The applicability of the SUR measurement for ultrafiltration of WWTP effluent

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The applicability of the SUR measurement for ultrafiltration of WWTP effluent

Proefschrift

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aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus prof. ir. K.C.A.M. Luyben,
voorzitter van het College voor Promoties,
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landbouwkundig ingenieur
geboren te Lisse.

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Voorwoord

Het is zover, mijn proefschrift is af. Vooral het laatste jaar heb ik vaak moeten zeggen dat het bijna af was, maar nu kan ik zeggen dat het écht afgerond is! Het voelt als op de top staan van een col na een stevige beklimming. Blij dat ik het heb bereikt, wetend dat het niet vanzelf ging. Juist dit maakt het schrijven van een proefschrift zo leuk. Zelf verantwoordelijk zijn voor de input, maar het zou nooit gelukt zijn zonder de support van velen. Daarom nu een goed moment om iedereen te bedanken, want zonder de steun van hen die in dit voorwoord vermeld staan, zou het nooit zover gekomen zijn. Mocht iemands naam niet genoemd zijn in dit proefschrift, ook jij bedankt!

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Zoals al eerder gezegd, onderzoek doe je niet alleen. Gelukkig maar! Tijdens mijn onderzoek heb ik veel plezier gehad tijdens het samenwerken met verschillende studenten. Christiaan van Assendelft,

Arthur Geilvoet, Arno Kooistra, Jaap Smit, Marie Thomas, Miguel Tavares, Jorg Trampé en Menno van der Zanden. Eén voor één hebben jullie een belangrijke bijdrage geleverd aan de totstandkoming van mijn proefschrift. Hopelijk zijn jullie net als ik door het onderzoek enthousiast geworden, en gebleven, over het mooie vakgebied van afvalwaterbehandeling.

In de periode dat ik werkzaam was bij de sectie Gezondheidstechniek heb ik de afdeling zien groeien en bloeien. Tijdens de gezellige koffiebreaks en de gezamenlijke presentaties heb ik niet alleen kennis gemaakt met afvalwater, maar ook met het schone drinkwater. Een speciale dank aan mijn collega promovendi van afvalwaterbehandeling. De MBR-adapten Adrien, Elif, Herman, Maria, Stefan en Pawel, de effluent nabehandelaars Viviane, Sandy en Sigrid, de rioleerders Cathelijne, Remy en Vincent en de echte pure influentman Aldo. Het delen van onderzoeksleed, persoonlijke reflecties en successen maakten elke dag mooi.

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Naast het werkzame leven is er gelukkig ook nog een andere kant van het leven. Een goed sociaal leven is voor mij heel belangrijk. Ik heb dat ook proberen te symboliseren door mijn paranimfen: Goos (broer) en Theo (vriend). Het doet mij goed dat jullie mij terzijde willen staan tijdens de verdediging van mijn proefschrift. Ook een woord van dank aan Niels en Leander. Onze dagelijkse e-mailwisseling, samen met Theo, moeten we voortzetten tot we niet meer kunnen. Dank ook aan de andere vrienden die betrokkenheid toonden door belangstelling te geven, maar ook gezelligheid, afleiding, vriendschap en wat nog meer. Tja, het poep roeren, is kennelijk zo interessant dat je er een boekje over kan schrijven.

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Tenslotte, wetend dat het begin en einde in Hem is, wil ik God danken voor de kracht en gezondheid die ik gekregen heb om dit werk voltooien.

Leiderdorp, 17 januari 2011
Arie Janssen

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1 Introduction

1.1 Development of wastewater treatment

There have been a lot of changes in wastewater treatment in the Netherlands since the mid-20th century. In the period 1960 – 1970 governmental and public awareness led to the introduction of the first part of Dutch environmental legislation, the Pollution of Surface Water Act (Wet Verontreiniging Oppervlaktewater, WVO). Here the focus was on the removal of oxygen consuming substances (ammonia and biodegradable organics). Later in the 80s – 90s the objective changed to the removal of nutrients (nitrogen and phosphorus) in order to decrease eutrophication of receiving water bodies. A modern wastewater treatment plant (WWTP) is now able to remove most nutrients up to the parts per million (ppm) level.

Nowadays, new treatment methods and developments to remove heavy metals and organic micro pollutants from WWTP effluent are under investigation. Recently, studies have shown environmental effects linked to the discharge of organic micro pollutants, such as feminisation of male fish and deformation of aquatic animals (Routledge, *et al*., 1998; Jobling, *et al.,* 2002). In an attempt to counter the unwanted presence of organic micro pollutants in the surface water, the European Water Framework Directive (WFD) was adopted in the year 2000. The WFD aims at a good ecological and biological status for all surface waters, coastal waters, transitional waters and groundwater in Europe by 2015. In total 33 components (nutrients, biological parameters, pesticides, heavy metals, hormone disrupters and medicinal substances) are listed in the WFD as priority compounds. Most likely, it will lead to stricter discharge limits for those 33 priority compounds at WWTPs. In addition to those 33 priority substances, new discharge limits will also be established for 'relevant area-specific' substances (STOWA, 2005).

In addition to the stricter standards the WFD encourages the integration of water reuse[1] options in an integrated approach of water resource management (Bixio and Wintgens, 2006). In this approach WWTP effluent is considered as a water source rather than a waste stream to augment water supply and to decrease the impact of human activities on the environment. In principle, WWTP effluent can be used for any purpose as long as adequate treatment is provided to meet the water quality for the intended use (Asano, 2002). For adequate treatment of WWTP effluent more advanced treatment methods are needed after the biological treatment. Therefore, nowadays the focus in wastewater treatment is changed to advanced treatment techniques to reclaim water from WWTP effluent and to meet future standards. In Table 1.1 an overview of advanced treatment techniques is given according to STOWA (2005).

[1] In this thesis *water reuse* refers to the use of treated wastewater for beneficial purposes such as agricultural irrigation and industrial cooling, while *wastewater reclamation* refers to the treatment or processing of wastewater to make it reusable. *Reclaimed water* is a treated effluent suitable for an intended water reuse application.

Table 1.1 – Overview of advanced techniques to treat WWTP effluent

Technique	Description
Biological techniques	
Membrane bioreactor (MBR)	An activated sludge system in which the sludge/water separation step takes place via membrane filtration instead of secondary clarifiers.
Advanced nitrogen removal	Removal of nitrogen molecules by nitrification and/or denitrification biomass in the form of a biofilm.
Advanced removal of organic contaminants	Biological removal of organic components by (1) physical/chemical bounding to biomass, (2) active uptake of dissolved organics by biomass and (3) degradation in the cells of biomass.
Pond treatment systems	Man made copies of natural wetland systems to exploit the treating processes (filtration, predation, biological removal nutrients, etc.) that occur in these systems.
Oxidative techniques	
Oxidisation processes	Process to oxidise (complex) organic compounds with the aid of strong oxidants like ozone (O_3) and hydrogen peroxide (H_2O_2).
Advanced oxidisation processes (AOP)	Combination of oxidation processes ($O_3 – H_2O_2$, O_3 – UV and UV – H_2O_2) that accelerate the oxidation reaction.
Chemical precipitation techniques	
Precipitation	The settling of ionic contaminants from a solution by the addition of chemicals.
Coagulation/Flocculation	The agglomeration of small particles to large flocs, caused by the use of coagulants such as ferric and aluminium chloride.
Coagulation/Flocculation in combination with suspended solids removal	The combination of coagulation, floc-formation and filtration (sand, membrane, etc.) to remove suspended particles and colloidal matter.
Adsorption techniques	
Activated carbon	The bonding of non-polar organic compounds due to Van der Waals forces.
Ion exchange	Bonding of ions to specific charged groups on the surface of a synthetic resin.
Bed filtration	The separation of particles from the water phase, while water flows through the pores in between the filter bed granules. The particles are removed by sieving, adsorption to the filter medium and settling onto the medium.
Screen- and membrane filtration	This type of filtration works by passing water through a filter of a defined pore size. In section 1.3 a more detailed description of this technique is given.
Disinfection	Destruction of micro organisms and the removal of viruses by filtration, physical disinfection (UV) and chemical disinfection (chlorine, ozone and AOP).
Integrated treatment techniques	Techniques that are added to or combined with conventional activated sludge systems like MBR, dosage of powdered activated carbon (PAC) in activated sludge process, etc..

1.2 Reclaiming water from WWTP effluent

Over the past two decades, the amount of municipal wastewater recovered has increased throughout the world (Levine and Asano, 2004). In 2004, Bixio *et al.* reported that worldwide over 3000 (municipal) wastewater reclamation sites existed. Especially in regions of the world faced with water scarcity, WWTP effluent is seen as a potential source for water reclamation. Some examples of water reclamation projects in the world are:

- The Water Factory 21 in Orange County, California (USA) where reclaimed water is recharged into the aquifer in order to stop salt intrusion. The first facility in operation since 1976 consisted of a series of treatment steps: flocculation, multi-media filtration, activated carbon adsorption, reverse osmosis and chlorination. The new facility is called Advanced Water Treatment (AWT) and has been commissioned in 2004. The AWT is designed to produce 325,000 m^3/d of microfiltration filtrate, among which 265,000 m^3/d will be further reclaimed with reverse osmosis. The system may even be expanded in the future to 491,000 m^3/d of product water (Bixio and Wintgens, 2006).
- The WWTP Baix Llobregat in Spain where wastewater is collected and treated from the southern part of Barcelona. Two reclaimed water flows with different qualities are produced at this plant. One for ecological flow, agricultural irrigation and wetlands and a second with a better quality for a barrier against seawater intrusion. The process scheme of the first flow is; regulation basin – flocculation/coagulation – filtration – ultraviolet – disinfection. The water to be used for the barrier against intrusion is additionally treated by microfiltration and reverse osmosis to obtain the required quality (Cazzura, 2008).
- The indirect potable use of WWTP effluent is applied in Wulpen (Belgium). Per year 2,500,000 m^3 WWTP effluent is infiltrated into the dunes after ultrafiltration, reverse osmosis and ultraviolet radiation. The purpose of infiltrating WWTP effluent is to reduce the extraction of natural groundwater for potable water production and hold back the saline intrusion at the coast. After a residence time of one to two months the water is recaptured and used for the production of drinking water (van Houtte and Verbauwhede, 2003).
- In Singapore the NEWater Project was implemented to supply industries and augment freshwater resources with reclaimed water. At the moment 4 water reclamation plants are in operation with a total capacity of about 95,000 m^3/d. The treatment technologies of the water reclamation plants differ, but the core of all the processes are membranes.
- The US $2.0 billion Western Corridor Recycled Water Project (WCRWP) is part of a capital works program of the Queensland Government to secure the future water supply for the region. Treated effluent is collected from WWTPs and further treated at three advanced water treatment plants incorporating microfiltration, reverse osmosis, advanced oxidisation and residual disinfection. The project will supply up to 182,000 m^3/day of purified recycled water for industrial and potable purposes (Traves *et al*., 2008).
- The only direct potable reuse project is operated in Windhoek (Namibia), one of the driest regions in Southern Africa. About 21,000 m^3/day of water (mixture WWTP effluent and

surface water) is produced per day by a complex treatment chain of coagulation, dual media filtration, ozonation, multi-stage activated carbon adsorption and ultrafiltration prior to chlorine disinfection (Menge *et al*., 2007).

- The DECO plant of Evides Industriewater (supplier of industrial water), in the southwest of the Netherlands (Zeeuws-Vlaanderen). This plant has been in operation since the year 2000 and produces an aggregate of 750 m^3/h demineralised water, 750 m^3/h cooling tower supply water and 1200 m^3/h ultra-pure water for the Dow Chemical Company. Part of the demineralised water is produced from effluent of a local communal WWTP in the vicinity of the plant. The applied technologies to upgrade the WWTP effluent to demineralised water are microfiltration followed by reverse osmosis; the design capacity is 2.2 million m^3/year (van Agtmaal *et al*., 2007).

Considering the given examples it is clear that wastewater can be reused for various purposes including a whole range of less advanced to more highly advanced purposes. The primary incentives for implementing water reuse were augmentation of water supplies and/or pollution abatement (Levine and Asano, 2004). Nowadays on an international scale, direct non-potable water reuse is currently the dominant mode for agricultural irrigation, industrial cooling water, river flow augmentation and other applications. In Table 1.2 a summary of the various possible applications from a global perspective is given.

Table 1.2 – Categories and description of water reuse applications from a global perspective according to Asano (2002)

Category	Description
Agricultural irrigation	Use of reclaimed water restricted to agricultural purposes and worldwide the largest current use.
Landscape irrigation	The second largest user of reclaimed water in industrialised countries and it includes the irrigation of parks, playgrounds, golf courses, etc.
Industrial reuse	The third major use of reclaimed water for a diversity of industries including power plants, pulp and paper and other industries with high rates of water utilisation.
Groundwater recharge	Include groundwater replenishment by assimilation and storage of reclaimed water in groundwater aquifers and the establishment of hydraulic barriers against saltwater intrusion in coastal areas.
Environmental and recreational uses	Involve non-potable uses related to land based water features such as the development of recreational lakes, marsh enhancement and stream flow augmentation.
Non-potable urban uses	Use of reclaimed water for fire protection, air conditioning, toilet flushing, construction water, flushing sanitary sewers, etc.
Indirect or direct potable reuse	Potable reuse occurs either by blending in water supply storage reservoirs or by the direct input of reclaimed water into the water distribution system.

1.3 Membrane filtration

1.3.1 Ultrafiltration membranes

Membrane filtration is a treatment process based on the physical separation of compounds from the water phase with the use of a semi-permeable barrier (membrane). Membranes for water treatment are pressure driven and can be divided into two categories based on their sieving mechanism (Mulder, 1996): porous and dense membranes. Porous membranes processes are microfiltration and ultrafiltration and their separation mechanism is sieving achieved by the pore size. The distinction between microfiltration and ultrafiltration is based on the size of the pores. Microfiltration features pore sizes of 100 nanometer (nm) up to a few micrometers (μm) while the ultrafiltration membranes feature smaller pores, from 1 – 100 nm. At lower pore size of the ultrafiltration membranes the charge of the membranes might be active as a separation mechanism as well. Nanofiltration and reverse osmosis are dense membranes that are able to retain dissolved salts and solids from water. By nanofiltration low molecular weight components and divalent ions (Ca^{2+}, Mg^{2+}, SO_4^{2-}, CO_3^{2-}, etc.) can be separated and furthermore monovalent ions (Na^+, K^+, Cl^-, NO_3^-, etc.) can be rejected by reverse osmosis. The driving force for porous and dense membrane processes in water

treatment is usually a hydraulic trans membrane pressure (TMP). It is obvious that the operating TMP of dense membranes is much higher than that of porous membranes. Table 1.3 outlines the typical pore sizes, required operational TMP and removable components of the different membrane processes for water treatment. It is mentioned that the pore size range related to each of the membrane processes is not fixed: slight variations can be found in the literature.

Table 1.3 – Classification of membrane processes with the pore size, pressure and removable components for water treatment (Mulder, 1996)

Membrane process	Pore size (nm)	Pressure (bar)	Removable component
Microfiltration	100 – 1000	0.1 – 2	Suspended solids, bacteria
Ultrafiltration	10 – 100	0.1 – 2	Macromolecules, viruses, proteins
Nanofiltration	1 – 10	4 – 20	Micropollutants, bivalent ions
Reverse osmosis	0.1 – 10	10 – 30	Monovalent ions, hardness

Many WWTP effluent reclamation plants rely heavily on membranes. Bixio and Wintgens (2006) state that the combination of microfiltration/ultrafiltration and reverse osmosis is the most applied scheme for the production of high quality water. Reverse osmosis itself has been used for desalination of WWTP effluent already since 1970 but the combination of microfiltration/ultrafiltration and reverse osmosis (double membrane system) has been developed recently. Initially pretreatment of reverse osmosis had been based on technologies like flocculation, lime clarification, recarbonation, settling and filtration. Compared to these pretreatment technologies microfiltration and ultrafiltration are superior in removing suspended solids, bacteria, large macro-molecules and pathogenic organisms, in order to provide a high quality feedwater to the reverse osmosis system. Although microfiltration and ultrafiltration are operated under similar process conditions, they differ in pore size characteristics (see Table 1.3). Ultrafiltration membranes are able to reject viruses completely where microfiltration does not. Also in the combination with reverse osmosis membrane differences are found. Kim *et al.*, (2002) present lower operating pressures and longer cleaning intervals when reverse osmosis membranes are fed with ultrafiltration filtrate instead of microfiltration filtrate.

The research described in this thesis focuses on the ultrafiltration membranes because of their good references. The ultrafiltration membranes have a proven capability to produce a stable and good filtrate quality in terms of particles and microbial parameters, regardless of the feedwater quality. Beside this the ultrafiltration membranes are able to operate at relatively high flux, low TMP and low energy costs, applying dead-end mode (te Poele, 2005).

1.3.2 Membrane process

In a membrane process three different streams can be defined (Figure 1.1). First the feed stream containing the constituents that have to be removed. The feed stream is split into two streams by the membrane; a clean water stream (the permeate) and the stream containing the

retained constituents (the concentrate). The separation is mostly forced by pressure in water treatment, the so-called (hydraulic) trans membrane pressure. Depending on the height of the TMP and the filtration resistance water will flow from the feed side through the membrane to the permeate side with a certain flow rate called the flux (J).

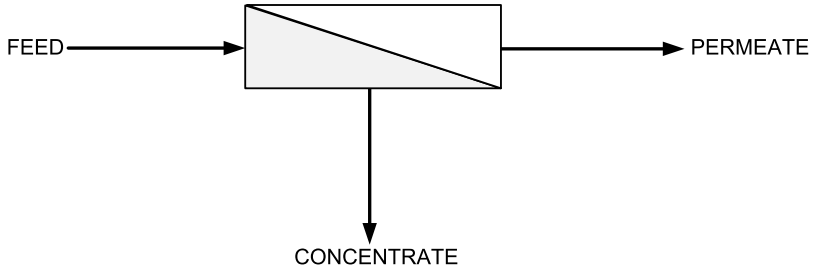


Figure 1.1 – Schematic representation of a membrane process

The membrane process can be designed in different ways. Generally three main aspects should be considered during the design of the membrane process: the membrane material, membrane configuration and mode of operation. These aspects will be described and discussed in the following section.

1.3.3 Membrane material

Membranes are made of various organic and inorganic materials. The most commonly applied in water treatment are the organic membranes with a wide variety of membrane materials, pore size, pore size distributions, membrane configurations and production processes. Compared to the organic membranes, inorganic membranes have superior mechanical strength and resistance to chemicals and temperature but the manufacturing costs are much higher. Although the prices of ceramic membranes are decreasing and therefore they are becoming more and more attractive (Baker, 2004) and will compete with organic membranes.

Formerly the organic membranes were made of cellulose, a natural material but nowadays most of the ultrafiltration membranes are mainly made of polymers with hydrophilic properties like polysulfone, polyethersulfone or polyvinylidene fluoride (Meier *et al*., 2006). In Table 1.4 the different membrane materials are summarized. The membrane structure can be isotropic or anisotropic. Whereas isotropic membranes have a uniform composition and structure, anisotropic (or asymmetric) membranes consist of different layers, prepared by a phase inversion process, each with different structures and permeability (Baker, 2004).

Table 1.4 – Different membrane materials (Mulder, 1996)

Organic		Inorganic	
Cellulose acetate	(CA)	*Ceramic*	
Polyetherimide	(Ultem)	Titanium Oxide	(TiO_2)
Polyacrylonitrile	(PAN)	Zircon Oxide	(ZrO_2)
Polysulphone	(PSU)	*Metallic*	
Polyethersulphone	(PES)	Aluminium Oxide	y-Al_2O_3
Teflon			
Polyvinylidenefluoride	(PVDF)		
Polyethylene	(PE)		

1.3.4 Configuration

The configuration of membranes generally differs in two ways based on their geometry: as flat sheet or tubular. The application of one of the configurations mainly depends on the feedwater characteristics. An ideal configuration would provide a high membrane area per volume area (packing density), high turbulence, low energy use, easy cleaning and operation. However, some of these features are conflicting and therefore result in a number of configurations in terms of membranes and membrane module. The two geometries and different features form the basis for four principle types of membrane modules used for wastewater treatment (Aptel and Buckley, 1996; Mulder, 1996):

- Tubular membranes; having an internal diameter larger than 3 mm and packing density of < 300 m^2/m^3, which are bundled in a module.
- Hollow fibre or capillary membranes; having an internal diameter less than 3 mm, which are bundled into a membrane module with some hundred or thousand fibres. The difference between hollow fibre and capillary membranes lies in the packing density, which is about 600 – 1200 m^2/m^3 and values of 30,000 m^2/m^3 respectively (te Poele, 2005).
- Plate or frame membranes; comprised of a series of flat sheet membranes and support layers and having a packing density of 100 – 400 m^2/m^3.
- Spiral wound membranes; flat membranes wound around a spacer, having a packing density of 300 – 1000 m^2/m^3 and used for nanofiltration and reverse osmosis.

In tubular and hollow fibre membranes the flow direction of feedwater can be inside-out or outside-in. During outside-in filtration permeate is collected inside the membrane and during inside-out it is collected outside the membrane fibre. This is illustrated in Figure 1.2.

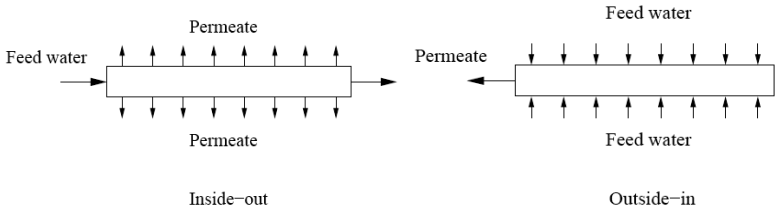


Figure 1.2 – Schematic representation of inside-out and outside-in filtration (te Poele, 2005)

1.3.5 Modes of operation

Membrane filtration can be operated basically in two modes: dead-end and cross-flow operation, illustrated in Figure 1.3. In the early days of membrane filtration with ultra- and microfiltration membranes, cross-flow filtration was the only applied mode of operation (van de Ven, 2008). In cross-flow filtration the feed flow is tangential to the membrane surface at a high speed. The aim of this high speed is to prevent the deposition of material of the feed solution on the membrane surface. During dead-end ultrafiltration all the deposits of the feed solution accumulate on the membrane surface and lead to an increase of pressure when the permeate flow is kept constant. Therefore, cross-flow filtration is more suitable for treating water with high solids content and higher permeate production can be achieved. However, the required velocity of cross-flow filtration leads to a high energy usage. To overcome the disadvantages of both modes of operations recently an intermediate mode of operation has been introduced: semi dead-end operation. In this mode the membrane is dead-end operated but accumulated deposits are frequently removed by hydraulic flushes. This type of operation is becoming an attractive filtration mode for WWTP effluent and is also applied during the research described in this thesis.

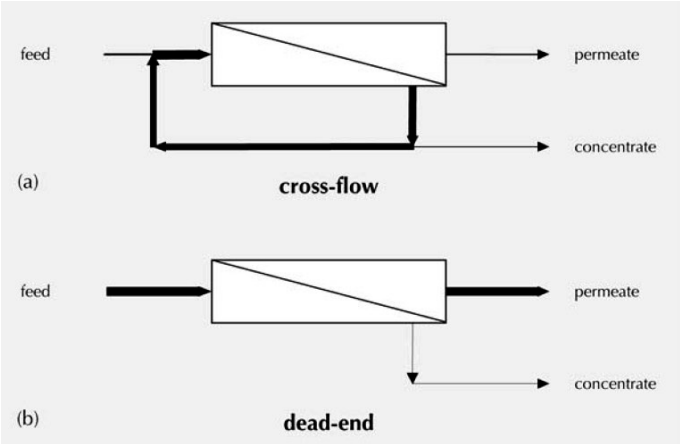


Figure 1.3 – Schematic representation of cross-flow filtration (a) and dead-end filtration (b) (Roorda, 2004)

In order to maintain stable operation during semi dead-end operation the membranes should be cleaned periodically. Cleaning can be performed either hydraulically, mechanically, chemically or by electrical cleaning. Hydraulic cleaning can be applied in two flow directions. A forward flush is a complete cross-flow cleaning method with high flow rates. With a backflush the flow rate is changed in the opposite direction, which means that backflush water is flowing from the permeate side to the feed side of the membrane and is discharged as concentrate.

After several filtration periods the performance declines, in spite of frequently applied hydraulic flushes. Chemical cleaning is needed to recover the performance of the membrane. This cleaning method can be applied by either starting with a forward or a back flush. Subsequently the membranes are soaked in chemicals for a certain period and finally the membranes are flushed with permeate, tap water or ultra pure water.

1.4 Ultrafiltration definitions

1.4.1 Flux and resistance

During ultrafiltration the TMP is the driving force for permeation. The permeate flow through the membrane is called flux (J) and is defined as the permeate volume through the membrane per unit of membrane area. The permeate flux is expressed as $m^3/m^2 \cdot s$ or more commonly $L/m^2 \cdot h$ and is given by the Equation 1.1 (Mulder, 1996).

$$J = \frac{dV}{dt} \cdot \frac{1}{A_{membrane}} \tag{1.1}$$

in which:
- J = flux ($m^3/m^2 \cdot s$), in this thesis expressed as ($L/m^2 \cdot h$)
- V = filtered volume (m^3)
- t = time (s)
- $A_{membrane}$ = membrane area (m^2)

The relation between the permeate flux and the trans membrane pressure under laminar conditions and through porous membranes can be described conveniently by an adaptation of the Darcy's law (Lojikine *et al*., 1992) and is introduced in Equation 1.2.

$$J = \frac{\Delta P}{\eta_p \cdot R_t} \tag{1.2}$$

in which: ΔP = trans membrane pressure (Pa), in this thesis expressed as (bar)
η_p = dynamic viscosity of the permeate (Pa·s)
R_t = total resistance to filtration resistance (m^{-1})

Equation 1.2 presents an inversely proportional relation between the permeate flux and the dynamic viscosity. In water and wastewater treatment it is usual to assume permeate viscosity equal to pure water (Manem and Sanderson, 1996) and therefore solely dependent on its temperature. In the literature several empirical relationships between temperature and pure water viscosity can be found; in this thesis the one as derived by Janssen and Warmoeskerken (1997) is used and shown in Equation 1.3.

$$\eta_p = 10^{-3} \cdot \exp^{0.580 - 2.520\theta + 0.909\theta^2 - 0.264\theta^3} \tag{1.3}$$

in which: θ = empirical factor: 3.6610·(T/(273.1+T)
T = temperature (°C)

When the object of filtration is not pure water (i.e. WWTP effluent) a contribution to the resistance may arise from the solute and other substances, which cause fouling (thoroughly described in chapter 2). Therefore the total filtration resistance (R_{total}) is often expressed as the sum of membrane resistance ($R_{membrane}$) and the additional resistance from fouling ($R_{fouling}$) as presented in Equation 1.4.

$$R_{total} = R_{membrane} + R_{fouling} \tag{1.4}$$

1.4.2 Filterability and reversibility

Dead-end ultrafiltration operation consists of subsequent filtration, backflush and chemical cleaning steps. The definitions of the various cycles are used throughout this thesis, as well as the definitions of *filterability* and *reversibility* used in this work are graphically given in Figure 1.4. Filterability is the increase of filtration resistance over time (or filtrated volume). A good filterability means that the increase of resistance is small. Reversibility is described as the extent to which the filtration resistance is returned to the original value after applying a hydraulic cleaning. If the filtration resistance after a backflush is equal to the filtration resistance at the start of the previous filtration period, than the fouling is considered to be completely reversible. Both terms, filterability and reversibility are related to the properties of the feedwater, membrane material and operational conditions. Therefore, the definitions are used in respect to these parameters. When the amount of irreversible fouling becomes too

high, or after a predetermined amount of backflush cycles, the membrane is chemically cleaned. Ideally, chemical cleaning removes all the irreversible fouling from the membrane.

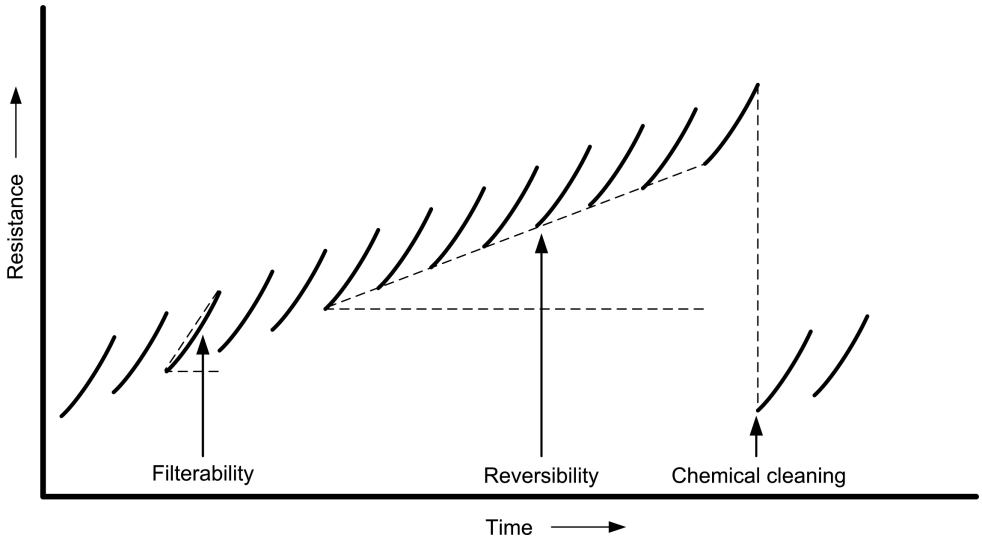


Figure 1.4 – Typical filtration curves illustrating filterability, reversibility and chemical cleaning

1.5 Background of this thesis

Since 1999 research at Delft University of Technology has been carried out on dead-end ultrafiltration of WWTP effluent. From 1999 – 2005 the research was performed in the framework of the project "Membrane filtration of effluent". The overall objective of the project was to develop filtration techniques for the large-scale distribution of reclaimed water, based on biologically treated effluent. Within the project the researchers focused on dead-end ultrafiltration. The aim of this part of the project was to gain more insight into the interaction between WWTP effluent and the ultrafiltration membranes in order to improve the design of ultrafiltration installations at lower costs.

The research at the Delft University of Technology has resulted until now in two dissertations. The first dissertation 'Filtration characteristics in dead-end ultrafiltration of WWTP effluent' published by Roorda (2004) deals with the filterability of WWTP effluent. In addition to pilot experiments at various WWTPs in the Netherlands the filterability of WWTP effluent was studied in detail on lab scale. These experiments resulted in a new parameter to describe the filtration characteristics of WWTP effluent during dead-end ultrafiltration, the Specific Ultrafiltration Resistance (SUR). With the SUR it is possible to measure even small differences in filtration characteristics of WWTP effluent (Roorda, 2004). The SUR was also used to measure the filterability of size fractions of the WWTP effluent. In chapter 3 the SUR equipment and measurement are described in detail.

The second dissertation deals with the physical and chemical mechanisms of membrane fouling during dead-end ultrafiltration of WWTP effluent (te Poele, 2005). To gain insight into this matter, photometric measurement methods are further developed in order to analyse potential membrane foulants in WWTP effluent. The analytical data of the foulants are compared with the actual filtration properties of WWTP effluent on lab and pilot scale. More details about the membrane foulants in WWP effluent were obtained by using different cleaning agents and methods in laboratory and pilot filtration tests. In chapter 2 the results of this dissertation are discussed in more detail.

In 2005 the research project was continued under the name "Ultrafiltration of WWTP effluent". This research focuses on the application of ultrafiltration of WWTP effluent for high quality industrial applications. Within the project two research topics were defined. The first topic deals with the improvement of filterability of WWTP effluent by pretreatment before ultrafiltration. Roorda (2004) stated earlier that an high initial filterability of WWTP effluent is required for stable ultrafiltration performance at fluxes as high as 100 $L/m^2 \cdot h$ and also showed that the filterability is dominated by particles in the range of 0.1 – 0.2 µm. Therefore for a high initial filterability of WWTP effluent particles in this particular size range should be removed or transformed during pretreatment for ultrafiltration. Pilot and lab scale experiments showed that with conventional pretreatment techniques (coagulation, sandfiltration and double layer filtration) only a moderate improvement of the initial filterability could be obtained. Therefore within this topic the application of alternative pretreatment technologies is investigated. The second topic deals with the SUR measurement and the operation of ultrafiltration installations. In this part the filterability of WWTP effluent measured as the SUR value and the performance of UF installations are evaluated. It is well known that the performance of UF installations depends on the filtration characteristics of the effluent. But this relation is not yet really quantified and therefore this part of the research focuses on the relation between SUR and process parameters like flux, trans membrane pressure and resistance increase.

1.6 Aim of this thesis

The research described in this thesis deals with the filterability and reversibility of WWTP effluent during dead-end ultrafiltration in order to optimise the process conditions and pretreatment technology of dead-end ultrafiltration. To gain insight into this matter, the following aspects are covered:

- Investigation of the relation between the SUR value of WWTP effluent and operating flux, trans membrane pressure and resistance.
- Identification of the effect of four pretreatment technologies: powdered activated carbon, granulated activated carbon filtration, dual media filtration and biological granulated

activated carbon filtration (1-STEP® filter). The effect is determined by fractionation, foulants and SUR measurements of untreated and pretreated WWTP effluent.

- Evaluation of the performance of a pretreatment technology (coagulation – dual media filter – coagulation) and full scale ultrafitration installation during the intake of WWTP effluent after buffering in a stabilization pond. For the evaluation SUR measurements and foulants analyses are performed.
- The effect of operational conditions flux and pretreatment on both fouling parameters, filterability and reversibility. To determine this effect a pilot ultrafiltration installation is used.
- Characterization of both filterability and reversibility of WWTP effluent with a revised version of the SUR equipment.

With the gained insight it should be possible to further implement or optimize the application of dead-end ultrafiltration of WWTP effluent at different locations and in different situations in order to increase the perspectives of water reuse for different applications.

1.7 Outline

For a general understanding of the terms and concepts in membrane filtration of WWTP effluent a concise introduction is presented in *Chapter 2 WWTP effluent and membrane filtration. Chapter 3 Research methodology* describes the material and methods used during the experiments. *Chapters 4 – 7*, the heart of the thesis, present the results of the experiments. *Chapter 4 Filtration properties and performance of ultrafiltration installations.*presents the results of tests at the WWTP Sas van Gent (full scale) and WWTP Horstermeer (pilot scale) to investigate the relation between the SUR value of WWTP effluent, flux, trans membrane pressure and resistance increase. *Chapter 5, Filtration properties and pretreatment*, focuses on the effect of powdered activated carbon, granulated activated carbon filtration, dual media filtration and biological granulated activated carbon filtration (1-STEP® filter) on the filterability of WWTP effluent. The next chapter, *Application of SUR in practice: A case study*, presents the added value of applying SUR measurements parallel to the operation of a full scale ultrafiltration – reverse osmosis installation. The effect of operation conditions on both filterability and reversibility are presented in *Chapter 7*. Finally in *Chapter 8* the results of the various experiments are evaluated and several recommendations for future research directions are proposed.

References

Agtmaal, J. van, Boks, P.A., Cornips, R. and Paping, L.L.M.J. (2007). *Evaluation of feed water sources and retrofitting of an Integrated Membrane System.* Proceedings 6th IWA Specialist Conference on Wastewater Reclamation an Reuse for Sustainability, October 9 – 12, 2007, Antwerp, Belgium.

Asano, T. (2002). *Water from (waste)water – the dependable water resource.* Water science and Technology, 45(8), 23 – 33.

Baker, R.W. (2004). *Membrane Technology and Applications*. 2nd Edition. John Wiley & Sons, Ltd, Chichester, United Kingdom.

Bixio, D. and Wintgens T. Editors (2006). *Water Reuse System Management Manual AQUAREC*. Project report European Comission, Luxembourg.

Cazurra, T. (2008). *Water reuse of south Barcelona's wastewater reclamation plant*. Desalination, 218, 43-51.

CBS (2006). *25 Jaar Statistiek Zuivering van afvalwater in vogelvlucht*. in Dutch, Voorburg/Heerlen, the Netherlands.

Houtte, E. van and Verbauwhede, J. (2003). *Drinkwater uit effluent via duinfiltratie (ET 16)*. Proceedings PAO Cursus 'Effluent van de toekomst', Delft, The Netherlands.

Jobling, S., Beresford, N., Nolan, M., Rodgers-Gray, T., Brighty, G.C., Sumpter, J.P. and Tyler, C.R. (2002) *Altered sexual maturation and gamete production in wild roach (Rutilus rutilus) living in rivers that receive treated sewage effluents.* Biology of reproduction, 66, 272 – 281.

Kim, S.L., Paul Chen, J. and Ting, Y.P. (2002). *Study on feed pretreatment for membrane filtration of secondary effluent.* Separation and Purification Technology, 29, 171 – 179.

Levine, A.D. and Asano T. (2004). *Recovering sustainable Water from Wastewater*. Environmental Science & Technology, June, 201 – 208.

Lojkine, M.H., Field, R.W. and Howell, J.A. (1992). *Crossflow microfiltration of cell suspensions: a review of models with emphasis on particle size effects.* Transition Inst. Chemical Engineering. 70. 149 – 164.

Manem, J. and Sanderson R. (1996). Membrane bioreactors. *Water Treatment Membrane Processes*. AWWARF/Lyonnaise des Eaux/WRC, McGraw-Hill, New York, USA.

Menge, J.G., Pisani, P. du and Merwe, B. van der. (2007). *Water quality control in a third world country: challenges to ensure good quality reclaimed in Windhoek, Namibia.* Proceedings 6th IWA Specialist Conference on Wastewater Reclamation an Reuse for Sustainability, October 9 – 12, 2007, Antwerp, Belgium.

Meier, P., Salehi, F., Kazner, C., Wintgens, T. and Melin T. (2006). *Ultrafiltration with pre-coagulation in drinking water production – Literature review.* Report Techneau project, report 06.

Mulder, M. (1996). *Basic principles of membrane technology*. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Routledge, E.J., Sheahan, D., Desbrow, C., Brighty, C., Waldock, M. and Sumpter, P. (1998). *Identification of estrogenic chemicals in STW effluent. 2. In vivo responses in trout and roach.* Environmental Science & Technology, 32, 1559 – 1565.

STOWA (2005). *Exploratory study for wastewater treatment techniques and the European Water Framework Directive*. Report 25, Utrecht, The Netherlands.

Traves, W.H., Gardner, E.A., Dennien, B. and Spiller, D. (2008). *Towards indirect potable reuse in South East Queensland*. Water Science & Technology, 58(1), 153 – 161.

Ven, W. van de (2008). *Towards optimal saving in membrane operation*. PhD thesis, Universtiteit Twente, Enschede, The Netherlands.

2 Wastewater treatment plant effluent and membrane filtration

2.1 Introduction

This chapter provides the background information for this thesis. In section 2.2 the municipal wastewater treatment process and the characteristics of WWTP effluent are described. Further in section 2.3 the fundamentals of membrane filtration for water treatment are addressed and in section 2.4 the main drawback of this process: fouling. The subsequent sections provide information about factors that affect this almost inevitable consequence of membrane filtration. The last section gives an overview of methods to indicate the fouling rate of WWTP effluent.

2.2 Wastewater treatment plant effluent

2.2.1 Treatment process

Nowadays in the Netherlands almost all the produced municipal and industrial wastewater (>98%) is treated in wastewater treatment plants (CBS, 2006). The municipal wastewater treatment plants (356 in 2007) with a total design capacity of 24,462,000 population equivalents (pe's) and a total volume of wastewater treated of approximately 2,100 Mm^3 in 2007 (CBS, 2009). In general almost all the current wastewater treatment plants in the Netherlands are based on the activated sludge process (Figure 2.1). Therefore the description of the treatment process in this section focuses on this process. The activated sludge process consists of three processes in series. The first step, the primary treatment, is usually mechanical aiming to remove coarse material, suspended solids and other undesired substances. Subsequently, in the second step, the "heart" of the process, the influent is mixed with biomass (activated sludge) and treated under aerobic and/or anoxic conditions. These conditions provide not only a removal of biodegradable COD but also a conversion of phosphorus and nitrogen. In the end the biologically treated water and the activated sludge are separated in a final clarifier using gravitational settling. The settled activated sludge is returned partly to the beginning of the biological process and the other part is wasted. In many cases the wasted sludge is treated on site together with the sludge of the primary step by digestion and dewatering. After dewatering it is usually transported to an incinerator.

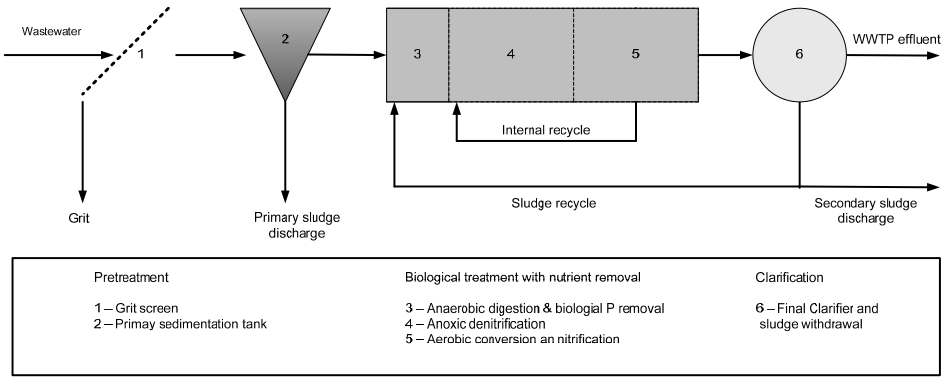


Figure 2.1 – Schematic flow scheme of an activated sludge process

2.2.2 Characteristics of WWTP effluent

The wastewater transported by the sewerage system and treated in a wastewater treatment plant consists of a mixture of municipal and industrial (pretreated) wastewater. Additionally, during storm weather events, the sewer is used for the transport of rainwater and is collected at the wastewater treatment plant. Considering these factors and the consequent variations in loads and concentrations of pollutants on different time and spatial scale an average influent quality for all wastewater treatment plants in the Netherlands is presented in Table 2.1.

Table 2.1 – Average quality of WWTP effluent in the Netherlands in 2007 and the current discharge standards

Parameter	Unit	Average concentration[1]	Discharge standards[2]
Chemical oxygen demand (COD)	mg O_2/L	43	125
Biochemical oxygen demand (BOD)	mg O_2/L	4	20 (without nitrification
Total nitrogen (N_{tot})	mg N/L	91	10 (> 20,000 pe) 15 (2,000 – 20,000 pe)
Total phosphorus (P_{tot})	mg P/L	1	1 (> 100,000 pe) 2 (2,000 – 100,000 pe)
Suspended solids (SS)	mg/L	11	30

[1)] Data 2007 (CBS, 2008)

[2)] VROM (1996)

Next to the constituents presented in Table 2.1 WWTP effluent contains a wide variety of trace compounds and elements, although they are not measured routinely. In Table 2.2 individual constituents are grouped into four broad categories: (1) the residual organic and inorganic colloidal and suspended solids, (2) dissolved organic constituents, (3) dissolved inorganic constituents and (4) biological constituents.

Table 2.2 – Typical residual constituents found in treated wastewater effluent and their impacts (Metcalf & Eddy, 2003)

Residual constituent	Effect
Inorganic and organic colloidal and suspended solids	
Suspended solids	- May cause sludge deposits or interfere with receiving water clarity - Can impact disinfection by shielding organisms
Colloidal solids	- May affect effluent turbidity
Organic matter (particulate)	- May shield bacteria during disinfection, may deplete oxygen resources
Dissolved organic matter	
Total organic carbon	- May deplete oxygen resources
Refractory organic	- Toxic to humans; carcinogenic
Volatile organic compounds	- Toxic to humans; carcinogenic; form photochemical oxidants
Pharmaceutical compounds	- Impact aquatic species (e.g., endocrine disruption, sex reversal)
Surfactants	- Cause foaming and may interfere with coagulation
Dissolved inorganic matter	
Ammonia	- Increases chlorine demand - Can be converted to nitrates and, in the process, can deplete oxygen resources - With phosphorus, can lead to the development of undesirable aquatic growths - Toxic to fish
Nitrate	- Stimulates algal and aquatic growth
Phosphorus	- Stimulates algal and aquatic growth - Interferes with coagulation - Interferes with lime-soda softening
Calcium and magnesium	- Increase hardness and total dissolved solids
Total dissolved solids	- Interfere with agricultural and industrial processes
Biological	
Bacteria	- May cause disease
Protozoan cyst and oocyst	- May cause disease
Viruses	- May cause disease

2.2.3 Particles in WWTP effluent

Particles in WWTP effluent are defined based on their size as dissolved, colloidal and suspended matter but there is not a sharp boundary in size that separates them. An overview of the different fractions, constituents and sizes of WWTP effluent is presented in Figure 2.2.

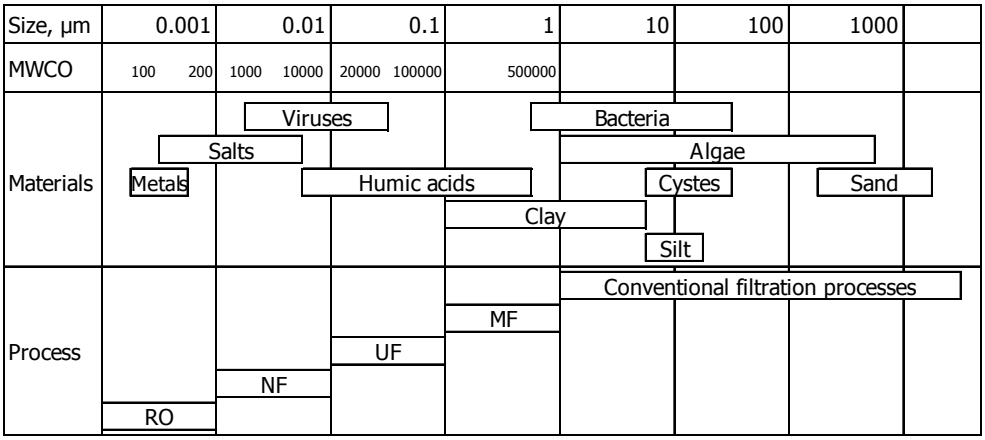


Figure 2.2 – Size of typical components in water and classification of membrane processes (adapted from van Dijk *et al.* 2001)

As written, there is not a sharp boundary between the particle sizes. Consequently there is some overlap between the same fractions. Van Nieuwenhuijzen (2002) classified wastewater constituents into different fractions: dissolved (< 0.1 µm), colloidal (0.45 µm – 2 µm), suspended (5 µm – 63 µm) and settable (> 63 µm). The fraction between colloidal and suspended was called supra colloidal and defined in the range 1.2 µm – 5 µm. However, other authors present different size ranges for the colloidal fraction. For example, Azema *et al.* (2002) and Levine *et al*. (1991) defined the colloidal fraction in the range of 0.001 µm – 1 µm and Metcalf and Eddy (2003) considered the colloidal fraction between 0.01 µm – 1 µm. Despite these different defined size ranges Adin (1999) reported that particles in WWTP effluent are mostly colloidal and negatively charged. This observation is confirmed by other researchers as well. For example, Abdessemed *et al.* (2002) showed that in WWTP effluent 58% of the organic load (COD) was found in a range larger than 0.1 µm, 13% of the COD was found within a range between 10 kDa and 0.1 µm and 29% was found in a fraction smaller than 10 kDa.

Also in this thesis to characterize WWTP effluent different fractions are distinguished. The classification of fractions is in line with earlier studies (Roorda, 2004; te Poele, 2005) and presented in Table 2.3.

Table 2.3 – Fractions distinguished in this thesis

Size (µm)	Fraction
> 0.45	Particles (suspended and settable material)
0.45 – 0.20	Colloids
0.20 – 0.10	Colloids
< 0.10	Dissolved material (macro molecules)

2.3 Membrane fouling

2.3.1 Definitions

As explained in chapter 1 membrane fouling is a natural consequence of the membrane separation process. The definitions of membrane fouling are several in literature and can be generally distinguished in two approaches. Some authors (van der Berg and Smolders, 1990 and Lojkine, 1992) use the term fouling to indicate strictly the 'tenacious' interactions that take place on the membrane and that would not be reversed by a release of the driving force (Ravazinni, 2008). The other approach includes within the term fouling each mechanism that leads to a decrease of performance during the filtration process. The approach, used in this thesis, is defined by the International Union for Pure and Applied Chemistry which defines fouling as *the process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores* (Koros *et al.*, 1996). Within this definition fouling is encountered in the two terms: filterability and reversibility. Filterability as the loss of performance e.g. during a filtration run and reversibility as the extent to which membrane performance can be regained after it was fouled during filtration.

2.3.2 Fouling mechanisms

In the membrane filtration process of WWTP effluent different fouling mechanisms may occur. Generally, five fouling mechanisms can be distinguished; each mechanism contributes to the total resistance over the membrane. The fouling mechanisms are schematically presented in Figure 2.3:

- *Concentration polarisation (R_{cp})*: The increased concentration of rejected solutes near the membrane surface resulting in e.g. scaling, raise of osmotic pressure and gel formation.
- *Pore blocking (R_{pb})*: Particles enter the membrane pores and get stuck in their openings, so that the number of pore channels available for permeation is reduced.
- *Pore narrowing, e.g. by adsorption (R_a)*: Particles, colloids and macro molecules that enter the membrane pores and adsorb to the pore wall, resulting in less open pore channels and decrease of the permeate flow.
- *Cake layer formation (R_c)*: Particles, colloids and macro molecules accumulate on the membrane surface forming a more or less dense and more or less permeable layer. The strength of the cake layer depends on the interaction between the substances themselves and interaction with the membrane.
- *Compression of cake layer (R_{cc})* (not shown in Figure 2.3): If the cake layer is compressible an increase of trans membrane pressure will compress the cake layer resulting in a higher resistance.

The type of fouling is highly dependent on the type of filtration process, i.e. microfiltration, ultrafiltration, nanofiltration and reverse osmosis. Concentration polarisation e.g. is of minor importance during dead-end ultrafiltration. The term originated from reverse osmosis applications and therefore left out of consideration in this thesis. Another fouling mechanism that is not incorporated in Figure 2.3 is the growth of micro organisms and their metabolic products of the membrane surface, so called biofouling. Once developed biofouling is very hard to remove as incomplete removal of (dead) micro organisms will spawn re-establishment of new biological activity (Schrader, 2006). Also in Figure 2.3 possible fouling mechanisms as compaction of membrane and capillary blocking are not included. Capillary blocking as fouling mechanism was established by Heijman *et al.* (2007). Heijman *et al.* (2007) investigated in a lab scale test whether there is heterogeneous fouling (capillary blocking) in dead-end ultrafiltration, the hydraulic cleaning with backflush and/or forward flush is not capable to remove all the cake fouling or pore blocking in the membrane module.

According to Figure 2.3 the total resistance (R_{total}) in equation 2.1 is based on the different fouling mechanisms and the initial membrane resistance, presented by a resistance-in-series relationship according to Bowen and Jenner (1995):

$$R_{total} = R_{membrane} + R_{cake} + R_{poreblocking} + R_{adsorption} \tag{2.1}$$

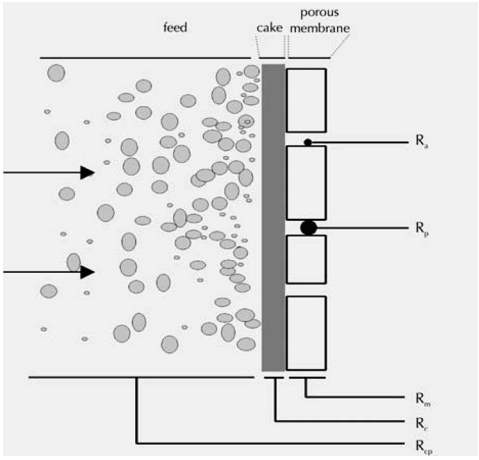


Figure 2.3 – The resistance of a fouled membrane by various fouling mechanisms, the driving force is from the left to the right: Ra = adsorption, Rp = pore blocking, Rm = initial membrane resistance, Rc = cake filtration, Rcp = concentration polarization (van den Berg, 1988)

2.3.3 Dynamics of the filter cake

In dead-end ultrafiltration of WWTP effluent cake filtration is assumed to be the predominant filtration mechanism for the increase of resistance (Roorda, 2004). Based on this assumption the total filtration resistance (R_{tot}) is the sum of membrane resistance (R_m) and cake resistance (R_c). Combining this assumption with the adapted Darcy's law (equation 1.2) the following relationship for the cake resistance can be derived (equation 2.2).

$$R_c = \frac{\Delta P}{\eta_t \cdot J} - R_m \tag{2.2}$$

in which: R_c = cake resistance (m^{-1})
ΔP = trans membrane pressure (Pa) or (bar)
η_t = feedwater dynamic viscosity (Pa·s)
J = flux ($m^3/m^2 \cdot s$)
R_m = membrane resistance (m^{-1})

The membrane resistance is considered as constant but the cake resistance increases due to the retention of particles within the cake layer. This is shown in equation 2.3. It assumes that material rejected by the membrane forms a cake layer, which causes cake resistance to increase proportionally to the amount of material delivered to the membrane surface.

$$R_c = \alpha_{av} \cdot c_v \cdot \frac{V}{A_m} \tag{2.3}$$

in which: α_{av} = average specific cake resistance (m/kg)
c_v = solids concentration in feedwater (kg/m^3)
V = permeate volume (m^3)
A_m = membrane area (m^2)

The compressibility of the formed cake layer during dead-end ultrafiltration of WWTP effluent was investigated by Roorda (2004) and Zheng, *et al*. (2010). Both authors indicate that the cake layer is compressible using equation 2.4. Therefore an increase of trans membrane pressure will result in more compression leading to a higher specific fouling resistance and a lower reversibility (Zheng, *et al*., 2010).

$$\alpha_{av} = \alpha_0 \cdot \Delta P^s \quad (2.4)$$

in which: α_0 = specific cake resistance at reference pressure (m/kg)
s = compressibility coefficient (s = 0 refers to no compression, the increase of s represents the layer is more compressed, s = 1 refers to complete compression)

2.4 Factors affecting fouling

As already mentioned the fouling mechanisms will result in a performance decrease of the separation step. The operational performance of membrane filtration is a function of time and many other variables. In general there are three major variables influencing membrane fouling: feedwater properties, operational conditions and membrane characteristics (Amy, 2008). Table 2.4 summarizes the main factors involved in the fouling process for each of these three variables.

Table 2.4 – Factors influencing membrane fouling

Feedwater properties	Operational conditions	Membrane characteristics
Chemical and physical properties feedwater solution	Flux	Clean water flux and/or resistance
Chemical properties of particles, colloids and dissolved material	Recovery	Pore size or molecular weight cut off
Particle size distribution	Pretreatment	Hydrophobicity/hydrophilicity
	Chemical cleaning	Surface charge
	Hydraulic flushes	Surface morphology

2.4.1 Feedwater properties

The potential membrane foulants in WWTP effluent can be present in the influent of the wastewater treatment plant, or originate from the activated sludge (particulate material) and bacterial metabolism. During the biological treatment the particle size distribution in wastewater changes as a result of new cell synthesis, flocculation, adsorption, enzymatic breakdown of macro molecules and biochemical oxidation (Levine *et al.*, 1985). Usually and also presented in Figure 2.1 the biomass and the biologically treated water (WWTP effluent) are separated by gravity in the final clarifier. Generally, the WWTP effluent consist of mainly organic compounds, suspended solids and nutrients (Metcalf and Eddy, 2003). In Table 2.5 an overview of these components is presented adopted from te Poele (2005) and Metcalf and Eddy (2003).

Table 2.5 – Constituents found in WWTP effluent that influence membrane fouling

Soluble biodegradable organics	-	Organics that escaped biological treatment.
	-	Organics formed an intermediate products in the biological degradation of the waster.
	-	Cellular components as a result of cell death or lysis .
Suspended organic material	-	Biomass produced during treatment that escaped separation in the final settling tank.
	-	Colloidal organic solids in the wastewater plant influent that escaped treatment and separation.
Nitrogen and phosphorus	-	Contained in biomass in effluent suspended solids.
	-	Soluble nitrogen as NH_4-N, NO_3-N, NO_2-N and organic–N.
	-	Soluble orthophosphates.
Non biodegradable organics	-	Those originally present in the wastewater influent.
	-	By-products of biological degradation

2.4.1.1 Organic fouling

From the constituents presented in Table 2.5 the organic compounds are mostly associated with fouling of ultrafiltration membranes. Fouling caused by these constituents (organic fouling) can be distinguished in three types (Amy, 2008) for drinking water treatment and wastewater reclamation/reuse:

- *allochthonous natural organic matter* (NOM) dominated by humic substances derived from runoff and leaching of vegetative debris from terrestrial sources within a watershed;
- *autochthonous or algal organic matter* (AOM) consisting of extracellular and intracellulare macromolecules and cellular debris;
- *wastewater effluent* (EfOM) consisting of background (drinking water) NOM plus soluble microbial products (SMPs) derived from biological wastewater treatment.

During filtration of WWTP effluent EfOM deposits on or in the membranes, reducing the permeate rate by the different filtration mechanisms. In particular the soluble microbial products (SMP) and/or (soluble) extracellular polymeric substances (EPS) are considered as the major foulants of EfOM during ultrafiltration of WWTP effluent (Jarusutthirak and Amy, 2001; te Poele, 2005; Rosenberger, *et al*., 2005; Jarusutthirak and Amy, 2007; Haberkamp, *et al*., 2008; Zheng *et al*., 2010). Both, SMP and (soluble) EPS are considered as similair substances. Laspidou and Rittmann (2002) compared the concepts of soluble EPS and soluble SMP and concluded that both are indeed identical. Furthermore, because of the analytical methods for their determination, soluble SMP and soluble EPS cannot be distinguished in engineering practice (te Poele, 2005). Both substances are produced by micro organisms and released into the liquid phase as part of the metabolism and due to biological or mechanical stress (te Poele, 2005). However, with respect to EPS two basic forms are distinguished: bound (or extracted EPS) or soluble EPS (Geilvoet 2010). Bound extracellular polymeric substances are of a biological origin, participate in the formation of microbial aggregates and consist of insoluble materials (sheats, capsular polymers, condensed gel, loosely bound polymers and attached organic material) (Laspidou and Rittmann 2002). Soluble EPS and

SMP consists of soluble macro molecules, colloids and slimes (te Poele, 2005) and can be defined as "the pool of organic compounds that are released into the solution from substrate metabolism (usually with biomass growth) and biomass decay" (Barker and Stuckey, 1999). The main components of soluble EPS and SMP are proteins and polysaccharides and to a minor extent, nucleic acids and lipids (Flemming and Wingender, 2001).

Which compound of SMP, either proteins or polysaccharides, are most strongly linked to membrane fouling is still an open question. For example, te Poele (2005) has demonstrated that the smaller colloids or organic molecules (< 0.10 µm) of WWTP effluent predominantly influence the reversibility during ultrafiltration. These organic molecules had a protein origin and the influence of polysaccharides was not clearly demonstrated. This observation was also noticed by Haberkamp (2008). In that research a model solution mixed with a natural EPS extract and real WWTP effluent were filtrated during cross flow filtration experiments. The model solution with bacterial EPS extract contained significantly more polysaccharides than proteins, whereas the WWTP effluent contained a larger proportion of proteins. The ultrafiltration of WWTP effluent resulted in a higher flux decline than the EPS model solutions. Therefore, it was indicated that proteins play an important role in ultrafiltration membrane fouling.

Other researchers underline the negative impact of proteins but take the effect of polyasaccharides and proteins in one term as biopolymers together. Zhing *et al.* (2009a) found during stirred cell experiments that dissolved substances of WWTP effluent larger than the ultrafiltration pore size attribute to more than 50% of the total fouling resistance. Within the dissolved substances, the concentration biopolymers detected by LC-OCD shows a quantitative correlation with the filterability of water samples in dead-end ultrafiltration. During the filtration process, biopolymers cover membrane pores firstly and lead to a steep flux decline. The impact of polysaccharides and colloids was shown by Jarusutthirak and Amy (2006) during the characterisation of different EfOM fractions in fouling of ultrafiltration and nanofiltration membranes. In addition, Cho *et al.* (1998) characterised clean and NOM fouled ultrafiltration and nanofiltration membranes and indicated polysaccharides or polysaccharide like substances as foulants.

Although the main components of soluble microbial products are the proteins (60%) and polysaccharides (40 – 95%) (Flemming and Wingender, 2001) also humic substances are a major part (Drewes and Fox, 1999; Hezjlar and Chudoba, 1986). Humic substances or fulvic substances are composed of phenol compounds, single sugars and amino acids and bound with ether, carbon and peptide bonds. Compared to the proteins and polysaccharides, humic substances are considered of minor relevance during ultrafiltration of WWTP effluent (Laabs *et al*., 2006; Haberkamp *et al*., 2008).

2.4.1.2 Size of organics

As stated in the previous section various researchers report that fouling is predominantly determined by soluble microbial products in WWTP effluent. To analyse SMP in WWTP effluent different methods (specific UV-absorbance, total organic carbon, colorimetric analyses and size exclusion chromatography) are applied but all these methods solely provide information about the characteristic concentration and not about the characteristic size. Therefore also investigations to characterize the size of (organic) colloidal particles causing fouling during ultrafiltration of WWTP effluent are performed by several researchers. Roorda (2004) performed SUR tests with different fractions of WWTP effluent and showed that the fraction between 0.1 – 0.2 µm predominantly determined the filtration characteristics. This finding is confirmed by research of te Poele (2005). Te Poele (2005) stated that organic colloids of size fraction < 0.45 and 0.10 µm reveal to be of major influence on the filterability. It is assumed that these colloids might possibly be cell fragments and large molecules (te Poele, 2005), which could not be measured by analytical methods like specific UV-absorbance, total organic carbon, colorimetric analyses and size exclusion chromatography. Furthermore, te Poele (2005) stated that the reversibility is mostly influenced by organic molecules (mainly proteins) < 0.10 µm. These findings of Roorda (2004) and te Poele (2005) underline the need to focus on both aspects of organic fouling: concentration and size. Therefore in this thesis the colorimetric analyses will be accompanied by fractionation tests in order to obtain a better understanding of fouling formation during ultrafiltration of WWTP effluent.

2.4.1.3 Divalent cations, pH and ionic strength

In addition to the composition and concentration of EfOM substances the concentration of divalent cations, pH and ionic strength influence the impact of organic fouling. The pH and ionic strength composition of feedwater influence the chemical interactions between the foulants and the membrane and between the foulant and fouling layer (Costa *et al*., 2006) Divalent cations like calcium and magnesium bridges with negatively charged functional groups within the bacterial EPS, which helps to aggregate and stabilize the matrix of biopolymer and microbes (Kim and Jang, 2006). This mechanism may result in a network structure of EPS on the membrane surface influencing the filtration rate.

2.4.2 Operational conditions

2.4.2.1 Operating mode

The permeate flux can be regarded as a measure for the fouling load on the membrane, because it determines how much foulants are transported towards the membrane. This unavoidable transport during permeation can be generally performed in three different operation modes: with constant trans membrane pressure, with constant permeate flux or with a combination of these. During constant trans membrane pressure operation the flux will decrease over time and during constant flux operation the trans membrane pressure will

increase over time. In the case of no fouling there will be no difference between both operation modes but from an engineering practice point of view the use of constant flux is preferable instead of constant trans membrane pressure. However many researchers use constant pressure operation for the benefit of conducting simple short term experiments (Lee *et al*., 2008). Whether the constant flux or the constant trans membrane pressure is more beneficial for fouling control is a difficult question. On the one hand different authors (Loijkine *et al*., 1992; Bourgeos *et al*., 2001; Tarabara *et al*., 2002) prefer for different reasons constant pressure but on the other hand other authors (Defrance and Jaffrin, 1999; Ho and Zydney, 2002) suggest constant flux as it usually results in higher permeate volume production. Differently, Vyas *et al*. (2002) suggest a combination of constant flow and constant pressure, which seems favourable to minimise fouling and optimise process performance.

2.4.2.2 Hydraulic flushes

As a result of the dead-end mode the membrane has to be cleaned often in order to remove the rejected compounds. The period (filtration run) between two hydraulic flushes may vary between 10 and 60 minutes depending on the feedwater quality, but a filtration period of 30 minutes is usually applied in practice. Depending on the type of hydraulic flush either feed water, permeate or ultrapure water are flushed into the membranes. The different types of hydraulic flushes are a forward flush (FF) and back flush (BF) as illustrated in Figure 2.4.

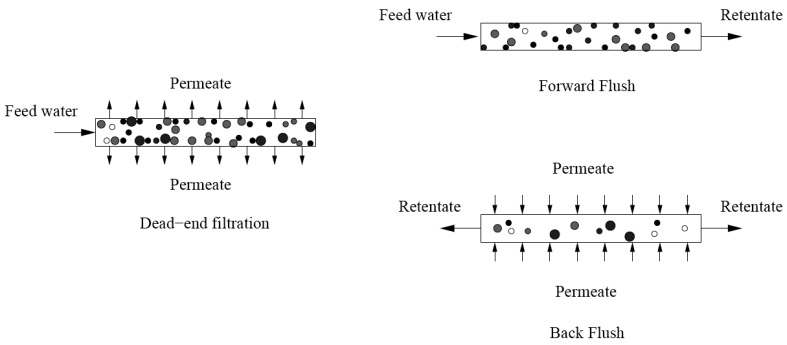


Figure 2.4 - Schematic drawing of hydraulic cleaning: forward flush and back flush (te Poele, 2005)

The forward flush is a turbulent cross flow along the feed side of the membrane surface. With small diameters of membrane fibres high cross flow velocities are needed to obtain turbulent flow. This velocity is many times higher compared to the velocity during dead-end filtration. A mixture of air and water can be used to improve the forward flush. The air is used to create turbulent flow in the membrane under process conditions where no turbulence is attained with the water flow. The air/water flush is patented as the AirFlush® and further investigated by Verberk (2005). For a forward flush feed water can be used in order to obtain a high recovery.

The back flush or backwash is a permeate flow reversed to the filtration mode. As a result, the retained material in the membrane pores and on the membrane is released. Permeate is used for the back flush in order to keep the permeate side of the membrane free of particles. The back flush flux is normally 2 to 2.5 times the flux during filtration. Typical duration of the back flush is 30 to 60 seconds. After removing the retained material from the pores and membrane surface it has to be transported out of the module. Because the amount of permeate used for back flush is limited (because of recovery) the transport of retained material may be insufficient. A combined back flush and forward flush can be used to overcome this problem.

2.4.2.3 Chemical cleaning

In order to maintain an optimal flux, membranes are often cleaned by periodical hydraulic flushes. Nevertheless after several filtration periods the performance declines, in spite of frequently applied hydraulic flushes. In that case the membranes need to be cleaned chemically in order to recover the flux to initial or acceptable values. The concentration of the chemicals and the cleaning time are important parameters for efficient use of a chemical cleaning procedure. In order to prevent membrane degradation during a chemical cleaning, the chemical properties of the membrane should be known. Effective cleaning must inhibit the redeposition of the foulants on the membrane surface. The chemicals that are used for cleaning can be classified in the following way (Table 2.6).

Table 2.6 – Classification of chemical cleaning agents and their mechanisms (te Poele, 2005)

Cleaning agent	Mechanism
Acid	Removing of crystallised salts, metal oxides and metal hydroxides.
Alkali	Removing of general organic fouling.
Active chlorine	Active chlorine as the active component to minimise organic fouling and micro-organisms.
Oxide	Oxide as the active component to diminish mainly organic fouling and micro-organisms, but can also be used to minimise other types of fouling.
Organic solvent	Removal of organics by solubility
Enzyme	Specific enzyme degrades specific organic foulants.

Beside the periodical chemical cleaning (often called chemical enhanced back flush) which is part of the automated process control of the installation also a more intensive cleaning might be necessary. The so-called “cleaning in place” (CIP) can last from a few hours to several days and is most of the time not automated. If the CIP is not able to clean the membranes, the membranes are replaced by new ones.

2.4.3 Pretreatment

The integration of an extra treatment step (pretreatment) in front of the membrane process is an important trend in the development of membrane filtration to improve the performance. In this thesis pretreatment refers to the operation of processes that are conducted in precedence to membrane filtration to improve the performance of membranes during filtration of WWTP effluent. Beside the improvement of the performance pretreatment is sometimes applied to enhance the rejection efficiencies. However, predominantly pretreatment is applied to control membrane fouling (Huang *et al*., 2007).

