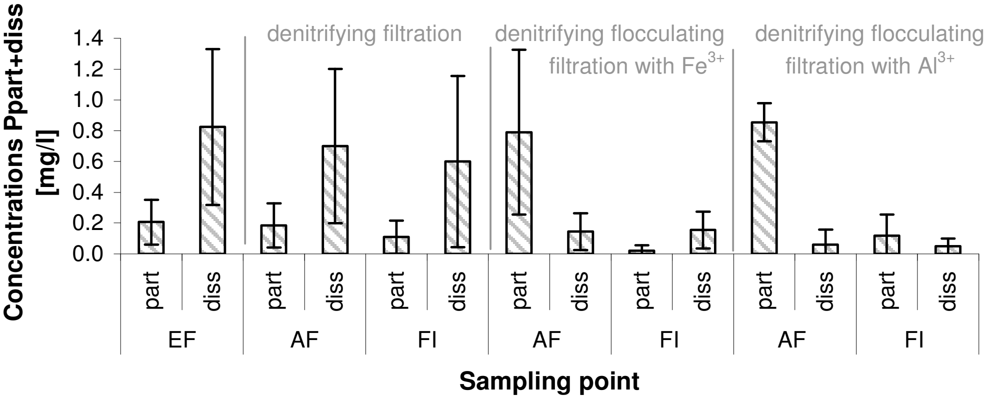

Figure 6. 13: Fractionation of average phosphate (Ppart+diss) in EF, AF and FI with standard deviation (±) for denitrifying and denitrifying flocculating filtration using $FeCl_3$ or PAC conducted at WWTP Beverwijk; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI)

The samples of the WWTP effluent EF were taken from the equalisation (buffer) tank, used for pre-settling prior to filtration. The samples for the flocculated effluent AF and the filtrate FI were taken directly above the anthracite layer and at the filtrate outlet.

The average concentrations for EF with more than 70% of total phosphate present in dissolved form (orthophosphate), seems comparable with the concentrations measured at AF for all filtration tests. Only a slightly less concentration is observed for AF during denitrifying filtration.

The phosphate distributions in the flocculated effluent AF during denitrifying flocculating filtration with both coagulants are comparable. While ferric chloride seems to result in slightly less particulate and more dissolved phosphate, the particulate fraction is removed more efficiently during filtration than with poly aluminium chloride. Furthermore, the high standard deviation of AF after dosing of iron indicates a higher spreading and less consistency of the analyses than for AF after aluminium dosing. For both coagulants the measured phosphate content is below 0.2 mg P/L.

6.3.4 Particle analyses

Comparable to the particle analyses presented in chapter 5 the countings were fractionated into the three cumulative particle classes 2-10 µm, 10-20 µm and 20-100 µm.

The average values for cumulative particle number (Figure 6. 14) and cumulative particle volume (Figure 6. 15) involve exclusively diluted measurements, in total

- 23 for the WWTP effluent EF
- 5 for the filter feed water AF and filtrate FI during denitrifying filtration
- 5 for the flocculated effluent AF and filtrate FI during denitrifying flocculating filtration with poly aluminium chloride (Al^{3+})
- 12 for the flocculated effluent AF and filtrate FI during denitrifying flocculating filtration with poly aluminium chloride (Fe^{3+})

The values for the cumulative particle numbers are displayed in normalised particle counts times 10^2 (1E+02).

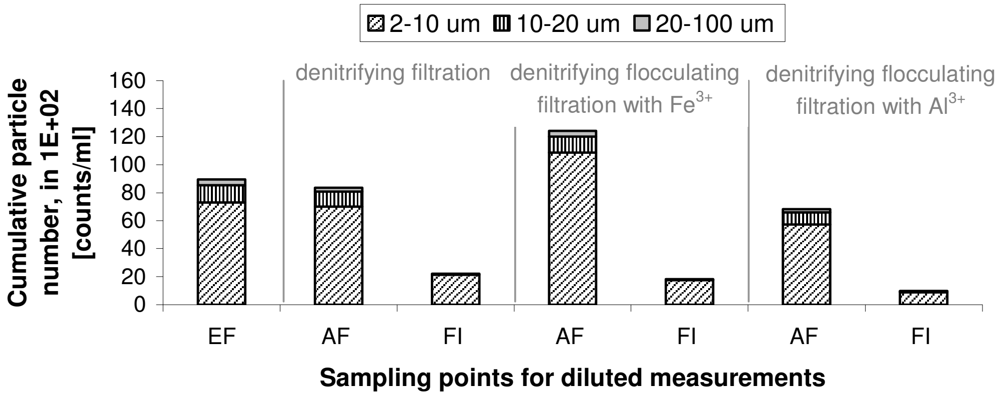

Figure 6. 14: Cumulative particle number for diluted samples conducted during denitrifying filtration and denitrifying flocculating filtration with ferric chloride and poly aluminium chloride (PAC) at WWTP Beverwijk; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI); in 1E+02 [counts/ml]

The distributions of particle numbers in EF and AF during denitrifying filtration are similar while the total number for AF during denitrifying flocculating filtration appears higher after doing of iron salt and lower after dosing of aluminium salt. The mass distributions of the feed water are not totally comparable during the different tests.

The comparison of the distributions of AF with coagulant dosing with EF or AF without coagulant dosing, does not show a significant shift in the distributions towards an increased number of bigger particles.

During filtration using iron as coagulant, larger particles (10-20 µm and 20-100 µm) are removed more efficiently than during filtration with aluminium dosing. This derives from the comparison of the filter feed water AF and the filtrate FI of these experiments. The number of particles 2-10 µm is significantly removed by filtration for both coagulants. After denitrifying flocculating filtration with iron a larger amount of small particles remains in the filtrate, according to the higher particle load measured in the flocculated WWTP effluent AF.

The observation of less efficient particle number removal applying ferric chloride could be confirmed by the numeric average diameter calculated for the same experiments, shown in Table 6. 4. Evaluating the average diameter in AF, the size slightly decreases with the application of ferric chloride and poly aluminium chloride compared to denitrifying filtration. After denitrifying flocculating filtration a smaller average diameter for FI (PAC) than for FI ($FeCl_3$) is found.

Table 6. 4: Overview of the numeric average diameter (± standard deviation) in µm for the particle analyses on diluted samples analysed during denitrifying and denitrifying flocculating filtration with ferric chloride or poly aluminium chloride at WWTP Beverwijk; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI)

	EF	AF	FI
All experiments	6.78 ± 0.59		
Denitrifying filtration		6.23 ± 0.72	3.93 ± 0.19
Denitrifying flocculating filtration (Fe^{3+})		5.91 ± 0.69	3.67 ± 0.32
Denitrifying flocculating filtration (Al^{3+})		6.03 ± 0.31	4.80 ± 0.40

The cumulative particle volumes for the sampling point AF do not indicate the expected increase of bigger particles after flocculation with iron and aluminium (Figure 6. 15). The filtration results of the fractionated particle volume shows more efficient removal for filtration with iron than for the other filtration experiments.

The total particle volume in the filtrate FI after denitrifying flocculating filtration with iron (Fe^{3+}) contains 30% less particle volume than the filtrate after filtration enhanced with aluminium salt. The present particle volume of the filtrate after denitrifying flocculating filtration with aluminium (Al^{3+}) is similar to the one measured after denitrifying filtration.

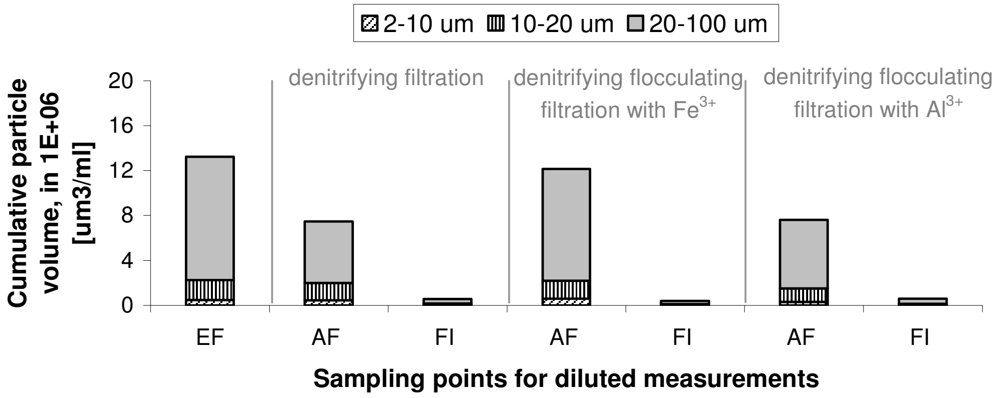

Figure 6. 15: Cumulative particle volume for diluted samples conducted during denitrifying filtration and denitrifying flocculating filtration with ferric chloride and poly aluminium chloride at WWTP Beverwijk; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI); in 1E+06 [µm^3/ml]

The distributions of cumulative numbers and volumes for the selected particle fractions showed to be corresponding during the different filtration tests. Although, the effect of a number increase of small particles (2-10 µm) on the total volume (normalised particle volumes times 10^6 or 1E+06) is very small compared.

The overall removal efficiencies of cumulative particle numbers and volumes, compared in Table 6. 5, indicate lower filtration results for all three particle fractions during flocculating filtration with poly aluminium chloride compared to filtration with ferric chloride.

Table 6. 5: Average number and volume removal [%] of particle fractions 2-10 µm, 10-20 µm, 20-100 µm for denitrifying and denitrifying flocculating filtration with ferric chloride or poly aluminium chloride at WWTP Beverwijk

	2-10 µm	10-20 µm	20-100 µm
Cumulative particle number			
Denitrifying filtration	69.3	94.5	97.0
Denitrifying flocculating filtration (Fe^{3+})	83.8	96.3	97.1
Denitrifying flocculating filtration (Al^{3+})	84.5	92.3	95.0
Cumulative particle volume			
Denitrifying filtration	80.5	94.8	92.7
Denitrifying flocculating filtration (Fe^{3+})	92.0	96.5	97.3
Denitrifying flocculating filtration (Al^{3+})	85.5	92.7	92.4

6.4 WWTP Horstermeer

6.4.1 Dosing conditions

The denitrifying flocculating filtration tests at WWTP Horstermeer aimed at stable long term results applying the experiences gained from earlier research conducted at WWTP Utrecht and Beverwijk. These experiences include especially adjustments of the backwash procedure and minimizing of the chemical dosing.

During the pilot investigations at WWTP Horstermeer poly aluminium chloride (PACl) is predominantly dosed to enhance the WWTP effluent for chemical phosphate precipitation. This product was chosen due to the low pH of averagely 6.6 measured in the WWTP effluent and its relatively good efficiency in the preliminary jar tests. Metal/orthophosphate ratios of Me/PO 4.8 to 11.7 (mol/mol) were applied in the filtration tests proportional to the PO_4-P concentration analysed in the effluent buffer. High Me/PO ratios of 9.2 to 11.7 were predominantly used to investigate simultaneous heavy metal removal. These results will be presented in chapter 7.

The methanol dosage during start-up of denitrification was maintained at a lower level than in earlier investigations, resulting in a dosing ratio CH_3OH/NO_3-N of 4.2 and a filtration rate of 6.3 m/h. After a week of good denitrification results, the methanol dosage was adjusted to the nitrate and free oxygen concentration in the effluent buffer using CH_3OH/NO_3-N ratios of 3.7-5.0. Frequent optimisation of backwash and adjustment of the methanol dosing resulted in the final dosing ratio CH_3OH/NO_3-N of 4.5 at a filtration rate of 10 m/h.

6.4.2 Filter run time and backwash

In the pilot scale filtration tests for simultaneous nutrient removal at the WWTP Horstermeer were conducted adopting the experiences of earlier pilot and lab scale investigations at the WWTP Utrecht and Beverwijk and in regards to even further backwash optimisation. These optimisation steps meant to achieve a more practical operation such as filter runtimes above 8 hours with a filtration rate of 10 m/h.

The backwash procedure BW2 applied during the pilot scale investigations at WWTP Utrecht, shown in Table 6. 1, was used as starting point for further adjustment of the procedure applied at WWTP Horstermeer (BW1.1, Table 6. 6).

Table 6. 6: Backwash procedure BW1.1 for pilot scale filter at WWTP Horstermeer

	Scale	phase 1	phase 2	phase 3	phase 4	phase 5
Time	[sec]	60	60	80	60	60
Air	[-]	On[1)]	Off	Off	Off	Off
Water	[-]	Off	On	On	On	On
Velocity	[m/h]	-	10	10	30	80-50

[1)] blower operation: 20 Hz, 20 Nm^3/h

In a later stage the backwash procedure was rearranged in several steps with decreasing backwash velocities to achieve better stratification of the filtration layers and to prevent rinsing of filtration material during backwash. This resulted in the backwash procedure BW 1.2 presented in Table 6. 7.

Table 6. 7: Backwash procedure BW1.2 for pilot scale filter at WWTP Horstermeer

	Scale	phase 1	phase 2	phase 3	phase 4	phase 5
Time	[sec]	60	180	120	360	60
Air	[-]	On[1)]	Off	Off	Off	Off
Water	[-]	Off	On	On	On	Off
Velocity	[m/h]	-	30	50	10	-

[1)] blower operation: 20 Hz, 20 Nm^3/h

Further adjustments led to the final procedure BW2 shown in Table 6. 8.

Table 6. 8: Backwash procedure BW2 for pilot scale filter at WWTP Horstermeer

	Scale	phase 1	phase 2	phase 3	phase 4
Time	[sec]	180	120	120	240
Air	[-]	On[1)]	Off	Off	Off
Water	[-]	On	On	On	Off
Velocity	[m/h]	5	30	50	10

[1)] blower operation: 20 Hz, 20 Nm^3/h

Additionally, an improvement of the filtration run time was investigated by applying a “bumping cleaning” of two minutes backwash with filtrate, every three hours with a rate of 50 m/h, aiming at the release of the nitrogen and carbon dioxide gasses formed during denitrification.

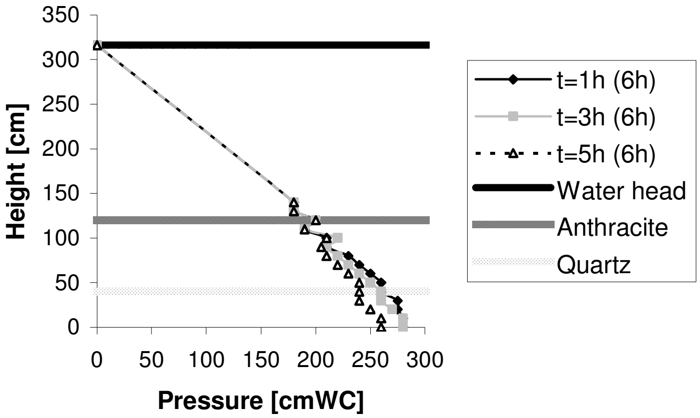

Figure 6. 16: Lindquist diagram obtained at WWTP Horstermeer with a filter run time of 6 hours with regular backwash procedure BW1.2, 40 mgCOD/L (CH_3OH/NO_3-N ratio = 4.5), 3.0 mgAl/L (MePO ~ 4 mol/mol) and a filtration rate of 10 m/h

Figure 6. 16 shows pressure readings for three different time steps within the filtration run time applying the backwash procedure BW1.2, a methanol dosing ratio of CH_3OH/NO_3-N of 4.5, metal-orthophosphate ratio of 4.0 (mol/mol) and a filtration rate of 10 m/h.

The pressure drops slightly within the filtration run time of only six hours due to fouling and entrapped nitrogen gas, and carbon dioxide in the filter bed. Even though, the pressure development can still be considered as linear development. No major clogging of the filter bed is indicated.

In the next tests the modified backwash procedure BW2 was applied with a frequency of 24 hours. The resulting pressure development during a filter run time of 24 hours is shown in Figure 6. 17.

An additionally the “bumping cleaning”, with a frequency of three hours, was introduced after 17 hours of run time. In the following hours of run time the pressure drops significantly within the next 4 hours.

After 24 hours the next normal backwash BW2 is performed.

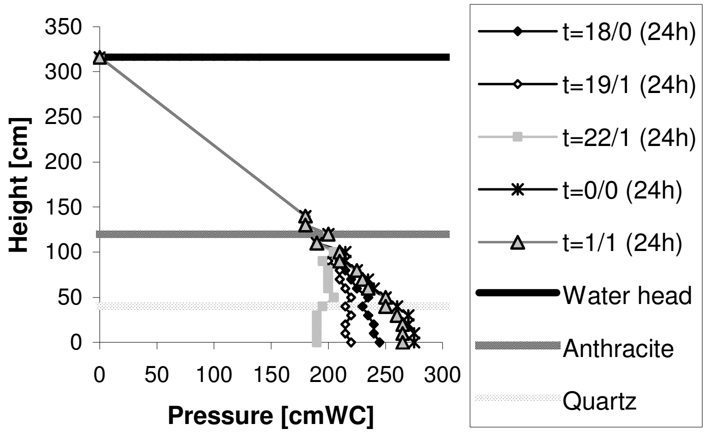

Figure 6. 17: Lindquist diagram obtained at WWTP Horstermeer with a filter run time of 24 hours with regular backwash procedure BW2 and bumping cleaning every 3 hours, 40 mgCOD/L (CH_3OH/NO_3-N ratio = 4.2), 3.0 mgAl/L (MePO ~ 4 mol/mol and a filtration rate of 10 m/h

The comparison of the first data curve after the main backwash BW2 at 24 hours of run time ‘t=0/0 (24h)’ with the data curve after the bumping cleaning (‘t=18/0 (24h)’) shows the incomplete reversibility of the clean bed pressure. This was caused by earlier clogging problems and extreme variations in suspended solids and nitrate concentrations measured in the WWTP effluent.

However, more stable feed water quality in the following days and weeks of operation approved the efficiency of the applied backwash regime achieving a stable filtration process.

6.4.3 Nutrients and other conventional parameters

During the experiments with pilot scale dual media filtration at the WWTP Horstermeer the start-up phase of denitrification is performed in a way similar to earlier research, with an applied filtration rate of 6.3 m/h and an overdose of methanol of 60%. According to the investigations at the previous locations, full denitrification occurred after one week.

In the following filtration runs the methanol dosing and the filtration rate were adjusted to the accomplished optimal backwash regime. Figure 6. 18 shows almost complete reduction of nitrate using a methanol dosing ratio of CH_3OH/NO_3-N = 4.5 in the shown period of 04-08-2005 until 23-10-2005. These operational conditions resulted in filtrate concentrations of even lower than 0.2 mg NO_3-N/L (N_{tot} < 1 mg/L) with a filtration rate of 10 m/h.

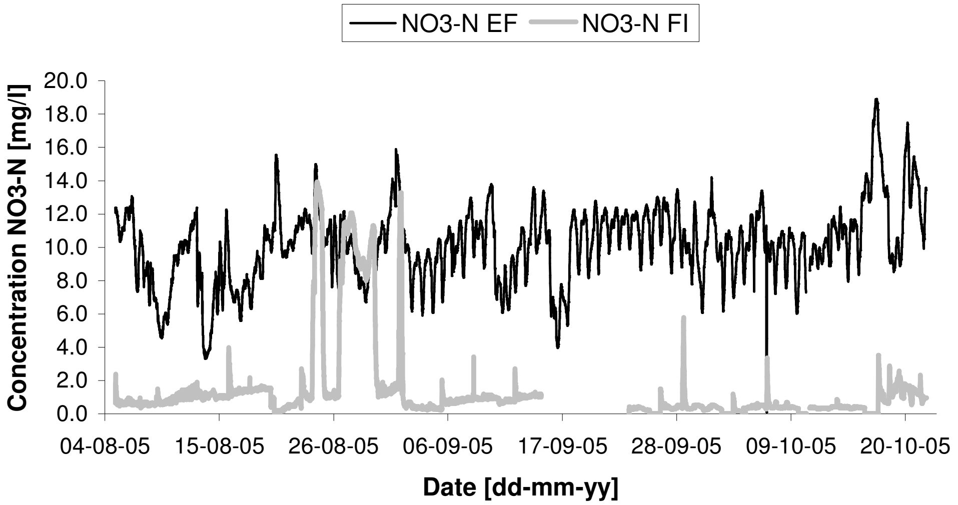

Figure 6. 18: Nitrate concentrations for WWTP effluent EF and filtrate FI during pilot scale denitrifying flocculating filtration at WWTP Horstermeer between 04-08-'05 and 23-10-'05; WWTP effluent (EF), filtrate (FI)

The concentrations in the WWTP effluent varied in that period between 4 and 19 mgNO_3-N/L. Short term increase of nitrate loading rates, even extreme daily peaks, resulted in incidentally increased nitrate concentrations in the filtrate. Also the turbidity values of the WWTP effluent in that period showed high variations between 5 and 14 NTU (see Appendix E).

During these extremely varying loading conditions the filter run time was reduced to 6 hours. Contradictory to the effect of backwash on the denitrification described for the filtration tests at WWTP Beverwijk, no significant nitrate removal deterioration was observed. No adverse effect due to oxygen intake into the filter bed during air scour or extreme loss of biomass due to vigorous backwash leading to less optimal denitrification occurred. At that point the first tests with the bumping cleaning were carried out with a two minutes flow rate of 50 m/h every three hours.

Although general high nitrate removal is observed, incidental methanol dosing problems between 24-08-'05 and 01-09-'05 resulted in high filtrate concentrations (Figure 6. 19). Restoring of the dosing within 72 hours resulted in fully recovered denitrification within 20 min.

The successive investigations in 2006, not displayed in Figure 6. 18, delivered comparable results of high nitrate removal achieved for less variable turbidity and nitrate concentrations in the feed water. A stable operation and high nitrate removal was obtained with a run time of 24 hours with a bumping cleaning of a two minutes and a flow rate of 50 m/h every three hours. Even the increase of the flow rate to 12.5 m/h resulted in similar high nitrate removal as with a flow rate of 10 m/h.

A nitrate conversion up to 3 $kgN/d/m^3_{bedvolume}$ at a process temperature of 20 ºC, was reached for denitrifying flocculating filtration, shown in Figure 6. 19. This conversion rate was restricted by the loading rate. Sometimes conversion rates up 3.5-4.0 $kgN/d/m^3_{bedvolume}$ were reached.

In later winter periods with a lower average temperature of 13 ºC in the WWTP effluent and a filtration rate of 12.5 m/h conversion rates of more than 3 $kgN/d/m^3_{bedvolume}$ were obtained.

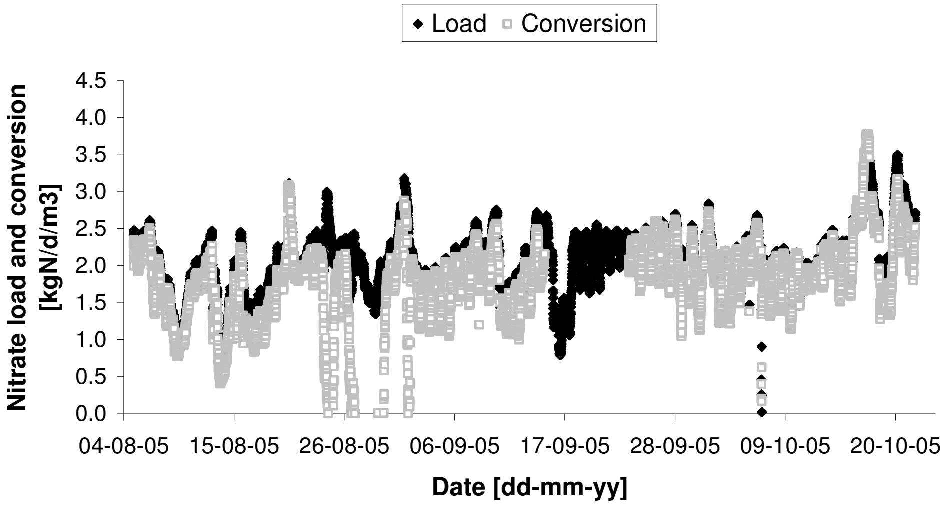

Figure 6. 19: Nitrate load measured in WWTP effluent and conversion after filtrate during pilot scale denitrifying flocculating filtration at WWTP Horstermeer between 04-08-'05 and 23-10-'05; WWTP effluent (EF), filtrate (FI)

Regular measurements at the WWTP within the research period, contracted to the laboratory "Waterproef", showed periodically peaks of Kjeldahl nitrogen concentrations above the expected 1-2 mg/L (see Appendix E). These high concentrations were addressed to incidentally high organic nitrogen (between 1 and 2 mg/L) and ammonium concentrations, which may be caused by incomplete bacterial decomposition of proteinaceous matter together with an inefficient nitrification due to low temperatures at the WWTP Horstermeer.

The online phosphate measurements, of orthophosphate ($PO_4\text{-}P_{<0.45\mu m}$) in the WWTP effluent, and total phosphate ($PO_4\text{-}P_{unfilt.}$) together with total phosphorus in the filtrate (P_{tot}), are presented in Figure 6. 20. The orthophosphate concentrations in the WWTP effluent show concentrations between 0.2 and 0.8 mg/L with peaks up to the detection limit of the analyser (1.6 mg/L). During operation with normal phosphate concentrations very low concentrations of total phosphate and total phosphorus in the filtrate were achieved the displayed period.

The organic phosphorus concentration present in the filtrate is determined by the difference between the total phosphate content, indicated by 'P (diss+part) of FI', and the total phosphorus concentration 'P_{tot} of FI'. It was calculated to a maximum of 0.1 mg/L and an average concentration of 0.06 mg/L. In periods of sufficient chemical phosphate precipitation and a stable filtration process, the total phosphorus varied around the aimed MTR quality concentration of 0.15 mgP_{tot}/L.

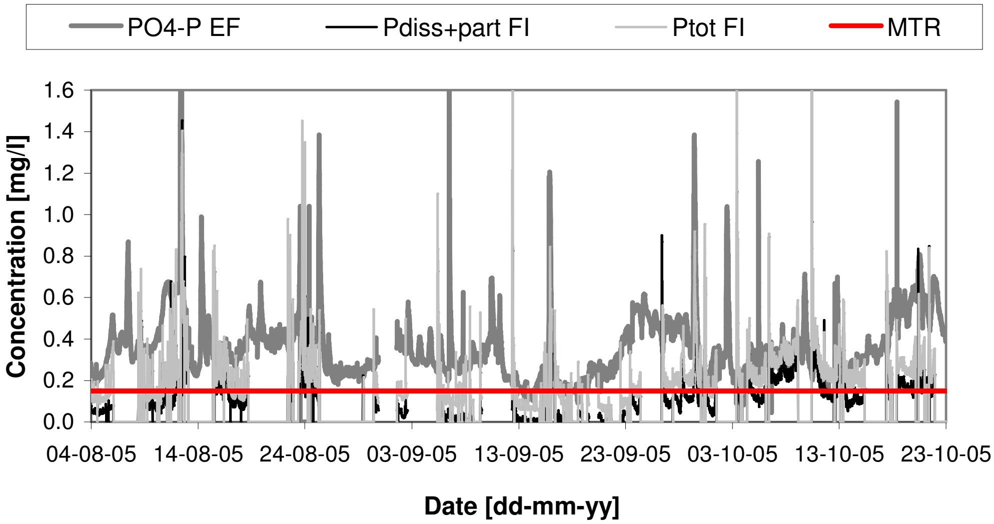

Figure 6. 20: Phosphorus concentrations for WWTP effluent, as orthophosphate (PO_4-$P_{<0.45 \mu m}$) and filtrate , as total phosphate P (diss +part), and total phosphorus P_{tot}, during pilot scale denitrifying flocculating filtration at WWTP Horstermeer between 04-08-'05 and 23-10-'05

In later stages of these investigations similar results were obtained with extended filter run time above 6 hours and up to 24 hours during less variable turbidity and nitrate loads.

In accordance to the overview given for the analyses at WWTP Beverwijk, the fractionated phosphate concentrations measured at WWTP Horstermeer are shown in Figure 6. 21. This figure presents the average values and their standard deviations of more than 500 analyses obtained for each operation, denitrifying and denitrifying flocculating filtration (with poly aluminium chloride, PACl).

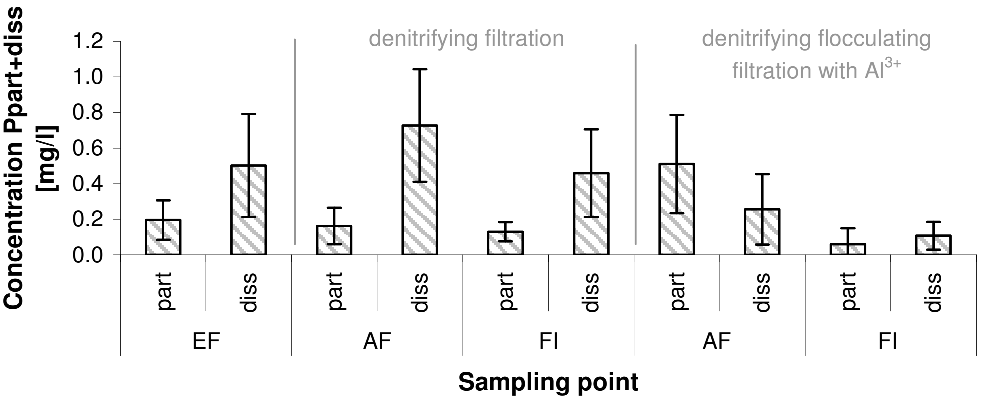

Figure 6. 21: Fractionation of average phosphate (Part+diss) in EF, AF and FI with standard deviation (±) for denitrifying and denitrifying flocculating filtration using poly aluminium chloride (PACl) conducted at WWTP Horstermeer; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI)

The concentrations for dissolved and particulate inorganic phosphorus are presented as average values with their standard deviations. The analyses were performed for the pre-sieved WWTP effluent EF, the flocculated effluent AF and the filtrate FI throughout the results obtained in 2005 and 2006.

During denitrifying flocculating filtration particulate phosphate and dissolved phosphate were removed with 82% and 37%, respectively. After denitrifying flocculating filtration using poly aluminium chloride, the orthophosphate concentration in the filtrate (FI) remained at an average concentration of 0.26 mg/L. This may indicate insufficient reaction of the metal with orthophosphate ions during chemical phosphate precipitation or an inefficient dose of aluminium salt. Even though the total phosphate concentration in the filter feed water (AF) during denitrifying flocculating filtration was lower than for denitrifying filtration, the removal of both phosphate fractions was only around 40%.

It should be noted, that the displayed analyses include all the phosphate measurements conducted for these fractions and at the described sampling points between 2005 and 2006. This includes also analyses conducted under sub-optimal operational conditions, such as, short filter run time, strongly fluctuating feed water quality and incidental filter clogging along the optimisation period of the backwash regime, etc.

An overview of all conducted manual measurements of several parameters, recorded and regularly updated in a database throughout the whole research period can be found in Appendix E.

6.4.4 Particle analyses

During the pilot scale investigations at WWTP Horstermeer particle analyses were also carried out as an additional measure for the evaluation of the coagulation and flocculation process, the filtration process and the final particle removal in possible relation to other parameters.

According to the previous presented analyses, particle numbers and volumes are displayed in Figure 6. 22, fractioned into three classes, 2-10 µm, 10-20 µm and 20 -100 µm. The average values are shown for diluted samples (ratio of 3 consisting of 2 parts demineralised water plus 1 part sample) conducted for the known sampling points

- in total 20 analyses of the WWTP effluent EF, and
- 10 analyses of each, the flocculated WWTP effluent (AF) and filtrate (FI), during the filtration experiments applying denitrifying and denitrifying flocculating filtration.

The total number of particles is removed only to a minor extent during denitrifying filtration. This may be addressed to incidental poor removal of bigger particle 10-20 µm and 20-100 µm.

Compared to the distribution in the WWTP effluent (EF) and the filter feed water (AF) during denitrifying filtration the distribution in AF after flocculation with poly aluminium chloride shifts towards bigger particle fractions. The total particle number is significantly reduced during denitrifying flocculating filtration. Most particles in the filtrate were in the range 2-10 µm with a small portion between 20-100 µm.

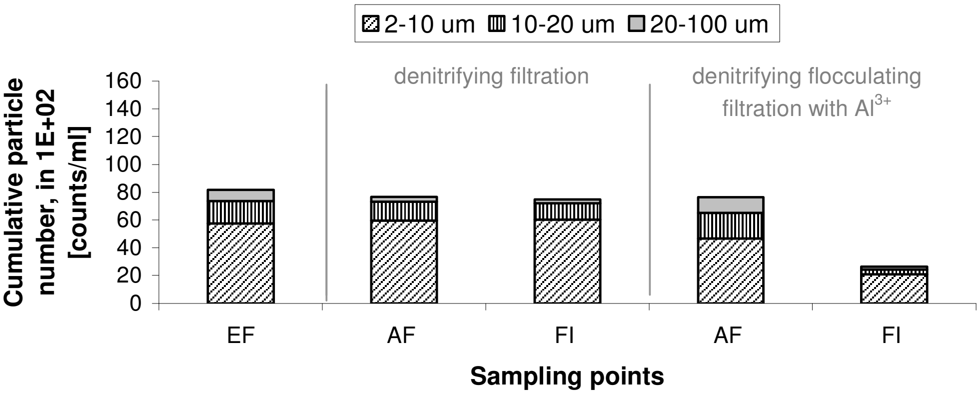

Figure 6. 22: Cumulative particle number for diluted samples conducted during denitrifying flocculating filtration with poly aluminium chloride (PACL) at WWTP Horstermeer; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI); in [1E+02]

These observations are confirmed by the calculation of the average diameters shown in Table 6. 9. High standard deviations however indicate a high dispersion of values used for calculation of the mean.

The increased numeric average diameter in the flocculated WWTP effluent (AF) during denitrifying flocculating filtration is in accordance with the shift towards bigger particles described before. Nevertheless, the relatively high average particle diameter in the filtrate (FI) confirms the inefficiency of particle removal during filtration applied for simultaneous nutrient removal.

Table 6. 9: Overview of the numeric average diameter (± standard deviation) in µm for the particle analyses on diluted samples analysed during denitrifying and denitrifying flocculating filtration with poly aluminium chloride at WWTP Horstermeer; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI)

	EF	AF	FI
All experiments	8.45 ± 2.82		
Denitrifying filtration		7.01 ± 0.60	6.89 ± 0.32
Denitrifying flocculating filtration (Al^{3+})		9.73 ± 2.83	7.18 ± 0.95

As shown before, by the average particle numbers, the volume of bigger particles was removed during denitrifying filtration. The slight change in particle size distributions after flocculation with poly aluminium chloride towards bigger particle size classes is visible in the cumulative particle volume distributions. The small numbers of particles between 20-100 µm measured in the filtrate (FI) after denitrifying flocculating filtration represents the biggest portion of the total particle volume in the filtrate.

It could be concluded that the particle distribution in the filter feed water (AF) for denitrifying flocculating filtration seems to be suitable for a good overall particle removal. Possible further optimisation of the dosing conditions, type and amount of coagulant dosing, or the adjustment of coagulation and flocculation conditions at the pilot scale installation may be necessary.

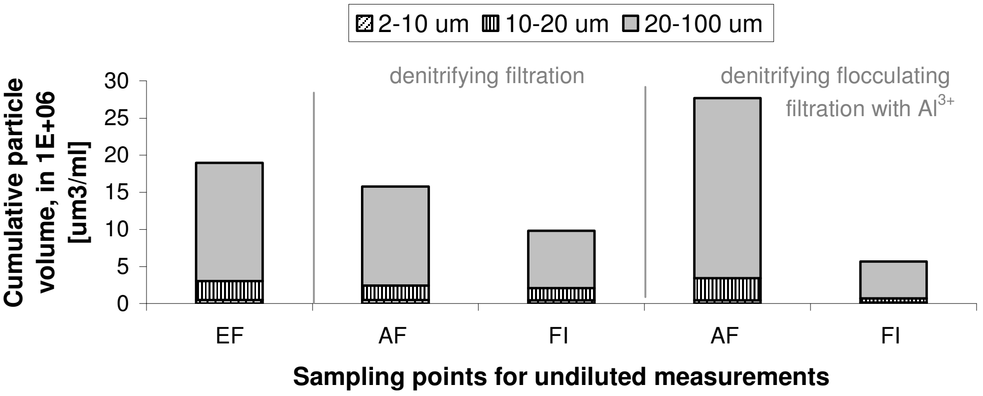

Figure 6. 23: Cumulative particle volume for diluted samples conducted during denitrifying flocculating filtration with poly aluminium chloride (PACl) at WWTP Horstermeer; WWTP effluent (EF), flocculated effluent (AF), filtrate (FI); in 1E+06 [µm^3/ml]

The average removal efficiencies for the cumulative particle numbers and volumes for the particle size classes 2-10 µm, 10-20 µm and 20-100 µm are shown in Table 6. 10. While the overall particle removal during denitrifying filtration is evaluated as weak, it improves significantly improves after coagulation and flocculation with poly aluminium chloride. However, the particle removal during denitrifying flocculating filtration could not yet be described as optimal.

Table 6. 10: Average number and volume removal [%] of particle fractions 2-10 µm, 10-20 µm, 20-100 µm for denitrifying and denitrifying flocculating filtration with poly aluminium chloride at WWTP Horstermeer

	2-10 µm	10-20 µm	20-100 µm
Cumulative particle number			
Denitrifying filtration	-1.2	15.6	16.2
Denitrifying flocculating filtration (Al^{3+})	55.1	81.2	82.8
Cumulative particle volume			
Denitrifying filtration	3.6	17.4	-45.4
Denitrifying flocculating filtration (Al^{3+})	67.1	82.4	79.4

6.5 Evaluation

Filter operation

The experiences of the preliminary investigations at the wastewater treatment plant WWTP Utrecht indicated the possibility of a multi purpose media filtration combining suspended solids, phosphorus and nitrogen removal. Even with theses fragmented results and unstable filtration on a long term basis, the possibility of simultaneous removal, at a low filtration rate of 6 m/h and filter run times of 4 hours, was demonstrated. Even though, operational adjustments had to be carried out in order to achieve stable and high removal of all assigned constituents. One of the major issues of focus was the backwash regime. At WWTP Utrecht the backwash was carried out without the air backwash phase due to an assumed affect of the oxygen intake on the efficiency of denitrification. Without proper duration and strengths of the backwash regime the filter material clogged regularly.

The following lab scale filtration tests at WWTP Beverwijk were conducted to confirm earlier investigations and to prove the applicability of a more practical filtration rate of 10 m/h. An air scour during water back flush seemed necessary for the sufficient breakage of the filter bed and separating the grains of the filter medium. Clogging of the filter bed due to the forming of ‘mud balls’ occurred less frequently than during previous pilot scale test without a simultaneous air and water back flush at a filtration rate of 10 m/h. Filter run times of 7 hours were possible although the pressure developments increasingly deviated from the optimal linear shape with increasing filter run time. This was caused by increasing fouling and also by the entrapped nitrogen and carbon dioxide gasses produced during the denitrification process.

The applied backwash regime resulted in reversibility of the starting pressure over the filter bed height directly after backwash.

The investigations for simultaneous nutrient removal on pilot scale were continued at the WWTP Horstermeer. Further optimisation was carried out, using the experiences of earlier investigations as a starting point, in order to achieve a stable process during long term filtration, including

- the increase of the filtration rate from 6.3 m/h to at least 10 m/h,
- the increase of the filtration run time from 4 hours to at least 10 – 24 hours, and
- backwash optimisation applying an adjusted regular backwash plus an additional ‘bumping cleaning’ for the release of entrapped gas in the filter bed.

The optimisation of the backwash regime towards a regular backwash together with a ‘bumping cleaning’ every 3 hours with 50 m/h, finally resulted in filtration run times up to 24 hours. The starting pressure along the filter bed after performed ‘bumping cleaning’ became almost reversible in periods of stable WWTP effluent quality. However, the process reliability depended strongly on variability of turbidity and nitrate loading conditions.

Nitrogen removal

Full denitrification was achieved after one week of start-up operation at all three research locations applying a filtration rate of 6.3 m/h and a methanol dosing ratio of 3.8 to 4.5 g CH_3OH/g NO_3-N. During stable denitrification and normal filter operation at a filtration rate of 10 m/h a ratio of 3.8 g CH_3OH/g NO_3-N at WWTP Beverwijk and 4.5 g CH_3OH/g NO_3-N at WWTP Horstermeer seemed to be appropriate. The methanol dosage of 30-40 mg/L measured as COD resulted in high nitrate removal and a sufficient reduction of oxygen. A breakthrough of methanol during the test was excluded but not finally proved by methanol analyses of the filtrate.

Nitrate concentrations between 0.5 to 3 mg/L in the filtrate were obtained during stable filtration process. For the filtration tests at WWTP Horstermeer high nitrate conversion rates up to 3 $kgN/d/m^3_{bedvolume}$ were observed with finale nitrate concentrations below detection limit. Even in later winter periods with lower average temperature in the WWTP effluent of 13 ° and higher filtration rates (12.5 m/h), high conversion rates with a maximum of 3 $kgN/d/m^3_{bedvolume}$ were achieved.

Though the nitrate removal revealed less after backwash in the first hour of a new filtration run, the denitrification increased with run time. This was confirmed by experiments conducted with a pilot scale discontinuous filter applied for simultaneous nutrient removal with a filtration rate of 10 m/h as described by Jonsson et al. (1997). The expected negative influence of oxygen intake in filter bed due to air backwash on denitrification seemed to be negligible. Loose denitrifying biomass could be rinsed during backwash but efficient backwash with proper frequency at a longer term seemed to form a strong biofilm on the filter material resulting in quick recovery of denitrification after backwash.

Incidental insufficient methanol dosing resulted in fully restored denitrification within 20 min after 72 hours of interrupted dosing. This could indicate the robustness of denitrification applied for this filtration technique.

Together with high nitrate removal in the denitrifying filter also sometimes high Kjeldahl nitrogen concentrations in the WWTP effluent were found which may possibly inhibit the overall goal of reaching MTR-quality 2.2 mgN_{tot}/L. The reason should be found in the wastewater treatment plant itself. Incomplete bacterial decomposition of proteinaceous matter even together with less optimal nitrification due to low temperatures could be the possible explanations. Future full scale applications of post-denitrifying filtration might require further optimisation at the wastewater treatment plants.

However, strong variations in loading conditions for nitrate and turbidity showed no significant affect on the process reliability.

Phosphorus removal

The efficiency of phosphorus removal was significantly improved by proportional dosing of metal salt to the orthophosphate in the WWTP effluent. The problem of insufficient dosing in between the periods of manual adjustments was eliminated by online dosing. But still a proper metal-orthophosphate MePO ratio depending on the applied coagulant (poly aluminium chloride or ferric chloride) should be found. For the investigations at WWTP Beverwijk and Horstermeer ratios of MePO between 2 - 4.8 mol/mol were investigated. High dosing ratios resulted in the most optimal results for phosphorus removal.

However, the filtrate concentrations varied around the MTR value of 0.15 mgP_{tot}/L with average concentrations of 0.1-0.3 mgP_{tot}/l, including a maximum of 0.1 mg/L organic phosphorus. Fractions of orthophosphate and particulate phosphate were determined to evaluate the inorganic phosphorus removal. The sub-optimal removal may be addressed to the coagulating and flocculating properties of the different coagulants. The phosphate distributions and their removal results within the filtration process were used for this evaluation. The distribution of dissolved and particulate phosphate after coagulation and flocculation is a result of the applied type of coagulant, dosed amount and the coagulation conditions of the applied installations.

In accordance to the remaining particles in the filtrate between 2-20 µm further fractionation of particulate phosphate between 0.45 – 20 µm maybe be valuable for the specification of the filtration results of these phosphorus fractions. Some results of performed fractionations will be presented in chapter 7.

Also, the biological assimilation of phosphorus should be considered, even though it does not have a large contribution to the total phosphorus removal than the removal through chemical phosphate precipitation. A maximum removal of 0.2 mgP/L was obtained from regular analyses.

The removal of organic phosphorus was limited to the filtration of particulate P_{org}, while dissolved organic phosphorus remained not to be removed during filtration with a concentration of max. 0.1 mg/L in the filtrate.

Particle analyses and removal

Particle counting and analyses were performed as an additional monitoring tool and provide a valuable parameter to supplement the conventional analyses. Similar behaviour of particle and particulate phosphate distributions in samples before and after filtration could be evaluated. Particles of 10-20 and 20-100 µm were found to be filterable fractions while the particles of 2-10 µm showed lower filtration results.

Nevertheless, particle analyses should be considered more like a qualitative tool for evaluation of the filter performance. For example, the particle mass and distribution of the flocculated WWTP effluent could possibly be used for a description for a parameter called ‘filterability’, characterising a filterable feed water particle distribution. A qualitative particle number together with a particle volume distribution of particles and size classified particulate phosphate distribution could be used. However, the development of these possible characteristics for filter monitoring is not yet established. Further fractionation of particulate phosphate in relation to particle measurements as well to manual turbidity analyses during the filtration process was performed and presented in chapter 7.

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7

Additional results and possibilities for further process optimisation

7.1. Introduction

This chapter presents additional results and possibilities for further process optimisation investigated during simultaneous nutrient and suspended solids removal. Different additional investigations were performed on the three wastewater treatment plants WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer and their outcomes will be presented.

First of all profile measurements of nitrate, phosphate phosphorus, turbidity and oxygen will be shown. These investigations indicate the contribution of the total filter bed height and the two filtration layers (anthracite above and quartz sand below) to the overall removal of these constituents.

Next to that, results will be presented for the fractionations of phosphate, turbidity and particles conducted during different process conditions. These investigations were obtained to show the effect of different distributions on the filtration results and possible relation of these three parameters used for the evaluation of the process performance of flocculating filtration.

The denitrifying flocculating filtration tests were performed with a dual media filter in particular. The value of an additional third layer was investigated with two lab scale filter columns, applied after the pilot scale dual media filter. One column was filled with fine garnet sand and the other with granular activated carbon. The improvement of filtration results or the possible removal of additional constituents was evaluated.

Simultaneous removal of heavy metals in jar tests and filtration tests will be described as well as results for removal of other substances. Investigated other substances were various endocrine disrupting compounds (EDC's) and polycyclic aromatic hydrocarbons (PAH's). They are listed as relevant or possibly relevant substances for Dutch wastewater treatment plant effluents and are described in Chapter 1.

This chapter closes with a final evaluation of the presented results in relation to other practical experiences in literature.

7.2. Profile measurements

7.2.1. Results

During the lab scale filtration tests at the wastewater treatment plant WWTP Beverwijk nitrate profiles over the filter bed height were measured to investigate the extent of removal at different sampling points. The sampling points were located on different heights along the anthracite layer (ø 1.6-2.5 mm, height 80 cm) and quartz sand layer (ø 0.8-1.25 mm, height 40 cm). Measurements were performed on two different dates,

- date 1: 5 days after start-up of denitrification at a filtration run time of 4 hours, and
- date 2: 2 days later (7 days after start-up of denitrification) and 20 minutes after backwash (BW).

The filter was operated at a filtration rate of 10 m/h.

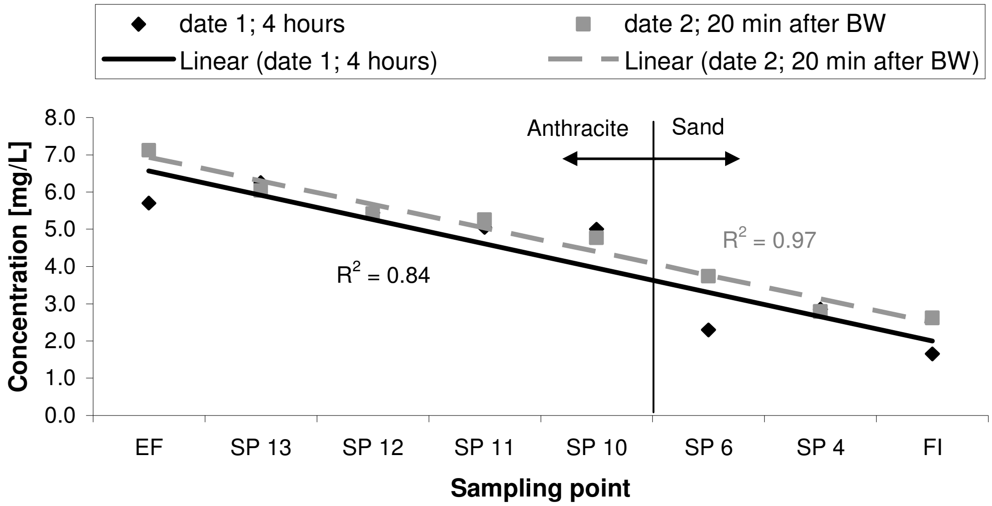

Figure 7. 1: Nitrate profile during lab scale denitrifying flocculating filtration at WWTP Beverwijk; BW: backwash, EF: WWTP effluent, SP 4-13: Sampling points at different heights, FI: filtrate

The concentration profile over the filter bed height in Figure 7. 1 showed a similar development for both dates. Beforehand it was assumed that most denitrification might occur within the anthracite layer due to a more open structure leading for increased biomass growth. The trend lines and high linear regression R^2 (> 0.8) on the other hand present a continued nitrate removal throughout both layers.

Abrasion and wash out of biomass during backwash applying an air backflush phase turned out to be less problematic than expected. This is shown by the comparison of profiles at date 1 measured 4 hours after the last backwash and at date 2 measured 20 minutes after backwash. Denitrification capacity was not greatly affected by the strong turbulence created during air backwash; the overall conversion rates stayed almost equal (>1 $kgN/d/m^3{}_{bedvolume}$).

Profile measurements were also performed during the pilot scale filtration tests at the WWTP Horstermeer. The concentrations were also measured at a filtration rate of 10 m/h applied for simultaneous nutrient removal. The measurements were performed at the sampling points of the WWTP effluent (EF), the flocculated WWTP effluent (AF), sampling points over filter bed (SP 1-15) and the filtrate (FI).

Figure 7. 2 shows a linear removal of phosphate, fractioned in particulate and dissolved form ($P_{diss+part}$), and turbidity. The linear regression results in $R^2 = 0.96$ and $R^2 = 0.80$, respectively. Comparable to the removal of nitrate described before, the removal of

phosphate and turbidity is also not determined by the filtration media indicating a contribution of the whole filter bed for the total removal of those parameters.

Contradictory to these results van der Graaf (2001) described predominant particle removal in the upper part of anthracite layer during flocculating filtration. Only minor improvement was observed in the second or even third filtration layer. A similar behaviour of phosphate was assumed when considering suspended matter with a phosphorus content of 1 - 4% (Van der Graaf, 1996).

Nevertheless, the presented results show that phosphate is removed throughout the whole filter bed. This can indicate continued flocculation, and adsorption possibly combined with biological phosphorus uptake in the filter bed. Consequently, the accumulation capacity of the whole filter bed may be used more efficiently and the filtration of suspended matter including particulate phosphate is improved.

Compared to the linear regression of the phosphate concentrations the correlation for the turbidity values turned out to be weaker. This can be addressed to lower accuracy of the light scattering principle applied for turbidity measurement especially when using small volumes for grab sampling.