In a paper of Huang *et al*. (2007) the mechanisms of pretreatment are summarized by three types of effect:

- *Physical effects*: pretreatment can increase the size of aquatic substances by aggregation (i.e., coagulation) or adsorbing them onto materials larger than membrane pore dimensions thereby enhancing their rejections by membrane filtration and reducing membrane fouling.
- *Chemical effects*: pretreatment can alter the nature (i.e., prefiltration, coagulation or adsorption) and magnitude (coagulation and softening) of interfacial interactions involved in the subsequent membrane filtration and mitigate the impact of undesirable interactions between aquatic substances and membrane surfaces.
- *Biological effects*: pretreatment can either enhance positive impacts (i.e., reduction of organic contaminant concentration in the feedwater by biodegradation) or reduce negative impacts (i.e., disinfection of feedwater to reduce biofilm formation) of biological processes in the performance of membrane filtration.

The effects are not strictly separated in process operations of pretreatment. Some pretreatment processes can have multiple effects on membrane filtration. For example, coagulation may results in two effects. Firstly, an increase of the particle size by the aggregation of aquatic substances (physical) and secondly a chemical reduction of dissolved and colloidal organic matter that may attribute to irreversible fouling (te Poele, 2005).

In general coagulation, prefiltration and adsorption are the most widely used pretreatment technologies in full scale applications. Beside these technologies oxidation and ion exchange are getting more attention. In the following sections the different pretreatment technologies are presented.

2.4.3.1 Coagulation

Metcalf and Eddy (2003) define coagulation as the process of destabilizing colloids and particles so that particle growth can occur as a result of particle collisions. Strictly the word coagulation refers to the destabilization process and the word flocculation to the formation of aggregates (flocks) consisting of destabilized colloidal and particulate matter and precipitated

hydroxides. In this thesis coagulation is used as a general term that includes both destabilization and flocculation. Most commonly cationic or metallic coagulants like aluminium and ferric salts are used for pretreatment. The operation of coagulation for membrane filtration can be directly (or so called direct in-line coagulation) or in combination with sedimentation or filtration. During these separation steps particles subject to membrane filtration can be effectively removed. Therefore the feedwaters after coagulation-sedimentation/filtration usually cause less fouling than raw feedwater (Huang *et al*., 2007). Roorda (2004) investigated the performance of an ultrafiltration pilot installation in combination with coagulation-filtration as pretreatment. Although some increase of performance was obtained, Roorda (2004) concludes that the technologies did not efficiently remove the 0.1 – 0.2 µm particles, which is the fraction that predominantly determines filtration characteristics. Direct in-line coagulation with ferric chloride was investigated by Decarolis *et al*. (2001) on pilot scale with a dead-end ultrafiltration system. This research showed that ferric chloride pretreatment enhanced membrane productivity by increasing particle floc size, which led to decreased pore plugging, reduced cake layer resistance and enhanced backwashing efficiency.

2.4.3.2 Prefiltration

In this thesis prefiltration as pretreatment for ultrafiltration and microfiltration involves the use of conventional packed bed filters i.e. rapid sand filters, dual media filter etc. Its main purpose is the removal of suspended (including coagulated flocs) and colloidal matter from raw WWTP effluent. Many authors (de Koning and van Nieuwenhuijzen, 1999; Bourgeous *et al*., 2001; Roorda, 2004; te Poele, 2005; Fan, *et al*., 2008) have reported about the effectiveness of prefiltration prior to ultrafiltration or microfiltration. De Koning and van Nieuwenhuijzen (1999) investigated the performance of the combination flocculation – filtration – ultrafiltration at two WWTPs (WWTP Ede and WWTP Elburg) in the Netherlands. Although the prefilter did not produce high removal efficiencies it appeared to be an important part of the total polishing concept. However, the results differed at the two treatment locations. At the WWTP Elburg prefiltration was required to achieve constant high fluxes and reliable operation at constant trans membrane pressure. Without prefiltration the flux rapidly decreased from an average of 90 $L/m^2 \cdot h$ to a low level of 40 $L/m^2 \cdot h$, without successful hydraulic cleaning procedures. Only chemical cleaning was effective for a short term. At the WWTP Ede successful and stable operation of ultrafiltration was possible at a trans membrane pressure of 0.6 – 0.9 bar without prefiltration. Nevertheless the content of suspended solids was higher in the effluent of WWTP Ede compared to WWTP Elburg. Therefore the authors suggested that the filterability of WWTP effluent is not related to amount of particles or suspended solids but effected by local circumstances. This phenomenon has been further investigated by Roorda (2004) who concluded that multi media filtration slightly improves the filterability of WWTP effluent with a maximum of 25%. But multi media filtration did not effectively decrease the 0.1 – 0.2 µm fraction of WWTP

effluent. This fraction is not measured during the analyses of turbidity and suspended solids. The results of Roorda (2004) have been confirmed by research of Bourgeous *et al*. (2001) and te Poele (2005). Bourgeous *et al*. (2001) showed that a thin cake layer developed for ultrafiltration of effluent after prefiltration, while a thicker cake layer developed for raw WWTP effluent. The thinner cake layer showed better filterability and was completely removed by a regular back flush. The cake layer found for raw WWTP effluent could not be removed completely with a backflush because of clogging membrane fibers. More or less the same observation was obtained by Fan, *et al*. 2008. Lab scale experiments showed that the combination of coagulation and prefiltration (1.5 µm) resulted in better ultrafiltration and microfiltration performance compared to coagulation-sedimentation and direct in-line coagulation.

Next to the mentioned prefiltration techniques, recent work has shown biofiltration as a promising pretreatment technology to reduce fouling (Mosqueda-Jimenez, 2006 and Huck and Sozański, 2008). Treatment combinations of natural biofiltration i.e. bank filtration and soil aquifer treatment and ultrafiltration/microfiltration are rarely applied in advanced water treatment. Nevertheless biofiltration processes offer simple and cost effective measures to reduce the content of bio available organic compounds (Zheng *et al*., 2009b). This effect has been shown by Zheng, *et al*. (2009b) with respect to the removal of biopolymers of WWTP effluent. Slow sand filtration of WWTP effluent removed the biopolymer peak and therefore decreased the fouling potential of water samples to ultrafiltration membranes.

2.4.3.3 Adsorption

In the context of this thesis adsorption refers to the use of activated carbons and other "pre-formed" adsorbents in precedence to membrane filtration. The adsorption process itself involves the uptake of aquatic substances by the surface of the activated carbon. Due to the high porosity and dispersity activated carbon has a relatively large specific surface area. In combination with ultrafiltration and microfiltration activated carbon can have two functions. Firstly, adsorption of small substances of WWTP effluent that can poorly be removed by the membranes. These substances can be of different origin e.g. humic substances, micropollutants, (organically bounded) heavy metals, etc. Secondly, the adsorbents may also serve as competitor with organic matter that can cause membrane fouling once being adsorbed on membrane surfaces (Crozes, *et al*., 1993).

Generally, activated carbon for water treatment can be separated in two size classifications: powdered activated carbon (PAC), which typically has a diameter of less than 0.074 mm, and granulated activated carbon, which has a diameter of larger than 0.1 mm (Metcalf and Eddy, 2003). The most intensively studied size for membrane filtration is probably powdered activated carbon. PAC is added into the feedwater to adsorb the small substances; it can be effectively rejected by ultrafiltration/microfiltration as its size is significantly greater than the

membrane pores. Consequently, PAC in the membrane concentrate may be recycled to maintain enough adsorbent concentration in feedwater. GAC is mostly applied in precedence to ultrafiltration/microfiltration in a filterbed and operated like a discontinuous sand/anthracite filter. This process is also referred to the name adsorptive filtration (Roorda, *et al*., 2005).

In respect to PAC and the effect on membrane fouling different findings are reported in the literature. In general it seems that the effect depends on the membrane material, operation mode, membrane configuration and type of feedwater. Mozia *et al*. (2005) tested the influence of PAC addition on flux decline of three different polymer ultrafiltration membranes. The tested membranes were prepared from polysulfone (PSF), cellulose acetate (CA) or polyacrylonitrile (PAN). The researchers presented the combination of PAC addition and membranes formed of PAN as most favourable considering the effectiveness of organics removal and the permeate flux. In contrast an earlier study performed by Lin *et al*. (1999) showed a significant decrease of flux of negatively charged polysulfone membranes due to PAC addition. One of the main differences between both studies was the applied membrane configuration; Lin *et al*. (1999) used a hollow fibre while Mozia *et al.* (2005) tested a flat sheet membrane. The optimization of the PAC-UF configuration in terms of increase of backwash pressure, greater linear velocity, feedwater direction and proper module design was also suggested by Oh *et al*. (2006) to get a suitable integrated system. Nevertheless good experience regarding ultrafiltration fouling control of WWTP effluent and PAC addition was obtained by Shon *et al*. (2004a) and Haberkamp *et al*. (2007). Shon *et al*. (2004a) presented similar reduction of flux decline for both flocculation and PAC adsorption. In spite of the same effect the underlying mechanisms were different. Flocculation resulted in higher reduction of colloidal matter and larger molecules than with PAC adsorption. The flocculation also achieved a removal of a portion of small molecular weight organics through the mechanisms of adsorption and complexation (Shon *et al*., 2004a). The PAC adsorption played a major role in removing small organics only. The removal of the colloidal portion (between 3500 Da to 0.45 µm) by adsorption was not significant. Haberkamp *et al*. (2007) observed as well a positive affect of PAC addition on flux decline during ultrafiltration but remarked that increased demand of activated carbon due to considerable consumption of adsorption capacity by low molecular weight substances, which do not presumably contribute tot membrane fouling, may be a limiting factor for the application of adsorption as the pretreatment process for fouling reduction.

Compared to the application of PAC addition less research is published about the use of granulated activated carbon (GAC) filtration in precedence to ultrafiltration and microfiltration. Nevertheless GAC and biological activated carbon filtration (BACF) give important perspectives in minimizing organic fouling. The main distinction between BACF and GAC filtration is the removal mechanism. BACF combines adsorption and biodegradation where GAC filtration only implies adsorption as a removal mechanism.

Regarding GAC filtration, Tsujimoto *et al*. (1998) found that this pretreatment step reduced irreversible fouling of ultrafiltration membranes treating a natural surface water. The combination of GAC – (cross flow) ultrafiltration and biologically treated sewage effluent was investigated by Shon *et al*. (2004b). In this study this combination was compared with other pretreatment steps for ultrafiltration: flocculation, PAC adsorption and flocculation followed by PAC adsorption. Compared to the other pretreatment technologies the flux decline after GAC filtration was more but it was still significantly better than raw biologically treated sewage effluent. The combination of GAC filtration and airflushed microfiltration with WWTP effluent was also subject of research by Roorda *et al*. (2005). In terms of stable operation performance the researchers found adsorptive filtration one of the best pretreatment steps for microfiltration.

2.4.3.4 Alternative pretreatment technologies

Advanced Oxidation Processes

In water treatment applications, advanced oxidation processes usually refer to a specific subset of processes that involve ozone (O_3), peroxide (H_2O_2), and/or ultra violet (UV) light. Usually only ozonation is applied for the treatment of wastewater. Park (2002) investigated ozonation of chemical wastewater as pretreatment for UF. It was concluded that the flux rate increased as the amount of ozone increased. Seo *et al*. (2001) concludes for ceramic membranes ozone is effective to control the membrane fouling by the increased organic degradation.

Ion exchange

Application of magnetic ion exchange (MIEX®) resin to membrane filtration has been studied at different scales in the past few years. Zhang *et al*. (2006) tested MIEX® as pretreatment to a submerged membrane system in the treatment of biologically treated wastewater. It was found that MIEX® resin could effectively remove the small molecular weight (500 – 1000 Da) organic matter and the hydrophilic portion of DOC. At optimal concentration MIEX® could remove as much as 60% of DOC in the wastewater resulting in a longer operational time of the membrane process. Combining MIEX® and PAC adsorption could even increase the removal rate (TOC removal of 80%). Generally the effect of MIEX® on ultrafiltration and microfiltration seems to be positive though the major mechanisms are unsolved.

Precoat

Removal of the 0.1 to 0.2 µm fraction might be done by the formation of a precoat (e.g. Diatomite, Powdered Activated Carbon (PAC), Kaolinite or iron oxide) on the membrane that acts as secondary membrane. Enhanced Pre-Coat Engineering (EPCE) results in a better restoration of the permeability after a backwash. Bigger particles seem to restore the membrane better after a backwash or enhanced backwash. Smaller particles seem to give a

lower rate of fouling. EPCE is a promising technique to increase membrane performance (Galjaard *et al*., 2001).

Integrated pretreatment technologies
As discussed previously, pretreatment technologies often remove only a certain type or range of compounds in WWTP effluent. It is, therefore, reasonable to consider that proper integration of multiple pretreatment can combine the benefits of each separate pre-treatment technology. For example, the technologies, coagulation – PAC adsorption – ultrafiltration are presented by different researchers (Shon *et al*., 2004a and Haberkamp *et al*., 2007) as an optimal combination in terms of flux decline and DOC removal. Roorda (2004) and te Poele (2005) presented coagulation – multi media filtration – ultrafiltration as a good integrated concept for stable operation performance. Generally, integration of pretreatment technologies can provide better feedwater quality for membranes, but the costs of the entire system may increase drastically (Huang *et al*., 2007).

2.4.4 Membrane characteristics

The effective pore size (Lozier *et al*., 2008) and surface charge of membranes (Schafer *et al*., 1998) are considered to be significant factors contributing to the reduction of permeate flux The effective pore size of a membrane is a membrane characteristic affecting the decrease of permeate flux. The lower the molecular weigh cut off (MWCO), the larger the membrane resistance. A negatively charged membrane will electrostatically repulse negatively charged functional groups associated with EfOM (Jarusutthirak and Amy, 2001). The more negative charge density on the membrane surface is correlated to the greater hydrophilicity of the membrane. Therefore, a less negative surface charge (more hydrophobic) may increase the deposition of hydrophobic EfOM leading to more adsorptive fouling.

The interaction between EfOM and the membrane characteristics like surface charge and MWCO has been investigated by Jarusutthirak and Amy (2001). They established a dependency of the flux decline, EfOM rejection and fouling mechanisms on the charge of membrane surface and the MWCO. The negatively charged surface led to the adsorption of the hydrophobic portion of EfOM.

The effect of the pore size of membranes on the retention of foulants of WWTP effluent is presented by Laabs *et al*. (2006). During filtration experiments with ultrafiltration membranes (pore size of 10 nm) the substances (organic colloids, polysaccharides and proteins) eluting in the polysaccharide peak of a size exclusion chromatogram changed completely e.g. the feedwater exhibits a clearly polysaccharide peak, the permeate sample contains no substances in this molecular weight/size range. But during the same experiment with microfiltration membranes (pore size of 50 nm) the substances of the polysaccharide peak remained in the

filtrate. Therefore Laabs *et al*. (2006) suggested that the fouling rate depends on the pore size of the membranes even when the size of the pores differs a little.

2.5 Fouling indicators and predictions

As written in chapter 1 the performance of the ultrafiltration process is determined by both filterability of the feedwater and by reversibility of the fouling layer. Both parameters are affected by the operational conditions and membrane characteristics but the more fundamental cause for membrane fouling are the properties of the feedwater. Therefore many researches (Boerlage *et al*., 2003; Roorda, 2004; Rosenberg *et al.,* 2005; Park *et al*., 2006; Kim and DiGiano, 2006 and Huang *et al*., 2008) underline the need for a general method to measure and predict the fouling potential of the feedwater to membrane filtration system. Such a method can be used at the design stage to assess required pretreatment and later to monitor the effectiveness and performance of a pretreatment system during plant operation (Park *et al*., 2006). Ultimately the fouling indices enable engineers to determine the design requirements without conducting pilot studies which need considerable time and expenses. There have been attempts to establish tests and indices to describe the filterability, respectively fouling potential of feed suspensions and solutions (Rosenberg *et al*., 2005):

- On-line measurements of flux and trans membrane pressure during continuous filtration can be used to calculate the fouling rate (slope of resistance over time). This can be done either in batch experiments (e.g. with side-stream modules) or in situ.
- Time to filter (TTF), modified fouling index (MFI), specific resistance to filtration (SRF) and specific ultrafiltration resistance (SUR) are used to describe filterability. Recently the unified modified fouling index (UMFI) is added to this list as a parameter to describe both filterability and reversibility. All these tests are based on the theory of cake filtration and are usually performed in dead-end mode on lab or bench scale.

2.5.1 Bench and lab scale tests

In this section briefly the MFI-(UF), UMFI and SUR are discussed.

2.5.1.1 Modified filtration index-(UF)

The MFI is measured in dead-end mode with a microfiltration membrane (0.45 µm) at constant pressure (2 bar) and continous measurement (every 30 seconds) of the produced filtrate. According to the cake filtration model (Hermia, 1982) at constant pressure the following relation between filtration time and filtrated volume can be derived (Mulder, 1996):

$$\frac{t}{V} = \frac{\eta \cdot R_m}{\Delta P \cdot A} + MFI \cdot V \qquad (2.5)$$

in which: t = filtration time (s)
V = filtrate volume (m^3)

η	=	water viscosity (N s/m^2)
R_m	=	membrane resistance (m^{-1})
ΔP	=	applied trans membrane pressure (bar or N/m^2)
A	=	membrane surface area (m^2)
MFI	=	membrane fouling index (s/m^3)

Equation 2.5 predicts a linear relationship between t/V and V during cake filtration. A high value of MFI indicates rapid fouling of the membrane.

The MFI is further developed by Boerlage *et al*. (2003) to the MFI-UF to measure and predict the particulate fouling potential for different feedwaters in membrane filtration installations. A disadvantage of the MFI-UF is the applied pressure. This pressure is 2 bar which is about 4 times more than usual in ultrafiltration applications. Especially during tests with WWTP effluent the applied pressure will result in unrepresentative values due to the compression of the formed cake layer.

2.5.1.2 Unified Membrane Filtration Index

The Unified Membrane Filtration Index (UMFI) has been recently reported by Huang *et al*. (2008) and presented as a parameter to quantify and compare the fouling on different scales (e.g., lab, bench and full scale) and different units (e.g., stirred-cell versus hollow fiber bench scale units). A value of UMFI (m^2/L) can be estimated from a data plot of inversed normalized flux (J_s/J_{s0}) versus hydraulic throughput (L/m^2) as shown in equation 2.6. Like the other filtration indices the UMFI is based on solely cake filtration and Darcy's law.

$$\frac{1}{J_s'} = 1 + \left(\frac{\alpha_c C_f}{R_m} \right) V_s \qquad (2.6)$$

in which:	J_s'	=	normalized specific flux (-)
	α_c	=	specific cake resistance (m^{-1})
	C_f	=	concentration of foulants (kg/m^3)
	R_m	=	membrane resistance (m^{-1})
	V_s	=	permeate throughput (L/m^2)

Based on this relationship, UMFI (m^2/L) is defined as follows:

$$UMFI = \frac{\alpha_c C_f}{R_m} \qquad (2.7)$$

For a filtration without hydraulic backwash, UMFI is related to the hydraulic property of the cake layer (α_c), the concentration of total foulants (C_f) and the hydraulic property of the clean

membrane. Its value is not affected by the operating mode. Temperature effects are also canceled out through the normalization of specific fluxes. If the concept is applied to a filtration with either frequent hydraulic backwashes or chemical cleaning (as with the pilot systems), the UMFI can still be calculated as a measure of the rate of hydraulically irreversible fouling or chemically irreversible fouling that occurs within certain unit permeate throughputs.

A critical note concerning the UMFI is that the value depends on the hydraulic property of the clean membrane (R_m). As a consequence it means the UMFI is not universal but differs for each type of membrane.

2.5.1.3 Specific Ultrafiltration Resistance

To measure the filterability of WWTP effluent Roorda (2004) developed a parameter called Specific Ultrafiltration Resistance (SUR). The SUR is calculated from the slope of a filtration curve (t/V versus V) that is measured in a period of 30 minutes of filtration over an ultrafiltration membrane at a constant temperature (~ 20 °C) and a trans membrane pressure (TMP) of 0.5 bar. The parameter is used during the experiments presented in this thesis and therefore is described in detail in chapter 3.

2.5.1.4 Vito Fouling Measurement

The Belgian company VITO (Flemish Institute for Technological Research) developed a filtration characterisation method called the Vito Fouling Measurement (VFM) (Braun *et al*., 2005). In contrast to the MFI-(UF), UMFI and SUR the VFM aims to characterise (by mathematical processing) all (complex) fouling phenomena (Brauns *et al*., 2002) instead of only cake filtration. As a consequence the VFM measurement does not results in one value like the MFI-(UF), UMFI and SUR but it will give a graphical presentation. By comparing filtration graphs of different types of feedwater information is gained about the fouling potential. However, since cake filtration is considered as the predominant mechanism during ultrafiltration of WWTP effluent (Roorda, 2004) this approach will not provide much extra information compared to the MFI-(UF), UMFI and SUR.

2.5.2 On-line measurements

In full scale or pilot installations, the filterability is measured in a different way. The filterability is measured as fouling rate (dR/dt) which is the increase in filtration resistance over time after one filtration period (see Figure 2.5) The filterability can be derived from on-line measurements of flux, TMP and temperature (te Poele, 2005), see equations 2.8 and 2.9

$$\frac{dR}{dt} \tag{2.8}$$

$$R = \frac{\Delta P}{\eta(T) \cdot J} \tag{2.9}$$

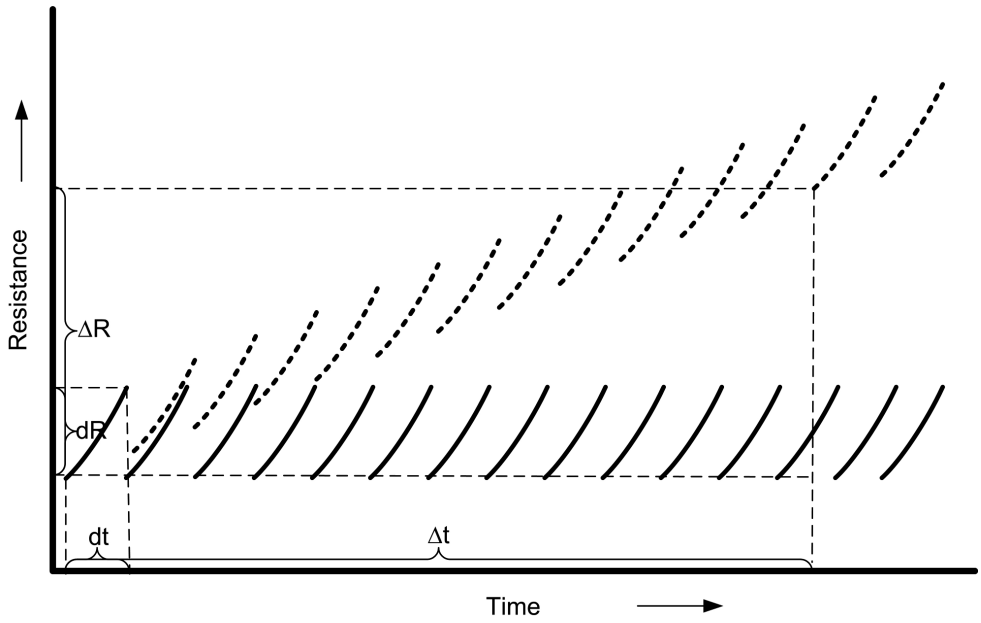


Figure 2.5 – Typical filtration curves illustrating filterability and reversibility

Reversibility is described as the extent within which the filtration resistance after applying a hydraulic cleaning is returned to the start value. If the filtration resistance after hydraulic cleaning is equal to the filtration resistance at the start of the previous filtration period, then the fouling is considered to be completely reversible (te Poele, 2005). Figure 2.5 shows two different filtration curves, with a different reversibility. The lowest filtration curve (covered line) is completely reversible. The reversibility can be determined after a few filtration periods and is illustrated in Figure 2.5 and described in equation 2.10.

$$\frac{\Delta R}{\Delta t} \tag{2.10}$$

2.6 Summary

The conventional method for the treatment of municipal wastewater is the activated sludge process. After the treatment process the micro-organisms (activated sludge) are separated from the treated water (WWTP effluent) by sedimentation but the WWTP effluent still consists of different constituents that may foul the membranes when it is tertiary treated by e.g. ultrafiltration. In this thesis fouling is defined as "the process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores".

In the literature soluble microbial products (SMP) and/or (soluble) extracellular polymeric substances (EPS) are considered as the major foulants of EfOM during ultrafiltration of WWTP effluent. But it has to be considered that in addition to soluble organic material, colloidal organic particles play an important role during ultrafiltration of effluent. With the analytical method that is mainly applied (specific UV-absorbance, total organic carbon, colorimetric analyses and size exclusion chromatography) only concentrations are characterized but not the size. Furthermore Roorda (2004) and te Poele (2005) have shown the importance of fractionation tests in order to quantify size of organic colloids. These studies have shown that especially the colloidal fraction of 0.1 – 0.2 µm causes fouling.

In order to remove fouling membranes are periodical chemically and hydraulically cleaned. In addition to these operational strategies the feedwater of membranes can also be pretreated. Main applied pretreatment technologies in practice are coagulation, prefiltration and adsorption. Unfortunately these pretreatment technologies do only partly remove the earlier mentioned fraction size of 0.1 – 0.2 µm. Therefore other pretreatment technologies have to be applied to remove this colloidal fraction. In the literature biofiltration (e.g. slow sand filtration) and the combination of coagulation – adsorption are considered as promising technologies. Therefore the performance of these technologies is investigated in the context of this thesis.

To measure the fouling rate/potential of WWTP effluent during ultrafiltration of WWTP effluent different methods are available. In this thesis the SUR measurement is used because the process conditions of this method are closely related to the process conditions applied in practice. Furthermore this measurement results in one value independent of the initial membrane resistance. On pilot and full scale the fouling is characterised in accordance to methods applied by other researchers.

References

Abdessemed, D., Nezzal, G. and Ben Aïm, R. (2002). *Fractionation of a secondary effluent with membrane separation*. Desalination, 146, 433 – 437.

Adin, A. (1999). *Particle characteristics: a key factor in effluent treatment and reuse.* Water Science and Technology, 40, 4, 67 – 74.

Amy, G. (2008). *Fundamental understanding of organic matter fouling.* Desalination, 231, 44 – 51.

Azema, N., Pouet, M-F., Berho, C. and Thomas, O. (2002). *Wastewater suspended solids study by optical methods.* Colloids and Surfaces A: Physochemical and Engineering Aspects, 204, 131 – 140.

Barker, D.J. and Stuckey, D.C. (1999). *A review of soluble microbial products (SMP) in wastewater treatment systems*. Water Research, 33, 14, 3063 – 3082.

Berg, G.B. van den (1988). *Concentration polarization in ultrafiltration – models en experiments.* PhD thesis, U *lux decline in ultrafiltration processes*. PhD thesis, Department of Chemical Technology, University of Twente, Enschede, The Netherlands.

Boerlage, S.F.E., Kennedy, M.D., Aniye, M.P., Abogrean, E., Tarawneh, Z.S. and Schippers, J.C. *The MFI-UF as water quality test and monitor*. Journal of Membrane Science, 2003, 271 – 289.

Bowen, W.R. and Jenner, F. (1995). *Theoretical descriptions of membrane filtration of colloids and fine particles: an assessment and review.* Advances in Colloid an Interface Science, 56, 141 – 200.

Bourgeos, K.N., Darby, J.L. and Tchobanoglous, G. *Ultrafiltration of wastewater: effects of particles, mode of operation, and backwash effectiveness*. Water Research, 35, 1, 77 – 90.

Brauns, E., Hoof, V. van, Molenberghs, B., Dotremont, C., Doyen, W. and Leysen, R. (2002). *A new method of measuring and presenting the membrane fouling potential*. Desalination, 150, 31 – 34.

Brauns, E., Dotremont, C., Hoof, V. van and Doyen, W. (2005). *Het meten van het membraan vervuilingspotentieel van water*. Afvalwaterwetenschap, 4, 1, 18 – 35.

CBS (2006). *25 Jaar Statistiek Zuivering van afvalwater in vogelvlucht*. (in Dutch), Voorburg/Heerlen, the Netherlands.

CBS (2008). *Zuivering van stedelijk afvalwater*. (in Dutch), Voorburg/Heerlen, the Netherlands, March 2008, www.StatLine.nl.

Cho, J., Amy, G., Pellegrino, J. and Yoon, Y. (1998). *Characterization of clean and natural organic matter (NOM) fouled NF and UF membrane, and foulants characterization.* Desalination, 2, 2, 101 – 108.

Costa, A.R., de Pinho, M.N. and Elimelech, M. (2006). *Mechanisms of colloidal natural organic matter in fouling of ultrafiltration*. Journal of Membrane Science, 281, 716 – 725.

Crozes, G., Anselme, C. and Mallevialle, J. (1993). *Effect of adsorption of organic-matter on fouling of ultrafiltration membranes*. Journal of Membrane Science, 84, 1 – 2, 61 – 77.

Decarolis, J., Hong, S. and Taylor, J. (2001). *Fouling behaviour of a pilot scale inside-out hollow fiber UF membrane during dead-end filtration of tertiary wastewater*. Journal of Membrane Science, 191, 165 – 178.

Defrance, L. and Jaffrin, M.Y. (1999). *Comparison between filtration at fixed transmembrane pressure and fixed permeate flux: application to a membrane bioreactor used for wastewater treatment.* Journal of Membrane Science, 152, 203 – 210.

Drewes, J.E. and Fox, P. (1999). *Fate of natural organic matter (NOM) during groundwater recharge using reclaimed water.* Water Science and Technology, 40, 9, 241 – 248.

Dijk, J.C. van, Rietveld, L.C., Verberk, J.Q.J.C. (2001). *Drinking water technology 1 ct4470 (in Dutch).* Lecture notes, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Fan, L., Nguyen, T., Roddick, F.A. and Harris, J.L (2008). *Low-pressure membrane filtration of secondary effluent in water reuse: Pre-treatment for fouling reduction*. Journal of Membrane Science, 320, 135 – 142.

Flemming, H-C. and Wingender J. (2001). *Relevance of microbial extracellular polymeric substances (EPSs) – Part 1: Structural and ecological aspects.* Water Science and Technology, 43, 6, 1 – 8.

Galjaard, G., Buijs, P., Beerendonk, E., Schoonenberg, F. and Schippers, J.C. (2001). *Precoating (EPCE®) UF membranes for direct treatment of surface water.* Desalination, 139, 1, 305 – 316.

Geilvoet, S.P. (2010). *The Delft Filtration Characterisation method*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Haberkamp, J., Ruhl, A.K., Ernst, M. Jekel, M. (2007). *Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting behaviour in ultrafiltration*. Water Research, 41, 3794 – 3802.

Haberkamp, J., Ernst, M., Böckelmann, U., Szewzyk, U. and Jekel, M. (2008). *Complexity of ultrafiltration membrane fouling caused by macromolecular dissolved organic compounds in secondary effluents*. Water Research, 42, 3153 – 3163.

Heijman, S.G.J., Vantieghem, M., Raktoe, S., Verberk, J.Q.J.C. and van Dijk, J.C. (2007). *Blocking of capillaries as fouling mechanism for dead-end ultrafiltration.* Journal of Membrane Science, 287, 1, 119 – 125.

Hejzlar, J. and Chudoba, J. (1986). *Microbial polymers n the aquatic environment-II: isolation form biologically nonpurified and purified municipal wastewater and analysis.* Water Research, 1986, 20, 10, 1217 – 1221.

Hermia, J. (1982). *Constant pressure blocking filtration laws – application to power-law non Newtonian fuids*. Transactions of the Institution of Chemical Engineers, 60, 183 – 187.

Huck, P.M. and Sozański, M.M. (2008). *Biological filtration for membrane pre-treatment and other applications: towards the development of a practically-oriented performance parameter*. Journal of Water Supply: Research and Technology-AQUA, 57, 4, 203 – 223.

Huang, H., Young, T. and Jacangelo, J.G. (2008). *Unified membrane fouling index for low pressure filtration of natural water: principles and methodology*. Environmental Science & Technology, 42, 714 – 720.

Huang, H., Young, T. and Jacangelo, J.G. (2007). *Pretreatment and its effect and implications on water treatment by low pressure membranes.* Proceedings 4th IWA leading-edge conference and exhibition on water and wastewater technologies, June 3-6, Singapore, Republic of Singapore.

Ho, C.C. and Zydney, A.L. (2002). *Transmembrane pressure profiles during constant flux microfiltration of bovine serum albumin*. Journal of Membrane Science, 209, 363 – 377.

Koning, J. de and Nieuwenhuiijzen, A.F. van. *Optimal combination of flocculating filtration and ultra filtration for advanced effluent treatment in the Netherlands*. Water Science and Technology, 40, 4 – 5, 285 – 292.

Jarusutthirak, C. and Amy, G. (2001). *Membrane filtration of wastewater effluents for reuse: effluent organic matter rejection and fouling.* Water Science and Technology, 42, 10, 225 – 232.

Jarusutthirak, C., Amy, G. and Croué, J.P. (2002). *Fouling characteristics of wastewater effluent organic matter (EfOM) isolates on NF and UF membranes*. Desalination, 145, 247 – 255.

Jarusutthirak, C. and Amy, G. (2007). *Understanding soluble microbial products (SMP) as component of effluent organic matter (EfOM)*. Water Research, 41, 2787 – 2793.

Kim, J. and DiGiano, F.A. (2006). *A two-fiber, bench scale-test of ultrafiltration (UF) for investigation of fouling rate and characteristics*. Journal of Membrane Science, 271, 196 – 204.

Kim, I.S. and Jang, N. (2006). *The effect of calcium on the membrane fouling in the membrane bioreactor (MBR)*. Water Research, 40, 2756 – 2764.

Koros, W.J., Ma, Y.H. and Shimidzu, T. (1996). *Terminology for membranes and membrane processes; IUPAC recommendations 1996*. Journal of Membrane Science, 120, 149 – 159.

Laabs, C.N., Amy, G.L. and Jekel. M. (2006). *Understanding the size and character of fouling-causing substances from effluent organic matter (EfOM) in low-pressure membrane filtration.* Environmental Science & Technology, 40, 14, 4495 – 4499.

Laspidou, C.S. and Rittmann, B.E. (2002). *A unified theory for extracellular polymeric substances , soluble microbial products, and active and inert biomass.* Water Research, 36, 2711 – 2720.

Lee, E.K., Chen, V. and Fane, A.G. (2008). *Natural organic matter (NOM) fouling in low pressure membrane filtration – effect of membranes and operation modes*. Desalination, 218, 257 – 270.

Levine, A.D., Tchobanoglous, G. and Asano, T. (1985). *Characterization of the size distribution of cantaminants in wastewater: treatment and reuse implications.* Journal WPCF, 57, 7, 805 – 816.

Levine, A.D., Tchobanoglous, G. and Asano, T. (1991). *Size distributions of particulate contaminants in wastewater and their impact on treatability*. Water Research, 25, 8, 911 – 922.

Lojkine, M.H., Field, R.W. and Howell, J.A. (1992). *Crossflow microfiltration of cell suspensions: a review of models with emphasis on particle size effects.* Transition Inst. Chemical Engineering, 70, 149 – 164

Lin, C-F., Huang, Y-J. and Hao, O.J. (1999). *Ultrafiltration processes for removing humic substances: effect of molecular weight fractions and PAC treatment*. Water Research, 33, 5, 1252 – 1264.

Lozier, J., Cappuci, L., Amy, G., Lee, N., Jacangelo, J.G., Huang, H., Young, T., Mysore, C., Emeraux, C., Clouet, J., Croué, J-P. and Heijman, B. (2008). *Natural organic matter fouling of low pressure membrane systems,* American Water Works Association Research Foundation, USA.

Metcalf and Eddy (2003). *Wastewater engineering, treatment and reuse*. Fourth Edition, McGraw-Hill, New York, United States of America.

Mozia, S., Tomaszewska, M. and Morawski. (2005). *Studies on the effect of humic acids and phenol on adsorption-ultrafiltration process performance*. Water Research, 39, 501 – 509.

Mosqueda-Jimenez, D.B and Huck, P.M. (2006). *Fouling analysis of ultrafiltration and nanofiltration*. Water Practice Technology, 1, 4, 90 – 99.

Mulder, M. (1996). *Basic principles of membrane technology*. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Nieuwenhuijzen, A.F. van (2002). *Scenario Studies into Advanced Particle Removal in the Physical – Chemical Pre-treatment of Wastewater*. PhD thesis, Delft University of Technology, Delft, The Netherlands.

Park, C., Kim, H., Hong, S. and Choi, S-I. (2006). *Variation and prediction of membrane fouling index under various feed water characteristics*. Journal of Membrane Science, 284, 248 – 254.

Park, Y.G. (2002). *Effect for reducing membrane-fouling in the UF membrane.* Desalination, 147, 43 – 48.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Ravazzini, A.M. (2008). *Crossflow Ultrafiltration of Raw Muncipal Wastewater*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H., Wortel, N.C. and Daalen, R. van (2005). *New process for treatment of organically fouled water; experiences with WWTP-effluent*. Desalination, 178, 27 – 40.

Rosenberger, S., Evenblij, H., te Poele, S., Wintgens, T. and Laabs, C. (2005). *The importance of liquid phase analyses to understand fouling in membrane assisted activated sludge processes – six case studies of different European research groups.* Journal of Membrane Science, 263, 113 – 126.

Schafer, A.I., Fane, A.G. and Waite, T.D. (1998). *Nanofiltration of natural organic matter: removal, fouling, and the influence of multivalent ions*. Desalination, 118, 109 – 122.

Schrader, G.A. (2006). *Direct nanofiltration of wastewater treatment plant effluent*. PhD thesis, Universtiteit Twente, Enschede, The Netherlands.

Seo, G.T., Lee, T.S., Moon, B.h. and Lim, J.H. (2001). *Ultrafiltration combined with ozone for domestic laundry wastewater reclamation and reuse*. Water Science and Technology, 1, 5 – 6, 387 – 392.

Shon, H.K., Vigneswaran, S., Kim, S.I., Cho, J. and Ngo, H.H. (2004a). *The effect of pretreatment to ultrafiltration of biologically treated sewage effluent: a detailed effluent organic matter (EfOM) characterization*. Water Research, 38, 1933 – 1939.

Shon, H.K., Vigneswaran, S., Kim, S.I., Cho, J. and Ngo, H.H. (2004b). *Effect of pretreatment on the fouling of membranes: application in biologically treated sewage effluent*. Journal of Membrane Science, 234, 111 – 120.

Tarabara, V.V., Hovinga, R.M. and Wiesner, M.R. (2002). *Constant transmembrane pressure versus constant permeate flux: effect of particle size on crossflow membrane filtration.* Environmental Engineering Science, 19, 6, 343 – 355.

Tsujimoto, W., Kimura, H., Izu, T. and Irie, T. (1998). *Membrane filtration and pre-treatment of GAC*. Desalination, 119, 323 – 326.

Verberk, J.Q.J.C. (2005). *Application of air in membrane filtration*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

VROM (1996). *Discharge requirements municipal wastewater.* (in Dutch). Lozingenbesluit WVO Stedelijk Afvalwater 24 February 1996. Ministry of Housing, Spatial Planning and the Environment, Staatscourant, 1996:150

Vyas, H.K., Bennett, R.J. and Marshall, A.D. (2002). *Performance of crossflow microfiltration during constant transmembrane pressure and constant flux operations*. International Dairy Journal, 12, 473 – 479.

Zheng, X., Ernst, M. and Jekel, M. (2009a). *Identification and quatification of major oganic foulants in treated domestic wastewater affecting filterability in dead-end ultrafiltration*. Water Research. 43, 1, 238 - 244.

Zheng, X., Mehrez, R., Jekel, M. and Ernst, M. (2009b). *Effect of slow sand filtration of treated domestic wastewater as pre-treatment to UF.* Desalination, 249, 2, 591 – 595.

Zheng, X., Ernst, M, Huck, P.M. and Jekel, M. (2010). *Biopolymer fouling in dead-end ultrafiltration of treated domestic wastewater.* Water Research, 44, 18, 5212 - 5221.

3 Research methodology

3.1 Introduction

The research presented in this thesis was conducted at different WWTPs in The Netherlands. At two WWTPs (Horstermeer and Maasbommel) tests on pilot scale were performed with an ultrafiltration pilot installation. The feedwater of the ultrafiltration pilot installation at WWTP Horstermeer was prefiltered WWTP effluent. The filters, a multi media filter and 1-STEP® filter, were part of a research carried out by Witteveen+Bos Consulting Engineers, Waternet (the municipal water chain company of Amsterdam) and Delft University of Technology. At the WWTP Maasbommel an ultrafiltration pilot installation and a granulated activated carbon filter were operated by the local water board. The operation of these units was part of a research program carried out by the Foundation for Applied Water Research (STOWA). At the WWTP Sas van Gent research has been carried out on a full scale ultrafiltration – reverse osmosis (UF – RO) plant. This plant, operated by Evides Industriewater treats the effluent of WWTP Sas van Gent. At the WWTP Berkel, the nearest wastewater treatment plant to the laboratory of the department of Sanitary Engineering at Delft University of Technology, batch samples of WWTP effluent were taken for lab scale experiments.

This chapter describes the various wastewater treatment plants, the produced effluent quality and the pilot and full scale ultrafiltration installations. Furthermore the applied methods like the SUR measurement, fractionation procedure and physical – chemical water analyses are described.

3.2 Research locations

3.2.1 WWTP Horstermeer and effluent quality

The municipal WWTP Horstermeer has been in operation since 1985 and currently treats the wastewater of 140,000 inhabitants residing in the surrounding areas of Naarden-Bussum, Hilversum-West and Nederhorst den Berg. The average flow of the WWTP is 26,000 m^3/day. The treatment in this plant consists of grit removal, primary sedimentation, aeration and final sedimentation. The effluent is discharged into the river Vecht, part of the river Rhine basin district. Due to the actual River Vecht program and possibly more stringent future norms the effluent quality has to improve. Probably the future norms for total nitrogen and total phosphorus will become 2.2 mg N/L and 0.15 mg P/L respectively. Considering the mean effluent quality parameters of the WWTP Horstermeer as presented in Table 3.1 it is obvious that post treatment is needed to fulfil these norms. Therefore possibilities (multi media filter and 1-STEP® filter) for further improvement have been investigated by Witteveen+Bos Consulting Engineers, Waternet and Delft University of Technology.

Table 3.1 – Average effluent quality of the WWTP Horstermeer in 2006, 2007 and 2008 (Scherrenberg *et al.*, 2007; STOWA, 2009)

		Year	
		2006	2007 and 2008
Parameter	Unit	Concentration	Concentration
COD	mg/L	35	33
BOD	mg/L	3.9	4.2
N_{total}	mg/L	14.5	13.7
$N_{Kjeldahl}$-N	mg/L	3.7	3.0
NO_3-N	mg/L	10.0	10.3
P_{total}	mg/L	0.8	0.9
PO_4-P	mg/L	0.4	0.4
Suspended Solids	mg/L	11	11.3

3.2.1.1 Experiments

At the WWTP Horstermeer several experiments have been performed during different periods. In Table 3.2 an overview is shown and also the chapter presenting the respectively results. The procedures of the experiments are described separately in these chapters.

Table 3.2 – Overview of experiments performed at the WWTP Horstermeer

Period	Description of experiment	Chapter
April, 2006	The SUR of (prefiltered) WWTP effluent in relation to the operational flux of the ultrafiltration pilot installation.	4
August, 2007 and April – July, 2008	Effect of flux and prefiltration (multi media filter or 1-STEP® filter) on the fouling rate of the ultrafiltration pilot installation.	7
April – July, 2008	Effect of prefiltration (multi media filter or 1-STEP® filter) on the SUR value, foulants and fractions of WWTP effluent.	5

3.2.1.2 Pilot installations

The pilot installations at WWTP Horstermeer consisted of a multi media filter, 1-STEP® filter and ultrafiltration pilot installation (Figure 3.1). Both, the multi media filter and 1-STEP® filter, were supplied with WWTP effluent, which passed through a 450 µm (curved) sieve and was then collected in the WWTP effluent buffertank. The curved sieve was used in order to protect the installations from non separated organic and inorganic material from the secondary clarifier. After the WWTP effluent buffer the water was supplied to both filters. Coagulant (poly aluminium chloride) was dosed in-line and mixed with the WWTP effluent in a static mixer (slide valve). The carbon source for denitrification (mostly methanol) was dosed in-line, closely after the static mixer. The coagulant and methanol dosage was applied proportionally to the concentration of orthophosphate and nitrate measured in the WWTP effluent buffer. After filtration the filtrate of both filters, the feedwater of the ultrafiltration pilot installation, was collected in separate filtrate buffertanks. The ultrafiltration pilot installation was fed with either multi media filtrate or 1-STEP® filtrate.

Analyses were obtained for various grab samples: WWTP effluent (after passing the curved sieve), WWTP effluent after dosing of coagulant and carbon source (only for multi media filter), filtrate multi media filter, filtrate 1-STEP® filter and feedwater ultrafiltration pilot installation.

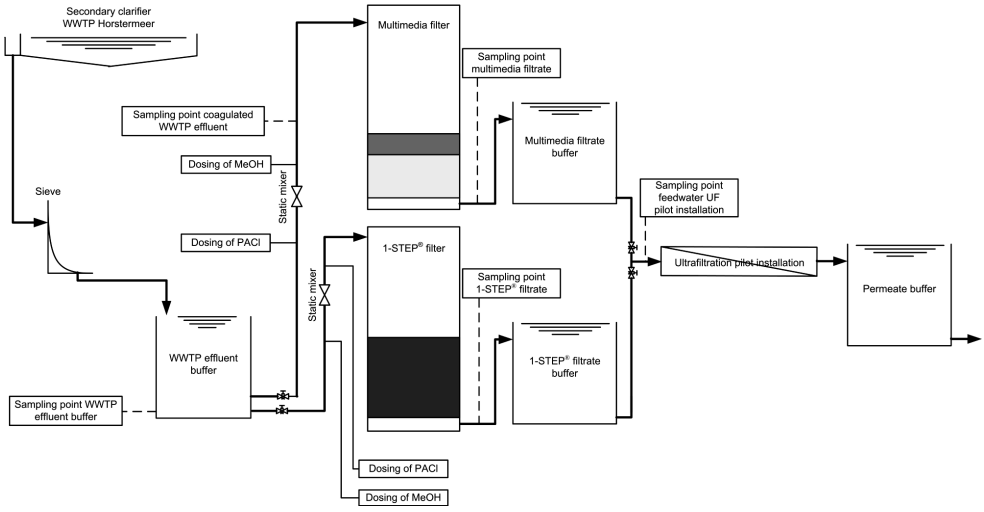


Figure 3.1 – Scheme of pilot installations at the WWTP Horstermeer

Multi Media Filter

In Figure 3.2 a picture of the used multi media filter, with a height of 4 meters and a diameter of 1 meter, is shown. The multi media filter is fed from the top with presieved and precoagulated (poly aluminium chloride) WWTP effluent. In addition to the coagulant, either methanol or acetic acid as a carbon source was dosed in-line for denitrification. The dosage of methanol or acetic acid was based on the actual nitrate and free oxygen concentrations in the WWTP effluent. The filtrate leaves the filter at the bottom and was collected in the multi media filtrate buffertank and formed the feedwater of the ultrafiltration installation. Additionally the collected filtrate was used to backwash the multi media filter.

Figure 3.2 – Multi media filter at the WWTP Horstermeer

During all experimental periods the multi media filter contained two filtration layers: upper layer, anthracite and the lower layer quartz sand. In Table 3.3 the properties of the filter media are shown during the different experimental periods.

Table 3.3 – Properties of the filter media of the upper and lower layer of the multi media filter at the WWTP Horstermeer

Period	Upper layer	Lower layer
April, 2006	Anthracite, 80 cm height, 2.0 – 4.0 mm grain diameter, 1,400 kg/m^3 density	Quartz sand, 40 cm height, 0,8 – 1.25 mm grain diameter, 2,600 kg/m^3 density
August, 2007	Anthracite, 80 cm height, 2.0 – 4.0 mm grain diameter, 1,400 kg/m^3 density	Quartz sand, 40 cm height, 1,25 – 1.5 mm grain diameter, 2,600 kg/m^3 density
April – July, 2008	Anthracite, 70 cm height, 2.0 – 4.0 mm grain diameter, 1,400 kg/m^3 density	Quartz sand, 80 cm height, 1,5 – 2.25 mm grain diameter, 2,600 kg/m^3 density

Cleaning of the filter bed was done by backwashing the filter. During a backwash the filter media expands, which causes scouring of the filter media and the removal of retained particles. At the WWTP Horstermeer for backwashing of the multi media filter two procedures were applied. The normal backwash procedure was applied approximately two times per day. The procedure of this backwash is shown in Table 3.4. Next to the normal backwash every three hours a ‘bumping cleaning’ was applied aiming to release the nitrogen and carbon dioxide gasses formed during denitrification. A bumping cleaning consists of two minutes backwash with filtrate at a rate of 50 m/h.

Table 3.4 – Backwash procedure of the multi media filter at WWTP Horstermeer during the experimental periods (Miska-Markusch, 2009; Scherrenberg 2011)

	Phase 1	Phase 2	Phase 3	Phase 4	Phase 5
April, 2006					
Time (s)	180	120	120	240	-
Air (-)	On[1]	Off	Off	Off	-
Water (-)	On	On	On	Off	-
Velocity (m/h)	5	30	50	10	-
August, 2007					
Time (s)	120	240	120	120	420
Air (-)	Off	On	Off	Off	Off
Water (-)	Off	Off	On	On	On
Velocity (m/h)	-	-	37.5	62.5	50
April – July, 2008					
Time (s)	120	240	120	120	420
Air (-)	Off	On	Off	Off	Off
Water (-)	Off	Off	On	On	On
Velocity (m/h)	-	-	37.5	62.5	50

[1)] blower operation: 20 Hz, 20 Nm3/h

The operational conditions like flow rate, filtration rate, filtration run, frequency bumping cleaning, methanol/acetic acid and poly aluminium chloride dosing ratio are reported separately in chapters 4, 5, and 7.

One Step Effluent Polishing (1-STEP®) filter

The One Step Effluent Polishing (1-STEP®) filter is a combination of a granulated activated carbon (GAC) filter and discontinuous media filter. It has been developed recently and operated on pilot scale by Witteveen+Bos Consulting Engineers, Waternet, Delft University of Technology and NORIT (STOWA, 2009). The media of the 1-STEP® filter is granulated activated carbon and operated for both, denitrification and simultaneous phosphorus removal. In addition the 1-STEP® filter is developed to achieve removal of priority compounds.

In Figure 3.3 a picture of the used 1-STEP® filter, with a height of 4 meters and a diameter of 1.13 meter, is shown. Like the multi media filter the 1-STEP® filter is fed from the top with presieved (450 µm) and precoagulated (poly aluminium chloride) WWTP effluent. Also either methanol or acetic acid was dosed for denitrification based on the actual nitrate and free oxygen concentrations in the WWTP effluent. The filtrate was collected in the 1-STEP® filtrate buffertank and formed the feedwater of the ultrafiltration installation.

Figure 3.3 – One Step Effluent Polishing (1-STEP®) filter at the WWTP Horstermeer

During the experimental period of April – July, 2008 the bed height of the granulated activated carbon was 1.9 meters. The properties of the granulated activated carbon are presented in Table 3.5.

Table 3.5 – Properties of the granulated activated carbon of the 1-STEP® filter at the WWTP Horstermeer

Medium characteristics	Unit	Value
Carbon type GAC	-	NORIT Vapure 612
Iodine number	mg/g	920
Apparent density	kg/m^3	510
Particle size	mm	1.70 – 3.35

Like the multi media filter the filter bed was flushed by applying two different types of backwash. The normal backwash procedure (Table 3.6) was applied approximately two times per day. In addition approximately every three hours a 'bumping cleaning' was applied with the same aim as described for the multi media filter. A bumping cleaning consisted of eight minutes backwash with filtrate at a rate of 15 m/h.

Table 3.6 – Backwash procedure of the 1-STEP® filter at the WWTP Horstermeer (STOWA, 2009)

	Phase 1	Phase 2	Phase 3	Phase 4	Phase 5
Time (s)	60	360	120	300	300
Air (-)	Off[1]	On	Off	Off	Off
Water (-)	On	Off	On	On	On
Velocity (m/h)	15	-	40	60	40

[1)] blower operation: 20 Hz, 20 Nm3/h

Ultrafiltration pilot installation

The ultrafiltration pilot installation operated at WWTP Horstermeer is displayed in Figure 3.4. This installation contained hydrophilic X-Flow membranes with a capillary diameter of 0.8 mm, a pore size of 20 – 30 nm (MWCO of 150 – 200 kDa) and prepared of a blend of polyethersulphone (PES) and polyvinylpyrrolidene (PVP). The installation is equipped with two 8-inch modules (S-225 FSCF PVC) with a length of 1.5 meter each and has a maximum capacity of 10 m^3/h. The total membrane area is 70 m^2. Further specifications of the pilot installation, membranes and modules are given in appendix A.

Figure 3.4 – Ultrafiltration pilot installation at the WWTP Horstermeer

During the experimental periods the installation was operated in dead-end mode at a constant flux. The permeate was collected in a permeate buffertank and used for backflushing and chemical cleaning of the membranes.

Interpretation of the ultrafiltration pilot installation data

The filtration data were logged every 10 seconds, thus providing the basis for a detailed visualisation and analysis of the filtration data. The following parameters were logged: time, flow (feedwater and backwash), temperature and pressure in the feedwater and permeate tubes. From the logged data the resistance of the filtration process was calculated according to the following equations:

$$R_t = \frac{\Delta P_{actualT}}{J_{actualT} \cdot \eta_{actualT}} \quad (3.1)$$

in which: R_t = total filtration resistance (m^{-1})
$\Delta P_{actualT}$ = trans membrane pressure at actual temperature (bar)
$J_{actual\ T}$ = flux at actual temperature ($L/m^2 \cdot h$)
$\eta_{actual\ T}$ = feedwater dynamic viscosity at actual temperature (Pa·s)

The membrane resistance of the used membranes type of the ultrafiltration membrane installation is approximately $0.8 \cdot 10^{12}$ m^{-1} (clean water permeability of 450 $L/m^2 \cdot h \cdot bar$ and feedwater temperature of 20 °C) (te Poele, 2005). At the start the initial filtration resistance is equal to the membrane resistance of a new membrane. Thereafter the start condition (filtration resistance) of subsequent filtration periods depends on the cleaning efficiency of the particular cleaning method.

The maximum trans membrane pressure for the membrane type used at the WWTP Horstermeer is 1 bar (te Poele, 2005). At the applied process conditions (constant flux and feedwater temperature), the maximum theoretical filtration resistance ($R_{max,th}$) can be calculated by equation 3.2. As shown, the $R_{max,th}$ depends on the applied flux and temperature of feedwater. The $R_{max,th}$ will decrease if the applied flux increases and/or the dynamic viscosity increases due to temperature decrease since the dynamic viscosity is related to temperature (see equation 1.3).

$$R_{max,th} = \frac{\Delta P_{max}}{J_{actualT} \cdot \eta_{actualT}} \quad (3.2)$$

in which: $R_{max,th}$ = maximum theoretical filtration resistance (m^{-1})
ΔP_{max} = maximum trans membrane pressure (1 bar)
$J_{actual\ T}$ = flux at actual temperature ($L/m^2 \cdot h$)
$\eta_{actual\ T}$ = dynamic viscosity of feedwater at actual temperature (Pa·s)

From the obtained data of the ultrafiltration pilot installation the filterability is expressed as fouling rate (dR/dt) and can be calculated within a filtration period. This is presented in Figure 3.5 and equation 3.3. By using this method, filterability of different feedwaters can be easily compared. The filterability, dR/dt, is expressed in m^{-1} per hour.

$$\frac{dR}{dt} = \frac{R_b - R_a}{t_b - t_a} \quad (3.3)$$

The reversibility is determined by ΔR/Δt, which is the increase of the filtration resistance over time after a series of filtration periods including back flushes and is measured by comparing the filtration resistance at the start of the first filtration period to the filtration resistance at the start of a filtration period after a serie(s) of filtration periods and is illustrated in Figure 3.5 and Equation 3.4. The reversibility is expressed in m^{-1} per hour. A high value for ΔR/Δt indicates a low reversibility, whereas a high reversibility is indicated by a low value for ΔR/Δt.

$$\frac{\Delta R}{\Delta t} = \frac{R_c - R_a}{t_c - t_a} \tag{3.4}$$

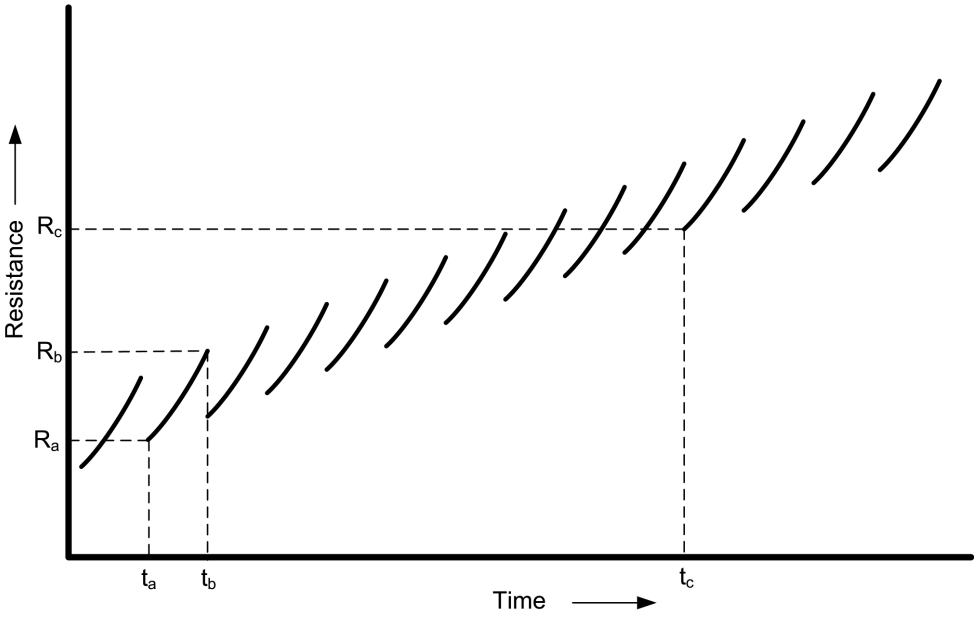


Figure 3.5 – Determination of the filterability (dR/dt) and reversibility (ΔR/Δt) of the ultrafiltration pilot data at WWTP Horstermeer

3.2.2 WWTP Maasbommel and effluent quality

The WWTP Maasbommel, owned and operated by the Waterboard Rivierenland, is situated in an agricultural area in The Netherlands and treats the wastewater of approximately 7,400 inhabitants (Van Betuw *et al*., 2007). The wastewater treatment plant consists of an influent screen (6 mm), an oxidation ditch and a secondary clarifier. No additional chemicals are dosed for nitrate or phosphate removal but since 2003 the remaining nutrients in the secondary clarifier effluent are removed by tertiary treatment. The tertiary treatment consist of a 2-step continuous sand filtration. The first sand filter functions to remove biologically nitrogen and the second filter has the function to remove phosphate and suspended solids. The

effluent of the tertiary treatment is discharged to a rural (agricultural) area with also a variety of recreational activities. But during the experiments at WWTP Maasbommel effluent was taken after the secondary clarifier. Therefore in Table 3.7 the characteristics of effluent after the secondary clarifier are shown.

Table 3.7 – Characteristics of the secondary clarifier effluent at WWTP Maasbommel during the period April, 2006 – April, 2007 (STOWA, 2007)

Parameter	Unit	Concentration
COD	mg/L	30
BOD	mg/L	<5
$N_{Kieldahl}$-N	mg/L	2.5
N_{total}	mg/L	6
P_{total}	mg/L	2

3.2.2.1 Experiments

At the WWTP Maasbommel during one period (February – May, 2007) two different experiments were performed simultaneously. The purpose of one of the experiments was to examine the effect of powdered activated carbon (PAC) addition on the filterability, concentration of foulants and fractions of WWTP effluent. This experiment was performed with an ultrafiltration pilot installation. At the same time, but in a different experiment, the effect of granulated activated carbon (GAC) filtration on the filterability, concentration of foulants and fractions of WWTP effluent was investigated. The results of both experiments are presented in chapter 5.

3.2.2.2 Pilot installations

The pilot installations at the WWTP Maasbommel consisted of a granulated activated carbon (GAC) filter and an ultrafiltration pilot installation (Figure 3.6). The GAC filter was supplied with WWTP effluent, which passed a screen of 5 mm. The feedwater of the ultrafiltration pilot installation passed in addition to the screen of 5 mm a sieve of 0.7 mm. Both, sieve and screen, were applied to protect the pilot installations from non separated organic and inorganic material from the secondary clarifier.

During the operation of the ultrafiltration pilot installation Nalmet® 8149 (hereafter Nalmet) and powdered activated carbon had been dosed to remove heavy metals and residual organic compounds (COD, herbicides, toxicity and endocrine disrupting compounds) respectively of the WWTP effluent. Nalmet is a polymeric substance consisting of sulphide and hydroxide groups to which dissolved and complex bounded metals attach (Geilvoet, 2007).

The GAC filter was operated without addition of any chemical. The filtrate was not collected in a buffertank but directly discharged to the sewage system of the WWTP Maasbommel.

During both experiments grab samples were taken at different places of the pilot installations (see Figure 3.6): WWTP effluent (after passing the first screen), settling tank (flocculation) of ultrafiltration pilot installation, fourth tank (powdered activated carbon) of ultrafiltration pilot installation and filtrate GAC filter.

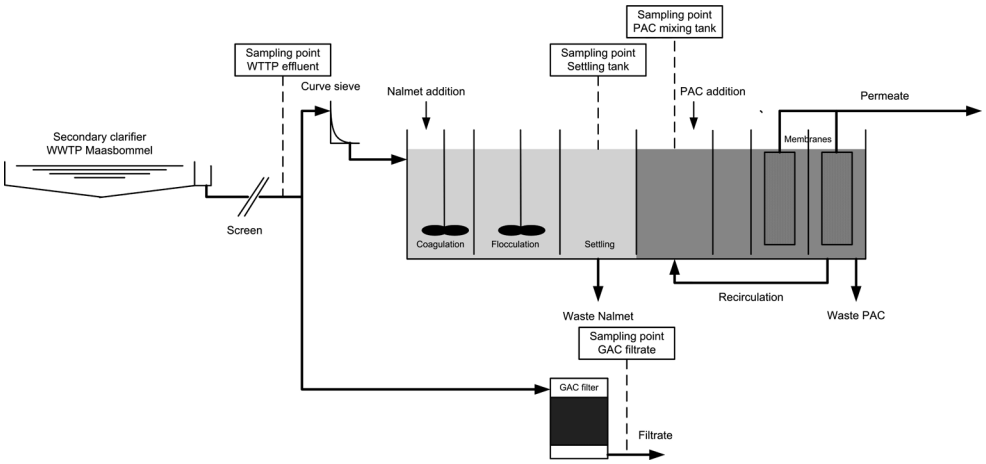


Figure 3.6 – Scheme of pilot installations at the WWTP Maasbommel

Ultrafiltration pilot installation

The ultrafiltration pilot installation (part of Figure 3.6) consisted of seven internally connected tanks with different volumes. In the first tank Nalmet was dosed to remove heavy metals. After mixing (coagulation and flocculation) in the first two tanks, the third tank is used for settling and discharging of suspended material. In the fourth tank powdered activated carbon is added and mixed. Finally in the last two tanks the treated WWTP effluent is extracted by submerged ultrafiltration membranes. In Table 3.8 the properties of the ultrafiltration pilot installation are given. The operational parameters during the experimental period are given in chapter 5.

Table 3.8 – Properties of the ultrafiltration pilot installation at the WWTP Maasbommel (STOWA, 2007)

Parameter	Unit	Value
Coagulation/Flocculation reactors		
Volume coagulation tank	m^3	8
Volume flocculation tank	m^3	9
Volume settling tank	m^3	9
Polymer type	-	Nalmet® 8149
Powdered activated carbon reactor		
Total volume contact tanks[1]	m^3	20
Carbon type PAC	-	Norit SAE SUPER
Activation method	-	Steam
Iodine number	-	1150
Methylene blue adsorption	g/g	0.28
Total surface area (BET)	m^2/g	1300
Apparent density	kg/m^3	425
Particle size > 150 µm	weight %	3
Particle size D50	µm	15
Ultrafiltration membranes		
Volume membrane tank	m^3	6
Membrane type	-	Zeeweed® ZW500-c-11
Membrane pore size	µm	0.1
Membrane surface area[2]	m^2	220

[1)] Including volume membrane tank.
[2)] One membrane module has been applied.

Granulated activated carbon filter

In Figure 3.7 the GAC filter with a diameter of 0.75 meter is shown. The secondary clarifier effluent is sieved (5 mm) only before it entered the GAC filter from the top. The filtrate leaves the filter at the bottom and was not collected in a filtrate buffer. Due to the absence of a filtrate buffer the GAC filter was backwashed once a week with process water of the wastewater treatment plant. During this weekly backwash the bed expansion was about 25%.

Figure 3.7 – Granulated activated carbon filter at the WWTP Maasbommel

The properties of the applied granulated activated carbon are given in Table 3.9. The process conditions during the experiment with the GAC filter are described in the experimental set up section of chapter 5.

Table 3.9 – Characteristics of the activated carbon of the GAC filter at the WWTP Maasbommel (Van Betuw *et al.*, 2007)

Parameter	Unit	Value
Carbon type GAC	-	NORIT GAC 830P
Activation methode	-	Steam
Iodine number	mg/g	1050
Total surface area	m^2/g	1150
Apparent density	kg/m^3	480
Particle size D10	mm	1.0

3.2.3 WWTP Sas van Gent and effluent characteristics

The UF-RO plant at the WWTP Sas van Gent is operated by Evides Industriewater (industrial water supply company) and produces demiwater from industrial effluent of a food producing factory. The wastewater treatment plant of this factory consists of an anaerobic (UASB) reactor and an oxidation ditch. After final sedimentation in a secondary clarifier the effluent is transported to the UF-RO plant. The average quality of the (industrial) effluent in 2006 measured by the operators is shown in Table 3.10. Compared to effluent of a municipal WWTP almost all values are higher and in particular the temperature and electrical conductivity but these differences are related to the origin (food producing factory) of the wastewater.

Table 3.10 – The average effluent quality of the WWTP Sas van Gent in 2006

Parameter	Unit	Value
Temperature	°C	32
pH	-	8.3
Suspended solids	mg/L	12.2
Turbidity	NTU	23.1
Electrical conductivity	µs/cm	5,090

3.2.3.1 Experiments

At the WWTP Sas van Gent during one period (August – December, 2006) two experiments have been performed. One experiment is described in chapter 4 and presents the relation between the SUR value and increase of resistance of the ultrafiltration units of the UF-RO plant. The other experiment is drawn in chapter 6. The aim of this experiment was to evaluate the performance of pretreatment and ultrafiltration units during the intake of WWTP effluent.

3.2.3.2 UF – RO plant

In Figure 3.8 a process flow diagram of the UF-RO plant at WWTP Sas van Gent is shown. As illustrated the effluent of WWTP Sas van Gent is pretreated before it enters the ultrafiltration units. First ferric chloride (2.5 mg Fe^{3+}/L) is dosed in-line and flocculated above the filter bed of the dual media filter. The dual media filter consists of a layer anthracite and sand. The main properties of the filter and filter media are shown in Table 3.11.