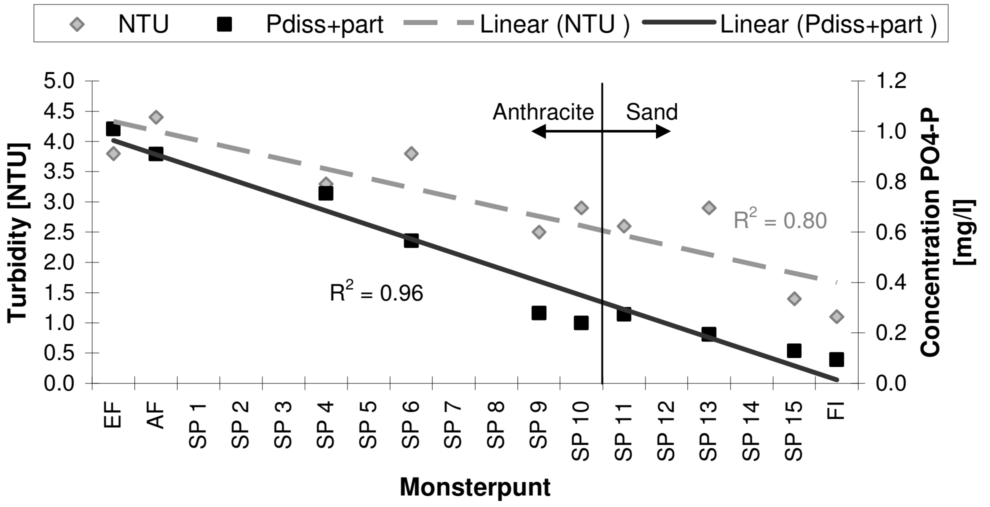

Figure 7. 2: Total phosphate ($P_{diss+part}$) and turbidity profile during pilot scale denitrifying flocculating filtration at WWTP Horstermeer; EF: WWTP effluent, AF: flocculated WWTP effluent, SP 1-15: Sampling points at different heights, FI: filtrate

The profile measurements in Figure 7. 3 show no difference of nitrate removal by the filtration medium resulting in the square of the correlation coefficient of R^2 of 0.98 for linear regression. Nitrate is removed to a very low concentration of less than 0.5 $mgNO_3$-N/L in the filtrate with a conversion rate of 2.5 kgN/d/m^3 $_{bedvolume}$. Removal of oxygen also takes place within the whole filter bed up to a level of approx. 1.0 mgO_2/L. This end concentration was frequently measured during other experiments.

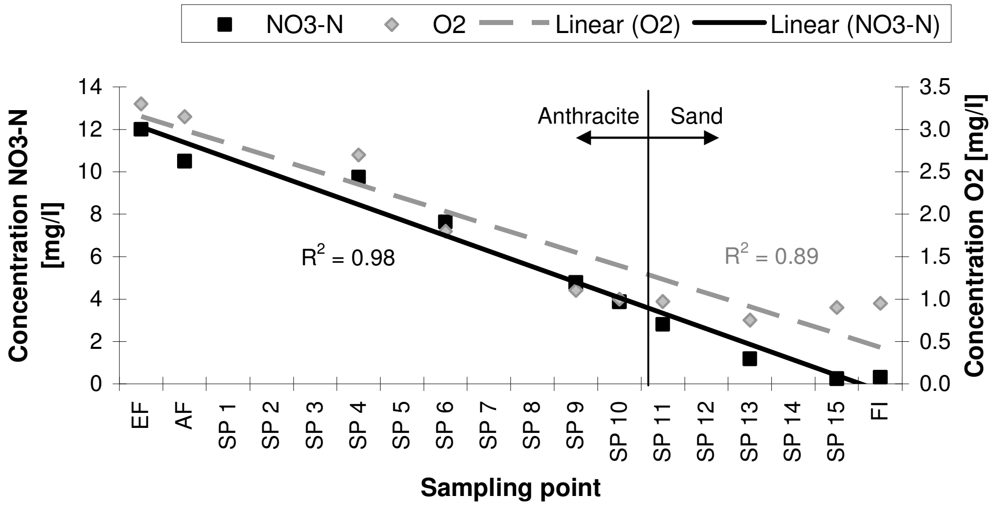

Figure 7. 3: Nitrate (NO_3-N) and free oxygen profile during pilot scale denitrifying flocculating filtration at WWTP Horstermeer; EF: WWTP effluent, AF: flocculated WWTP effluent, FI: filtrate

These high correlation coefficients for the presented profile measurements were confirmed with several additional measurements obtained during stable operation.

7.2.2. Conclusions

Van der Graaf (2001) *et al.* reported major particle removal in the first centimetres of the upper filter layer. That would consequently mean that not the whole filter bed is used for the accumulation and filtration of particulate matter. This would also count for the precipitated phosphate. Also considering the different surface properties of sand and anthracite available for biomass growth is expected to determine visibly the nitrate removal over the two layers.

However, the results for the profile measurements presented in this chapter indicated the effective usage of the whole filter bed. The combination of flocculating filtration, with inline coagulant dosing, and floc forming before filtration in the water head of 1.96 m above the filter bed, resulted in depth filtration using the available accumulation capacity of the filter effectively.

Decreasing concentrations of total phosphate($P_{part+diss}$), turbidity, nitrate (NO_3-N) and oxygen (O_2) are measured at the sampling points along the filter bed height. High correlations with a square of the linear regression of R^2 above 0.8 were found for those parameters showing the use of the whole filter bed for the removal.

The low average nitrate concentrations in the filtrate for the pilot plant investigation at WWTP Horstermeer indicate the sufficiency of the available biomass with the chosen filtration rate of 10 m/h and methanol dosing ratio of 4.8 gCH_3OH/gNO_3-N. Even if the oxygen levels in the filtrate are above the optimal concentration for denitrification no inhibition of nitrate removal is observed. This indirectly leads to the conclusion that the chosen filter bed configuration is very effective for the simultaneous nutrient removal to very low levels.

7.3. Fractionation of phosphorus, turbidity and particles

7.3.1. Results

The results of the phosphate fractionation in dissolved and particulate form obtained from the filtration experiments at the wastewater treatment plant (WWTP) Horstermeer, described in Chapter 6, led to the investigation applying a more detailed fractionation. The particle size range of 2-30 µm was already specified as particle size range which is difficult to filtration with the applied granular medium filtration. Therefore the fractionation aimed to show the distribution of particulate phosphate in this range and their filtration results.

Six pilot scale filtration tests, operated as denitrifying flocculating filtration, were performed using two different poly aluminium chloride products (PACl and PAX11) and variable initial mixing. The variation of the initial mixing energy was conducted by narrowing a cut-off valve before the installed static mixer. The dosing of both aluminium products occurred with a fixed metal-orthophosphate Me/PO ratio of 4.7.

Samples of the flocculated WWTP effluent (AF) and filtrate (FI) were collected for the determination of total phosphate ($P_{diss+part}$) and turbidity (manual turbidity meter, Hach-Lange). The analyses were conducted for the unfiltered samples and the five permeates, pre-filtered samples using polycarbonate membrane filters with the pore sizes of 20 µm, 10 µm, 2 µm, 0.45 µm.

It was expected beforehand that the fractionation may demonstrate changes in particulate phosphate distributions due to:

- continued adsorption, precipitation, coagulation and flocculation processes in the filter bed during filtration,

- the use of two different products of poly aluminium chloride, PACl and PAX11, containing different sulphate and active aluminium (Al_2O_3) concentrations influencing the floc strength and formed particle size,
- variation of initial mixing by narrowing a cut-off valve before the static mixer.

The operational details for the performed fractionation tests are presented in Table 7. 1.

Table 7. 1: Details of filter operation for fractionation tests at WWTP Horstermeer; PACl/PAX 11: poly aluminium chloride

Test No	Date	Filter run time; current (total)	Type poly aluminium chloride	Status cut-off valve
Test 1	04-05-’06	9 (10 hours)	PACl	open
Test 2	12-06-’06	3 (15 hours)	PACl	open
Test 3	22-06-’06	13 (15 hours)	PACl	half closed
Test 4	23-10-’06	4 (24 hours)	PACl	half closed
Test 5	13-11-‘06	16 (24 hours)	PAX11	half closed
Test 6	01-12-’06	4 (24 hours)	PAX11	half closed

Figure 7. 4 shows the distributions of total phosphate in the flocculated effluent. The comparison of Tests 1 and 2 with the Tests 3 and 4 indicates no major differences in distributions varying the initial mixing energy.

The particulate phosphorus for the Tests 1 to 4 seems to mainly consist of phosphorus particulates with a size of 0.45 µm - 2 µm representing 10-70% of the total inorganic phosphorus concentration. Flocs with a size above 20 µm represent approx. 10-20%. The particle fraction considered as easily filterable with media filtration in the range >2 µm to >20 µm is indicated (grey shaded columns).

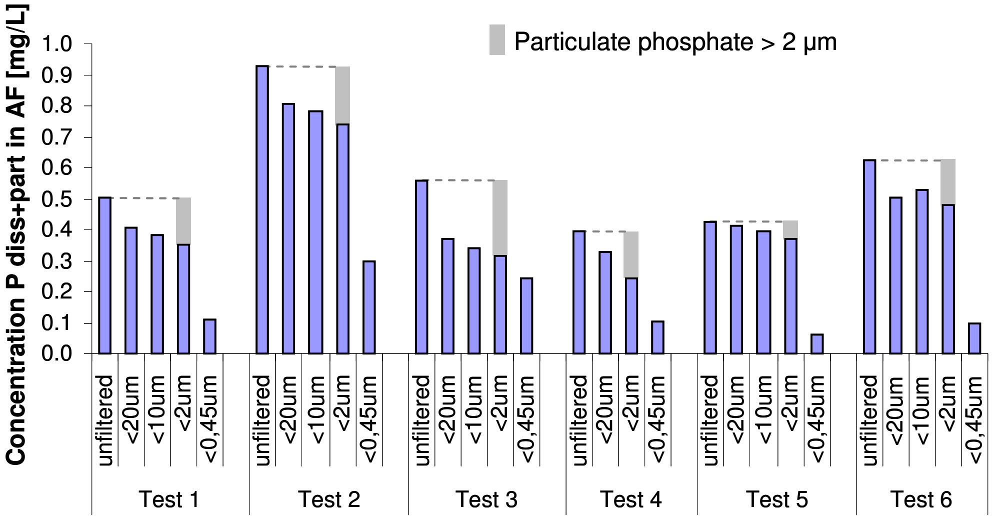

Figure 7. 4: Fractionation of total phosphate ($P_{diss+part}$) for the differently pre-treated samples of the flocculated WWTP effluent (AF) for the denitrifying flocculating filtration tests at WWTP Horstermeer

The choice of poly aluminium chloride shows an effect on the phosphate distribution. Max. 85% of the particulate fraction after flocculation with PAX11 is present between 0.45 µm and 2 µm. The concentration measured for this small particle size range is increased towards the other fractions.

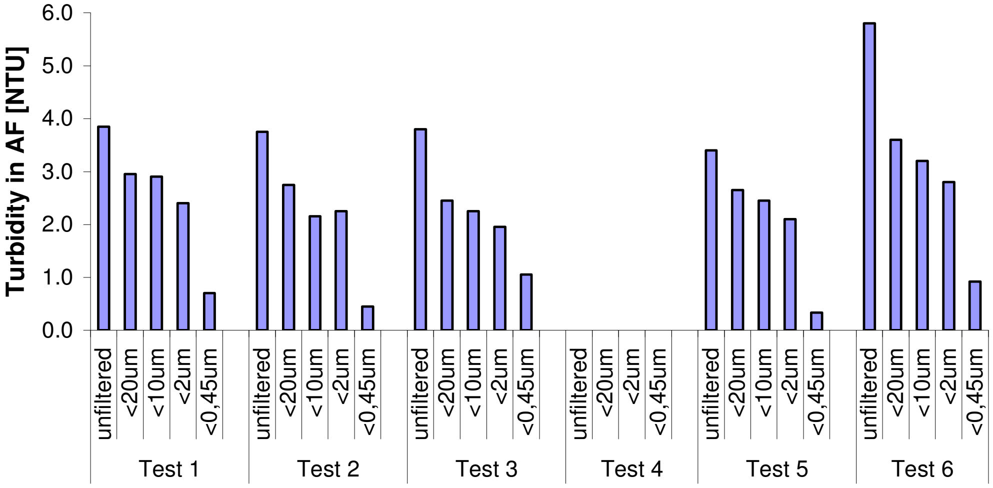

Figure 7. 5: Turbidity analyses for the differently pre-treated samples of the flocculated WWTP effluent (AF) for the denitrifying flocculating filtration tests at WWTP Horstermeer

The turbidity analyses (Figure 7. 5) of the same samples show similar distributions compared to the total phosphate. Turbidity response is decreasing like the phosphorus concentrations in the series of fractionations. But also the distribution of the fractions seems comparable for turbidity and phosphorus. The phosphorus fractions causing the highest response of phosphorus and turbidity are found above 20 µm and between 0.45 µm - 2 µm.

The linear regression of the turbidity values versus total phosphate concentrations measured in the different permeates obtained for the flocculated effluent is shown in Figure 7. 6. Good correlations of $R^2 > 0.9$ are found for total phosphate concentrations between 0.3 and 0.9 mg/L. For a phosphate concentration above 1 mg/L only a poor correlation of $R^2 = 0.71$ is found.

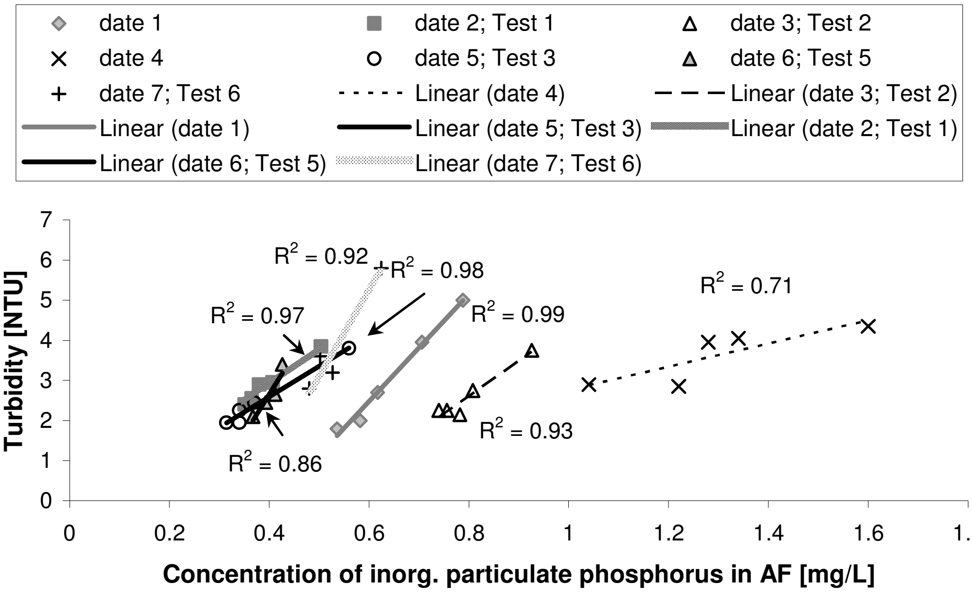

Figure 7. 6: Concentration of turbidity [NTU] versus particulate phosphate P_{part} [mg/L] in the flocculated WWTP effluent (AF) for different dates over all permeates, linear regression and correlation coefficients (R^2) conducted at pilot scale investigations at WWTP Horstermeer

The comparison of Figure 7. 4 and Figure 7. 7 before and after filtration indicate that the particles above 2 µm are greatly removed (indicated by the grey shaded columns), up to 90% for the tests with poly aluminium chloride and up 75% for the tests with PAX11.

Even if the dissolved fraction seems to be slightly removed, possibly due to biological assimilation or continued precipitation in the filter bed, a remaining concentration of averagely 0.1 mg/L (range 0.025-0.25 mg/L) was measured in the filtrate.

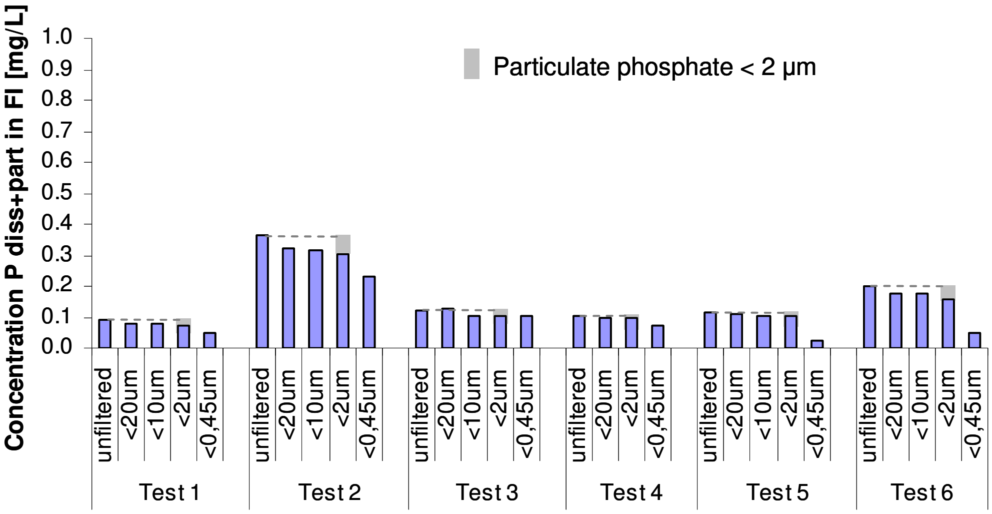

Figure 7. 7: Fractionation of total phosphate ($P_{diss+part}$) for the differently pre-treated samples of the filtrate (FI) for the denitrifying flocculating filtration tests at WWTP Horstermeer

For the tests 1 to 4 the dissolved fraction still represents 50-90% of the total phosphate.

A different picture is shown for the tests 5 and 6 using another poly aluminium product PAX11 as coagulant. This product contains a higher active aluminium (Al_2O_3) and lower sulphate (SO_4^{2-}) content than PACl. After flocculation approximately 30% remains as orthophosphate and 70% of the total phosphate is in particulate form in the size range of 0.45 to 2 µm. This fraction is already described before as problematic for granular medium filtration.

However, the results indicate that processes of precipitation, coagulation and flocculation previous to filtration and the choice of coagulant influence the phosphate distribution and consequently the overall removal. A good filterable floc size and strength can be achieved with optimisation of these processes.

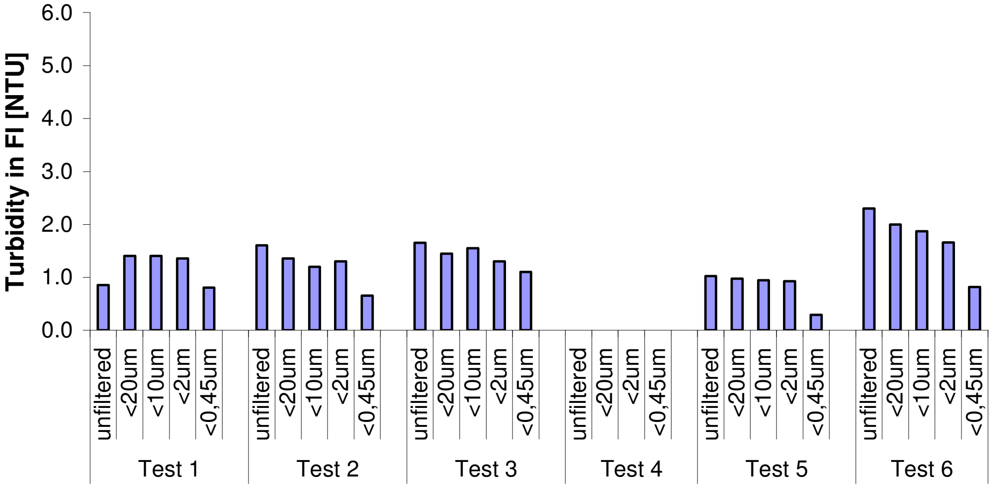

Figure 7. 8: Turbidity analyses for the differently pre-treated samples of the filtrate (FI) for the denitrifying flocculating filtration tests at WWTP Horstermeer

The manual turbidity analyses of the same differently pre-treated samples of the filtrate (FI) in Figure 7. 8 show similar distributions like described before for the phosphate distributions. The turbidity values measured for these low turbidity permeates seems to be very well related to the present small particulate phosphate flocs between 0.45 and 2 µm.

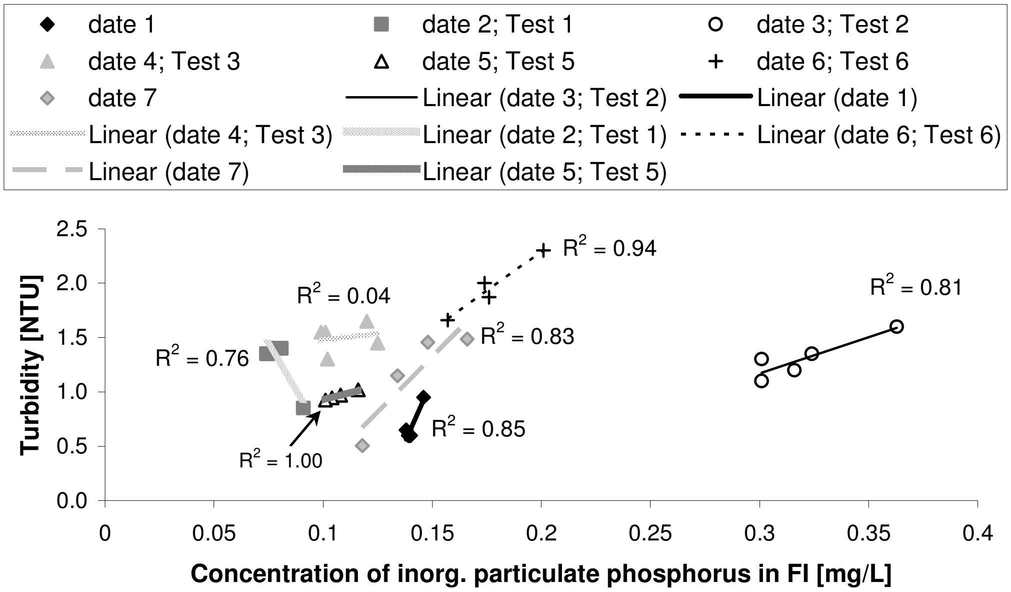

Figure 7. 9: Concentration of turbidity [NTU] versus total phosphate [mg/L] in the filtrate FI for different dates over all permeates, linear regression and correlation coefficients (R^2) conducted at pilot scale investigations at WWTP Horstermeer

The linear regression and the square of the correlation coefficient R^2 for the fractionations obtained for the filtrate resulted in values above 0.8 for total phosphate concentrations between 0.1 and 0.85 mg/L.

7.3.2. Conclusion

The phosphate fractionations performed with two different initial mixing conditions and dosing of two poly aluminium chloride products indicated minor influence in phosphate distributions due to narrowing the cut-off valve before the static mixer.

The dosing of another poly aluminium chloride product (PAX11) on the other hand resulted in a high concentration of particulate phosphate between 0.45 and 2 µm. However, this fraction can only be partly removed by granular dual medium filtration and is considered as problematic to filtration. The poly aluminium chloride PACl seems to form more particulate phosphate bigger than 2 µm than PAX 11. This resulted consequently in better filtration results.

It can be concluded that the complete reaction of orthophosphate in the first place and the creation of filterable particulate phosphate in the second place seems necessary for an optimal removal. This can be achieved by optimisation of the precipitation, coagulation and flocculation processes before filtration and a proper choice of the coagulant type.

Further, turbidity showed a strong relation to particulate phosphate (>0.45 µm to >20 µm) in the range of 0.1-0.9 $mgP_{part+diss}/L$ at the sampling points for the flocculated WWTP effluent (AF) and the filtrate (FI). The correlation of turbidity measurements and total phosphate concentrations indicated R^2 above 0.8 for that concentration range. This observation is confirmed by the correlation of measurements for three fractionations conducted for the WWTP effluent (EF) shown in Figure 7. 9.

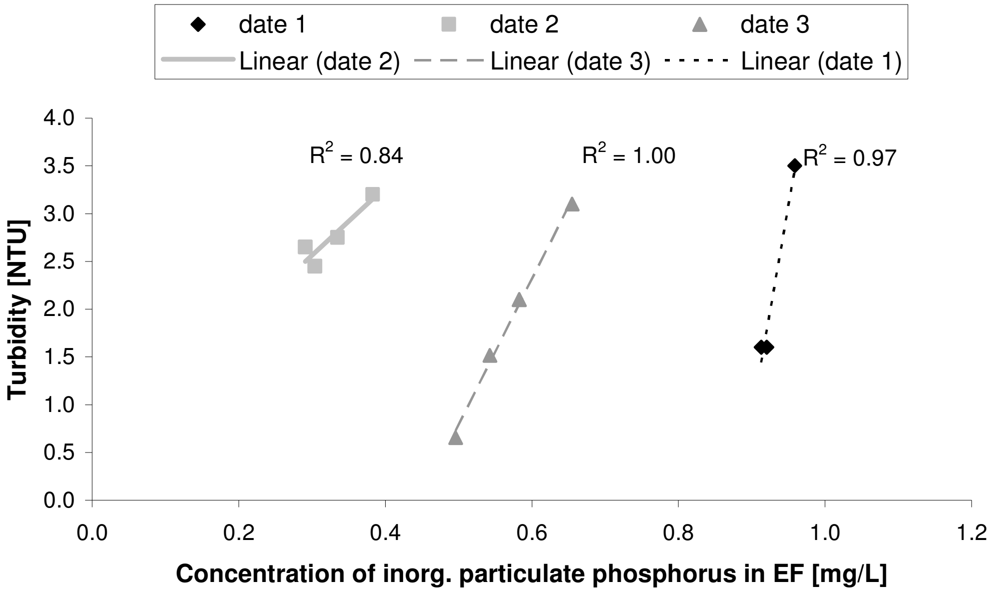

Figure 7. 10: Concentration of turbidity [NTU] versus total phosphate [mg/L] in the WWTP effluent (EF) for different dates over all permeates, linear regression and correlation coefficients (R^2) conducted at pilot scale investigations at WWTP Horstermeer

The poor correlation for total phosphate concentrations below 0.1 mg/L may be due to greater influence of the accuracy of the applied cuvette test (0.006 mg/L; Hach Lange) at low concentrations. The steep slope of the trend lines of the linear regressions could indicate that during fractionation not only particulate phosphate is removed. Other suspended matter like humic acids and other colloids affecting turbidity may be also removed due to fractionation. Simultaneously to high phosphate content (>0.85 mg/L) these substances may lead to higher turbidity values in bigger fractions resulting in a weak correlation of turbidity versus total phosphate above that concentration.

7.4. Removal of conventional parameters with three layer filtration

7.4.1. Results

In 2005, the first year of investigations at the wastewater treatment plant (WWTP) Horstermeer, the possible improvement of removal efficiencies with a third layer following the dual media filter was tested. Two lab scale columns were filled with different filter materials. One consisted of a fine garnet sand layer (0.5 – 0.8 mm grain diameter, 30 cm height) and the other of an activated carbon layer (1.6 mm grain diameter, 1.2 m height). The feed water was distributed from the buffer tank storing the filtrate of the dual media installation. The two filters were operated in parallel without any further chemical conditioning.