Table 3.11 – Properties of the dual media filter of the UF-RO plant at WWTP Sas van Gent

Parameter	Unit	Value
Surface area	m^2	18
Anthracite (upper layer)		
Density	kg/m^3	1400
Bedheight	m	0.8
Fraction size	mm	2.5 – 4.0
Sand (lower layer)		
Density	kg/m^3	2600
Bedheight	m	0.7
Fraction size	mm	1.4 – 2.0

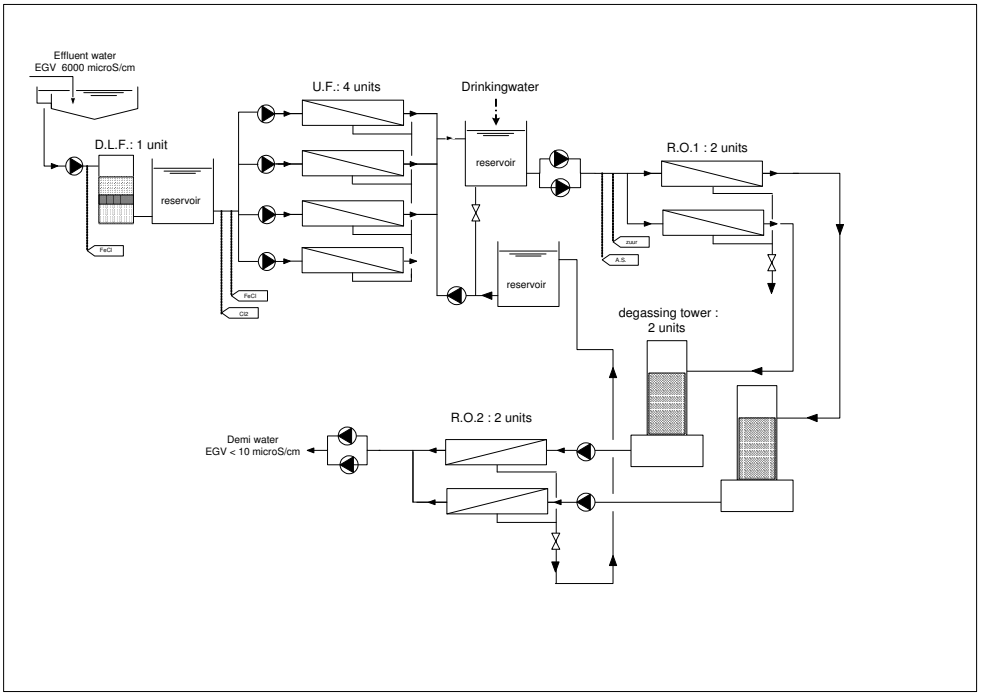


Figure 3.8 – Process flow diagram of the UF-RO plant at WWTP Sas van Gent (Evides Industriewater, 2005)

The filtrate of the dual media filter is collected in a buffertank. The filtrate is the feedwater of the ultrafiltration units but is also applied to backwash the dual media filter. The dual media filter is backwashed under normal conditions three or four times per day. The applied backwash procedure is given in Table 3.12.

Table 3.12 – Backwash procedure of the dual media filter of the UF-RO plant at WWTP Sas van Gent

	Phase 1	*Phase 2*
Time (s)	300	540
Air (-)	On[1]	Off
Water (-)	Off	On
Velocity (m/h)	60	60

[1)] blower operation: 1080 Nm^3/h

From the filtrate buffertank the water is pumped to the ultrafiltration units. Before it enters these ferric chloride (1 mg Fe^{3+}/L) is dosed in order to increase the filtration performance. Beside the ferric chloride sodium hypochlorite is dosed to prevent biological growth in the ultrafiltration units.

The ultrafiltration part of the UF-RO plant consists of 4 ultrafiltration units with a total membrane area of 3,120 m^2. The ultrafiltration units are operated at a fixed production flow of approximately 80 m^3/h. This means the flux of each filtration unit is not constant but depends on the production flow (flux) of the other units. Therefore the flux of each unit can vary between 20 – 60 L/m^2·h. In Table 3.13. the properties of the ultrafiltration units are summarized. Further particulars are given in chapter 4 and 6.

Table 3.13 – Properties of the ultrafiltration units of the UF-RO plant at WWTP Sas van Gent

Parameter	Unit	Value
Ultrafiltration units 1 and 2		
Surface area per unit	m^2	720
Number of membrane modules per unit	-	18
Membrane type	-	Norit UFCM5LE (40 m^2)
Ultrafiltration units 3 and 4		
Surface area per unit	m^2	840
Number of membrane modules per unit	-	24
Membrane type	-	Norit UFCM5 (35 m^2)

As shown in Figure 3.8, the permeate of the ultrafiltration units is collected in an ultrafiltration permeate buffer tank. The ultrafiltration permeate is further polished by the reverse osmosis membranes in two steps. The concentrate of the first reverse osmosis (RO1) is discharged to the surface water and the permeate is fed to the second reverse osmosis (RO2) after degassing. The concentrate of the second reverse osmosis is used to backflush the ultrafiltration units. As the reverse osmosis is beyond the scope of this thesis the properties of are not presented in this section.

3.2.4 WWTP Berkel and effluent characteristics

The WWTP Berkel is designed for 27,000 population equivalents. The wastewater is treated in an oxidation ditch and after final sedimentation the effluent is discharged into the surface water. In Table 3.14 the effluent characteristics after final sedimentation are given.

Table 3.14 – Characteristics of the effluent of WWTP Berkel in 2002 (Roorda, 2004)

Parameter	Unit	Concentration
COD	mg/L	38.8
BOD	mg/L	2.9
Suspended solids	mg/L	5.5
N_{total}	mg/l	1.6
P_{total}	mg/L	7.3

3.2.4.1 Experiments

At the WWTP Berkel no pilot or full scale experiments were done but at various times grab samples of the effluent were taken for lab scale experiments in the laboratory of Sanitary Engineering. The results of the experiments with effluent of WWTP Berkel are presented in chapters 5 and 7.

3.3 Specific Ultrafiltration Resistance

Roorda (2004) developed a parameter for the evaluation of filtration characteristics during dead-end ultrafiltration of WWTP effluent and proposed the Specific Ultrafiltration Resistance as a parameter. This parameter provides useful information about the filterability of the WWTP effluent and can be measured in a short period of 30 minutes. The SUR is described as the cake layer resistance per unit of filtered feedwater per m^2 membrane area and is expressed in m^{-2}. Low SUR values indicate relatively high filterability, whereas high SUR values indicate the opposite (rapid membrane fouling and poor filterability).

In this section the theoretical description, the experimental set-up and the measuring procedure are described.

3.3.1 Theoretical description

As shown and expressed in section 2.3.3 during dead-end ultrafiltration of WWTP effluent the membrane resistance is assumed as constant but the cake resistance increases due to the retaining of particles within the cake layer. The assumptions of constant membrane resistance (equation 2.2) and increase of cake resistance proportional to the amount of particles delivered to the membrane surface (equation 2.3) can be combined and integrated ($t_0 = 0$, $T_t = t$, $V_0 = 0$, $V_t = V$) in one equation and a relation for t/V and V can be derived assuming constant trans membrane pressure and temperature. Equation 3.5 presents the derivation resulting in a linear relationship between t/V and V, depending on trans membrane pressure and viscosity (temperature).

$$\frac{t}{V} = \frac{\eta_T \cdot R_m}{\Delta P \cdot A_m} + \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P \cdot A_m^{\,2}} \cdot V \qquad (3.5)$$

in which:

t	=	time (s)
V	=	filtered volume (m^3)
η_T	=	dynamic viscosity ($N \cdot s/m^2$ or $Pa \cdot s$)
R_m	=	membrane resistance (m^{-1})
ΔP	=	trans membrane pressure (N/m^2 or Pa)
A_m	=	membrane area (m^2)
α_{av}	=	average specific cake resistance (m/kg)
c_v	=	solids concentration in feedwater (kg/m^3)

During dead-end ultrafiltration of WWTP effluent all parameters, except the average specific cake resistance (α_{av}) and the solids concentration in the feedwater (c_v), are known or can be measured. Therefore the slope of the curve (t/V versus V) can be used to calculate the product of average specific cake resistance and the solids concentration in the feedwater ($\alpha_{av} \cdot c_v$). This is graphically shown with a theoretically characteristic filtration curve in Figure 3.15. This

figure distinguishes three filtration mechanisms at constant trans membrane pressure; initially pore blocking occurs, which is followed by cake filtration and finally cake filtration with compression of the cake layer (Roorda, 2004).

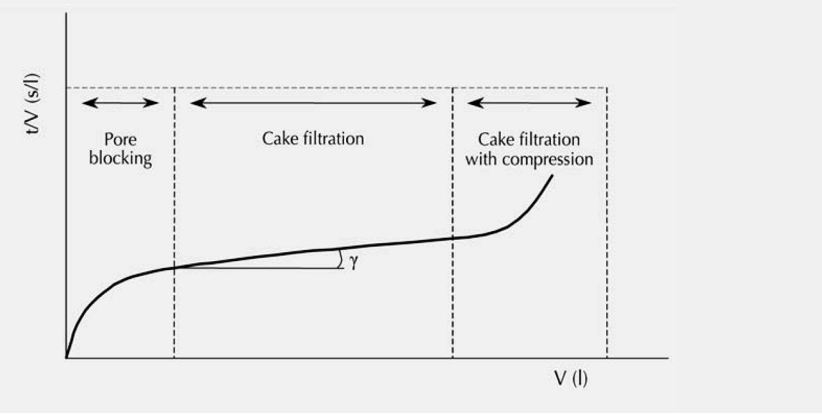


Figure 3.15 – Picture of three theoretically occurring filtration mechanism (pore blocking, cake filtration and cake compression) during dead-end ultrafiltration at constant pressure; tan γ is used to calculate the Specific Ultrafiltration Resistance (Roorda, 2004)

The SUR (m^{-2}) expressed in equation 3.6 is defined as the product of the average specific cake resistance (α_{av}) and the solids concentration of the feedwater (c_v).

$$SUR = \alpha_{av} \cdot c_v = \frac{d\left(\frac{t}{V}\right)}{d(V)} \cdot \frac{2 \cdot \Delta P \cdot {A_m}^2}{\eta_T} \tag{3.6}$$

When the SUR is measured cake filtration is assumed as the dominant filtration mechanism. To prove this assumption theoretical information about the fouling mechanisms during constant pressure dead-end filtration experiments can be analyzed based on the blocking laws summarized by Hermia (1982).

$$\frac{d^2t}{dV^2} = k \cdot \left(\frac{dt}{dV}\right)^{\beta} \tag{3.7}$$

in which: t = operation time (s)
V = cumulative permeate volume (m^3)
k = fluid characterisation constant ($s^{1-\beta} \cdot m^{3\cdot(\beta-2)}$)
β = constant that indicates fouling mechanism (-)

When equation 3.7 is drawn in log-scale graph, the slope of the curve is equal to the β-value, and four major fouling mechanisms can be theoretically derived. In Table 3.15 the β-values for the various filtration mechanisms are presented.

Table 3.15 – β-values for various filtration mechanism (Hermia, 1982)

Blocking filtration low	β
Complete blocking	2
Standard blocking	1.5
Intermediate blocking	1
Cake filtration	0

Roorda (2004) analysed the filtration mechanism according to equation 3.3 for dead-end ultrafiltration of WWTP effluent with and without pretreatment. It showed that β-values are in the same order of magnitude as the theoretical values. The values did not clearly indicate one filtration mechanism, but β-values were mostly found between 0.0 and 1.0. This indicates that in most cases the cake filtration law could be used for description of the filtration mechanism (Roorda, 2004).

3.3.2 Experimental setup

In Figure 3.16 the experimental setup of the SUR equipment is shown. The equipment consists of two pressurized vessels (B and C) with a volume of 10 liters. One pressure device is filled with demiwater (B), the other (C) is filled with the feedwater from which the filtration curves and the SUR are calculated. These vessels are connected to a three-way valve (E), so the only choices are demiwater or feedwater. Both vessels are connected to a pressure device (A). On the permeate side of the membrane, the pressure is equal to atmospheric pressure. The pressure difference over the membrane (H), indicated as the trans membrane pressure, is equal to 0.5 bar. Connector G, which is a valve, is used to connect the membrane module with the tube of the three-way valve (E). With the valve J, the filtration mode of the membrane can be changed from cross flow to dead-end. The permeate of the membrane is collected in an erlenmeyer on a mass balance (I). Both the mass balance and the manometer (F) are connected to computer (K), wich registers the mass of the Erlenmeyer and the pressure difference continuously over time. The filtration is carried out in a dead-end mode and takes 30 minutes. The temperature of the feedwater must be adjusted to 20 °C.

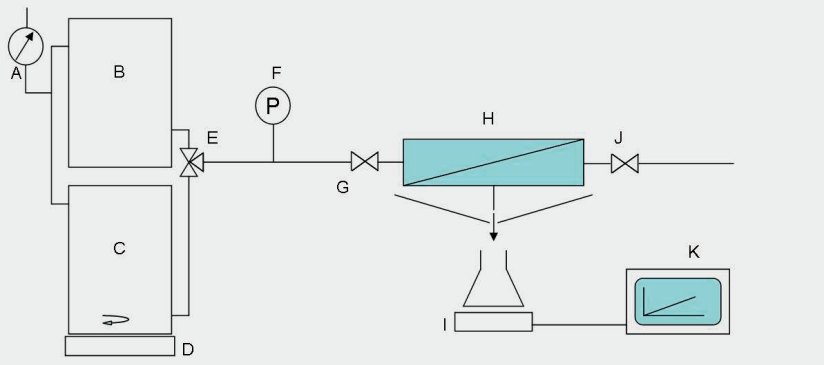


Figure 3.16 – SUR equipment containing; A pressure valve, B demiwater vessel, C wastewater vessel, D Magnetic stirrer and E three-way valve, F digital pressure sensor, G connector, J valve to change the mode of the membrane, H membrane module, I mass balance with Erlenmeyer and K computer with measuring program (Roorda, 2004)

One of the important aspects of the SUR equipment is the membrane module. This module contains three capillary ultrafiltration membranes of Norit X-Flow with an internal diameter of 0.8 mm, a MWCO of 150 – 200 kDa and prepared of PES (PVP). One membrane module provides a membrane area of about $10 \cdot 10^{-4}$ m^2. The same type of membranes were used in the ultrafiltration pilot installation at the WWTP Horstermeer and the full scale UF-RO plant at the WWTP Sas van Gent.

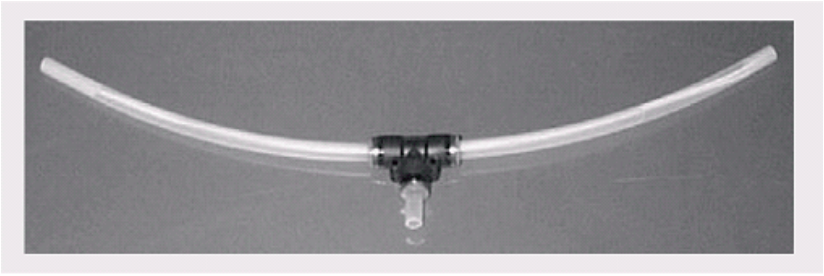


Figure 3.17 – Standard membrane module of the SUR equipment (Roorda, 2004)

3.3.3 Experimental procedure

Before a new membrane module is used it is first of all soaked for 30 minutes in a 400 mg/L sodium hypochlorite (NaOCl) solution. After soaking the sodium hypochlorite solution was flushed out by demiwater and then the clean water flux (CWF) is measured in order to determine the properties of the membrane module. The CWF of a new membrane module has to be in the range of 300 – 400 L/m^2·h at a trans membrane pressure of 0.5 bar and temperature of 20 °C. After the determination of the membrane properties the filtration with WWTP effluent starts. The filtration period with WWTP effluent is 30 minutes and after this period the membrane module is firstly cross flow flushed with demiwater and then chemically cleaned with a solution of 400 mg/L sodium hypochlorite or 1.25 w/w-% Divos 120CL (chemical membrane cleaning agent of JohnsonDiversey) at 40 °C. The temperature was increased in order to enhance the cleaning rate. The chemical cleaning procedure consist of first flushing and then soaking for approximately 10 minutes. After chemical cleaning the properties of the membrane module are checked again by filtrating demiwater at 20 °C during 15 minutes. If the CWF is changed less than 10% compared to the initial value the membrane module was (re)used in the next test. If not, a new membrane module is taken and used for the next SUR tests. When the membrane modules are not in use, they are stored in a sodium hypochlorite solution of 400 mg/L.

An example of the filtration curve when the SUR is measured is shown in Figure 3.17, where a screenshot of the SUR measurement program is shown. The three lines shown are the flux, trans membrane pressure and the resistance.

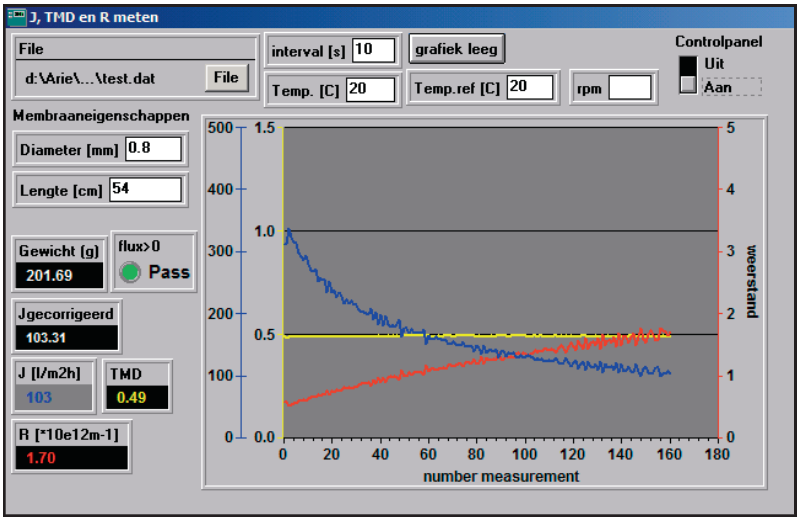


Figure 3.17 – Registration of flux, trans membrane pressure and resistance for determining the SUR (Geilvoet, 2007)

Given the measured mass (m) and the density of the water (ρ) the filtrated volume (V) can be calculated. The filtration time divided by the filtrated volume (t/V) can be plotted against the filtrated volume (V) in a graph. An example is shown Figure 3.18. The tan γ is used to calculate the SUR value. Roorda (2004) recommended to measure the SUR value between 10 and 30 minutes of filtration in order to obtain stable SUR values. In the first minutes of filtration the SUR was mostly less stable because of the development of a cake layer. After ten minutes a stable cake layer was formed.

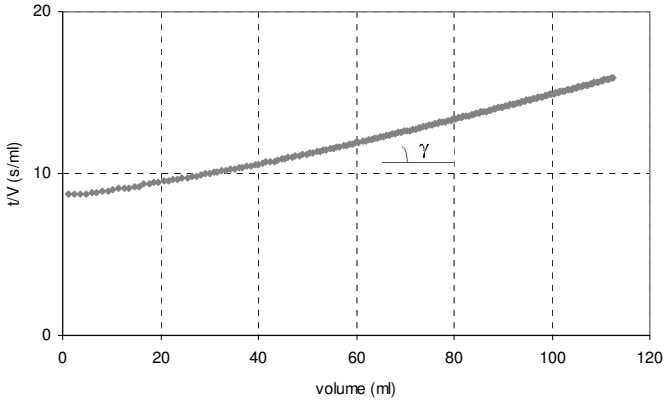


Figure 3.18 – Relation between the ratio of filtration time (t) to the filtrated volume (V) and volume (V)

The accuracy of the SUR measurement has been determined by Roorda (2004).These results showed an accuracy of more than 95% (Roorda, 2004). This observation was verified during tests performed in April, 2006. During these tests the accuracy of the SUR measurement was determined with effluent of WWT Berkel and Horstermeer. One sample of each WWTP was measured three times with a different membrane module. These determinations resulted in SUR values and variations of $8.4 \cdot 10^{12}$ $m^{-2} \pm 0.14 \cdot 10^{12}$ m^{-2} (WWTP Berkel) and $10.7 \cdot 10^{12}$ $m^{-2} \pm 0.28 \cdot 10^{12}$ m^{-2} (WWTP Horstermeer). Based on these results it was concluded that the filterability of WWTP effluent can be determined accurately with the SUR measurement as stated by Roorda (2004).

3.4 Fractionation

Fractionation of WWTP influent was applied by van Nieuwenhuijzen (2002) and fractionation of WWTP effluent by Roorda (2004) and te Poele (2005). In this thesis as well as in Roorda (2004) and te Poele (2005) the fractionation method was used to investigate the influence of different fractions of WWTP effluent on the total filterability of WWTP effluent.

The fractionation experiments were carried out in the laboratory of Sanitary Engineering at Delft University of Technology. Four fractions are formed by fractionation. The first fraction was either formed by a curved sieve with a mash size of 450 µm (WWTP Horstermeer) or by filtration over a sieve with a mash size of 400 µm (WWTP Maasbommel and WWTP Berkel). The main purpose of the first fraction was to remove non separated organic and inorganic material from the secondary clarifier of the WWTPs. The other fractions were formed by filtrating under vacuum the filtrate of the first fraction over successively 0.45 µm cellulose acetate (CA), 0.2 µm cellulose acetate and 0.1 µm cellulose nitrate (CN) of Sartorius. This fractionation method resulted in four different fractions:

- > 0.45 µm
- 0.45 – 0.2 µm
- 0.2 – 0.1 µm
- <0.1 µm.

The fractionation experiments can be generally executed in two ways: direct filtration and step by step filtration. The two methods are illustrated in Figure 3.19 Van Nieuwenhuijzen (2002) used the direct filtration method. All water in that research was first filtered over a 63 µm sieve and successively over a 7-8 µm paper filter. After the paper filter direct filtration was applied over four different filters. Roorda (2004) and te Poele (2005) used the step by step filtration method. A disadvantage of direct filtration could be the formation of a cake layer on the filters, especially for the filters with the smaller pore sizes. When this occurs, only particles which are much smaller than for example 0.1 µm could get through a 0.1 µm CN filters.

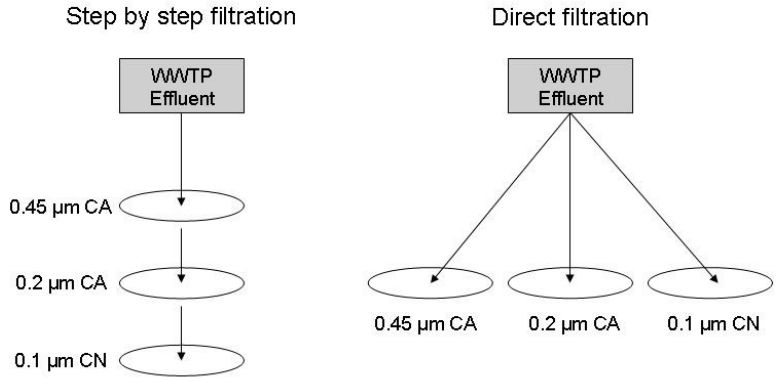


Figure 3.19 – Two different fractionation methods; step by step and direct filtration (Geilvoet, 2007)

To check the difference between the two fractionation methods one sample of WWTP Berkel (April, 2007) was fractionated, using the two methods and subsequently all the SUR values of the fractions were determined. The results are shown in Figure 3.20.

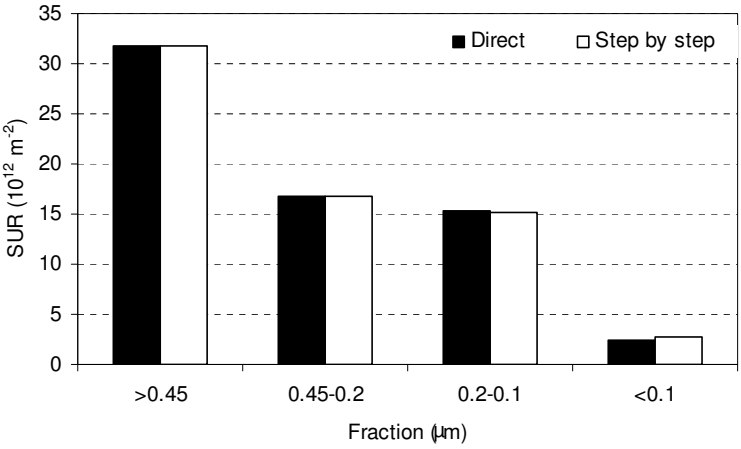


Figure 3.15 – SUR values of different fractions of effluent WWTP Berkel applying two different fractionation methods: direct fractionation and step by step fractionation

From Figure 3.20 it is concluded that the applied fractionation method would not have made a difference. For step by step filtration the SUR is slightly lower in the 0.2 – 0.1 µm fraction, compared with the direct filtration ($15.1 \cdot 10^{12}$ m^{-2} versus $15.3 \cdot 10^{12}$ m^{-2}), but slightly higher in the < 0.1 µm fraction. These differences fall within the standard deviation of 5%. Therefore the same fractionation method (step by step) as Roorda (2004) and te Poele (2005) was applied. To minimize clogging of the filters maximally 200 ml of sample was filtered with one filter.

One other aspect that could influence the results, is applying a constant pressure with the fractionation. Roorda (2004) used vacuum for every fraction, except for the 0.1 µm fraction. This last separation step was filtered under a constant pressure of 2.0 bar. Both Van Nieuwenhuijzen (2002) and te Poele (2005) do not mention the use of constant pressure. It is not applied for the 0.1 µm fraction in this research either.

3.5 Physical-chemical analyses

3.5.1 Dissolved organic carbon

The TOC Cell Test of Spectroquant® (Category number 1.14878.0001) with a measuring range of 5.0 – 80.0 mg/L is used to analyse the DOC concentration. The TOC analysis is conducted on the filtrate over a 0.45 µm cellulose acetate filter (Sartorius). Therefore the measured value corresponds to dissolved organic carbon (DOC). Before filtration of the sample first the filter was flushed with demiwater to remove (organic) residuals. The amount of DOC is expressed in mg/L.

3.5.2 Colour

For the determination of colour the NEN 1056 11.2 method was used. The absorption of colour at 455 nm in a 4 cm quartz glass cuvette is measured by spectrophotometer (Milton Roy spectromic 401). The amount of colour is expressed in mg Pt/L. Because light absorption is used for this measurement, the sample is filtrated over 0.45 µm (Sartorius, cellulos acetate) before determination. Otherwise non dissolved particles could block the beam of light resulting in inaccurate measurements.

3.5.3 Humic substances

Humic substances are characterised by UV-absorption at 254 nm. UV-absorption is measured at 254 nm in a 1 cm quartz glass cuvette by a UV-VIS spectrophotometer (Perking-Elmer Lambda 16). The amount of humic substances is expressed in cm^{-1}. Because UV light is used for this measurement, the sample is filtrated over 0.45 µm (Sartorius, cellulose acetate).

3.5.4 Polysaccharides

The sample for polysaccharides analyses is obtained after filtration over a 0.45 µm filter (Sartorius, cellulos acetate). The applied measuring method is in accordance with the variation of te Poele (2005) on the Rosenberg's method (Rosenberger, 2003). The colorimetric is based on the method of Dubois *et al*. (1956). The absorption of the formed colour is measured at 487 nm in a 4 cm glass cuvette. The amount of polysaccharides is expressed in mg/L. A detailed description of the method is given in appendix B.

3.5.5 Proteins

For the analysis of proteins the improved method of te Poele (2005) is used. Like the other analyses the sample is filtrated over a 0.45 µm filter (Sartorius, cellulos acetate) before analyses. The analysis method is a variant of the method of Rosenberger (2003), also a modified form of the method of Lowry *et al*. (1951). Absorption of the formed colour is measured at 750 nm in a 4 cm glass cuvett. The amount of proteins is expressed in mg/L. A detailed description of the method is given in appendix B.

3.5.6 PUVA

To compare different water samples te Poele (2005) introduced a new parameter: the ratio of ultraviolet absorption by humic substances to the amount of proteins (PUVA) which is shown in Equation 3.8.

$$PUVA = \frac{UV_{254}}{C_{proteins}} \tag{3.8}$$

in which: UV_{254} = UV-absorptions at 254 nm (m^{-1})
$C_{proteins}$ = concentration of proteins by Lowry *et al*. (1951) (mg/L).

Low values for PUVA (L/m·mg) indicate a high concentration of proteins, whereas high values are a symptom of a (rather) high concentration of humic substances (te Poele, 2005).

References

Betuw, W. van, Kruit, J., Segers, J., Piron, D. and Uijterlinde, C.A. (2007). *Opportunities for Effluent Reuse at Municipal Wastewater Treatment Plant Maasbommel*. Proceedings 6th IWA Specialists Conference on Wastewater Reclamation and Reuse for Sustainability 2007, October 9-15, Antwerp, Belgium.

Dubois, M., Gilles, K.A., Hamilton, J.K., Rebers, P.A. and Smith F. (1956). *Colourmetric method for determination of sugars and related substances*. Analytical Chemistry, 28, 350-356.

Evides Industriewater/Grontmij, *Basisgegevens DECO*, Internal document (in Dutch), Middelburg, 2005.

Geilvoet, A.J. (2007). *Fouling modification during pretreatment for dead-end ultrafiltration of wwtp effluent.* MSc thesis. Delft University of Technology, Delft, The Netherlands.

Lowry, O.H., Rosebrough, N.J., Farr, A.L. and Randall, R.J. (1951). *Protein measurement with the Folin phenol reagent*. Journal of Biological Chemistry, 193, 265-275.

Miska – Markusch, V. (2009). *Effluent filtration for more than particle removal.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Nieuwenhuijzen, A.F. van (2002). *Scenario Studies into Advanced Particle Removal in the Physical – Chemical Pre-treatment of Wastewater*. PhD thesis, Delft University of Technology, Delft, The Netherlands.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands

Rosenberger, S. (2003). *Charkterisierung von belebtem Schlamm in Membranbelebungsreaktoren zur Abwasserreiniging*. PhD thesis, Technical University of Berlin, Berlin, Germany.

Scherrenberg, S.M., Menkveld, H.W.H., Neef, R. and Graaf, J.H.J.M. van der (2007). *Comparison of dual media filtration with continuous sand filtration*. Proceedings 10th IWA Specialised Conference on Design, Operations and Economics of Large Wastewater Treatment Plants, Vienna, Austria.

Scherrenberg, S.M. (2011) *Reaching ultra low phosphorus concentrations by* filtration techniques. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

STOWA (2007). *Effluentnabehandeling op de rwzi Maasbommel*, in Dutch, Report 17, Utrecht, The Netherlands.

STOWA (2009). *1-STEP® filter als effluentpolishingstechniek (in Dutch)*. Report 34, Utrecht, The Netherlands.

4 Filtration properties and performance of ultrafiltration installations

4.1 Introduction

As discussed in chapter 2 the performance of ultrafiltration installations treating WWTP effluent decreases over time due to the fouling of membranes. Generally, in practice the actual performance and fouling rate of ultrafiltration installations (operating at constant flux) are determined mostly on-line by the trans membrane pressure or permeability. In this way of operation it is possible to adjust at any moment the process settings to control the performance of the ultrafiltration installation. But this approach has some disadvantages. A major disadvantage concerns the moment when for example the maximum trans membrane pressure is exceeded due to a bad WWTP effluent quality. At that moment the "bad" WWTP effluent has already entered the ultrafiltration installation resulting in operational problems. Considering this possible "bad quality" and the operational consequences (reduced production time, extra consumption of chemical for cleaning, etc.) many researchers and operators in practice are looking for tools to predict the fouling rate and loss of performance of ultrafiltration installations. When it is possible to predict and monitor the filtration properties and the quality of effluent on-line, major improvements in operation may be accomplished. Monitoring and its direct follow-up via process adjustments will result in a stable performance with increase of membrane lifetime and decrease of cost per cubic meter of produced water.

In this chapter the possibility to use the SUR measurement as a predicting tool is investigated. Therefore firstly the relation between operational flux and SUR values has been examined and secondly the relation between SUR value and increase of resistance. These parameters are operational parameters and are often used in practice to evaluate the performance of ultrafiltration plants.

4.2 The SUR value of feedwater and the operational flux of an ultrafiltration installation

4.2.1 Experimental setup

The purpose of this experiment was to determine a relation between the operational flux and the SUR value of the feedwater of an ultrafiltration pilot installation. Therefore the ultrafiltration pilot installation as described in chapter 3 had been operated during a period of three weeks (05-04-06 – 26-04-06) at the WWTP Horstermeer. During this period the permeate flux was increased several different times and the frequency and the duration of chemical cleanings (CC) was changed (Table 4.1). The chemical cleaning included soaking of the membrane modules of the ultrafiltration pilot installation during 1 or 1.5 hours in a Divos 120Cl solution (1.25 %-w/w). The frequency and duration of the backflushes (BF) were constant during the total period: after a filtration period of 15 minutes a backflush of 45 seconds at a flow of 17 m^3/h. In Table 4.1 the operational parameters of the ultrafiltration pilot installation during the experimental period are summarized.

Table 4.1 – Operational parameters of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05-04-06 – 26-04-06

Period	Flow (m^3/h)	Actual T (°C)	Flux (L/m^2·h)	BF interval (h)	CC interval (h)	CC duration (h)
05-04-06 – 06-04-06	3	14	43	0.25	12	1.5
10-04-06 – 12-04-06	4	14	57	0.25	8	1.5
18-04-06 – 20-04-06	5	15	71	0.25	8	1.5
24-04-06 – 26-04-06	6	16	86	0.25	4	1

The maximum theoretical filtration resistance ($R_{max,th}$) has been used to evaluate the process performance of the ultrafiltration pilot installation. If the $R_{max,th,}$ calculated by Equation 3.2, was reached within 24 hours the process performance was qualified as sub critical. Therefore for the qualification of the process performance the maximum (theoretical) increase of the filtration resistance of 24 hours was calculated. This calculation is shown in Equation 4.1 and indirectly distracted from the datasheet of the membrane supplier as presented in the thesis of te Poele (2005). Of course, the 24 hours are subjective but this interval was also applied by te Poele (2005) and therefore used in this thesis. Consistent use of this time interval makes it possible to qualify and compare process performances of ultrafiltration installations.

$$\frac{\Delta R_{\max}}{t_{\min}} = \frac{R_{\max,th} - R_{membrane}}{24} \tag{4.1}$$

in which: $\Delta R_{max}/t_{min}$ = maximum rate of filtration resistance increase (1/m·h)
$R_{max,th}$ = maximum theoretical filtration resistance (m^{-1})
$R_{membrane}$ = membrane resistance of clean membranes ($0.8 \cdot 10^{12}$ m^{-1})

The maximum rate of the filtration resistance increase was compared with the actual rate of filtration resistance increase between two chemical cleanings. If the value of the actual rate exceeded the maximum rate the process performance was qualified as super critical. The performance was qualified as sub critical if the value of the actual rate was below the maximum rate. In Table 4.2 the maximum theoretical resistance values and rates are given during the experimental period.

Table 4.2 – The maximum theoretical resistance values and the maximum rates of the filtration resistance increase of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05-04-06 – 26-04-06.

Period	$R_{max,th}$ (10^{12}·m^{-1})	$\Delta R_{max}/t_{24h}$ (·10^{12} 1/m·h)
05-04-06 – 06-04-06	7.15	0.26
10-04-06 – 12-04-06	5.39	0.19
18-04-06 – 20-04-06	4.45	0.15
24-04-06 – 26-04-06	3.77	0.12

Next to the operation of the ultrafiltration pilot installation the SUR value of the feedwater of the ultrafiltration pilot installation was measured 2 or 3 times per day. Samples of the feedwater were taken after the buffertank of the multi media filter just before the ultrafiltration pilot installation (see Figure 3.1). During the experimental period the multi media filter was operated for denitrification and simultaneous phosphorous removal. Therefore methanol and poly aluminium chloride (PACl) were dosed in-line to the feedwater of the filter. In Table 4.3 the operational parameters of the multi media filter are given.

Table 4.3 – Operational parameters of the multi media filter at the WWTP Horstermeer during the period 03-04-06 – 27-04-06 (Miska-Markusch, 2009)

Period	Flow rate (m^3/h)	Filtration rate (m/h)	Filter run (h)	Frequency 'bumping cleaning' (d^{-1})	Methanol dosing (gCH_3OH/gNO_3-N)	PACl in metal orthophosphate ratio (mol/mol)
05-04-06 – 06-04-06	5	6.3	12	8	4.5	4.8
10-04-06 – 27-04-06	8	10	10	12	4.5	4.8

After sampling the feedwater of the ultrafiltration pilot installation the samples were heated up to 20 °C and further the SUR measurements were performed on-site with a mobile SUR equipment. The samples for foulants analyses were also taken and after filtration (0.45 µm) stored in a refrigerator (5 °C) on-site. Subsequently these samples were transported to the laboratory of Sanitary Engineering of Delft University of Technology. Then the foulants were analysed within at least one week after sampling.

4.2.2 SUR values

In Table 4.4 the average SUR values of the (sieved) WWTP effluent and the feedwater of the ultrafiltration pilot installation pilot are shown. The SUR values of the WWTP effluent varied between $13 \cdot 10^{12}$ m^{-2} and $27 \cdot 10^{12}$ m^{-2}. This variation did not seem exceptional compared to the variation of effluent SUR values at other WWTPs in the Netherlands. For instance, Roorda (2004) presented SUR values of effluent in the range of $6.0 \cdot 10^{12}$ m^{-2} to $40 \cdot 10^{12}$ m^{-2} at the WWTP Berkel. Due to the complexity of a wastewater treatment process it is difficult to explain and even more to predict the SUR values of WWTP effluent. Several factors e.g. changing of weather conditions, changing of operational conditions of the WWTP, varying influent water quality, etc. influence the performance of the wastewater treatment process and subsequently the SUR value of WWTP effluent.

Not only the SUR values of the WWTP effluent varied during the experimental period but also the SUR values of the feedwater of the ultrafiltration pilot installation. As shown in Table 4.4, the SUR values varied between $9 \cdot 10^{12} \cdot m^{-2}$ and $25 \cdot 10^{12}$ m^{-2}. This range is more or less comparable with the range of the WWTP effluent ($13 \cdot 10^{12}$ m^{-2} – $27 \cdot 10^{12}$ m^{-2}). Nevertheless the SUR values of the feedwater of the ultrafiltration pilot installation were expected to be less due to the filtration step (multi media filter) before the ultrafiltration pilot installation. But as presented in Table 4.4 there was no direct relation between the SUR values of the WWTP effluent and the feedwater of the ultrafiltration pilot installation. A possible explanation could be the position of the sampling point. The samples were taken at the outlet of the buffertank (bottom buffertank) just before the feedwater enters the ultrafiltration pilot installation. Probably the SUR values have been influenced by (uncontrolled) processes in the buffertank (e.g. biological re-growth, settling of particulate organic matter on the bottom, dead zones, etc.). Another important factor could be of course the performance of the multi media filter. It might be possible that the multi media filter did not perform optimal during the whole period. However the influence of these mentioned factors was not investigated because it was not within the scope of this chapter. In chapter 5 the effect of multi media filtration on the SUR values of WWTP effluent is more thoroughly investigated.

Table 4.4 – Average SUR values of the WWTP effluent and the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05/04/06 – 26/04/06

	WWTP effluent		Feedwater ultrafiltration pilot installation	
Date	Average SUR (10^{12}·m^{-2})	Number of samples	Average SUR (10^{12}·m^{-2})	Number of samples
05-04-06	-	-	14.4	3
06-04-06	18.9	1	14.5	2
10-04-06	20.1	3	23.3	3
11-04-06	14.1	3	9.8	3
12-04-06	23.6	4	10.8	2
18-04-06	15.4	2	24.6	3
19-04-06	17.9	3	11.6	3
20-04-06	26.1	2	12.7	3
24-04-06	13.6	2	10.5	2
25-04-06	21.9	3	10.2	3
26-04-06	26.6	3	9.2	3

4.2.3 Foulants

In Table 4.5 the average concentrations of foulants during the experimental period is shown except for the days 05-04-06 and 06-04-06. During these days no samples of the feedwater of the ultrafiltration pilot installation were taken.

In general the concentrations of foulants of the feedwater of the ultrafiltration pilot installation were more or less comparable with the concentrations of foulants found at other WWTPs in the Netherlands. Te Poele (2005) published colour concentrations in the range of 35 – 136 mg Pt/L, polysaccharide concentrations of 2.7 – 4.7 mg/L, protein concentrations of 10.1 – 25.3 mg/L, humic substances amounts of 0.252 – 0.608 cm^{-1} and PUVA ratios of 2.0 – 2.8 L/mg·m at different WWTPs in the Netherlands. But in contrast to this study, te Poele (2005) took the WWTP effluent samples directly after the final clarifier of the different WWTPs. In this study the samples were taken directly after multi media filtration (including methanol and poly aluminium chloride dosage). Therefore small differences (e.g. concentrations of polysaccharides) may be explained by the operation of the multi media filter. From other investigations (Janssen *et al.*, 2010) it is known that the operation of a multi media filter filtration (including methanol and poly aluminium chloride dosage) could negatively influence the concentrations of polysaccharides. Apart from the concentrations of polysaccharides it could be noticed that the feedwater quality of the ultrafiltration pilot installation was not extraordinary compared to effluent of other WWTPs in the Netherlands.

Table 4.5 – The concentrations of foulants of the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05-04-06 – 26-04-06

Date	DOC (mg/L)	Polysaccharides (mg/L)	Proteins (mg/L)	Colour (mg Pt/L)	Humic substances (cm^{-1})	PUVA (L/mg m)
10-04-06	32.5	6.9	7.8	39	0.211	2.7
11-04-06	24.7	8.4	9.3	47	0.205	2.2
12-04-06	42.6	7.3	12.1	50	0.241	2.0
18-04-06	30.2	9.3	10.3	50	0.244	2.4
19-04-06	25.7	6.8	8.7	35	0.210	2.4
20-04-06	22.8	5.9	9.8	58	0.229	2.3
24-04-06	21.4	6.9	9.1	50	0.219	2.4
25-04-06	25.7	8.3	10.1	50	0.237	2.3
26-04-06	24.3	7.1	9.3	42	0.218	2.3
Average	27.8	7.4	9.6	52.3	0.224	2.3

4.2.4 Performance ultrafiltration pilot installation

In Figure 4.1 the process performance of the ultrafiltration pilot installation during the period 05-04-06 – 06-04-06 is shown. The filtration resistance at the start was around $0.8 \cdot 10^{12}$ m^{-1} which means the membranes were clean. During the period the operation of the ultrafiltration pilot installation was sub critical. On 05-04-06 and 06-04-06 the average $\Delta R/\Delta t$ values were respectively 0.09 1/m·h and 0.08 1/m·h. This is significantly below the $\Delta R_{max}/t_{24h}$ value of 0.26 1/m·h. Also the chemical cleaning on 05-04-06 revealed to be sufficient enough since the filtration resistance came back to the level at the start of the experiment. On 06-04-06 the filtration resistance at the start was about $0.9 \cdot 10^{12}$ m^{-1} what means the membranes were slightly fouled but this remained fouling did not present any influence on the process performance. Generally it can be concluded that the process performance of the ultrafiltration pilot installation was sub critical under the given process conditions during the period 05-04-06 – 06-04-06.

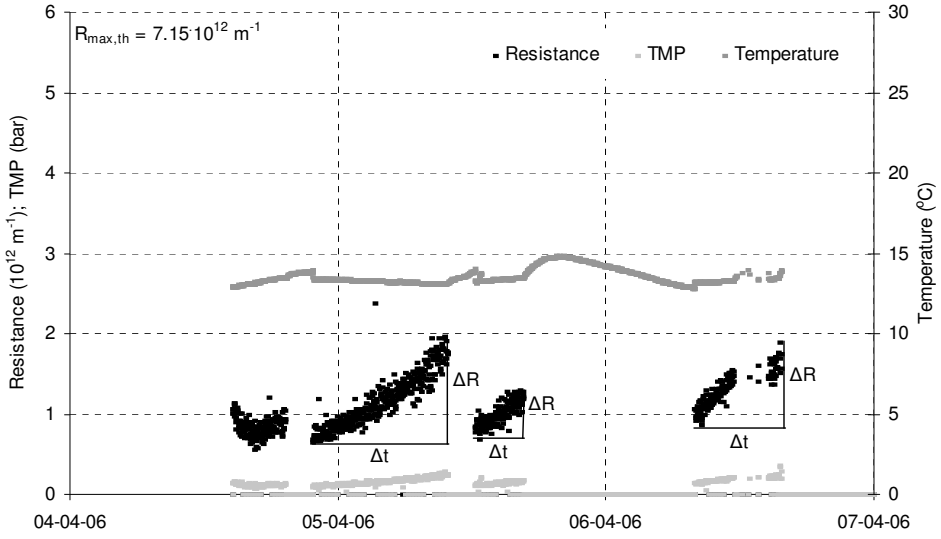


Figure 4.1 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer at a flux of 43 L/m^2·h

In Figure 4.2 the process performance of the ultrafiltration pilot during the second period (10-4-2006 – 12-04-2006) is presented. At the start of the period (10-04-06) the filtration resistance of membrane was around $0.8 \cdot 10^{12}$ m^{-1}. On the other days the filtration resistance increased till approximately $1.0 \cdot 10^{12}$ m^{-1} after a chemical cleaning. It means that still residual fouling remained on the membranes and the chemical cleaning procedure was not sufficient enough to obtain the initial membrane resistance ($0.8 \cdot 10^{12}$ m^{-1}) at the start of a filtration period.

During the first filtration period (10-04-06) a sharp increase of the filtration resistance can be observed from Figure 4.2. The $\Delta R/\Delta t$ value of this filtration run was 0.39 1/m·h. This is almost two times more than the $\Delta R_{max}/t_{24h}$ value of 0.19 1/m·h. Therefore the process performance was qualified as super critical during this day. During the other days (11-04-06 – 12-04-06) the average $\Delta R/\Delta t$ values were respectively 0.27 1/m·h and 0.14 1/m·h. This meant that only on 12-04-06 a sub critical process performance was obtained. Therefore it seemed that a flux of 57 L/m^2·h will lead to a more critical performance than for instance an operational flux of 43 L/m^2·h in the first period.

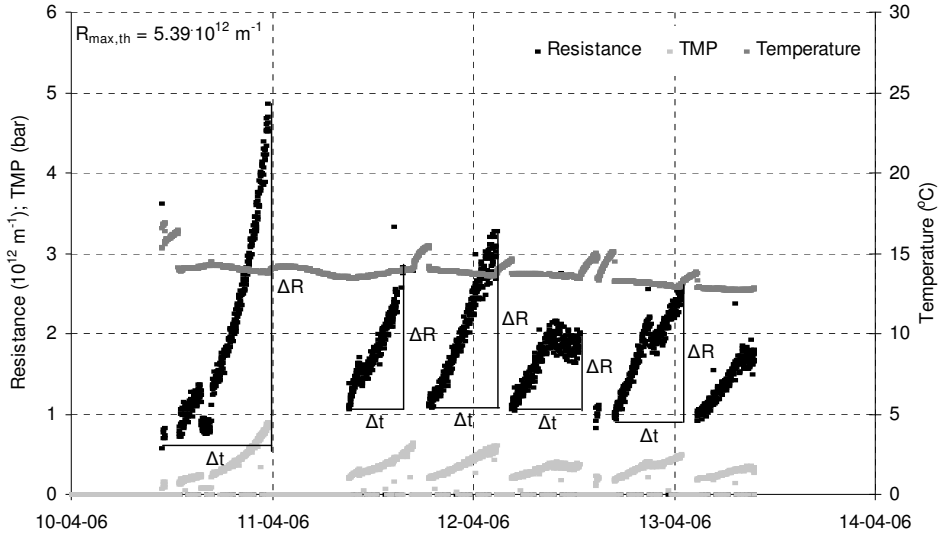


Figure 4.2 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer at a flux of 57 L/m^2·h

Figure 4.3 presents the performance of the ultrafiltration pilot installation between 18-4-06 and 20-4-06. Like the previous period a sharp increase of the filtration resistance ($\Delta R/\Delta t$ = 0.63 1/m·h) was noticed during the first filtration period. Considering the $\Delta R_{max}/t_{24h}$ value of 0.15 1/m·h it is evident that the process performance was considered as super critical. Furthermore during the next day (19-04-06) the process performance was super critical as well. The average $\Delta R/\Delta t$ value was 0.33 1/m·h during this day. Only on the third day (20-04-06) the process performance was sub critical. On this day the $\Delta R/\Delta t$ value was 0.10 1/m·h which is significantly below the $\Delta R_{max}/t_{24h}$ value.

The sharp increase of the first filtration might be explained by the average SUR value of the feedwater. The average SUR value was $24.6 \cdot 10^{12}$ m^{-2} which is significantly higher than the SUR values of the other days. Nevertheless the SUR value of the feedwater did not explain the different process performance of 19-04-06 and 20-04-06. On both days the average SUR values were measured in the range of $12.0 \cdot 10^{12}$ m^{-2} but the process performance differed. Therefore next to the quality of the feedwater other aspects might also influence the process performance of the ultrafiltration pilot installation. Probably the efficiency of the chemical cleaning played a role. On the 19-04-06 the chemical cleaning was less efficient than during the next day (20-04-06). This aspect of the process performance will be further discussed in the next section of this chapter.

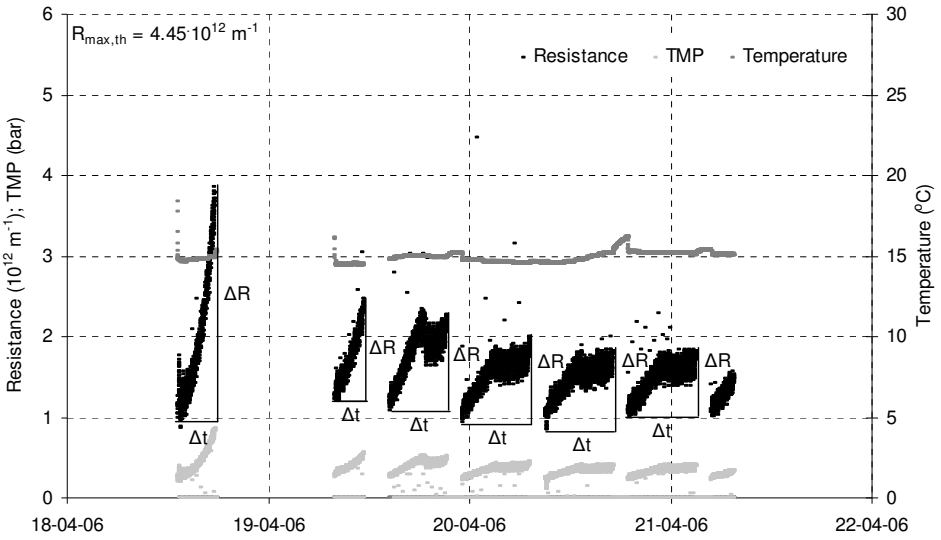


Figure 4.3 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer at a flux of 71 L/m^2·h

The results of the last period are shown in Figure 4.4. The flux during this period was 86 L/m^2·h at the actual temperature. This relative high flux resulted in an super critical process performance during the whole period. The ΔR/Δt values varied between 0.68 1/m·h and 0.16 1/m·h which is considerably higher than the $\Delta R_{max}/t_{24h}$ of 0.12 1/m·h. Therefore it might be concluded it is not possible to obtain sub critical process performance at a flux of 86 L/m^2·h. Even when the SUR values of the feedwater are relatively low. As shown in Table 4.5 the average SUR values were about 10·10^{12} m^{-2} during the whole period.

In Figure 4.4 also the effect of chemical cleanings can be observed. It is obviously that the chemical cleanings were insufficient. During the whole period an increase of the filtration resistance at the start of a new filtration run can be noticed. This effect might be also related to the flux rate. Evidently at higher fluxes a larger amount of foulants passing along the membrane surface increasing probably the adsorption of foulants. Therefore at higher fluxes more intensive chemical cleaning and/or other combination of chemicals should be considered.

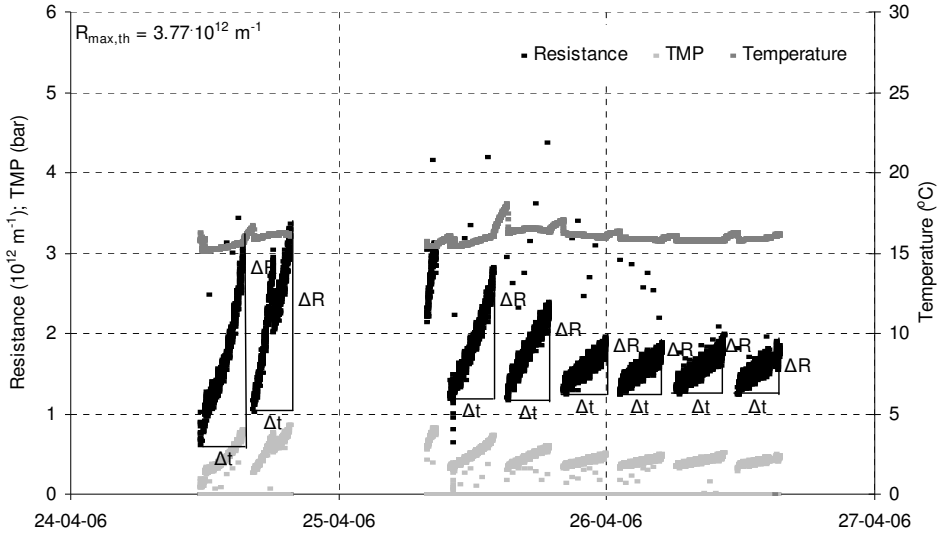


Figure 4.4 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate of the WWTP Horstermeer at a flux of 86 L/m^2·h

In Figure 4.5 the results of the process performance ($\Delta R/\Delta t$) of the pilot ultrafiltration installation are summarized together with the maximum rate of filtration resistance increase during 24 hours ($\Delta R_{max}/t_{24h}$). The figure clearly shows that at lower fluxes (40 – 50 L/m^2·h) the process performance can be considered as sub critical. But when the flux increases the process performance becomes critical or super critical. Fluxes of more than approximately 80 L/m^2·h seemed to result always in super critical process performance where fluxes in the range of 50 L/m^2·h and 80 L/m^2·h show some variation. In this flux range probably other aspects like for instance cleaning efficiency and water quality play an important role in the performance of the filtration process.

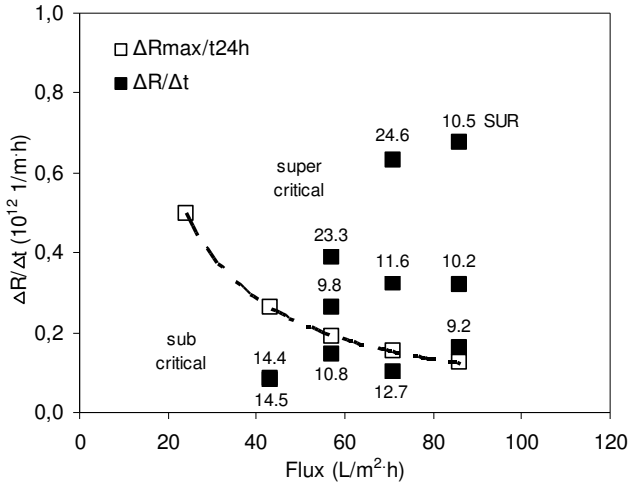


Figure 4.5 – Process performance (ΔR/Δt) and SUR values compared with the maximum filtration resistance increase during 24 hours (R_{max}/t_{24h}) of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05-04-06 – 26-04-06

4.2.5 Discussion

As stated in the introduction the purpose of this experiment was to compare the measured SUR values of the feedwater with the process performance of the ultrafiltration pilot installation. The performance was qualified by the maximum slope of filtration resistance during 24 hours ($\Delta R_{max}/t_{24h}$). This value was compared with the actual slope between two chemical cleanings (ΔR/Δt). When the value of the actual slope exceeded the maximum slope the process was qualified as super critical. Sub critical was obtained when the value of the actual slope was lower than the value of the maximum slope. In Table 4.6 the overall process performance together with the SUR values, flux and ΔR/Δt value are summarized.

Table 4.6 – Performance of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 05/04/06 – 26/04/06

Date	Flux ($L/m^2 \cdot h$)	ΔR/Δt (10^{12} 1/m·h)	Average SUR (10^{12} m^{-2})	Performance (critical)
05-04-06	43	0.09	14.4	sub
06-04-06	43	0.08	14.5	sub
10-04-06	57	0.39	23.3	super
11-04-06	57	0.27	9.8	super
12-04-06	57	0.14	10.8	sub
18-04-06	71	0.63	24.6	super
19-04-06	71	0.33	11.6	super
20-04-06	71	0.10	12.7	sub
24-04-06	86	0.68	10.5	super
25-04-06	86	0.32	10.2	super
26-04-06	86	0.16	9.2	super

Table 4.6 shows the dependency of the performance of the ultrafiltration pilot installation on the SUR value of feedwater. With an operational flux of 43 $L/m^2 \cdot h$ and an average SUR value

of approximately $14 \cdot 10^{12}$ m^{-2} sub critical performance was observed. But when the flux was increased (57 L/m^2·h and 72 L/m^2·h) super critical performance occurred. At high SUR values ($23.3 \cdot 10^{12}$ m^{-2} and $24.6 \cdot 10^{12}$ m^{-2}) the $\Delta R/\Delta t$ values were high as well resulting in super critical performance. At lower SUR values ($10 \cdot 10^{12}$ m^{-2} – $12 \cdot 10^{12}$ m^{-2}) the process performance became less clear. Both sub and super critical performance were noticed with more or less similar SUR values of the feedwater. In this range of SUR values probably other aspects like the efficiency of cleaning may play a role as well. Operating at fluxes of 86 L/m^2·h resulted in this study at all times in super critical performance. Even when the SUR value was about $9.0 \cdot 10^{12}$ m^{-2}. In Figure 4.6 the findings of Table 4.6 are summarized in an operating window showing the relation between the SUR value of feedwater and the applicable flux.

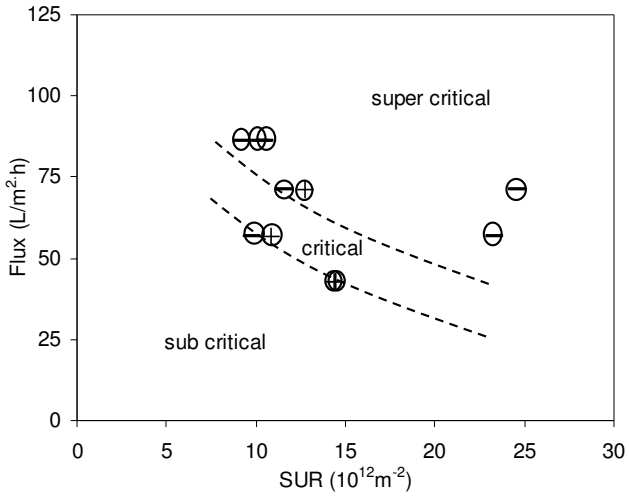


Figure 4.6 – Operating process performance window showing the relation between SUR values and flux of the ultrafiltration pilot installation at the WWTP Horstermeer

In Figure 4.6 three areas are defined: sub critical, critical and super critical. The areas are arbitrary although they give an indication about the relation between the flux and obtained SUR values. The results differ from the conclusions of Roorda (2004). Roorda (2004) stated that SUR values below $10 \cdot 10^{12}$ m^{-2} seem to be a precondition for stable i.e. sub critical performance with high fluxes ($\geq$ 100 L/m^2·h, 15 °C). However the results presented in this chapter show that significantly lower SUR values ($< 5 \cdot 10^{12}$ m^{-2}) seem to be a precondition for sub critical performance at high operational fluxes. Especially, when the chemical cleaning procedures are considered. During this experiment the chemical cleaning were intensive to obtain comparable filtration resistance at the start of each filtration period. Normally, in practice less intensive chemical cleanings are applied.

Next tot the flux and chemical cleanings, the production of permeate is a parameter to evaluate as well. The production of permeate is the aim of membrane filtration. The produced

permeate volume results from the integration of the flow over time minus the loss of permeate for back flushes and chemical cleanings. In Figure 4.7 the net production and recovery during the different periods of the experiment are shown. Usually micro- and ultrafiltration systems operate at a 90 to 95% recovery (Bixio and Wintgens, 2006). The maximum recovery obtained in this experiment was 80% with a feedwater flow of 4 m^3/h.

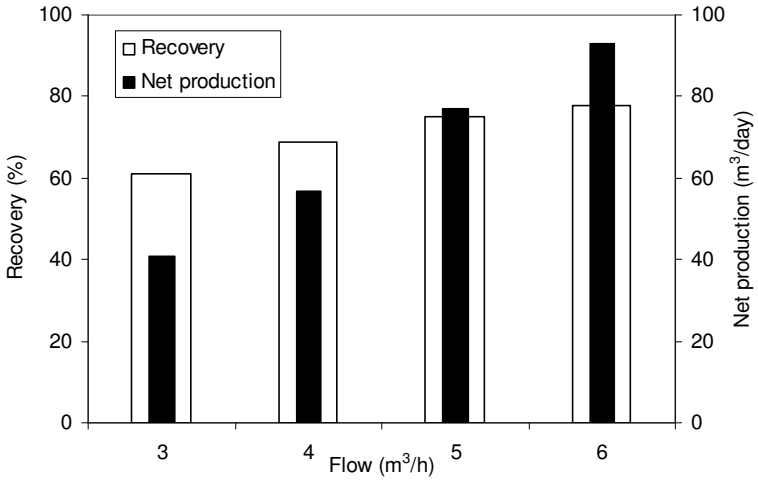


Figure 4.7 – Recovery and net production at different flow rates and operational conditions of the ultrafiltration pilot installation at the WWTP Horstermeer

Summarizing the experiment it seems that fluxes of 40 L/m^2·h – 60 L/m^2·h are preferable at the WWTP Horstermeer. At these fluxes rather stable performance is obtained and probably the frequency of chemical cleanings and back flushes can be decreased to get a better recovery and higher net permeate production. Nevertheless, these new settings can only be applied when the SUR values are $\leq 10 \cdot 10^{12}$ m^{-2}. When the SUR values of the feedwater are in the range of $15 \cdot 10^{12}$ m^{-2} – $30 \cdot 10^{12}$ m^{-2} probably an extra incidental chemical cleaning will be necessary or a lower flux should be considered. This will prolong the ultrafiltration operation time and decrease potential operational problems.

4.3 The SUR value of feedwater and the increase of filtration resistance of an ultrafiltration installation

4.3.1 Experimental setup

A second experiment to investigate the relation between the SUR values of ultrafiltration feedwater and the increase of trans membrane pressure and filtration resistance had been performed at the WWTP Sas van Gent. The increase of filtration resistance over time is a generally used parameter to measure the fouling rate of full scale and pilot installations (Rosenberg *et al*., 2005; te Poele, 2005) and is explained in chapter 3 of this thesis. The operational parameter, increase of trans membrane pressure over time, is less used in scientific research but in practice a useful tool for operators to evaluate the actual performance of a ultrafiltration installation. In this section both parameters are related to the SUR values of the feedwater of an ultrafiltration installation in order to determine the possibility to apply the SUR measurement to predict actual ultrafiltration process performance.

The experiment had been performed during the period 01-09-06 – 11-10-06 at the WWTP Sas van Gent (for details of the ultrafiltration installation see chapter 3). The samples of the feedwater were taken just before the water entered the ultrafiltration units of the installation (Figure 4.8). Process data had been collected from two of the four units; ultrafiltration unit 1 and 4. Both units are more or less identical but only the lifetime of the membranes modules was different. The membrane modules of ultrafiltration unit 1 had been replaced approximately 6 months before the start of the experiment. The membrane modules of ultrafiltration unit 4 were already 4 years in operation and therefore expected to be more fouled and chemically worn than the membrane modules of ultrafiltration unit 1.

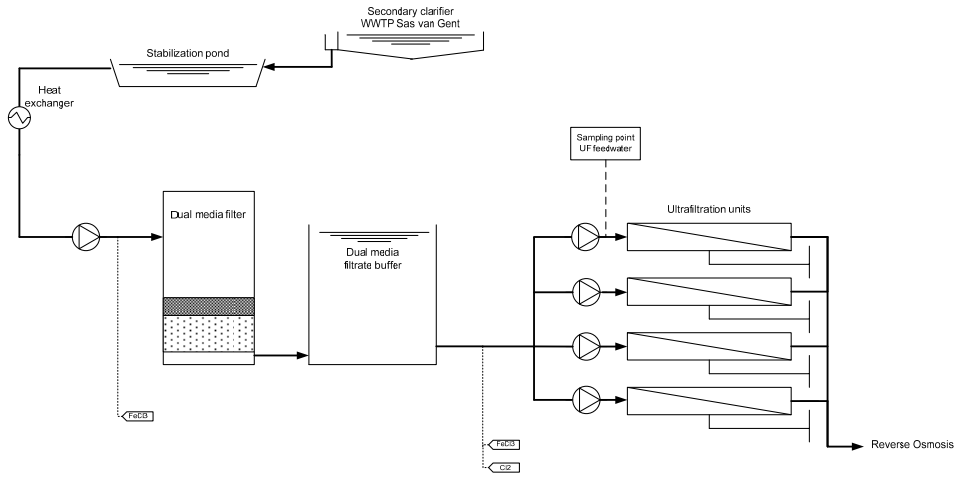


Figure 4.8 – Schematic overview of the WWTP Sas van Gent, stabilization pond, dual media filter, filtrate buffer and ultrafiltration units inclusive the sampling point ultrafiltration feedwater

As shown in Figure 4.8 the (industrial) WWTP effluent was not directly fed to the ultrafiltration plant. During the experimental period the WWTP effluent was firstly buffered in a stabilization pond during 2 – 3 days. After the stabilization pond the (stabilized) WWTP effluent was pumped to the dual media filter. The dual media filter was operated with upstream addition of ferric chloride (2.5 mg Fe^{3+}/L) to remove suspended solids. The filtrate of the dual media filter was collected in a buffertank and further pumped to the ultrafiltration installation. Between the buffertank and the ultrafiltration installation ferric chloride (1.0 mg Fe^{3+}/L) and sodium hypochlorite (NaOCl) were dosed to decrease the fouling rate of the ultrafiltration membranes.

The samples of the ultrafiltration feedwater had been taken just before the water entered the ultrafiltration installation (Figure 4.8). After sampling the samples were warmed to 20 °C and the SUR value was measured directly on-site with a mobile SUR equipment. The analyses of the foulants were done within a week after sampling at the laboratory of Sanitary Engineering of Delft University of Technology. Therefore, the samples for analyses were stored after filtration (0.45 µm) on site in a refrigerator.

4.3.2 SUR values

Table 4.7 shows the SUR values of the WWTP effluent after the stabilization pond and of the feedwater of the ultrafiltration installation. During the experimental period the SUR values of the (stabilized) WWTP effluent were measured in the range of $9 \cdot 10^{12}$ m^{-2} – $21 \cdot 10^{12}$ m^{-2}. This observed range of SUR values is comparable with variations of SUR values at other WWTPs in the Netherlands. For instance in the previous section of this chapter a range of SUR values of $13 \cdot 10^{12}$ m^{-2} – $27 \cdot 10^{12}$ m^{-2} was noticed at the WWTP Horstermeer. Furthermore Roorda (2004) presented SUR values in the range of $6 \cdot 10^{12}$ m^{-2} – $40 \cdot 10^{12}$ m^{-2}. However less variation of the SUR value of WWTP effluent was expected due to the buffering effect of the stabilization pond. In this pond the WWTP effluent was buffered during 2 – 3 days and this may lead to more stable SUR values of the WWTP effluent. This effect was not observed and will be further discussed in chapter 6 of this thesis.

Next to the SUR values of WWTP effluent, Table 4.7 shows the SUR values of the feedwater of the ultrafiltration plant. Due to the different pretreatment steps (coagulation – dual media filter – coagulation) the SUR value of the WWTP effluent decreased in average with 54% resulting in SUR values of ultrafiltration feedwater in the range of $5 \cdot 10^{12}$ m^{-2} and $10 \cdot 10^{12}$ m^{-2}. According to Roorda (2004) it means that the filterability of the ultrafiltration feedwater was good. Although considering the results of section 4.2 lower ($<5 \cdot 10^{12}$ m^{-2}) SUR values of the ultrafiltration feedwater are preferred for stable operational process performance.

Table 4.7 – SUR values of the WWTP effluent (after stabilization pond) and the feedwater of the ultrafiltration installation at the WWTP Sas van Gent during the period 01-09-06 – 11-10-06

Date	WWTP effluent (after stabilization pond) SUR (10^{12} m^{-2})	Feedwater ultrafiltration installation SUR (10^{12} m^{-2})
01-09-06	19.0	5.7
05-09-06	21.2	6.8
06-09-06	19.0	10.2
07-09-06	19.0	8.7
12-09-06	17.6	6.4
14-09-06	12.2	6.2
19-09-06	16.7	6.8
21-09-06	13.1	7.3
25-09-06	9.8	5.6
26-09-06	11.5	7.1
28-09-06	18.0	6.9
02-10-06	18.5	6.1
04-10-06	15.5	6.4
09-10-06	10.5	5.7
11-10-06	8.6	4.6

4.3.3 Foulants

Next to the SUR values of the feedwater of the ultrafiltration installation the concentration of foulants had been monitored (see Table 4.8). The origin of the wastewater (food producing factory) of the WWTP Sas van Gent may be observed by the concentration of proteins. This concentration is roughly two times more than for instance the concentration of proteins in the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer (see Table 4.5). In spite of the high concentration of proteins the PUVA value was equal to the PUVA value of the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer. This means that next to the concentration of proteins the amount of humic substances was twice of the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer. Consequently the feedwaters presented similar PUVA values.

As is generally known the water quality of WWTP effluent depends on the origin of the wastewater (municipal, industrial, etc.). Therefore it is difficult to compare the water quality of the WWTP Sas van Gent with the water quality of other municipal WWTPs in Netherlands. In spite of this difference it is noticed that the concentration of polysaccharides and colour of the effluent of WWTP Sas van Gent are close or even in the range of other WWTPs in the Netherlands. te Poele (2005) presented colour concentrations in the range of 35 – 139 mg Pt/L and concentration of polysaccharides of 2.7 – 4.7 mg/L. Therefore in general it is concluded that apart from the concentrations of DOC and colour, the concentration of foulants of the WWTP effluent of Sas van Gent were only slightly higher than other WWTPs in the Netherlands.

Table 4.8 – Water quality of the feedwater of the ultrafiltration installation at the WWTP Sas van Gent during the period 01-09-06 – 11-10-06

Date	DOC (mg/L)	Polysaccharides (mg/L)	Proteins (mg/L)	Colour (mg Pt/L)	Humic substances (cm^{-1})	PUVA (L/mg·m)
01-09-06	17.9	6.4	16.0	47.8	0.372	2.3
05-09-06	21.6	6.5	19.8	81.0	0.538	2.7
06-09-06	-	-	-	-	-	-
07-09-06	23.7	7.0	19.4	66.0	0.485	2.5
12-09-06	27.9	7.0	24.4	66.0	0.500	2.0
14-09-06	24.0	6.9	25.4	70.1	0.499	2.0
19-09-06	19.2	6.5	21.6	56.9	0.431	2.0
21-09-06	19.2	-	-	-	-	-
25-09-06	18.3	7.2	17.0	73.3	0.330	1.9
26-09-06	-	-	-	-	-	-
28-09-06	24.2	8.3	20.2	74.1	0.339	1.7
02-10-06	-	6.2	16.9	56.9	0.413	2.4
04-10-06	-	-	-	-	-	-
09-10-06	19.8	6.4	19.0	52.8	0.413	2.2
11-10-06	-	-	-	-	-	
Average	21.6	6.8	20.0	64.5	0.432	2.2

4.3.4 The increase of resistance

The ultrafiltration installation at the WWTP Sas van Gent had been designed to deliver continuously a fixed amount of permeate volume per hour. This mode of operation resulted frequently in a varying fluxes per ultrafiltration unit. For instance when one unit of the four was out of production due to a chemical cleaning in place or backflush, the other three units had to operate with a higher flux in order to produce the fixed amount of permeate volume. Consequently the varying flux influenced the increase of the trans membrane pressure and filtration resistance during a filtration run. Therefore only process data were collected when the flux was more or less stable (standard deviation $\leq 5\%$) during a filtration run.

In accordance to Darcy's law the actual resistance was calculated as presented in Equation 4.2.

$$R_{actual} = \frac{P_{actual}}{\eta_{actual} \cdot J_{actual}} \tag{4.2}$$

in which: R_{actual} = filtration resistance (m^{-1})
P_{actual} = trans membrane pressure at actual temperature (bar)
η_{actual} = dynamic viscosity of feedwater at actual temperature (Pa·s)
J_{actual} = actual flux ($L/m^2 \cdot h$)

Equation 4.2 was applied to calculate the actual filtration resistance. The next equation (4.3) presents the procedure to measure the slope of the filtration resistance during one filtration run of approximately 20 minutes.

$$\frac{dR}{dt} = \frac{R_{actual,\,t=x} - R_{actual,\,t=0}}{t_x - t_0} \qquad (4.3)$$

in which: dR = filtration resistance difference (m^{-1})
dt = time difference (hour)
$R_{actual,\,t=x}$ = actual filtration resistance the end of filtration run (m^{-1})
$R_{actual,\,t=0}$ = actual filtration resistance at the start of filtration run (m^{-1})
t_x = time at the end of filtration run (hour)
t_0 = time at the start of filtration run (hour)

In Table 4.9 the results of the SUR values of feedwater and the dR/dt of ultrafiltration unit 1 and 4 are presented. As discussed previously the flux of ultrafiltration unit 1 and 4 was not constant all the time. Therefore half of the process data could not be used for the evaluation. As shown in Table 4.9 only the process data of 9 days were used. During these days the filterability of the feedwater was $\leq 10 \cdot 10^{12}$ m^{-2}. According to Roorda (2004) it meant the filterability was good.

Next to the different SUR values of feedwater the dR/dt values of the ultrafiltration unit 1 and 4 differed. At the same SUR values of feedwater the ultrafiltration 1 presented significantly lower dR/dt values than the ultrafiltration unit 4. This is clearly shown in Figure 4.9 were the obtained dR/dt values are plotted against the SUR values of feedwater.

Table 4.9 – SUR values of feedwater and the dR/dt values of the ultrafiltration unit 1 and 4 of the ultrafiltration installation at the WWTP Sas van Gent during the period 01-09-06 – 11-10-06

Date	Feedwater	Ultrafiltration unit 1		Ultrafiltration unit 4	
	SUR (10^{12} m^{-2})	Average flux ($L/m^2 \cdot h$)	dR/dt (10^{12} $1/m \cdot h$)	Average flux ($L/m^2 \cdot h$)	dR/dt (10^{12} $1/m \cdot h$)
05-09-06	6.8	23.3	0.17	26.9	0.70
06-09-06	10.2	52.1	0.39	59.5	2.24
07-09-06	8.7	26.5	0.31	-	-
12-09-06	6.4	22.3	0.11	26.0	0.24
14-09-06	6.2	52.1	0.24	59.1	1.12
19-09-06	6.8	-	-	53.7	0.93
25-09-06	5.6	22.8	0.07	26.1	0.63
28-09-06	6.9	33.1	0.13	38.7	0.75
09-10-06	5.7	-	-	26.1	0.29

The results of Figure 4.9 indicate a relation between the SUR values of feedwater and dR/dt values of the ultrafiltration unit 1 and 4. But despite the significant correlation different relations were observed for the ultrafiltration units 1 and 4. The ultrafiltration unit 4 presented significantly higher dR/dt values at the same SUR values of feedwater than ultrafiltration unit 1. An explanation for this difference might be the different condition of the membrane modules of the units. As written earlier in this section the membrane modules of ultrafiltration

unit 1 had been replaced half a year before the experiment contrary to the membrane modules of ultrafiltration unit 1. These membrane modules had not been replaced and were already approximately 4 years in operation. Therefore it could be expected that these membrane modules were more fouled and probably chemically affected. Considering this difference in life time and condition it seemed that the extra filtration resistance due to fouling resulted in a higher operational pressure, leading to a higher increase of trans membrane pressure and filtration resistance per hour.

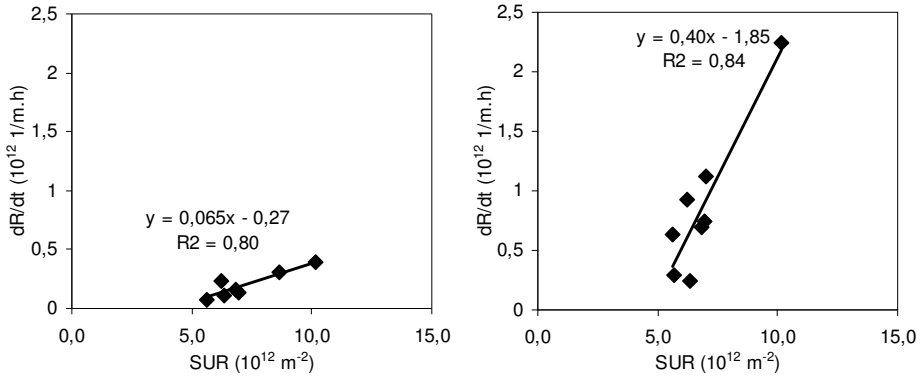


Figure 4.9 – The relationship between the SUR values of feedwater and the dR/dt values of the ultrafiltration unit 1 (left) and 4 (right) during the period 01-09-06 – 11-10-06

4.3.5 Discussion

The purpose of this experiment was to determine a relation between the SUR value of feedwater and the operational parameters of an ultrafiltration installation. As operational parameters the dR/dt of the ultrafiltration installation was taken. Although this parameters is only an indicator of short term fouling (filterability) they also could give an indication about long term fouling since the short term fouling and long term fouling (reversibility) are considered to be related (Roorda, 2004; te Poele, 2005). The impact of long term fouling might be an explanation for the different relations as shown in Figure 4.9. Due to the different life time and condition it was assumed that the membrane modules of ultrafiltration unit 4 are fouled more severely. This remaining fouling seemed to influence the dR/dt values. This (negative) influence of remaining fouling is also described by Roorda (2004). Insufficient hydraulic or chemical cleanings and consequently the presence of irreversible fouling will lead to a more rapid increase of trans membrane pressure and filtration resistance. This process is accelerated by the filtration properties of WWTP effluent. Compared to other water sources the fouling caused by dead-end ultrafiltration of WWTP effluent is highly compressible (Roorda, 2004; Zheng *et al.*, 2010). Therefore higher operational trans membrane pressure may lead to a compression of the formed cake layer during dead-end ultrafiltration of WWTP effluent and consequently decrease the reversibility of fouling during

a hydraulic cleaning. However this aspect was not investigated during the experiment but could explain the observed difference and underline the need of efficient chemical and hydraulic cleaning.

Considering the findings of this section and the unavoidable differences (lab scale versus full scale measurement of the filterability) between the SUR measurement of feedwater and the interpretation of operational parameters it seemed that the SUR measurement can be applied to evaluate the filterability of (dead-end) ultrafiltration installations.

4.4 Evaluation

In this chapter the possibility to use the SUR measurement as a predicting tool for the process performance of an ultrafiltration installation is investigated. With the first experiment the relation between the SUR value of feedwater and the operational flux of a pilot ultrafiltration installation has been examined. The flux is an important parameter for the design of a (full scale) ultrafiltration installation. Normally this parameter is obtained after thorough pilot scale experiments but these experiments are mostly time consuming and costly. Therefore as an alternative these experiments might be replaced by SUR measurement to have an indication about the design flux. The experiment has shown that SUR values below $5 \cdot 10^{12}$ m^{-2} are required for sub critical operation at high operational fluxes. This finding was partly confirmed by earlier research of Roorda (2004). Roorda (2004) reported a SUR value of $10 \cdot 10^{12}$ m^{-2} as a requirement for stable operation at high operational fluxes but unfortunately the definition of stable operation was not clearly specified in the thesis of Roorda (2004) and makes comparison therefore difficult. In the study presented in this chapter SUR values of $10 \cdot 10^{12}$ m^{-2} were needed for sub critical process performance at operational fluxes of 40 – 60 $L/m^2 \cdot h$. When the SUR values were found in the range 15 – $30 \cdot 10^{12}$ m^{-2} the process performance became super critical and lower fluxes or extra cleanings were needed. Although the process performance depends on local conditions, efficiency of the WWTP process, source wastewater, etc. A classification is made of the operational fluxes versus the SUR value of feedwater (Table 4.10). At these SUR values of feedwater the ultrafiltration installation is expected to operate stable at the given operational fluxes.

Table 4.10 – The SUR values of feedwater and corresponding operational flux.

SUR feedwater (10^{12} m^{-2})	Flux ($L/m^2 \cdot h$)
≤ 5	60 – 80
5 – 10	40 – 60
10 – 15	20 – 40
≥ 15	≤ 20

During the second experiment, presented in section 4.3, the SUR value of feedwater was related to the operational parameter (dR/dt) of a (full scale) ultrafiltration installation. The results showed a rather good relation but the slope of the relation depends on the condition of

the membrane modules. Membranes that are fouled more will lead to a higher filtration resistance at the same SUR value of feedwater. Therefore this aspect should be well considered when the settings of operational parameters are assumed based on the SUR value of feedwater and this finding underlines the need of SUR values of feedwater below $5 \cdot 10^{12}$ m^{-2} for sub critical process performance. The influence of feedwater at low SUR values is less for fouled membranes (Figure 4.9).

4.5 Conclusions

- The process performance of the ultrafiltration pilot installation at WWTP Horstermeer depends on the SUR values of the feedwater. For sub critical process performance with fluxes of 40 – 60 L/m^2·h the SUR value of the feedwater may be $\leq 10 \cdot 10^{12}$ m^{-2}.

- For sub critical process performance at higher fluxes (60 – 80 L/m^2·h) SUR values below $5 \cdot 10^{12}$ m^{-2} are required.

- The process parameter, dR/dt, of the ultrafiltration installation at the WWTP Sas van Gent revealed a significant relation with the SUR value of ultrafiltration feedwater.

- The relation between the process parameter dR/dt of the ultrafiltration installation at the WWTP Sas van Gent and the SUR value of feedwater seemed to be dependent on the condition of the membrane module.

References

Bixio, D. and Wintgens T. Editors (2006). *Water Reuse System Management Manual AQUAREC*. Project report European Comission, Luxembourg.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den, Geilvoet, A.J., Menkveld, H.W.H., J-C., Schrotter and Graaf, J.H.J.M. van der. (2010). *Prefiltration of wastewater effluent: effects on foulants and performance during dead-end ultrafiltration.* Desalination, 250, 2, 855 – 860.

Miska – Markusch, V. (2009). *Effluent filtration for more than particle removal.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent*. Phd thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Rosenberg, S., Evenblij, H., Poele, S. te, Wintgens, T. and Laabs, C. (2005). *The importance of liquid phase analyses to understand fouling in membrane assisted activated sludge processes – six case studies of different European research groups.* Journal of Membrane Science, 263, 113 – 126.

Zheng, X., Ernst, M, Huck, P.M. and Jekel, M. (2010). *Biopolymer fouling in dead-end ultrafiltration of treated domestic wastewater.* Water Research, 44, 18, 5212 – 5221.

5 Filtration properties and pretreatment

5.1 Introduction

An important trend in the development of ultrafiltration of WWTP effluent is the integration of pretreatment in order to improve the process performance of membrane installations. In addition to the improvement of process performance, pretreatment is applied to enhance the rejection efficiencies of aquatic substances (e.g. organic contaminants, heavy metals, etc.). But in general pretreatment is mainly applied to control membrane fouling (Huang *et al*., 2007). The major pretreatment technologies applied in practice are coagulation, adsorption and prefiltration (Huang *et al*., 2007). Of these technologies, coagulation is most widely adopted with aluminium or iron salts as coagulants. Consequently, many researchers have investigated the ins and outs of this technology. Also te Poele (2005) and Roorda (2004) have investigated the effect of (in-line) coagulation and prefiltration (continuous sand filtration and multi media filtration) on the concentrations of foulants in WWTP effluent, fractions of WWTP effluent and the performance of an ultrafiltration pilot installation. Both researchers concluded that with these pretreatment technologies only a moderate improvement of the filterability can be obtained. The particles of the size fraction < 0.45 µm still remained in the WWTP effluent after pretreatment. Unfortunately, particles of this size fraction have been considered to be of a major influence on the filterability of WWTP effluent (Roorda, 2004; te Poele, 2005; Zheng *et al*., 2009). Therefore, both te Poele (2005) and Roorda (2004) recommended to investigate the effect of alternative pretreatment technologies in order to remove the size fraction < 0.45 µm and consequently improve the filterability of WWTP effluent. Considering these recommendations research has been undertaken to investigate the effect of four alternative pretreatment technologies: powdered activated carbon, granulated activated carbon filter, multi media filter and 1-STEP® filter. In this chapter the experimental set-ups, results and conclusions of the investigations are presented. The study is undertaken with two specific objectives: (1) to determine the effect of the four pretreatment technologies on the (ultra)filtration properties and concentrations of foulants of WWTP effluent and (2), to determine in which particle size range the most particles or colloidals are removed by the pretreatment technologies.

5.2 Powdered Activated Carbon

5.2.1 General

Powdered activated carbon is often combined with ultrafiltration and microfiltration to alleviate membrane fouling and to improve the effectiveness of the removal of contaminants. (Shon *et al*., 2004a; Mozia *et al*., 2005; Lee *et al*., 2005). In spite of the wide application the effect of powdered activated carbon on membrane fouling is still unclear and seems to be dependent on the membrane material, operation mode, membrane configuration and type of feedwater. In the literature (see chapter 2) both reduction of membrane fouling as well as significant decreases of flux by powdered activated carbon addition are published. Therefore experiments on lab and pilot scale have been conducted to investigate the effect of powdered activated carbon addition on the filtration properties of WWTP effluent during dead-end ultrafiltration. In this section the results of these experiments are presented and discussed. On lab scale two experiments have been performed with batch samples of effluent of WWTP Berkel and Horstermeer. On pilot scale one experiment at the WWTP Maasbommel has been performed.

5.2.2 Lab scale experiments

On lab scale two experiments have been performed. The first experiment was carried out to investigate the effect of two different types of powdered activated carbon on the filtration properties of WWTP effluent. The types of powdered activated carbon have been selected in consultation with the supplier Norit. The second experiment was done to investigate the potential of powdered activated carbon to increase the initial filterability of WWTP effluent.

All lab scale experiments have been performed at the laboratory of Sanitary Engineering of Delft University of Technology. The effluent samples have been taken from the WWTP Berkel and WWTP Horstermeer. The effluent sample of WWTP Berkel was sieved (400 µm) during sampling and the effluent sample of WWTP Horstermeer was taken after the curved sieve (450 µm) of the pilot plant. The experiments in the laboratory were done on the same day as sampling or after storage the sample for the maximum of one day in a refrigerator (5 °C).

5.2.2.1 Experimental setup

Effect of two different types of powdered activated carbon

Two litres of (sieved) WWTP effluent were mixed with a dosage of powdered activated carbon for 1 hour at 100 rpm in a 2.2 liter vessel. After mixing the powdered activated carbon and WWTP effluent were separated by paper filtration (Schleichel&Schuell 589[2]). The filtrate was collected and then the SUR of the filtrate was measured two times. Also the blanc (WWTP effluent without dosage of powdered activated carbon) was filtrated with the paper filter prior to the SUR measurement of the filtrate. During mixing of the WWTP effluent and powdered activated carbon the temperature of the mixture was kept at 20 °C. The applied

concentrations of powdered activated carbon during this experiment were 0, 10, 25 and 50 mg/L. Foulants were analysed according to the procedures presented in chapter 3.

Two types of powdered activated carbon were selected for the first experiment: NORIT SAE SUPER and NORIT SA UF. In this section NORIT SAE SUPER and NORIT SA UF correspondent respectively to PAC-A and PAC-B. Both types of powdered activated carbon are coal based. Coal based carbon was selected since its low surface acidity had a higher natural organic matter uptake by preferentially removing aromatic compounds from natural organic matter than wood based carbon (Oh *et al*., 2006). Table 5.1 shows the main properties of the types of powdered activated carbon. These data have been provided by Norit.

Table 5.1 – Properties of the powdered activated carbon used in the experiments (provided by Norit)

	Unit	PAC-A	PAC-B
Activation method	-	Steam	Steam
Iodine number	-	1150	1100
Methylene blue adsorption	g/g	0.28	0.24
Phenol adsorption	weight %	-	5
Total surface area	m^2/g	1300	1200
Apparent density	kg/m^3	425	160
Particle size > 150 µm	weight %	3	-
Particle size D_{50}	µm	15	7
Ash content	weight %	-	10
pH	-	Alkaline	Alkaline
Moisture	weight %	-	2

As shown in Table 5.1 the main differences between both types of powdered activated carbon are the particle size (D_{50}) and the apparent density (kg/m^3). The particle size of PAC-B is approximately half the particle size of PAC-A. The apparent density relates to the porosity of the powdered activated carbon (Dikkenburg, 2006). PAC-B consists of more mesopores (classification of pores between the macro and micropores) per volume unit than PAC-A. Consequently, at the same concentration of powdered activated carbon in the mixture the volume of PAC-B is bigger than PAC-A.

Potential of PAC

To investigate the (maximum) potential of powdered activated carbon to improve filterability of the WWTP effluent two experiments were performed with the effluent of WWTP Berkel on different days. The experimental procedure was exactly the same as the previous experiment (effect of different types of powdered activated carbon). Only the applied concentrations of powdered activated carbon differed. During the first experiments the concentrations of powdered activated carbon were 0, 50, 100 and 200 mg/L and at the second experiment 0, 100, 200, 300, 400 and 500 mg/L.

5.2.2.2 Effect of two different types of powdered activated carbon

SUR values

In Table 5.3 the results of the experiments are presented. Small differences between the initial SUR values of PAC-A and PAC-B were observed at both WWTPs. This is probably a consequence of the experimental procedure. The experiments with PAC-A were performed on the day of sampling and transporting. The experiments with PAC-B were performed with the same WWTP effluent sample but one day later. In between the WWTP effluent samples were stored at 5 °C but taken into account the variation of the SUR measurement, the effect of storage was minimal.

Generally, Table 5.3 presents a decrease of the SUR values when the concentrations of PAC-A and PAC-B increase. Especially when the concentrations of PAC-A and PAC-B were about 50 mg/L the decline was considerable. Nevertheless, this decline is more obvious to PAC-B than PAC-A. This observation is also displayed in Figure 5.1 where the relative SUR values of Table 5.2 are given. The maximum SUR decrease with PAC-A is 18% and 7% for respectively effluent of the WWTP Berkel and WWTP Horstermeer. In comparing, the initial SUR value decreased by 40% (WWTP Berkel) and 35% (WWTP Horstermeer) when 50 mg/L of PAC-B was applied. At lower concentrations (10 and 25 mg/L) the relative difference between PAC-A and PAC-B was less, but in general the trend was similar. Therefore, PAC-B seems to be more effective than PAC-A considering the decrease of the initial SUR value.

Table 5.2 – SUR values of the effluent of WWTP Berkel and WWTP Horstermeer after addition, mixing and paper filtration of different dosages of PAC-A and PAC-B

	WWTP Berkel		WWTP Horstermeer	
PAC (mg/L)	PAC-A SUR (10^{12} m^{-2})	PAC-B SUR (10^{12} m^{-2})	PAC-A SUR (10^{12} m^{-2})	PAC-B SUR (10^{12} m^{-2})
0	13.6	14.3	10.9	11.3
10	12.6	12.5	11.2	10.1
25	12.4	11.8	11.5	10.0
50	11.1	8.6	10.1	7.8

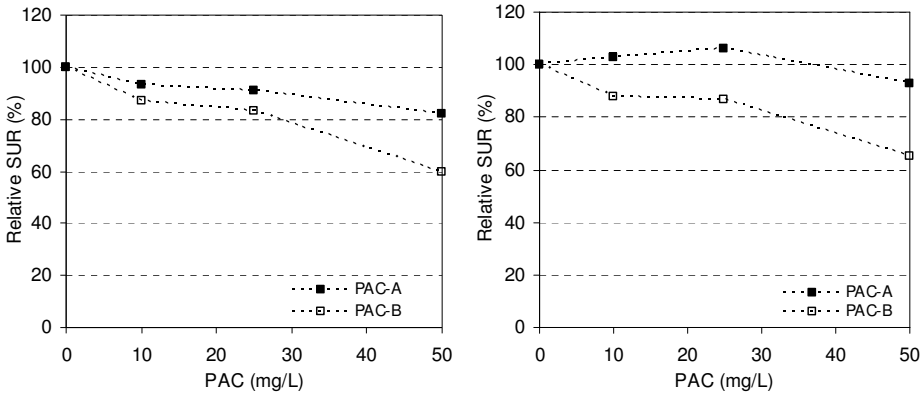
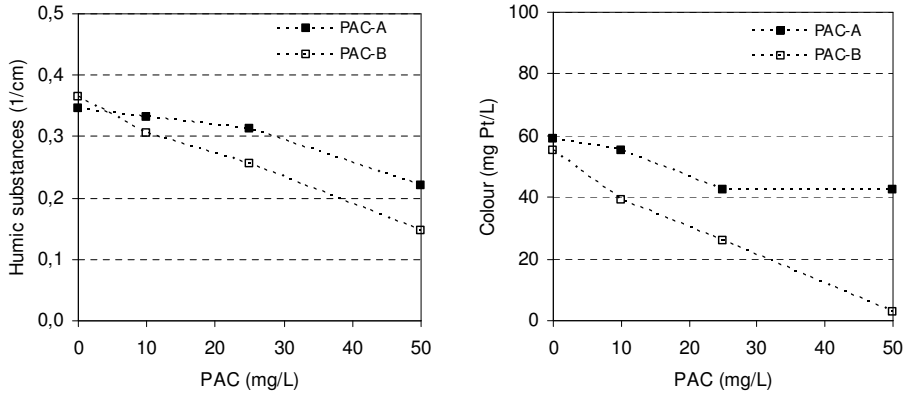


Figure 5.1 – Relative SUR values of the effluent of WWTP Berkel (left) and WWTP Horstermeer (right) after addition, mixing and paper filtration of different dosages of PAC-A and PAC-B

Foulants

Figure 5.2 displays the concentrations of foulants after mixing (1 hour at 20 °C) the effluent of WWTP Berkel with different dosages of PAC-A and B. Generally, all foulants present more or less the same trend. An increasing dosage of powdered activated carbon resulted in lower concentrations except for polysaccharides. Polysaccharides presented a lower adsorption affinity compared to the other compounds. Next to this observation, Figure 5.2 presents a significant difference. At the same dosage of powdered activated carbon, temperature and mixing conditions PAC-B adsorbed more foulants than PAC-A. However, for polysaccharides this difference was less significant than for the other foulants.



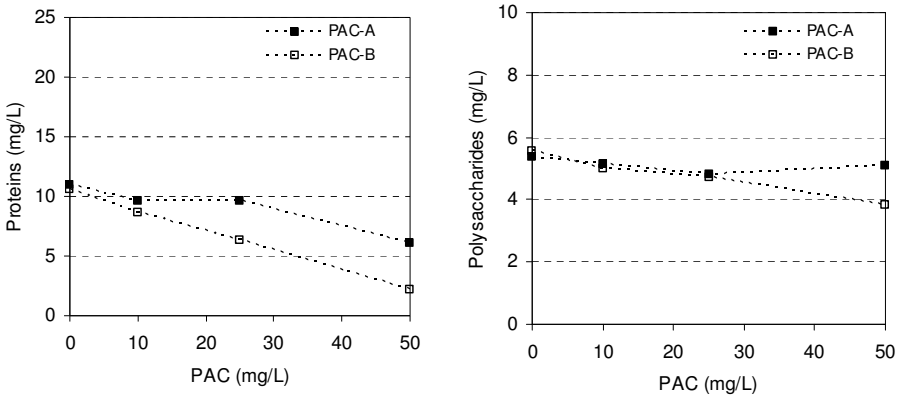
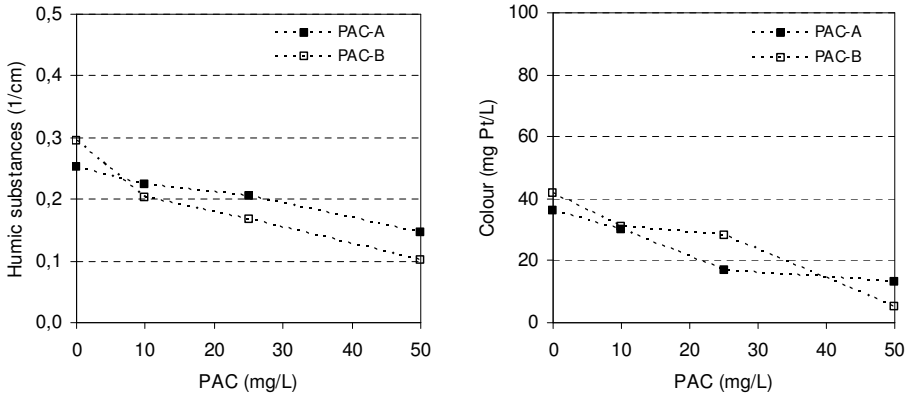


Figure 5.2 – Concentrations of humic substances, colour, proteins and polysaccharides in the water phase of the effluent of WWTP Berkel after addition and mixing of different PAC-A and PAC-B dosages

Figure 5.3 displays the results of the experiment with the effluent of WWTP Horstermeer. As expected, the graphs of this figure present the same trend as Figure 5.2. Increasing dosages of powdered activated carbon resulted in lower concentrations of foulants in the water phase. Nevertheless the difference between PAC-A and B was less significant than the results of WWTP Berkel (Figure 5.2). But still the results indicate that PAC-B adsorbs more foulants than PAC-A considering the defined mixing conditions and temperature.



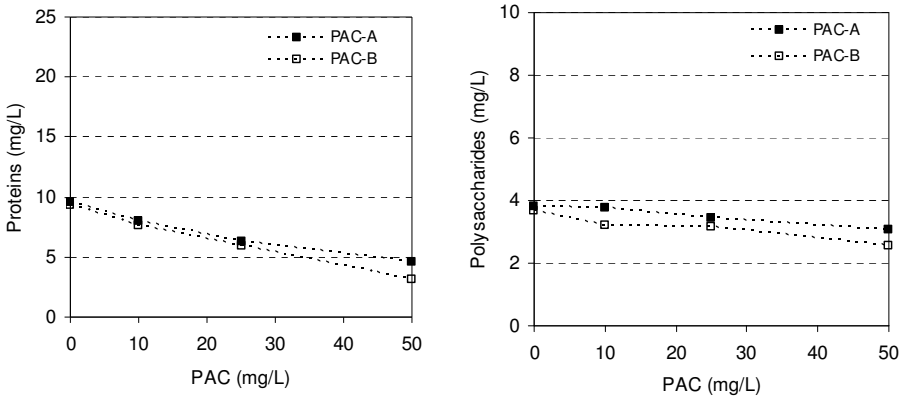


Figure 5.3 – Concentrations of humic substances, colour, proteins and polysaccharides in the water phase of the effluent of WWTP Horstermeer after addition and mixing of different PAC-A and PAC-B dosages

5.2.2.3 Potential of PAC

SUR values

The potential of PAC-B to decrease the SUR value of WWTP effluent was further determined during two experiments (I and II) with effluent of the WWTP Berkel. For both experiments batch samples were taken on two different days. Unfortunately the initial SUR values (powdered activated carbon concentration is 0 mg/L) of both effluent samples presented a large difference. The initial SUR values of the first and second experiment were respectively $31.2 \cdot 10^{12}$ m^{-2} and $11.3 \cdot 10^{12}$ m^{-2} (Table 5.4). This wide range was also determined by Roorda (2004). Roorda (2004) presented SUR values in the range of $6 \cdot 10^{12}$ – $40 \cdot 10^{12}$ m^{-2} of the effluent of WWTP Berkel. Therefore it was concluded that the measured initial SUR values in this thesis are not extraordinary. Nevertheless more similar initial SUR values are preferred for this kind of experiment.

Furthermore Table 5.3 shows for both experiments a significant decrease of the SUR values due to powdered activated carbon addition, mixing and (paper) filtration. Especially in the first phase a rapid decline was observed. The results of experiment I showed a strong decrease of the SUR value between the powdered activated carbon concentrations of 0 and 50 mg/L. For experiment II this strong decrease was observed between the powdered activated carbon concentrations of 0 and 100 mg/L. Nevertheless, increasing dosages of PAC-B still resulted in a decrease of the SUR value but the rate was less compared to the first phase (0 – 100 mg/L).

Table 5.3 – SUR values of the effluent of WWTP Berkel after addition, mixing and paper filtration of different PAC-B dosages

Experiment	I	II
PAC (mg/L)	SUR (10^{12} m^{-2})	SUR (10^{12} m^{-2})
0	11.3	31.2
50	6.6	-
100	5.6	20.0
200	4.1	13.6
300	-	13.8
400	-	8.7
500	-	5.9

In Figure 5.4 the relative SUR values of Table 5.4 are presented. As noticed already earlier, a rather similar trend was observed during both experiments. An initial fast decrease of the SUR value was followed by a more gradual and smaller decrease. Figure 5.4 shows also that the minimal SUR value (i.e. maximal decrease of the SUR value) was not obtained during both experiments. The SUR value did not stabilize even when relative high dosages of PAC-B were applied. On the other hand the results showed that the combination of powdered activated carbon addition and mixing followed by filtration may result in a good filterability of the WWTP effluent, even when the initial SUR values of the WWTP effluent are high.

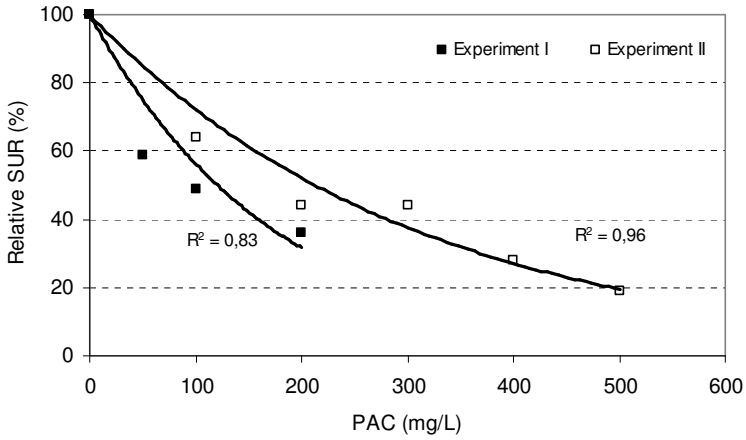


Figure 5.4 – Relative SUR values of the effluent of WWTP Berkel after addition, mixing and paper filtration of different PAC-B dosages

Foulants

Figure 5.5 presents the changes in concentrations of humic substances, colour, proteins and polysaccharides in the water phase of the effluent of WWTP Berkel. In spite of the different initial SUR values (Table 5.3) the initial concentrations of the foulants were more or less similar. Also after addition and mixing the effluent samples with 100 and 200 mg/L of PAC-B the concentrations of foulants were almost similar. Like the results of the SUR measurements,

especially in the first phase (0 – 100 mg/L of PAC-B), the adsorption was effective. Increasing dosages still resulted in lower foulants concentration in the water phase, but the effectiveness was less compared to the first phase. Contrary to the SUR values the concentrations of foulants became more or less stable. Increasing dosages (beyond 100 mg/L) did not result in further significant removal of foulants. Therefore the results indicate that a PAC-B dosage in the range of 50 – 150 mg/L is capable to adsorb the foulants of the effluent of WWTP Berkel, assuming conditions of temperature and mixing as in this experiment.

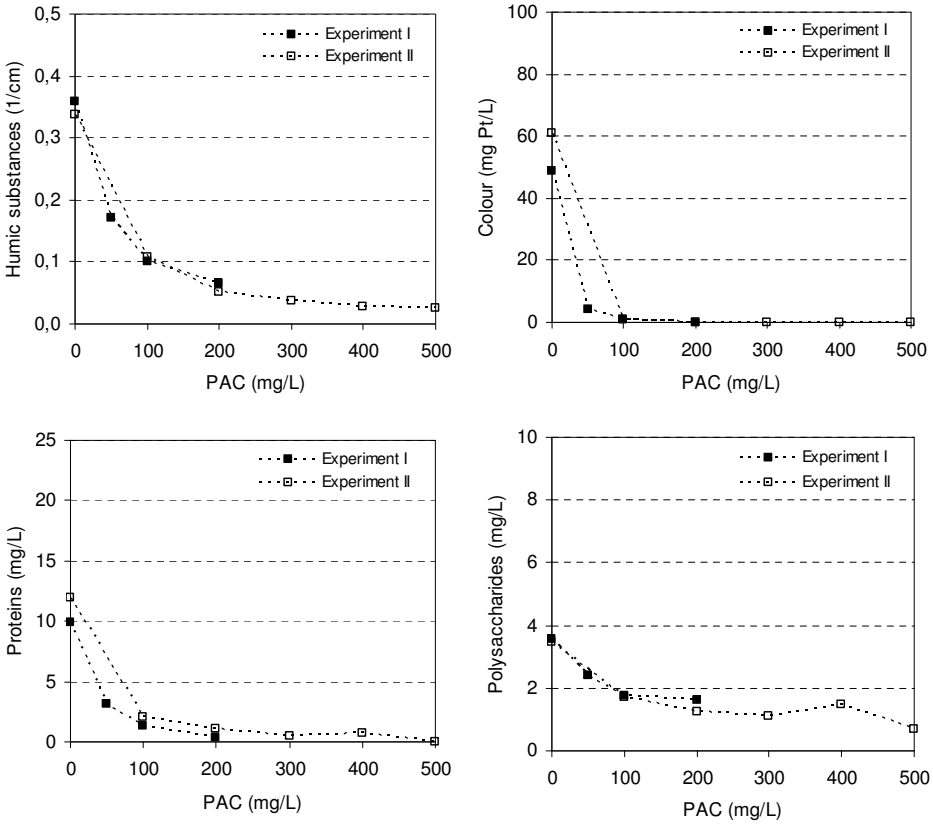


Figure 5.5 – Concentrations of humic substances, colour, proteins and polysaccharides in the water phase of the effluent of WWTP Berkel after addition and mixing of PAC-B dosages

5.2.2.4 Discussion of lab scale experiments

The procedure of the first experiment to determine and to compare the effect of two different types of powdered activated carbon differed from the procedure of Norit to measure adsorptive capacity of powdered activated carbon. Norit stipulates that, first, the optimum contact time has to be determined, followed by experiments to measure the total adsorptive capacity. Then, secondly, the adsorption isotherm must be plotted to compare the performance of the different types of powdered activated carbon. In variance with Norit's recommendation it was decided that the contact time in practice should be limited to 1 hour. Because the contact time was the same in all experiments, this departure from the rule has no influence on the results of the comparison of both types of PAC.

The difference between the performance of PAC-A and PAC-B can be explained by two parameters of the powdered activated carbon. As shown in Table 5.1 the particle size (D_{50}) of PAC-A was approximately two times larger than that of PAC-B. From literature (Matsui *et al*., 2004) it is known that a smaller particle size results in a larger total outer surface area and faster adsorption kinetics. Another aspect that may explain the difference is the presence of mesopores. PAC-B consists of relative more mesopores than PAC-A (Dikkenburg, 2006). This difference resulted also in a lower apparent density of PAC-B compared to PAC-A because more mesopores means more space and area for adsorption.

Generally the results of both experiment showed that powdered activated carbon addition and mixing followed by filtration resulted in adsorption of foulants and an increase of the filterability of WWTP effluent. Especially the humic substances, colour and proteins were adsorbed. This finding is confirmed in literature. Haberkamp *et al*. (2007) reported that (powdered) activated carbon adsorbs organic compounds of a wide range of molecular weights. Haberkamp *et al*. (2007) also noticed that low molecular weight substances (i.e. colour and humic substances) consume more of the adsorption capacity than biopolymers (i.e. proteins and polysaccharides). Similar observations were found during both experiments. The humic substances and colour were more rapidly removed than proteins and polysaccharides.

Furthermore the results indicate no clear relation between filterability and the measured samples. For example during the second experiment (potential of PAC) different initial SUR values were measured but the concentrations of foulants were more or less similar. This discussion item will be further investigated and discussed in sections 5.4 – 5.6.

In spite of the results the actual relation between powdered activated carbon addition and the SUR values seems to be in some way inefficient: only with high dosages of powdered activated carbon the SUR reaches the required low values. Therefore, more attention should be paid to optimize the efficiency of the powdered activated carbon addition, for example by

combining with the addition of coagulants or recirculation of the partially loaded powdered activated carbon.

5.2.3 Pilot scale experiment

5.2.3.1 Experimental setup

At the WWTP Maasbommel research had been performed by the local water board as part of a research program of the Foundation for Applied Water Research (STOWA). The objective of the STOWA research was to assess the performance of advanced treatment technologies with the aim of enhancing the effluent quality to the level of the Dutch Maximum Tolerable Risk (van Betuw *et al.*, 2007). One of the tested advanced treatment technologies was the combination of coagulation/flocculation (Nalmet) and powdered activated carbon adsorption followed by ultrafiltration. This ultrafiltration pilot installation was operated during the period February – May, 2007. The main operational parameters are given in Table 5.4. Further detailed information about the coagulant/flocculant, type of powdered activated carbon and membranes can be found in chapter 3.

Table 5.4 – Operational parameters of the flocculation reactor, powdered activated carbon reactor and ultrafiltration membranes during the experimental period February – May, 2007

Parameter	Unit	Value
Coagulation/Flocculation reactor		
Dosage Nalmet	L/m^3	0.03
Flow	m^3/h	5
Coagulation time	h	1.6
Flocculation time	h	1.8
Powdered activated carbon reactor		
Flow	m^3/h	5
Dosage PAC	mg/L	40
Concentration PAC in contact tank	g/L	3
Residence time PAC	d	± 30
Ultrafiltration membranes		
Flux	$L/m^2 \cdot h$	22.7

During the experimental period, at five times grab samples were taken of the WWTP effluent behind the 5 mm screen, the coagulation/flocculation reactor and the powdered activated carbon reactor. Samples of the coagulation/flocculation reactor were taken from the settling tank and samples of the powdered activated carbon reactor from the contact reactor (see Figure 3.6). After collecting, the samples were transported directly to the laboratory of Sanitary Engineering of Delft University of Technology to carry out foulants analyses, SUR measurements and fractionations (Table 5.5). If possible the fractionations and SUR measurements were performed on the day of sample collection. If not, the samples were stored at most one day at 5 °C in a refrigerator. For foulants analyses the samples were filtered on the day of sampling. Then the foulants were analysed within at least one week after filtration.

Table 5.5 – Dates of sampling, measurements and analyses of the WWTP effluent, settling tank and PAC contact tank of the ultrafiltration pilot installation at the WWTP Maasbommel during the experimental period February – May, 2007

Sample date	WWTP effluent	Settling tank	PAC contact tank
26-02-07	SUR and foulants	SUR and foulants	SUR and foulants
05-03-07	SUR and foulants	SUR and foulants	SUR and foulants
13-03-07	SUR, foulants and fractionation	SUR, foulants and fractionation	SUR, foulants and fractionation
04-04-07	SUR, foulants and fractionation	SUR, foulants and fractionation	SUR, foulants and fractionation
07-05-07	SUR, foulants and fractionation	SUR, foulants and fractionation	SUR, foulants and fractionation

5.2.3.2 SUR values

Figure 5.6 shows the SUR values of the effluent of WWTP Maasbommel, the settling tank and the powdered activated carbon contact tank of the ultrafiltration pilot installation. Before the SUR measurements of the samples of the powdered activated carbon contact tank the samples were filtered over a 0.45 µm cellulose acetate filter to separate the powdered activated carbon and water.

After coagulation, flocculation and settling of Nalmet the SUR values in the settling tank were most of the time lower than the WWTP effluent. Only the first measurement (26-02-07) showed an increase but this was caused by an improper SUR measurement. The other four measurements presented an average SUR decrease of 47%. Surprisingly after the settling tank, in the powdered activated carbon contact tank, the SUR values increased drastically, even when the samples were filtrated over a 0.45 µm cellulose acetate filter. Considering the particle size of the applied powdered activated carbon (D_{50} = 15µm) it was expected that the carbon would be completely separated by filtration over a 0.45 µm filter. Therefore other aspects may play a role like the retention of small particles or the pulverization of the powdered activated carbon. However the role of these aspects was not investigated during the experiment.

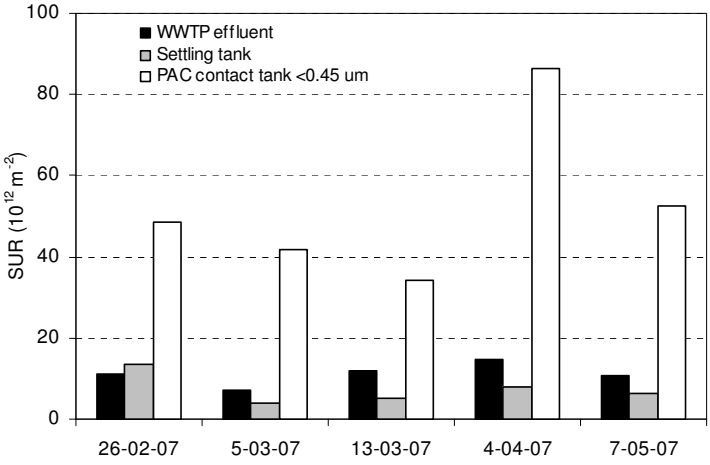


Figure 5.6 – SUR values of the effluent of WWTP Maasbommel, settling tank and powdered activated carbon contact tank (< 0.45 µm) of the ultrafiltration pilot installation during the period February – May, 2007

5.2.3.3 Foulants

The results of the foulants analyses are presented in Figure 5.7. In general it was noticed that the concentrations of DOC, proteins, humic substances and colour decreased after the different treatment steps. Considering the results of the lab scale experiments a removal of these compounds was expected. Especially the low molecular weight compounds (humic substances and colour) show a higher tendency to activated carbon (Tomaszewska and Mozia, 2002) than e.g. proteins. Contrary to the partly adsorbed compounds (DOC, proteins, humic substances and colour) the concentration of polysaccharide increased in the powdered activated carbon tank. It seemed that the polysaccharides were less tended to adsorb than the other compounds. This observation proves the findings of the lab scale experiments. These experiments showed also less removal of polysaccharides compared to other foulants.

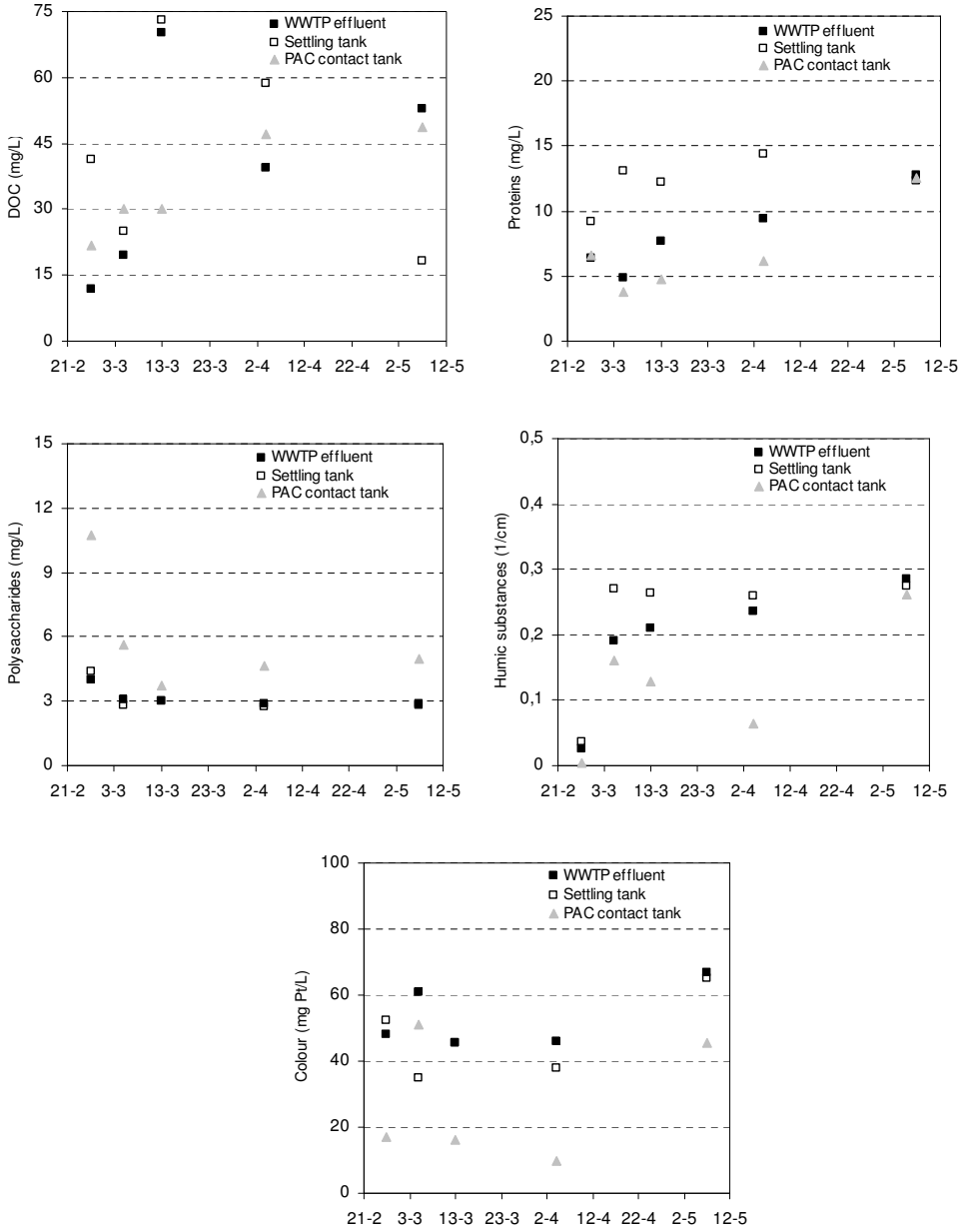


Figure 5.7 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour of the WWTP effluent, settling tank and powdered activated carbon contact tank of the ultrafiltration pilot installation at the WWTP Maasbommel during the period February – May, 2007

5.2.3.4 Fractionation

Figure 5.8 displays the results of the fractionation measurements of the different treatment steps of the ultrafiltration pilot installation. All the fractions of the samples were measured except the samples of the powdered activated contact tank. Of these samples the fraction > 0.45 µm had not been measured. Considering the pore size of the filter (0.45 µm) and the particle size of the applied powdered activated carbon (D_{50} = 15µm) it was assumed that the filtrate did not contain any particle of applied powdered activated carbon. Nevertheless in Figure 5.8 a drastic increase of the SUR value can be observed in the powdered activated carbon contact tank during all the sample dates. Therefore it seemed that the fraction 0.1 – 0.2 µm accumulated in the powdered activated carbon contact tank resulting in high SUR values. An explanation for this could be the membrane pore size of the ultrafiltration membranes. As shown in chapter 3 the membrane pore size is 0.1 µm. Beside a part of the powdered activated carbon in the ultrafiltration tank was circulated back to the powdered activated carbon contact tank. Evidently, the slurry with a high content of particles in the size range of 0.1 – 0.2 µm circulated in both the powdered activated carbon and ultrafiltration tank. Unfortunately this suggestion was not proven by additional measurements of the recirculation flow because no fractionation measurements were performed of this flow.

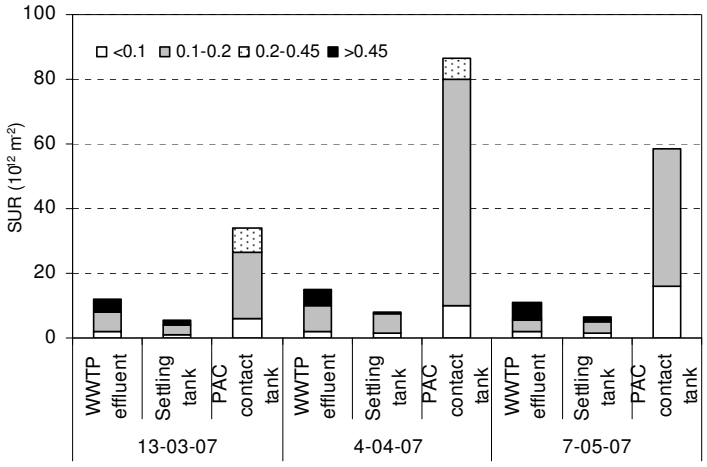


Figure 5.8 – Contribution per fraction to the total SUR value of the WWTP effluent, settling and PAC contact tank of the ultrafiltration pilot installation at the WWTP Horstermeer during the period February – May, 2007

Despite the increased SUR values in the powdered activated carbon contact tank a decrease of the SUR values of WWTP effluent was observed in the upstream tank (settling tank). After coagulation, flocculation and settling the average relative SUR decrease was 47%. It seemed from Figure 5.8 that this decrease may mainly be caused by the settlement of particles > 0.45 µm. During the three measurements the average relative SUR decrease of this fraction was

79%. This observation is in line with results of Roorda (2004). That tests presented also a decrease of the impact of the fraction > 0.45 µm of WWTP effluent after coagulation. The overall relative SUR decrease presented by Roorda (2004) after coagulation was found to be in the range of 20% to 30%. This is less than the overall relative SUR decrease displayed in Figure 5.8 (47%) but the difference between those percentages depends of course on the configuration of the coagulation/flocculation process and the local circumstances.

5.2.3.5 Discussion of pilot scale experiments

The results of the foulants analysis corresponded to the findings of the lab scale experiments. The proteins, humic substances and colour were partly adsorbed by the powdered activated carbon. These observations are also reported by different researchers during other conducted studies (Shon *et al*., 2004b; Mozia *et al*., 2005; Haberkamp *et al*., 2007). Despite the decrease of foulants the SUR values increased drastically in the powdered activated carbon tank. These results suggested there was no correlation between the concentration of foulants and the SUR values. This suggestion is confirmed by studies of te Poele (2005). Furthermore the results indicate that the increased SUR values in the powdered activated carbon tank were caused by the accumulation of particles in the size range of 0.1 – 0.2 µm. For example the powdered activated carbon was concentrated 75 times (concentration in PAC tank 3 g/L and dosage of PAC 40 mg/l). Although air was supplied in the ultrafiltration tank for air scourcing this could not prevent the accumulation of these particles (0.1 – 0.2 µm). Air (oxygen) could probably stimulate the biodegradation of these particles. However the investigation of this effect was not included in the experimental set-up.

As mentioned the particle size range of 0.1 – 0.2 µm dominated the filterability during the SUR measurements. Considering the results of this study it seemed in hindsight that the configuration of the ultrafiltration pilot installation was not well designed. During the design of an ultrafiltration (pilot) installation the accumulation of small particles (< 0.2 µm) should be prevented. This suggestion is supported by the reported operational experience of the ultrafiltration pilot installation during the whole research period (April, 2006 – May, 2007). In the report (STOWA, 2007) the researchers reported decreasing permeability values and even fibre breakage was observed. The fibre breakage seemed to be related to the scouring effect of powdered activated carbon (STOWA, 2007).

Contrary to the drastic increase of the SUR values the coagulation and flocculation of the Nalmet resulted in a decrease of the SUR value of WWTP effluent. Compared to other experiences with coagulants (Roorda, 2004) the Nalmet performed well but unfortunately only predominately the fraction > 0.45 µm was removed. Considering the possibility to remove heavy metals with Nalmet the combination of Nalmet and ultrafiltration may be an option for (industrial) applications in water reuse.

5.3 Granulated Activated Carbon Filter

5.3.1 General

The combination of granulated activated carbon filtration and membrane filtration is presented by a few researchers in the literature. Despite the few numbers of publications several authors presented promising results of this combination. For example, Tsujimoto *et al*. (1998) suggested that granulated activated carbon pretreatment is effective to prevent irreversible fouling and to obtain stable operation during ultrafiltration of river water with cellulose acetate membranes. Another study of Roorda *et al*. (2005) presented results of the combination of granulated activated carbon and nanofiltration (ANF-process). This process was applied to treat WWTP effluent and presented similar costs to direct nanofiltration due to higher fluxes, less chemical cleanings, etc.. Considering these findings and experiences the effect of granulated activated carbon filtration on the filterability, foulants and fractions of WWTP effluent was investigated in the context of this thesis. In this section the experimental setup and results are described and discussed.

5.3.2 Experimental setup

At the WWTP Maasbommel a granulated activated carbon filter was operated during a period of 11 weeks from February - May, 2007. A detailed description of the granulated activated carbon filter had been given in section 3.2. Before the start of the experimental period (February, 2007) the filter had been filled with fresh media. During the operational period at five times samples were taken of the WWTP effluent (behind 5 mm screen) and filtrate of granulated activated carbon filter (see Figure 3.6). The samples of the granulated activated carbon filter were taken 20 minutes after the samples of WWTP effluent and before the weekly backwash. After sampling different actions were performed (Table 5.6). Firstly the collected samples were transported directly to the laboratory of Sanitary Engineering of Delft University of Technology. Then secondly, if possible the fractionations and SUR measurements were performed on the day of sampling. If not, the samples were stored at most one day at 5 °C in a refrigerator. For foulants analyses the samples were filtered on the day of sampling and analysed within at least one week after filtration. In between the filtrated samples were stored at 5 °C in a refrigerator.

Table 5.6 – Dates of sampling, measurements and analyses of the WWTP effluent and the granulated activated carbon filtrate of the WWTP Maasbommel during the experimental period February – May, 2007

Sample date	WWTP effluent	GAC filtrate
26-02-07	SUR and foulants	SUR and foulants
05-03-07	SUR and foulants	SUR and foulants
13-03-07	SUR, foulants and fractionation	SUR, foulants and fractionation
04-04-07	SUR, foulants and fractionation	SUR, foulants and fractionation
07-05-07	SUR, foulants and fractionation	SUR, foulants and fractionation

During the operational period no additional chemicals were dosed upstream or in the granulated activated carbon filter. Despite the absence of chemical dosages (e.g. methanol)

biological activity occurred in the filter as expected. But this biological activity was not stimulated and considered as a fact.

The operational parameters of the GAC filter during the experimental period are summarized in Table 5.7.

Table 5.7 – Operational parameters of the granulated activated carbon filter at the WWTP Maasbommel during the experimental period February – May, 2007 (Van Betuw *et al.*, 2007)

Parameter	Unit	Value
Capacity	m^3/h	1 – 1.2
Empty Bed Contact Time	minutes	20
Volume activated carbon	m^3	0.4
Surface area	m^2	0.45
Hydraulic load	m/h	2.25 – 3
Backwash flow	m^3/h	13
Bed expansion at backwash	%	25
Number of backwashes	1/week	1

5.3.3 Results

5.3.3.1 SUR values

Figure 5.9 presents the SUR values of effluent from WWTP Maasbommel and the filtrate of the granulated activated carbon filter during the experimental period. Like the obtained experience at other WWTPs the SUR values of the effluent fluctuated over the time. The SUR values of the WWTP effluent were measured within the range of $7 \cdot 10^{12} – 15 \cdot 10^{12}$ m^{-2}.

Surprisingly, on the sample dates (26-2-07 and 5-3-07) the SUR values of the WWTP effluent and the granulated activated carbon filtrate were almost similar but on the other three sample dates the SUR values were significantly lowered. The SUR values decreased with 37% (13-3-07), 22% (4-4-07) and 28% (7-5-07). The difference between the results on the first sample dates (26-2-07 and 5-3-07) and the other sample dates (13-3-07, 4-4-07 and 7-5-07) may be related to the renewal of the granulated activated carbon in the filter. Just before the experimental period and the first sample dates the granulated activated carbon was renewed. From the literature (Shon, *et al*., 2004a) it is known that granulated activated carbon filters need some time to stabilize in order to function properly. In a similar experiment these authors took the first samples after 45 days of operation of a fixed bed adsorption system.

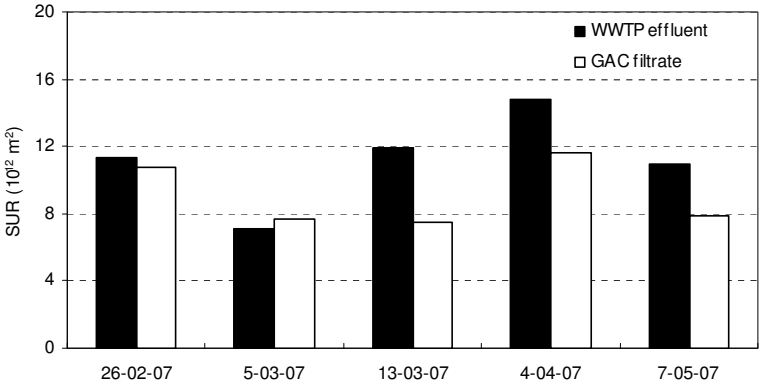
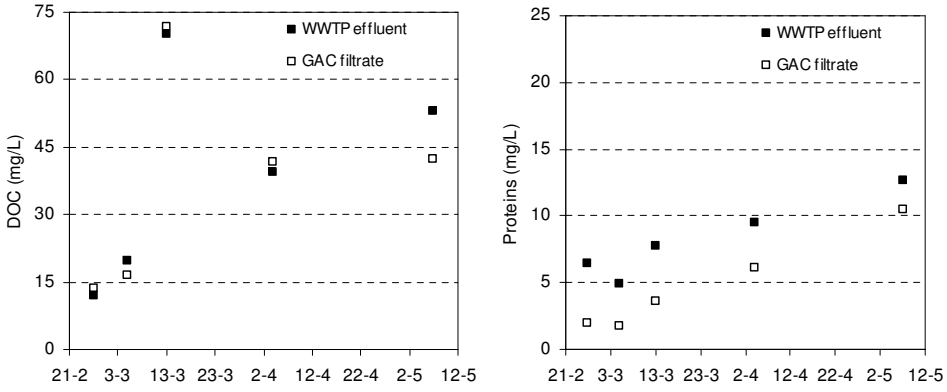


Figure 5.9 – SUR values of the WWTP effluent and the granulated activated carbon filtrate of the WWTP Maasbommel during the period February – May, 2007

5.3.3.2 Foulants

Figure 5.10 shows the results of the foulants analyses of the WWTP effluent and the granulated activated carbon filtrate. The DOC concentrations of both, the WWTP effluent and the granulated activated carbon filtrate were found to be in the range of 10 – 70 mg/L. Except for the last sample date (7-5-07) no significant removal of DOC was obtained. However, this observation was not proved by the results of the other foulants. The concentrations of proteins, humic substances and colour were note worthily lowered by the granulated activated carbon filter. Nevertheless the concentrations of polysaccharides did not present a significant decrease. These concentrations were almost similar before and after the granulated activated carbon filter.



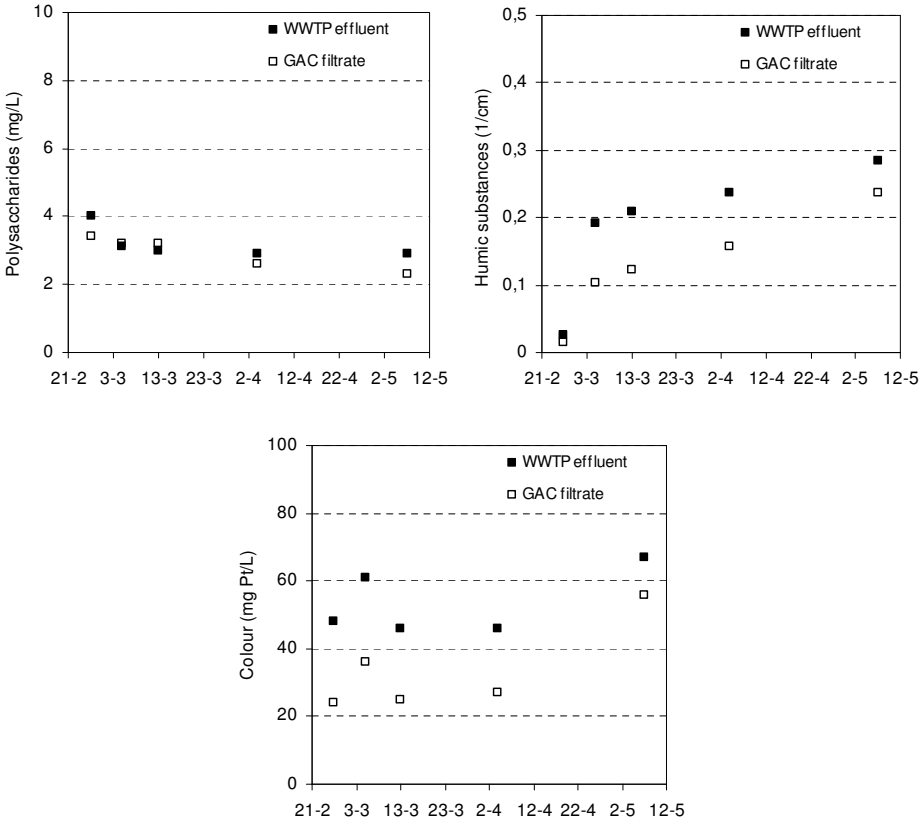


Figure 5.10 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour of the WWTP effluent and the granulated activated carbon filtrate of the WWTP Maasbommel during the period February – May, 2007

5.3.3.3 SUR values and foulants

In order to investigate a relationship between the filterability and concentrations of foulants, Figure 5.11 shows the obtained SUR values of WWTP effluent and GAC filtrate (see Figure 5.9) with the accompanying foulants concentrations (see Figure 5.10). The outcome do not confirm any relation between one the foulants and the filterability of WWTP effluent and GAC filtrate (e.g. higher concentrations of foulants did not result automatically in higher SUR values).

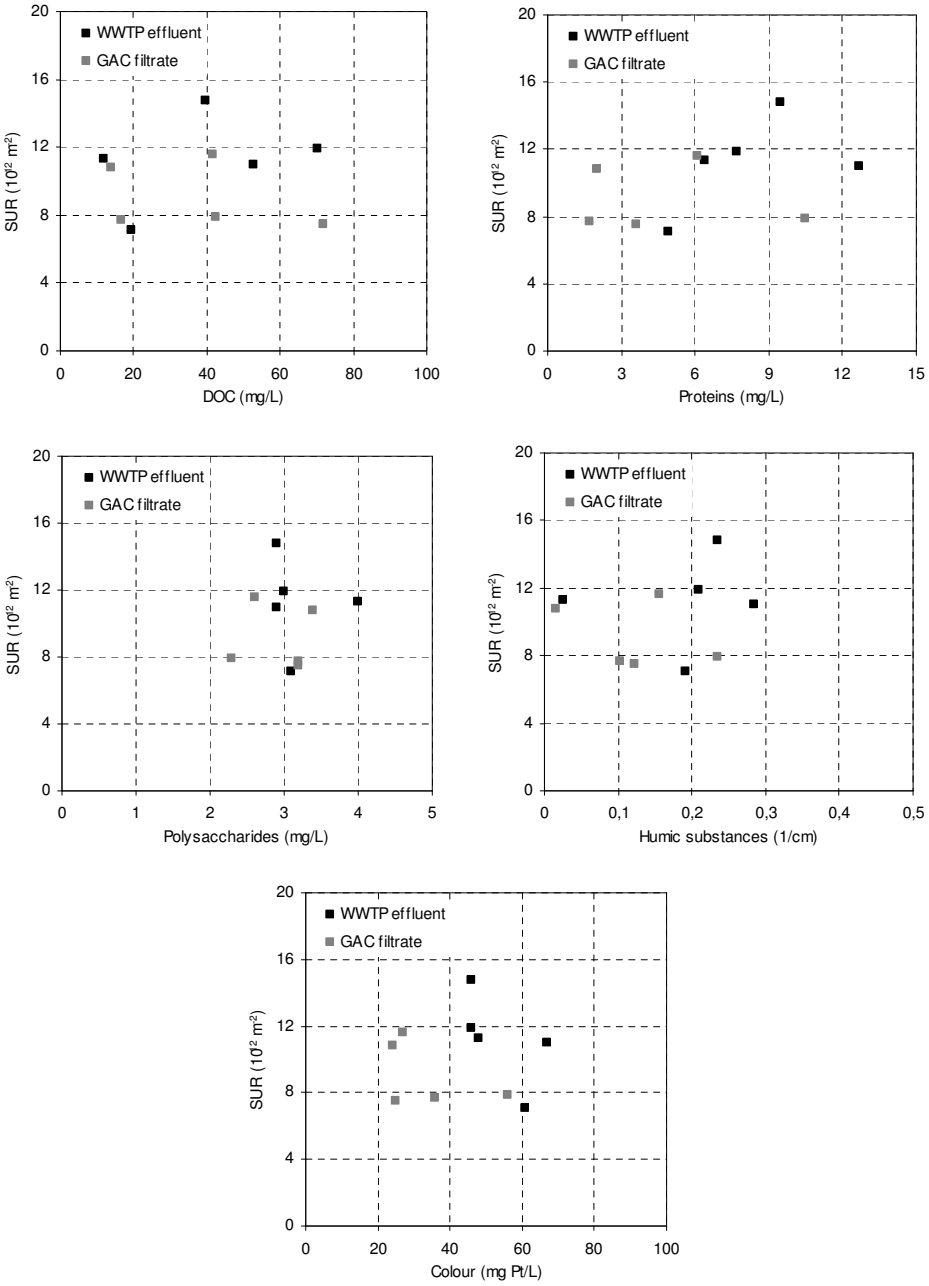


Figure 5.11 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour versus SUR values of the WWTP effluent and the granulated activated carbon filtrate of the WWTP Maasbommel during the period February – May, 2007

5.3.3.4 Fractionation

Figure 5.12 displays the contribution per fraction to the total SUR value of the effluent from WWTP Maasbommel and the granulated activated carbon filtrate. At three times samples were taken for fractionation. The fractionation results of the WWTP effluent show that the filterability is mainly dominated by the fractions > 0.45 µm and 0.1 – 0.2 µm. The contribution of the fraction > 0.45 µm to the total SUR value of WWTP effluent varied between 30% (4-4-07) and 45% (7-5-07). The fraction 0.1 – 0.2 µm contributed for 30% (7-5-07) to 54% (4-4-07).