A filtration rate of 10 m/h was applied for the column with the fine sand layer. The activated carbon column was operated at a flow rate maintaining an empty bed contact time of 10 min resulting in a filtration rate of 7 m/h. This empty bed contact time was chosen in order to gain results under similar process conditions with earlier performed tests at WWTP Utrecht (Van Oene, 2004; Te Poele *et al.*, 2004; Menkveld *et al.*, 2005).

Table 7. 2 gives an overview of all conducted measurements expressed as average concentrations with standard deviation (stdev) for the sampling points:

- Filtrate buffer (FI_buffer): storage of dual media filtrate and feed water for lab scale installations,
- Filtrate fine sand filter (FI_fs), and
- Filtrate activated carbon filter (FI_ac).

In order to retrieve measurable results the filtrate of the dual media filter still had to contain various components; this was achieved by the application of suboptimal operating conditions of the dual media filter.

Table 7. 2: Average concentrations and standard deviations (stdev) for several parameters measured in the filtrate buffer (FI_buffer), filtrate fine sand (FI_fs) and filtrate activated carbon (FI_ac) in 2005 at the WWTP Horstermeer (Menkveld *et al.*, 2005a and 2006)

		Filtrate buffer (FI_buffer)		Filtrate fine sand (FI_fs)		Filtrate activated carbon (FI_ac)	
Parameter	Unit	average	stdev	average	stdev	average	stdev
CZV*	[mg/L]	44.3	14.5	35.7	11.4	24.5	9.2
N_{tot}*	[mg/L]	5.3	4.6	3.8	3.9	3.6	4.4
NO_3-N	[mg/L]	3.1	3.8	1.6	2.8	1.0	2.0
P_{tot}*	[mg/L]	0.36	0.23	0.25	0.15	0.25	0.15
PO_4-P*	[mg/L]	0.30	0.23	0.23	0.28	0.19	0.17
Sulphate	[mg/L]	57.7	13.9	58.5	11.4	58.3	12.8
Turbidity	[NTU]	2.2	2.0	1.21	0.68	1.20	0.67
pH	[-]	6.45	0.34	6.65	0.28	6.78	0.27
O_2	[mg/L]	2.9	1.0	2.5	1.0	2.2	0.8

* Unfiltered samples used for analyses

The concentrations of nitrate measured in the filtrate buffer tank are slightly higher compared with the nitrate concentrations established in the filtrate outlet at the dual media filter (see Chapter 6). Residual easily degradable organic matter in the feed water seems to enhance continued denitrification in both lab scale columns. Further removal of the chemical oxygen demand (COD), nitrate (NO_3-N) and free oxygen (O_2) is observed. This is confirmed by the increased pH in both filtrates compared to the filter feed water (FI_buffer).

Furthermore, also the total phosphorus concentration is further reduced, corresponding to lowering the turbidity values. It is remarked that the phosphorus removal derives exclusively from the filtration of particulate phosphate. The organic phosphorus is not significantly reduced, also not by adsorption in the activated carbon filtration.

Furthermore, the turbidity values for both third layer filtrates are lower than the dual media filtrate.

7.4.2. Conclusion

For simulation of a third layer in the dual media filter two lab scale columns are fed with dual media filtrate from the buffer tank. The application of this lab scale installation externally is not totally representative to an integrated third layer. Also some change of feed water characteristics before feeding the lab scale columns needed to be considered for this configuration.

Anyhow, trends for improved removal of the selected parameters are indicated. The final filtrate quality after third layer filtration is improved compared to dual media filtration regarding nitrate, chemical oxygen demand, turbidity and total phosphate. The removal of total phosphate is attributed to the filtration of remaining particulate phosphate which is higher in the activated carbon column than in the fine sand column. The turbidity is removed in both columns to almost equal values.

A decrease of oxygen and nitrate concentrations indicated further denitrification in the lab scale columns.

7.5. Simultaneous removal of heavy metals

7.5.1. Jar tests

Next to the removal of the so-called conventional parameters, the possibility of simultaneously removing other substances like e.g. heavy metals was another issue of investigation. Indicative jar tests preceded the filtration tests at the wastewater treatment plant (WWTP) Horstermeer.

These tests were conducted with water from the WWTP effluent buffer applying the process parameters, e.g. coagulation, flocculation time and energy input, used in the pilot scale installation. A standard sedimentation time of 30 min was applied for all jar tests with following analyses of the supernatant. Standard solutions for heavy metals (Nickel: 1000 mg Ni/L, Zinc: 100 mg Zn/L, Copper: 100 mg Cu/L) were dosed in order to get measurable concentrations for the direct determination with cuvette analyses.

The removal efficiencies for the heavy metals were compared under three different dosing conditions, dosing of (1) poly aluminium chloride (PACl), (2) ferric chloride or (3) powdered activated carbon (Norit W35). The dosing of coagulant aimed the removal mechanism co-precipitation with metal ions. It was refrained from the pH adjustment which might enhance the efficiencies of this mechanism. The expected negative effect on the denitrifying biomass led to the omission of pH adjustment when thinking of pilot or full scale filtration tests for simultaneous removal.

The dosed amount of the coagulant is given as MePO ratio (mol metal per mol orthophosphate). For the jar test with powdered activated carbon, the supernatant is pre-filtered over a 63 μm suspended solids filter before performing the analyses.

The results of heavy metal removal with co-precipitation, determined for unfiltered samples (Table 7. 3), showed a relatively high removal of copper (79%) at a dosing of aluminium with MePO = 4.4 (mol metal per mol PO_4-P). The co-precipitation of nickel with aluminium resulted in a maximum removal of 15%, with a high metal dosing ratio of 11.4 MePO. Zinc and copper are removed with 38% and 20%, respectively, when using ferric chloride as coagulant.

Table 7. 3: Summary of maximum removal efficiencies, for co-precipitation and adsorption, during jar tests conducted at WWTP Horstermeer; in %; MePO: metal-orthophosphate ratio (Menkveld *et al.*, 2005a)

	Removal efficiencies [%]		
Metal	Co-precipitation with Fe^{3+}	Co-precipitation with Al^{3+}	Adsorption with powdered activated carbon
Zinc	38 (MePO 4.4)	nd	>95 (1 g/l PAC) *
Nickel	nd	15 (MePO 11.4)	>95 (3 g/l PAC) *
Copper	20 (MePO 3.1)	79 (MePO 4.4)	nd

* : Removal efficiency of above 95% cannot be shown due to the measuring range of the cuvette analyses; nd: not determined

The dosage of powdered activated carbon resulted in a removal efficiency of above 95% for zinc with 1 g/L carbon. A similar removal is achieved for nickel with an addition of 3 g/L activated carbon.

The partial removal of the three selected heavy metals indicated a promising performance for the combination of co-precipitation and/or adsorption within the media filtration applied for simultaneous nutrient removal.

7.5.2. Dual media filtration

The removal of heavy metals was investigated during the pilot scale project on the WWTP Horstermeer. In order to get heavy metal concentrations measurable with the used cuvette analyses the filter feed water, the pre-sieved WWTP-effluent, was spiked with a heavy metal solution (with copper, nickel and zinc). This solution was prepared of metal chlorides and dosed to maintain a concentration of 120-150 μg/L of each metal.

Poly aluminium chloride (PACl) was dosed as coagulant for chemical phosphate precipitation with a metal-orthophosphate ratio MePO of 5.5 to 9.2. Methanol is applied as a carbon source for denitrification in dosing ratios of 4.8 gCH_3OH/gNO_3-N.

Heavy metal concentrations are obtained at the sampling points (1) AF, the flocculated WWTP effluent, filter feed water after dosing of poly aluminium chloride (PACl), methanol and heavy metal solution, and (2) FI, the filtrate.

The analyses of the raw samples are indicated as “unfiltered”, representing the total inorganic metal content while the analyses of the pre-filtered samples described with “<

0.45 µm” represent the dissolved metal fraction. The difference between the two measurements at each sampling point represents the particulate fraction of the heavy metals. The results are displayed as average concentrations measured for three different test conditions:

- Test 1: MePO = 5.5; CH_3OH/NO_3-N = 4.8; average concentrations of heavy metals out of 5 measurements conducted during 2 days,
- Test 2: MePO = 0; CH_3OH/NO_3-N = 4.8; average concentrations of heavy metals out of 14 measurements conducted during 8 days.
- Test 3: MePO = 9.2; CH_3OH/NO_3-N = 0; average concentrations of heavy metals out of 5 measurements conducted during 2 days.

In order to investigate the extent of biological uptake of heavy metals through denitrifying biomass contributing to the heavy metal removal in Test 3, the dosage of methanol was stopped for two days before performing the analyses.

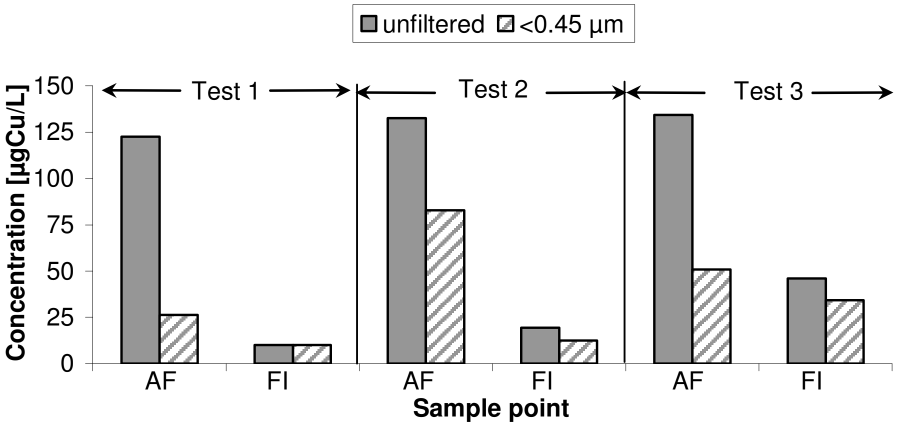

Figure 7. 11: Concentrations of total (unfiltered) and dissolved (<0.45 µm) copper for the flocculated WWTP effluent (AF) and filtrate (FI) during the filtration tests, Test 1, Test 2 and Test 3, performed at WWTP Horstermeer; in [µg/L]

Figure 7. 11 gives the results for the copper analyses during the three different test conditions. The bulk of copper in AF is present in particulate form (difference between “unfiltered” and “< 0.45 µm”) and is highly removed due to filtration in Test 1. Also the dissolved fraction of copper is removed in the filter (difference between “< 0.45 µm” in sampling points AF and FI). At sampling point FI the concentrations reached the detection limit of the analyses resulting in a removal efficiency of copper, measured in the unfiltered sample, higher than 92%.

In Test 2 the fraction of dissolved copper in AF appeared to be higher than in Test 1. Even though, the copper removal is high with filtration. The concentrations reached almost the detection limits of the analyses. The removal efficiency of the fraction of the total inorganic copper content (“unfiltered”) is 85%.

Under the test conditions with inactivated biomass but with dosing of coagulant in Test 3, the removal efficiency of the total inorganic copper is 66% and therefore less than in the previous Test 1 and 2.

The measurements for nickel present remarkably different results compared with the results for copper (Figure 7. 12). After dosing (AF), nickel is found mainly in dissolved form (“< 0.45 µm”). The dual media filter did not retain this fraction and the removal is only 15% during Test 1. Test 2 shows a slightly higher removal efficiency of 25%. In test 3, with inactivated biomass, no removal of nickel is observed. The concentration of dissolved nickel even slightly increases in the sampling point FI compared to AF.

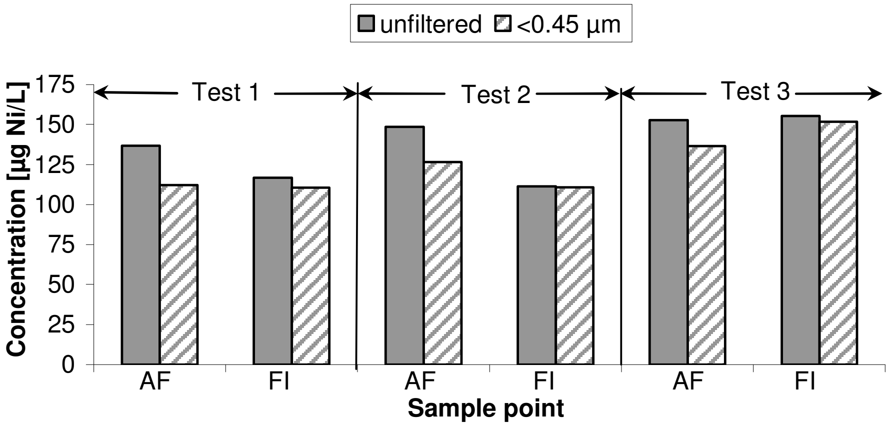

Figure 7. 12: Concentrations of total (unfiltered) and dissolved (<0.45 µm) nickel for the flocculated WWTP effluent (AF) and filtrate (FI) during the filtration tests, Test 1, Test 2 and Test 3, performed at WWTP Horstermeer; in [µg/L]

Figure 7. 13 shows the fractionation of zinc for the three filtration tests. In Test 1 and 2 the major concentration of zinc is in dissolved form but is partly removed in the dual media filter with approximately 60%. In Test 3 the removal of particulate zinc is less than in the previous tests. Even an increase of the dissolved fraction was observed.

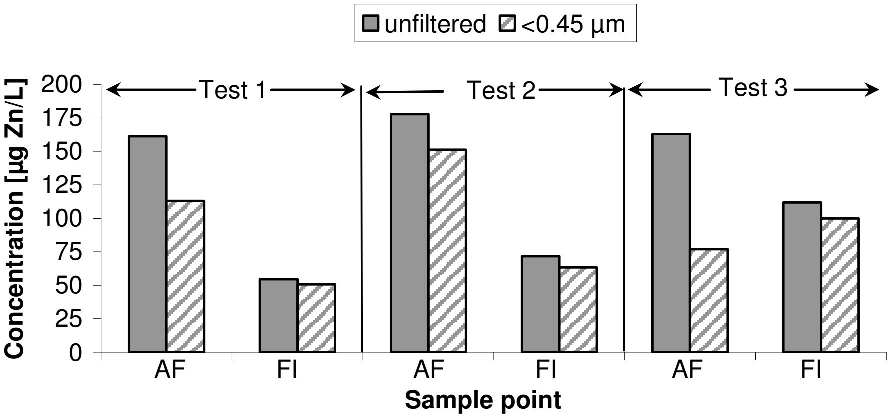

Figure 7. 13: Concentrations of total (unfiltered) and dissolved (<0.45 µm) zinc for the flocculated WWTP effluent (AF) and filtrate (FI) during the filtration tests, Test 1, Test 2 and Test 3, performed at WWTP Horstermeer; in [µg/L]

7.5.3. Three layer filtration

Additional to the filtration results of heavy metals with dual media filtration further removal with following third layer filtration through fine sand or activated carbon was investigated. For these filtration tests the dosing of heavy metal solution before dual media filtration was increased to the double amount in order to maintain measurable concentrations before and after third layer filtration

Figure 7. 14 shows the heavy metal analyses in the filtrate buffer (sampling point FI_buffer), feed water of the lab scale installation, and the filtrates of the two single media columns (Fine sand FI_fs and activated carbon FI_ac) conducted at the same time as the analyses for dual media filtration. The analyses were conducted on unfiltered samples without pre-filtration. The evaluation of the heavy metal analyses occurred for:

- Copper: average concentration out of 4 measurements during 2 days
- Nickel: average concentration out of 11 measurements during 5 days
- Zinc: average concentration out of 11 measurements during 5 days.

For the copper analyses only the measurements with the highest copper concentrations are displayed in Figure 7. 14 in order to indicate a removal in the lab scale installation within the measuring range of the used analyse method (cuvette analyses).

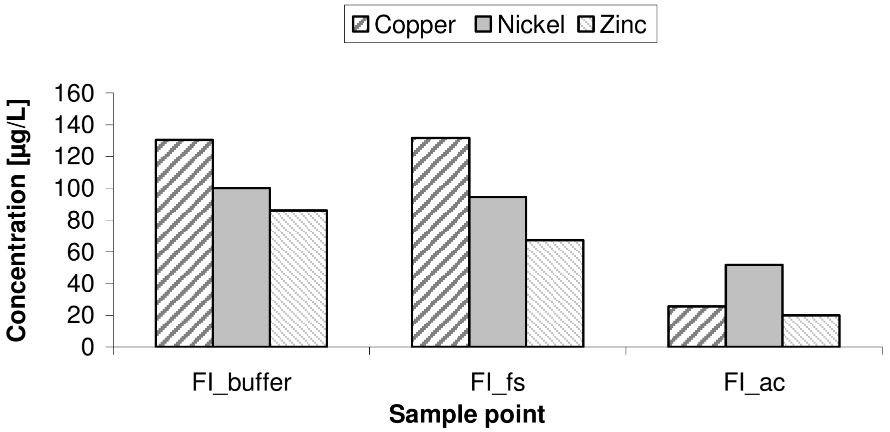

Figure 7. 14: Removal of copper, nickel and zinc in the lab scale installation consisting of one fine sand (FS) and activated carbon (AC) column applied at WWTP Horstermeer, in [µg/L]

The removal efficiencies in the activated carbon column are higher than in the fine sand column. The heavy metal present in the filtrate buffer (FI_buffer) passed almost completely through the lab scale column filled with fine sand. Only a small amount of nickel and zinc was removed with filtration resulting in efficiencies of 13% and 22%, respectively.

On the other hand removal efficiencies of 81% for copper, 67% for nickel and > 88% for zinc are achieved with activated carbon filtration.

At least, these results indicated relatively high removal for all three heavy metals through media filtration applied for simultaneous nutrient removal combined with activated carbon adsorption.

7.5.4. Conclusions

The preliminary jar tests performed with addition of coagulant (poly aluminium chloride and ferric chloride) aimed at the removal with co-precipitation. The highest removal is found for the precipitation of copper with aluminium ions. This may be explained by the behaviour of copper forming easily complexes with filterable suspended matter (Zwolsman, 2005).

The removal of heavy metals during dual media filtration reached the highest value for all three selected metals during operation for simultaneous nutrient removal, with efficiencies of >92%, 10-20% and 60% for copper, nickel and zinc, respectively. These removal efficiencies derived from the combination of the main removal mechanisms like filtration, adsorption, co-precipitation and biological uptake. The third layer filtration with activated carbon also resulted high heavy metal removal. Above 80% of copper and zinc, and >60% of nickel is removed; most probably due to adsorption and biological uptake.

The high removal with activated carbon adsorption was already indicated during the jar tests.

Uzun and Güzel (2000) reported the removal of the heavy metals manganese, iron, nickel and copper from aqueous solution using activated carbon (Merck standard M – 2514). On the other hand, complexation of heavy metals with organic matter with following adsorption of the organics is more likely the responsible mechanism.

Consequently, the integration of activated carbon adsorption within the denitrifying flocculating filter offers the additional removal of at least the complexed heavy metal fractions.

7.6. Removal of other substances

7.6.1. Results

During the pilot scale investigations at the wastewater treatment plant WWTP Utrecht and WWTP Horstermeer also the removal of several other substances, considered as endocrine disrupting compounds (EDC’s) and polycyclic aromatic hydrocarbons (PAH’s), was investigated. These substances are partially considered in the list of priority (hazardous) substances in the annex of the Water Framework Directive WFD. Due to their presence in Dutch wastewater treatment plant effluent several of these substances are considered as ‘relevant’ or ‘possibly relevant’ substances.

Only a few measurements of some selected compounds bisphenol A, nonylphenol and polycyclic aromatic hydrocarbons were performed occasionally. The omission of an extended and regular screening is attributed to the preliminary assumption of insignificant removal during denitrifying flocculating filtration.

However, the concentration of bisphenol A in the effluent at WWTP Utrecht was calculated to 140 ng/L as an average of two measurements. During denitrifying flocculating filtration with a dual media filter 30 ng/L was removed.

The removal of nonylphenol was analysed as well for two different days. From the results no clear removal was indicated for the removal with dual media filtration. One time nonylphenol was reduced with 20% and the other time an increase of 30% was found.

During the pilot scale research at the WWTP Horstermeer the Water Board Waternet conducted an effluent screening twice regarding the substances considered in the annex of the WFD, especially focussing on polycyclic aromatic hydrocarbons (PAH's).

The following Table 7. 4 shows the concentrations of PAH's measured in the WWTP effluent and in the filtrate of the dual media filter. Most of the substances even in the filter feed water are present in concentrations below the measuring range of the applied method. Clear removal through dual media filtration can be observed for the substances acenaftene, acenaphtylene, naftalene and pyrene.

Table 7. 4: Concentrations of polycyclic aromatic hydrocarbons (PAH's) measured during effluent screening at WWTP Horstermeer; conducted at 09-02-06

		WWTP-effluent [µg/l]		Filtrate dual media [µg/l]
Acenaphttylene		0.05	<	0.05
Acenaftene, Anthracene, Benzo(a)anthracene, Benzo(a)pyrene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Chrysene, Dibenzo(a,h)antracene	<	0.01	<	0.005
Benzo(g,h,i)perylene	<	0.01	<	0.008
Fenanthrene	<	0.01	<	0.029
Fluorene	<	0.01	<	0.22
Fluoranthene		0.02		0.007
Ideno(1,2,3-c,d)pyrene	<	0.01	<	0.0022
Naftalene		0.01	<	0.0039
Pyrene		0.01	<	0.0012
Sum PAH	<	0.16	<	0.2
Pentachlorphenol	<	0.01	<	0.01

For the other compounds removal can not be justified because already the concentration measured in the filter feed water is obtained below the detection limit of the applied method. Further screenings may necessary in order to gain more representative results.

7.6.2. Conclusions

The removal of micro pollutants during filtration experiments showed to be rather low. Next to bisphenol A also several polycyclic aromatic hydrocarbon showed to be reduced with denitrifying flocculating filtration. Other researchers dedicated the removal of several PAH's, antibiotics, hormones, drugs and fragrances to biodegradation or sorption to sludge (Brubaker and Stroo, 1992; Ternes and Joss, 2006; Göbel *et al.*, 2006).

Considering the short detention time within the filter bed (<20 min) the main removal mechanisms may be the filtration due to the adsorption of those substances to remaining sludge particles.

7.7. Evaluation

The additional results presented in this chapter confirm the suitability of chosen filter configuration and operation in regard to the removal of parameters like phosphorus, turbidity, nitrogen and oxygen. Profile measurements during denitrifying flocculating filtration indicated the removal of these parameters within the whole filter bed resulting in high linear correlation coefficients. No determination of the removal by the filter material is found.

The performed fractionations of total phosphate defined particulate phosphate between 0.45 µm and 2 µm as most difficult for filtration. Minimising this fraction may be achieved by the optimisation of the initial processes like coagulation and precipitation. Especially the choice of coagulant clearly influences the effectiveness of the reaction between orthophosphate and metal, the strength and size of the created metal-orthophosphate flocs positively. High removal (70-90%) of particulates in the size of 2 µm to >20 µm indicates the targeted particle size in order to gain proper phosphorus removal.

A strong linear relationship is found for turbidity versus total phosphate (0.1-0.9 mg/L) for the different fractions obtained for single sample fractionations. The filterability of other colloids and humic substances during one sample fractionation might influence the relationship of the two parameters and determine the slope of the trend line for linear regression. However, the establishment of these fractionations indicate the filterability of a phosphate distribution and even the removal of particulate phosphate can be related to turbidity measurements. Even if the utilisation is not yet fully established this fractionations may offer chances for improvement of filter monitoring when phosphorus removal is targeted.

Following single medium lab scale filter applied after pilot scale dual media filtration aimed the simulation of an integrated third layer within the dual media filter and the investigation of the possible improvement of the filtrate quality. Besides almost equal further removal of nitrogen, turbidity and phosphorus, the activated carbon filter removed significantly heavy metals. The removal of heavy metals may be explained by indirect adsorption through complexation with organic matter while.

The integration of an activated carbon layer within the denitrifying flocculating filter may improve the applied filtration step aiming a filtrate quality beyond low nutrient and suspended solids concentrations.

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8

Evaluation of all filtration experiments

8.1 General observations

This study investigates the ability of denitrifying flocculating filtration with a discontinuous dual media filter to perform advanced removal of suspended matter and nutrients simultaneously from wastewater treatment plant (WWTP) effluent. The involvement of chemical and biological processes within filtration impedes evaluation, or even prediction, with traditional filtration theory as described for discrete particles. On-site filtration tests are therefore still very informative, useful and even necessary.

With current filtration practices in the Netherlands granular medium filtration is successfully applied for suspended solids removal, and may even be combined with phosphorus removal via flocculating filtration. A prerequisite for the simultaneous application of biological denitrification was the exclusion of negative impacts on the removal of suspended solids and phosphorus. Therefore, filtration performances with and without denitrification were compared. The backwash procedure and frequency was a major operational issue and had to be adapted in order to achieve comparable filter run times with flocculating filtration. The integration of a “bumping cleaning” within the backwash procedure led to increased filter run times. A filtration rate of 10 m/h, which is commonly used design filtration rate, was chosen as a practical and successful operation parameter during the filtration tests.

Investigations into optimal dosing ratios and online controlled dosing are very important during filter operation when chemicals are applied. Regular confirmation of concentrations measured with online equipment was of great value.

Pressure readings, analyses of the filtrate quality with conventional parameters and particle countings were used to evaluate and ensure similar filtration performances with or without denitrification. The value of additional monitoring with particle analyses was exhibited when evaluating the coagulation, flocculation and filtration processes resulting in further optimisation and understanding the particle removal mechanisms.

A clear specification of the measured substances, especially the phosphorus species, and a corresponding pre-treatment of the samples enhanced interpretation of the final results.

8.2 Filter configuration

Dual media filtration

A filter bed with a total bed height of 1.2 m, consisting of a lower layer of 40 cm quartz sand (diameter: 1.5-2.25 mm, density: 2,600 kg/m^3) and an upper layer of 80 cm anthracite (diameter: 2.0-4.0 mm, density: 1,400 kg/m^3) was selected for the described filtration tests. This filter bed configuration is only one of many possible configurations of various filter materials and layer heights. However, this configuration was identified in optimisation research, conducted by Van Nieuwenhuijzen (1996), as very suitable to achieve long filter run times without major clogging problems. The combination of a coarse layer on top and fine layer on the bottom leads to utilisation of the accumulating capacity of the whole filter bed. Large particles are captured in the upper part while smaller particles can penetrate into the sand layer and are filtered deeper in the filter bed. Literature commonly uses the term ‘depth filtration’ to describe this mechanism (Metcalf and Eddy, 2003). As experience of filtration for phosphorus removal grows, the trend is towards finer filtration materials. STOWA (2006) suggests grain sizes of 0.85-2.5 mm for anthracite and 0.6-1.25 mm for sand when applying discontinuous filtration for P-removal. This is mainly attributed to the fact that phosphate precipitates often appear to be very small. Investigations within this thesis identified that the bulk of phosphate fractions breaking through the filter bed are in the size range of 0.45 – 2 µm.

On the other hand the chosen filter bed configuration with relatively coarse filtration material is very suitable for the application of denitrification. Biological activity results in a biomass film covering the filter grains and consequently narrowing the pore volume. A consequence of using finer filter material may also be the formation of ‘mud-balls’ and persistent clogging of the filter bed over time. Occasionally this also occurred with the chosen filter bed. However optimisation of the backwash regime solved this problem.

Evidently, the narrowed pore volume due to biofilm development even had a positive effect on the filtration results of suspended solids.

Profile measurements for phosphorus, turbidity, oxygen and nitrate confirmed efficient usage of the whole filter bed. Linear removal of all parameters was found for lab and pilot scale filtration where the removal efficiency was not influenced by the filtration material (anthracite or quartz sand).

Application of a third layer

In order to evaluate the improvement to the filtrate by the application of an additional third layer, two single media lab scale columns were applied after dual media filtration. One column was filled with fine garnet sand (0.5 – 0.8 mm grain diameter, 30 cm height) while the other one consisted of a granular activated carbon layer (1.6 mm grain diameter, 1.2 m height). Both were fed with filtrate from the dual media filter which was collected in the filtrate buffer. Even if filtration results after an external third layer are not totally representative to the results of an integrated third layer, possible further quality improvements were observed. The removal of fine particles was especially improved. Suspended matter, expressed with turbidity, particulate organic as well as total phosphate, was further removed.

Furthermore, both columns removed particulate heavy metals breaking through the dual media filter. The removal of copper, zinc and nickel was higher with activated carbon filtration than with fine sand filtration.

However, it should be noted that the integration of a third layer may result in economical concerns for integration within the dual media filtration step. A fine garnet sand layer leads to higher backwash velocities required for filter bed expansion and removal of foulants. The time for saturation and regeneration has to be considered for the integration of granular activated.

8.3 Filter operation

Floc versus flocculating filtration

Filtration aiming at advanced suspended solids and phosphorus removal can generally be applied in the configuration of *floc filtration* or *flocculating filtration.* Floc filtration stands for the filtration of flocs after chemical enhancement for coagulation and flocculation in a separate chamber or tank. In-line coagulation followed by forming of flocs and separation within the filter bed is described by the term flocculating filtration (Van Nieuwenhuijzen, 1996). The second configuration is often applied with discontinuous filtration in combination with a water head above the filter bed. This water volume could be interpreted as flocculator and flocs can grow depending on the retention time before entering the filter bed. The term flocculating filtration in this thesis therefore describes the combination of floc and flocculating filtration.

Van Nieuwenhuijzen (1996) stated that this application leads to better filtrate quality due to efficient usage of the total accumulating capacity and possible additional filtration effects of accumulated particles (‘ripening’). During this research it could be confirmed that this application led to low phosphorus and suspended solids concentrations in the final effluent.

However, particle analyses and phosphate fractionation (dissolved and particulate) showed almost no difference in distributions before and after flocculation. A shift to larger particle sizes, following the flocculation process, within the piping and water volume above the filter bed was not identified but also did not appear to be essential for phosphorus removal. This suggests that, the initial precipitation and coagulation processes are more important for optimal particle and phosphorus removal than the following flocculation process.

Filtration rate

In the first filtration tests for simultaneous nutrient removal at the wastewater treatment plant (WWTP) Utrecht the discontinuous filter was operated at a low filtration rate of 6 m/h. Subsequent filtration tests at WWTP Beverwijk and Horstermeer indicated similar good performance of suspended solids, phosphorus and nitrogen removal with a filtration rate of 10 m/h. This filtration rate is a more commonly applied rate in actual filtration applications. Similar good removal performances were established in a later project stage even with a filtration rate of 12.5 m/h and may be even higher filtration rates are possible with similar removal performances. Generally, STOWA (2006) suggests filtration rates of 5-30 m/h for discontinuous filtration but only for the removal of suspended solids.

It is noted that the retention time of the chemically conditioned filter feed water in the water volume above the filter bed is determined by the fixed water head together with the filtration rate. This retention time is comparable with a flocculation time and was assumed to influence the floc strength and size prior to filtration. During the filtration tests the filtration rate of 10 m/h and a water head of almost 2 m above the filter bed resulted in a flocculation time of 11 minutes.

Backwash

When the hydraulic resistance across the filter reaches a maximum set-point or if the filtrate quality deteriorates it is necessary to backwash the filter. The backwash aims at, (1) the removal of impurities adhering to the filter grain surfaces by the shearing action, and (2) the expansion of the filter bed increasing the pore space and allowing the accumulated foulants to be more easily removed with the wash water. In case of denitrifying flocculating filtration the backwash is optimal when the foulants are removed efficiently without removing the complete biomass by excessively strong shearing forces. Another problem causing clogging of the filter bed is the development of entrapped gas during biological activity. Regular release of the gas can ensure efficient filtration run times.

The optimal backwash investigated on both lab and pilot scale during denitrifying flocculating filtration resulted in 4 backwash phases. One combined air-water phase is followed by three water backwash phases with decreasing velocities aiming for filter bed stratification. The total backwash lasts 11 minutes every 24 hours. Every three hours a ‘bumping’ cleaning of 2 minutes with a backwash water rate of 50 m/h releases the developed gas produced by denitrification (CO_2 and N_2). Metcalf and Eddy (2003) suggest a hydraulic surge for 3-5 minutes every 2 to 4 hours with a flow rate of 12 m/h. This frequency and duration is comparable with the application within the presented investigations.

During the investigations of an optimal backwash procedure and frequency mud-ball formation in the filter material was frequently observed. This caused incidental rinsing of filter material, especially anthracite, due to greater bed expansion. In the backwash theory the filter bed expansion is influenced by the backwash rate and water temperature. Clements (2004), Clements and Haarhoff (2004) and Van Meer (2006) indicated increased bed expansion by biomass growth of even more than 10 % for sand and activated carbon filters. This can lead to a decrease of the backwash water rate.

However, after optimisation of the backwash regime (normal plus ‘bumping’ cleaning) at WWTP Horstermeer major ‘mud-ball’ formation was avoided indicating no need for decreasing the backwash rate.

Usually discontinuous filters are designed for a backwash water usage of 3-8% of the hydraulic load (STOWA, 2006). The application of a ‘bumping’ cleaning reduced the water usage below 6%. This is positive when comparing discontinuous filtration with continuous filtration where the design value for backwash water usage is approximately 5-10% of the hydraulic load (STOWA, 2006).

Although high turbulence is created during backwash causing shear forces in the filter bed and scouring of the grains, biofilm loss is negligible. Obviously an adhered and strongly attached biofilm is created and supported by the coarse surface of the anthracite and sand grains. The denitrification process was found to recover within 30 minutes after backwash.

Pressure readings along the filter bed indicate the recovery of the clean water pressure profile after each backwash and comparability with the pressure profile for single flocculating filtration. The investigated backwash seems to be sufficient to recover the filter bed quality and ensure stable process performance.

Chemical dosing

Ferric chloride (or iron(III)chloride) or poly aluminium chloride are used as coagulants. Initial dosing concentrations for filtration tests were obtained by preliminary jar tests. The optimal metal concentration results from (1) a high removal of particulate phosphate through sedimentation and filtration over 0.45 µm membrane filter, and (2) low concentrations of remaining orthophosphate and dissolved metal. It is noted that filtration mechanisms can not be simulated by sedimentation in a jar and sample filtration. The resulting optimal dosing concentrations from jar tests were lower, about 3 mol metal per mol orthophosphate (MePO ratio), than the optimal concentrations investigated in the subsequent filtration tests, for example MePO = 4.8 mol/mol at WWTP Horstermeer.

The dosed quantity of metal salt is expressed with the metal-bound orthophosphate MePO ratio because orthophosphate is the phosphorus fraction targeted by precipitation.

During filtration tests the dosing of coagulant is adjusted proportional to the continuously measured orthophosphate (PO_4-$P_{<0.45\mu m}$) concentration in the filter feed water (WWTP effluent) in order to avoid over- or under-dosing.

Generally, the coagulant dosing ratio for filtration of WWTP effluent is high compared to, for example, ratios applied for chemical phosphorus precipitation during biological treatment but is in accordance with STOWA (2006). This high dose rate is a necessity to ensure sufficient presence of metal ions able to precipitate with a low concentration of orthophosphate and may be even to compensate the low retention times in the filter unit for flocculation.

Methanol (CH_3OH) was chosen as carbon source for denitrification because of its relatively low costs and small sludge production compared to other carbon compounds (Koch and Siegrist, 1997) and was proved to work satisfactorily. Although methanol is easily biodegradable, it is not available for all microorganisms because it is classified as a C1 compound (Nurse, 1980; Timmermans and Van Haute, 1983; Zumft, 1997; Dos Santos, 2004). Disadvantageous for the application of methanol are the high safety precautions required. The use of an alternative carbon sources, like acetate, bio-ethanol, etc., with similar biodegradability to methanol might be reasonable.

8.4 Monitoring

Conventional analyses

The filtration process was monitored by online and manual analyses. For instance orthophosphate, total phosphorus, nitrate, temperature and turbidity were analysed online in the WWTP effluent and filtrate. The online analyses can indicate the long term process performance and are therefore very useful for later full scale operation.

The manual analyses are used to directly obtain the concentrations of parameters which are not measured online and to determine the effect of process adjustment. Manual analyses are also conducted to control the reliability of the online analysers because online analysers can be still rather sensitive to malfunctions and require regular maintenance.

Online turbidity meters measured turbidity in the filter feed water and the filtrate. These values can be used as an indication of suspended solids removal during filtration. Analyses of grab samples with a mobile turbidity meter were conducted to confirm the online measurements. The applied online devices were not self-cleaning and fouled over time, with rapid deterioration after 24 hours of operation. Although regular cleaning took place the interpretation of the total values seems to be doubtful.

The investigated relationship of particulate phosphate and turbidity in chapter 7 may be used for online monitoring when flocculating filtration is applied.

Nevertheless, a trend for the daily turbidity pattern of the WWTP effluent and stable low filtrate turbidity values (below 1.5 NTU) are obtained online.

Particle analysis

Particle analysis was investigated as an additional monitoring tool to conventional analyses. Particles in the size range 2-100 µm were counted in grab samples. The device used, MetOne PCX100, had a concentration limit of 10,000 particles per mL sample. For some samples the particle number exceeded this concentration limit, especially the filter feed water, the WWTP effluent after chemical enhancement for phosphorus precipitation and completed flocculation (expressed with the abbreviation AF). Sample dilution and recalculation of the counts to an undiluted sample was applied. A dilution rate of 3 was chosen for the investigations presented in this thesis.

Different parameters can be used for evaluation. The average diameter, the distribution of particle size or particle volume is appropriate for the identification of a site specific fingerprint of a WWTP effluent. Cumulative counts or volumes could be used to describe total removal efficiencies.

Investigation into the appropriate use of particle analyses showed that the particle volume is preferable to the particle number. This conclusion is based on the result of relating the total particle number versus particle volume. It showed that one certain particle volume may be related to different particle numbers.

Particle distributions at different stages of the process (e.g. before and after coagulation, flocculation, filtration) can be evaluated and may even influence processes when necessary. For example during the filtration tests at all three WWTP's no significant growth of particle size or flocs was measured after the completed flocculation time. This could be a consequence of some operational parameters, for example mixing energy too low during the coagulation phase or flocculation time too short prior to filtration. Creation of a different particle distribution by optimisation of one of those parameters may lead to a better filtration result of, for example, particulate phosphate.

The cumulative particle volume removal, calculated by the filter feed water and the filtrate, was used for the interpretation of the overall filtration performance. This filtration performance was related to the particulate phosphate removal. Unfortunately phosphate fractions could not be correlated to particle fractions in the range 2-100 µm. The results of phosphate fractionations showed the removal of particulate phosphate above 2 µm. Phosphate particulates still present in the filtrate had a size of 0.45 - 2 µm and are not measured with the employed particle counting device. Nevertheless, this particle size range is too small to be removed with granular medium filtration.

The relationship of particulate phosphate and turbidity is clear; these two parameters are directly related. This is obtained in the analyses using fractionation of the samples over 0.45 µm, 2 µm, 5 µm, 10 µm and 20 µm polycarbonate membrane filters.

However, the relationship with other conventional parameters is difficult may be due to the different analytical principles they use. The particle counter is based on the light blocking method, turbidity applies light scattering measurements and wavelengths indicate phosphorus concentration through colouring chemical reactions. Even if relationships with other conventional parameters are difficult and maybe even confusing, this monitoring tool has definitely proven its additional value in understanding the filtration process.

8.5 Removal of suspended solids, nitrogen and phosphorus

The only limiting requirement on the additional application of denitrification was that the optimal removal of suspended solids and phosphorus was to remain undisturbed. The phosphorus removal efficiencies during the operation as single flocculation filtration and simultaneous denitrifying flocculating filtration were comparable. Also the average concentration of orthophosphate reached in the filtrate was comparable for both operations for three WWTP's applying filtration on lab and pilot scale. Relatively high metal-orthophosphate ratios, MePO of 4-5 mol/mol, were found to be most effective for the phosphate precipitation. High dosing ratios are also suggested by several researchers (Hultman *et al.*, 1994; Van der Graaf and Van Nieuwenhuijzen, 1998; De Haas, *et al.*, 2001, Metcalf and Eddy, 2003) and are required for two reasons;

1. to increase the natural low suspended solids content in WWTP effluent necessary for floc formation, and
2. to ensure the precipitation of phosphate present in concentrations below 1 mg/L.

Efficient chemical phosphate precipitation and the effective filtration of particulate inorganic and organic phosphorus is essential to achieve a low total phosphorus concentration of $0.15 mgP_{tot}/L$ in the filtrate. From fractionation tests the small particulate phosphate fraction between 0.45 and 2.0 μm appears most difficult for filtration. A comparison of different coagulants (poly aluminium chloride) seems to influence the amount of phosphate present in this size range.

However, the fraction of dissolved organic phosphorus can not be coagulated and filtered or adsorbed. Measurements at all three WWTP's indicated a concentration of max. 0.1 mg/L in the filtrate. This confirmed the outcomes of several other researchers, such as. Van Lierop (2004). At this concentration level dissolved organic phosphorus represents two thirds of the total phosphorus concentration required for MTR-quality of 0.15 mgP_{tot}/L, and is a significant contribution of the total phosphorus concentration.

Orthophosphate concentrations below 0.1 mg/L, obtained during the tests at WWTP Utrecht and reported as minimum concentration for denitrification by Hultman *et al.* (1994), had no inhibiting effect on the biological activity. It is most likely that the phosphate ions partially bound in metal-bound orthophosphate precipitates are still available for biological uptake.

Biological phosphorus removal due to the assimilation of denitrifying biomass is measured with a maximum of 0.1 mg/L. A similar value is reported by Eichinger (1994).

The start-up of denitrification lasted one week and involved an increased methanol dose during reduced filtrate rate. Wilderer *et al.* (1994) and Koch and Siegrist (1997) reported an adaptation period for biomass for filtration of a few days to weeks which is comparable with the findings presented in this thesis. High denitrification rates above 3 $kgNO_3\text{-}N/d/m^3_{bedvolume}$ were observed for water temperatures of 13-20 °C with a dosed amount of methanol 4.7 $kgCH_3OH/kgNO_3\text{-}N$. Nitrate was removed to concentrations below the detection limit of the used manual cuvette analyses of 0.5 $mgNO_3\text{-}N/L$. However, a very low total nitrogen concentrations (2.2 gN_{tot}/L) in the final effluent may still be difficult to achieve in the presence of high organic or ammonium concentrations. This occasionally occurred during the research at WWTP Beverwijk and Horstermeer where high kjeldahl-nitrogen concentrations were intermittently observed. These high concentrations of organic and ammonium nitrogen were probably caused by a less optimal nitrification due to temporary overloading or low temperatures at the WWTP.

And again, optimal wastewater treatment prior to the tertiary treatment with denitrifying flocculating filtration is essential.

8.6 Removal of other substances

Heavy metals

The three heavy metals, namely copper, zinc and nickel, were chosen due to their significance for Dutch wastewater treatment plant effluent in general and their importance in national and future international requirements.

Filtration tests at WWTP Horstermeer indicated significant removal of copper and zinc during denitrifying flocculating filtration. Copper seems to form more easily than zinc complexed particulates with filterable material. The variation of the dosing of poly aluminium chloride influenced the distribution between particulate and dissolved form for both metals but did enhance the overall removal. Nickel was hardly removed because it exists predominantly in dissolved form (below 0.45 μm). The removal for all three metals decreased slightly during inactivation of the denitrifying biomass (Miska *et al.*, 2006).

High removal of heavy metals with (powdered) activated carbon was indicated by preliminary jar tests. Possible explanations could be indirect or direct removal.

Indirect adsorption of heavy metals to absorbable organics could be a very likely removal mechanism. Some microorganisms and biomass are able to accumulate heavy metals. This mechanism is called bio-sorption and defined as sorption and/or complex forming based on the chemical properties of the biomass (Volesky, 1994). Dissolved organic matter and sludge flocs are defined as weak acids (Wang *et al.*, 1998, Wang, 2000) able to form complexes with heavy metals ions. Adsorption of these organic complexes subsequently results in removal of heavy metals. Generally, sorption can be less at neutral pH values and increases with increasing alkalinity.

Overall, the three chosen heavy metals were removed with denitrifying flocculating filtration, resulting in efficiencies of above 92%, 15% and 66% for copper, nickel and zinc respectively. Even higher removal may be achieved in combination with activated carbon adsorption.

Other (possibly) relevant substances

During the filtration experiments at WWTP Utrecht and WWTP Horstermeer slight removal of bisphenol A, some nonylphenol compounds and polycyclic aromatic hydrocarbons was observed. Incidental measurements of several polycyclic aromatic hydrocarbons (PAH's) during pilot scale filtration at WWTP Horstermeer indicated removal of some PAH's. Many other substances targeted with the Water Framework Directive were not removed.

However, the limited available analyses do not allow general conclusions about possible removal during denitrifying flocculating filtration but indicate the necessity for more research.

8.7 Performance compared to continuous filtration

General

The performance of discontinuous filtration may be compared with continuous filtration due to their competitive application in practice. Three removal performances are evaluated and compared for the application of a filtration system for combined nutrient removal; nitrogen, suspended solids and phosphorus removal. For continuous filters the following advantages, compared with discontinuous operation, can be described:

1. no extreme disturbances of the filter bed due to backwash in the end of the filtration run, and
2. no occurrence of filter bed clogging due to 'mud-balls' forming.

Nitrogen

The removal of nitrogen can be evaluated by concentrations and removal efficiencies but also by the nitrate conversion rate per cubic meter bed volume. Typical design conversion rates for nitrate vary between 0.5 – 2.0 kg $kgNO_3\text{-}N/d/m^3_{bedvolume}$ for discontinuous and continuous filters (Metcalf and Eddy, 2003). STOWA (2006) reported a maximum nitrate conversion of 1.5-2.5 $kgNO_3\text{-}N/d/m^3_{bedvolume}$. Accordingly, the continuous filtration step at WWTP Uithoorn is designed for a conversion rate of 2.0 $kgNO_3\text{-}N/d/m^3_{bedvolume}$ (Piekema and Daamen, 2003).

The nitrogen removal for discontinuous filtration investigated at WWTP Horstermeer resulted in nitrate conversion rates of maximal 4 $kgNO_3\text{-}N/d/m^3_{bedvolume}$ at 20°C water temperature and a filtration rate of 10 m/h, and 3 $kgNO_3\text{-}N/d/m^3_{bedvolume}$ at 13°C water temperature and a filtration rate of maximal 12.5 m/h.

Phosphorus and particle removal

The evaluation of particle removal in both filtration systems shows a higher removal of particles between 2 and 5 µm with discontinuous filtration. The dosing of coagulant during continuous filtration, in the presented case ferric chloride was dosed, improves the removal of the particle volume in that size range. Above 5 µm the volume removal efficiencies differ only with maximal 10% and are therefore relatively comparable in both systems. The removal of particles in all size ranges and especially the small particles 2 - 5 µm is important when aiming at high phosphorus removal.

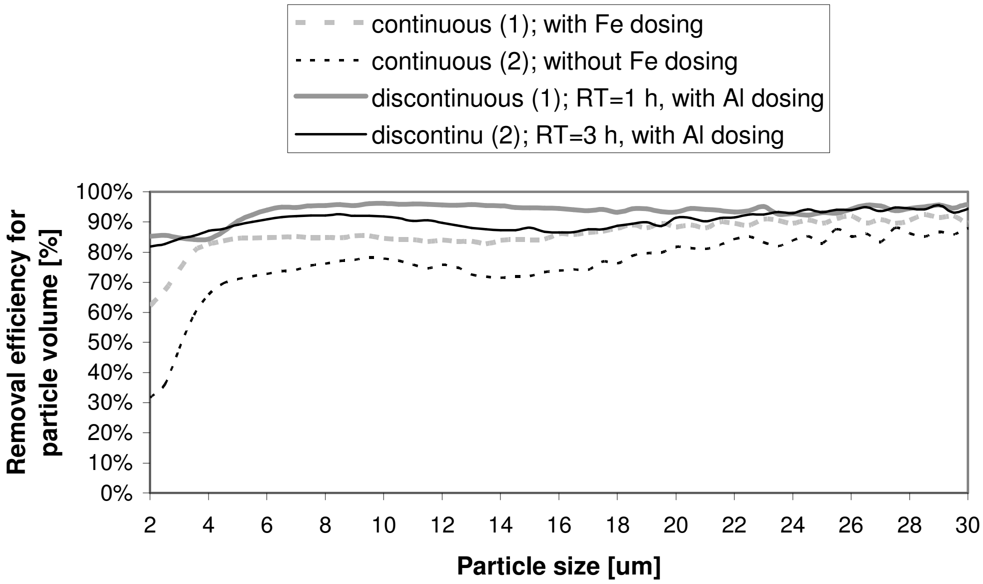

Figure 8. 1: Comparison of particle volume removal 2-30 µm for continuous and discontinuous filtration obtained at WWTP Horstermeer; RT: Run time, Fe: ferric chloride, al: poly aluminium chloride

The results for phosphorus removal seem comparable for both filtration systems. An average concentration of 0.2 mgP_{tot}/L was found for all three research locations, WWTP Utrecht, Beverwijk and Horstermeer, applying denitrifying flocculating filtration in a discontinuous mode. Similar results were obtained during pilot investigations with continuous filtration applied for simultaneous nutrient removal at WWTP Maasbommel (Van Lierop, 2004, Kiestra *et al.*, 2004) and WWTP Horstermeer (Paques, 2006).

Therefore, the advantage of discontinuous filtration to remove small particles 2-5 µm more efficiently does not significantly improve the removal of phosphate. The results for phosphorus fractionation show that most of the particulate phosphate breaking through the filter measures a size of 0.45 - 2 µm, and is therefore much smaller than what is counted with the actual used particle analyser (MetOne PCX100).

Particles between 0.45 - 2 µm should be measured in order to confirm and even complete the presented results for particle removal related to particulate phosphate removal.

8.8 Further developments

The research on combined nutrient removal in one discontinuous filtration unit showed promising results for application at modern Dutch wastewater treatment plants. This is one positive development for advanced effluent polishing reaching an effluent quality which could comply with the more stringent effluent criteria aimed by the European Water Framework Directive (European Commission, 2000). Lab scale tests at WWTP Beverwijk and pilot investigations at WWTP Utrecht and Horstermeer provide sufficient experimental reference to prove the process in full scale for the research goal. Hence, investigations are continuing and extending after the completion of the presented PhD research period in April 2007. The investigations include further long term filtration tests at WWTP Horstermeer and full scale investigations at WWTP Leiden South-West (STOWA, 2008).

The combination of activated carbon and filtration especially seems to deliver a promising option for further process optimisation. Also the optimisation of coagulation and flocculation may increase the reliability of reaching very low phosphorus concentrations in the filtrate.

8.9 Conclusions

The most important results of the presented investigations on the three wastewater treatment plants WWTP Utrecht, WWTP Beverwijk and WWTP Horstermeer are summarised below:

1. Dual media filtration in a discontinuous mode operated with denitrifying flocculating filtration is applicable for modern Dutch WWTP effluent reaching very low suspended solids and nutrient concentrations. Even MTR-quality of 2.2 mgN_{tot}/L for total nitrogen and 0.15 mgP_{tot}/L for total phosphorus can be achieved.
2. A stable and robust filtration process is obtainable aiming at the combined removal of suspended solids, phosphorus, nitrogen and other substances. A prerequisite for reaching the very stringent Water Framework Directive requirements is a stable quality of the WWTP effluent; ammonia level should be very low (<1mg/L), the orthophosphate concentration should not exceed 0.6-0.7

mg/L and the nitrate concentration should not overload the denitrification rate of 3-4 kgNO_3-N/d/$m^3_{bedvolume}$.

3. Particle analyses and total phosphate fractionation indicated initial precipitation and coagulation to be the most important processes responsible for optimal removal of suspended matter and phosphorus. The flocculation process prior to filtration plays only a minor role.