The fractionation of the granulated activated carbon filtrate presents a different picture in Figure 5.12. The filtrate filterability was mainly dominated by the fraction 0.1 – 0.2 µm. This fraction was responsible for 53% (average of the three samples) of the total SUR value. The contribution of the fraction > 0.45 µm in the granulated activated carbon filtrate was between 10% (13-3-07) and 15% (4-4-07). Therefore it may be concluded that on the one hand granulated activated carbon filtration removed significantly the fraction > 0.45 µm but on the other hand did it not decrease the impact of the fraction 0.1 – 0.2 µm on the filterability.

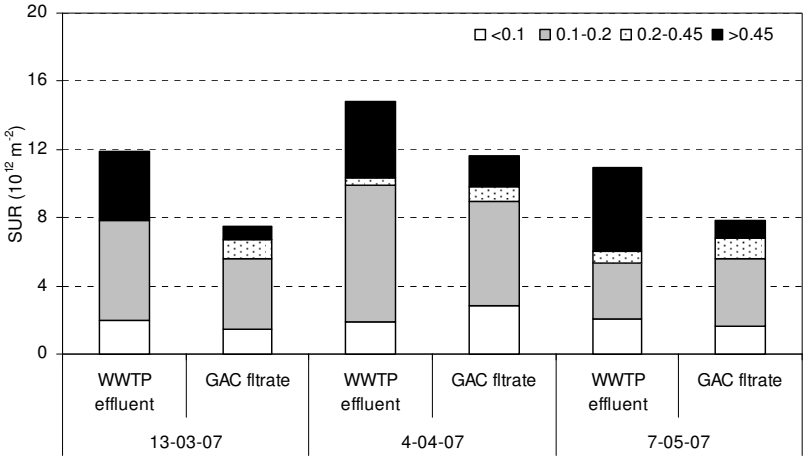


Figure 5.12 – Contribution per fraction to the total SUR value of the WWTP effluent and the granulated activated carbon filtrate of the WWTP Maasbommel during the period February – May, 2007

5.3.4 Discussion

In spite of the fact that only five samples were taken and analysed it seemed that granulated activated carbon filtration reduces the SUR value of WWTP effluent (Figure 5.13). During the last three sampling dates, when the granulated activated carbon filter was expected to be stabilized, the average relative SUR decrease was 29%. This relative decrease is also found during coagulation and multi media filtration of WWTP effluent. Roorda (2004) reported

relative SUR decrease values of approximately 20% to 30% for coagulation and multi media filtration but showed also that these values were greatly depending on local conditions. Yet, in terms of SUR decrease, it seemed that granulated activated carbon filtration performed similar as coagulation and multi media filtration.

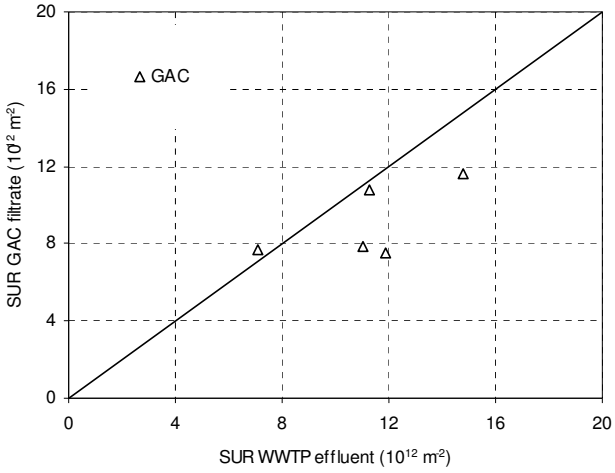


Figure 5.13 – Relation between the SUR values of WWTP effluent and the SUR values of GAC filtrate of the WWTP Maasbommel during the period February – May, 2007

The fractionation results showed that the decrease of the SUR value was mainly due to the removal of the fraction > 0.45 µm. The average contribution of this fraction was respectively 36% and 13% in WWTP effluent and granulated activated carbon filtrate. This is in contrast with the fraction 0.1 – 0.2 µm. This fraction was not influenced during granulated activated carbon filtration. Therefore it seemed that mainly the particles > 0.45 µm were entrapped in the granulated activated carbon filter. This observation was also made by Roorda (2004) for coagulation and multi media filtration. These pretreatment technologies entrapped also mainly the fraction > 0.45 µm .

Some foulants were removed during granulated activated carbon filtration. Especially the low molecular weight substances like colour and humic substances were partly removed. From the literature it is known that activated carbon has a high tendency to take up these aquatic substances (Huang, *et al.*, 2007). Therefore it can be assumed that these compounds were adsorbed during filtration. In addition to these compounds also the proteins (high molecular weight substance) were slightly removed. Proteins are high molecular weight substances and these substances do have a lower tendency to adsorb than low molecular weight substances

(Haberkamp *et al.*, 2007). Although, it seemed proteins were adsorbed as well. Proteins are too small to be filtered considering the particle size of the applied granulated activated carbon.

The compilation of the foulants concentrations and SUR values of both WWTP effluent and granulated activated carbon filtrate did not show any significant relation. This observation proves the results found by te Poele (2005). Te Poele (2005) showed no clear relations between the filterability and amount of foulants retained during lab and pilot scale experiments.

5.4 Multi Media Filter

5.4.1 General

Multi media filtration is a commonly applied pretreatment technology before ultrafiltration of WWTP effluent. In combination with ultrafiltration it is mainly exploited to increase the performance of the ultrafiltration unit by removing the particles of the WWTP effluent. This combination had been intensively investigated on pilot scale by Roorda (2004) and te Poele (2005) at different WWTPs in The Netherlands. Except for one pilot study (WWTP Utrecht) during all these studies the multi media filter was operated without upstream addition of a carbon source for denitrification. Mainly, only a coagulant was added upstream the multi media filter to improve the filtration performance. It was assumed by te Poele (2005) that the dosage of a carbon source (methanol) did not influence the ultrafiltration performance. This assumption is disputable due to the possible formation of extra (soluble) microbial products by denitrifying bacteria. Therefore research was carried out to investigate the performance of multi media filtration (in combination with methanol and coagulant addition) as pretreatment technology for ultrafiltration. In this section the results of the SUR measurements, fractionations and foulants are presented.

5.4.2 Experimental setup

At the WWTP Horstermeer tests were carried out during a period of 11 weeks (April – July, 2008) . The multi media filter was already in operation before the experimental period but the media (sand and anthracite) had been renewed just before the tests. During the experimental period the multi media filter was operated for denitrification and simultaneous phosphorous removal and was fed with WWTP effluent that passed a 450 µm curved sieve (see Figure 3.1). For the simultaneous phosphorous removal and removal of suspended solids poly aluminium chloride was dosed in-line into feedwater of the filter. For denitrification acetic acid or methanol was dosed based on the actual nitrate and free oxygen concentrations in the WWTP effluent. In Table 5.8 the operational parameters of the multi media filter are summarized.

Table 5.8 – Operational parameters of the multi media filter at the WWTP Horstermeer during the period April – July, 2008.

Parameter	Unit	Value
Flow	m^3/h	5.5 – 8.4
Hydraulic load	m/h	10
Filter run	h	12
Surface area	m^2	0.8
Frequency 'bumping cleaning'	1/d	8
Acetic acid dosing (April, 21 – June, 2)	g COD/g NO_3-N	3
Methanol dosing (June, 3 – July, 1)	g COD/g NO_3-N	4
PACl in metal orthophosphate ratio	mol/mol	4

Samples were taken of WWTP effluent (after the curved sieve), coagulated WWTP effluent (after static mixer) and filtrate of multi media filter. The samples of the filtrate of multi media filter were taken 20 minutes after the samples of WWTP effluent and at least two hours after a backwash. Samples of WWTP effluent and coagulated effluent were taken almost at the same time. After sampling different actions are performed which are summarized in Figure 5.14. The collected samples were transported directly to the laboratory of Sanitary Engineering of Delft University of Technology. Then, if possible the fractionations and SUR measurements were performed on the day of sampling. If not the samples were stored at most one day at 5 °C in a refrigerator. For foulants analyses the samples were filtered on the day of sampling and analysed within at least one week after filtration. In between the filtrated samples were stored at 5 °C.

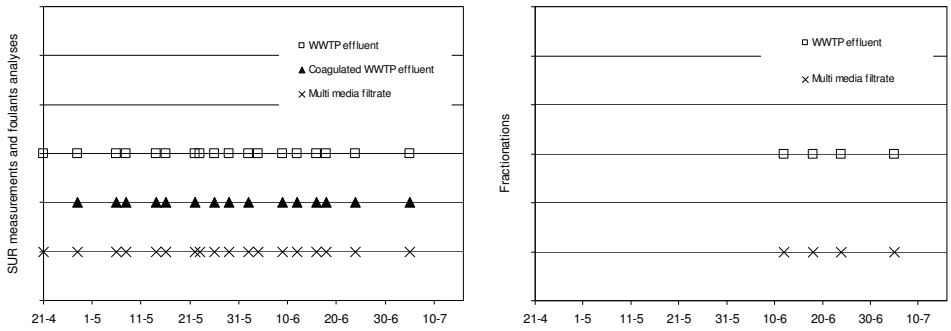


Figure 5.14 – Dates of sampling of the WWTP effluent, coagulated WWTP effluent and multi media filtrate and performed actions (SUR measurements and foulants analysis (left) and fractionations (right)) during the experimental period April – July, 2008

5.4.3 Results

5.4.3.1 SUR values

In Figure 5.15 it is shown that the SUR values of WWTP effluent varied between $5 \cdot 10^{12}$ m^{-2} and $17 \cdot 10^{12}$ m^{-2}. Based on the SUR values of WWTP effluent the experimental period can be divided in two phases. During the first phase, in the months April and May, SUR values in the

range of $10 \cdot 10^{12}$ – $17 \cdot 10^{12}$ m^{-2} were measured. During the second phase, in the months June and July, the SUR values of the WWTP effluent were lower and measured in the range of $5 \cdot 10^{12}$ – $7 \cdot 10^{12}$ m^{-2}. It is hard to explain the difference between both phases because the reasons can be various (weather conditions, temperature, operational conditions WWTP, etc.) and are complex as well. In spite of this variation the SUR values were not extraordinary if the range of SUR values ($5 \cdot 10^{12}$ – $30 \cdot 10^{12}$ m^{-2}) found at other WWTPs in The Netherlands is considered (Roorda, 2004).

During the first phase (April and May) the SUR values of WWTP effluent were most significantly decreased by multi media filtration. Generally, the average (relative) SUR decrease was 30% during these months. This is in contrast with second phase (June and July). During these months hardly any decrease of the SUR value was observed. Surprisingly, the multi media filter performed well during this phase in contrast to the first phase. In the first phase fungus appeared in the filter media and clogged sometimes the multi media filter. After changing the carbon source (acetic acid to methanol) the fungus disappeared and the filter performed well but this did not result in a significant decrease of the SUR values.

Figure 5.15 presents also the SUR values of coagulated WWTP effluent. During the whole experimental period the SUR values of coagulated WWTP effluent are noticeably lower than the SUR values of multi media filtrate. The average decrease of the SUR value of coagulated WWTP effluent was 45% during the whole period. Even when the SUR values of the WWTP effluent (June and July) were relatively low. But in spite of this decrease, after filtration the SUR values had increased. Therefore, it seemed that beside filtration also other processes occurred in the multi media filter resulting in an increase of the SUR values.

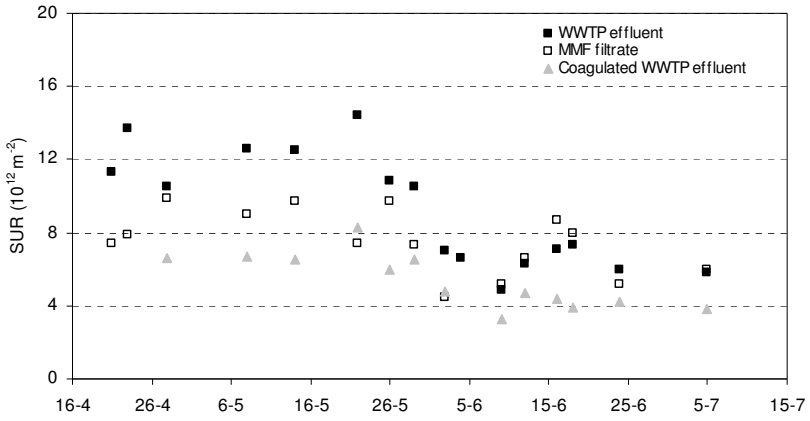


Figure 5.15 – SUR values of the WWTP effluent, coagulated WWTP effluent and multi media filtrate of the WWTP Horstermeer during the period April – July, 2008

5.4.3.2 Foulants

The results of the foulants analyses are presented in Figure 5.16.

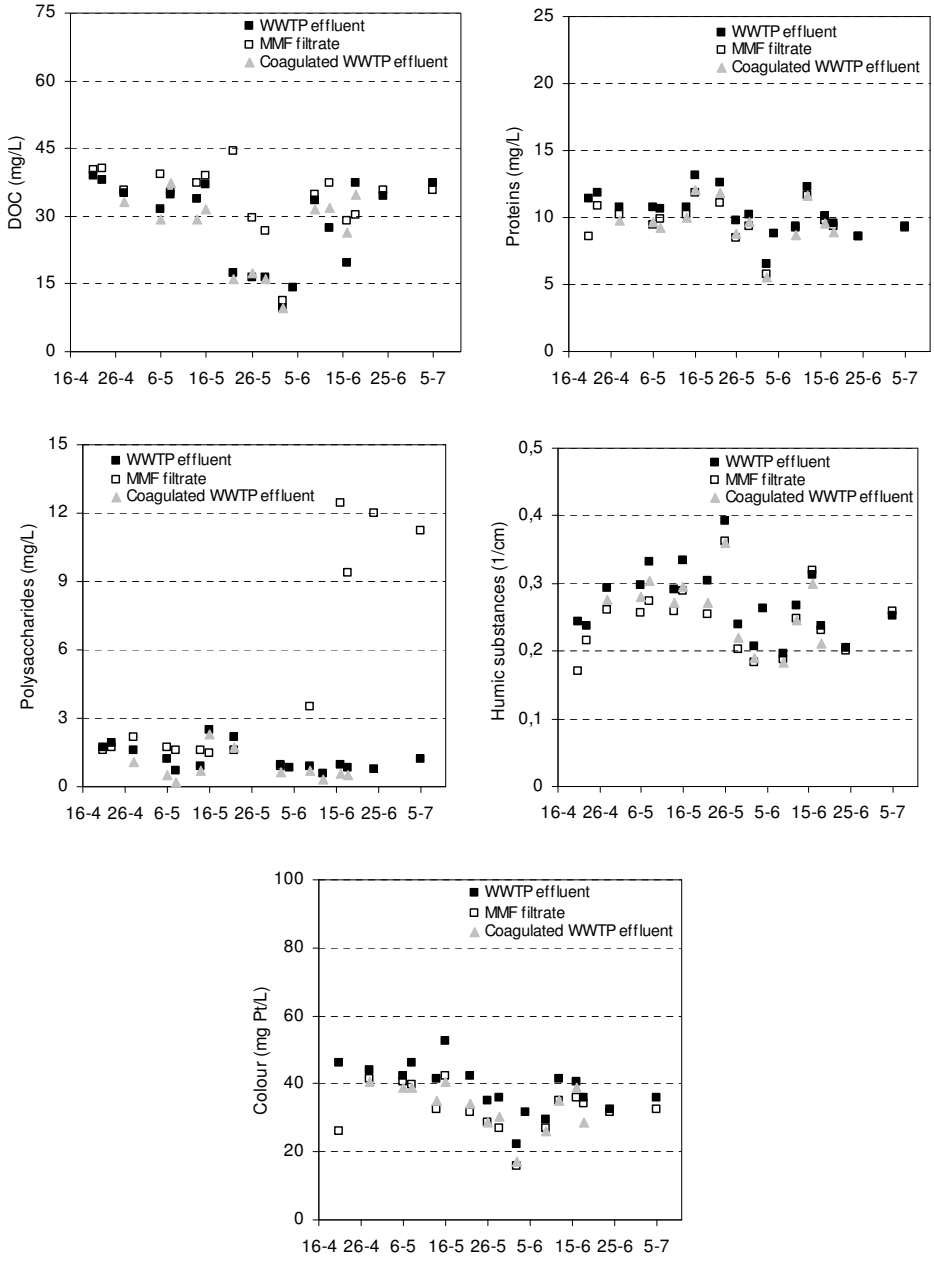


Figure 5.16 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour of the WWTP effluent, coagulated WWTP effluent and multi media filtrate of the WWTP Horstermeer during the period April – July, 2008

During the experimental period the incoming DOC concentrations (WWTP effluent) varied between 10-40 mg/L. The DOC concentrations did not decrease by coagulation (average 0% ± 12%) and presented a high variation. Subsequently the DOC concentrations increased with an average of 26% (± 42%) after multi media filtration. Regarding the concentrations of the proteins no significant effect was observed during both coagulation and multi media filtration. This observation was noticed during the whole experimental period. A different profile was observed regarding the concentrations of the polysaccharides. The concentrations of the polysaccharides presented in the last months (June and July) a huge increase after multi media filtration. This may be caused by the change of carbon source (methanol in stead of acetic acid) and the disappearance of fungus in the multi media filter. Just before (June, 3rd) the taking of the samples in these months the multi media filter was disinfected with chlorine and afterwards methanol was used as carbon source for dentrification. These actions could have resulted in a release of biomass related products like polysaccharides. But suggestion was not confirmed by additional analyses. Regarding the humic substances and colour a slight removal was observed. Therefore it seemed that the coagulation was mainly responsible for the removal. The concentrations of humic substances and colour after the coagulation and multi media filter were more or less similar. This observation is confirmed in other studies as well. Te Poele (2005) also observed a removal of the humic substances and colour humic during different pilot studies.

5.4.3.3 SUR values and foulants

In Figure 5.17 the obtained SUR values of WWTP effluent and multi media filtrate (see Figure 5.15) with the accompanying foulants concentrations (see Figure 5.16 are plotted. This figure does not show any clear relation between foulants and filterability but some differences can be distinguished. The DOC and polysaccharide present hardly any relation whereas proteins, humic substances and colour present some relation but insignificant. Therefore it is not possible to establish any relation between filterability and foulants from Figure 5.17.

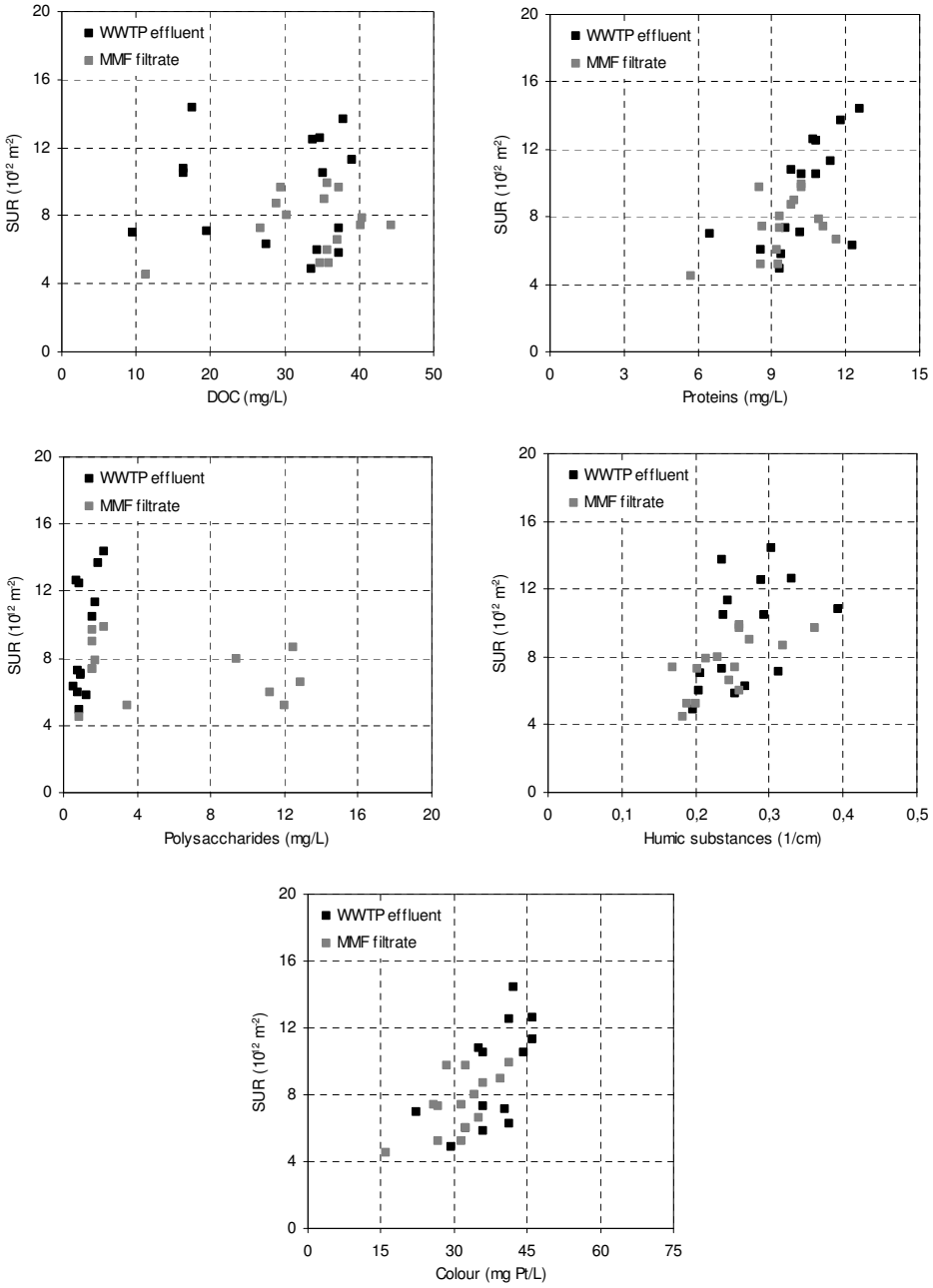


Figure 5.17 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour versus SUR values of the WWTP effluent, coagulated WWTP effluent and multi media filtrate of the WWTP Horstermeer during the period April – July, 2008

5.4.3.4 Fractionation

During the experimental period four times samples of WWTP effluent and multi media filtrate were taken for fractionation. The results are presented in Figure 5.18.

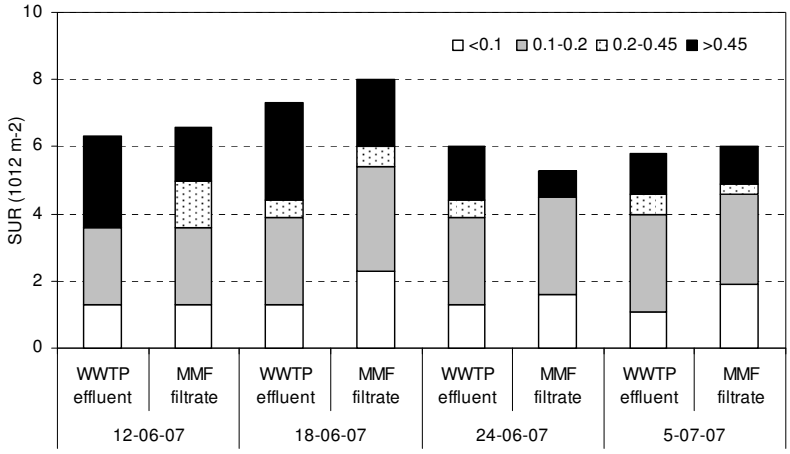


Figure 5.18 – Contribution per fraction to the total SUR value of the WWTP effluent and multi media filtrate of the WWTP Horstermeer during the period April – July, 2008

The fractionation results showed that the fraction 0.1 – 0.2 µm contributes for almost half (41%) of the total SUR value of the effluent of WWTP Horstermeer. The largest fraction (> 0.45 µm) contributes considerably to the filterability, but still to a lesser extent (32%). In total both fractions contributed for approximately three quarter of the total SUR of the effluent of WWTP Horstermeer. But after multi media filtration the contribution of each fraction changed. The contribution of the fraction > 0.45 µm decreased to 21%. But unfortunately the contribution of the other fractions (< 0.1 µm and 0.1 – 0.2 µm) increased after multi media filtration. Now these fractions contributed approximately three quarters of the total SUR of the multi media filtrate. Therefore it seemed that during multi media filtration these fractions were not removed and it even suggested that extra substances were formed within the size range of < 0.1 µm during multi media filtration.

5.4.4 Discussion

The SUR values of the effluent of WWTP Horstermeer decreased after multi media filtration. Especially during the first months (April and May, 2007) the average decrease was significant (28%). This value is comparable with results published by Roorda (2004). As mentioned earlier in this chapter, Roorda (2004) found relative SUR value decreases of 20% to 30% for multi media filtration and coagulation. But in contrast to the first months, during the last two months (June and July, 2007) the average SUR decrease was 4%. This difference between both periods is related to the initial SUR value of WWTP effluent and is shown in Figure

5.19. When the SUR values of the WWTP effluent are below $10 \cdot 10^{12}$ m^{-2} the possibility to lower the SUR value become minimal during multi media filtration.

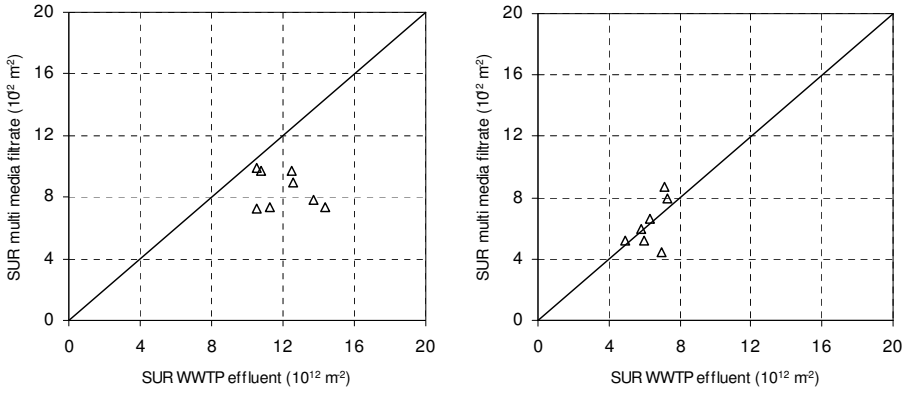


Figure 5.19 – Relation between the SUR values of WWTP effluent and the SUR values of multi media filtrate of the WWTP Horstermeer during the periods April – May, 2007 (left) and June – July, 2007 (right)

Next to the different SUR values of the WWTP effluent during both phases of the experimental period the kind of dosed carbon source differed. During the first months acetic acid was dosed and during the last two months methanol. The dosage of acetic acid resulted in operational problems like rapid clogging of the multi media filter. Probably rapid clogging explain partly the different performance of the multi media filter. Rapid clogging could result in a better entrapment of particles in the filter but resulted also in shorter filtration times to keep the desired filtration velocity. Therefore the carbon source was changed to methanol in order to prevent operational problems.

In general the multi media filter did not remove significantly any foulant. Taken into account the general size (< 0.1 µm) of foulants (te Poele, 2005) this observation could be expected. Multi media filters are mainly operated to remove particles of a larger size range (Miska – Markusch, 2009) of WWTP effluent. This observation is confirmed by the fractionation results. These results presented only a slight removal of the particle size range > 0.45 µm whereas the fraction of 0.1 – 0.2 µm did not change. Even the contribution of the size fraction < 0.1 µm increased in the multi media filtrate. Probably this increase is caused by the formation of soluble microbial products during multi media filtration in this configuration, because in Figure 5.16 also some small increase of proteins can be observed after multi media filtration. However this aspect was not investigated, therefore it can only be assumed.

Furthermore the compilation of the foulants concentrations and SUR values of both WWTP effluent and MMF filtrate did not present any significant relation. This observation proves

once more the results found by te Poele (2005) and confirms the observation of Roorda (2004) that particles and colloids had a great influence on the filterability.

5.5 1-STEP® Filter

5.5.1 General

The One Step Total Effluent Polishing Filter (1-STEP® filter) is a combination of the granulated activated carbon filter and the multi media filter. The filter media of the 1-STEP® filter is activated carbon but like the multi media filter the 1-STEP® filter is operated for denitrification and simultaneous phosphorus removal. Beside the removal of these compounds the 1-STEP® filter has been developed to achieve removal of small organic compounds and organically bound heavy metals. Furthermore the 1-STEP® filter is also considered to be a promising technology for pretreatment of membrane filtration. The combination of filtration and adsorption in one concept may combine the advantages of the multi media filtration and granulated activated carbon filtration. Therefore the effect of the 1-STEP® filter on the filterability, foulants and fractions of WWTP effluent was investigated. In this section the results are presented and discussed.

5.5.2 Experimental set up

Like the experiments with the multi media filter the experiments with the 1-STEP® filter were conducted at the WWTP Horstermeer. The tests were performed during a period of 17 weeks (March – July, 2008). Halfway the experimental period (May, 2008) the filter media was renewed once. During the experimental period the 1-STEP® filter was operated for denitrification and simultaneous phosphorus removal. The feedwater of the filter, WWTP effluent, passed the 450 µm curved sieve and was collected in a feedwater buffer. For the simultaneous removal of phosphorus and suspended solids poly aluminium chlorine was dosed in-line into the feedwater of the filter. The coagulation took place with a static mixer and the flocculation took place in the water volume above the filter medium. The flocculation time was approximately 8 minutes. For denitrification acetic acid or methanol was dosed based on the actual nitrate and free oxygen concentrations in the feedwater. In Table 5.9 the operational parameters of the 1-STEP® filter during the experimental period are given.

Table 5.9 – Operational parameters of the 1-STEP® filter at the WWTP Horstermeer during the period March – July, 2008

Parameter	Unit	Value
Flow	m^3/h	8 – 10
Hydraulic load	m/h	8 – 10
Filter run	h	12
Frequency 'bumping cleaning'	1/d	7 – 9
Bed height	m	1.9
Surface area	m^2	1.0
Empty Bed Contact Time	minutes	11 – 14
Acetic acid dosing (March, 3 – May, 22)	g COD/g NO_3-N	5
Methanol dosing (May, 22 – July, 1)	g COD/g NO_3-N	3 – 5
PACl in metal orthophosphate ratio	mol/mol	3 – 5

During the experimental period samples were taken of the WWTP effluent (after the curved sieve) in the feedwater buffer and of the filtrate of the 1-STEP® filter. The latter samples were taken 20 minutes after the samples of WWTP effluent and at least two hours after a backwash. After sampling different actions were performed which are summarized in Figure 5.20. The collected samples were transported directly to the laboratory of Sanitary Engineering of Delft University of Technology. Then, if possible, the fractionations and SUR measurements were conducted on that very day. If not the samples were stored for at most one day at 5 °C in a refrigerator. For foulants analyses the samples were filtered on the day of sampling and analysed within at least one week after filtration. In between the filtrated samples were stored at 5 °C.

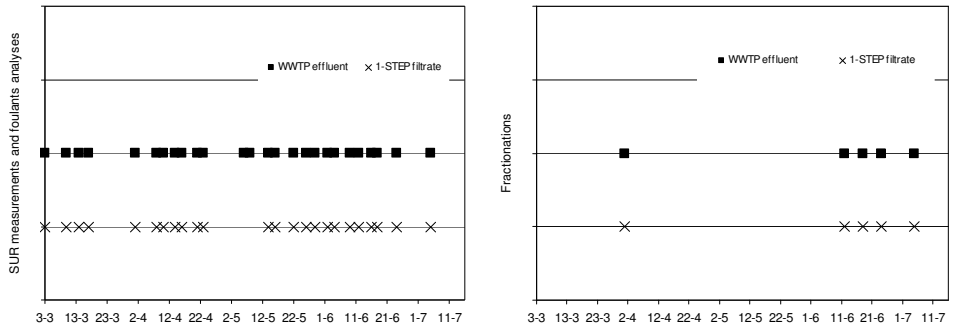


Figure 5.20 – Dates of sampling of WWTP effluent and 1-STEP® filtrate and performed actions (SUR measurements and foulants analyses (left) and fractionations (right)) during the experimental period March – July, 2008

5.5.3 Results

5.5.3.1 SUR values

In Figure 5.21 the results of the SUR measurements are shown. In the figure also the moment of the renewal of the filter media is illustrated. In the phase before the renewal of the filter media the average SUR value of the 1-STEP® filtrate was $5 \cdot 10^{12}$ m^{-2}. Unfortunately two times (April 14th and 16th) the SUR values of 1-STEP® filtrate did not present a significant decrease, because some operational problems occurred with the 1-STEP® filter. In general a relative SUR decrease of 60% was observed before the renewal, whilst after the that the average relative SUR decrease was 50% with an average SUR value of $8 \cdot 10^{12}$ m^{-2} (WWTP effluent). Except one measurement all the SUR values of the 1-STEP® filter were around or below $4 \cdot 10^{12}$ m^{-2} after the renewal of the filter media. Also before the renewal the SUR values were measured in this range. It means that during the whole experimental period, 1-STEP® filtration resulted in very good filterable water. Even when the SUR values of the WWTP effluent were more than $10 \cdot 10^{12}$ m^{-2}.

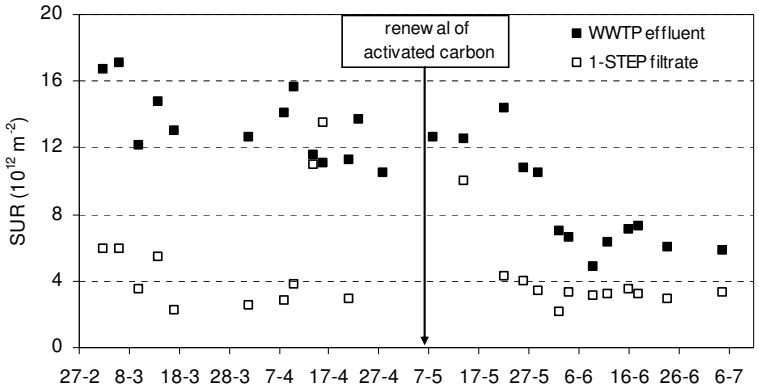
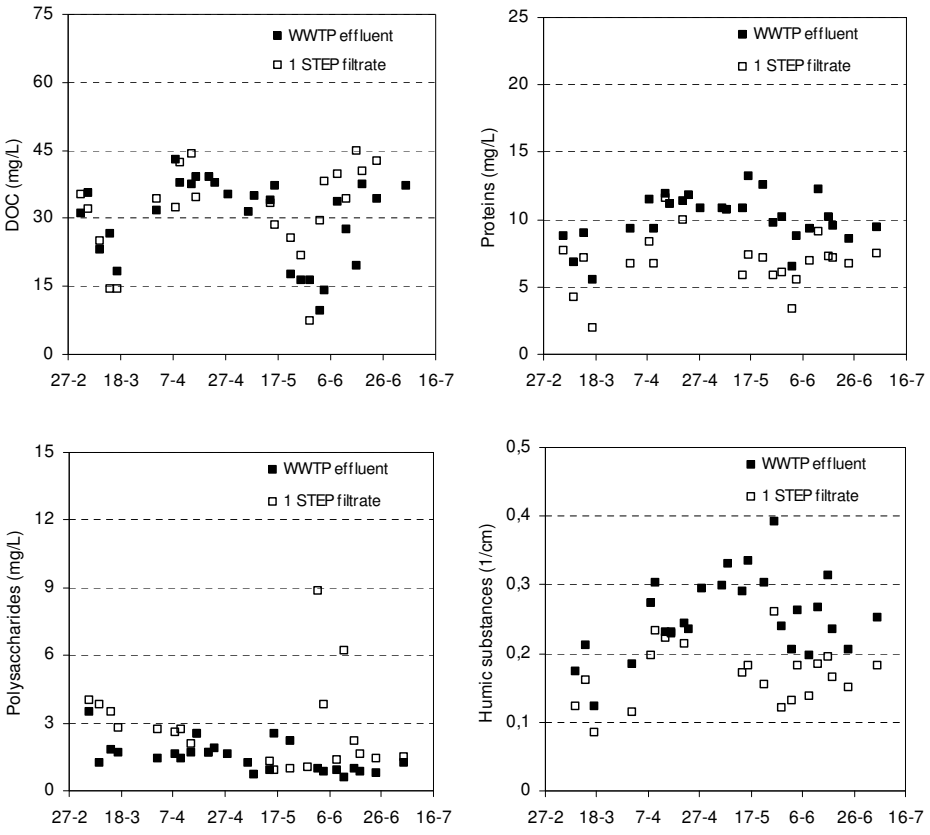


Figure 5.21 – SUR values of the WWTP effluent and 1-STEP® filtrate of the WWTP Horstermeer during the period March – July, 2008

5.5.3.2 Foulants

In Figure 5.22 the concentrations of foulants of the WWTP effluent and 1-STEP® filtrate are displayed. As shown, the effect of 1-STEP® filtration on the DOC concentration was negative i.e. the average DOC concentration increased about 15% after filtration. This trend was also observed during the experiments with the multi media filter (section 5.4). However, the concentrations of proteins presented a different picture. During the whole experimental period the concentrations of proteins decreased with an average of 30%. In contrast to the proteins the concentrations of polysaccharides increased nearly always. Like the experiments with the multi media filter a sudden huge increase of the polysaccharides concentration in 1-STEP® filtrate was observed in June, 2008. It seemed the change of carbon source played a role again. Since May, 22nd methanol was dosed instead of acetic acid. Furthermore the concentrations of humic substances and colour presented a significant decrease during the whole period. The average decrease of the humic substance and colour were respectively 30 and 41%.



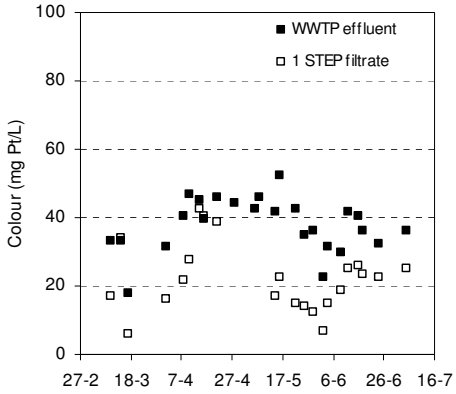
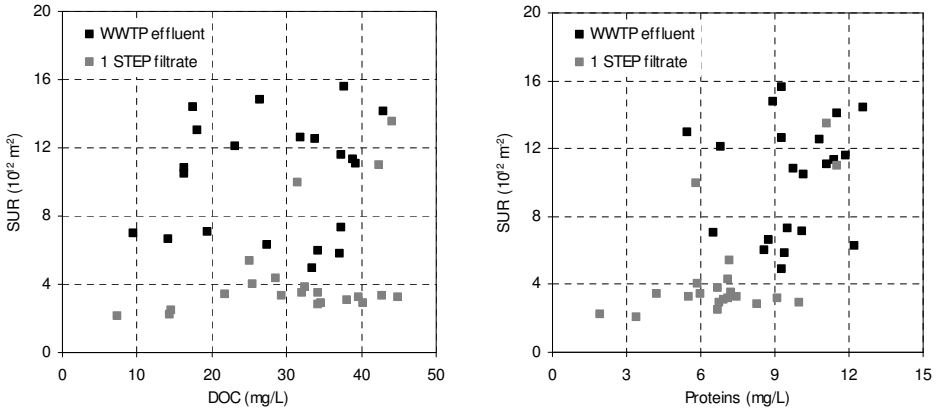


Figure 5.22 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour of the WWTP effluent and 1-STEP® filtrate of the WWTP Horstermeer during the period March – July, 2008

5.5.3.3 SUR values and foulants

In Figure 5.23 the values of the Figure 5.21. and 5.22 are related in order to determine any relation between the SUR values and foulants of WWTP effluent and 1-STEP® filtrate. Like the presented figures in the previous section again no correlation can be established i.e. only a concentration of a certain foulant cannot be used to predict the filterability of WWTP effluent neither 1-STEP® filtrate.



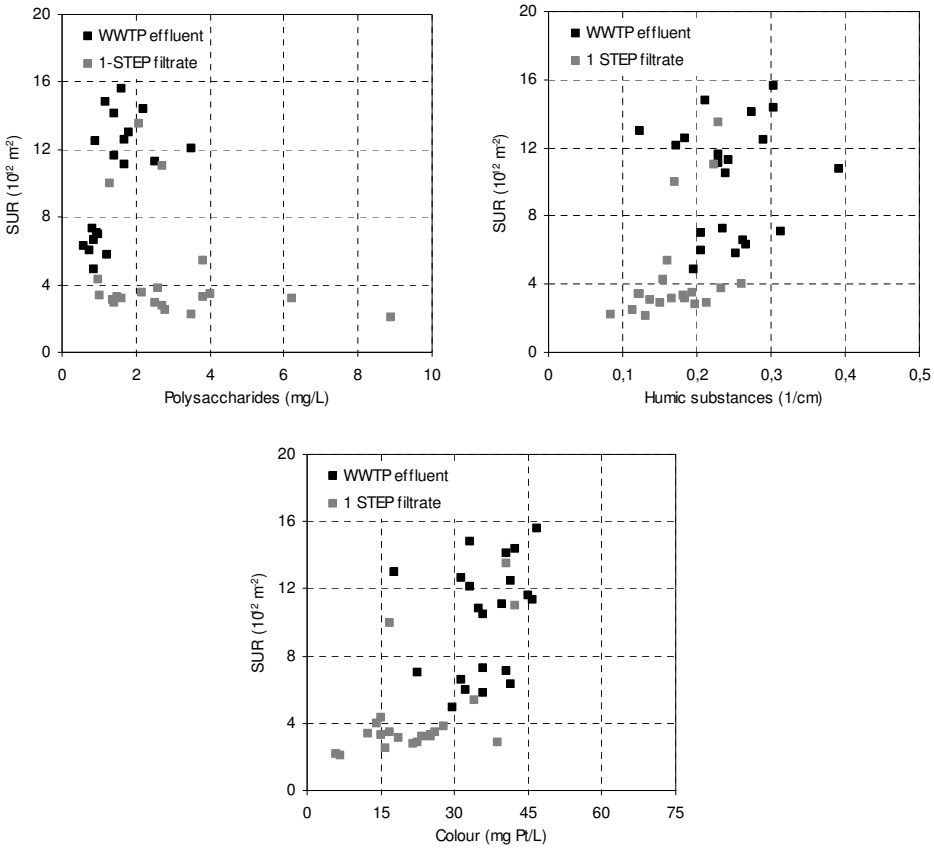


Figure 5.23 – Concentrations of the dissolved organic carbon, proteins, polysaccharides, humic substances and colour versus SUR values of the WWTP effluent and 1-STEP® filtrate of the WWTP Horstermeer during the period March – July, 2008

5.5.3.4 Fractionation

At five times fractionation tests were performed of both, effluent and 1-STEP® filtrate of the WWTP Horstermeer. The results are plotted in Figure 5.24. Like shown previously (section 5.4) the fraction 0.1 – 0.2 µm contributed mainly (46%). to the total SUR value of the effluent. Furthermore the fraction > 0.45 µm accounted for 32% of the total SUR value of the WWTP effluent. But after the 1-STEP® filter this picture changed. The contribution of both fractions, 0.1 – 0.2 µm and > 0.45 µm, decreased significantly. The average decrease of the fraction > 0.45 µm and 0.1 – 0.2 µm was respectively 68% and 50%. The fraction < 0.1 µm did not change consistently. Therefore the results indicate that 1-STEP® filtration results in low SUR values due to the removal of both fractions 0.1 – 0.2 µm and > 0.45 µm.

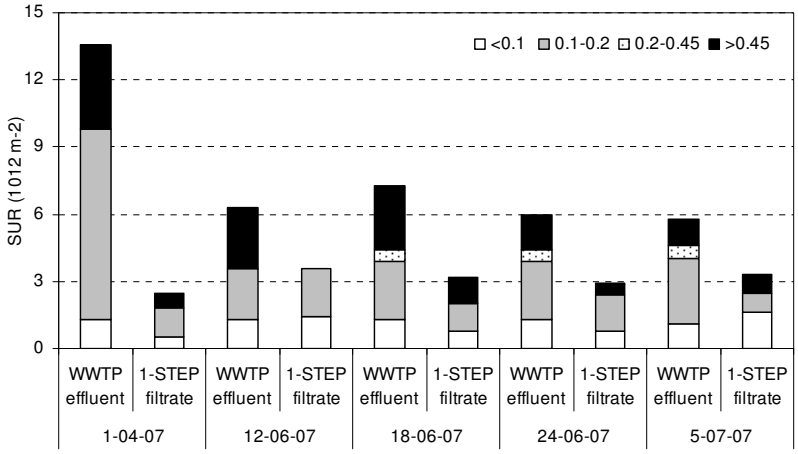


Figure 5.24 – Contribution per fraction to the total SUR value of the WWTP effluent and 1-STEP® filtrate of the WWTP Horstermeer during the period March – July, 2008

5.5.4 Discussion

The average relative SUR decrease during 1-STEP® filtration at WWTP Horstermeer was 56% during the whole experimental period. This is significantly higher than the other pretreatment technologies. In the same experimental period the average relative SUR decrease of the multi media filter was found to be in the range of 4% - 28% depending on the initial SUR value. In contrast to the multi media filter it seemed that the relative SUR decrease during 1-STEP® filtration was less dependent on the initial SUR value. During the whole experimental period it was possible to achieve SUR values of the 1-STEP® filtrate below approximately $5 \cdot 10^{12}$ m^{-2} (see Figure 5.25). This result may mainly be achieved thanks to the removal of both fractions, 0.1 – 0.2 µm and > 0.45 µm, of the WWTP effluent. With the other tested pretreatment technologies it was only possible to remove partly the fraction > 0.45 µm.

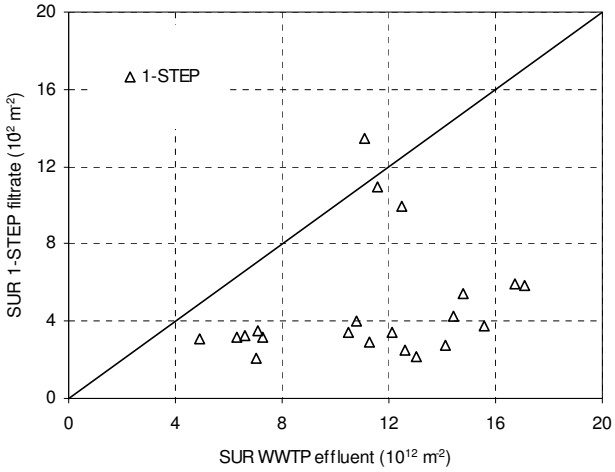


Figure 5.25 – Relation between the SUR values of WWTP effluent and the SUR values of 1-STEP® filtrate of WWTP Horstermeer during the period March – July, 2008

Considering the concentrations of foulants and fractionation results it seemed that during 1-STEP® filtration the different processes, filtration, flocculation and adsorption, play a role. Concerning adsorption, the concentrations of the proteins, humic substances and colour were significantly removed during 1-STEP® filtration. This was not noticed during the experiment with the multi media filter. The main difference between the multi media filter and 1-STEP® filter, is the type of filter media. Therefore, it may indicate that the activated carbon was responsible for the partial removal (adsorption) of some foulants, although the concentrations of DOC increased during 1-STEP® filtration. This observation was also noticed with the multi media filter. Probably this is related to the dosage of methanol or acetic acid upstream both filters. It was not checked but it could be that a part of the methanol and acetic acid was not consumed by the denitrifying bacteria. The DOC is an overall parameter (te Poele, 2005) and could include the unconverted methanol and acetic acid.

5.6 Evaluation

5.6.1 Powdered activated carbon

Experiments with powdered activated carbon were performed on both lab and pilot scale. The lab scale experiment presented a significant decrease of the initial SUR value of WWTP effluent with increasing dosages of powdered activated carbon. Next to the dosages, the properties of the carbon played an important role. The carbon with a relative smaller particle size (D_{50}) and consisting of relatively more mesopores showed the most significant SUR decrease. This effect of smaller particle size was also investigated by Matsui *et al*. (2005). These researchers started from plant supplied powdered activated carbon, ground that down to

submicrometre particles and used these as an adsorbant before microfiltration for drinking water treatment. The microground powdered activated carbon adsorbed natural organic matter much more rapidly and had a higher adsorptive capacity that ordinary powdered activated carbon (Matsui *et al*., 2005). Consequently the use of submicrometre powdered activated carbon permitted not only shorter contact times but also a 75% reduction in dose (Matsui *et al*., 2005). However the particle size of powdered activated carbon applied during the lab scale experiments had been much larger (D_{50} of 7 and 15 µm) than the microground powdered activated carbon (0.8 and 0.6 µm) of Matsui *et al*. (2005). Therefore the potential of powdered activated carbon to decrease the SUR value of WWTP effluent may increase when particle size could be further reduced, because it was also noticed during the lab scale experiments that the actual relation between the powdered activated carbon addition and the SUR values seems to be in some way inefficient. Only with high dosages of powdered activated carbon the SUR values of WWTP effluent reached attractively low values.

Another option to optimize the efficiency of the powdered activated carbon is to combine it with the addition of coagulants. Several authors (Haberkamp *et al*., 2007; Shon *et al*., 2004b) have published about this combination. Shon *et al*. (2004b) reported no filtration decline during crossflow ultrafiltration of WWTP effluent after pretreatment of flocculation and adsorption. Separately, both pretreatment technologies showed a flux decline in the same study. Haberkamp *et al*. (2007) also investigated the effect of coagulation and adsorption with WWTP effluent and concluded that this combination largely enhances the removal of biopolymers in WWTP effluent. Therefore it seemed the combination of coagulation/flocculation and powdered activated carbon could optimize the efficiency of the application of powdered activated carbon but the configuration of the pretreatment technologies in combination with ultrafiltration has to be well considered. During the pilot scale experiment at the WWTP Maasbommel, which was also a combination of coagulation/flocculation – adsorption – ultrafiltration, the results seemed to be influenced by the configuration of ultrafiltration pilot installation. As presented, in this configuration the particles of WWTP effluent in the size range of 0.1 – 0.2 µm accumulated resulting in high SUR values (i.e. bad filterability).

5.6.2 Prefiltration

The experiment with the granulated activated carbon filter was conducted at the WWTP Maasbommel and the experiments with the multi media filter and 1-STEP® filter at the WWTP Horstermeer. In Figure 5.26 the SUR values of all the experiments are plotted in one picture. From this figure it is evident that 1-STEP® filtration resulted in the most significant decrease of the SUR value. As presented in section 5.5 the average relative SUR decrease during the whole period was 56%. This value was slightly dependent on the SUR value of WWTP effluent in contrast to the multi media filter. As shown in section 5.4. the SUR value of multi media filtrate depended highly on the SUR value of WWTP effluent. For granulated

activated carbon filtration the number of samples was too low to determine a relation between the SUR value of WWTP effluent and granulated activated carbon filtrate.

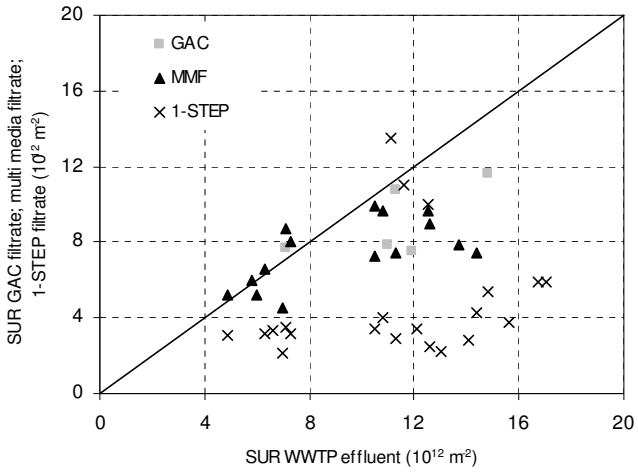


Figure 5.26 – Relation between the SUR values of WWTP effluent and the SUR values of granulated activated carbon filtrate (WWTP Maasbommel), multi media filtrate and 1-STEP® filtrate (WWTP Horstermeer).

The different performance of the filters can be explained by the results of the fractionation experiments. In Table 5.10 an overview of the average percentage of SUR decrease of the different fractions of WWTP effluent is given. In the table 100% means a complete removal of a certain fraction and 0% means no change before and after the filter.

Table 5.10 – The average percentages of SUR decrease of the different fractions of WWTP effluent after granulated activated carbon filtration (WWTP Maasbommel), multi media filter and 1-STEP© filter (WWTP Horstermeer)

	Granulated activated carbon filter	Multi media filter	1-STEP® filter
> 0.45 µm	73% (± 11%)	33% (± 18%)	68% (± 25%)
0.2 – 0.45 µm	-363% (± 466%)	43% (± 60%)	100% (± 0%)
0.1 – 0.2 µm	11% (± 28%)	-6% (± 12%)	50% (± 31%)
< 0.1 µm	-1% (± 41%)	-43% (± 38%)	17% (± 43%)

In Table 5.10 it is shown that all the filters did remove the fraction > 0.45 µm. Both filters filled with granulated activated carbon (granulated activated carbon filter and 1-STEP® filter) present the highest removal percentages. The relative high removal percentage of the granulated activated carbon filtration probably be could explained by the hydraulic load (2.25 – 3 m/h) and the frequency of the backwash (1/week). These process conditions may result in better filtration performance than the multi media filter and 1-STEP® filter with hydraulic

loads of approximately 10 m/h and filter runs of 12 hours, although, the difference between the multi media filter and 1-STEP® filter cannot be explained by the process conditions because these were more or less similar (see Table 5.9. and 5.10). Also the particles of the filter media of both filters were in the same particle size range. Therefore other reasons should explain the difference. It may be the properties of filter media and not the particle size that played a role. Activated carbon has more surface area per volume unit than sand or anthracite and therefore more possibilities (area) for particles (and bacteria) to attach or adsorb. This suggestion could also explain the relatively high removal of the fractions 0.1 – 0.2 µm and < 0.1 µm. Compared to the other filters the 1-STEP® filter presented the highest removal of these fractions resulting in low SUR values of the filtrate. Considering all these findings it might be that bioadsorption was the dominant mechanism in the 1-STEP® filter. In the 1-STEP® filter biological activity was stimulated by methanol dosing but this was also done in the multi media filter. Nevertheless in the multi media no removal of the fractions 0.1 – 0.2 µm and < 0.1 µm was observed, even the fraction of < 0.1 µm increased. Combining these differences with the results of the granulated activated carbon filter (some removal of the fraction 0.1 – 0.2 µm) it is suggested that both adsorption and biological activity play a role in the 1-STEP® filter resulting in low SUR values of the filtrate.

The adsorption capacity of the 1-STEP® filter is also shown in Table 5.11. Like the granulated activated carbon filter the 1-STEP® filter removed the proteins, humic substances and colour of the WWTP effluent. The tendency of these compounds to activated carbon is also reported by other researchers (Haberkamp *et al.*, 2007; Shon *et al.*, 2004b). However the removal of these foulants could not explain the decrease of the SUR value. For example during 1-STEP® filtration the concentrations of polysaccharides increased but the SUR values decreased. This confirms the earlier observations in this chapter that filterability is related to (organic) particles and colloids instead of foulants.

Table 5.11 – The average decrease percentages of the concentration of foulants of WWTP effluent after granulated activated carbon filtration (WWTP Maasbommel), multi media filter and 1-STEP© filter (WWTP Horstermeer)

	Granulated activated carbon filter	Multi media filter	1-STEP®filter
DOC (%)	3 (± 14)	-26 (± 42)	-15 (± 37)
Proteins (%)	48 (± 21)	8 (± 6)	30 (± 15)
Polysaccharides (%)	7 (± 12)	-21 (± 55)	-40 (51)
Humic Substances (%)	36 (± 12)	10 (± 8)	30 (± 13)
Colour (%)	39 (± 13)	16 (± 11)	42 (± 22)

In general the prefiltration experiments show that the 1-STEP®filter was able to remove the particles dominating the filterability of WWTP effluent. It may be that bioadsorption and filtration are the responsible mechanism but this was not proven by additional measurements.

5.7 Conclusions

- Lab scale experiments with two different types of powdered activated carbon showed different results in terms of SUR decrease. The main difference between both types of powdered activated carbon was the particle size (D_{50}). The powdered activated carbon with the smallest (7 µm) particle size presented lower SUR values at the same dosages as the powdered activated carbon with a bigger particle size (15 µm).

- Foulants were adsorbed by powdered activated carbon during the lab scale experiments. Especially, the proteins, humic substances and colour were adsorbed instead of the polysaccharides.

- The application of the combination of powdered activated carbon and ultrafiltration in practice should be well considered. The design of this combination seemed to play an important role in the filterability of the feedwater of the ultrafiltration unit.

- The combination of adsorption, coagulation/flocculation and filtration in one filter (1-STEP®filter) resulted in a stable decrease of the SUR value of WWTP effluent. Even when the SUR values of WWTP effluent were less than $10 \cdot 10^{12}$ m^{-2}.

- From the investigated three prefiltration technologies the 1-STEP® filter was the only technology that showed significant removal of the fraction 0.1 – 0.2 µm of WWTP effluent. It seemed that the combination of adsorption and biological activity was responsible for this phenomena.

- From all the measured foulants of the WWTP effluent especially the proteins, humic substances and colour were removed by granulated activated carbon filtration and 1-STEP® filtration. Adsorption is probably the dominant mechanism responsible for this removal.

- During the experiment with the granulated activated carbon filter, multi media filter and 1-STEP® filter no significant relation between the SUR values and foulants concentrations of WWTP effluent and filtrate was observed.

References

Amy, G. (2007) *Fundamental Understanding of Organic Matter Fouling of Membranes*. (2008) Desalination, 231, 44-51.

Betuw, W. van, Kruit, J., Segers, J., Piron, D. and Uijterlinde, C.A. (2007). *Opportunities for Effluent Reuse at Municipal Wastewater Treatment Plant Maasbommel,* Proceedings 6th IWA Specialists Conference on Wastewater Reclamation and Reuse for Sustainability 2007, October 9-15, Antwerp, Belgium.

Dikkenburg, J. van den (2006). Personal communication. Norit.

Geilvoet, A.J. (2007). *Fouling modification during pretreatment for dead-end ultrafiltration of wwtp effluent.* MSc thesis. Delft University of Technology, Delft, The Netherlands.

Haberkamp, J., Ruhl, A.K., Ernst, M. Jekel, M. (2007). *Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting behaviour in ultrafiltration*. Water Research, 41, 3794-3802.

Haberkamp, J., Ernst, M., Bockelmann, U., Szewzyk, U. and Jekel, M. (2008) *Complexity of ultrafiltration membrane fouling caused by macromolecular dissolved organic compounds in secondary effluents*. Water Research 42, 3153-3161.

Huang, H., Young, T. and Jacangelo, J.G. (2007). *Pretreatment and its effect and implications on water treatment by low pressure membranes.* Proceedings 4th IWA leading-edge conference and exhibition on water and wastewater technologies, June 3-6, Singapore, Republic of Singapore.

Huck, P.M. and Sozanski, M.M. (2008) *Biological filtration for membrane pre-treatment and other applications: towards the development of a practically-oriented performance parameter*. Journal of Water Supply: Research and Technology-AQUA 57.4, 203-224.

Lee, C.W., Bae, S.D., Han, S.W., and Kang, L.S. (2005). *Application of UF hybrid processes for reuse of secondary effluent.* Proceedings IWA Specialty Conference 'Wastewater Reclamation & Reuse for Sustainability'. November 8-11, Jeju, Korea.

Matsui, Y., Murase, R., Sanogawa, T., Aoki, N., Mima, S., Inoue, T. and Matsushita, T. (2004). *Micro-ground powdered activated carbon for effective removal of natural organic matter during water treatment*. Water Science and Technology: Water Supply, 4(6), 155-163.

Matsui, Y., Murase, R., Sanogawa, T., Aoki, N., Mima, S., Inoue, T. and Matsushita, T. (2005). *Rapid adsorption pre-treatment with submicrometre powdered activated carbon particles before microfiltratont*. Water Science and Technology, 51(6-7), 249-256.

Menkveld, H.W.H., Miska, V. and Gorter, K. (2006). *Op weg naar EKW, praktijkonderzoek RWZI Horstermeer naar de aanvullende verwijdering van nutriënten, zware metalen en prioritaire stiffen.* Voortgangsrapportage 2, Witteveen+Bos, Deventer, The Netherlands.

Menkveld, H.W.H., Scherrenberg, S.M., Postma, P. and Kloeze, A. te. (2008). *Op weg naar EKW, praktijkonderzoek RWZI Horstermeer naar aanvullende verwijdering van nutriënten, zware metalen en prioritaire stoffen.* Voortgangsrapportage 4 januari t/m december 2007, Intern report (in Dutch), Witteveen+Bos Consulting Engineers, Deventer, The Netherlands.

Miska – Markusch, V. (2009). *Effluent filtration for more than particle removal.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Mozia, S., Tomaszewska, M. and Morawski, A.W. (2005). *Studies on the effect of humic acids and phenol on adsorption-ultrafiltration process performance.* Water Research, 39 (2-3), 501-509.

Oh, H., Yu, M., Takizawa, S. and Ohgaki., S. (2006). *Evaluation of PAC behavior and fouling formation in an integrated PAC-UF membrane for surface water treatment*. Desalination, 192, 54-62.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H., Wortel, N.C. and Daalen, R. van (2005). *New process for treatment of organically fouled water; experiences with WWTP-effluent*. Desalination, 178, 27-40.

Shon, H.K., Vigneswaran, S., Kim, I.S., Cho, J. and Ngo, H.H. (2004a). *Effect of pretreatment on the fouling of membranes: application in biologically treated sewage effluent.* Journal of Membrane Science, 234 (1-2), 111-120.

Shon, H.K., Vigneswaran, S., Kim, S.I., Cho, J. and Ngo, H.H. (2004b). *The effect of pre-treatment to ultrafiltration of biologically treated sewage effluent: a detailed effluent organic matter (EfOM) characterization*. Water Research, 38, 1933 – 1939.

STOWA (2007). *Effluentnabehandeling op de rwzi Maasbommel*. Report 2007-17 (in Dutch), Utrecht, The Netherlands.

Tomaszewska, M. and Mozia, S. (2002). *Removal of organic matter from water by PAC/UF system.* Water Research, 36 (16), 4137-4143.

Tsujimoto, W., Kimura, H., Izu, T. and Irie, T. (1998). *Membrane filtration and pre-treatment by GAC.* Desalination, 119, 323-326.

Zheng, X., Ernst, M. and Jekel, M. (2009). *Identification and quatification of major oganic foulants in treated domestic wastewater affecting filterability in dead-end ultrafiltration*. Water Research. 43, 1, 238 – 244.

6 Application of the SUR measurement in practice: A case study

6.1 Introduction

During ultrafiltration of WWTP effluent the fouling is controlled by periodically cleaning the membranes. Cleaning is performed either hydraulically, mechanically, chemically or sometimes by electrical cleaning. Next to cleaning, the fouling of the membranes can be controlled by pretreatment e.g. coagulation often serve as a pretreatment for ultrafiltration membranes. The ultrafiltration membranes remove particulate (organic) matter, which results in a stable performance of the reverse osmosis membranes. But as written in chapter 5 also for ultrafiltration membranes often a pretreatment step is placed to enhance the filtration rate and to decrease the fouling rate. Next to these reasons pretreatment is often applied to minimize the negative impact of quality variations of the feedwater. Especially at industrial wastewater treatment plants high variations in time and concentration of the effluent quality can be observed. The reasons for these variations can be several i.e. poor operation of the wastewater treatment plant, discontinuous discharge from the industrial batch processes, complex biodegradable batches (new products), wastewater coming from incidental industrial cleaning processes, etc. Due to these reasons and the complexity of the treatment processes the effluent water quality is sometimes unpredictable. Especially when the effluent quality is bad (bad quality event) in terms of filtration and fouling rate operational problems will occur.

At the WWTP Sas van Gent the described problem of bad water quality sometimes appeared and disturbed the operation of the UF-RO installation. Even the combination of coagulation and dual media filtration before the UF-RO installation was not always able to prevent the occurring operational problems. The operational problems often resulted in rapid fouling of the ultrafiltration and reverse osmosis membranes and extra chemical cleanings were needed. Therefore it was suggested to tackle these problems by the implementation of an extra buffering zone between the wastewater treatment plant and the dual media filter; a stabilization pond. The stabilization pond has a high buffer capacity (2 – 3 days) and was expected to be able to equalize the effluent water quality. Nevertheless also disadvantages of using a stabilization pond were expected. The stabilization pond is an open water and therefore the temperature will change and algae may grow and influence negatively the filtration and fouling. To gain an insight into the expected advantages and disadvantages research was needed. Therefore in this chapter the performance of pretreatment and the ultrafiltration units is evaluated during the intake of WWTP effluent after buffering in the stabilization pond. For this evaluation the SUR measurement was applied accompanied with foulants analyses of the WWTP effluent.

6.2 Filterability and foulants of WWTP effluent after secondary clarifier and stabilized WWTP effluent

6.2.1 Experimental setup

As stated in the introduction the research had been performed at the WWTP Sas van Gent. During a period of six weeks (August, 31 – October, 12, 2006) the UF-RO installation was fed with WWTP effluent that flowed through a stabilization pond. During normal operation of the wastewater treatment plant and UF-RO installation the effluent flow is divided into two streams. One part of the flow is directly pumped to the UF-RO installation and the other part is discharged to the surface water after flowing through the stabilization pond. During this experiment the situation was changed. The total flow of WWTP effluent was pumped to the stabilization pond and the UF-RO installation was fed with the WWTP effluent after it passed the stabilization pond. The residence time of the WWTP effluent in the stabilization pond was 2 – 3 days. In Figure 6.1 an impression of the stabilization pond is given. A pump had been placed at the outlet of the stabilization pond to transport the stabilized WWTP effluent to the UF-RO installation. Therefore a temporary pipeline had been constructed between the pump and UF-RO installation with a length of approximately 500 meters. Because of design criteria (the water temperature) of the UF-RO installation a heat exchanger was placed halfway the pipeline to keep the feed water temperature above 20 °C.

Figure 6.1 – Impression of the stabilization pond and pump for transporting the stabilized WWTP effluent to the UF-RO installation at the WWTP Sas van Gent

To compare the filterability and the concentrations of foulants of WWTP effluent and of the stabilized WWTP effluent, samples were taken at the outlet of the secondary clarifier of the wastewater treatment plant (WWTP effluent) and before the pretreatment steps at the UF-RO installation (stabilized WWTP effluent). A schematic overview of the wastewater treatment plant, stabilization pond, dual media filter and the ultrafiltration units including the sampling and coagulant dosing points is presented in Figure 6.2.

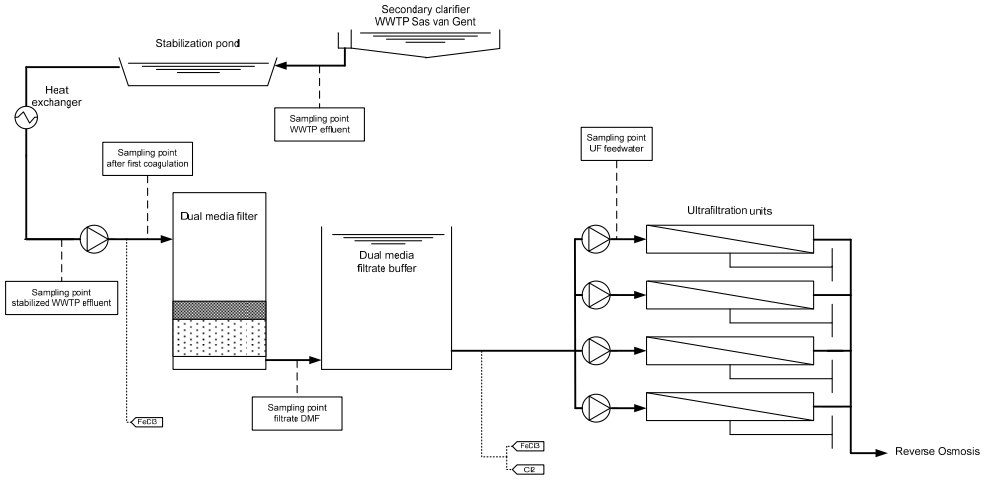


Figure 6.2 – Schematic overview of the WWTP Sas van Gent, stabilization pond, dual media filter, filtrate buffer and ultrafiltration units inclusive sampling points and coagulant dosing points

After sampling the SUR measurements were directly performed on site. The analyses of the foulants were done within a week after sampling at the laboratory of Sanitary Engineering of Delft University of Technology. Therefore, the samples for analyses were stored on site in a refrigerator.

6.2.2 SUR values

Figure 6.3 represents the SUR values of the WWTP effluent with and without using the stabilization pond during the experimental period. From this figure it is obvious that the SUR values of WWTP effluent increased during the passage of the stabilization pond. Except two times (October, 4th and 5th) the SUR values of the WWTP effluent after the secondary clarifier were always below $10.0 \cdot 10^{12}$ m^{-2} and the average was $5.8 \cdot 10^{12}$ m^{-2} ($\pm$ $2.9 \cdot 10^{12}$ m^{-2}). It means that the average filterability of WWTP effluent before the stabilization pond was good and suitable for membrane filtration. However, after the pond passage the picture changed. The SUR values almost doubled and resulted in an average SUR value of $14.7 \cdot 10^{12}$ m^{-2} ($\pm$ $4.5 \cdot 10^{12}$ m^{-2}). This means the filterability became worse and the stabilized WWTP effluent was not directly applicable to ultrafiltration.

As discussed, the expected buffering effect of the stabilization pond (more steady and lower SUR values) did not occur during the experiment. Even the opposite effect was observed. Probably the presence of algae in the stabilization pond played an important role. The algae were clearly visible in the stabilization pond and the taken samples. Therefore from a few samples the chlorofyl-a concentrations have been analyzed. The chlorofyl-a concentrations varied and were found in the range of 90 – 1,420 µg/L (Trampé, 2007). From the literature

(Babel and Takizawa, 2000) it is known that algae can have a negative effect on the performance of membrane installations. Therefore the presence of algae might explain the findings as represented in Figure 6.3.

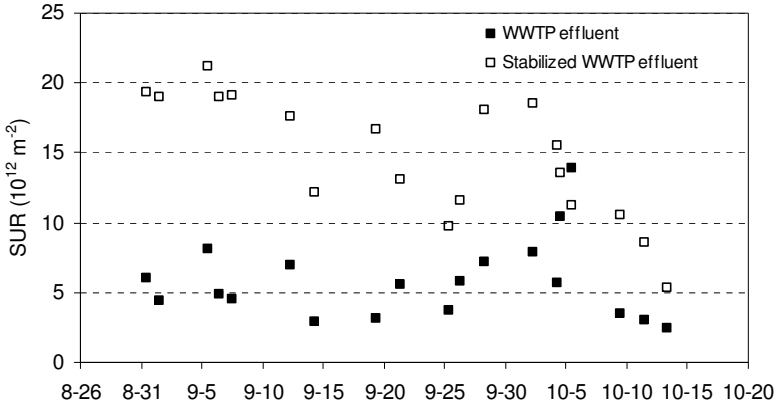


Figure 6.3 – The SUR values of WWTP effluent and stabilized WWTP effluent at the WWTP Sas van Gent during the period August – October, 2006

6.2.3 Foulants

Next to the measurements of SUR values the concentrations of several foulants have been analysed. Table 6.1 presents the average concentrations of foulants of WWTP effluent and stabilized WWTP effluent during the experimental period. The trends of the concentrations of foulants are presented in Figure 6.4.

Table 6.1 – The average concentrations of foulants of WWTP effluent and stabilized WWTP effluent at the WWTP Sas van Gent during the period August – October, 2006

Parameter	Unit	WWTP effluent	Stabilized WWTP effluent
Proteins	mg/L	38.8 (± 25.6)	19.7 (± 4.5)
Polysaccharides	mg/L	7.6 (± 1.6)	6.9 (± 1.0)
DOC	mg/L	23.3 (± 4.9)	22.5 (± 6.7)
Humic substances	1/cm	0.50 (± 0.21)	0.42 (± 0.13)
Colour	mg Pt/L	86.4 (± 26.7)	67.4 (± 15.1)
PUVA	L/mg·m	1.6 (± 0.9)	2.2 (± 0.7)

In contrast to the results of the SUR measurements the stabilization pond showed a clear effect on the concentrations of foulants. All concentrations of foulants decreased during the pond passage. Although the concentrations of foulants after pond passage are still high compared to concentrations analysed at other WWTPs in the Netherlands, this may be related to the wastewater source (food producing factory) of the WWTP Sas van Gent. The other WWTPs are mainly fed with domestic wastewater.

Next to the observed decrease of the concentrations of foulants a more steady water quality can be noticed from Table 6.1 and Figure 6.4 but this was not the case for the DOC concentrations. It might be that the DOC concentrations were influenced by the presence of algae in the stabilization pond. During the DOC analyses all dissolved carbon including possible rests and products of algae are measured instead of specific compounds of DOC.

As expected from the decrease of the foulants concentrations the PUVA value (the ratio of ultraviolet absorption to proteins) changed after the stabilization pond. Before the stabilization pond the concentrations of proteins were high compared to humic substances but after the stabilization pond the PUVA value was comparable with other WWTP effluent values. The concentrations of proteins decreased more than the amount of humic substances. Anyhow, considering the different wastewater sources, te Poele (2005) found PUVA values in the same range (2.0 – 2.8 L/mg·m) at other WWTPs in the Netherlands.

6.2.4 Discussion

The decrease of filterability i.e. increase of the SUR value by pond passage is also shown by other researchers. te Poele (2005) investigated twice at the WWTP Tilburg Noord the SUR values after a secondary clarifier and after the pond system. The average SUR value after final sedimentation was $8.0 \cdot 10^{12}$ m^{-2} and after the pond system $11.1 \cdot 10^{12}$ m^{-2}. Geilvoet (2007) presented also an increase of SUR values after an artificial pond system but it is difficult to compare the configurations (dimensions, vegetation, retention time, etc.) and circumstances (temperature, weather conditions, etc.) of the stabilization pond of WWTP Sas van Gent with the systems of te Poele (2005) and Geilvoet (2007). Nevertheless, in general a negative impact of pond systems on the SUR values can be observed. This observation (pollution by algae and following consequences) is also reported by Bixio and Wintgens (2006) as a possible disadvantage of maturation ponds.

The effect of pond passage on the concentrations of foulants in WWTP effluent was also investigated by te Poele (2005) and Geilvoet (2007) but unfortunately both researchers noticed different findings. Te Poele (2005) found a slight increase of all the concentrations of foulants where Geilvoet (2007) observed equal or lower concentrations of foulants after pond passage. In particular the concentrations of proteins presented a strong decrease during pond passage in the research of Geilvoet (2007). This last observation is in accordance with the result presented in this section but as already emphasized earlier the configurations of the stabilization ponds were completely different. This makes it difficult to compare the results but in contrast to the filterability (SUR) no significant negative impact of pond passage on the concentrations of foulants of WWTP effluent can be noticed.

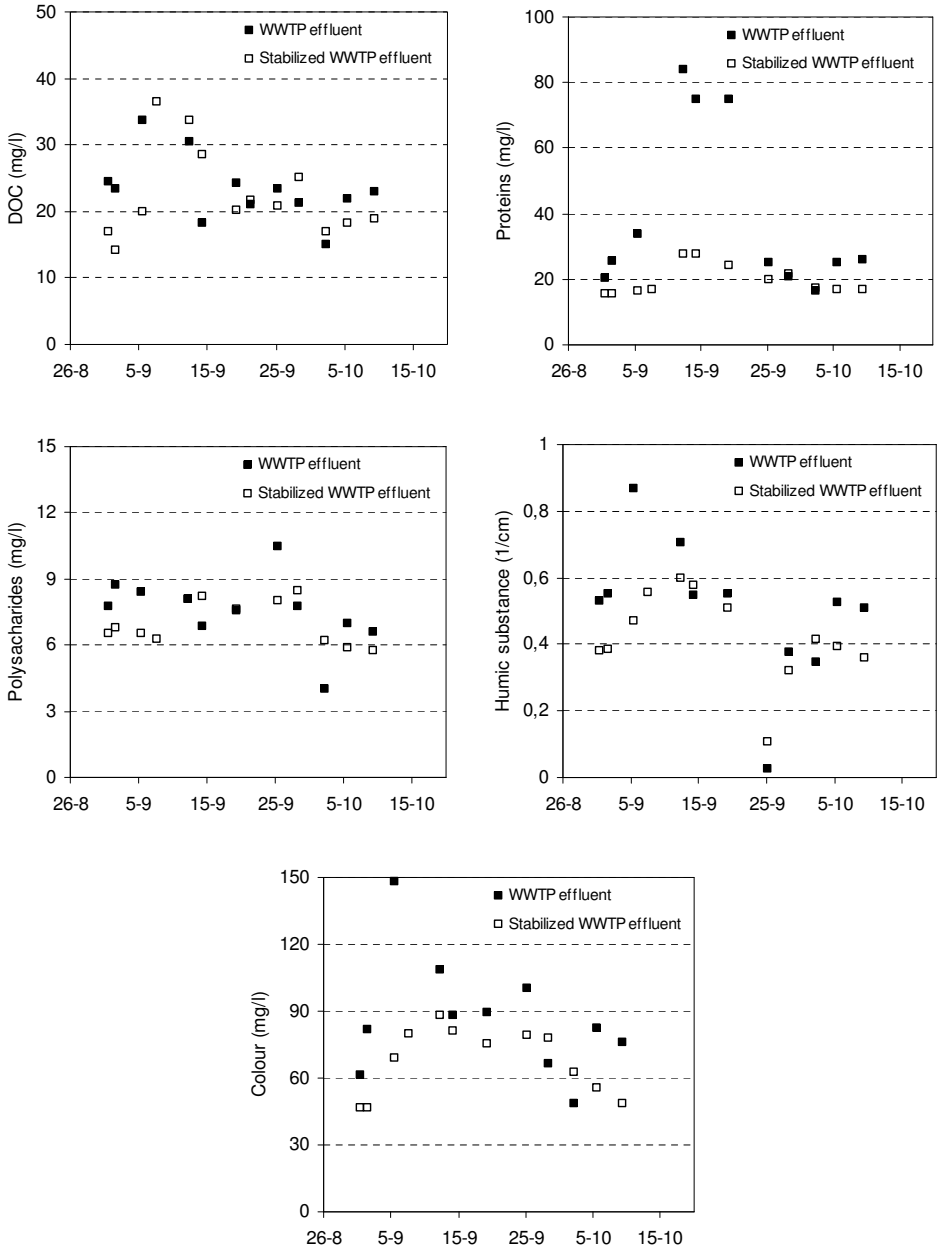


Figure 6.4 – The concentrations of DOC, proteins, polysaccharides, humic substances and colour of the WWTP effluent and stabilized WWTP effluent at the WWTP Sas van Gent during the period August – October, 2006

6.3 Performance of the pretreatment steps

6.3.1 Experimental setup

Also the effect of using different water sources (WWTP effluent after the secondary clarifier and stabilized WWTP effluent) on the performance of the pretreatment steps before the ultrafiltration units of the UF-RO installation at the WWTP Sas van Gent has been investigated. As presented in Figure 6.2 the pretreatment steps consist of (first) coagulation, dual media filtration and (second) coagulation. For both coagulation steps ferric chloride was used as coagulant. During the first coagulation step 2.5 mg Fe^{3+}/L was dosed and mixed by the valves and curves in the pipeline. Some flocculation took place in the upper water layer of the dual media filter. The operational parameters of the dual media filter are summarized in Table 6.2.

Table 6.2 – Operational parameters of the dual media filter of the UF-RO installation at the WWTP Sas van Gent during the period August – September, 2006

Parameter	Unit	Value
Flow	m^3/h	167
Hydraulic load	m/h	9.3
Filter run	h	8
Backwash velocity	m/h	60

After the dual media filter the filtrate was collected in a buffertank. The following pretreatment step, the second coagulation (1.0 mg Fe^{3+}/L), was placed between the buffer tank and the ultrafiltration units and the mixing was done by the pumps. Next to the addition of ferric chloride, sodium hypochlorite was dosed at this dosing point to prevent biological growth in the system.

The period of the experiments was divided in two phases. During the first phase, from August, 31th till October, 12th, the UF-RO installation and dual media filter had been fed by stabilized WWTP effluent. During the second phase, October, 13th till December, 12th, the UF-RO installation had been directly fed with WWTP effluent after the secondary clarifier. Due to the availability of just one dual media filter it was not possible to compare stabilized WWTP effluent and WWTP effluent at the same time. Therefore the experiment had been divided in two phases.

During both phases of the experimental period samples were taken. Samples were taken of the WWTP effluent (after the secondary clarifier or after the stabilization pond), after the first coagulation, dual media filtrate and after the second coagulation (ultrafiltration feedwater). In Figure 6.2 the sampling points are shown and in Figure 6.5 the different actions after sampling are summarized. The SUR measurements were directly performed after sampling on site. The analyses of the foulants were done within a week after sampling at the laboratory of Sanitary Engineering of Delft University of Technology. Therefore, the samples for analyses were stored on site in a refrigerator.

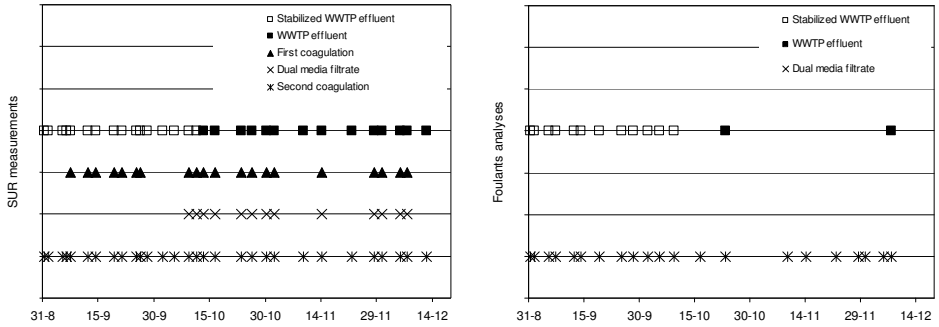


Figure 6.5 – Dates of sampling of the WWTP effluent, stabilized WWTP effluent, after first coagulation, dual media filtrate and after second coagulation and performed actions (SUR measurements left and foulants analyses right) during the period August – December, 2006

6.3.2 SUR values

6.3.2.1 First coagulation

Table 6.3 presents the SUR values before and after the first coagulation and the relative SUR decrease. Despite the fluctuating SUR values of the WWTP effluent the results show a significant effect of the coagulation during both phases. During the first phase (stabilized WWTP effluent) the average relative SUR decrease was 24% (± 9%) and during the second phase (WWTP effluent) 19% (± 20%). These different relative SUR decrease values may be related to the SUR values before coagulation. As presented in the previous section and in Table 6.3 the stabilized WWTP effluent shows higher SUR values than the WWTP effluent after the secondary clarifier. To gain more insight into this difference the results of Table 6.3 are graphically presented in Figure 6.6.

Table 6.3 – The SUR values of the stabilized WWTP effluent, WWTP effluent, after first coagulation and the relative SUR decrease of the UF-RO installation at the WWTP Sas van Gent during the period September – December, 2006

Date	(Stabilized) WWTP effluent SUR (10^{12} m^{-2})	First coagulation SUR (10^{12} m^{-2})	Relative SUR decrease (%)
	Stabilized WWTP effluent		
06-09-06	19.0	16.5	13
07-09-06	19.0	13.0	32
12-09-06	17.6	11.2	36
14-09-06	12.2	10.9	11
19-09-06	16.7	11.5	31
21-09-06	13.1	10.0	24
25-09-06	9.8	8.1	17
26-09-06	11.5	9.9	14
09-10-06	10.5	7.3	30
11-10-06	8.6	6.2	28
Average	13.8 (± 3.9)	10.5 (± 3.0)	24 (± 9)
	WWTP effluent		
13-10-06	2.4	2.0	17
16-10-06	5.1	6.3	-24
23-10-06	4.9	3.5	29
26-10-06	5.5	4.4	20
30-10-06	4.0	3.7	8
01-11-06	3.9	3.3	15
14-11-06	4.4	4.6	-5
22-11-06	12.1	7.3	40
28-11-06	10.3	5.9	43
30-11-06	15.6	8.9	43
05-12-06	21.8	14.7	33
07-12-06	22.5	19.9	12
Average	9.4 (± 7.1)	7.0 (± 5.3)	19 (± 20)

Figure 6.6 shows no significant decrease of the SUR value when the SUR values of WWTP effluent are around $5.0 \cdot 10^{12}$ m^{-2}. This observation could only be made during the second phase. Only in this period SUR values of about $5.0 \cdot 10^{12}$ m^{-2} had been measured. During the first phase the SUR values of stabilized WWTP effluent were almost always more than $10 \cdot 10^{12}$ m^{-2}. As represented in Figure 6.6 these higher SUR values resulted in a more significant effect of coagulation compared to SUR values of $5.0 \cdot 10^{12}$ m^{-2}.

Figure 6.6 also shows a slight difference between stabilized WWTP effluent and WWTP effluent after the secondary clarifier when the initial SUR values were more than $10.0 \cdot 10^{12}$ m^{-2}. The average SUR decrease of stabilized WWTP effluent and the WWTP effluent were respectively 24% and 34%. This resulted in a difference of 10% when the SUR values of both stabilized WWTP and WWTP effluent were more than $10.0 \cdot 10^{12}$ m^{-2}. In comparison the total average SUR decrease was 24% and 19% of respectively the stabilized WWTP and WWTP effluent. This difference may be related to the different water quality as shown in Table 6.1. The composition of stabilized WWTP effluent differed from the WWTP effluent after the secondary clarifier.

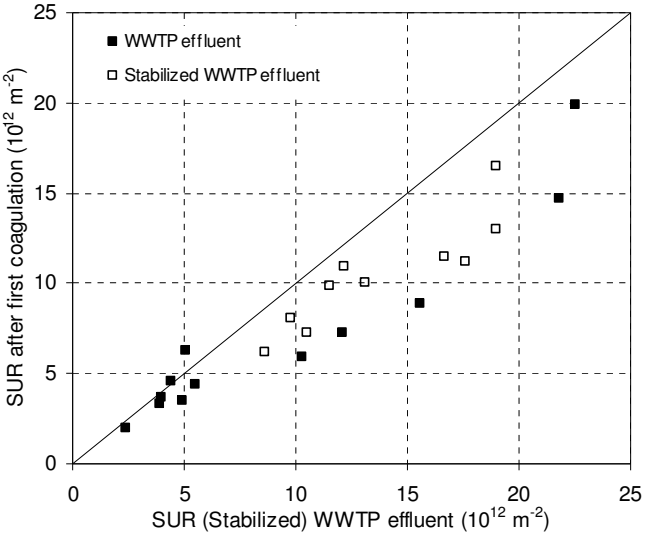


Figure 6.6 – Relation between the SUR value of stabilized WWTP effluent, WWP effluent after the secondary clarifier and the SUR value after the first coagulation of the UF-RO installation at the WWTP Sas van Gent during the period September – December, 2006

6.3.2.2 Dual media filter

The effect of the following pretreatment step, the dual media filter, on the SUR values after first coagulation is presented in Table 6.4 and Figure 6.7. As shown in Figure 6.5 during the first phase only two times the SUR value of dual media filtrate had been measured. Therefore it is not possible to compare the SUR decrease of both phases. Nevertheless Figure 6.7 represents a similar trend as the results of the previous section. When the SUR values after the first coagulation were around $5.0 \cdot 10^{12}$ m^{-2} the effect of the dual media filter became less. Below SUR values after first coagulation of $5.0 \cdot 10^{12}$ m^{-2} the average decrease was 13%. When the SUR values after first coagulation were higher ($> 5.0 \cdot 10^{12}$ m^{-2}) the average SUR decrease was 28%. Therefore it seemed that the relative SUR decrease is related to the SUR value of the feedwater of the dual media filter.

Table 6.4 – The SUR values after the first coagulation and dual media filter together with the relative SUR decrease of the UF-RO installation at the WWTP Sas van Gent during the period September – December, 2006

Date	First coagulation SUR (10^{12} m^{-2})	Dual media filtrate SUR (10^{12} m^{-2})	Relative SUR decrease (%)
	Stabilized WWTP effluent		
09-10-06	7.3	6.0	18
11-10-06	6.2	4.6	26
Average	6.8	5.3	22
	WWTP effluent		
13-10-06	2.0	4.3	-115
16-10-06	6.3	3.6	43
23-10-06	3.5	3.2	9
26-10-06	4.4	3.4	23
30-10-06	3.7	3.4	8
01-11-06	3.3	3.1	6
14-11-06	4.6	4.6	0
28-11-06	5.9	5.3	10
30-11-06	8.9	7.2	19
05-12-06	14.7	9.0	39
07-12-06	19.9	14.3	28
Average	7.0 (± 5.5)	5.6 (± 3.4)	18 (± 14)[1]

[1] The outlier of 13-10-2006 is not taken into account

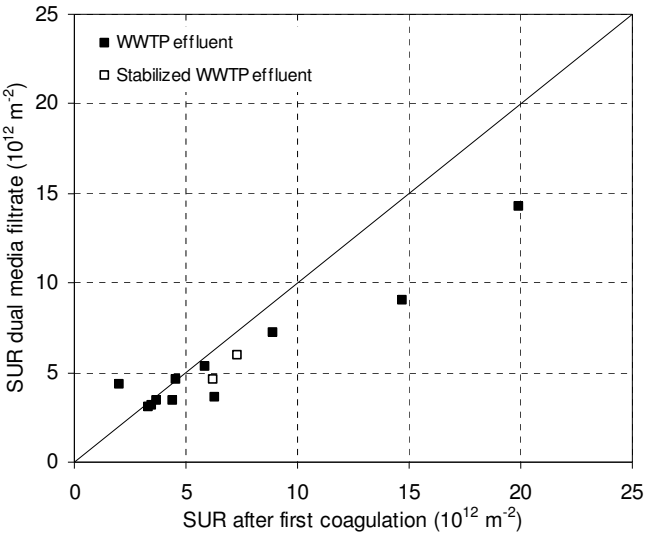


Figure 6.7 – Relation between the SUR values after first coagulation and the SUR values of dual media filtrate of the UF-RO installation at the WWTP Sas van Gent during the period September – December, 2006

6.3.2.3 Second coagulation

The effect of the second coagulation (1.0 mg Fe^{3+}/L), the last pretreatment step before the ultrafiltration units, on the SUR values of dual media filtrate are presented in Table 6.5 and Figure 6.8. During both phases of the experimental period only once the SUR value of dual media filtrate was above $10.0 \cdot 10^{12}$ m^{-2}. Consequently the relative SUR decrease was lower compared to the pretreatment steps before the second coagulation. The total average SUR decrease during the second phase was 12%. Just like the results of the previous pretreatment steps it seemed again that the relative SUR decrease depends on the initial SUR value. When the SUR values of dual media are above $5.0 \cdot 10^{12}$ m^{-2} the decrease was significantly. Below initial SUR values of $5.0 \cdot 10^{12}$ m^{-2} the effect became less significant as illustrated in Figure 6.8.

Table 6.5 – The SUR values of the dual media filter and after second coagulation together with the relative SUR decrease of the UF-RO installation at the WWTP Sas van Gent during the period September – December, 2006

Date	Dual media filtrate SUR (10^{12} m^{-2})	Second coagulation SUR (10^{12} m^{-2})	Relative SUR decrease (%)
	Stabilized WWTP effluent		
10-09-2006	6.0	5.7	5
10-11-2006	4.6	4.6	0
Average	5.3	5.2	2.5
	WWTP effluent		
10-13-2006	4.3	4.5	-5
10-16-2006	3.6	3.3	8
10-23-2006	3.2	2.8	13
10-26-2006	3.4	3.5	-3
10-30-2006	3.4	3.3	3
11-01-2006	3.1	3.0	3
11-14-2006	4.6	3.6	22
11-28-2006	5.3	4.4	17
11-30-2006	7.2	5.8	19
12-05-2006	9.0	8.3	8
12-07-2006	14.3	7.8	45
Average	5.6 (± 3.4)	4.6 (± 1.9)	12 (± 14)

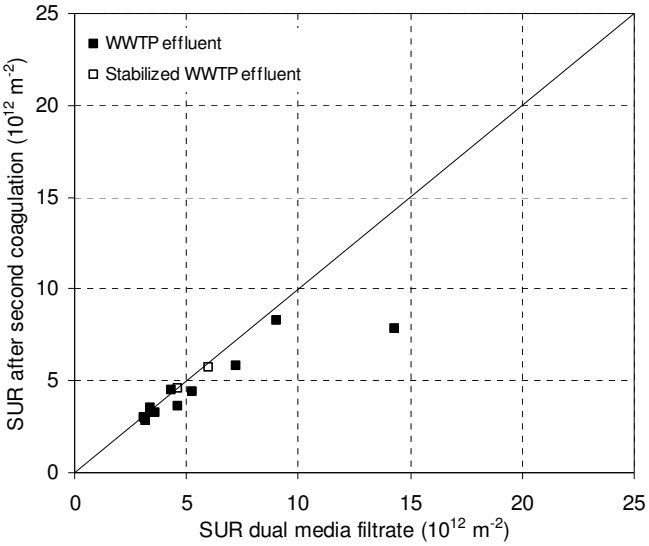


Figure 6.8 – Relation between the SUR values of dual media filtrate and the SUR values after the second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

6.3.2.4 First coagulation – dual media filter – second coagulation

Table 6.6 displays the overall effect of the pretreatment steps on the SUR values of stabilized WWTP effluent and WWTP effluent after the secondary clarifier. Considering the stabilized WWTP effluent the average SUR value decreased with more than 50% after the different pretreatment steps. This resulted almost all times in SUR values below $10.0 \cdot 10^{12}$ m^{-2}. A similar trend was observed during the second phase when the UF-RO installation was fed with WWTP effluent after the secondary clarifier. During this phase the average SUR decrease (40%) was less but the initial SUR values were lower as well. The average SUR value of WWTP effluent after the secondary clarifier was $9.2 \cdot 10^{12}$ m^{-2} ($\pm$ $6.5 \cdot 10^{12}$ m^{-2}) instead of $15.3 \cdot 10^{12}$ m^{-2} ($\pm$ $4.2 \cdot 10^{12}$ m^{-2}) for stabilized WWTP effluent.

In contrast to the initial SUR values of WWTP effluent and stabilized WWTP effluent the SUR values after second coagulation were rather steady. The absolute variations of the SUR values of stabilized WWTP effluent and WWTP effluent after the secondary clarifier were respectively $4.2 \cdot 10^{12}$ m^{-2} and $6.5 \cdot 10^{12}$ m^{-2}. After the pretreatment steps these values were significantly lower; the absolute variation of stabilized WWTP effluent was $1.4 \cdot 10^{12}$ m^{-2} and of WWTP effluent $1.7 \cdot 10^{12}$ m^{-2}. This is an important observation regarding the operation of ultrafiltration installations. For a good and stable process performance of membrane installations stable or constant water quality of the feedwater is preferred (Lazarova, *et. al*.,

2007). Therefore it is evident that the pretreatment steps contribute to a better process performance in two ways. Firstly pretreament lowers the initial SUR value and secondly it decreases the absolute variation of the initial SUR values.

Table 6.6 – The SUR values of stabilized WWTP effluent, WWTP effluent after the secondary clarifier and after second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

Date	(Stabilized) WWTP effluent SUR (10^{12} m^{-2})	Second coagulation SUR (10^{12} m^{-2})	Relative SUR decrease (%)
	Stabilized WWTP effluent		
31-08-06	19.3	7.9	59
01-09-06	19.0	5.7	70
05-09-06	21.2	6.8	67
06-09-06	19.0	10.2	46
07-09-06	19.0	8.7	54
12-09-06	17.6	6.4	63
14-09-06	12.2	5.2	57
19-09-06	16.7	6.8	59
21-09-06	13.1	7.3	44
25-09-06	9.8	5.6	43
26-09-06	11.5	7.1	38
28-09-06	18.0	6,9	62
02-10-06	18.5	6.1	67
05-10-06	11.2	6.7	40
09-10-06	10.5	5.7	46
11-10-06	8.6	4.6	47
Average	15.3 (± 4.2)	6.7 (± 1.4)	54 (± 11)
	WWTP effluent		
13-10-06	5.3	4.5	15
16-10-06	5.1	3.3	35
23-10-06	4.9	2.8	43
26-10-06	5.5	3.5	36
30-10-06	4.0	3.3	18
01-11-06	3.9	3.0	23
09-11-06	4.3	3.4	21
14-11-06	4.4	3.6	18
22-11-06	12.1	5.8	52
28-11-06	10.3	4.4	57
30-11-06	15.6	5.8	63
05-12-06	21.8	8.3	62
07-12-06	22.5	7.8	65
12-12-06	8.4	4.8	43
Average	9.2 (± 6.5)	4.6 (± 1.7)	40 (± 18)

To gain more insight into the effect of pretreatment changes the SUR values of stabilized WWTP effluent and WWTP effluent after the secondary clarifier are related to the SUR values after first coagulation, dual media filter and second coagulation. In Figure 6.9 the relation between the SUR values of stabilized WWTP effluent and the SUR values after first and second coagulation is presented. Unfortunately the SUR value of dual media filtrate had been measured only twice and therefore is not plotted in the figure. In spite of this lack and the poor relations a distinguished effect can be noticed between the first and second

coagulation. After the first coagulation the SUR decreased with 24% and subsequently after multi media filtration and second coagulation with 54%.

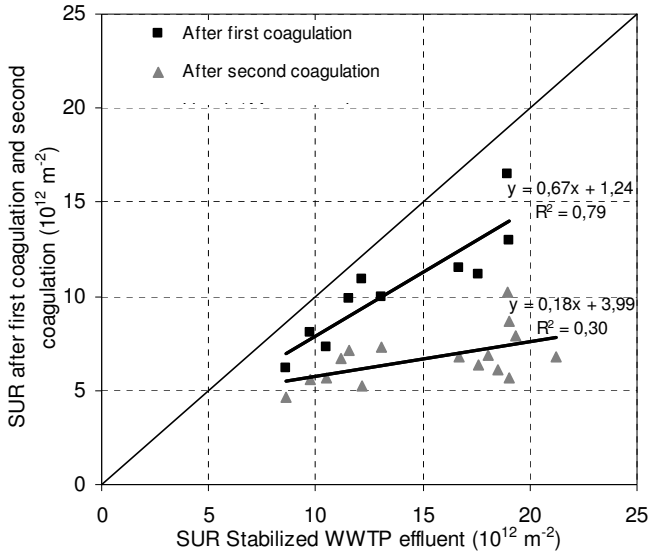


Figure 6.9 – Relation between the SUR values of stabilized WWTP effluent and the SUR values after first coagulation and second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

Figure 6.10 displays the relation during the second phase when WWTP effluent after the secondary clarifier was used as feedwater for the UF-RO installation. This figure shows that each pretreatment step contributed significantly to the total SUR decrease of WWTP effluent. Nevertheless the contribution of each pretreatment highly depends on the SUR value of WWTP effluent. When the initial SUR values were around $5.0 \cdot 10^{12}$ m^{-2} the effect of all the pretreatment steps was minimal. But with increasing SUR values of the WWTP effluent the contributions of each pretreatment step increased as well.

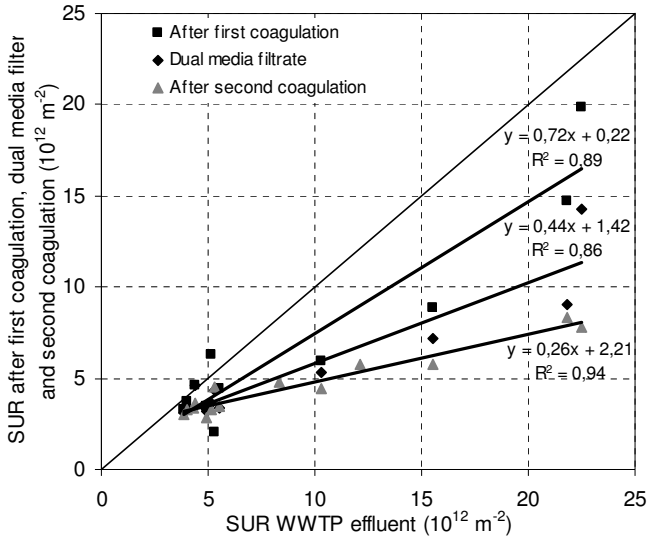


Figure 6.10 – Relation between the SUR values of WWTP effluent and the SUR values after first coagulation, dual media filter and second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

6.3.3 Foulants

Table 6.7 presents the average concentrations of foulants before and after the different pretreatment steps upstream the UF-RO installation. During the second phase (October – December, 2006) of the experimental period only two times the concentrations of foulants of the WWTP effluent had been analysed. Therefore it is difficult to draw any conclusion about the different concentrations of foulants of WWTP effluent after second coagulation. But more results are available of the first phase. During this phase no significant difference is observed between the foulants concentrations of stabilized WWTP effluent and after second coagulation. This is in contrast with the results of the SUR measurements. These results, presented in the previous section, showed a significant decrease of the initial SUR value after pretreatment. This may mean that the decrease of the SUR values did not directly correlate with the foulants concentrations. This observation was also noticed in chapter 5 and is also supported by the research of te Poele (2005).

Table 6.7 – Average concentrations of foulants of stabilized WWTP effluent, WWTP effluent after the secondary clarifier and after second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

		After stabilization pond August – October, 2006		After secondary clarifier October – December, 2006	
Parameter	Unit	WWTP effluent	Second coagulation	WWTP effluent[1]	Second coagulation
DOC	mg/L	22.5 (± 6.7)	21.6 (± 3.3)	26.1	22.2 (± 3.5)
Proteins	mg/L	19.7 (± 4.5)	19.9 (± 3.7)	28.0	24.3 (± 1.4)
Polysaccharides	mg/L	6.9 (± 1.0)	6.5 (± 0.7)	7.3	5.9 (± 1.0)
Humic substances	1/cm	0.42 (± 0.13)	0.43 (± 0.08)	0.56	0.45 (± 0.09)
Colour	mg Pt/L	67.4 (± 15.1)	64.0 (± 13.6)	98.7	73.7 (± 15.7)
PUVA	L/mg·m	2.2 (± 0.7)	2.2 (± 0.3)	2.0	1.9 (± 0.4)

[1] Average of two values

6.3.4 Discussion

The (relative) effect of the pretreatment steps on the SUR values of stabilized WWTP effluent and WWTP effluent after the secondary clarifier is summarized in Table 6.8. In general the effect of the pretreatment steps is more significant for stabilized WWTP effluent than for WWTP effluent after the secondary clarifier. But as earlier discussed this difference may be related to the initial SUR value. As shown in Table 6.6 the average SUR values of stabilized WWTP effluent and WWTP effluent after the secondary clarifier were respectively $15.3 \cdot 10^{12}$ m^{-2} ($\pm$ $4.2 \cdot 10^{12}$ m^{-2}) and $9.2 \cdot 10^{12}$ m^{-2} ($\pm$ $6.5 \cdot 10^{12}$ m^{-2}). The influence of the initial SUR value was also presented in the Figures 6.6. and 6.7. When the SUR values of stabilized WWTP effluent and WWTP effluent were below $5.0 \cdot 10^{12}$ m^{-2} the effect of pretreatment was very little. Therefore it seemed important to accompany this kind of experiment with measurements of the initial SUR values in order to evaluate the performance properly. Too often pilot experiments are performed with one unit and the effect of varying filterability is not taken into account. Therefore this kind of experiments should be accompanied with regular measurements of the filterability of the feedwater. Despite of this shortcoming the results of this chapter are in line with findings of other researchers (Bourgeous *et al*., 2001; Fan *et al*., 2008; Haberkamp *et al*., 2007; Roorda, 2004; Shon *et al*., 2004; te Poele, 2005). Considering the different scale, circumstances and concentrations of coagulant all these researchers presented generally a positive effect of coagulation and filtration on the initial filterability of WWTP effluent. For instance Roorda (2004) presented a relative SUR decrease of approximately 20 – 30% after coagulation and filtration of WWTP effluent. The initial SUR values during these experiments were $12.4 \cdot 10^{12}$ m^{-2} and $17.0 \cdot 10^{12}$ m^{-2}. These average SUR decrease values are more or less in the range of values found in this chapter.

Table 6.8 – Summary of the average relative SUR decrease of stabilized WWTP effluent and WWTP effluent (after the secondary clarifier) after first coagulation, dual media filter and second coagulation of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

	Stabilized WWTP effluent	WWTP effluent
Pretreatment	Average SUR decrease (%)	Average SUR decrease (%)
WWTP effluent – first coagulation	24 (± 9)	19 (± 20)
First coagulation – dual media filter	22[1]	18 (± 14)
Dual media filter – Second coagulation	2.5[1]	12 (± 14)
WWTP effluent – first coagulation – dual media filter – second coagulation	54 (± 11)	40 (± 18)

[1)] Average of two values

The effect of pretreatment on the concentrations of foulants during the first phase of the research period was minimal. These results are in contrast with the findings of the SUR measurements. During the second phase the effect was more significant but only two foulants analyses of the WWTP effluent after the secondary clarifier had been performed. Anyhow this difference seemed to be related to the concentration of foulants. In section 6.2.3 it was shown that the concentration of foulants of WWTP effluent after the secondary clarifier are generally higher than in the stabilized WWTP effluent. Probably not only the concentration of foulants played a role but also the general water matrix. As shown in section 6.2.3 not only the concentrations of foulants changed but for instance also the PUVA (the ratio of ultraviolet absorption to proteins) values. From literature (Huang *et al*., 2007) it is known that the (organic carbon) composition and in particular the presence of humic substances water plays an important role during coagulation.

Furthermore the trend of the foulants concentrations cannot be related with the trend of the SUR values. A significant decrease of the SUR value did not correlate with a decrease of the concentration of foulants. Earlier research of Roorda (2004) and also the results of chapter 5 of this thesis determined the fraction 0.1 – 0.2 µm as the size fraction that predominantly contributes to the SUR value of WWTP effluent. Te Poele (2005) suggested that organic colloids or submicron particles like cell fragments and macro molecules may be the main components of this fraction. However the applied analytical methods of this research did not determine specifically this fraction. The samples for chemical analyses had been filtered over a 0.45 µm filter prior to the analyses. Therefore it may be possible that organic substances are analysed that do not relate to SUR value of feedwater. More specific analyses and methods (i.e. fractionation) has to be considered to determine the possible relation between the foulants and SUR value of WWTP effluent.

6.4 Performance of the ultrafiltration membrane units

6.4.1 Experimental setup

The influence of both water sources, WWTP effluent after the secondary clarifier and stabilized WWTP effluent, on the performance of the ultrafiltration units had been investigated during the research period (August – December, 2006) as well. As shown in chapter 4 the increase of trans membrane pressure and resistance in relation to the SUR value of ultrafiltration feedwater depends on the condition of membranes. More fouled membranes presented a higher increase of trans membrane pressure and resistance per filtration run than less fouled membranes at the same SUR value of ultrafiltration feedwater. Therefore in this experiment the relation was investigated during both phases of the research period to identify the impact of both water sources on the performance of the ultrafiltration membranes.

During both phases the ultrafiltration units had been operated under normal conditions like described in chapter 3. The ultrafiltration units delivered a fixed amount of cubic meters per hour. This operation mode resulted frequently in a varying flux during a filtration run. Consequently this variation influenced the resistance increase during a filtration run. Therefore only data were collected when the flux was stable (standard deviation $\leq 5\%$) during a filtration run. The procedure of the calculations of resistance increase is described thoroughly in chapter 4.

Samples were taken just before the feedwater entered the ultrafiltration units (see Figure 6.2). Subsequently the SUR measurements had been directly performed on site and compared with the process data of the ultrafiltration units 1 and 4. As described in chapter 4 both units were almost identical but the lifetime and the conditions of the membrane modules were different. The membranes of the ultrafiltration unit 1 were replaced half a year before the experiment. The membranes of the ultrafiltration unit 4 were 4 years old and expectedly therefore probably more affected by fouling, chemical cleanings, etc.

Between the first phase and the second phase of the research period the membrane modules of the ultrafiltration unit 4 were intensively cleaned with hydrochloric acid and oxalic acid to remove residual fouling. The membrane modules of ultrafiltration unit 1 were not intensively cleaned during the whole research period.

6.4.2 Results

Table 6.9 displays the results of the whole research period. Unfortunately many process data of the ultrafiltration units were not useful due to the mentioned varying flux. Therefore during the second phase only a few points for the relation between the SUR value and increase of trans membrane pressure and resistance are presented.

During the whole period the filterability of the ultrafiltration feedwater was suitable for ultrafiltration. As shown in Table 6.9 all the SUR values were mostly below $10.0 \cdot 10^{12}$ m^{-2}.

Despite the availability of just a few data during the second phase the effect of the chemical cleaning of ultrafiltration unit 4 between both phases on the process performance was significantly. During the second phase the rate of resistance increase was much lower than during the first phase with uncleaned membrane modules. This effect is graphically presented in Figure 6.11 together with the results of ultrafiltration unit 1.

Table 6.9 – The SUR values of the ultrafiltration feedwater and the dR/dt values of the ultrafiltration units 1 and 4 of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

Date	Feedwater	Ultrafiltration unit 1		Ultrafiltration unit 4	
	SUR (10^{12} m^{-2})	Average flux ($L/m^2 \cdot h$)	dR/dt (10^{12} $1/m \cdot h$)	Average flux ($L/m^2 \cdot h$)	dR/dt (10^{12} $1/m \cdot h$)
	Stabilized WWTP effluent after pretreatment				
05-09-06	6.8	23.3	0.17	26.9	0.70
06-09-06	10.2	52.1	0.39	59.5	2.24
07-09-06	8.7	26.5	0.31	-	-
12-09-06	6.4	22.3	0.11	26.0	0.24
14-09-06	5.2	52.1	0.24	59. 1	1.12
19-09-06	6.8	-	-	53.7	0.93
25-09-06	5.6	22.8	0.07	26.1	0.63
28-09-06	6.9	33.1	0.13	38.7	0.75
09-10-06	5.7	-	-	26.1	0.29
	WWTP effluent after pretreatment				
26-10-06	3,5	22.7	0.14	-	-
30-10-06	3,3	32.4	0.13	37.2	0.20
14-11-06	3.6	-	-	59.1	0.24
22-11-06	5.8	-	-	26.7	0.16
28-11-06	4,4	23.3	0.16	26.7	0.15
05-12-06	8.3	-	-	25.8	0.18
07-12-06	7.8	-	-	58.8	0.31

Figure 6.11 shows no significantly different relation between the SUR values of ultrafiltration feedwater and the dR/dt values of the ultrafiltration unit 1 during both phases. In other words the relation was more or less similar. But in contrast to the results of ultrafiltration unit 1 the results of the ultrafiltration unit 4 presented a significant difference between both phases. During the first phase the dR/dt values were much higher at the same SUR values of ultrafiltration feedwater. This effect may be related to the intensive chemical cleaning between both phases. As described earlier the membrane modules of the ultrafiltration unit 4 were intensively cleaned (cleaning-in-place) with hydrochloric acid and oxalic acid between

both phases. This cleaning in place was not applied to the membrane modules of the ultrafiltration unit 1. These membrane modules were only hydraulically (backflushes) and chemically cleaned according to the normal operational procedure and presented more or less similar relations during both phases. This finding may suggest that fouling caused by the filtration of stabilized WWTP effluent can be removed by either hydraulic, chemical cleaning and (chemical) cleaning in place. In general it appeared that stabilized WWTP effluent had no significant positive or negative impact on the membranes modules. The fouling could be removed by hydraulic and/or chemical cleaning.

When the membrane modules are fouled (first phase of the ultrafiltration unit 4) the SUR values of the ultrafiltration feedwater will have a significant impact on the operational parameters. Fouled membranes present significantly higher dR/dt values at the same SUR values of ultrafiltration feedwater. Moreover this observation is thoroughly discussed in chapter 4 of this thesis.

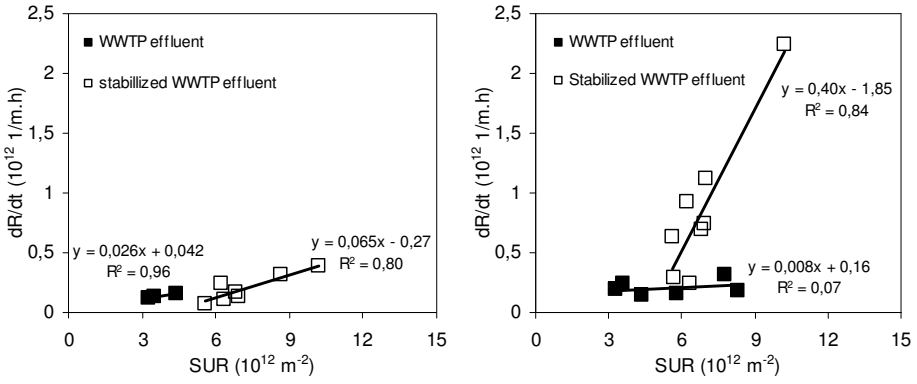


Figure 6.11 – Relation between SUR values of ultrafiltration feedwater and the resistance slope per filtration run of ultrafiltration unit 1 (left) and 4 (right) of the UF-RO installation at the WWTP Sas van Gent during the period August – December, 2006

6.4.3 Discussion

The purpose of the experiment was to evaluate the performance of the ultrafiltration units of the UF-RO installation during the intake of stabilized WWTP effluent (first phase) and WWTP effluent after the secondary clarifier (second phase). For the evaluation the operational parameter, dR/dt, was plotted against the SUR value of ultrafiltration feedwater. This method was also used in chapter 4 of this thesis and resulted in a different relation between fouled and unfouled membrane modules. Fouled membrane modules will result in higher dR/dt values than unfouled membranes at the same SUR values of ultrafiltration feedwater. This observation is confirmed by the results of section 6.4.2. The results of ultrafiltration unit 4 presented a significant impact of the chemical cleaning in place on the

process performance. During the first phase the impact of the SUR value of feedwater on the dR/dt value was much higher than during the second phase. This effect was not observed for the ultrafiltration unit 1. During both phases the relation between the SUR values and dR/dt values was almost similar but these membrane modules were cleaned according to the standard operational procedures. Therefore it seemed that the formed fouling of stabilized WWTP effluent could be removed either hydraulically (backflush) and chemically. This observation was also found in the literature. Babel (2000) investigated the effect of algae fouling on the membrane performance. In this study it was shown that *Chlorella* algae cause significant fouling of both cellulose ester and PVDF membranes with a pore size of 0.45 µm. But after chemical and hydraulic cleaning the cake resistance reduced with more than 99%.

6.5 Evaluation

The objective of this chapter was to evaluate the performance of pretreatment and the ultrafiltration units during the intake of WWTP effluent after buffering in the stabilization pond by using the SUR measurement and foulants analyses. After pond passage the SUR values of WWTP effluent increased significantly. This was also confirmed by other researchers (te Poele, 2005; Geilvoet, 2007). Nevertheless, after coagulation, dual media filtration and coagulation the SUR values lowered significantly and became steady as well. In spite of this effect the SUR values of ultrafiltration feedwater were still higher during the first phase but this may be related to the higher initial SUR values of the stabilized WWTP effluent.

In contradiction to the SUR values the concentrations of foulants in the ultrafiltration feedwater were higher when WWTP effluent was used directly after the secondary clarifier. But this higher concentration did not result in higher SUR or dR/dt values. Therefore it seemed that the concentrations of foulants cannot be related to the ultrafiltration process performance.

The formed fouling during ultrafiltration of stabilized WWTP effluent seemed to be hydraulically and chemically reversible. After a (chemical) cleaning in place of the membrane modules of ultrafiltration unit 4 the dR/dt values decreased significantly at the same SUR values of ultrafiltration feedwater. The membranes of ultrafiltration unit 1 were only hydraulically and chemically cleaned and presented no significant difference between both phases. Therefore it seemed that the stabilized WWTP effluent did not significantly affect the long term operational performance of the ultrafiltration units.

In addition to the foulants concentrations and SUR values also other aspects played a role during operation of the UF-RO installation. Firstly the temperature and the linked viscosity of the ultrafiltration is an important aspect. Higher temperatures of feedwater required less energy for ultra- and reserve osmosis filtration when the same flux is applied. After the

secondary clarifier the average temperature in 2006 was 32 °C (Table 3.10) but during the intake of stabilized WWTP effluent the temperature varied between 20 – 25 °C. Considering the season (August – October, 2006) lower temperatures can be expected during wintertime and consequently impact on the energy consumption of the UF-RO installation. A second aspect is the sludge production. Due to algae growth in the stabilization pond more sludge production is expected during the pretreatement and ultrafiltration steps of the UF-RO installation. No calculations regarding this aspect were made but it will influence the operational costs as well.

Considering the foulants concentrations, SUR values and other aspects the implementation of a stabilization pond seemed not to be beneficial compared to the intake of WWTP effluent after the secondary clarifier. However, this conclusion cannot be made for periods with very high SUR values (bad quality event) of WWTP effluent after the secondary clarifier. Unfortunately such an event did not occur during the first phase of the research period.

6.6 Conclusions

- The SUR values of WWTP effluent after the secondary clarifier increased significantly during buffering in the stabilization pond at WWTP Sas van Gent. Contrarily the concentrations of foulants decreased significantly during the buffering in the stabilization pond.

- Considering the contradiction between the increase of SUR values and the decrease of concentrations of foulants it seemed that the presence of algae in the stabilization pond played an important role in the increase of the SUR values.

- The SUR values of stabilized WWTP effluent and WWTP effluent lowered and became more steady after the different pretreatment steps. Moreover, the effect of the pretreatment steps (coagulation – multi media filter – coagulation) is dependent on the SUR values before pretreatment.

- Considering the operational procedure of the ultrafiltration unit it seemed that retained fouling of stabilized WWTP effluent can be hydraulically and/or chemically removed.

- In general the results of this study show that the use of stabilized WWTP effluent is not considerably beneficial compared to the direct intake of WWTP effluent after the secondary clarifier at the WWTP Sas van Gent.

References

Babel, S. and Takizawa, S. (2000). *A study on membrane fouling due to algal deposition*. Water Science and Technology, 41 (10-11), 327-335.

Bixio, D. and Wintgens T. Editors (2006). *Water Reuse System Management Manual AQUAREC*. Project report European Comission, Luxembourg.

Bourgeous, K.N., Darby, J.L. and Tchobanoglous, G. (2001). *Ultrafiltration of wastewater: effects of particles, mode of operation, and backwash effectiveness*. Water Research, 35 (1), 77 – 90.

Fan, L., Nguyen, T., Roddick, F.A. and Harris, J.L. (2008). *Low-pressure membrane filtration of secondary effluent in water reuse: Pre-treatment for fouling reduction*. Journal of Membrane Science, 320, 135 – 142.

Geilvoet, A.J. (2007). *Fouling modification during pretreatment for dead-end ultrafiltration of wwtp effluent.* MSc thesis. Delft University of Technology, Delft, The Netherlands.

Haberkamp, J., Ruhl, A.S., Ernst, M. and Jekel, M. (2007). *Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting fouling behaviour in ultrafiltration*. Water Research, 41, 3794 – 3802.

Huang, H., Young, T. and Jacangelo, J.G. (2007). *Pretreatment and its effect and implications on water treatment by low pressure membranes.* Proceedings 4th IWA leading-edge conference and exhibition on water and wastewater technologies, June 3-6, Singapore, Republic of Singapore.

Lazarova, V., Gallego, S., Molina, V.G. and Rougé, P. *Problems of operation and main reasons of membranes in tertiary treatment systems.* Proceedings 6th IWA Specialist Conference on Wastewater Reclamation an Reuse for Sustainability, October 9 – 12, 2007, Antwerp, Belgium.

Shon, H.K., Vigneswaran, S., Kim, I.S., Cho, J. and Ngo, H.H. (2004). *Effect of pretreatment on the fouling of membranes: application in biologically treated sewage effluent.* Journal of Membrane Science, 234 (1-2), 111-120.

Trampé, J. (2007). *The SUR parameter for evaluating WWTP effluent ultrafiltration*, Training report (in Dutch), Evides Industriewater, Middelburg, The Netherlands.

7 Filterability and reversibility

7.1 Introduction

As presented in the previous chapters and reported as well by other researchers (Heijman *et al*., 2005; Huang *et al*., 2009) the general parameters to determine foulants (DOC, polysaccharides, proteins, humic substances and colour) cannot be applied to predict directly the fouling rate of dead-end ultrafiltration installations treating WWTP effluent. Therefore in this thesis the SUR measurement was presented as a possible tool to indicate and predict the fouling rate. However where other bench scale tests (Heijman *et al*., 2005; Kim and DiGiano, 2006; Huang *et al*., 2008) have the possibility to mimic full scale operation, the current SUR measurement does not. The SUR measurement only indicates the increase of cake resistance during a single filtration run (*filterability*) but does not give information about the *reversibility* of WWTP effluent. Nevertheless Roorda (2004) reported that a good filterability goes hand in hand with a good reversibility of WWTP effluent. This relation is also endorsed by te Poele (2005) and Lozier *et al*. (2008) but some other researchers (Heijman *et al*., 2005) show doubts about this relation. In general this relation is complex and depends on many factors. It firstly depends on the feedwater quality, secondly on the membrane properties (polymeric composition, clean water permeability, pore size or molecular weight cutoff, contact angle (an index of hydrophobicity), zeta potential (an index of surface charge and surface roughness) and thirdly on the operational conditions of an ultrafiltration installation. The main operational conditions that influence fouling of membranes are in accordance with Amy (2008): flux, recovery, pretreatment, chemical cleaning and hydraulic backwashing.

In this chapter the influence of the two operational conditions (flux and pretreatment) on both fouling parameters, filterability and reversibility, are investigated based on the interpretation of ultrafiltration pilot data. Furthermore a new configuration of the SUR equipment (SUR+ equipment) is introduced. With this equipment it is possible to mimic full scale operation of dead-end ultrafiltration installations in order to evaluate the filterability and reversibility of WWTP effluent under well-defined conditions.

7.2 Filterability and reversibility during the operation of an ultrafiltration pilot installation

7.2.1 Experimental setup

The experiments presented in this section had been performed at the WWTP Horstermeer during two different periods. During the first one (August, 2007) the ultrafiltration pilot installation was fed with filtrate of a multi media filter and during the second (May – July, 2008) with filtrate of a One Step Effluent Polishing (1-STEP®) filter.

The multi media filter, as described in chapter 3 and presented in Figure 7.1 was operated for denitrification and simultaneous phosphorous removal.

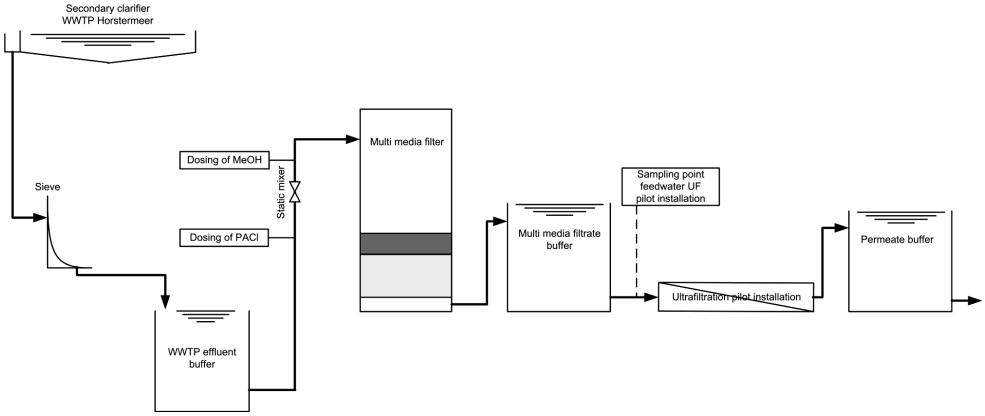


Figure 7.1 – Scheme of the multi media filter and ultrafiltration pilot installation at the WWTP Horstermeer

The feedwater of the multi media filter was WWTP effluent that passed a 450 µm curved sieve. The multi media filter consists of two filtration layers with anthracite on the top and quartz sand on the bottom. The multi media filter was backwashed with air and filtrate that was collected in the filtrate buffer. For denitrification, methanol was dosed. The dosage was based on the actual nitrate and oxygen concentrations of the (sieved) WWTP effluent. To remove phosphorous and suspended solids poly aluminium chloride was dosed in-line into the feedwater of the multi media filter. In Table 7.1 the operational parameters of the multi media filter during the first period are summarized.

Table 7.1 – Operational parameters of the multi media filter at the WWTP Horstermeer during the period August, 2007

Parameter	Unit	Value
Flow	m^3/h	10
Hydraulic load	m/h	12.5
Filter run	h	4 – 12
Surface area	m^2	0.8
Frequency 'bumping cleaning'	1/d	4
Methanol dosing	g MeOH/g NO_3-N	3.7 – 5.0
PACl in metal orthophosphate ratio	mol/mol	4

The 1-STEP® filter in combination with the pilot ultrafiltration installation was operated from May – July, 2008. Like the multi media filter the 1-STEP® filter was operated for denitrification and simultaneous phosphorous removal but the filter media of both filters differed. The 1-STEP® filter contained granulated activated carbon as filter media instead of anthracite and quartz sand. The feedwater of the 1-STEP® filter, WWTP effluent, passed a 450 µm curved sieve and was collected in the WWTP effluent buffer (see Figure 7.2).

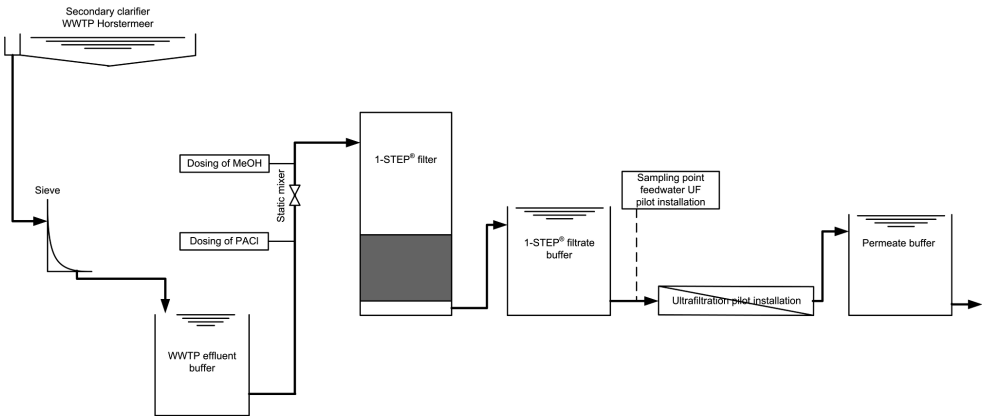


Figure 7.2 – Scheme of the 1-STEP® filter and ultrafiltration pilot installation at the WWTP Horstermeer

For the simultaneous removal of phosphorous and suspended solids, poly aluminium chloride was dosed in-line the feed water of the filter. The coagulation took place by a static mixer and flocculation occurred in the water volume above the filter medium, the flocculation time was approximately 8 minutes. For denitrification methanol was dosed based on the actual nitrate and oxygen concentrations in the WWTP effluent. In Table 7.2 the operational parameters of the 1-STEP® filter during the experimental period are given.

Table 7.2 – Operational parameters of the 1-STEP® filter at the WWTP Horstermeer during the period May – July, 2008

Parameter	Unit	Value
Flow	m^3/h	8 – 10
Hydraulic load	m/h	8 – 10
Filter run	h	12
Frequency 'bumping cleaning'	1/d	7 – 9
Bed height	m	1.9
Surface area	m^2	1.0
Empty Bed Contact Time	minutes	11 – 14
Methanol dosing	g MeOH/g NO_3-N	3 – 4
PACl in metal orthophosphate ratio	mol/mol	3 – 6

The ultrafiltration pilot installation described in chapter 3 was in operation during both periods. The feedwater of the ultrafiltration pilot installation was taken either of the filtrate buffer of the multi media filter or the 1-STEP® filter. During both periods the operational flux of the ultrafiltration pilot installation was varied several times as shown in Table 7.3. The frequency and the duration of the backflush (BF) were constant during both periods: after a filtration run of 15 minutes a backflush of 17 m^3/h during 45 seconds. In order to remove the fouling chemically the membranes were soaked for 2 hours in a Divos 120Cl solution (1.25 %-w/w). Mostly the chemical cleaning (CC) interval was 12 hours but only once (16-07-08 – 18-07-08) it was changed to 8 hours. Table 7.3 gives a summary of the operational parameters of the ultrafiltration pilot installation during both periods.

Table 7.3 – Operational parameters of the ultrafiltration pilot installation at the WWTP Horstermeer during the periods August, 2007 and May – July, 2008

Period	Flow (m^3/h)	Average actual T (°C)	Flux ($L/m^2 \cdot h$)	BF interval (h)	CC interval (h)	CC duration (h)
Multi media filter						
02-08-07 – 05-08-07	2	21.4	29	0.25	12	2
06-08-07 – 12-08-07	4	21.5	57	0.25	12	2
13-08-07 – 16-08-07	3	21.5	43	0.25	12	2
1-STEP® filter						
19-05-08 – 26-05-08	3	18.1	43	0.25	12	2
17-06-08 – 19-06-08	4	20.2	57	0.25	12	2
16-07-08 – 18-07-08	6	20.9	86	0.25	8	2

The logged process data of the ultrafiltration pilot installation were evaluated for the filterability and the reversibility. The filterability is determined by dR/dt, which is the fouling rate within a filtration run (period between two backflushes). During both periods the (average) filterability was measured as the mean filterability of all filtration runs between two chemical cleanings. The reversibility is calculated by the filtration resistance over time, $\Delta R/\Delta t$, comparing the filtration resistance at the start of a filtration period and at the end of the filtration period. The measurements of the filterability and reversibility are illustrated in Figure 3.5 of section 3.2.1.

Next to the operation of the ultrafiltration pilot installation also the SUR values of the feedwater were measured. Samples during the period of August, 2007 were taken after the filtrate buffertank of the multi media filter. The samples of the period of May – July, 2008 (1-STEP® filter) were taken directly after the 1-STEP® filter in front of the filtrate buffertank.

7.2.2 Multi media filter and ultrafiltration pilot installation

7.2.2.1 Results

In the Figures 7.3, 7.4 and 7.5 the filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate are presented. During the first experimental period (02-08-07 – 05-08-07) the ultrafiltration pilot installation was operated at a constant flux of 29 L/m^2·h and the filtration resistance at the start was about $0.7 \cdot 10^{12}$ m^{-1}. Taking into account the membrane resistance of a clean membrane ($0.8 \cdot 10^{12}$ m^{-1}) it indicates the membranes were clean at the start of the experimental period. During the successive filtration periods the filtration resistance increased till a maximum of approximately $1.2 \cdot 10^{12}$ m^{-1} but after chemical cleaning the filtration resistance returned to the initial level of about $0.7 \cdot 10^{12}$ m^{-1}. This means the chemical cleanings were sufficient enough to restore the filtration resistance after a filtration period.

During the period represented in Figure 7.3 the SUR values of the feedwater of the ultrafiltration pilot installation were measured two times. On 03-08-07 and 05-08-07 these values were respectively $6.6 \cdot 10^{12}$ m^{-2} and $9.4 \cdot 10^{12}$ m^{-2}. These values in combination with the relative low flux (29 L/m^2·h) evidently resulted in a stable process performance.

During the whole period the average dR/dt and ΔR/Δt values were respectively $0.27 \cdot 10^{12}$ 1/m·h and $0.03 \cdot 10^{12}$ 1/m·h. In Table 7.4 these values are given for each separate filtration period of Figure 7.3. The ΔR/Δt values are more or less steady in contrast to the dR/dt values. The latter values might be related to the filterability of the feedwater. As described in the previous paragraph the SUR values were $6.6 \cdot 10^{12}$ m^{-2} and $9.4 \cdot 10^{12}$ m^{-2} during respectively the first and fourth filtration period.

Table 7.4 – Filtration properties of multi media filtrate at the WWTP Horstermeer during the period 02-08-07 – 05-08-07

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.25	0.03
2	0.17	0.03
3	0.22	0.03
4	0.34	0.02
5	0.40	0.04
Average	0.27	0.03

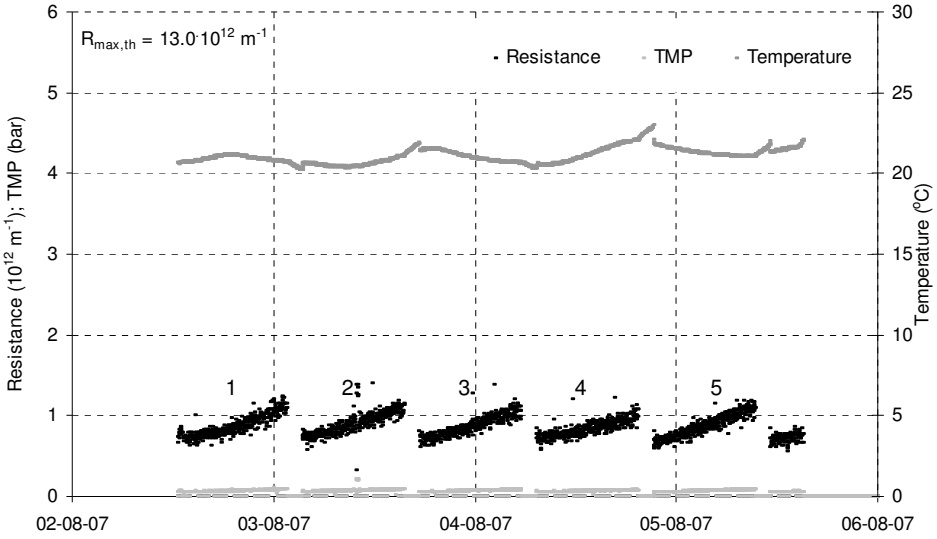


Figure 7.3 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer in the period of 02-08-07 – 05-08-07 at a flux of 29 L/m^2·h

During the period of 06-08-07 – 11-08-07 (Figure 7.4) the ultrafiltration pilot installation was operated at a flux of 57 L/m^2·h. The initial filtration resistance of the membranes at the start of the experimental period (06-08-07) was about 0.9·10^{12} m^{-1}. This means the membranes were fouled to a very small extent at the start of the experimental period. After the start up, the (initial) filtration resistance of a filtration period increased repeatedly after each chemical cleaning. This effect was especially noticed during the last filtration periods (5 to 8). It means the chemical cleanings were not sufficient enough in spite of the relative long soaking time (2 hours). Furthermore, these inadequate chemical cleanings seemed to influence the filterability (dR/dt) and reversibility (ΔR/Δt) values as shown in Table 7.5.