   Firstly, the optimal reaction of orthophosphate with metal ions to metal-bound orthophosphate flocs is essential. Secondly the formation of a filterable floc size >2 µm is necessary in order to achieve a high removal of particulate phosphate. The strength of formed flocs should be ensured as filtration involves high shearing forces within the pore space. Investigation of the optimal type of coagulant is important to improve floc strength.

4. Particle analysis is a valuable tool also in relation to other conventional parameters and improves the understanding of particle related filtration mechanisms. The comparison of particle distributions between 2-100 µm before and after filtration may lead to optimisation of the precipitation, coagulation and filtration processes aiming at a high removal of phosphate precipitates and other suspended matter.

5. Integration of the adsorption process can improve the removal of other organic micro pollutants like heavy metals, polycyclic aromatic hydrocarbons, pesticides and endocrine disrupting compounds simultaneous to removal of nutrients and suspended solids. This may be achieved by additional dosing of powdered activated carbon or integration of a granular activated carbon layer in the denitrifying flocculating filter.

6. The application of the existing and well known conventional filtration in discontinuous operational mode combining different removal processes can lead to a reliable treatment step, delivering an improved effluent quality offering further possibilities for optimisation.

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A

Substances Water Framework Directive

A.1 List of (possible) relevant substances

The following Table A. 1 shows the relevant and possibly relevant substances indentified due to their importance in Dutch wastewater treatment plant effluent including compounds listed in the European Water Framework Directive and Dutch directive 'Vierde Nota Waterhuishouding'. This list includes nutrients, microorganisms, organic micro pollutants, pesticides, heavy metals (plus metalloids), hormone disrupting and medicinal substances.

Table A. 1: (Possible) relevant substances defined for Dutch wastewater treatment plant effluent, (STOWA, 2005)

Group	Substance
Nutrients	Total-phosphorus *
	Total-nitrogen *
Microorganisms	Intestinal enterococci *
	Escherichia coli *
	Viruses *

Group	Substance
Organic micro pollutants	4-chloroaniline **
	Octyl phenol *
	Nonyl phenol *
	Di(2-ethylhexyl)phthalate (DEHP) *
	Nezo(a)pyrene *
	Fluoranthene *
	Benzo(b)fluoranthene *
	Benzo(k)fluoranthene *
	Benzo(g, h, i)pyrene **
	Ideno(1, 2, 3-cd)pyrene *
	Anthracene *
	Naphthalene *
	Trichloromethane **
	C10-13 chloroalkane **
	Bromated diphenyl ethers
Pesticides	Hexachlorocyclohexane/HCH/Lindane *
	Pentachlorophenol (PCP) **
	Simazine **
	Atrazine *
	MCPA **
	Mecoprop (MCPP) **
	Diuron *
	Bentazon **
	Pyrazon/chloridazon **
Group	Substance
Heavy metals & metalloids	Arsenic **
	Cadmium *
	Chromium **
	Lead *
	Mercury **
	Nickel *
	Copper *
	Zinc *
Hormone disrupting & medicinal substances	Not mentioned in the WFD

* relevant substance; ** possible relevant substance

B

Operation of test installations

B.1 Operation of pilot scale filtration at WWTP Utrecht

The operation of the dual media filter during pilot scale investigations at the WWTP Utrecht is described in detail in Table B. 1. The automatic backwash was initiated after reaching filter run time set point, the maximum water head above the filter bed. The water level above the filter material was kept constant at 1.96 m during normal filtration.

Table B. 1: Operation of pilot-scale dual media filter at WWTP Utrecht between 02-03-'04 – 08-07-'04

Period	Flow rate [m³/h] / Filtration rate [m/h]	Run time [h]	Back wash procedure, BW	Methanol dosing* [gCH$_3$OH/g NO$_3$-N]	FeCl$_3$/PACl [mg Me^{3+}/L]
02-03 / 08-03	8 / 10	24	BW 1	-	-
09-03 / 19-03	8 / 10	16	BW 1	-	0.5/0
20-03 / 24-03	6 / 7.5	16	BW 1	-	1.2/0
25-03 / 29-03	6 / 7.5	16	BW 1	-	0/1.0
30-03 / 08-04	6 / 7.5	16	BW 1	3.0	0/1.0
09-04 / 14-04	5 / 6.3	16	BW 1	4.8	-
15-04 / 20-04	5 / 6.3	8	BW 1	4.2	-
21-04 / 22-04	5 / 6.3	8	BW 2	4.2	0/1.0
23-04 / 04-05	5 / 6.3	8	BW 3	4.2	-

Period	Flow rate [m³/h] / Filtration rate [m/h]	Run time [h]	Back wash procedure, BW	Methanol dosing* [gCH_3OH/g NO_3-N]	$FeCl_3$/PACl [mg Me^{3+}/L]
05-05 / 18-05	5 / 6.3	8	BW 3	4.2	1.0/0
19-05 / 31-05	5 / 6.3	8	BW 3	4.2	0/2.0-2.5
01-06 / 04-06	6 / 7.5	8	BW 1	-	0/2.0
05-06 / 07-06	8 / 10	8	BW 1	-	0/2.0
08-06 / 14-06	10 / 12.5	8	BW 1	-	0/2.0
15-06	-	8	BW 1	-	0/2.0
16-06 / 17-06	8 / 10	8	BW 1	-	0/2.0
18-06 / 21-06	10 / 12.5	4	BW 1	-	0/2.0
22-06 / 25-06	13.6 / 17	4	BW 1	-	0/2.0
26-06 / 30-06	8 / 10	4	BW 1	-	0/2.0
01-07 / 05-07	8 / 10	4	BW 1	-	-
06-07 / 08-07	13.6 / 17	4	BW 1	-	0/2.0

*3 g CH_3OH is necessary for the conversion of 1 g NO_3-N. The additional amount of 1.5 en 2 g CH_3OH is used for the removal of oxygen.

B.2 Operation of lab scale filtration at WWTP Beverwijk

Table B. 2 shows the operational modes for the lab scale columns applied for filtration tests at WWTP Beverwijk. The installation was operated manually and with a free raising water table.

Table B. 2: Operation of lab-scale dual media filter at WWTP Beverwijk between 22-06-'04 – 22-11-'04

Period	Flow rate [l/h] / Filtration rate [m/h]	Run time [h]	Backwash procedure, BW	Methanol dosing* [gCH_3OH/ gNO_3-N]	$FeCl_3$/PAC [mg Me^{3+}/L]
22-06 / 26-06	262 / 20	7-24	BW 1	-	-
29-06 / 12-07	131 / 10	7-24	BW 1	-	-
29-07 / 10-08	131 / 10	7-24	BW 1	-	3-4 / 0
23-08 / 26-08	262 / 20	7-24	BW 1	-	3-4 / 0
09-09 / 13-09	131 / 10	7-24	BW 1	-	0 / 2-3
14-09 / 20-09	262 / 20	7-24	BW 1	-	0 / 2-3
22-09 / 27-09	131 / 10	7-24	BW 1	-	3-4 / 0

Period	Flow rate [l/h] / Filtration rate [m/h]	Run time [h]	Backwash procedure, BW	Methanol dosing* [gCH_3OH/ gNO_3-N]	$FeCl_3$/PAC [mg Me^{3+}/L]
28-09 –06-10	/ 6.5	7-24	BW 2	-	-
07-10 / 12-10	/ 6.5	7-24	BW 2	4.8	-
13-10 / 01-11	131 / 10	7-24	BW 2	3.8	2-4 / 0
02-10 / 09-11	131 / 10	7-24	BW 2	3.8	0 / 2-3.5
10-11 / 11-11	131 / 10	7-24	BW 2	3.8	-
12-11 / 18-11	131 / 10	7-24	BW 1	-	3-4 / 0
22-11	131 / 10	7-24	BW 1	-	-

*3 g CH_3OH is necessary for the conversion of 1 g NO_3-N. The additional amount of 1.5 en 2 g CH_3OH is used for the removal of oxygen.

B.3 Operation of pilot scale filtration at WWTP Horstermeer

The same dual media filter installation as used before in the investigations at WWTP Utrecht was applied during pilot scale investigations at the WWTP Horstermeer. The operational parameters, using a constant water head of 1.96 m, are described in detail in Table B. 3. The backwash was initiated automatically after reaching the filter run time set point, the maximum water head above the filter bed.

Table B. 3: Operation of dual media filter at WWTP Horstermeer between 01-03-’05 – 28-10-’05

Period	Flow rate [m³/h] / Filtration rate [m/h]	Run time [h]	Back wash procedure BW	Methanol dosing* [$kgCH_3OH$/ $kgNO_3$-N]	PACl in metal-ortho-phosphate ratio MePO
01-03 / 10-03	8 / 10	18	BW 1.1	-	-
11-03 / 24-03	5 / 6.3	18	BW 1.1	-	-
24-03 / 01-04	5 / 6.3	12	BW 1.1	-	-
01-04 / 13-04	5 / 6.3	12	BW 1.1	-	-
13-04 / 20-04	8 / 10	10	BW 1.1	-	-
20-04 / 22-04	8 / 10	8	BW 1.1	-	2.2 – 2.5
22-04 / 25-04	8 / 10	6	BW 1.1	-	2.5 – 2.8
25-04 / 28-04	8 / 10	4	BW 1.1	-	3.0 – 4.0

Period	Flow rate [m³/h] / Filtration rate [m/h]	Run time [h]	Back wash procedu re BW	Methanol dosing* [kgCH_3O H/ kgNO_3-N]	PACl in metal-ortho-phosphate ratio MePO
28-04 / 13-05	8 / 10	4	BW 1.1	-	3.0 – 4.0
18-05 / 14-06	5 / 6.3	6	BW 1.1	4.5	-
14-06 / 29-06	8 / 10	6	BW 1.1	4.5	10.7
30-06 / 05-07	8 / 10	6	BW 1.1	4.5	9.2
05-07	8 / 10	6	BW 1.1	4.5	0
06-07 / 07-07	8 / 10	6	BW 1.1	4.5	9.2
08-07 / 10-07	8 / 10	6	BW 1.1	4.5	0
11-07 / 13-07	8 / 10	6	BW 1.1	4.5	9.2
14-07	8 / 10	6	BW 1.1	5.0	9.2
15-07 / 17-07	8 / 10	6	BW 1.1	5.0	0
18-07 / 21-07	8 / 10	6	BW 1.1	5.0	9.9
22-07 / 25-07	8 / 10	6	BW 1.1	5.0	11.7
26-07 / 29-07	8 / 10	6	BW 1.1	5.0	5.5
31-07 / 01-08	8 / 10	6	BW 1.1	5.0	4.8
02-08 / 05-08	8 / 10	6	BW 1.1	5.0	0
06-08 / 08/08	8 / 10	6	BW 1.1	5.0	11.6 – 4.8
08-08 / 01-09	8 / 10	6	BW 1.2	5.0	3.5 – 4.8
01-09 / 19-09	8 / 10	6	BW 1.2	4.7	4.8
19-09 / 22-09	8 / 10	5	BW 1.2	4.7	4.8
22-09 / 27-09	8 / 10	4	BW 1.2	4.7	4.8
28-09 / 04-10	8 / 10	4	BW 2	4.5	4.8
04-10 / 20-10	8 / 10	4	BW 2	3.7	4.8
21-10 / 25-10	8 / 10	4	BW 2	4.5	4.8
25-10 / 28-10	8 / 10	4	BW 2	Change carbon source	4.8

*3 g CH_3OH is necessary for the conversion of 1 g NO_3-N. The additional amount of 1.5 en 2kg CH_3OH is used for the removal of oxygen.

During the research period different carbon sources were investigated between 28-10-’05 - 22-12-’05. The operational conditions of these tests are not presented in this thesis. The following filtration tests between 01-03-’05 – 31-12-’06 are presented in Table B. 4.

Table B. 4: Operation of dual media filter (with applied backwash procedure BW2) at WWTP Horstermeer between 01-03-'06 – 31-12-'06

Period	Flow rate [m3/h]	Filtration rate [m/h]	Filter run time /Frequency backwash BW* [h]	Frequency "bumping cleaning" ** [h]	Methanol dosing* [gCH_3OH/gNO_3-N]	PAClin metal-ortho-phosphate ratio MePO
01-03 / 08-03	5	6.3	10		4.5	4.8
08-03 / 14-03	5	6.3	6		4.5	4.8
14-03 / 31-03	5	6.3	12	3	4.5	4.8
31-03 / 03-04	5	6.3	10	2	4.5	4.8
03-04 / 10-04	5	6.3	12	3	4.5	4.8
10-04 / 27-04	8	10	10	2	4.5	4.8
27-04 / 04-05	8	10	10	2	4.5	4.8
04-05 / 09-05	8	10	14	2	4.5	4.8
09-05 / 15-05	8	10	15	3	4.5	4.8
15-05 / 08-06						
08-06 / 18-06	8	10	10	2	4.5	4.8
18-06 / 05-09	8	10	15	3	4.5	4.8
05-09 / 09-11	8	10	24	3	4.5	4.8
04-12 / 22-12	10	12.5	24	3	4.5	4.8
22-12 / 31-12	5	6.3	24	3	4.5	4.8

* BW: normal backwash, ** : " bumping cleaning" consisting of high water flow rate, ***: theoretically 3 kg CH_3OH is necessary for the conversion of 1 kg NO_3-N. The amount of 1.5 en 2 kg CH_3OH additionally was dosed for the removal of oxygen.

B.4 Product specifications

Different chemicals were applied during the different filtration tests. The methanol solution dosed as carbon source consisted of a chemical pure methanol product (97%) diluted with tap water.

The product specification of the applied coagulants such as ferric chloride and the poly aluminium chloride products (PACl, PAC) are presented in this paragraph.

$FeCl_3$ - Iron(III)chloride 40% (Chemproha Chemie Partner)

Chemical formula	$FeCl_3$
CAS number	7705-08-0
EINECS number	231-729-4

Test item	Unit	Specification
Purity	mass-%	40 ± 1.0
Density at 20°C	kg/L	1.415 ± 0.015
Composition:		
Hg	mg/kg	max. 0.1
Pb	mg/kg	max. 1.0
Cr	mg/kg	max. 25.0
Ni	mg/kg	max. 50.0
Cd	mg/kg	max. 1.0
Cu	mg/kg	max. 2.0
Zn	mg/kg	max. 25.0

PACl - Aqualenc (Chemproha Chemie Partner)

Chemical name	polyaluminiumchloride, polyaluminiumhydroxychloridesulphate, PAC
Chemical formula	$Al_2(SO_4)xCly(OH)z$
Molecular weight	very high
CAS number	39290-78-3

Test item	Unit	Specification

Density at 20°C	kg/L	1.165
Viscosity at 20°C	mPas	4.5
pH value at 20°C	-	2.9 ± 0.1
Composition:		
Al_2O_3	gew %	8.3
SO_4	gew %	4.2 ± 0.5
Cl	gew %	4.8 ± 0.5
Ca	gew %	0.2
Fe	mg/kg	60
Mn	mg/kg	10
Zn	mg/kg	10
Heavy metals (as lead)	mg/kg	5

PAC - PAX 11 (Kemira Chemicals B.V.)

Test item	Unit	Specification
Al_2O_3	%	10.2 ± 0.4
Al^{3+}	%	5.4 ± 0.2
Cl^-	%	9.6 ± 0.5
F_{tot}	%	<0.01
SO_4^{2-}	%	2.5 ± 0.5
Alkalinity	%	46 ± 5
Density	g/cm^3	1.21 ± 0.02
pH	-	2.5 ± 0.5
Crystallization temp	°C	-8
Viscosity at 25 °C	mPas	10 ± 5

PAC - PAX 14 (Kemira Chemicals B.V.)

Test item	Unit	Specification
Al_2O_3	%	ca. 13.6

Al	%	ca. 7.2
Cl^-	%	ca. 20
Alkalinity	%	ca. 30
Density	g/cm^3	1.31
pH	-	ca. 1
Crystallization temp	°C	-15
Viscosity at 23 °C	mPa	24
at 0 °C	mPa	50

C

Phosphorus analyses

C.1 Procedures for phosphorus analyses

For the measurements and the proper interpretation of phosphorus measurements the following procedure was followed considering different sample treatment:

1. Orthophosphate (PO_4-P) analyses required filtration over 0.45 µm membrane filters. The material of the utilised membrane filters was cellulose acetate. This pre-filtration was necessary in order to analyse only the dissolved inorganic fraction of phosphorus, the so called orthophosphate. It was determined by directly adding a substance such as ammonium molybdate to the acidified sample which forms a coloured complex with phosphate.

   The analyses of this fraction was identified with the abbreviation "*diss"* used for the actual orthophosphate concentration; $PO_4\text{-P} = P_{diss}$.

2. Orthophosphate analyses of a raw sample, which was not filtered over 0.45 µm, will additional obtain metal-orthophosphate flocs (inorganic particulate fraction) as orthophosphate. Due to the change of the pH while acidification of the sample for the cuvette analyses this particulate inorganic fraction will dissolve and indicate a higher concentration than the actual orthophosphate in the sample. The abbreviation "*diss+part*" was introduced for this phosphorus fraction; $P_{diss+part}$.

3. The difference in the orthophosphate analyses conducted on the raw and filtered sample represents the inorganic particulate phosphate concentration indicated by the abbreviation *"part"*, i.e. $P_{diss+part} - P_{diss} = P_{part.}$
4. Within this thesis particulate phosphorus concentrations were fractionated for different particulate sizes. For this fractionation samples were filtered over poly carbonate membrane filters with the pore size of 20, 10, 5 and 2 µm. Compared to the analyses of the raw sample and the 0.45 µm permeate, phosphorus fractions in different size ranges can be defined and evaluated in regard to their possible removal.
5. Analyses of a sample using a digestion step (by 100 ºC) will achieve the hydrolysis of organic phosphorus. The total phosphorus analyses in a raw sample contains then the total content of inorganic dissolved, inorganic particulate and organic phosphorus (dissolved plus particulate), i.e. $P_{tot} = P_{diss} + P_{part} + P_{org}$. For the 0.45 µm filtered sample only the dissolved inorganic and organic compounds was determined.

D

Particle analyses at WWTP Beverwijk

D.1 Details particle analyses conducted at WWTP Beverwijk

The following Table D. 1, Table D. 2, Table D. 3, Table D. 4, Table D. 5 and Table D. 6 present the details for the particle analyses conducted at WWTP Beverwijk. Particle counts, volumes and removal efficiencies are shown for the sampling points EF, AF and FI during the filtration tests aiming for blank filtration and flocculating filtration tests.

Table D. 1: Minimum, maximum and average particle counts and volumes (± stdev) for undiluted and diluted samples taken from the sampling point EF over all filtration experiment (number of measurements n=22)

	undiluted			diluted		
	min	max	average ± stdev	min	max	average ± stdev
Particle counts						
Average diameter	5.2	11.6	7.8 ± 1.1	5.4	8.6	6.9 ± 0.7
2-100 µm	6,127	10,724	8,720 ± 1,338	8,683	20,628	12,806 ± 3,191
2-10 µm	4,768	8,528	6,305 ± 1,020	6,944	15,372	10,277 ± 2,644

	undiluted			diluted		
	min	max	average ± stdev	min	max	average ± stdev
10-20 µm	941	3,447	1,701 ± 529	929	4,544	1,964 ± 721
20-100 µm	265	1,509	524 ± 261	226	1,594	564 ± 253
< 10 µm [%]	80	54	74	87	70	80
> 10 µm [%]	20	46	26	13	30	20
Particle volume						
2-100 µm	7.3E+06	4.0E+07	1.4E+07 ± 6.6E+06	6.1E+06	4.5E+07	1.7E+07 ± 7.2E+06
2-10 µm	3.1E+05	8.3E+05	5.5E+05 ± 1.5E+05	4.7E+05	1.3E+06	7.6E+05 ± 2.3E+05
10-20 µm	1.3E+06	5.4E+06	2.4E+06 ± 8.7E+05	1.2E+06	6.7E+06	2.8E+06 ± 1.1E+06
20-100 µm	5.5E+06	3.4E+07	1.1E+07 ± 5.7E+06	4.0E+06	3.7E+07	1.3E+07 ± 6.1E+06
< 10 µm [%]	7	2	4	15	3	7
> 10 µm [%]	93	98	96	85	97	93

Table D. 2: Average particle counts (+stdev) for undiluted and diluted samples taken from the sampling points AF and FI during blank filtration (number of measurements n=6)

	AF		FI	
	undiluted	diluted	undiluted	diluted
Particle counts				
Average diameter	6.70 ± 1.48	6.40 ± 0.56	3.33 ± 0.82	3.93 ± 0.21
2-100 µm	8,221 ± 258	9,927 ± 2,271	2,186 ± 1,029	2,234 ± 1,389
2-10 µm	5,556 ± 909	8,158 ± 1,897	1,839 ± 1,154	2,148 ± 1,280
10-20 µm	1,356 ± 451	1,428 ± 486	43 ± 12	76 ± 40
20-100 µm	395 ± 123	340 ± 122	15 ± 3	10 ± 6
% < 10 µm	79	82	97	96
% > 10 µm	21	18	3	4

Table D. 3: Average particle volumes (+stdev) for undiluted and diluted samples taken from the sampling points AF and FI during blank filtration (number of measurements n=6)

	AF		FI	
	undiluted	diluted	undiluted	diluted
Particle volume				
2-100 µm	1,1E+07 ± 3.3E+06	9.0E+06 ± 3.4E+06	7.6E+05 ± 3.2E+05	5.2E+05 ± 2.8E+05
2-10 µm	4.4E+05 ± 1.0E+05	5.6E+05 ± 1.5E+05	6.6E+04 ± 7.5E+04	8.0E+04 ± 8.0E+04
10-20 µm	1.9E+06 ± 6.5E+05	2.0E+06 ± 6.7E+05	5.4E+04 ± 1.2E+04	9.8E+04 ± 5.1E+04
20-100 µm	8.6E+06 ± 2.7E+06	6.5E+06 ± 2.8E+06	6.4E+05 ± 3.4E+05	3.4E+05 ± 2.4E+05
% < 10 µm	4	6	9	16
% > 10 µm	96	94	91	84

Table D. 4: Average particle counts (+stdev) for undiluted and diluted samples taken from the sampling points AF and FI during flocculating filtration with $FeCl_3$ (number of measurements n=10)

	AF		FI	
	undiluted	diluted	undiluted	diluted
Particle counts				
Average diameter	7.87 ± 1.02	6.44 ± 0.33	4.43 ± 0.47	4.17 ± 0.36
2-100 µm	7,647 ± 1,793	10,686 ± 4,275	1,130 ± 632	1,182 ± 568
2-10 µm	5,653 ± 1,070	8,822 ± 3,734	1,070 ± 603	1,120 ± 553
10-20 µm	1,482 ± 569	1,468 ± 512	45 ± 29	56 ± 18
20-100 µm	512 ± 295	396 ± 156	15 ± 7	7 ± 3
% < 10 µm	74	83	95	95
% > 10 µm	26	17	5	5

Table D. 5: Average particle volumes (+stdev) for undiluted and diluted samples taken from the sampling points AF and FI during flocculating filtration with $FeCl_3$ (number of measurements n=10)

	AF		FI	
	undiluted	diluted	undiluted	diluted
Particle volume				
2-100 µm	1.3E+07 ± 8.1E+06	1.1E+07 ± 3.5E+06	1.6E+06 ± 2.6E+06	3.6E+05 ± 1.8E+05

	AF		FI	
	undiluted	diluted	undiluted	diluted
2-10 µm	4.4E+05 ± 2.5E+05	6.0E+05 ± 3.1E+05	8.1E+04 ± 1.2E+05	3.9E+04 ± 1.7E+04
10-20 µm	1.9E+06 ± 1.2E+06	2.1E+06 ± 7.3E+05	2.3E+05 ± 5.2E+05	7.0E+04 ± 2.3E+04
20-100 µm	1.0E+07 ± 6.8E+06	7.8E+06 ± 2.6E+06	1.3E+06 ± 1.9E+06	2.5E+05 ± 1.7E+05
% < 10 µm	8	10	4	11
% > 10 µm	92	90	96	89

Table D. 6: Average particle counts and volumes (+stdev) for undiluted and diluted samples taken from the sampling points AF and FI during flocculating filtration with PACl (number of measurements n=8)

	AF		FI	
	undiluted	diluted	undiluted	diluted
Particle counts				
Average diameter	7.02 ± 1.04	5.88 ± 0.50	5.79 ± 2.26	5.18 ± 1.27
2-100 µm	8,655 ± 1,653	14,217 ± 2,959	2,114 ± 2,221	2,641 ± 2,726
2-10 µm	6,944 ± 832	12,378 ± 2,456	1,654 ± 1,476	2,254 ± 2,147
10-20 µm	1,312 ± 605	1,447 ± 449	313 ± 502	296 ± 432
20-100 µm	399 ± 240	392 ± 154	146 ± 257	91 ± 154
% < 10 µm	80	87	78	85
% > 10 µm	20	13	22	15
Particle volume				
2-100 µm	1.0E+07 ± 5.9E+06	9.9E+06 ± 4.0E+06	4.2E+06 ± 7.2E+06	2.4E+06 ± 3.8E+06
2-10 µm	6.3E+05 ± 1.7E+05	9.0E+05 ± 2.3E+05	1.3E+05 ± 1.6E+05	1.7E+05 ± 1.9E+05
10-20 µm	1.8E+06 ± 8.9E+05	1.9E+06 ± 6.2E+05	4.6E+05 ± 7.6E+05	4.1E+05 ± 6.1E+05
20-100 µm	7.6E+06 ± 4.9E+06	7.1E+06 ± 3.3E+06	3.6E+06 ± 6.2E+06	1.8E+06 ± 3.0E+06
% < 10 µm	6	9	3	8
% > 10 µm	94	91	97	92

E

Additional results during simultaneous nutrient removal

E.1 Online turbidity measurements obtained at WWTP Horstermeer

The following Figure E. 1, Figure E. 2 and Figure E. 3 shows the online turbidity measurements for the filtration tests aiming at simultaneous nutrient removal. The values are shown for the WWTP effluent (EF) and the filtrate (FI) of the pilot scale dual media filter applied at WWTP Horstermeer between August and October 2005.

The turbidity values of the WWTP effluent showed variation between 6 and 16 NTU. Incidental peeks up to 20 NTU may be caused by increased wash out of sludge particles from the final sedimentation tank during rain weather conditions. But also fouling of the turbidity meter was found to be responsible for sudden value increase.

After filtration the turbidity was reduced to 1 to 2 NTU during normal filter operation. Increased turbidity values > 4 NTU was also dedicated to fouling of the online measuring device.

However, these figures indicate more stable filtrate turbidity values in periods with less variable effluent turbidity; between 6 and 12 NTU.

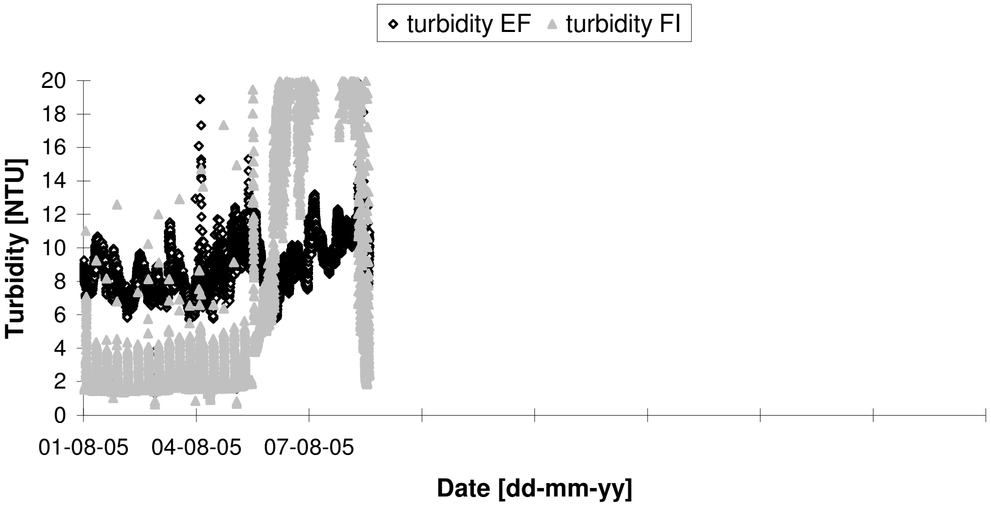

Figure E. 1: Online turbidity for the sieved WWTP effluent EF and the filtrate FI during denitrifying flocculating filtration with a pilot scale dual media filter at WWTP Horstermeer for August 2005

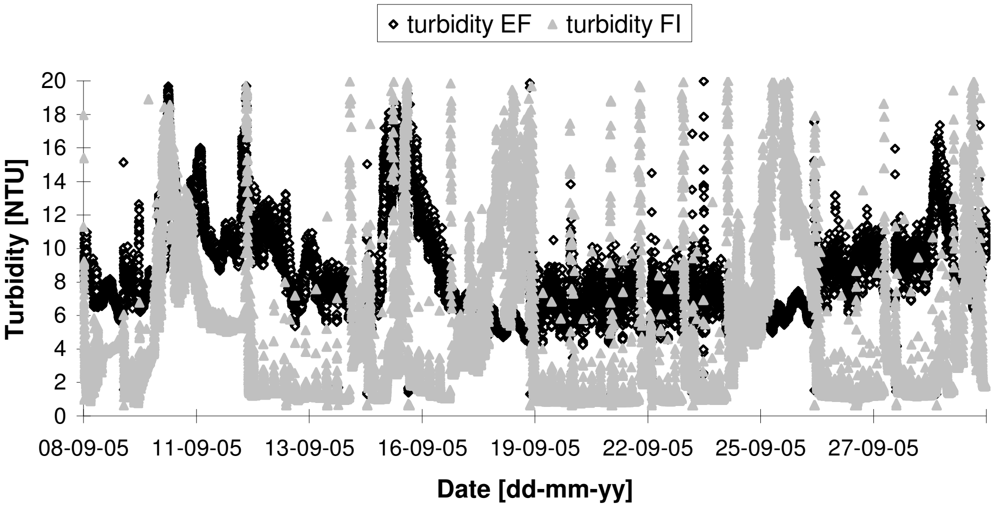

Figure E. 2: Online turbidity for the sieved WWTP effluent EF and the filtrate FI during denitrifying flocculating filtration with a pilot scale dual media filter at WWTP Horstermeer for September 2005

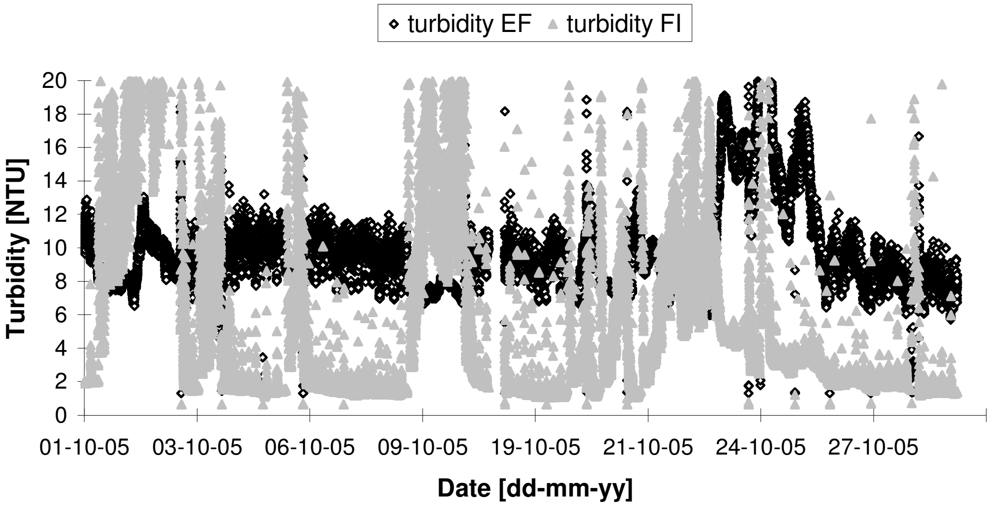

Figure E. 3: Online turbidity for the sieved WWTP effluent EF and the filtrate FI during denitrifying flocculating filtration with a pilot scale dual media filter at WWTP Horstermeer for October 2005

E.2 Kjeldahl and ammonium nitrogen measurements obtained at WWTP Horstermeer

Kjeldahl and ammonium nitrogen concentrations were measured within the regular monitoring campaign at WWTP Horstermeer. Figure E. 4 gives an overview of those analyses between 2005 and 2006 when pilot filtration tests were performed.

In this figure the development of Kjeldahl nitrogen is in accordance to ammonium nitrogen resulting in a relatively constant average organic nitrogen concentrations between 1-2 mgN$_{org}$/L, which derives form the difference between the Kjeldahl and ammonium nitrogen concentrations.

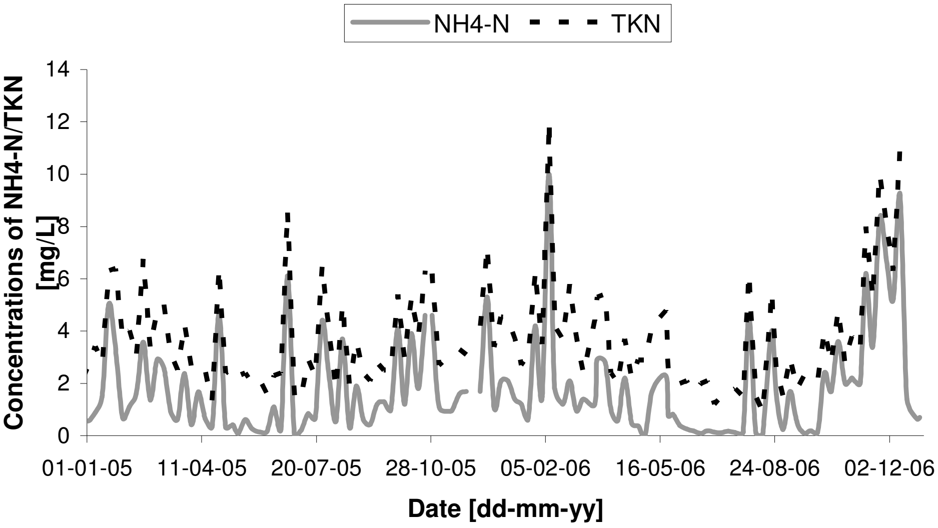

Figure E. 4: Analyses of ammonium (NH$_4$-N) and Kjeldahl nitrogen (TKN) conducted within the regular measuring campaign at the WWTP Horstermeer

E.3 Summary of average values and standard deviations for manual measured parameters at WWTP Horstermeer

Table E. 1and Table E. 2 give a summary of parameters (average and standard deviations) that were monitored frequently during the filtration experiments at WWTP Horstermeer between 2005 and 2006.

The average concentrations were calculated for all results during the experiments in 2005 and 2006, eventually including also measurements obtained during sub-optimal process conditions. Consequently, the concentrations of the selected parameters after filtration may be used to indicate the minimum achievable filtrate quality with the displayed feed water characteristics.

Table E. 1: Average concentrations and standard deviations for several parameters obtained with manual analyses during pilot scale investigations between June – December 2005 at WWTP Horstermeer

		WWTP effluent (EF)		Filter feed water (AF)		Filtrate (FI)	
Parameter	Unit	average	stdev	average	stdev	average	stdev
CZV*	[mg/L]	32.1	7.5	76.5	26.6	38.4	22.7
N_{tot}^*	[mg/L]	11.3	4.3			3.0	2.5
NO_3-N	[mg/L]	9.1	3.3	9.3	2.7	1.6	1.5
NH_4-N	[mg/L]	1.9	0.9			1.2	0.01
P_{tot}^*	[mg/L]	0.72	0.29	0.72	0.43	0.35	0.30
PO_4-P*	[mg/L]	0.65	0.41	0.64	0.35	0.31	0.30
Sulphate	[mg/L]	57.5	16.4	57.1	17.0	60.7	15.9
Turbidity	[NTU]	4.4	1.7	5.6	2.2	1.4	0.8
pH	[-]	6.37	0.25	6.32	0.34	6.39	0.34
O_2	[mg/L]	6.7	1.3	6.6	1.3	2.3	1.4
Temperature	[°C]	21.1	0.4			21.3	0.4

*: measurements of unfiltered samples

Table E. 2: Average concentrations and standard deviations for several parameters obtained with manual analyses during pilot scale investigations between March – December 2006 at WWTP Horstermeer

		WWTP effluent (EF)		Filter feed water (AF)		Filtrate (FI)	
Parameter	Unit	average	stdev	average	stdev	average	stdev
CZV*	[mg/L]	33.0	10.5	77.2	27.6	38.3	28.5
N_{tot}^*	[mg/L]	12.6	3.7			4.4	2.0
NO_3-N	[mg/L]	9.3	5.5			1.9	3.0
NH_4-N	[mg/L]	0.8	1.1			0.6	1.1
P_{tot}^*	[mg/L]	0.85	0.40			0.31	0.27
PO_4-P*	[mg/L]	0.70	0.42			0.24	0.21
Turbidity	[NTU]	4.8	2.9			1.5	1.2
pH	[-]	6.3	0.2			6.4	0.2
O_2	[mg/L]	2.7	0.7			1.0	0.5
Temperature	[°C]	19.1	2.9				

*: measurements of unfiltered samples

List of figures and tables

List of figures

List of tables

Summary

In the Netherlands the quality of wastewater treatment plant (WWTP) effluents discharged to surface water bodies is becoming an increasingly severe issue in water management practice. Constituents such as phosphorus, nitrogen and micropollutants causing eutrophication and pollution at the receiving water body are originated by these point sources. Their discharge concentrations will be therefore strongly regulated in the future due to new regulations which derived from the European Water Directive. Low nutrient levels, 2.2 mgN_{tot}/L for total nitrogen and 0.15 mgP_{tot}/L for total phosphorus, defined as 'maximum tolerable risk" (MTR) concentrations in the Dutch legislation, and the reduction of micropollutants up to zero-discharge for several substances will be required. The advanced removal through an effluent polishing step combining the removal of several substances may lead to a serious option for existing wastewater treatment plants in the near future.

As to the improvement of the WWTP effluent quality, conventional filtration appears to take a major place in future treatment scenarios in the Netherlands and other European Countries. The removal of suspended matter, including inorganic dissolved phosphate, enhanced by prior precipitation, coagulation and flocculation, and the biological removal of nitrogen and organic matter with denitrification are processes that may be combined within one filtration step.

The application of filtration within the modern Dutch WWTP effluent treatment is still new. This has led to intensive investigations in the last years on different scales at various WWTP's. One possible conventional filtration technique is rapid media (deep bed) filters operated in a continuous or discontinuous mode. The selection of the process depends on the aimed purpose either to reach very low suspended solids and/or phosphorus concentrations in the final effluent. The particle distribution in WWTP effluent between 2-100 μm and the probable presence of phosphate particulates within this range require a filtration technique which is capable to remove sufficiently even small particles. Practical experience indicated a better removal of small particles (2-5 μm) including phosphate precipitates with dual media discontinuous filtration compared to mono media continuous filtration. Dual media discontinuous filtration is therefore the selected process applied in the presented investigations for advanced treatment of secondary WWTP effluent.

The idea of a multi purpose filtration step aiming at the removal of suspended solids and phosphorus even combined with the removal of nitrogen and maybe other micropollutants led to the conclusion that standard filtration practice has to be reconsidered and optimised. Current models used to predict filtration performances are predominantly based on the traditional filtration theory which is defined for discrete particle removal. This approach seems to fail for the application of filtration for combined nutrient removal dealing with simultaneous physical, chemical and biological processes. The mutual interactions of these processes affecting the filtration mechanisms, changes of particle distributions throughout filtration, and fluctuations of the feed water quality are expected to interfere with the predictability of the filtration performance. Standard operational parameters such as optimal filtration rates, backwash procedure and frequencies may need to be evaluated and adjusted during filtration experiments in order to achieve optimal simultaneous suspended solids and nutrient removal. Practical experience with this multi purpose filtration with actual WWTP effluent still has to be extended.

The starting point for the investigations is the average quality of WWTP effluent in the Netherlands, after extended nutrient removal, which can generally be described with concentrations of suspended solids ~ 10 mgSS/L, total phosphorus < 2 mgP_{tot}/L and total nitrogen < 10 mgN_{tot}/L. This effluent quality shows the opportunity for further improvement and was available at the chosen research locations.

Filtration tests at different scales were carried out on three different locations in the Netherlands in order to gain practical experience for this new application. The results are presented in this thesis. Initial results were gained with pilot scale filtration at WWTP Utrecht, followed by verifying lab scale filtration tests at WWTP Beverwijk. Long term filtration on pilot scale with more optimized operation was finally conducted at WWTP Horstermeer.

The emphasis of the filtration experiments was to investigate the applicability of multi purpose filtration for the Dutch WWTP effluent and the most optimal filter operation in order to achieve the reduction of suspended solids and nutrients to very low levels. Any adverse effect of the simultaneous denitrification on the suspended solids and phosphorus removal needed to be excluded. The optimisation of the dosing conditions and filter backwash seemed to be important operational issues in order to develop a stable filtration process and high removal efficiencies. Next to the monitoring of conventional water quality parameters and pressure measurements, the additional value of particle analysis, as a new and potential tool for monitoring and control, was an important research topic. The applicability of particle analysis during effluent filtration and the relationship of particles with other conventional parameters were assumed to improve filter monitoring.

The performed filtration tests included investigations for

- single suspended solids removal (blank filtration),
- combined suspended solids and phosphorus removal (flocculating filtration),
- combined suspended solids and nitrogen removal (denitrifying filtration),
- simultaneous removal of suspended solids, phosphorus and nitrogen (denitrifying flocculating filtration).

Preliminary jar tests with WWTP effluent were performed to select an appropriate coagulant type and the dosage. Poly aluminium chloride was indicated as the most optimal coagulant for almost all jar tests. The metal-orthophosphate ratios of MePO 3.0 (mol/mol) appeared to be a sufficient dose with good results for the reaction of metal ions with dissolved inorganic phosphate ions (orthophosphate).

However, to be able to evaluate phosphorus removal, either during jar tests or later filtration experiments, the precise definition of phosphorus fractions seemed to be essential. Inorganic and organic phosphorus fractions, either in particulate or dissolved form (<0.45 μm), can be analysed depending on sample treatment. The change of their distribution before and after chemical precipitation and following filtration determines the efficiency of these processes and helps to optimise the chemical dosing conditions. The organic particulates and inorganic precipitates will be removed by filtration depending on their size.

The analyses of the total inorganic phosphorus (or total phosphate) of the unfiltered filtrate sample versus the orthophosphate concentration before and after filtration indicated the particulate phosphate removal during the filtration experiments. Additional total phosphorus analyses were used to observe the overall phosphorus removal including organic phosphorus fractions and to evaluate whether a very low filtrate concentration of 0.15 mg/L (MTR) could be achieved or not.

Filtration results on the three WWTP's were mainly obtained at a filtration rate of 10 m/h. A metal salt dosage proportional to the online measured orthophosphate concentration the WWTP effluent ensured sufficient dosages and prevented an over or under dosage. Compared to the results in preliminary jar tests higher dose rates, such as MePO ratios of 4.0-4.8 (mol/mol) at WWTP Horstermeer, resulted in satisfactory phosphorus removal. Over all filtration experiments a total phosphorus concentration of averagely 0.15 mg/L was obtained with feed water (WWTP effluent) concentrations below 0.8 mgPO_4-P/L. The phosphorus removal during single flocculating filtration and simultaneous denitrifying flocculating filtration showed similar results excluding any adverse effect of denitrification. Also filtrate turbidity was structurally below 2 NTU, independent of the filter operation and the feed water turbidity confirming the comparable removal of suspended solids during different filter operations.

The organic phosphorus concentration (dissolved plus particulate) of the three WWTP effluents was calculated to be 0.1 – 0.3 mg/L. The particulate organic phosphorus is completely removed during filtration while a concentration of << 0.1 mg/L of dissolved organic phosphorus could not be removed by filtration. This indicates that in situations with high concentrations of dissolved organic phosphorus in a WWTP effluent aiming to reach the MTR level may be unsuccessful.

To enhance denitrification methanol was used as external carbon source due to its high biological degradability. Increased dosing during start-up indicated good denitrification after one week of operation. During normal operation methanol-nitrate ratios of 4.5-4.7 gCH_3OH/gNO_3-N were applied, considering an overdose of 40 % for the reduction of free oxygen. A high removal of nitrate nitrogen resulted in final concentrations of << 2.0 $mgNO_3$-N/L with conversion rates up to 3 $kgN/kgm^3_{bedvolume}$.d, even with oxygen concentrations of 1.0 mg/L in the filtrate. Total nitrogen concentrations of < 2.2 mgN_{tot}/L (MTR) were frequently achieved. Comparable phosphorus and nitrogen removal results with similar conversion rates were found even with a filtration rate of 12.5 m/h, investigated at a later stage and during a winter period (average 13ºC in the WWTP effluent) at WWTP Horstermeer.

However, the performance of the biological processes at the WWTP's prior to filtration showed to be a critical issue when final low nitrogen concentrations are required. Incidental increased Kjeldahl-nitrogen concentrations were observed during the experiments at the WWTP's, which may be attributed to increased ammonium concentrations due to less optimal nitrification. In this case, denitrifying filtration is not capable of reaching low total nitrogen levels (MTR). Sub-optimal performance of biological treatment at the WWTP should therefore be avoided.

In order to gain a stable multi purpose filtration and to achieve the above described low nutrient levels the backwash regime needed to be adjusted. The main backwash procedure was optimised with a combined air scour and water back flush in the first phase and decreasing water back flush velocities in the following phases. This ensured the breakage of the bed, separation of the filter grains and proper stratification of the filter material. A subsequent backwash procedure ("bumping cleaning") applying a strong water back flush (50 m/h) every 3 hours was implemented to release the nitrogen gas formed by denitrification. This backwash regime resulted in filter run times up to 24 hours. The monitoring of the pressure development over the filter bed during filter run time was helpful in order to observe the increasing filter bed resistance, indicate filter bed clogging and to ensure effectiveness of the applied backwash procedure and frequency. Especially the filtration tests at WWTP Horstermeer showed reversibility of the filter bed pressure after "bumping cleaning" and gradual increase of filter bed resistance in time below the critical resistance. Although nitrate removal deteriorated after main backwash, most likely due to rinsing of biomass, the original level was restored after one hour of operation. At that stage of the investigations the applied backwash regime showed to be sufficient.

Further investigations including profile measurements showed continuing removal of nitrogen, oxygen, turbidity and phosphate particulates throughout the applied dual media filter bed proving efficient deep bed filtration and the usage of the complete accumulation capacity. The chosen filter bed, 40 cm of quartz sand below (0.8-1.25 mm) and 80 cm of anthracite above (2.0-4.0 mm) seemed to be an appropriate configuration for this multi purpose filtration.

The removal of particulate phosphate during dual media filtration was observed more closely by phosphate fractionation. Fractionations were performed with filtration of samples over different membrane filters (0.45 µm, 2 µm, 5 µm, 10 µm and 20 µm cellulose acetate and polycarbonate filters) and showed the fraction 0.45 µm and 2 µm to be the most difficult fraction for filtration. The concentration of this fraction seemed to be influenced by the choice of coagulant. The formation of the fraction 0.45 - 2 µm after initial precipitation and coagulation and its removal during filtration obviously depend on the active aluminium (Al_2O_3) concentrations of the applied poly aluminium chloride. Furthermore, a strong relation between turbidity and the determined phosphate fractions was found for total phosphate concentrations in the WWTP effluent between 0.1 and 0.9 mgP/L. This relation may be used to improve filter monitoring for flocculating filtration.

Investigations of three layer filtration aimed to evaluate the further improvement of the final effluent quality after dual media filtration. Therefore, the dual media filter was operated for denitrifying flocculating filtration and followed up by two single layer filters, one filled with fine sand (0.5 – 0.8 mm grain diameter and 30 cm height) and the other filled with activated carbon (1.6 mm grain diameter and 1.2 m height). The filtrates of both single layer filters indicated a further removal of turbidity, total phosphates, nitrate nitrogen and chemical oxygen demand.

Next to the removal of suspended solids and nutrients attention was given to the removal of heavy metals and other substances. The removal of three selected heavy metals, copper, nickel and zinc was investigated during the filtration experiments. Denitrifying flocculating filtration resulted in a removal of copper (>92%), zinc (60%) and nickel (10-20%). The removal of other substances, such as several polycyclic aromatic hydrocarbons (PAH's) and endocrine disrupting compounds (EDC's), appeared to be very low. However, the removal of heavy metals and other substances can be exclusively attributed to the filtration of filterable matter that formed complexes with these constituents prior to filtration.

The jar tests with powdered activated carbon and third layer filtration with granular activated carbon indicated high removal of the three selected heavy metals through adsorption, thus most likely also indicating indirect removal through complexation with organic matter. Although, the removal of other organic micropollutants with third layer activated carbon filtration was not investigated within these research published literature reports that removal of several substances is most likely. The integration of the adsorption mechanisms within filtration aiming simultaneous removal of suspended solids and nutrients could therefore improve the value of this multi purpose filtration step.

The application of particle analysis aimed to investigate a possible additional value for filter monitoring also in combination with conventional analyses, such as particulate phosphate and turbidity. For a proper utilisation sample dilution showed to be of great importance when small particles have to be counted accurately. Furthermore, the particle volume appeared to be the more reliable parameter in comparison to particle counts.

Particle size and volume distributions, and the average particle diameter were determined to evaluate the change of the particle distributions during coagulation, flocculation and filtration. The increase of total particle number and volume after completed coagulation and flocculation was monitored. Although, an expected growth in particle size after flocculation was missing. Obvious development of small flocs during flocculating filtration can be attributed to sweep coagulation as one of the two involved coagulation processes. The formation of small flocs has obviously no negative effect on the suspended solids and total phosphorus removal. Investigation of the direct relationship of particles, turbidity and phosphate particulates resulted in inconsistent results. Total particle numbers were well related with turbidity measurements while a good correlation was found between the removal efficiencies of phosphate particulates and the total particle volume for blank filtration and flocculating filtration (with poly aluminium chloride). However, the linear regression indicated that the correlation decreases for flocculating filtration compared to filtration without metal salt dosing. This observation may be directly linked to the particulate phosphate distributions observed during the fractionation experiments. The formation of phosphate particulates between 0.45 - 2 µm as most difficult for filtration increases due to metal salt addition but can not be measured with the applied particle counter because of its particle size limit of 2 µm. The application of a device counting in a lower particle range may lead to a more accurate relationship between particles and phosphate particulates.

It is concluded, that the removal of suspended solids, phosphorus and nitrogen can be combined in just one conventional effluent filtration step with optimised operation resulting in a stable filtration performance on a long term. Particle countings and phosphate fractionations are not established as independent monitoring parameters but they improved the understanding of the process where removal of small particles and phosphate fractions shows to be very important.

Samenvatting

Nog steeds is een groot deel van eutrofiërende en verontreinigende stoffen, zoals fosfor, stikstof en microverontreinigingen, in het Nederlandse oppervlaktewater afkomstig van rioolwaterzuiveringsinstallaties (rwzi's). Zodoende is in het waterbeheer de verbetering van de kwaliteit van rwzi effluent als puntbron van deze stoffen een aandachtspunt dat aan importantie wint. De lozingseisen van deze stoffen zullen daarom in de toekomst worden aangescherpt door met name het van kracht worden van nieuwe voorschriften. De Europese Kaderrichtlijn Water is hier één van. De toekomstige verankering van deze richtlijn in de Nederlandse wetgeving zal in een aantal gevallen leiden tot de eis naar lagere concentraties van nutriënten (totaal stikstof < 2,2 mgN_{tot}/L en totaal fosfor < 0,15 mgP_{tot}/L), gedefinieerd als het maximum toelaatbaar risiconiveau (MTR). Daarnaast zal zelfs een reductie van bepaalde microverontreinigingen tot nullozing worden vereist. De geavanceerde verwijdering door middel van één nazuiveringstrap voor de gecombineerde verwijdering van meerdere stoffen is een serieuze optie voor bestaande rwzi's in de nabije toekomst.