Table 7.5 – Filtration properties of multi media filtrate at the WWTP Horstermeer during the period 06-08-07 – 11-08-07

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.63	0.11
2	0.52	0.06
3	0.51	0.12
4	0.69	0.14
5	1.01	0.15
6	1.13	0.25
7	1.00	0.25
8	1.07	0.31
Average	0.82	0.17

The dR/dt and ΔR/Δt values show an increasing trend during the whole period and especially during the last filtration periods (5 to 8) a sharp increase can be observed. Therefore this effect might be related to the efficiency of the chemical cleanings because the different dR/dt and ΔR/Δt values cannot be directly related to the SUR values of the feedwater. During the experimental period the SUR values of the multi media filtrate had been measured three times as presented in Table 7.6. This table shows a small increase of the SUR values during the period but this variation cannot be directly related to the dR/dt and ΔR/Δt values. Therefore it seemed that other aspects like the efficiency of the chemical cleaning played an important role.

Table 7.6 – The SUR values of the feedwater of the ultrafiltration pilot installation at the WWTP Horstermeer during the period 06-08-07 – 11-08-07

Date	SUR (10^{12} m^{-2})
07-08-07	6.0
09-08-07	7.7
10-08-07	8.3

As discussed, an aspect that might have influenced the values of the dR/dt and ΔR/Δt measurements are the insufficient chemical cleanings. Due to lacking chemical cleaning, fouling remained on the membranes of the ultrafiltration pilot installation resulting in a higher initial filtration resistance value at the start of a subsequent filtration period (see Figure 7.4). Consequently, Figure 7.4 shows that a higher initial filtration resistance value resulted in higher operational trans membrane pressure values at the start of a filtration period. Considering this observation and the fact of a highly compressible cake layer of WWTP effluent during dead-end ultrafiltration (Roorda, 2004) it seemed that inadequate chemical cleaning leads to a compression of the cake layer. In Figure 7.4 an exponential increase of the trans membrane pressure and filtration resistance can be noticed during the last filtration periods. These kind of graph is typical for cake compression presented by Roorda (2004) as well. Therefore it seemed sufficient chemical cleaning is a precondition to compare accurately the dR/dt and ΔR/Δt values.

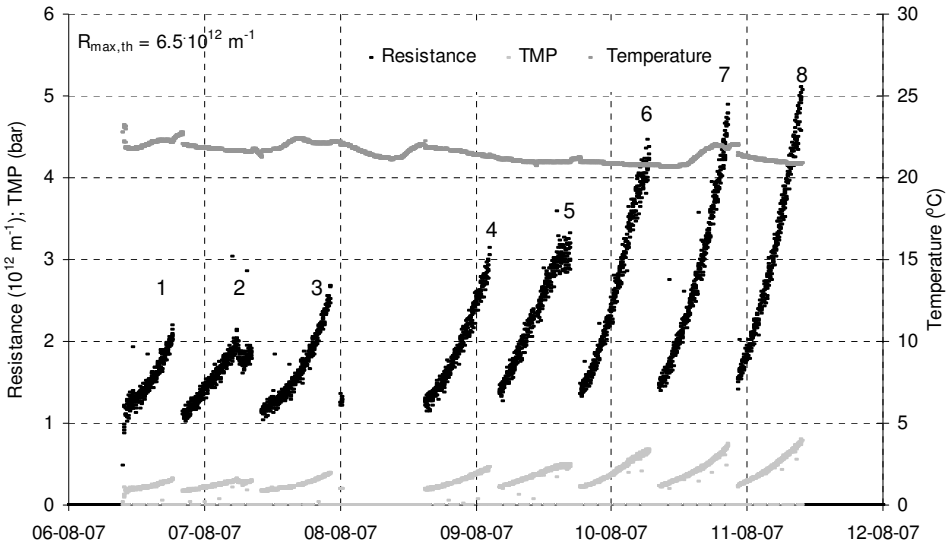


Figure 7.4 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer in the period of 06-08-07 – 11-08-07 at a flux of 57 L/m^2·h

After the period with an operational flux of 57 L/m^2·h, the flux was decreased till 43 L/m^2·h during the period 13-08-07 – 15-08-07. The obtained data of the ultrafiltration pilot installation are presented in Figure 7.5. As shown the initial filtration resistance at the start of the period was around 1.5·10^{12} m^{-1} which means the membranes were fouled. But in contrast to the previous experimental period after a chemical cleaning the filtration resistance came back to the initial filtration resistance level (1.5·10^{12} m^{-1}). Therefore it seemed the chemical cleaning was sufficient enough resulting in a restoration of the initial filtration resistance level. However the chemical cleanings did not suffice to obtain completely clean membranes. As described earlier the filtration resistance of clean membranes is about 0.8·10^{12} m^{-1}. Therefore a more intensive chemical cleaning was preferred at the start of filtration period in order to obtain a good comparison with the dR/dt and ΔR/Δt values of other periods.

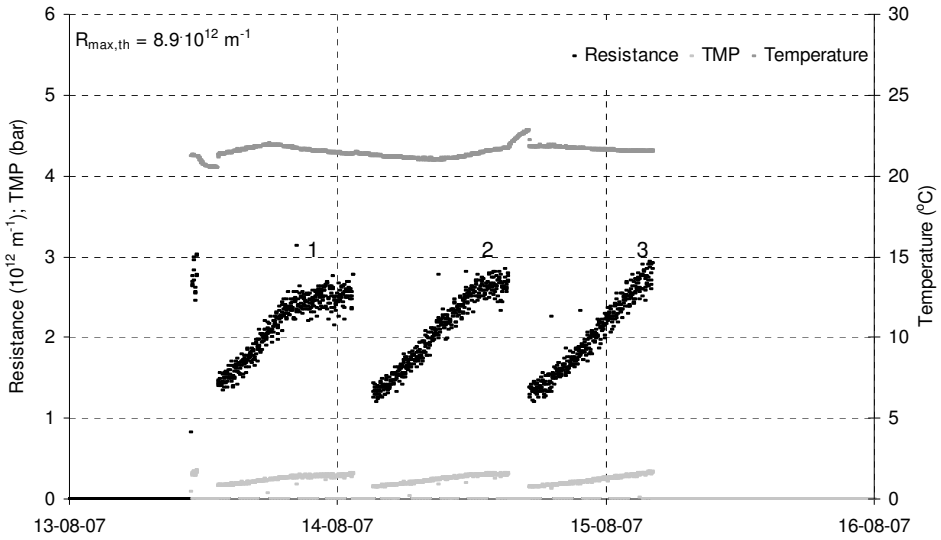


Figure 7.5 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of multi media filtrate at the WWTP Horstermeer in the period of 13/08/07 – 15/08/07 at a flux of 43 L/m^2·h

In Table 7.7 the dR/dt and ΔR/Δt values of the period are summarized. Compared to the previous periods the values are quite stable. However this was not the situation for the SUR values. During the period the SUR values of the ultrafiltration feedwater had been measured on 13-08-07 and 14-08-07. The SUR values were respectively $18.3 \cdot 10^{12}$ m^{-2} and $9.0 \cdot 10^{12}$ m^{-2}. These values did not relate with the dR/dt and ΔR/Δt values of Table 7.7. Therefore it seemed again that other aspects played a role as well.

Table 7.7 – Filtration properties of multi media filtrate at the WWTP Horstermeer during the period 13-08-07 – 15-08-07

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.79	0.09
2	0.74	0.12
3	0.83	0.13
Average	0.79	0.11

7.2.2.2 Discussion

In Table 7.8 the filtration properties of the multi media filtrate during pilot ultrafiltration are summarized.

Table 7.8 – Filtration properties of multi media filtrate during pilot ultrafiltration at the WWTP Horstermeer during the period August, 2007

Periods	Flux ($L/m^2 \cdot h$)	SUR (10^{12} m^{-2})	dR/dt (10^{12} $1/m \cdot h$)	$\Delta R/\Delta t$ (10^{12} $1/m \cdot h$)
02-08-07 – 05-08-07	29	6.6 and 8.4	0.27 (± 0.09)	0.03 (± 0.01)
06-08-07 – 12-08-07	57	6.0, 7.7 and 8.3	0.82 (± 0.26)	0.17 (± 0.09)
13-08-07 – 16-08-07	43	18.3 and 9.0	0.79 (± 0.04)	0.11 (± 0.02)

As shown in Table 7.8 the dR/dt and $\Delta R/\Delta t$ values of the multi media filtrate were the lowest at a flux of 29 $L/m^2 \cdot h$ and became generally higher, i.e. decrease of both filterability and reversibility, when the flux was increased. This observation might be explained by the larger amount of foulants that passed along the membrane surface and therefore an increase of foulant adsorption when the flux was increased. However this interpretation does not explain the almost similar dR/dt values at a flux of 43 $L/m^2 \cdot h$ and 57 $L/m^2 \cdot h$. These results might be related to the SUR values of ultrafiltration feedwater. The SUR values of the multi media filtrate at a flux of 43 $L/m^2 \cdot h$ were higher compared to the period with an operational flux of 57 $L/m^2 \cdot h$. From 13-08-07 – 16-08-07 the measured SUR values were 18.3 and $9.0 \cdot 10^{12}$ m^{-2}. During the other period (06-08-07 – 12-08-07) the average SUR value was $7.3 \cdot 10^{12}$ m^{-2}. Of course this explanation only explicates partly the difference but emphasizes the added value of SUR measurements parallel to the operation of the ultrafiltration pilot installation in order to gain a better understanding of the process performance in relation to the filtration properties of ultrafiltration feedwater.

Next to the effect of the flux on the filterability also an effect on the reversibility was noticed. As shown in Table 7.8 an increase of the flux resulted in higher $\Delta R/\Delta t$ values i.e. decrease of the reversibility. As discussed above it is evident that a higher flux will lead to an increase of the $\Delta R/\Delta t$ value. But this is not the only relation. As shown in the Figures 7.3 – 7.5 the reversibility seems also dependent on the efficiency of the chemical cleaning and the condition of the membranes at the start of the filtration periods. Both, insufficient chemical cleaning and/or fouled membranes may result in higher $\Delta R/\Delta t$ and dR/dt values. For instance during the period 06-08-07 – 12-08-07 (Figure 7.4) the flux was doubled in comparison to the period of 02-08-07 – 05-08-07 (Figure 7.3). But in spite of the almost similar feedwater quality (SUR values) the dR/dt and $\Delta R/\Delta t$ values increased respectively 3 and 6 times. Considering the doubled flux an increase of the filtration resistance of approximately 2 times was expected. Therefore other aspects beside the flux like the condition of the membranes and the efficiency of chemical cleanings seem to influence the dR/dt and $\Delta R/\Delta t$ value as well and have to be taken into account when theses values are evaluated.

7.2.3 1-STEP® filter and ultrafiltration pilot installation

7.2.3.1 Results

In the following figures (Figure 7.6, 7.7 and 7.8) the filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of 1-STEP® filtrate are presented. During the period 19-05-08 – 26-05-08 the ultrafiltration pilot installation was operated at a constant flux of 43 L/m^2·h. At the start the initial filtration resistance was around $1.0 \cdot 10^{12}$ m^{-1} but increased till $1.6 – 2.0 \cdot 10^{12}$ m^{-1} within a filtration period. But after a chemical cleaning the filtration resistance came back to initial filtration resistance of $\pm 1.0 \cdot 10^{12}$ m^{-1} which means the chemical cleanings were sufficient. Nevertheless the chemical cleanings were not sufficient enough to obtain clean membranes i.e. a initial filtration resistance of about $0.8 \cdot 10^{12}$ m^{-1}.

Furthermore during the period the SUR values of the ultrafiltration feedwater were about $4 \cdot 10^{12}$ m^{-2}. On 22-05-08 and 26/05-08 the SUR values were respectively $4.3 \cdot 10^{12}$ m^{-2} and $4.0 \cdot 10^{12}$ m^{-2}. This means the filterability was very good resulting in a steady ultrafiltration performance as shown in Figure 7.6.

During the period also the dR/dt and ΔR/Δt values were measured. Despite of the steady ultrafiltration performance the dR/dt values varied in the range of $0.2 – 0.5 \cdot 10^{12}$ 1/m·h with an average value of $0.33 \cdot 10^{12}$ 1/m·h. A clear explanation for this variation cannot be given because the SUR values of the feedwater seemed to be relatively constant. Therefore it seemed this variation is not extraordinary and is just a result of the ultrafiltration process operation. The average ΔR/Δt value during the whole period was $0.04 \cdot 10^{12}$ 1/m·h which is quite low if the $\Delta R_{max,th}/t_{24h}$ value of $0.30 \cdot 10^{12}$ 1/m·h is considered. As a consequence of this low value a steady process performance was obtained. All the retained fouling could be removed either hydraulically or chemically.

Table 7.9 – Filtration properties of 1-STEP® filtrate at the WWTP Horstermeer during the period 19-05-08 – 26-05-08

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.27	0.05
2	0.47	0.07
3	0.41	0.06
4	0.47	0.05
5	-	-
6	0.23	0.04
7	0.20	0.04
8	0.26	0.04
9	0.32	0.02
10	0.20	0.03
11	0.31	0.02
12	0.44	0.03
Average	0.33	0.04

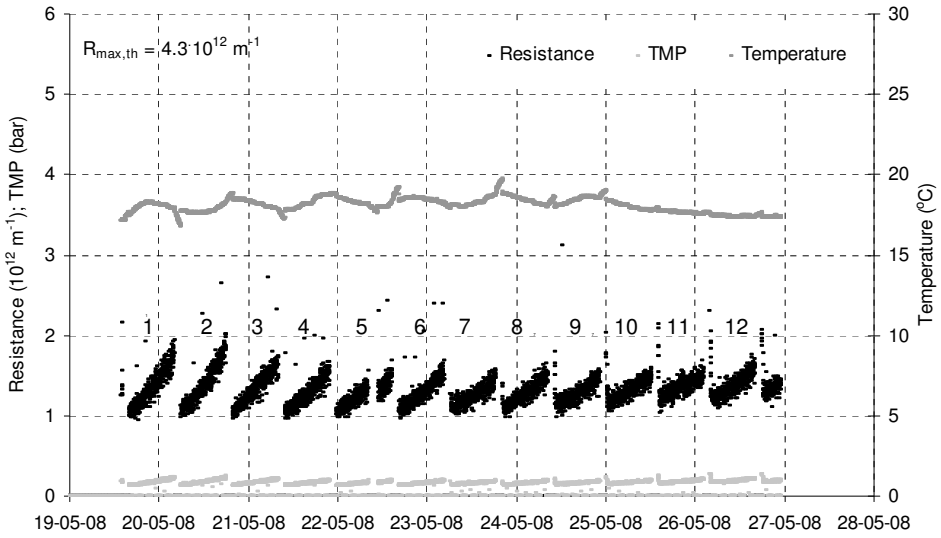


Figure 7.6 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of 1-STEP® filtrate at the WWTP Horstermeer in the period 19-05-08 – 26-05-08 at a flux of 43 L/m^2·h

Figure 7.7 represents the results of the period from 17-06-08 to 19-06-08. Unfortunately the ultrafiltration membranes were already fouled to a large extent at the start of the period. The initial filtration resistance at the start was ± 2.0·10^{12} m^{-1} where normally clean membranes should present a value of ± 0.8·10^{12} m^{-1}. Nevertheless the applied chemical cleanings revealed to be sufficient enough, since the filtration resistance came back to ± 2.0·10^{12} m^{-1} after the successive chemical cleanings. These combined with a very low SUR value (3.2·10^{12} m^{-2}) of the ultrafiltration feedwater on 18-06-08 resulted in average dR/dt and ΔR/Δt values of respectively 0.62·10^{12} 1/m·h and 0.09·10^{12} 1/m·h. In Table 7.10 the dR/dt and ΔR/Δt values of the whole period are summarized.

Table 7.10 – Filtration properties of 1-STEP® filtrate at the WWTP Horstermeer during the period 17-06-08 – 19-06-08

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.67	0.10
2	0.57	0.08
3	0.63	0.09
4	0.61	0.09
Average	0.62	0.09

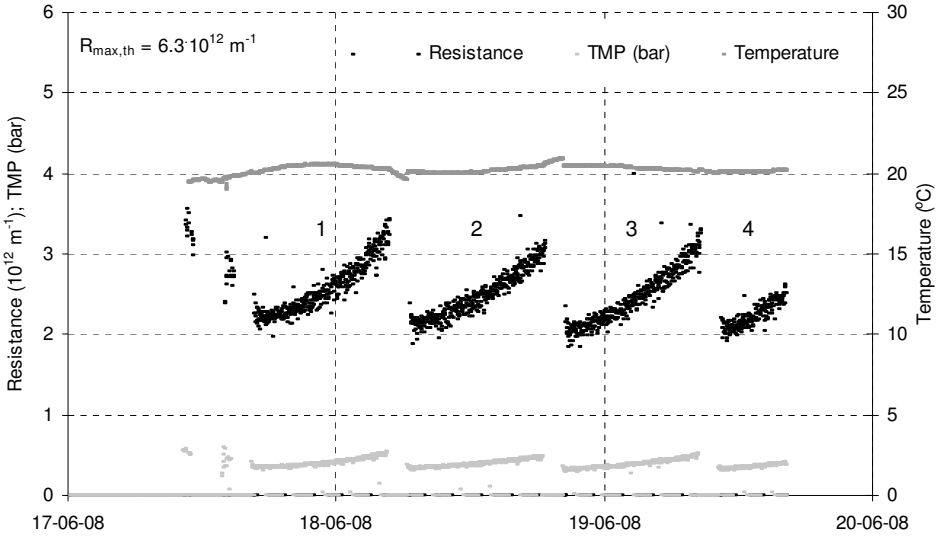


Figure 7.7 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of 1-STEP® filtrate at the WWTP Horstermeer during the period 17-06-08 – 19-06-08 at a flux of 57 L/m^2·h

During the last period (16-07-08 – 17-07-08) the flux was increased till 86 L/m^2·h and the chemical cleaning interval was decreased till 8 hours (Figure 7.8). Like the previous period represented in Figure 7.7 the membranes were fouled to a large extent (filtration resistance ± 2.0·10^{12} m^{-1}) at the start of the period. But in contrast to the preceding period the chemical cleanings were not sufficient enough to remove the retained fouling. The filtration resistance at the start of each filtration period increased slightly. It seemed these insufficient chemical cleanings influenced the dR/dt and ΔR/Δt values of the multi media filtrate. During the first filtration period these values were lower than during the subsequent filtration periods. This is shown in Table 7.11.

Table 7.11 – Filtration properties of 1-STEP® filtrate at the WWTP Horstermeer during the period 16-07-08 – 18-07-08

Filtration period	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)
1	0.70	0.16
2	0.87	0.18
3	0.85	0.19
Average	0.81	0.17

During the period no SUR values of the ultrafiltration feedwater had been measured. Therefore the influence of the feedwater quality on the dR/dt and ΔR/Δt values cannot be determined. However like the previous periods (Figure 7.6 and 7.7) low SUR values of about 5.0·10^{12} m^{-2} can be expected because the pretreatment step and settings were similar.

Therefore the results of Table 7.11 subscribe again the need for sufficient chemical cleaning to obtain steady process performance. As shown the dR/dt and ΔR/Δt values increased due to insufficient cleaning and consequently this will result in unsteady process performance of the ultrafiltration pilot installation after a certain time.

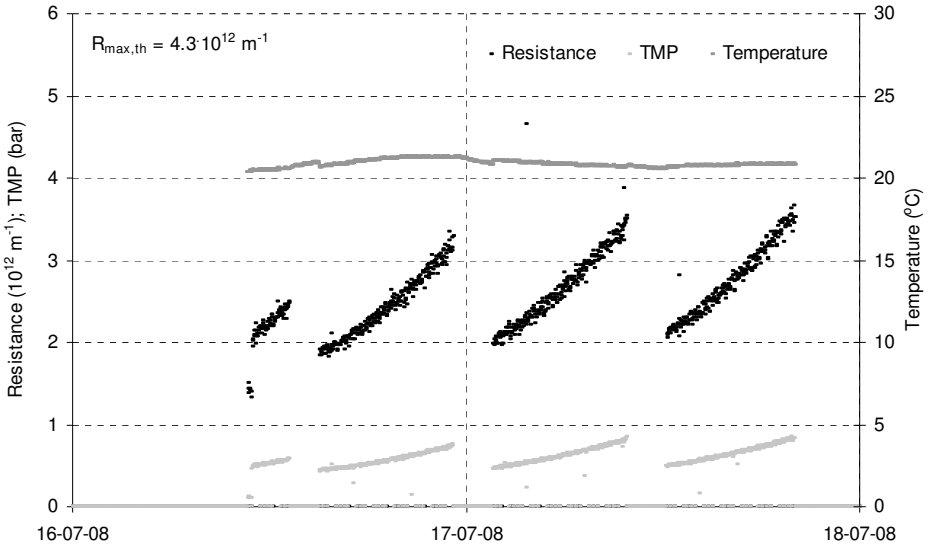


Figure 7.8 – The filtration resistance, trans membrane pressure and actual temperature during pilot ultrafiltration of 1-STEP® filtrate at the WWTP Horstermeer during the period 16-07-08 – 18-07-08 at a flux of 86 L/m^2·h

7.2.3.2 Discussion

In Table 7.12 the filtration properties of 1-STEP® filtrate during pilot ultrafiltration are summarized.

Table 7.12 – Filtration properties of 1-STEP® filtrate during pilot ultrafiltration at the WWTP Horstermeer during the period May – July, 2008

Periods	Flux (L/m^2·h)	dR/dt (10^{12} 1/m·h)	ΔR/Δt (10^{12} 1/m·h)	SUR (10^{12} m^{-2})
19-05-08 – 26-05-08	43	0.33 (± 0.04)	0.04 (± 0.02)	4.3 and 4.0
19-06-08 – 29-06-08	57	0.62 (± 0.04)	0.09 (± 0.01)	3.2
16-07-08 – 17-07-08	86	0.81 (± 0.09)	0.17 (± 0.04)	-

Like the results of Section 7.2.2 a relation between the flux and both filterability and reversibility can be observed from Table 7.8. The lowest dR/dt and ΔR/Δt values were obtained at an operational flux of 43 L/m^2·h. Further increase of the flux resulted at all times in higher dR/dt and ΔR/Δt values. However this was not a direct relation i.e. two times increase of the flux did not directly result in double dR/dt and ΔR/Δt values. But as previously

discussed (Section 7.2.2) next to the flux, other aspects like the trans membrane pressure, initial membrane resistance, cake compression, cleaning efficiency, etc. influence the dR/dt and $\Delta R/\Delta t$ values as well. Because of this complication it is difficult to directly relate the filterability and reversibility values of the pilot experiments. However in spite of this complexity at least a reasonable relation between the dR/dt and $\Delta R/\Delta t$ can be observed. However to have a better insight into this relation more well defined experiments on lab-scale have to be performed. In this way some aspects that influence the comparison can be eliminated.

Summarizing the findings of this section and considering the relation between flux and the dR/dt and $\Delta R/\Delta t$ values it seemed that most stable process performance can be obtained when the membranes are clean (low initial filtration resistance), ultrafiltration feedwater very well filterable (low SUR value) and an operational flux below approximately 60 $L/m^2 \cdot h$.

7.2.4 Evaluation

In Figure 7.9 the effect of the flux on both the dR/dt and $\Delta R/\Delta t$ values is presented. As expected both values became higher with increasing flux. During the pilot experiments with 1-STEP® filtrate this relation was more linear than during the experiments with multi media filtrate. Therefore this figure confirms the fact that filterability and reversibility are influenced by the operational flux. But at the pilot this relation was influenced by different circumstances. It seemed that the efficiency of chemical cleaning, initial condition of the membranes, varying water, occurring cake compression, etc. played an important role in the interpretation of the ultrafiltration pilot data. Therefore the influence of these aspects have to be taken into account when the reversibility and filterability of ultrafiltration pilot data are evaluated.

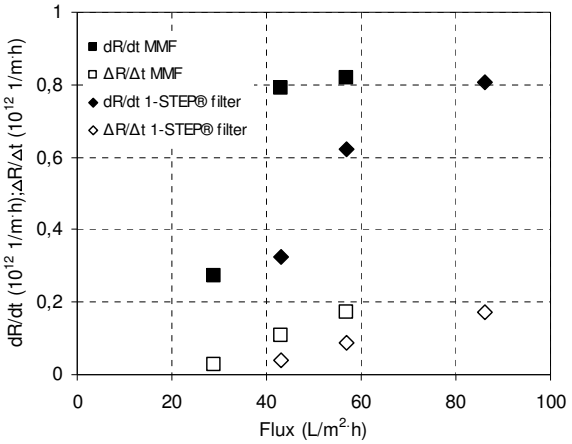


Figure 7.9 – Filtration properties (dR/dt and ΔR/Δt) at different flux rates during pilot ultrafiltration of multi media and 1-STEP® filtrate at the WWTP Horstermeer

In addition to the effect of the flux, Figure 7.9 presents the effect of pretreatment on the dR/dt and ΔR/Δt values of the feedwater of the ultrafiltration pilot installation. At the same operating flux (43 L/m^2·h and 57 L/m^2·h) the dR/dt and ΔR/Δt values of the 1-STEP® filtrate are significantly lower than the multi media filter. This observed difference was confirmed by the SUR measurements of the 1-STEP® and multi media filtrate. Generally, the SUR values of multi media filtrate were above $7.0 \cdot 10^{12}$ m^{-2} while the SUR values of 1-STEP® filtrate were found in the range of $3 \cdot 10^{12}$ m^{-2} – $4 \cdot 10^{12}$ m^{-2}. Considering these SUR values and the results presented in chapter 5 the different dR/dt values are evident. But additionally to the dR/dt values also the ΔR/Δt values differed. The ΔR/Δt values of the 1-STEP® filtrate are significantly lower than the multi media filtrate. This means that 1-STEP® filtrate is more hydraulically reversible than the filtrate of the dual media filter. This observation might be related to the results presented in chapter 5. In this chapter it was noticed that 1-STEP® filtration in contrast to multi media filtration significantly decreases the concentration of proteins of WWTP effluent. Proteins as part of the soluble microbial products are broadly considered as a compound responsible for long term (irreversible) fouling (te Poele, 2005; Amy, 2008; Haberkamp, 2008). Therefore the lower ΔR/Δt values of the 1-STEP® filtrate during pilot ultrafiltration might be related to the removal of proteins in the 1-STEP® filter.

In Figure 7.10 all dR/dt and ΔR/Δt values of the Figures 7.3 – 7.8 are plotted in one figure. As shown, this compilation of dR/dt and ΔR/Δt values resulted in a reasonable relation between filterability and reversibility. This relationship was also suggested by other researchers (Roorda, 2004; te Poele, 2005; Lozier *et al*., 2008) and so confirmed by this figure. Of course the relationship is disturbed by different factors during the pilot scale experiment (pretreatment, initial membrane resistance, different fluxes, recovery, fluctuating feedwater quality, etc.) and therefore additional experiments are performed to prove this relation. These

experiments are performed under well defined conditions and presented in the next section of this chapter.

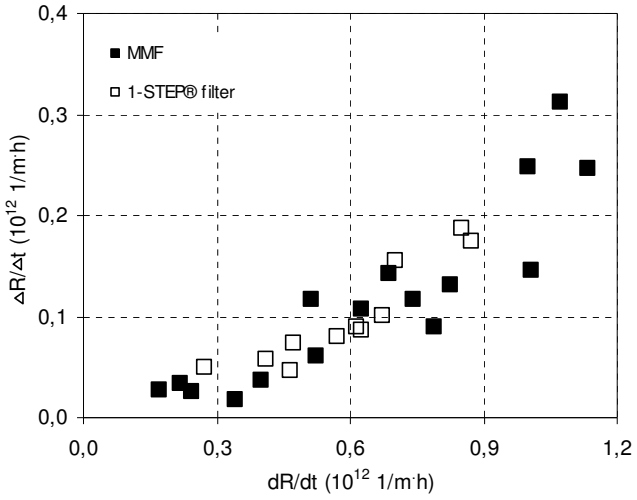


Figure 7.10 – Filtration properties (dR/dt and ΔR/Δt) of multi media and 1-STEP® filtrate during pilot ultrafiltration at the WWTP Horstermeer

7.3 Filterability and reversibility during lab scale experiments

7.3.1 General

As emphasized in the previous section there was a need to measure the filterability and reversibility of ultrafiltration feedwater under well defined conditions in order to evaluate a possible relation between both parameters. Therefore the SUR equipment as described in section 3.3 was rebuild to mimic full and pilot scale ultrafiltration operation including the backflush in order to measure both filterability and reversibility of WWTP effluent. The main differences between the new (SUR+) and previous SUR equipment are the position of the membrane module and the way of measuring the production of permeate (see Figure 7.11). The SUR+ equipment consists of a vertical placed membrane module instead of the horizontal one of the former equipment. By using a vertical placed membrane module it is easier to get rid of air at the permeate side of membranes inside the module. This is important to perform an accurate backflush. Furthermore the permeate is collected in a pressurized buffer tank instead of an erlenmeyer and the produced permeate is measured with a mass flow meter instead of a mass balance. Collection of permeate in an erlenmeyer made it impossible to apply a backflush with the permeate itself. For this reason this change of the configuration was made.

Figure 7.11 - Pictures of the SUR equipment (left) and the SUR+ equipment (right)

In this section the SUR+ equipment and results are presented. Starting point of the SUR+ equipment was that it has to operate at constant pressure and be able to measure the SUR value and reversibility ($\Delta R/\Delta t$) of WWTP effluent during a single test.

7.3.2 New equipment and operation

In Figure 7.12 a schematic picture of the SUR+ equipment is given. The pressurized buffer tank B (volume = 10 litre) is filled with demineralised water to measure the clean water flux at the start of the test. Pressurized buffer tank C (volume = 10 litre) is filled with the sample and stirred to be well mixed. In pressurized buffer tank M (volume = 10 litre) the permeate during filtration is collected. To keep the buffer tanks under constant pressure two pressure devices (A1 and A2) were installed. Pressure device A1 keeps the pressure of the buffer tanks B and C at 1.5 bar. Consequently pressure device A2 keeps the pressure of buffer tank M at 1.0 bar in order to have a trans membrane pressure of 0.5 bar over the membrane module (G).

The pressurized buffer tanks B and C are connected to the membrane module by means of a three-way valve (E) in between. This valve makes it possible to switch easily between buffertank B or C. Between the permeate side and feed side of the membrane module (G) a digital pressure difference device is placed to check and log the trans membrane pressure over the membrane module. The device is hooked up to the computer (K). The permeate flow is continuously measured by a mass flow meter (F).

The produced permeate can be collected or wasted. To collect the permeate in the buffertank during dead-end operation valves H and L have to be closed and valve N open. To apply a backflush valves F and L have to be closed and valve H open. These actions will results in a constant backflush pressure of 1.0 bar.

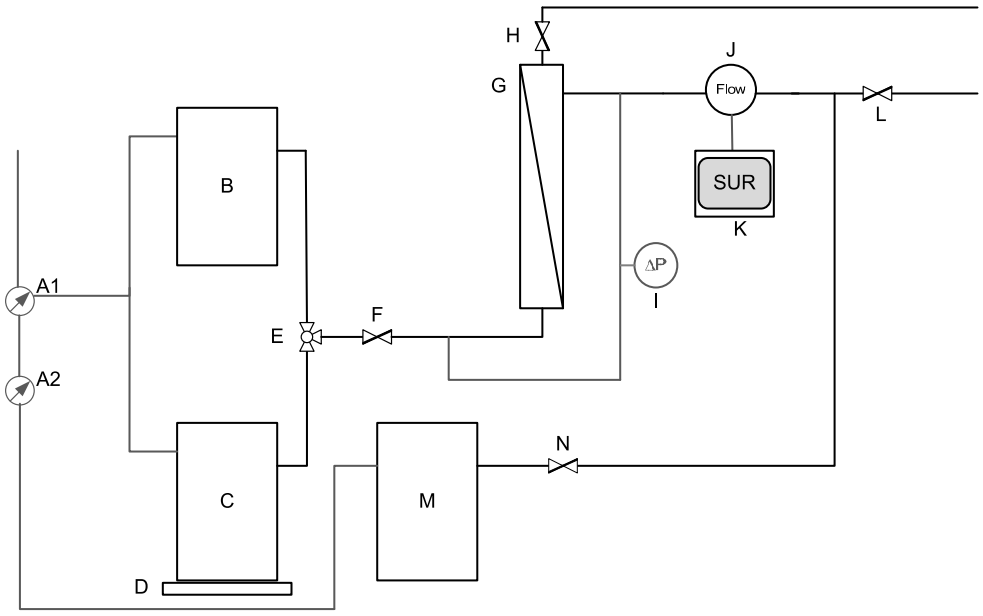


Figure 7.12 – Schematic picture of the SUR+ equipment containing; A1 and A2 pressure devices, B demiwater buffer tank (1.5 bar), C sample buffer tank (1.5 bar), D magnetic stirrer, E three-way valve, F and H valves to change the operation mode of the membrane, G membrane module, I digital pressure sensor, J flow meter, K computer with measuring program, L and N valves to change the direction of the permeate and M permeate vessel (1.0 bar)

7.3.3 Measuring protocol

Before the start of the test the buffer tanks B and C are filled with respectively demineralised water and the sample. During the tests the demineralised water and the WWTP effluent sample were always kept at 20 °C. The measuring protocol of the test consists of the following steps:

- **Measuring clean water flux.** This is required to assess the clean water permeability of the membranes in the module at 20 °C. The permeability of demineralised water as measured on a single membrane capillary is approximately 600 – 700 L/m^2·h·bar at 20 °C.
- **Filtrating wastewater effluent.** The WWTP effluent sample is filtrated under standard conditions (trans membrane pressure = 0.5 bar and temperature = 20 °C) during a period of 30 minutes.
- **Backflushing.** The membrane module is back flushed with permeate during 30 seconds at a pressure of 1 bar.
- **Filtrating WWTP effluent.** The WWTP effluent sample is filtrated under standard conditions (trans membrane pressure = 0.5 bar and temperature = 20 °C) during a period of 10 minutes.

- **Backflush.** The membrane module is back flushed with permeate during 30 seconds at a pressure of 1 bar.
- **Filtrating WWTP effluent**. The WWTP effluent sample is filtrated under standard conditions (trans membrane pressure = 0.5 bar and temperature = 20 °C) during a period of 10 minutes.
- **Backflush.** The membrane module is back flushed with permeate during 30 seconds at a pressure of 1 bar.
- **Filtrating WWTP effluent.** The WWTP effluent sample is filtrated under standard conditions (trans membrane pressure = 0.5 bar and temperature = 20 °C) during a period of 10 minutes.
- **Backflush.** The membrane module is back flushed with permeate during 30 seconds at a pressure of 1 bar.
- **Filtrating WWTP effluent.** The WWTP effluent sample is filtrated under standard conditions (trans membrane pressure = 0.5 bar and temperature = 20 °C) during 1 to 2 minutes.
- **Chemical cleaning.** A chemical cleaning is performed with 1.25% Divos 120 Cl at 40 °C until the membrane is clean. The criteria of a clean membrane are defined in the first step of this measuring protocol.

7.3.4 Results of new SUR equipment

7.3.4.1 General

The first experiments of the SUR+ equipment focused only on the SUR measurement except for the reversibility test. The purpose was to check the accuracy and reproducibility of the SUR+ equipment. Effluent was taken from the WWTP Berkel and WWTP Horstermeer. After sampling, the WWTP effluent samples were transported to the laboratory of Sanitary Engineering of Delft University of Technology. Before the SUR measurements the samples were sieved (400 µm) and measurements were performed at 20 °C.

During the first experiment the SUR value of WWTP effluent was measured at the same time with the SUR+ and SUR equipment. After measuring the obtained values of both equipments were compared. During the second experiment the SUR of one sample was measured two or three times with the SUR+ equipment in order to check the reproducibility of this measurement.

7.3.4.2 Comparison SUR value of SUR and SUR+ equipment

In Figure 7.13 the SUR values of WWTP effluent measured with the SUR and SUR+ equipment are compared. The measured SUR values were found in the range of $4 – 18 \cdot 10^{12}$ m^{-2}. Unfortunately this range did not cover the whole range of SUR values found at other WWTPs in the Netherlands. Roorda (2004), reported SUR values in the range of $5 – 30 \cdot 10^{12}$ m^{-2} at different WWTPs in the Netherlands. Nevertheless, Figure 7.13 presents a good

relation between the SUR values of the SUR equipment with the mass balance (old set up) and the mass flow meter (SUR+ equipment). Therefore it was concluded that the modifications did not significantly influence the absolute SUR values of WWTP effluent.

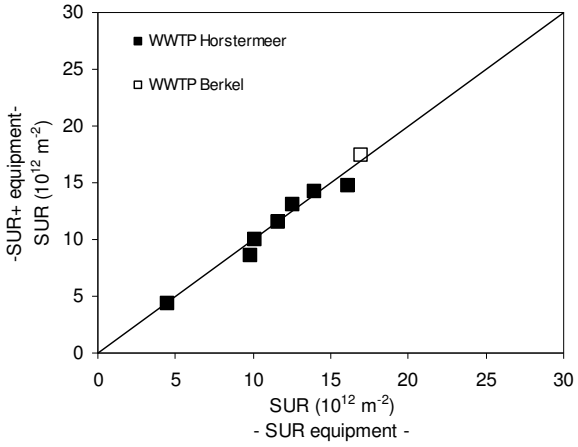


Figure 7.13 – Comparison of the SUR values of effluent WWTP Horstermeer and WWTP Berkel measured with the SUR and SUR+ equipment

7.3.4.3 Reproducibility SUR value new set up

The reproducibility of the SUR measurements of the SUR+ equipment was tested. Therefore the SUR value of one samples was measured several times. The average SUR value of these measurements was calculated as well as the relative deviation from the first measurement:

$$\frac{SUR_n}{SUR_1} \tag{7.1}$$

in which:
n = 2, 3 or 4

The results are summarized in Table 7.13.

Table 7.13 – Results of the reproducibility measurements of the SUR+ equipment

WWTP	Measurement	SUR (10^{12} m^{-2})	Average SUR (10^{12} m^{-2})	SUR_n/SUR_1 (-)
Horstermeer	1	13.6	13.6	
	2	14.1		1.04
	3	13.1		0.96
Berkel	1	17.2	17.0	
	2	16.8		0.98
Horstermeer	1	13.2	12.9	
	2	12.9		0.98
	3	12.7		0.96
Horstermeer	1	9.2	9.3	
	2	9.1		0.99
	3	9.6		1.04
	4	9.2		1.00
Horstermeer	1	15.6	15.2	
	2	14.7		0.94
Horstermeer	1	4.3	4.4	
	2	4.2		0.98
	3	4.7		1.09

In Table 7.13 the first column shows the origin of the WWTP effluent sample, the second one the number of the measurement and the last two columns show the average SUR value and the ratio between the n^{th} measurement and the first. Between the measurements the membrane modules were chemically cleaned to have them pristine at the start of the next measurement cycle.

Generally, Table 7.13 shows a good reproducibility of the SUR measurements of the SUR+ equipment. The relative deviation varied between 0.94 – 1.09 and this deviation seemed to be independent on the average SUR values. The range of SUR values was rather broad (4 – $17 \cdot 10^{12}$ m^{-2}) but the relative deviation values were comparable. This indicates the SUR+ equipment is capable to measure accurately the SUR values of WWTP effluent in a fairly broad range.

7.3.5 Reversibility test

7.3.5.1 General

In Figure 7.14 a schematic presentation of the complete test is given. Firstly, the WWTP effluent sample is filtrated in accordance with the normal SUR procedure (trans membrane pressure = 0.5 bar and temperature of 20 °C). After 30 minutes the membrane module is back flushed during 30 seconds at a pressure of 1 bar (see measuring protocol described in section 7.3.3). After this the WWTP effluent sample is filtrated again during 10 minutes at a trans membrane pressure of 0.5 bar and afterwards again a backflush is applied. This procedure is repeated 3 times after the SUR measurement. Therefore the complete test takes approximately

60 minutes where during the first 30 minutes the filterability (SUR and dR/dt) is determined and during the last 30 minutes the reversibility (ΔR/Δt).

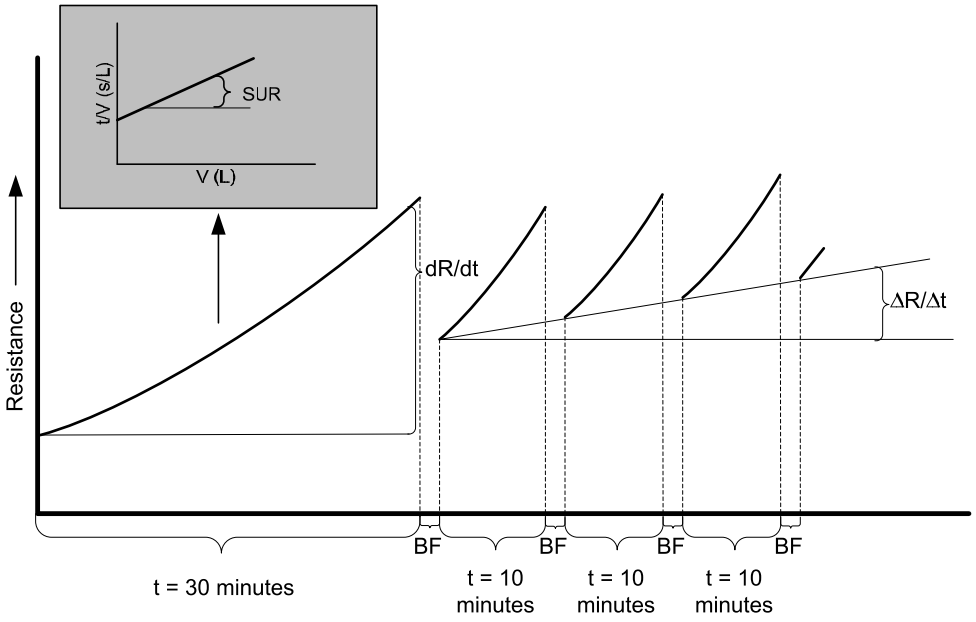


Figure 7.14 – Determination of the filterability (SUR and dR/dt) and reversibility by ΔR/Δt

In this section the accuracy of the reversibility test is presented. Firstly it had been checked if four backflushes were enough to obtain a reliable ΔR/Δt value. Secondly the reproducibility of the reversibility test is presented and finally one sample was diluted to investigate the effect of different foulants concentrations on the SUR and ΔR/Δt value.

7.3.5.2 Number of backflushes

To investigate the influence of the number of backflushes on the ΔR/Δt value three effluent samples were taken on different days at the WWTP Horstermeer. Each sample was measured in duplo and the average values are summarized Table 7.14. Unfortunately the SUR values of the WWTP effluent were relatively low during the sampling days. Therefore it was difficult to measure significant differences between the samples. However in spite of the low SUR values still different ΔR/Δt values were obtained during the measurements. Considering these small differences Table 7.14 shows no significant effect of the number of backflushes on the ΔR/Δt value. Therefore it seems that four backflushes are enough to gain an indication of the ΔR/Δt value of WWTP effluent during this test.

Table 7.14 – The SUR and ΔR/Δt values of effluent of WWTP Horstermeer at different numbers of backflushes (BF)

WWTP	SUR (10^{12} m^{-2})	ΔR/Δt, 4 BF (10^{12} 1/m·h)	ΔR/Δt, 10 BF (10^{12} 1/m·h)
Horstermeer	7.9	0.07	0.06
Horstermeer	3.8	0.04	0.04
Horstermeer	5.4	0.05	0.07

7.3.5.3 Reproducibility of the reversibility

In Table 7.15 the reproducibility of the complete test is presented. Like the previous experiment effluent samples were taken of the WWTP Horstermeer but now only on two different days. Unfortunately on both days the filterability of the WWTP effluent was very good. This resulted in almost similar results during both days. Only the third measurement of the first sample presented a difference with the previous measurements. However the second sample showed no significant difference throughout the measurement. Therefore, despite of the low SUR values these results indicate a reproducible test for SUR and ΔR/Δt.

Table 7.15 – Reproducibility of the ΔR/Δt value during two measurements with effluent of the WWTP Horstermeer

WWTP	SUR (10^{12} m^{-2})	ΔR/Δt, 4 BF (10^{12} 1/m·h)
Horstermeer	4.3	0.05
	4.2	0.05
	4.7	0.03
Horstermeer	4.3	0.03
	4.8	0.03
	4.9	0.04

7.3.5.4 Dilution experiment

Next to the reproducibility experiments only once a dilution experiment was performed to investigate the accuracy of the complete test. One sample was taken from the WWTP Horstermeer and diluted with demineralised water at the laboratory. The dilution factors were 2 and 4 as showed in Table 7.16.

Table 7.16 – Results of dilution experiment with effluent WWTP Horstermeer and demineralised water

WWTP	Dilution	SUR (10^{12} m^{-2})	ΔR/Δt, 4 BF (10^{12} 1/m·h)
Horstermeer	0	14.0	0.08
	2	6.0	0.04
	4	2.6	0.02

As expected and also shown by Roorda (2004) an almost linear relation was found between SUR decrease and effluent dilution (Figure 7.15). In Figure 7.15 the SUR and ΔR/Δt value of 0% WWTP effluent (100% demineralised water) is defined as 0% of the SUR and ΔR/Δt value of a 100% WWTP effluent sample. Next to the almost linear relation of SUR decrease a

linear relation was observed for ΔR/Δt values. Both relations may indicate that the SUR and ΔR/Δt values are related to the concentration of foulants. Although the latter experiment and the previous ones indicate that the reversibility can be measured accurately with the SUR+ equipment and related measuring procedure, confirmation would require the experiment to be repeated.

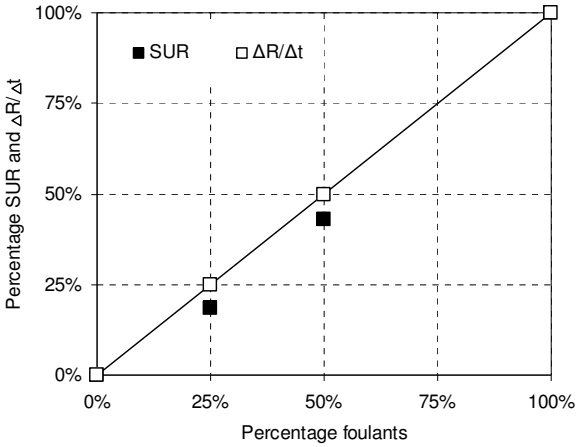


Figure 7.15 – Percentage of SUR and ΔR/Δt as a function of dilution for one experiment using effluent of the WWTP Horstermeer

7.3.6 Discussion

In this section an adapted version of the SUR equipment was presented: the SUR+ equipment. With this new equipment tests were performed to determine both the filterability and reversibility of WWTP effluent under well defined conditions. The defined conditions of the SUR+ measurement are summarized in Table 7.17.

Table 7.17 – The defined conditions of SUR+ measurement

Conditions	Unit	
Initial clean water permeability	$L/m^2 \cdot h \cdot bar$ (20 °C)	600 – 700
Trans membrane pressure during filtration	bar	0.5
Filtration time	minutes	$30 + 3 \times 10$
Backflush trans membrane pressure	bar	1.0
Backflush time	seconds	30
Numbers of backflushes	-	4
Temperature of feedwater	°C	20

The defined conditions were tested during different experiments. Firstly it was shown that the measured SUR values of the SUR and SUR+ equipment were comparable and reproducible. These results allow to use the SUR+ equipment also for just only the SUR measurement. The different ways to measure the permeate production did not significantly influence the SUR value of WWTP effluent. The other experiments focused on the reversibility (ΔR/Δt) part of

the test. The results of these tests seemed also reproducible and it was shown that four backflushes might be enough to obtain an indication of the ΔR/Δt value. The accuracy of the total test was determined at the hand of a dilution experiment. This experiment presented a good relation between the percentage of foulants and the percentage of the SUR and ΔR/Δt value. Considering these findings it was concluded that the SUR+ equipment and the accompanying measuring procedure might be useful to investigate the relation between filterability and reversibility of WWTP effluent.

7.4 Evaluation

In Figure 7.13 the dR/dt and ΔR/Δt values of effluent samples of WWTP Horstermeer during the period June – July, 2008 are plotted in one picture together with the values of Figure 7.10 . Both relations show that with increasing dR/dt values, the ΔR/Δt value increased as well. Although the relations are not similar it proves the result of the previous section (section 7.2). In this section a relation between the filterability and reversibility of ultrafiltration feedwater was suggested based on pilot-scale experiments. An explanation for the possible relation could be presence of foulants, colloids and particles in WWTP effluent. Generally these compounds are considered as the cause of fouling during ultrafiltration. At higher SUR values larger amounts of these compounds can be expected in comparison to lower SUR values. Consequently, it is assumable that a part of these compounds not only affect the filterability but also the reversibility of WWTP effluent. From the literature it is known that the compounds related to both reversibility and filterability do have the same origin and comparable characteristics (te Poele, 2005; Amy, 2008; Zheng *et al*., 2009). Therefore this might explain the relations as shown in Figure 7.16.

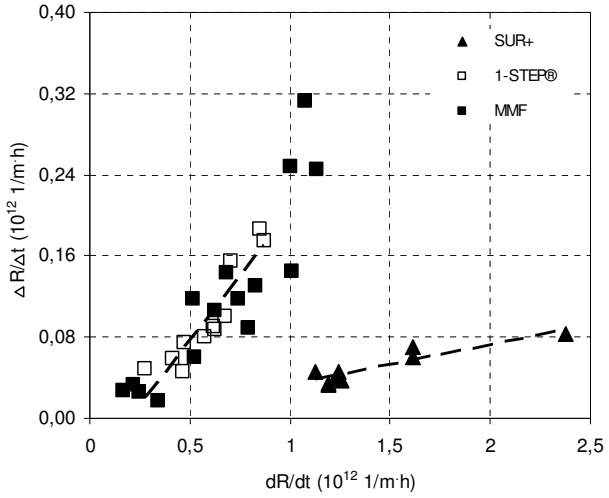


Figure 7.16 – The filtration properties (dR/dt and ΔR/Δt) of effluent of WWTP Horstermeer in the period June – July, 2008 measured with the SUR+ equipment and the filtration properties (dR/dt and ΔR/Δt) of multi media and 1-STEP® filtrate during pilot ultrafiltration at the WWTP Horstermeer as shown also in Figure 7.10

However in spite of the similarity (filterability related to reversibility) the relations present also a considerably difference. The dR/dt values of the pilot experiments are significantly lower than the values measured with the SUR+ equipment. This difference could be related to the different flux of the pilot experiments and the SUR+ measurement. Due to operation at constant pressure (0.5 bar) the initial flux of the SUR+ measurement is about 300 – 400 $L/m^2 \cdot h$ and decreases until approximately 100 $L/m^2 \cdot h$ after 30 minutes. Contrarily the constant operational fluxes of the pilot experiments were in the range of 29 – 86 $L/m^2 \cdot h$. Therefore different dR/dt values are evident and expected as shown in Figure 7.16.

Next to the different dR/dt values, which are related to the flux, the ΔR/Δt values of the pilot experiments and SUR+ measurements differed. As shown in Figure 7.16, compared to the ΔR/Δt values of the SUR+ measurements, the ΔR/Δt values of the pilot experiments are notably higher at the same dR/dt values. This difference could suggest that the reversibility of ultrafiltration feedwater also depends on the configuration of the membrane installation i.e. lab scale (SUR+) versus pilot scale (pilot ultrafiltration) in this research. During the SUR+ measurements the backflush conditions were well defined and also the configuration of the small membrane module (only three fibers inside) could result in an "optimal" backflush performance. This in contrast to the large membrane modules (35 m^2 each) of the pilot ultrafiltration installation. In this installation the backflush performance could be less efficient due to e.g. unequal distribution of backflush water, plugging of membrane fibres, etc.

Considering this difference between the SUR+ equipment and pilot ultrafiltration installation it is evident that the backflush efficiency of the pilot should be improved in order to operate at higher SUR values of WWTP effluent.

7.5 Conclusions

- Both filterability and reversibility of filtered WWTP effluent (multi media filter and 1-STEP® filter) are influenced by the operational flux of the ultrafiltration pilot installation.

- The efficiency of chemical cleaning and the initial condition of membranes influence the filterability and reversibility values of filtered WWTP effluent (multi media filter and 1-STEP® filter) during dead-end ultrafiltration.

- At the same operational flux of the ultrafiltration pilot installation, 1-STEP® filtrate presented significantly lower dR/dt and $\Delta R/\Delta t$ values than multi media filtrate. This difference might be related to the SUR values of both filtrates. The SUR values of multi media filtrate were above $7.0 \cdot 10^{12}$ m^{-2} and of the 1-STEP® filtrate in the range of $3 \cdot 10^{12}$ m^{-2} – $4 \cdot 10^{12}$ m^{-2}.

- The reversibility and filterability of filtered (multi media filter and 1-STEP® filter) and raw effluent of the WWTP Horstermeer are related i.e. low SUR values (high filterability) go hand in hand with a high reversibility.

- The SUR+ can be used as a method for measuring both filterability (SUR and dR/dt) and reversibility ($\Delta R/\Delta t$) of WWTP effluent in dead-end ultrafiltration. The measurement includes 1 hour (30 minutes and three times 10 minutes) of filtration and four backflushes at 1 bar during 30 seconds.

References

Amy, G. (2008). *Fundamental understanding of organic matter fouling.* Desalination, 231, 44 – 51.

Haberkamp, J., Ernst, M., Böckelmann, U., Swewzyk, U. and Jekel, M. (2008). *Complexity of ultrafiltration membrane fouling caused by macromolecular dissolved organic compounds in secondary effluent.* Water Research, 42, 3153 – 3161.

Heijman S.G.J., Amy G.L. and Gijsbertsen, A. (2005). *Water quality test for dead end ultrafiltration.* Proceedings AWWA WQTC Conference, 1 – 7, Quebec, Canada.

Huang, H., Young, T.A. and Jacangelo, J.G (2008). *Unified membrane fouling index for low pressure membrane filtration of natural waters: principles and methodology*. Environmental Science & Technology, 42, 714 – 720.

Huang, H., Young, T. and Jacangelo, J.G. (2009). *Novel approach for the analysis of bench scale, low pressure membrane fouling in water treatment*. Journal of Membrane Science, 334, 1 – 9.

Kim, J. and DiGiano, F.A. (2006). *A two fiber, bench-scale test of ultrafiltration (UF) for investigation of fouling rate and characteristics*. Journal of Membrane Science, 271, 196 – 204.

Lozier, J., Cappuci, L., Amy, G., Lee, N., Jacangelo, J.G., Huang, H., Young, T., Mysore, C., Emeraux, C., Clouet, J., Croué, J-P. and Heijman, B. (2008). *Natural organic matter fouling of low pressure membrane systems,* American Water Works Association Research Foundation, USA.

Poele, S. te (2005). *Foulants in ultrafiltration of wwtp effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp- effluent*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Zheng, X., Ernst, M. and Jekel, M. (2009). *Identification and quatification of major oganic foulants in treated domestic wastewater affecting filterability in dead-end ultrafiltration*. Water Research. 43, 1, 238 – 244.

Zheng, X., Ernst, M, Huck, P.M. and Jekel, M. (2010). *Biopolymer fouling in dead-end ultrafiltration of treated domestic wastewater.* Water Research, 44, 18, 5212 – 5221.

8 General evaluation

8.1 Introduction

The study presented in this thesis was initiated to investigate further treatment of WWTP effluent for high quality industrial applications using dead-end ultrafiltration membranes. Focuses of the study were the process performance of ultrafiltration installations in relation to the filtration properties of feedwater and the pretreatment options up front to ultrafiltration. During all the investigations the SUR parameter, developed by Roorda (2004), played a crucial role. With this parameter it is possible to characterize the filterability of WWTP effluent within a short time frame of 30 minutes.

In the subsequent sections the main findings of this study concerning the two major topics are presented. Section 8.2 discusses the results on process performance and filtration properties of ultrafiltration feedwater, while in section 8.3 the findings about pretreatment of ultrafiltration feedwater are compared and evaluated. The results themselves are described chapter 5 and 6, respectively 4 and 7. Overall conclusions and recommendations are summarised in section 8.4. The recommendations are offered with a view to future research concerning the SUR parameter as well as to the optimisation of the ultrafiltration process in general.

8.2 Process performance of ultrafiltration installations

8.2.1 SUR values and operational parameters.

Roorda (2004) was the first to report about the SUR value of ultrafiltration feedwater in relation to the operational flux of a (pilot) ultrafiltration installation, although the conclusion was rather general. Roorda (2004) stated that SUR values below $10 \cdot 10^{12}$ m^{-2} are a precondition for stable ultrafiltration performance with high fluxes ($\geq$ 100 $L/m^2 \cdot h$). Furthermore, it was concluded that ultrafiltration of feedwater with high SUR values will be accompanied with very extensive cleaning procedures in order to maintain high fluxes. In this thesis these observations of Roorda (2004) were further deepened in order to gain more specific operational criteria for ultrafiltration operation of WWTP effluent.

In chapter 4 of this thesis criteria have been developed for the evaluation of the process performance of ultrafiltration operation. These criteria have not been defined by Roorda (2004) and therefore it is unclear what exactly is meant by a stable process performance in that thesis. But it has to be emphasized that the development of process criteria to qualify process performance is difficult and also arbitrary. From the literature no standardized or universal process criteria are available. Most researchers and practitioners of membrane processes use their own criteria which are of course subjective. In this thesis the maximum rate of filtration resistance increase in 24 hours ($\Delta R_{max}/t_{24h}$) has been introduced for the purpose of quantifying and qualifying the process performance of the ultrafiltration pilot installation. This value was indirectly extracted from the datasheet of the membrane supplier

as presented in the thesis of te Poele (2005). Firstly, based on the maximum allowed trans membrane pressure of 1 bar, constant flux and feedwater temperature the maximum theoretical filtration resistance ($R_{max,th}$) was calculated. Secondly the $R_{max,th}$ value was divided by 24 hours (i.e. 1 day) resulting in the $\Delta R_{max}/t_{24h}$ value. The $\Delta R_{max}/t_{24h}$ value so incorporates a chemical cleaning interval of 24 hours. Of course, this interval is arguable but is also used by te Poele (2005) during comparable experiments. However, it is obvious that the $\Delta R_{max}/t_{24h}$ parameter is subjective as well but in this thesis the same definition is constantly applied. This approach makes it possible to qualify ultrafiltration process performance in relation to the SUR value of ultrafiltration feedwater. To qualify the process performance the $\Delta R_{max}/t_{24h}$ value was compared with the actual rate of filtration resistance increase ($\Delta R/\Delta t$) between two chemical cleanings. This comparison resulted in three categories of process performance: sub critical, critical and super critical. Sub critical process performance relates more or less to stable process performance as defined by Roorda (2004) and super critical to unstable process performance. Critical performance, the middle category, means the performance can be either sub or super critical. In this category the performance depends highly on external factors like e.g. the efficiency of the chemical cleaning, condition of the membranes, etc. In Table 8.1 an overview of the qualification of the process performance is given.

Table 8.1 – Qualification of the ultrafiltration process performance based on the $\Delta R_{max}/t_{24h}$ and $\Delta R/\Delta t$ values

Qualification process performance	Value
Sub critical	$\Delta R_{max}/t_{24h} > \Delta R/\Delta t$
Critical	$\Delta R_{max}/t_{24h} \sim \Delta R/\Delta t$
Super critical	$\Delta R_{max}/t_{24h} < \Delta R/\Delta t$

The effect of different chemical cleaning intervals on the qualification of ultrafiltration process is shown in Figure 8.1. On the left of this figure, Figure 4.5 is plotted in which the process performance ($\Delta R/\Delta t$) and SUR values were compared with the maximum filtration resistance increase during 24 hours (R_{max}/t_{24h}). On the right, the same figure is plotted but now including the maximum filtration resistance increase during 12 and 48 hours (R_{max}/t_{12h} and R_{max}/t_{48h}) and it shows clearly the effect of different cleaning intervals. Increase of the cleaning interval (24 → 48 hours) leads to super critical performance at lower SUR values and decrease of cleaning interval (24 → 12 hours) leads to sub critical performance at higher values. This effect of the different cleaning intervals must be well considered when the ultrafiltration process performance is evaluated i.e. selection of other cleaning intervals results in a different evaluation of the process performance.

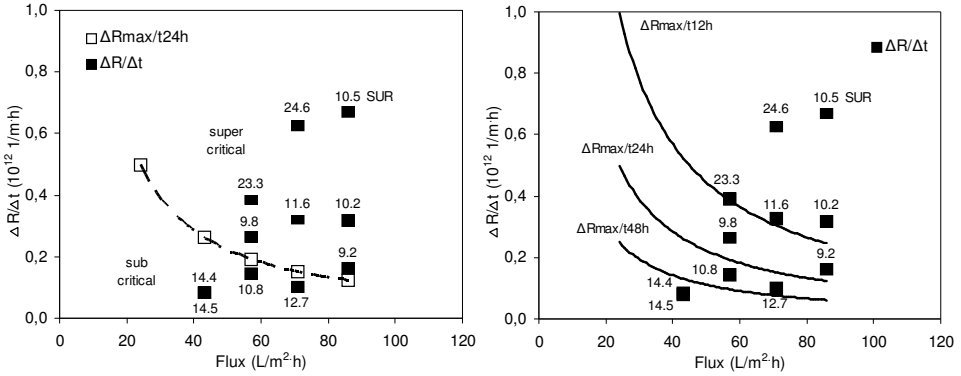


Figure 8.1 – Process performance (ΔR/Δt) and SUR values compared with the maximum filtration resistance increase during 24 hours (left) and the effect of changing chemical cleaning intervals (right)

The findings of Roorda (2004) were partly confirmed in this thesis. In contradiction to Roorda (2004) during the research described in this thesis considerable lower SUR values ($< 5 \cdot 10^{12}$ m^{-2}) were found as a precondition for sub critical process performance. When the SUR values are about $5 \cdot 10^{12}$ m^{-2} it seems possible to obtain (sub) critical process performance at fluxes of 60 – 80 L/m^2·h. But when the SUR values are higher i.e. about $10 \cdot 10^{12}$ m^{-2} the operational flux has to be about 40 – 60 L/m^2·h for (sub) critical process performance. At the same SUR value ($10 \cdot 10^{12}$ m^{-2}) Roorda (2004) noticed a significant higher flux (100 L/m^2·h). But as already discussed, Roorda (2004) did not present a definition of stable process performance. This makes the comparison complex but at a rough estimate it can be stated that the reported flux of Roorda (2004) did not correspond with the observations reported in this thesis

In Figure 8.2 the observations of this thesis are illustrated. This figure, the operating window presented in chapter 4, shows the relation between the SUR and flux. Also, in addition to Figure 4.6, the obtained process performance data of chapter 7 are included in this figure. As shown, these data confirm the operating window of chapter 4. Except one time all data points (sub critical) are in the area of sub critical process performance. It indicates the operating window is useful for the prediction of process performance and flux in relation to the SUR value.

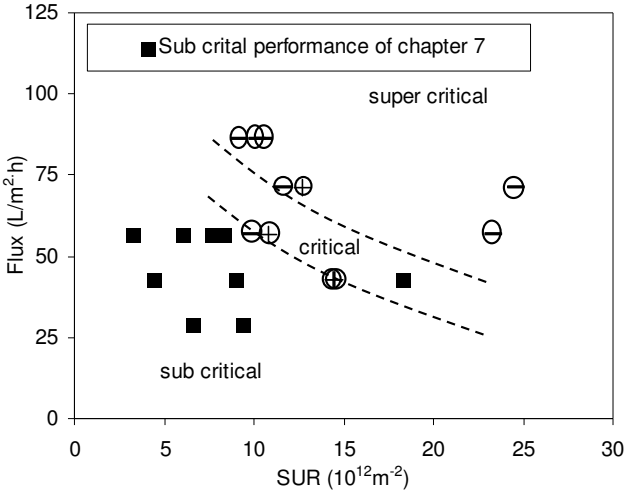


Figure 8.2 – Operating process performance window.

Next to the filtration properties of feedwater the efficiency of the chemical cleaning played an important role which is illustrated in Figure 8.3. This figure presents two possible situations. The black line represents an ideal process performance. During a filtration period the resistance/trans membrane pressure increases due to the remained fouling after hydraulic cleanings. After a certain period the membranes are chemically cleaned and the resistance/trans membrane pressure comes back to the initial level at the start. During the successive filtration periods this mode of operation continued and resulted in similar initial resistance/trans membrane pressure after chemical cleaning. A non ideal ultrafiltration process performance is illustrated by the dotted line in Figure 8.3. After a chemical cleaning the resistance/trans membrane pressure value does not restore because of insufficient chemical cleaning. During the following filtration periods this insufficient chemical cleaning results in an acceleration of the resistance/trans membrane pressure increase. This acceleration might be a consequence of the compressibility of the formed cake layer. As known from Roorda (2004) and Zheng *et al*. (2010b) the formed cake layer of WWTP effluent during ultrafiltration is highly compressible. Therefore higher trans membrane pressure could result in less hydraulic and chemical reversible fouling layers. Further the dotted filtration profile will result in different actual rates of filtration resistance increase ($\Delta R/\Delta t$) over time. Therefore it is necessary to keep an eye on cleaning efficiency when comparing actual filtration resistance increase ($\Delta R/\Delta t$) and maximum rate of increase in 24 hours. In other words insufficient chemical cleaning will lead sooner or later to critical or super critical process performance.

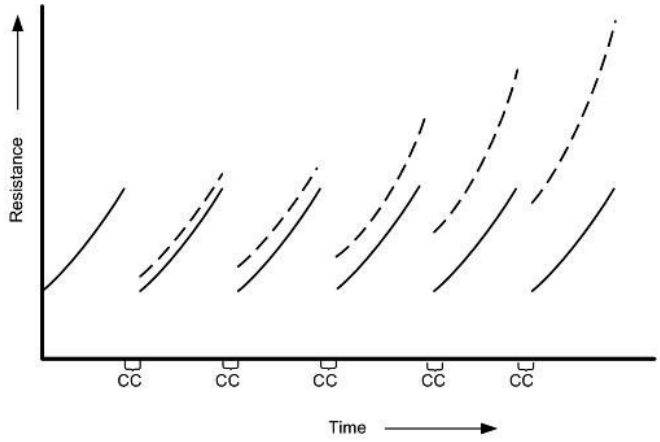


Figure 8.3 – Development of the resistance/trans membrane pressure (TMP) during constant flux ultrafiltration of WWTP effluent; the black line represents the ideal situation with sufficient chemical cleaning (CC); after chemical cleaning the resistance/trans membrane pressure restores completely; the dotted line represents a non ideal situation, insufficient chemical cleaning results in higher resistance/trans membrane pressure at the start of the subsequent filtration period, the resistance/trans membrane pressure increases disproportional due to cake compression.

8.2.2 Filterability and reversibility

In the previous section the filterability of WWTP effluent was related to the operational performance of an ultrafiltration installation. But the filterability of WWTP effluent is just one aspect influencing the performance. Other factors like chemical and hydraulic cleaning efficiency, flux, trans membrane pressure, cake compression, pretreatment, etc. influence the performance as well. In chapter 7 the impact of some of these aspects (flux and pretreatment) on the operational performance were investigated.

The effect of the operational flux on both the filterability and reversibility was notably shown during pilot experiments at the WWTP Horstermeer i.e. increase of the flux resulted in higher dR/dt and $\Delta R/\Delta t$ values (decrease of the filterability and reversibility) as illustrated in Figure 8.4, which is a copy of Figure 7.9.

A similar result, like Figure 8.4, was found by te Poele (2005). One explanation for the relation could be the increase of the amount of foulants that are passing the membrane surface and therefore an increase of foulants adsorption influencing both filterability and reversibility. Another explanation could be the compressibility of the fouling confirmed by Zheng *et al.* (2010b). Higher fluxes and/or higher trans membrane pressures result in more compression, leading to a lower reversibility (Zheng *et al.*, 2010b). Therefore to operate at higher fluxes lower initial SUR values are required in order to obtain sub critical process performance. To

obtain lower initial SUR values the WWTP effluent should be pretreated upstream the ultrafiltration installation.