Conventionele filtratie voor de verbetering van de rwzi effluentkwaliteit blijkt een steeds belangrijkere plaats in te nemen in toekomstige zuiveringsscenario's in Nederland en andere Europese landen. De verwijdering van zwevende stof samen met anorganisch gebonden fosfaat, na precipitatie, coagulatie en flocculatie, en de biologische verwijdering van stikstof en organisch materiaal met denitrificatie zijn daarbij processen die gecombineerd kunnen worden in één filtratiestap.

In de huidige Nederlandse situatie is de toepassing van filtratie als nazuiveringstap van het moderne effluent van rwzi's nog steeds een nieuwe applicatie. Dit heeft de laatste jaren geleid tot intensief onderzoek op verschillende schaalgroottes en op verschillende rwzi's als proeflocatie. Een potentiële conventionele filtratietechniek is snelfiltratie (dieptefiltratie) die zowel continu als discontinu bedreven kan worden. De selectie van het toe te passen proces hangt af van de nagestreefde effluent kwaliteit. De omvang van aanwezige deeltjes van het effluent van een rwzi ligt tussen de 2-100 µm. Gezien de aanwezigheid van fosfaat in deze fracties is er een techniek beoogd die geschikt is om zelfs de kleinste deeltjes succesvol te verwijderen. Praktijkervaring heeft aangetoond dat dubbellaags discontinue filtratie tot een betere verwijdering tendeert ten aanzien van kleine deeltjes (2-5 µm) inclusief fosfaat precipitaten, dan enkellaags continue filtratie.

Dubbellaags discontinue filtratie is daarom gekozen voor de toepassing in dit onderzoek naar de verdergaande zuivering van secundair rwzi effluent.

Het idee van een filtratieproces dat meerdere zuiveringsaspecten dient, zoals de verwijdering van zwevende stof, fosfaat, stikstof en misschien zelfs voor de reductie van andere microverontreinigingen, heeft geleid tot de conclusie dat de gestandaardiseerde aanpak voor filtratie opnieuw overwogen en geoptimaliseerd moet worden. Bestaande filtratiemodellen, die ingezet worden voor het voorspellen van de filtratiewerking, zijn hoofdzakelijk gebaseerd op de traditionele filtratietheorie. Deze is wederom gedefinieerd voor discrete deeltjesverwijdering. Deze aanpak blijkt niet te werken voor de toepassing van filtratie voor de gecombineerde nutriëntenverwijdering waarin fysische, chemische en biologische processen simultaan plaatsvinden. Het is te verwachten dat interactie tussen deze processen de voorspelbaarheid van het te behalen filtratierendement zullen beïnvloeden. Hierbij valt te denken aan filtratiemechanismen, veranderingen in deeltjesverdeling gedurende de filtratie en fluctuaties in de kwaliteit van het voedingswater. De standaard in te stellen bedrijfsvoeringsparameters zoals optimale filtratiesnelheid en de aan te houden terugspoelprocedures en –frequenties zullen tijdens de filtratietesten geëvalueerd en bijgesteld moeten worden voor het behalen van een optimale simultane verwijdering van zwevende stof en nutriënten. Aangezien de praktijkervaring met nafiltratie van rwzi effluent voor het reduceren van zowel zwevende stof als verschillende nutriënten nog relatief weinig is toegepast is verder onderzoek van groot belang.

Het uitgangspunt voor het onderzoek is de inschatting van de gemiddelde kwaliteit van rwzi effluent in Nederland, na geavanceerde nutriëntenverwijdering. Deze kan algemeen beschreven worden met concentraties voor zwevend stof ~ 10 mgSS/L, totaal fosfor < 2 mgP_{tot}/L en totaal stikstof < 10 mgN_{tot}/L. Deze effluentsamenstelling biedt kans voor verdergaande kwaliteitsverbetering en is aanwezig op de geselecteerde onderzoekslocaties.

Aan de hand van filtratietesten op verschillende schaalgrootte en op drie verschillende locaties in Nederland is praktijkervaring met de nieuwe applicatie opgedaan. De resultaten uit dat onderzoek zijn eveneens opgenomen in dit proefschrift. De initiële filtratie experimenten zijn uitgevoerd op pilotschaal op de rwzi Utrecht, gevolgd door labschaal filtratietesten op de rwzi Beverwijk. De duurproeven op pilotschaal, ondersteund met geoptimaliseerde bedrijfsvoering, zijn afsluitend uitgevoerd op de rwzi Horstermeer.

De nadruk voor de filtratieproeven was gericht op de toepasbaarheid van filtratie voor het Nederlandse rwzi effluent met gecombineerde zuiveringsmechanismen en het vinden van de meest optimale bedrijfsvoering voor het behalen van lage concentraties voor zwevende stof en nutriënten. Een eventueel nadelig effect van simultane denitrificatie op de zwevende stof en fosfaatverwijdering moest hierin uitgesloten worden. De optimalisatie van het doseringsregime en terugspoelingprocedures waren belangrijke operationele aandachtspunten voor het verkrijgen van een stabiel filtratieproces gekoppeld aan hoge verwijderingrendementen. De toegevoegde waarde van deeltjestellingen als een nieuw instrument voor het monitoren en de procescontrole was naast het meten van conventionele waterkwaliteitsparameter en drukmetingen een interessante onderzoekstopic. Er werd aangenomen dat de toepassing van deeltjestellingen in het te filtreren effluent en de relatie van deeltjes met andere conventionele parameters het monitoren tijdens filtratie kan verbeteren. De uitgevoerde filtratietesten bestonden uit een stapsgewijze uitbreiding van de proeven voor:

- zwevende stofverwijdering (blanco filtratie),
- gecombineerde zwevende stof en fosfaatverwijdering (vlokkingsfiltratie),
- gecombineerde zwevende stof en stikstofverwijdering (denitrificerende filtratie),
- simultane verwijdering van zwevende stof, fosfaat en stikstof (denitrificerende vlokkingsfiltratie).

Ter bepaling van een geschikte soort coagulant en de startdosering voor initiële filtratietesten zijn bekerglasproeven met rwzi effluent uitgevoerd. In bijna al deze proeven was polyaluminiumchloride het meest optimale coagulant. Met het oog op de reactie van de metaalionen met het opgelost anorganische fosfaat (orthofosfaat) leidde een metaal-orthofosfaat verhouding van MePO 3,0 (mol/mol) tot goede resultaten.

Voor de beoordeling van fosfaatverwijdering zowel tijdens bekerglasproeven als latere filtratietesten is het belangrijk om de fosforfracties te definiëren. Anorganische en organische fosforfracties, in gebonden of opgeloste vorm (<0,45 μm), kunnen afhankelijk van de monstervoorbehandeling geanalyseerd worden. De verandering van de verdeling voor en na chemische precipitatie en navolgende filtratie bepaalt de efficiëntie van deze processen en maakt een optimalisatie van de chemische dosering mogelijk. Organisch gebonden en anorganische neerslag kunnen vervolgens, overigens afhankelijk van de deeltjesgrootte, door middel van filtratie verwijderd worden.

De analyse van totaal anorganisch fosfor (of totaal fosfaat) van een ongefilterd monster versus de orthofosfaat concentratie die bepaald is voor en na de filtratie, toont daarbij de verwijdering van gebonden fosfaat aan tijdens de proeven. Additionele totaal fosfor bepalingen zijn uitgevoerd voor het vaststellen van de totale fosfor verwijdering. Deze bepalingen zijn inclusief organische fosforfracties en ter beoordeling van het behalen van zeer lage filtraatconcentraties richting de MTR-kwaliteit van 0,15 mg/L.

De filtratieresultaten op de drie rwzi’s zijn verkregen bij een filtratiesnelheid van 10 m/h. Om te lage dosering of overdosering te vermijden vond metaalzoutdosering op fosfaat debietsproportioneel plaats, gebaseerd op online gemeten orthofosfaat in het rwzi effluent. Vergeleken met de resultaten van de voorafgaande bekerglasproeven is een acceptabele fosfaatverwijdering bereikt met hogere metaal-orthofosfaat verhoudingen dan vooraf ingeschat. Aangaande het onderzoek op de rwzi Horstermeer zijn bijvoorbeeld bij een MePO 4,0-4,8 (mol/mol)-verhouding de beste resultaten bereikt. Voor alle uitgevoerde filtratieproeven kan geconcludeerd worden dat een totaal fosfor concentratie van 0,15 mgP_{tot}/L behaald wordt bij een orthofosfaatconcentratie van onder 0,8 mg/L in het voedingswater (rwzi effluent). De totaal fosforverwijdering tijdens vlokkingsfiltratie en simultane denitrificerende vlokkingsfiltratie lieten een vergelijkbaar resultaat zien waarbij een eventueel nadelige effect van denitrificerende biomassa uitgesloten is. Ook werd de filtraattroebelheid, onafhankelijk van de toegepaste bedrijfsvoering en troebelheid in het voedingswater, structureel onder 2 NTU gemeten. Dit bevestigt een vergelijkbare verwijdering van zwevende stof tijdens de verschillende filtratieproeven.

Het organische fosfaatgehalte (opgelost plus gebonden) van de drie rwzi’s was berekend tussen 0,1 – 0,3 mg/L. De gebonden fracties werden compleet verwijderd terwijl een concentratie van << 0,1 mg/L opgelost organisch fosfor in het filtraat niet verder verwijderd kon worden door middel van filtratie. Hoge concentraties van opgelost organisch fosfor in het rwzi effluent kunnen als fractie van totaal fosfor problematisch verwijderd worden als het behalen van het MTR-niveau een eis is.

Ter bevordering van denitrificatie is methanol gebruikt als externe koolstofbron vanwege zijn hoge biologische afbreekbaarheid. Een verhoogde dosering tijdens de opstart van de denitrificatie leverde al vanaf de tweede week na opstarten goede resultaten. Tijdens een normale bedrijfsvoering zijn methanol-nitraat verhoudingen van 4,5-4,7 gCH_3OH/gNO_3-N toegepast, inclusief een overdosering van 40% voor de verwijdering van vrije zuurstof. De goede denitrificatie resulteerde in filtraatconcentraties van << 2,0 mgNO_3-N/L met omzettingen van boven de 3 kgN/d/kg$m^3_{bedvolume}$ en bij zuurstofconcentraties van 1,0 mg/L in het filtraat. Totaal stikstofconcentraties van < 2,2 mgN_{tot}/L (MTR) zijn frequent aangetoond. Vergelijkbare resultaten voor fosfor, stikstofverwijdering en omzetting zijn zelfs behaald bij een filtratiesnelheid van 12,5 m/uur. Dit is op een later tijdstip en in de winterperiode (gemiddeld 13°C in het rwzi effluent) aangetoond op de proeflocatie op de rwzi Horstermeer.

De werking van de voorafgaande biologische processen op de rwzi’s voorafgaande aan de filtratiestap bleek een kritiek aspect te zijn. Dit aspect speelt vooral als lage eindconcentraties voor stikstof gewenst zijn. Incidenteel verhoogde Kjeldahl-stikstof concentraties, die zijn gemeten tijdens de testen op de rwzi’s, zijn toe te schrijven aan verhoogde ammoniumconcentraties veroorzaakt door een minder goed werkend nitrificatieproces. In dit geval is denitrificerende filtratie in zijn capaciteit beperkt om voldoende lage totaal stikstofconcentraties (MTR) te bereiken. Een niet optimale werking van de biologische voorzuivering op de rwzi moet daarom vermeden worden.

Voor het behalen van een stabiele filtratie voor de diverse zuiveringsparameters en het bereiken van lage concentraties van nutriënten moest de terugspoeling aangepast worden. De reguliere terugspoelprocedure is uiteindelijk geoptimaliseerd door het toepassen van een gecombineerde lucht-water spoeling in de eerste fase en teruglopende watersnelheden in de aansluitende fases. Deze combinatie zorgt voor het breken van het filterbed, scheiding van de filterkorrels en voldoende stratificatie van het filtratiemateriaal. Een tweede terugspoelprocedure ("bumping cleaning") werd geïntroduceerd met een hoge watersnelheid (50 m/uur) die om de 3 uur werd aangewend, zodat het tijdens de denitrificatie gevormde stikstofgas uit het filterbed wordt gedreven. Het toepassen van dit terugspoelregime met de gecombineerde procedures, reguliere spoeling plus "bumping cleaning", resulteerde in filterlooptijden tot 24 uur. Drukmetingen over het filterbed tijdens de filtratieruns zijn noodzakelijk om de optredende weerstandstoename over het bed te monitoren, verstoppingen zichtbaar te maken en de efficiëntie van het toegepaste terugspoelregime aan te tonen. Vooral de filtratietesten op rwzi Horstermeer laten zien dat de schoonbedweerstand na de "bumping cleaning" zich herstelt en de graduele toename van de filterbedweerstand in tijd beneden de kritische weerstand is aangetoond. Ondanks dat de nitraatverwijdering iets verslechtert na de reguliere terugspoeling, wat waarschijnlijk veroorzaakt werd door het uitspoelen van denitrificerende biomassa, is het originele niveau weer hersteld na een uur bedrijf. Op dit punt van het onderzoek is geconstateerd dat het toegepaste spoelregime toereikend is.

Verder werd een continue verwijdering van stikstof, zuurstof, en gebonden fosfaat en de vermindering van de troebelheid over het dubbellaags filterbed aangetoond door profielmetingen. Dit duidt op een efficiënte werking van dieptefiltratie en de complete benutting van de bergingscapaciteit. Het gekozen filterbed, bestaande uit 40 cm kwartzand (0,8-1,25 mm) beneden en 80 cm antraciet (2,0-4,0 mm) boven bleek een acceptabele configuratie voor filtratie die toepasbaar is voor het zuiveren op zwevende stof en verschillende nutriënten.

De verwijdering van gebonden fosfaat tijdens de dubbellaags filtratie is nader onderzocht door fractioneringsmetingen van fosfaat uit te voeren. Deze fractioneringen werden uitgevoerd door middel van filtratie van watermonsters over verschillende membraanfilters (0,45 μm, 2 μm, 5 μm, 10 μm en 20 μm cellulose acetaat en polycarbonaat filters). De fosfaatfractie tussen 0,45 μm en 2 μm was het meest problematisch voor filtratie. De concentratie van deze fractie leek beïnvloedbaar door de coagulantkeuze. Het ontstaan van de fractie 0,45 - 2 μm na initiële precipitatie en coagulatie en de verwijdering ervan tijdens filtratie was afhankelijk van de concentratie van actief aluminium (Al_2O_3) in het toegepaste polyaluminiumchloride product. Verder is een goede relatie gevonden tussen de troebelheid en de bepaalde fosfaatfracties; wat met name geldt voor totaal fosfaatconcentraties tussen 0,1 en 0,9 mgP/L in het rwzi effluent. Dit verband kan gebruikt worden voor een verbeterde monitoring van vlokkingsfiltratie.

Het deelonderzoek met betrekking tot filtratie met drie filterlagen is uitgevoerd om verdergaande verbetering van de eindwaterkwaliteit aan te tonen na dubbellaags filtratie. Het dubbellaags filter werd hiervoor bedreven als denitrificerend vlokkingsfilter gevolgd door twee enkellaags labschaal kolommen, waarvan de ene kolom gevuld was met fijn zand (0,5 – 0,8 mm korrelgrootte en 30 cm hoogte) en de andere met actief korrelkool (1,6 mm korrelgrootte en 1,2 m hoogte). De filtraten van beide kolommen lieten een verdergaande verwijdering zien van totaal fosfaat en nitraat stikstof en een afname van troebelheid en het chemisch zuurstofverbruik.

Naast de verwijdering van zwevende stof en nutriënten is ook aandacht besteed aan de verwijdering van enkele zware metalen en andere stoffen. De verwijdering van drie geselecteerde zware metalen, koper, zink en nikkel is onderzocht tijdens verschillende filtratietesten. Denitrificerende vlokkingsfiltratie resulteerde daarbij in verwijderingsrendementen van >92% voor koper, 60% voor zink en 10-20% voor nikkel. De verwijdering van andere stoffen, zoals enkele polycyclische aromatische koolwaterstoffen en hormoonverstorende stoffen, was laag. Echter, de verwijdering van zware metalen en andere stoffen wordt uitsluitend toegeschreven aan de filtratie van filtreerbaar materiaal dat vooraf complexen heeft gevormd met deze stoffen.

De bekerglasproeven met actief poederkool en de filtratietesten met het nageschakelde actief korrelkoolfilter gaven een hoog verwijderingsrendement aan voor de drie bovengenoemde zware metalen. Adsorptie wordt hierbij als belangrijkste verwijderingsmechanisme gezien, waarbij een indirecte verwijdering aangetoond werd door middel van complexvorming met organisch materiaal. De verwijdering van andere microverontreinigingen met dubbellaagsfiltratie in combinatie met actieve kooladsorptie is binnen dit onderzoek niet onderzocht. Er zijn voldoende aanwijzingen in de bestaande literatuur dat de verwijdering van enkele microverontreinigingen mogelijk is. De integratie van het adsorptiemechanisme in het filtratieproces gecombineerd met de verwijdering van zwevende stof en nutriënten kan daarom leiden tot een meerwaarde van het filtratieproces toegepast voor meerdere te zuiveren stoffen.

Het deelonderzoek aangaande het aspect deeltjestellingen was gericht op het bepalen van een meerwaarde van dit additionele meetinstrument. Hierbij is ook gekeken naar de additionele combinatie hiervan met meer conventionele analyses, zoals testen naar gebonden fosfaat en de bepaling van troebelheid. Verdunning van watermonsters was zeer belangrijk bij de nauwkeurigheid en betrouwbaarheid van het tellen van kleine deeltjes. Verder werd het deeltjesvolume als meer betrouwbare parameter ingeschat dan het deeltjesaantal.

Deeltjesgrootte- en volumeverdelingen, en de gemiddelde deeltjesgrootte zijn bepaald voor de beoordeling van de veranderingen in deeltjesverdelingen tijden coagulatie, flocculatie en filtratie. De toename van het totale deeltjesaantal en volume na voltooide coagulatie en flocculatie is vastgesteld, waaruit blijkt dat de toename in deeltjesgrootte na flocculatie lager was dan verwacht. Waarschijnlijk ontstonden tijdens de vlokkingsfiltratie proeven merendeels kleine deeltjes, wat toegeschreven kan worden aan “sweep-coagulatie” als een van de twee belangrijkste coagulatiemechanismen. De gevormde kleine deeltjes lijken geen negatief effect te hebben op de verwijdering van zwevende stof en totaal fosfor. Directe verbanden tussen deeltjes, troebelheid en gebonden fosfaat waren inconsistent. Het totale deeltjesaantal was gerelateerd aan troebelheidsmetingen waarbij een goede correlatie is gevonden voor de verwijderingsrendementen van gebonden fosfaat en het totale deeltjesvolume voor blanco filtratie en vlokkingsfiltratie (met polyaluminiumchloride). Deze correlatie lijkt echter te verzwakken voor vlokkingsfiltratie vergeleken met de blanco filtratie zonder voorafgaande metaalzoutdosering. Deze bevinding kan gekoppeld worden aan de fosfaatverdeling afkomstig van de fractioneringsproeven. De gebonden fosfaatfractie 0,45 - 2 μm die het meest problematisch is voor filtratie wordt bevorderd door het toevoegen van metaalzout. Niettemin kan deze niet gemeten worden met de toegepaste deeltjesteller vanwege het meetbare minimum van 2 μm. De toepassing van een deeltjesteller met een lagere meetrange kan leiden tot een nauwkeurigere relatie tussen deeltjes en gebonden fosfaat.

Er is geconcludeerd dat de verwijdering van zwevende stof, fosfor en stikstof gecombineerd kan worden in slechts één conventionele filtratiestap met geoptimaliseerde bedrijfsvoering. Hierbij is aangetoond dat een langdurige en stabiele filtratiewerking mogelijk is. Deeltjestellingen en fosfaat fractioneringen zijn daarbij nog niet volledig ontwikkeld als onafhankelijke parameter voor het monitoren of procescontrole maar zouden een beter inzicht kunnen geven in het filtratieproces waarbij de verwijdering van kleine deeltjes en fosfaatfracties zeer belangrijk wordt.

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2008

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Curriculum Vitae

Identification

Name	Miska-Markusch
First name	Viviane
Date of birth	31 August 1974
Place of birth	Hildesheim, Germany

Education

1987 – 1995	Higher Secondary Education at ‘Gymnasium Langenhagen’, in Germany.
1995 – 2002	Civil Engineering at ‘Leipniz University of Hanover’, master degree in civil engineering, in Germany.

Work experience

2003 – 2006	EU-project AQUAREC – Integrated concepts for water reuse
2003 – 2007	PhD student at the Delft University of Technology, faculty of Civil Engineering and Geoscience, department of Sanitary Engineering under supervision of prof. ir. J.H.J.M. van der Graaf.
2007 – recent	Water treatment specialist at DHV B.V. in Amerfoort


STELLING 746:
DE MEESTE AIO'S DOEN VEEL LANGER.....
ZUCHT
STEUN
DAN VIER JAAR OVER HUN ONDERZOEK
STEUN
ZUCHT
DEZE VERTRAGING IS INHERENT AAN......
ZUCHT
.....HET VERSCHRIKKELIJKE EINDE DAT HUN WACHT!
ZUCHT
GM

Propositions

Pertaining to the dissertation

"Effluent filtration for more than particle removal"

Viviane Miska-Markusch, Delft, 7 May 2009

1. Conventional multimedia filtration transformed into a new concept for suspended solids, phosphorus and nitrogen removal to very low levels (P_{tot}<0.15 mg/L and N_{tot}<2.2 mg/L) will play an important role in order to meet future water quality standards. (*this dissertation*)
2. Well-known strategies for filtration and modeling using the traditional discrete-particle-approach have to be reconsidered, optimized and verified by practical multi purposes filtration experiments. (*this dissertation*)
3. The optimisation of operational parameters, such as chemical dosing conditions and backwash procedures are the key to ensure a stable long-term filtration performance. (*this dissertation*)
4. Particle analysis demonstrates additional value to conventional analyses and visualising particle distribution changes during filtration but phosphate fractionations led the attention towards even smaller particles. Therefore, particle analysis is not yet established as independent parameter for monitoring and process control. (*this dissertation*)
5. Worldwide privatization of water and wastewater companies may lead to a water crisis in the 21st century even after we have overcome the actual financial crisis.
6. It is within realm of possibilities that in due time a team of highly trained and properly equipped scientific men may be able to analyze the water characteristics, the filtering media characteristics and determine with mathematical exactness the results which may be obtained under a given set of conditions or for any number of different sets of conditions (*Stanley, 1973*). This is a clear explanation for the fact that filtration experiments will always be site and time specific and even well established knowledge needs to be optimized and polished all over again.
7. When filter grains are visualised as pink balls and particles are described as perfectly round shaped suspended objects it is possible to explain the filtration research to non-technical people as my relatives.
8. Even for a knowledge emigrant learning the language is a prerequisite for integration. (*own experience*)
9. Research and TaiChi can have repelling approaches.
10. "Ich habe fertig!" translated to "I have ready!" (*Giovanni Trapattoni, former Italian coach of German team FC Bayern München*) is a citation with a high Johan Cruijff content.

These propositions are considered opposable and defendable and as such have been approved by the supervisor Prof. ir. J.H.J.M. van der Graaf.

Stellingen

Behorende bij het proefschrift

“Effluent filtration for more than particle removal”

Viviane Miska-Markusch, Delft, 7 Mei 2009

1. Conventionele multimedia filtratie getransformeerd in een nieuw concept voor verwijdering van zwevende stof, fosfor en stikstof tot lage waardes ($P_{tot}<0{,}15$ mg/L and $N_{tot}<2{,}2$ mg/L) zal een belangrijke rol spelen in het behalen van toekomstige waterkwaliteitseisen. (*dit proefschrift*)
2. Bekende strategieën voor filtratie en modellering gebaseerd op discrete deeltjes moeten heroverwogen, geoptimaliseerd en geverifieerd worden door middel van praktijkonderzoek, vooral als filtratie wordt toegepast voor meerdere doeleinden. (*dit proefschrift*)
3. De optimalisatie van bedrijfsvoeringstechnische instellingen, zoals chemische doseringen en terugspoelprocedures, zijn de sleutel om een langdurig en stabiel filtratieproces zeker te stellen. (*dit proefschrift*)
4. Deeltjestellingen hebben een toegevoegde waarde bij de ondersteuning van resultaten met conventionele analyses en visualiseren van veranderingen in deeltjesverdelingen tijdens filtratie, maar fosfaatfractioneringen hebben tot aandacht voor kleinere deeltjes geleid. Daarom hebben deeltjestellingen zich nog niet bewezen als onafhankelijk parameter voor het monitoren en procescontrole. (*dit proefschrift*)
5. Wereldwijde privatisering van bedrijven voor water- en afvalwaterbehandeling zal leiden tot een watercrisis in de 21^de^ eeuw, zelfs nadat wij de actuele financiële crisis achter ons hebben gelaten.
6. Het is in het bereik van mogelijkheden dat binnen zekere tijd een groep van hoog opgeleide en goed uitgeruste wetenschappers in staat is om de karakteristieken van het water en filtratiemedium te analyseren en de resultaten met wiskundige precisie te bepalen welke verkregen zijn onder een gegeven set van omstandigheden of een aantal sets van verschillende omstandigheden (*Stanley, 1973*). Dit maakt duidelijk waarom filtratieonderzoek altijd locatie- en tijdsafhankelijk is en al goed ontwikkelde kennis opnieuw geoptimaliseerd en verfijnd moet worden.
7. Indien filtermateriaal als roze balletjes en deeltjes als perfecte zwevende rondjes worden voorgesteld is het mogelijk het filtratieonderzoek begrijpbaar te maken aan niet-technische mensen zoals mijn familie.
8. Zelfs voor kennisemigranten is het leren van de taal een voorwaarde voor integratie. (*eigen ervaring*)
9. De benaderingen van onderzoek en TaiChi kunnen elkaar tegenspreken.
10. “ Ich habe fertig!” vertaald naar “ Ik heb klaar!” (*Giovanni Trapattoni, vroegere Italiaanse coach van het Duitse voetbalteam FC Bayern München*) is een citaat met een hoog Johan Cruijff-gehalte.

Deze stellingen worden opponeerbaar en verdedigbaar geacht en zijn als zodanig goedgekeurd door de promoter Prof. ir. J.H.J.M. van der Graaf.