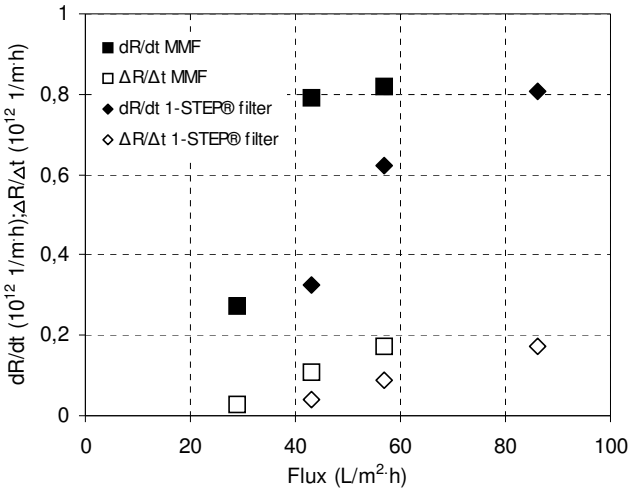


Figure 8.4 – Filtration properties (dR/dt and ΔR/Δt) at different flux rates during pilot ultrafiltration of multi media and 1-STEP® filtrate at the WWTP Horstermeer

Figure 8.4 also presents the effect of two different pretreatment technologies (multi media filter and 1-STEP® filter) on the filterability and reversibility. As shown, ultrafiltration of 1-STEP® filtrate leads to significant lower dR/dt and ΔR/Δt values than multi media filtrate at fluxes in the range of 43 and 57 L/m^2·h. This finding was confirmed by results on lab scale presented in chapter 5. These results are evaluated in the next section together with the performance of other pretreatment technologies.

Next to the effect of pretreatment also the efficiency of the backflush influence the operational performance. During lab scale experiment with the SUR+ equipment significantly higher dR/dt values were obtained compared to pilot scale experiments but the dR/dt values were measured more or less in the same range. This is schematically illustrated in Figure 8.5

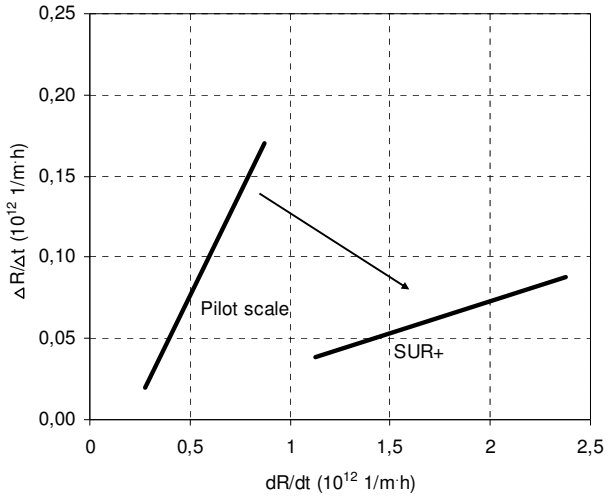


Figure 8.5 – Filtration properties of ultrafiltration feedwater obtained during pilot and lab scale experiments.

During the lab scale the backflush conditions were well defined and the membrane area of the module was about $10 \cdot 10^{-2}$ m^2 (3 membrane fibres of about 25 cm). Therefore the obtained $\Delta R/\Delta t$ values of the lab scale experiments were considered as the maximum attainable reversibility values at the measured dR/dt values. Considering this different relation of dR/dt and $\Delta R/\Delta t$ values of the lab and pilot scale experiments also attention should be paid to the performance of the backflush of ultrafiltration installation. A low SUR value of WWTP effluent during ultrafiltration has to be accompanied with optimal backflush condition in order to obtain sub critical process performance because of the observed relation between filterability and reversibility.

8.2.3 Condition of membranes

In chapter 4 and 6 the performance of the ultrafiltration units of the UF-RO installation at WWTP Sas van Gent was investigated. During two periods the UF-RO installation was fed with WWTP effluent after a stabilization pond (first phase) and WWTP effluent directly taken after a secondary clarifier. The results of the experiments showed a significant difference between the fouled and less fouled membranes as illustrated in Figure 8.6.

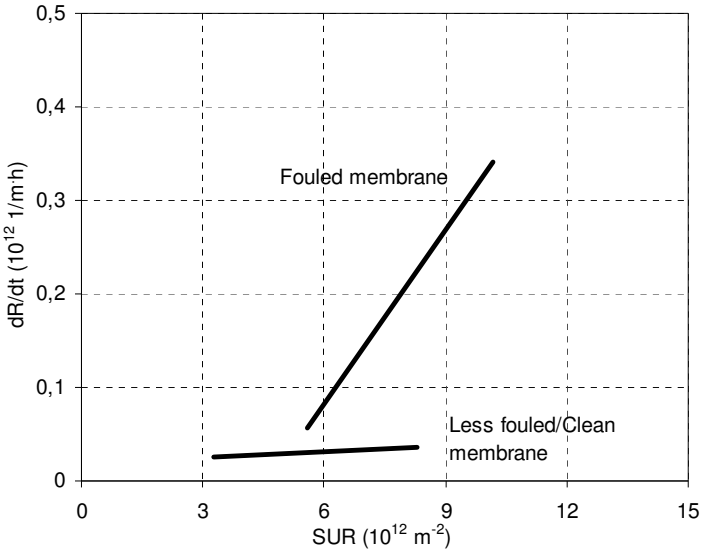


Figure 8.6 – Relationship between SUR values and filtration properties during pilot and lab scale experiments.

From the operational journal and actual permeability values it was known that the membranes of one unit (ultrafiltration unit 1) were more fouled than the membranes of another unit (ultrafiltration unit 4) before and during the first phase. This difference in fouling resulted in considerable higher dR/dt values for ultrafiltration unit 1 compared to ultrafiltration unit 4. Therefore it is concluded that the condition of membranes does influence the process performance.

The influence of the condition of the membranes was confirmed by the results of the second phase of the experiment. Between phase 1 and 3 the membrane modules of ultrafiltration were intensively cleaned during a cleaning-in-place (CIP). This cleaning resulted in lower dR/dt values during the second phase at same SUR values of WWTP effluent. Furthermore the effect of chemical cleaning in place showed that the formed fouling of the first phase (stabilized effluent) was hydraulically reversible and as stated earlier this experiment proved again that clean membranes are needed (i.e. efficient hydraulic and chemical cleaning) in order to obtain sub critical process performance. With the SUR measurement it was possible to relate this finding to the initial SUR value of WWTP effluent.

8.3 Pretreatment of ultrafiltration feedwater

In the previous section the need for sufficient chemical and hydraulic cleaning to control membrane fouling was emphasized. Another option is to pretreat WWTP effluent upstream an ultrafiltration installation. This integration of pretreatment and ultrafiltration, to improve the process performance, is an important trend in the development of membrane filtration (Huang

et al., 2007) and is underlined by the evaluation in the previous section. In addition to efficient chemical and hydraulic cleaning the SUR value of ultrafiltration feedwater should be $\leq 5 \cdot 10^{12}$ m^{-2} for sub critical process performance at higher fluxes. But, to obtain this SUR value the ultrafiltration feedwater has to be pretreated in most cases. Therefore in this thesis (chapter 5 and 6) the effect of different pretreatment technologies on the SUR value of WWTP effluent was investigated. Beside the SUR value also the effect of pretreatment on the concentration of foulants and fractions of WWTP effluent were investigated and are evaluated in this section.

8.3.1 Effect pretreatment on foulants

In chapter 5 and 6 the effect of different pretreatment technologies on the concentrations of foulants of WWTP effluent were presented. The foulants are measured as DOC, colour, proteins, polysaccharides and humic substances during the experiments. From all these compounds several researchers suggested the main foulants to be proteins and polysaccharides i.e. soluble microbial products and/or biopolymers (Jarusutthirak and Amy, 2001; te Poele, 2005; Rosenberger, *et al*., 2005; Jarusutthirak and Amy, 2007; Haberkamp, *et al*., 2008; Zheng, *et al*., 2010b). Considering this suggestion, the evaluation of the effect of pretreatment technologies on foulants will mainly focus on these substances.

In chapter 5 the results of different pretreatment technologies with activated carbon as adsorption and/or filtration medium were presented. On lab scale two different types of powdered activated carbon have been tested in combination with (paper) filtration. Furthermore on pilot scale the effect of powdered activated carbon has been investigated at the WWTP Maasbommel. As for the foulants, both the lab and pilot experiments presented comparable results. The experiments showed a significant adsorption of the humic substances, colour and proteins of WWTP effluent. This observation is also noticed in studies of other researchers (Shon *et al*., 2004a; Haberkamp *et al*., 2007). For example, Haberkamp *et al*. (2007) concluded that activated carbon adsorbs organic compounds of a wide range of molecular weights but most of the adsorption capacity was used by low molecular weight substances (i.e. colour and humic substances) instead of high molecular weight substances (i.e. biopolymers). Consequently, Haberkamp *et al*. (2007) investigated the combination of coagulation and adsorption. In contrast to adsorption, coagulation removes predominantly organic macromolecular substances of WWP effluent (te Poele, 2005; Haberkamp *et al*., 2007). Therefore, regarding the removal of foulants, the combination of coagulation and adsorption seems to be more beneficial than adsorption only. As shown in chapter 5 high dosages of powdered activated carbon were needed to remove considerable amounts of foulants. Combining adsorption with coagulation will probably result in lower dosages of powdered activated carbon and consequently lower operational costs.

The adsorption experiments have not been performed only with powdered activated carbon but also with granulated activated carbon at the WWTP Maasbommel and WWTP Horstermeer. At the WWTP Maasbommel a "conventional" granulated activated carbon filter has been tested. Like the experiments with powdered activated carbon a considerable removal of proteins, humic substances and colour has been noticed during this test. The same result was obtained with the 1-STEP® filter at WWTP Horstermeer. During a research period of four months the removal of respectively proteins, humic substances and colour was on average 30%, 30% and 41%. These observations at WWTP Maasbommel and WWTP Horstermeer were confirmed by another study of Shon *et al*. (2004b). In that study a biological granulated activated carbon filter as pretreatment technology was tested on lab scale and the results showed also removal of hydrophilic organic matter which is the extracellular enzyme of microorganisms in WWTP effluent. Considering the obtained results and the literature it seemed that granulated activated carbon is able to adsorb part of the foulants.

Next to the investigations with the 1-STEP® filter, the multi media filter has been tested at the WWTP Horstermeer. During a period of approximately three months samples of WWTP effluent and filtrate were taken for foulants analyses and SUR measurements. The results of the foulants analyses of the multi media filter were different from the results of the 1-STEP® filter. In contrast to the 1-STEP® filter the multi media filter did not remove significantly any foulant. This absence of foulants removal might be related to the different filter media of both filters i.e. quartz sand/anthracite (multi media filter) versus granulated activated carbon (1-STEP® filter). Therefore it seemed that adsorption is needed to remove foulants at filtration rates of about 10 m/h. When the filtration rates are much lower (slow sand filtration or bank filtration) it seems that only sand is adequate to remove foulants. In a pilot scale investigation of Zheng *et al*. (2010a) considerable amounts of dissolved organic foulants/biopolymers were removed by slow sand filtration. Moreover additional experiments showed that the biopolymers were removed by biodegradation (Zheng *et al*., 2010a).

The effect of a stabilization pond on the removal of foulants has been investigated at the WWTP Sas van Gent. This experiment presented a significant decrease of the concentration of proteins and to a lesser extent also removal of colour and humic substances. Nevertheless, this decrease of concentrations of foulants did not result in significant lower SUR values due to the presence of algae in the stabilization pond.

In chapter 5 the concentrations of foulants were also compared with the SUR values of WWTP effluent and different filtrates in order to determine a relation between both. These comparisons suggested that the concentrations of foulants and corresponding SUR values cannot be related for any compound. This observation has been also made by te Poele (2005) during more or less similar experiments. Instead of (dissolved) foulants te Poele (2005) stated that particles and colloids are of substantial influence on the filterability of WWTP effluent.

Therefore during the research presented in this thesis fractionation experiments have been performed which will be evaluated in the next section of this chapter. In contrast to the observations in this thesis other researchers determined a proportionally between the biopolymer content and filterability of WWTP effluent (Zheng *et al*., 2010b). However, this observed difference seemed to be related to the applied analytical methods. During the experiments presented in this thesis the protein and polysaccharide concentrations in the water phase (< 0.45 µm) were determined according to the modified methods of Rosenberger (2003) which are based on the colorimetric methods described by Lowry *et al*. (1951) and Dubois *et al*. (1956). This method does not provide information about the size of proteins and polysaccharides. During the analyses both the colloidal and dissolved proteins and/or polysaccharides are analysed. Therefore this method does not distinguish dissolved proteins and polysaccharides that can pass the membrane and colloidal proteins and polysaccharides that are retained by the membrane. In the research of Zheng *et al*. (2010b) the biopolymers were analysed after size exclusion chromatography. Using this method (size exclusion) means that only a part (dissolved) of biopolymers are measured. Consequently the colloidal biopolymers are not taken into account with this method. Considering these shortcomings of both methods (colorimetric and size exclusion chromatography) fractionation experiments have been applied additionally in this thesis to unravel the effect of pretreatment.

8.3.2 Effect pretreatment on the fractions of WWTP effluent

At the WWTP Horstermeer and WWTP Maasbommel different fractionation experiments, presented in chapter 5, have been performed in order to unravel the effect of pretreatment on the initial SUR values of WWTP effluent. From previous research conducted by Roorda (2004) and te Poele (2005) it was known that especially the colloids in the size range of 0.1 – 0.45 µm are of major influence on the filterability. But in contradiction to te Poele (2005) the results of Roorda (2004) focused in particular on the fraction size of 0.1 – 0.2 µm. The importance of this particular size range was proven by the experiments shown in chapter 5. For example in the effluent of WWTP Horstermeer (feedwater of the multi media filter and 1-STEP® filter) the average contribution of this size range was 46% and 5% for the size range of 0.45 – 0.2 µm. This observation was also proved by the fractionation test with effluent of WWTP Maasbommel. During these tests (three) the average contribution of the fraction 0.1 – 0.2 µm was 44% and of the fraction 0.45 – 0.2 µm only 3%. Therefore it can be concluded that the results of this thesis confirm the results of Roorda (2004).

The dominant role of the fraction size 0.1 – 0.2 µm on the filterability of WWTP effluent was also shown during the ultrafiltration pilot test at WWTP Maasbommel. The design of the ultrafiltration pilot resulted in an accumulation of colloids. Therefore the SUR values in the PAC contact tank increased substantially. This decrease of filtration performance (i.e. permeability) was also mentioned in the report of the study (STOWA, 2007). Therefore the

important role of the fraction 0.1 – 0.2 µm should be considered well in the design phase of ultrafiltration treatment plants.

A possible way to reduce the impact of the fraction 0.1 – 0.2 µm is pretreatment. This possibility was investigated at the WWTP Maasbommel and WWTP Horstermeer. At the WWTP Maasbommel a granulated activated carbon filter was operated and at the WWTP Horstermeer a multi media filter and 1-STEP® filter. Granulated activated carbon filtration resulted in a significant removal of the fraction size > 0.45 µm but hardly any effect was observed regarding the fraction 0.1 – 0.2 µm of WWTP effluent. Almost similar results were obtained during the experiment with the multi media filter. These results revealed a slight removal of the > 0.45 µm fraction size but surprisingly the contribution of the fraction 0.1 – 0.2 µm increased after filtration. This in contrast to the results of the 1-STEP® filter. After 1-STEP® filtration the contribution of both fractions, 0.1 – 0.2 µm and > 0.45 µm, decreased significantly. Therefore it was possible to achieve SUR values below approximately $5 \cdot 10^{12}$ m^{-2} during the whole experimental period, independent on the initial SUR value of WWTP effluent. In Figure 8.7 these findings are summarized in one picture illustrating which applied pretreatment technology removes which fraction.

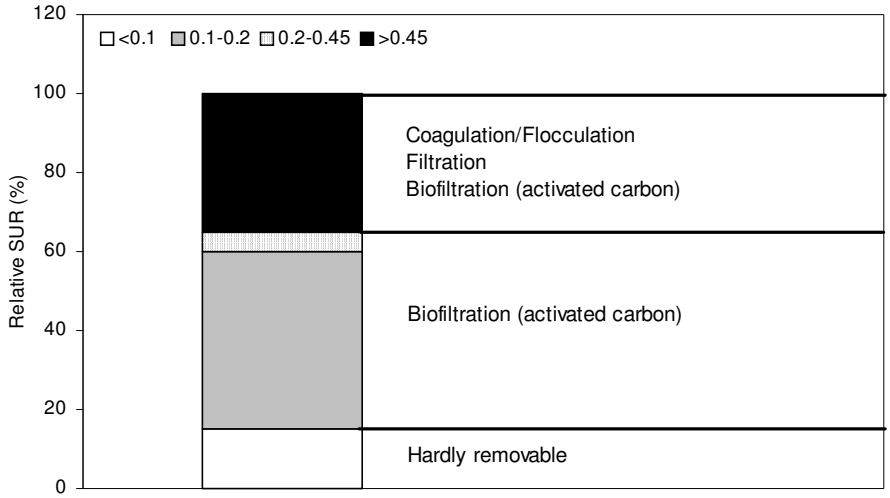


Figure 8.7 – Fraction size distribution of WWTP effluent and pretreatment.

During the experiments at WWTP Horstermeer no deep research was carried out to investigate the different mechanisms in the 1-STEP® filter that are responsible for the removal of both fractions. But it is obvious that coagulation/flocculation and filtration were responsible for the removal of the fraction > 0.45 µm. This phenomenon was also observed in other studies by e.g. Roorda (2004). But the mechanism that effects the removal and/or conversion of the fraction 0.1 – 0.2 µm is less clear. However, after slow sand filtration Zheng

et al. (2009) presented also during comparable experiments a considerable decrease of the contribution of the fraction size 0.026 – 0.45 µm to fouling resistance of ultrafiltration membranes. Like the 1-STEP® filter, slow sand filtration could be considered as biofiltration and therefore both studies suggest that biological activity played an important role in the removal or conversion of the fraction 0.1 – 0.2 µm. Furthermore another study by Remy et al. (2010) suggests that addition of powdered activated carbon resulted in stronger sludge flocs in membrane bioreactor. Probably a similar process occurred in the 1-STEP® filter, resulting in a retention of colloids and microbial products. To gain more insight into these removal and conversion mechanisms in the 1-STEP® filter more research is needed.

8.3.3 Initial SUR value

In chapter 5 and 6 it revealed that the SUR value after pretreatment depends highly on the initial SUR value of WWTP effluent. In chapter 6 it was shown that the subsequent pretreament steps of coagulation – dual media filtration – coagulation did not affect the SUR values when these were initially around $5 \cdot 10^{12}$ m^{-2}. But with higher SUR values the contribution of each step increased as well. An similar observation was gained during the experiment presented in chapter 5 and illustrated in Figure 8.8.

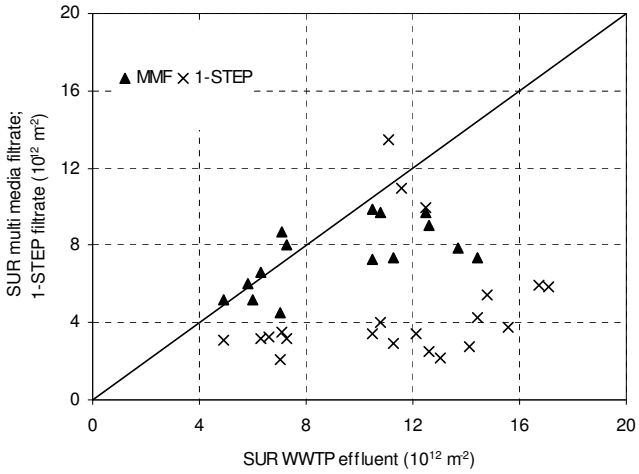


Figure 8.8 – Relation between the SUR values of WWTP effluent and the SUR values of multi media filtrate and 1-STEP® filtrate

Regarding the multi media filter the possibility to lower the initial SUR value was minimal when the SUR values of WWTP effluent were below $10 \cdot 10^{12}$ m^{-2}. At higher SUR values than $10 \cdot 10^{12}$ m^{-2} the average relative SUR decrease was 28%. The same average decrease was observed by Roorda (2004) during multi media filtration. However during the experiments with the 1-STEP® filter the relative SUR decrease showed to be less dependent on the initial

SUR value. During the whole experimental period the average relative SUR decrease was 56%. Even when the initial SUR values were around $5 \cdot 10^{12}$ m^{-2} it was possible to lower the SUR value. These differences between the 1-STEP® filter on the one hand and coagulation and dual or multi media filtration on the other hand might be related to the results of the fractionation tests. The 1-STEP® filter was able to remove both fractions (0.1 – 0.2 µm and > 0.45 µm) whereas the other pretreatment technologies only remove the fraction > 0.45 µm.

In Figure 8.9 a schematic overview of the initial SUR value, pretreatment method and possible process performance is shown. This picture is based on the observations described in the previous sections. The figure shows that when the initial SUR value is below $5 \cdot 10^{12}$ m^{-2} sub critical process performance can be obtained without pretreatment. But when the SUR values are in the range of $5 \cdot 10^{12}$ m^{-2} – $10 \cdot 10^{12}$ m^{-2} pretreatment is required. For critical process performance the conventional pre-treatment technologies are applicable but for sub critical performance biofiltration with activated carbon is needed. At higher initial SUR values (5 – $10 \cdot 10^{12}$ m^{-2}) the process performance becomes more and more critical. And the efficiency of the chemical and hydraulic cleaning becomes more important. Further when the SUR values are ≥ $15 \cdot 10^{12}$ m^{-2} the process performance becomes problematic and intensive cleanings will be needed or extra pretreatment steps.

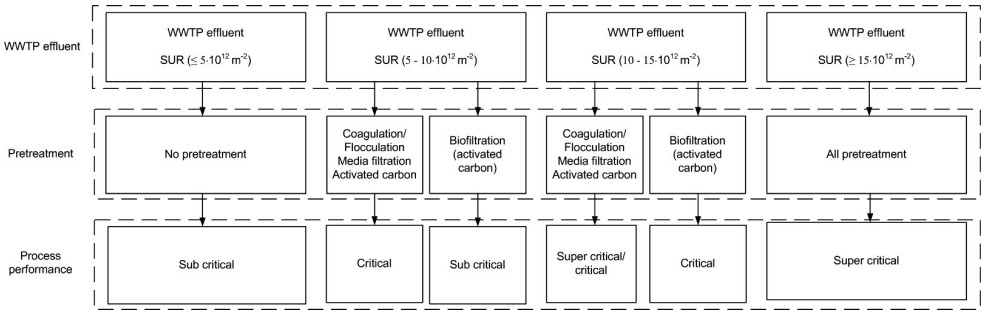


Figure 8.9 – Schematic overview of the initial SUR value of WWTP effluent in relation to required pre-treatment steps and process performance.

8.4 Recommendations and general conclusions

8.4.1 Recommendations

As demonstrated in this thesis the SUR value of WWTP effluent is a dynamic parameter. The maximum frequency that was applied with the SUR measurement at WWTP Horstermeer was 3 times per day. This frequency of measurements provide useful information but to gain a better indication on the hydrodynamic behaviour the frequency has to increase. Therefore an automatic SUR measurement should give an added value to indicate the filterability dynamics. In this way pretreatment or ultrafiltration performance can be controlled and optimised more frequently in order to obtain (sub) critical process performance.

The SUR+ measurement was introduced for measuring both the filterability of WWTP effluent and reversibility of the fouling layer. The results of the SUR+ measurement are reproducible and therefore this equipment could be used to perform further research on the reversibility of the fouling layer. But it has to be considered that reversibility is a reflection of the physical cleaning efficiency. Therefore it is difficult to translate reversibility values obtained during lab scale experiment into practice.

It was shown that effective chemical cleanings are needed to obtain sub critical performance at higher SUR values. Therefore the cleaning procedure and type of chemical should be well considered. An alternative for chemical cleaning could be enzymatic cleaning. Te Poele (2005) showed a complete recovery of the clean water flux after enzymatic (protease) cleaning.

In chapter 7 different reversibility values were presented for pilot and lab scale experiments with WWTP effluent of the same SUR values. Therefore it was suggested that the reversibility depends on the hydraulic regime in the membrane modules. Research into these hydraulics during a backflush could probably lead to an optimization of the backflush efficiency. Another option could be to apply a different type of backflush water instead of ultrafiltration permeate. Recent research (Li, *et al*., 2009) has shown beneficial effects when demiwater is applied for backflush during ultrafiltration.

In this thesis the evaluation of the effect of pretreatment was only based on the ultrafiltration performance. But in practice, ultrafiltration is mostly only a part of a complete treatment line. Therefore the performance of pretreatment technologies should be evaluated in the context of a treatment scheme. For example, an disadvantage of methanol dosage could be the enhancement of the biofouling rate of reverse osmosis membranes when these are placed after ultrafiltration.

8.4.2 Conclusions

The most important conclusions of the presented investigations on lab and pilot scale are summarised below:

- The process performance of ultrafiltration installations treating WWTP effluent depends on the SUR value. To operate at higher fluxes (60 – 80 L/m^2·h) SUR values below $5 \cdot 10^{12}$ m^{-2} are required for sub critical process performance. When the SUR values are around $10 \cdot 10^{12}$ m^{-2} operational fluxes should be 40 – 60 L/m^2·h in order to obtain sub critical process performance.

- The process performance of ultrafiltration installation depends on the condition of the membranes and efficiency of hydraulic and chemical cleaning. At higher SUR values (≥ $10 \cdot 10^{12}$ m^{-2}) effective cleaning becomes more important and a precondition for sub critical process performance.

- Both, lab and pilot scale experiment, indicate that filterability of WWTP effluent and reversibility of the fouling layer are related i.e. high filterability will also mean high reversibility.

- The filterability of WWTP effluent is mainly determined by the fraction of WWTP effluent with a size range (0.1 – 0.2 µm) five to twenty times the pore diameter of the applied membrane material.

- No significant relation between the filterability of WWTP effluent and foulants concentrations (DOC, humic substances, colour, proteins and polysaccharides) is noticed during dead-end ultrafiltration.

- Pretreatment of WWTP effluent with coagulation/flocculation, multi/dual media filtration and granulated activated carbon filtration resulted in a small to moderate increase of filterability. This increase of filterability depends on the initial SUR value of WWTP effluent. Pretreatment of WWTP effluent with the 1-STEP® filter resulted in a considerable increase of the filterability (SUR value ≤ $5 \cdot 10^{12}$ m^{-2}) and was less dependent on the initial SUR value.

- Instead of multi media filtration and granulated activated carbon filtration the experiments with the 1-STEP® filter show a significant removal of both fraction sizes, 0.1 – 0.2 µm and > 0.45 µm, that contribute to the SUR value of WWTP effluent.

- Next to a higher filterability, 1-STEP® filtration resulted in a higher reversibility of the fouling layer during dead-end ultrafiltration on pilot scale.

- During 1-STEP® filtration removal or conversion of the fraction 0.1 – 0.2 µm was shown but the responsible mechanisms were still unclear. Therefore research should be performed to unravel this phenomenon in order to optimize this type of filtration.

8.4.3 Final remark

During the research presented in this thesis the SUR measurements played a crucial role. It was demonstrated that the SUR measurements can be used to control and even predict the process performance of full and pilot scale installations. Further, the necessity of effective pretreatment was stressed by SUR measurements. Considering these results it is concluded that the SUR measurement is a useful tool to comprehend and optimize the filtration process of WWTP effluent.

References

Amy, G. (2008). *Fundamental understanding of organic matter fouling.* Desalination, 231, 44 – 51.

Dubois, M., Gilles, K.A., Hamilton, J.K., Rebers, P.A. and Smith F. (1956). *Colourmetric method for determination of sugars and related substances*. Analytical Chemistry, 28, 350-356.

Haberkamp, J., Ruhl, A.K., Ernst, M. Jekel, M. (2007). *Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting behaviour in ultrafiltration*. Water Research, 41, 3794-3802.

Haberkamp, J., Ernst, M., Böckelmann, U., Szewzyk, U. and Jekel, M. (2008). *Complexity of ultrafiltration membrane fouling caused by macromolecular dissolved organic compounds in secondary effluents*. Water Research, 42, 3153 – 3163.

Huang, H., Young, T. and Jacangelo, J.G. (2007). *Pretreatment and its effect and implications on water treatment by low pressure membranes.* Proceedings 4th IWA leading-edge conference and exhibition on water and wastewater technologies, June 3-6, Singapore, Republic of Singapore.

Jarusutthirak, C. and Amy, G. (2001). *Membrane filtration of wastewater effluents for reuse: effluent organic matter rejection and fouling.* Water Science and Technology, 42, 10, 225 – 232

Jarusutthirak, C. and Amy, G. (2007). *Understanding soluble microbial products (SMP) as component of effluent organic matter (EfOM)*. Water Research, 41, 2787 – 2793

Li, S., Heijman, S.G.J., Verberk, J.Q.J.C., Verliefde, A.R.D., Kemperman, A., Dijk, van J.C. and Amy, G. (2009). *Impact of backwash composition on ultrafiltration fouling control*. Journal of Membrane Science, 344, 17 – 25.

Lowry, O.H., Rosebrough, N.J., Farr, A.L. and Randall, R.J. (1951). *Protein measurement with the Folin phenol reagent*. Journal of Biological Chemistry, 193, 265-275.

Poele, S. te (2005) *Foulants in ultrafiltration of wwtp effluent*. PhD thesis, Department of Sanitary Engineering, Delft University of Technology, Delft, The Netherlands.

Remy, M., Potier, V., Temming, H. and Rulkens, W. (2010). *Why low powdered activated carbon addition reduces membrane fouling in MBRs*. Water Research, 3, 861 – 867.

Roorda, J.H. (2004). *Filtration characteristics in dead-end ultrafiltration of wwtp-effluent.* PhD thesis, Department of Sanitary Engineering, Delft University of Technology, The Netherlands.

Rosenberger, S. (2003). *Charkterisierung von belebtem Schlamm in Membranbelebungsreaktoren zur Abwasserreiniging*. PhD thesis, Technical University of Berlin, Berlin, Germany.

Rosenberger, S., Evenblij, H., te Poele, S., Wintgens, T. and Laabs, C. (2005). *The importance of liquid phase analyses to understand fouling in membrane assisted activated sludge processes – six case studies of different European research groups.* Journal of Membrane Science, 263, 113 – 126.

Shon, H.K., Vigneswaran, S., Kim, S.I., Cho, J. and Ngo, H.H. (2004a). *The effect of pretreatment to ultrafiltration of biologically treated sewage effluent: a detailed effluent organic matter (EfOM) characterization*. Water Research, 38, 1933 – 1939.

Shon, H.K., Vigneswaran, S., Kim, S.I., Cho, J. and Ngo, H.H. (2004b). *Effect of pre-treatment on the fouling of membranes: application in biologically treated sewage effluent.* Journal of Membrane Science, 234, 111-120.

STOWA (2007). *Effluentnabehandeling op de rwzi Maasbommel*. Report 2007-17 (in Dutch), Utrecht, The Netherlands.

Zheng, X., Ernst, M. and Jekel, M. (2009). *Identification and quatification of major oganic foulants in treated domestic wastewater affecting filterability in dead-end ultrafiltration*. Water Research. 43, 1, 238 – 244.

Zheng, X., Ernst, M, and Jekel, M. (2010a). *Pilot-scale investigation on the removal of organic foulants in secondary effluent by slow sand filtration prior to ultrafiltration.* Journal of Membrane Science, 44, 10, 3203 – 3213.

Zheng, X., Ernst, M, Huck, P.M. and Jekel, M. (2010b). *Biopolymer fouling in dead-end ultrafiltration of treated domestic wastewater.* Water Research, 44, 18, 5212 – 5221.

Appendix A Ultrafiltration pilot installation and membrane properties

Ultrafiltration pilot installation

The ultrafiltration installation used in this research is designed by Rossmark Waterbehandeling and Witteveen+Bos Consulting Engineers and is schematically shown in Figure 1.

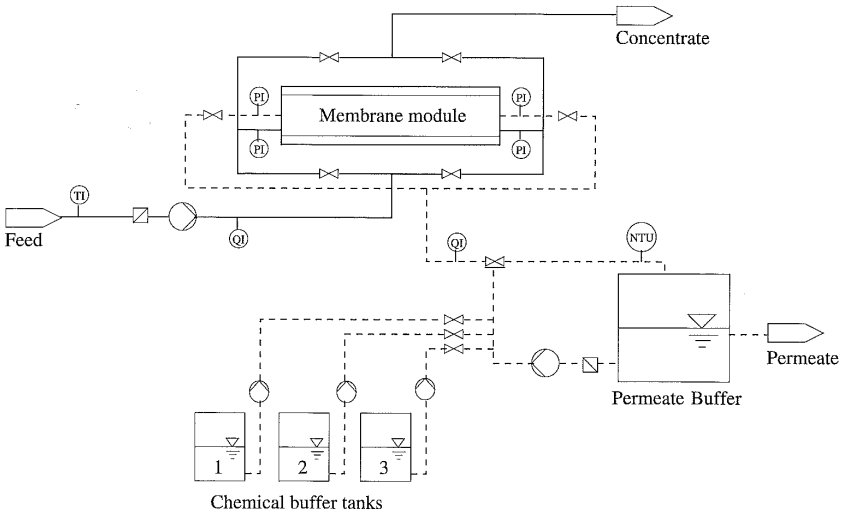


Figure 1 – Flow diagram of the pilot ultrafiltration installation

Membrane module

The membrane module used in the ultrafiltration installation is the 8" Capfil Membrane element PVC, type S-225 FSFC of Norit X-Flow. A schematic picture of the element with the specification is given in Figure 2 and Table 1.

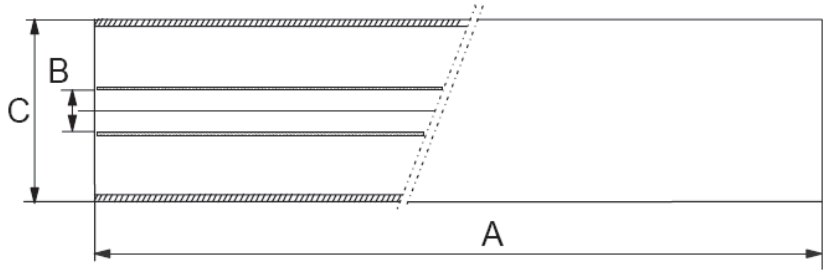


Figure 2 – Scheme of the 8" Capfil Membrane element PVC, type S-225 FSFC of X-Flow.

Table 1 – Module specifications of S-225 FSFC

Parameter	Unit	Value
Hydraulic membrane diameter	mm	0.8
Membrane area	m^2	35
Element length A	mm	1527.5
Permeate collector inner diameter B	mm	42.6
Element outer diameter	mm	200

Materials of construction are:

- PVC housing
- polyethersulfone or PVC flow distributor
- epoxy potting

The operating specifications are presented in Table 2.

Table 2 – Module operating specifications of S-225 FSFC

Parameter		Specification
Maximum system pressure	0 – 20 °C	600 kPa
	20 – 25 °C	500 kPa
	25 – 30 °C	400 kPa
	30 – 35 °C	350 kPa
	35 – 40 °C	300 kPa
Maximum transmembrane pressure	0 – 40 °C	300 kPa
Maximum back flush pressure	0 – 40 °C	300 kPa
Maximum operation temperature		40 °C

Ultrafiltration membranes

The membranes used in the ultrafiltration pilot installation and lab scale set-up (SUR) are the Capfil ultrafiltration membrane, type UFC M5 of Norit X-Flow.

Basic characteristics:

- hydrophilic polyethersulfone membrane
- cappilary membrane of 0.8 mm
- structure asymmetric/microporous
- developed for inside-out filtration
- developed for use in large scale processes for water purification
- high performance and very good anti-fouling behaviour
- membrane elements can be backflushed for efficient membrane cleaning

Membrane composition

- Hydrophilic membrane composed of a blend of polyvinylpyrrolidone and polyethersulfone
- M5: contains glycerine for pore protection and bisulfite for prevention of microbial growth.

The membrane performance data is presented in Table 3.

Table 3 – Ultrafiltration membrane performance data

Parameter	Unit	Specification	Remarks
Trans membrane pressure	kPa	-300 +300	
Pore size	nm	25 – 30	
Molecular weight cutoff	kDa	150 – 200	on 1 wt% PVP at 1 bar
pH feed	-	2 – 12	
Temperature	°C	1 – 80	
Chloride exposure	ppm·h	250000	500 ppm max. at 0 – 40 °C

The permeability of clean water (at least UF permeate quality) as measured on a single membrane capillary is approximately 600 – 700 L/m^2·h·bar at 20 °C. The permeability of clean water as measured on a module is approximately 300 – 400 L/m^2·h·bar at 20 °C.

Since the resistance of the membrane to solvents strongly depends on the actual process conditions, the indications given in Table 4 should be considered as guidelines.

Table 4 – Resistance of the ultrafiltration membrane to solvents

Solvent	Resistance[1]
Acids	++
Bases	++
Organic esters, ketones, ethers	-
Aliphatic alcohols	+
Aliphatic hydrocarbons	+
Halogenated hydrocarbons	-
Aromatic hydrocarbons	-
Polar organic solvents	-
Oils	++

[1] ++: no change in membrane properties; +: membrane properties may slightly change; -: significant change in membrane properties

The cleaning agents which can be used are listed in Table 5.

Table 5 – Possible cleaning agents to use for the cleaning of the ultrafiltration membrane

Chemical	Specification
NaOCl (active chloride)	200 ppm max. at 40 °C and max. 30 minutes per day
H_2O_2	100 – 200 ppm at 40 °C
NaOH + EDTA	pH ≤ 12 + 1 wt%
HCl	pH ≥ 1
Citric acid	2 wt%
Enxymatic compounds	-

It is recommended to keep the pH between 1 and 13 and not to exceed a temperature of 80 °C (depending on the module type) during cleaning and/or desinfection.

Appendix B Polysaccharides and proteins

Polysaccharides

For the analysis of polysaccharides the by Rosenberger (2003) modified method of Dubois *et al.* (1956) is used. For the calibration D(+)-glucose (J.T.Baker), in a concentration range between 0.5 - 10 mg/L is used. The concentration can be calculated with the measured extinction and calibration curve.

Reagents

A: 5 % Phenol solution in demineralised water
B: 95 - 97 % sulphuric acid

Method

A sample of 4 mL is filled in a round tube and 2 mL reagent A is added. After mixing 10 mL reagent B is added in a spout in order to get a proper mixing. Thereafter the mixture is stored for 10 minutes at room temperature, mixed again and incubated for 30 minutes at room temperature. The adsorption is then measured in a 4 cm cuvett at a wavelength of 487 nm with a UV-VIS spectrophotometer against a reference sample of demineralised water. The formed colour complex will be stable for a long time. The samples are measured twice and the mean value is calculated. The amount of polysaccharides is expressed in mg/L.

Proteins

For the analysis of proteins the by Rosenberger (2003) modified method of Frølund *et al.* (1996), based on the method of Lowry et al. (1951) is used. For the calibration Albumin bovine, BSA, (Acros) fraction V, in a concentration range between of 0 – 25 mg/L is used. The concentration can be calculated with the measured extinction and calibration curve.

Reagents

A: 143 mM NaOH and 270 mM Na_2CO_3 in demineralised water
B: 57 mM CuSO4 in demineralised water
C: 124 mM Na_2-tatrate, $C_4H_4Na_2O_6$, or Na-K-tatrate, $C_4H_4NaKO_6$, in demineralised water
D: mixture of reagents A, B en C in the relation of 100:1:1
E: Folin-Ciocalteu phenol reagent 1:2 dilution with demineralised water

Reagents A, B, C en E can be stored unlimited, reagent D has to be prepared daily.

Method

A sample of 5 mL and in addition 7 mL reagent D, is filled in a round tube and mixed in a tube mixer. After that the mixture is stored for 10 minutes at room temperature. Then immediately after addition of 1 mL reagent E the mixture must be mixed fast and powerful because the Folin-Ciocalteu phenol reagent is only for a short time stable in the alkaline

environment. The formation of the colour complex will be finished before starting the measurement. The sample-solution mixtures must be incubated for 45 minutes at room temperature. The adsorption is then measured in a 4 cm cuvett at a wavelength of 750 nm with a UV-VIS spectrophotometer against a reference sample of demineralised water. The formed colour complex will be stable for about 45 – 60 minutes. The samples should be measured twice and the mean value has to be calculated. The amount of proteins is expressed in mg/L.

Materials

- VIS Photo-spectrometer: Milton Roy Spectronic 401
- Mass balance: Mettler AT261 Delta range, range 0 - 200 g ± 0.0001 g
- Tube mixer: Vortex-genie 2, scientific industries
- Glass test tubes: Schott duran 16X160 mm with screw pod
- Plastic test tubes: 25 mL with screw pod
- Pipettes:
  - 200 - 1000 µL ± 5 µL, Finnpipette Digital
  - 1 - 5 mL ± 0.05 mL, Finnpipette Digital
  - 2 - 10 mL ± 0.1 mL, Finnpipette
  - 100 mL ± 0.08 mL, glass pipette, DIN AS, Hirschmann EM Techcolor
- 4 cm quarts or glass cuvett

References

Dubois, M., Gilles, K.A., Hamilton, J.K., Rebers, P.A. and Smith F. (1956). *Colourmetric method for determination of sugars and related substances*. Analytical Chemistry, 28, 350-356.

Frølund, B., Palmgren, R., Keiding, P.H., Nielsen, K. (1996). *Extraction of extracellular polymers from activated sludge using a cation exchange resin.* Water Science and Technology, 30, 8, 1749 – 1758.

Lowry, O.H., Rosebrough, N.J., Farr, A.L. and Randall, R.J. (1951). *Protein measurement with the Folin phenol reagent*. Journal of Biological Chemistry, 193, 265-275.

Rosenberger, S. (2003). *Charkterisierung von belebtem Schlamm in Membranbelebungsreaktoren zur Abwasserreiniging*. PhD thesis, Technical University of Berlin, Berlin, Germany.

Summary

Membrane filtration is one of the main applied advanced treatment technologies to upgrade wastewater treatment plant (WWTP) effluent to a higher quality. By using ultrafiltration membranes a complete removal of suspended solids and bacteria can be guaranteed. But this process also has a major drawback, the so-called membrane fouling, i.e. the decrease of filtration performances because of accumulation of material at the membrane surface. To gain insight in the process of membrane fouling the Specific Ultrafiltration Resistance (SUR) parameter is developed. This parameter allows determination of the filterability of WWTP effluent during dead-end ultrafiltration within a short time (30 minutes).

In this thesis two main research topics can be distinguished; pretreatment and performance. Pretreatment deals with the initial filterability of WWTP effluent before ultrafiltration. A high initial filterability of WWTP effluent is required for stable ultrafiltration performance and therefore WWTP effluent has to be pretreated (coagulation, filtration, etc.). Therefore within this research the application of different pretreatment technologies is investigated. The second topic, performance, deals with the SUR parameter and the operation of ultrafiltration installations. The performance of ultrafiltration installations depends on the filtration characteristics of WWTP effluent but this is not yet really quantified in relation to the SUR value. Therefore experiments were performed with pilot and full scale ultrafiltration installations to gain insight into this relation.

An short overview of the development of wastewater treatment since the mid 20th century in the Netherlands is presented in *chapter 1*. An important recent development is the introduction of advanced treatment technologies i.e. ultrafiltration. To understand this process the ultrafiltration process is explained and described as well. Important terms of the ultrafiltration process are filterability (i.e. increase of filtration resistance over time within a filtration run) and reversibility (i.e. the extent to which the filtration resistance is returned to the original value after applying a hydraulic cleaning) which have been used to evaluate process performance. These parameters were measured on lab (SUR), pilot and full scale. Furthermore in chapter 1 the background and aim (i.e. optimise the process conditions and pretreatment technology of dead-end ultrafiltration) of this thesis are presented.

In *chapter 2* is discussed in more detail the combination of WWTP effluent and membrane filtration. In the literature the soluble microbial products (SMP) and/or (soluble) extracellular polymeric substances are considered as major foulants during ultrafiltration of WWTP effluent. But in addition to these compounds other researchers have shown the major impact of the colloidal fraction 0.1 – 0.2 µm on the filterability of WWTP effluent. Unfortunately the literature also shows these fraction and compounds are hardly removable by conventional pretreatment technologies (filtration, coagulation, etc.). Therefore other pretreatment

technologies (e.g. biofiltration and coagulation followed by adsorption) are suggested in the literature and some of them are investigated in the context of this thesis.

Furthermore different methods to characterize the fouling rate of WWTP effluent are discussed. In this thesis the SUR parameter is used because the process conditions of this measurement are closely related to actual practice.

In *chapter 3* the approach of the research is presented. The research presented in this thesis has been carried out on different locations in the Netherlands. At the WWTP Horstermeer a pilot ultrafiltration installation was operated during several periods. With this installation the relation between the SUR values of ultrafiltration feedwater and the operational flux was determined. Furthermore the effect of two pretreatment technologies (multi media filter and 1 –STEP® filter) was thoroughly investigated on pilot and lab scale (SUR, foulants and fractionation). At the WWTP Maasbommel also the effect of different pretreatment technologies (powdered and granulated activated carbon) has been researched. Experiments with a full scale ultrafiltration installation have been performed at the WWTP Sas van Gent. The operational data of this UF-RO plant were compared with SUR values of the ultrafiltration feedwater. In addition the effect of buffering WWTP effluent in a stabilization pond was determined.

Next to a description of the research at different locations the background of different methods (SUR parameter, foulants analyses and fractionation procedure) were shown in that chapter.

Chapter 4 discusses the relation between the SUR value and two process parameters; operational flux and resistance increase. To qualify process performance the $\Delta R_{max}/t_{min}$ was introduced. This parameter, the maximum rate of filtration resistance increase (1/m·h), is compared with the actual rate of filtration resistance increase between two chemical cleanings. In this thesis a chemical cleaning interval of 24 hours was taken. If the value of the actual rate exceeded the maximum rate, the process performance was qualified as super critical. The performance was qualified as sub critical if the value of the actual rate was below the maximum rate. Based on these criteria it was concluded that for sub critical process performance with fluxes of 40 – 60 L/m^2·h the SUR value of the feedwater should be $\leq 10 \cdot 10^{12}$ m^{-2}. To operate at higher fluxes (60 – 80 L/m^2·h) SUR values below $5 \cdot 10^{12}$ m^{-2} are required for sub critical process performance.

The relation between resistance increase (dR/dt) and the SUR values was determined at the WWTP Sas van Gent. A significant relation was shown, but the slope depends on the condition of the membranes of the ultrafiltration installation i.e. at the same SUR value of fouled membranes showed higher dR/dt values compared to clean membranes.

The objectives of *chapter 5* were (1) to determine the effect of four pretreatment technologies on the filtration properties and concentrations of foulants of WWTP effluent and (2), to

determine in which particle size range most of the particles or colloidal are removed by pretreatment.

Powdered activated carbon was tested on lab and pilot scale. The laboratory tests showed an increase of filterability and decrease of some foulants (humic substances, colour and proteins) after treatment with powdered activated carbon. However, this observation was not confirmed during pilot tests at the WWTP Maasbommel. In this experiment even an decrease of the filterability was observed due to accumulation of the colloidal fraction. This result is inherent to the design of that particular pilot plant. Furthermore, both experiments showed that high amounts of powdered activated carbon were needed to observe significant improvement of the filterability. This observation makes this pretreatment technology less economically less attractive.

Moreover the effect of three prefiltration technologies (granulated activated carbon filter, multi media filter and 1-STEP® filter) was investigated. From all these technologies only the operation of the 1-STEP® filter resulted in a significant removal of the colloidal fraction of 0.1 – 0.2 µm. A clear explanation for this difference cannot be given but it seems that the combination of adsorption and biological activity is responsible for this observation.

In *chapter 5* also the relation between the SUR values and concentrations of foulants (DOC, proteins, polysaccharides, humic substances and colour) was investigated. None of the results showed a significant relation. Consequently it seems that filterability is more related to colloidal particles than to foulants as analysed in this thesis.

In *chapter 6* results in practice are presented of an experiment at the WWTP Sas van Gent. The UF-RO installation at this WWTP has to deal with fluctuating effluent quality and therefore during a period of 2 months the effect of buffering in a stabilization pond was investigated. Based on SUR measurements the results of this experiment showed no beneficial effect of buffering. Next to this experiment the performance of each pretreatment step (coagulation – filtration – coagulation) of the UF-RO installation was investigated. These results showed that the effect of these pretreatment steps depends on the initial SUR value. Around and below a SUR value of $5 \cdot 10^{12}$ m^{-2} none of the pretreatment steps had hardly any effect. At higher SUR values however e.g. $10 \cdot 10^{12}$ m^{-2} the effect became more significant.

Furthermore from the comparison of process data and SUR values of the ultrafiltration feedwater it was concluded that remained fouling of stabilized WWTP effluent can be removed hydraucally and/or chemically.

The influence of two operational conditions (flux and pretreatment) on both filterability and reversibility are presented in c*hapter 7*. As expected an increase of the flux resulted in a decrease of filterability and reversibility but this relation is highly dependent on different aspects like cleaning efficiency, cake compression, initial filtration resistance, etc. The effect of pretreatment was investigated during two pilot experiments. At the same operational flux, 1-STEP® filtrate presented a significant higher filterability and reversibility than multi media

filtrate. This difference could also be related to the SUR values. The SUR values of multi media filtrate and 1-STEP® filtrate were respectively above $7.0 \cdot 10^{12}$ m^{-2} and in the range of $3 \cdot 10^{12}$ m^{-2} – $4 \cdot 10^{12}$ m^{-2}.

To gain a better indication of the relation between filterability and reversibility of WWTP effluent experiments were performed with the newly developed SUR+ equipment. With this equipment and accompanying method it is possible to measure all filtration properties in about 1 hour. The results of the SUR+ confirmed the findings of the pilot scale test. Both, filterability and reversibility are related but the relation depends on the configuration of the membrane module.

In the evaluation chapter (*chapter 8*) all findings of this thesis are discussed and concluded. Overall, it was concluded that the SUR measurement is a useful tool to comprehend and optimize the ultrafiltration process of WWTP effluent. Based on the results of this thesis special attention should be given to pretreatment and cleaning. These two aspects are the key factors to stable ultrafiltration filtration performance of WWTP effluent.

Samenvatting

Door de verwachte toekomstige lokale en wereldwijde waterschaarste wordt RWZI-effluent steeds vaker beschouwd als een bron voor hergebruiktoepassingen, zoals bijvoorbeeld in de industrie of de landbouw. In deze ontwikkeling speelt membraanfiltratie een belangrijke rol. Het onderzoek, beschreven in dit proefschrift, gaat in op de toepassing van ultrafiltratie voor de nabehandeling van RWZI-effluent. Een beperking van dit proces is het vervuilen van de membranen tijdens het filtreren. Door deze vervuiling moeten membranen regelmatig hydraulisch en chemisch gereinigd worden wat onder andere resulteert in productieverlies en verslechtering van membraaneigenschappen. Om inzicht te verkrijgen in het proces van membraanvervuiling is de Specifieke Ultrafiltratie Weerstand (SUR)-parameter ontwikkeld. De SUR wordt op labschaal bepaald gedurende een periode van 30 minuten en het resultaat van de meting geeft een indicatie van de filtreerbaarheid van RWZI-effluent.

Het onderzoek beschreven in dit proefschrift richt zich op twee aspecten van (dead-end) ultrafiltratie van RWZI-effluent: voorbehandeling en performance. Voorbehandeling (coagulatie, filtratie, etc.) is gericht op het verbeteren van de initiële filtreerbaarheid van RWZI-effluent. Een goede initiële filtreerbaarheid van RWZI-effluent is namelijk een voorwaarde voor een stabiele bedrijfsvoering van ultrafiltratie-installaties. Tijdens het onderzoek zijn verschillende voorbehandelingtechnologieën met elkaar vergeleken en beoordeeld op hun prestatie. Het tweede onderwerp van onderzoek, de performance, heeft betrekking op de relatie tussen de SUR en de prestatie van ultrafiltratie-installaties. De performance van ultrafiltratie-installaties is afhankelijk van de filtratie-eigenschappen van RWZI-effluent. Daarom is gekeken of de SUR hier ook een rol in kan spelen. Op proef- en praktijkschaal zijn verschillende experimenten uitgevoerd om inzicht te krijgen in deze relatie.

De ontwikkeling van de behandeling van afvalwater in Nederland sinds het midden van de 20^{e} eeuw is in hoofdstuk 1 beschreven. Een belangrijke, recente ontwikkeling is de steeds verdergaande zuivering van RWZI-effluent met behulp van geavanceerde technologieën, zoals bijvoorbeeld ultrafiltratie. Belangrijke begrippen van het ultrafiltratieproces zijn filtreerbaarheid (toename van filtratieweerstand tijdens een filtratierun) en de reversibiliteit (verwijderbaarheid van membraanvervuiling na een hydraulische reiniging). Beide begrippen zijn in het proefschrift gebruikt voor het evalueren van het ultrafiltratieproces en zijn zowel op labschaal (SUR) als op proef- en praktijkschaal gemeten. Verder zijn in hoofdstuk 1 de achtergrond en de doelstellingen (het optimaliseren van procescondities en voorbehandeling in relatie tot filtreerbaarheid) van dit proefschrift beschreven.

In hoofdstuk 2 wordt de combinatie van RWZI-effluent met membraanfiltratie beschreven. In de literatuur worden de Soluble Microbial Products (SMP) en/of de opgeloste extracellulaire

polymere stoffen beschouwd als de belangrijkste membraanvervuilende componenten tijdens ultrafiltratie van RWZI-effluent. Maar naast deze stoffen hebben andere onderzoekers aangetoond dat met name de colloïdale fractie 0,1 tot 0,2 µm van grote invloed is op de filtreerbaarheid van RWZI-effluent. Echter uit de literatuur blijkt dat juist deze stoffen en de colloïdale fractie nauwelijks worden verwijderd door conventionele voorbehandelings-technologieën als filtratie, coagulatie, etc.. Daarom zijn in dit proefschrift op basis van de literatuur andere voorbehandelingtechnologieën (bijvoorbeeld biofiltratie en coagulatie in combinatie met adsorptie) voorgesteld en onderzocht.

Verder zijn in hoofdstuk 2 verschillende methoden beschreven en bediscussieerd voor het karakteriseren van membraanvervuiling. In dit proefschrift wordt de SUR-parameter gebruikt omdat de procescondities van deze meting aansluiten bij de praktijk.

In hoofdstuk 3 is de aanpak van de verschillende deelonderzoeken beschreven. Op de RWZI Horstermeer is gedurende verschillende perioden onderzoek gedaan met een proefschaal ultrafiltratie-installatie. Met deze installatie is onder andere de relatie bepaald tussen de SUR-waarden van het voedingswater en de operationele flux. Op deze locatie is ook het effect van twee voorbehandelingtechnologieën, het multimediafilter en het 1-STEP® filter, op proef- en labschaal onderzocht. Op de RWZI Maasbommel is ook het effect van verschillende voorbehandelingtechnologieën (poeder- en granulair actieve kool) onderzocht. Experimenten met een praktijkschaal ultrafiltratie-installatie zijn uitgevoerd op de RWZI Sas van Gent. De operationele data van deze praktijkinstallatie werden gerelateerd aan de SUR-waarden van het voedingswater. Daarnaast is op deze locatie het effect van de implementatie van een stabilisatievijver onderzocht op de SUR-waarden en de concentraties van membraanvervuilende stoffen in RWZI-effluent.

Naast de aanpak is in hoofdstuk 3 de achtergrond beschreven van de verschillende meet- en analysemethoden (SUR-parameter, fractionering en analyses membraanvervuilende stoffen).

De relatie tussen de SUR-waarden en de performance van ultrafiltratie-installaties is beschreven in hoofdstuk 4. Om de performance te kwalificeren is hier de $\Delta R_{max}/t_{min}$ geïntroduceerd. Deze parameter, de maximaal toegestane snelheid van de filtratieweerstands-toename (1/m·h), is vergeleken met de werkelijke snelheid van de filtratieweerstandstoename tussen twee chemische reinigingen. In dit proefschrift is het tijdsinterval tussen twee chemische reinigingen 24 uur. Wanneer de waarde van de werkelijke snelheid de maximale snelheid overschreed, werd het performance gekwalificeerd als ‘super critical’. De performance werd als ‘sub critical’ gekwalificeerd indien de waarde van de werkelijke snelheid lager was dan de maximale snelheid. Op basis van deze criteria is geconcludeerd dat voor een ‘sub critical’ performance met fluxen van 40 – 60 $L/m^2 \cdot h$ de SUR-waarde van het voedingswater $\leq 10 \cdot 10^{12}$ m^{-2} moet zijn. Voor hogere fluxen (60 – 80 $L/m^2 \cdot h$) zijn SUR-waarden lager dan $5 \cdot 10^{12}$ m^{-2} vereist voor ‘sub critical’ performance.

De relatie tussen de weerstandstoename (dR/dt) en de SUR-waarden is onderzocht op de RWZI Sas van Gent. Een significante relatie werd aangetoond. Deze relatie bleek echter afhankelijk te zijn van de conditie van de membranen in de ultrafiltratie-installatie. Vervuilde membranen lieten bij dezelfde SUR-waarden hogere dR/dt-waarden zien dan bij schonere membranen.

De doelstellingen van hoofdstuk 5 zijn (1) het vaststellen van het effect van vier voorbehandelingtechnologieën wat betreft filtratie-eigenschappen en concentraties membraanvervuilende stoffen en (2) het bepalen welke grootte van de deeltjes en/of colloïden worden verwijderd of omgezet door voorbehandeling.

Actieve poederkool werd getest op lab- en proefschaal. De laboratoriumexperimenten toonden een toename van de filtreerbaarheid en een vermindering van bepaalde membraanvervuilende stoffen aan (humusachtig stoffen, kleurhoudende componenten en proteïnen) na behandeling met actieve poederkool. Deze waarneming werd echter niet bevestigd door een proefschaalexperiment op de RWZI Maasbommel. Tijdens dit experiment werd een daling van de filtreerbaarheid waargenomen door ophoping van de colloïdale fractie. Dit resultaat bleek inherent te zijn aan het ontwerp van de ultrafiltratie-installatie. Daarnaast toonden zowel de lab- als proefschaalexperimenten aan dat hoge concentraties actieve poederkool nodig zijn voor een verbetering van de filtreerbaarheid. Dit maakt deze voorbehandelingtechnologie economisch gezien minder aantrekkelijk.

Ook het effect van drie filtratietechnologieën (granulair actieve kool, multimediafilter en 1-STEP® filter) werd onderzocht en beschreven in hoofdstuk 5. Van de drie filtratietechnologieën lieten alleen de resultaten van het 1-STEP® filter een aanzienlijke verwijdering van de colloïdale fractie van 0,1 - 0,2 μm zien. Een duidelijke verklaring kan hier niet voor worden gegeven, maar het lijkt erop dat de combinatie van adsorptie en de biologische activiteit verantwoordelijk is voor dit resultaat.

In hoofdstuk 5 is ook de relatie tussen de SUR-waarden en de concentraties van membraanvervuilende stoffen (Dissolved Organic Carbon, proteïnen, polysacchariden, humusachtige stoffen en kleurhoudende componenten) onderzocht. Geen van de relaties toonde een significant verband. Het lijkt erop dat de filtreerbaarheid meer verwant is aan colloïdale deeltjes dan aan membraanvervuilende stoffen zoals geanalyseerd is in dit proefschrift.

In hoofdstuk 6 zijn de resultaten van praktijkproeven op RWZI Sas van Gent gepresenteerd. De UF-RO-installatie op deze RWZI had te maken met een sterk fluctuerende kwaliteit van het effluent. Om deze reden is gedurende een periode van 2 maanden het bufferend effect van een stabilisatievijver onderzocht door middel van SUR-metingen. In tegenstelling tot de verwachtingen was er echter geen gunstig effect van buffering door onder andere algengroei in de stabilisatievijver. Naast dit experiment is de prestatie van elke voorbehandelingstap (coagulatie - filtratie - coagulatie) van de UF-RO-installatie onderzocht. Deze resultaten

toonden aan dat het effect van de voorbehandeling afhankelijk is van de initiële SUR-waarde. Beneden een SUR-waarde van $5 \cdot 10^{12}$ m^{-2} bleek de voorbehandeling nauwelijks effect te hebben. Bij hogere SUR-waarden ($\geq 10 \cdot 10^{12}$ m^{-2}) was het effect van de voorbehandeling echter groter.

Verder bleek uit een vergelijking van de procesdata en de SUR-waarden dat de achtergebleven vervuiling van het gestabiliseerd RWZI-effluent hydraulisch en/of chemisch kon worden verwijderd.

De invloed van flux en voorbehandeling op de filtreerbaarheid en reversibiliteit is gepresenteerd in hoofdstuk 7. Zoals verwacht resulteerde een toename van de flux in een daling van de filtreerbaarheid en de reversibiliteit. Deze relatie is echter sterk afhankelijk van verschillende aspecten, zoals de efficiëntie van reiniging, cakecompressie, initiële membraanweerstand, etc.. Het effect van voorbehandeling is onderzocht tijdens twee proefschaalexperimenten. Filtratie van 1-STEP® filtraat, bij dezelfde operationele flux, resulteerde in een betere filtreerbaarheid en reversibiliteit dan multimediafiltraat. Dit verschil kan ook worden gerelateerd aan de SUR-waarden. De SUR-waarden van multimediafiltraat en 1-STEP® filtraat waren respectievelijk $\geq 7 \cdot 10^{12}$ m^{-2} en $3 \cdot 10^{12}$ m^{-2} – $4 \cdot 10^{12}$ m^{-2}.

Om een betere inzicht te krijgen in de relatie tussen filtreerbaarheid en reversibiliteit zijn experimenten uitgevoerd met de nieuw ontwikkelde SUR+-opstelling. Met deze opstelling en de bijbehorende meetmethode is het mogelijk om beide filtratie-eigenschappen van RWZI-effluent in ongeveer 1 uur te bepalen. De resultaten van de SUR+-metingen bevestigden de bevindingen van de proefschaalexperimenten. Filtreerbaarheid en reversibiliteit zijn verwant, maar afhankelijk van de configuratie en terugspoelsnelheden.

In het laatste hoofdstuk zijn alle bevindingen van dit proefschrift geëvalueerd en samengevat. In het algemeen kan worden geconcludeerd dat de SUR-meting een nuttig instrument is voor het begrijpen en optimaliseren van het ultrafiltratieproces van RWZI-effluent. In het bijzonder moet aan de voorbehandeling en reiniging aandacht worden besteed. Deze aspecten zijn belangrijk voor een stabiele bedrijfsvoering van ultrafiltratie-installaties.

Curriculum Vitae

Arie Janssen was born on November 25th, 1976 in Lisse, The Netherlands. After completing secondary education (HAVO) at the Driestar College in Gouda he studied Environmental Science at the Hogeschool Delft (nowadays Hogeschool Inholland). After this study he started his study MSc.-study Environmental Hygiene at the Wageningen University. He finished his studies at the Sub-department of Environmental Technology with a master thesis, entitled 'The possibilities for application of oligochate worms in wastewater treatment with a membrane bioreactor'.

In 2003 he started as a environmental researcher at the Lettinga Associates Foundation (LeAF) in Wageningen. His work concerned research on sludge reduction by anaerobic and physical-chemical pretreatment of municipal wastewater. In 2004 he started his PhD research within the research group of prof.ir. J.H.J.M. van der Graaf at the department of Sanitary Engineering, within the project 'Ultrafiltration of WWTP effluent'. The results of this research are described in this thesis. In 2008 he joined Evides Industriewater as a process engineer and was mainly involved in two projects dealing with treatment and reuse of WWTP effluent and desalination of seawater. At the moment he is in between jobs.

List of publications

2005

Poele, S. te and Janssen, A.N. (2005). *Membraan filtratie van RWZI-effluent.* Delft, PAO course "Op weg naar nieuw effluent".

2006

Janssen, A.N., Thomas, M., Koning, J. de and Graaf, J.H.J.M. van der (2006). Modification of filtration characteristics by PAC addition during dead-end ultrafiltration of WWTP effluent. *Young Researchers 2006,* 77 – 84. London: IWA publishing.

Menkveld, H.W.H., Nieuwenhuijzen, A.F. van and Janssen, A.N. (2006). *Membraanfiltratie van RWZI-effluent de schakel naar hergebruik.* DHV, Amersfoort, Nationaal Watersymposium 2006.

Janssen, A.N. and Menkveld, H.W.H. (2006). *Ultrafiltratie RWZI-effluent is sterk te verbeteren*. Land + water, *46* (6/7), 20 – 21.

2007

Janssen, A.N. and Broek, W.B.P. van den (2007). *Membraanfiltratie van effluent op praktijkschaal.* PAO course ”Op weg naar KRW-effluent.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den, Koning, J. de, Menkveld, H.W.H., Schrotter, J-C., Teeuw, H. and Graaf, J.H.J.M. van der (2007). *Monitoring of SUR to control and enhance the performance of ultrafiltration installations treating WWTP effluent.* Proceedings Membranes in Drinking and Industrial Water Treatment, Harrogate, United Kingdom.

Janssen, A.N. and Graaf, J.H.J.M. van der (2007). *Dead-end ultrafiltration of wwtp effluent: pretreatment and performance.* The 2nd Workshop on Innovation of Membrane Technology for Water and Wastewater Treatment. Sapporo, Japan.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den and Graaf, J.H.J.M. van der (2007). *Evaluation of two treatment concepts for the reuse of WWTP effluent at an UF-RO plant*. Proceedings of 6th IWA Specialist Conference on Wastewater Reclamation and Reuse for Sustainability, Antwerp, Belgium

Janssen, A.N. and Broek, W.B.P. van den (2007). *Membraanfiltratie van effluent op praktijkschaal.* Afvalwaterwetenschap, 6 (4), 252 – 266.

2008

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. and Graaf, J.H.J.M. van der (2008). *Is buffering the answer to overcome the fluctuation at an UF-RO plant.* Proceedings of WEF conference Membrane Technology 2008, Atlanta, Georgia, USA.

Janssen, A.N., Kooistra, R.A. and Graaf, J.H.J.M. van der (2008). *SUR+: a short and accurate test to monitor the total fouling rate of WWTP effluent during dead-end ultrafiltration.* Proceedings of WEF conference Membrane Technology 2008, Atlanta, Georgia, USA.

Janssen, A.N. and Scherrenberg, S.M. (2008). *Verslag van de zesde IWA conferentie 'guiding the grownth of water reuse'.* Afvalwaterwetenschap, 7, 2, 134 – 137.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den, Geilvoet, A.J., Menkveld, H.W.H., J Schrotter, J-C and Graaf, J.H.J.M. van der (2008). *Prefiltration of wastewater effluent: Effects on foulants and performance during dead-end ultrafiltration.* Proceedings Membranes in Drinking and Industrial Water Treatment, Toulouse, France.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den, Koning, J. de, Menkveld, H.W.H., Schrotter, J-C., Teeuw, H. and Graaf, J.H.J.M. van der (2008). *Monitoring of SUR to control and enhance the performance of ultrafiltration installations treating WWTP effluent.* Desalination, 231, 1-3, 99 – 107.

2009

Scherrenberg, S.M., Kloeze, A. M. te, Janssen, A. N., Nieuwenhuijzen, A. F. van, Menkveld, H. W. H., Bechger, M. and Graaf, J. H. J. M. van der (2009). *Advanced treatment of WWTP effluent with filtration leading to a pretreatment technique for membrane filtration*. Proceedings of 7th IWA world congress on water reclamation and reuse, Brisbane, Australia.

2010

Schurer, R., Janssen, A.N., Villacorte, L. and Kennedy, M. (2010). *Evides Pilot Seawater Desalination: efficacy of coagulant in ultrafiltration pre-treatment in integral UF-RO membrane desalination.* Proceedings Membranes in Drinking and Industrial Water Treatment, Trondheim, Norway.

Janssen, A.N., Agtmaal, J. van, Broek, W.B.P. van den, Geilvoet, A.J., Menkveld, H.W.H., J Schrotter, J-C and Graaf, J.H.J.M. van der (2010). *Prefiltration of wastewater effluent: Effects on foulants and performance during dead-end ultrafiltration.* Desalination, 250, 2, 855 – 860.

Scherrenberg, S.M., Kloeze, A. M. te, Janssen, A. N., Nieuwenhuijzen, A. F. van, Menkveld, H. W. H., Bechger, M. and Graaf, J. H. J. M. van der (2010). *Advanced treatment of WWTP effluent with filtration leading to a pretreatment technique for membrane filtration*. Water Science and Technology, 62, 9, 2083 – 2089.