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Environmentally friendly treatment of highly potent pharmaceuticals in hospital wastewater - Mermiss

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Preface

The initial purpose of the project was to develop a treatment technology for biological removal of pharmaceuticals in hospital wastewater based on a biofilm solution rather than the conventional approach with activated sludge. The advantage of the biofilm approach is that slow growing bacteria can be maintained in the wastewater systems as they can form protected biofilms on carrier material, typically made of plastic. Furthermore, biofilm systems can be designed for proliferation of bacteria utilizing different substrates available in the wastewater by having multiple reactors in series where each reactor maintains its own bacteria communities.

This approach is termed moving bed biofilm reactor principle (MBBR), which in earlier published studies have been investigated in terms of removal of pharmaceuticals. The ability of MBBR to degraded pharmaceuticals has been investigated in a number of studies that all identify MBBR as superior compared to activated sludge in degrading pharmaceuticals in full scale wastewater treatment plants (Falås et al., 2012 a,b, Vieno and Sillanpää, 2014; Zupanc et al., 2013). Therefore, an initial study tested and optimized the staged MBBR approach on a side stream originating from Dep. of Oncology. Only aerobic treatment was carried out in the raw wastewater from the department. Three MBBR operating in series with a total volume of 9 liters were operated for promoting bacteria specialized in degradation of pharmaceuticals. The results generated here formed the basis of this project as “proof–of-concept” and have been published in Casas et al., 2015a and in a Danish report (Kragelund et al., 2014,).

The present project investigated the potential of the MBBR technology in detail; MBBR alone or in combination with activated sludge termed HYBAS™ (a type of IFAS= `Integrated fixed film in activated sludge´); and by examining different types of wastewater containing pharmaceuticals e.g. hospital wastewater as well as conventional municipal wastewater. Bench scale plants (liter scale) and pilot-scale plants (m^3) have been operated at the above-mentioned test sites, with continuous feeding for six to nine months at each location to ensure fully adapted biology in relation to the actual wastewater. Besides the optimized biological treatment step, also chemical polishing by ozonation was applied in pilot-scale.

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- Aarhus University Hospital: Thomas Moeller
- Krüger Veolia: Christina Sund, & Kim Sundmark
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Introduction

In Denmark and other European countries, there has been an increasing focus on the presence of pharmaceuticals in the surrounding environment originating from wastewater discharge. Different actions have been initiated across Europe and Switzerland currently the only country regulating pharmaceuticals and micropollutants in their wastewater. They have ambitious goals to remove in total 50% of micropollutants by upgrading 1/7 of all wastewater plants using activated carbon and ozonation (Mulder et al., 2015).

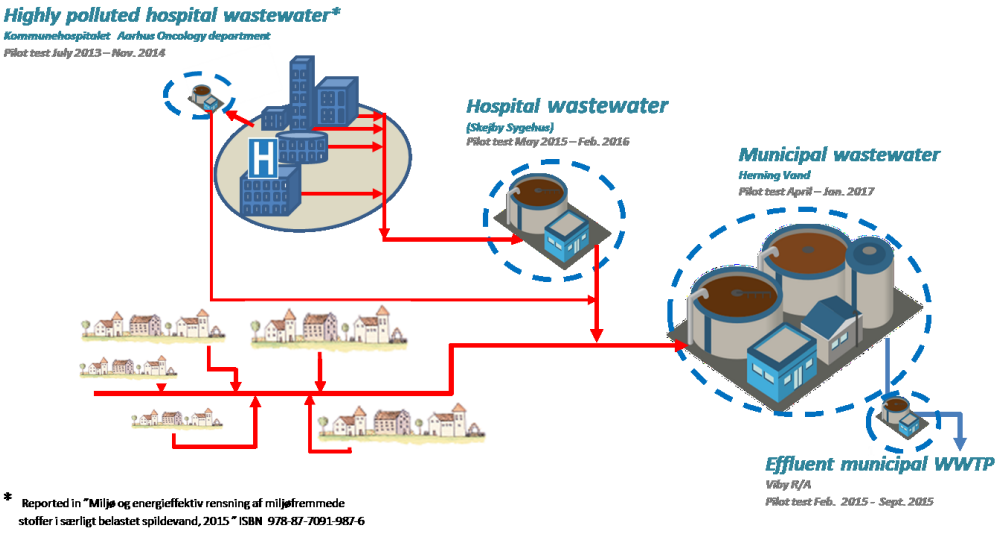
In Denmark, several studies have been conducted in order to map the pharmaceuticals, especially in hospital wastewater discharge. Comprehensive mappings of pharmaceuticals used at different Danish hospitals have been subject to investigation since 2000 and onwards. One of the early findings, based on mapping of pharmaceuticals, revealed that approx. 1-4% of the entire consumption of pharmaceuticals took place at the hospital; the remaining part was consumed in the private sector (Mose-Pedersen, 2007). Based on these mappings, hospitals were identified as point source polluters due to the discharge of pharmaceuticals in wastewater, and therefore hospitals are proposed to be regulated in line with industries. To regulate hospitals as point sources, a task group established by the Association of Local Governments Denmark (KL) proposed a guideline. The list containing guiding limit values for 36 pharmaceuticals was published in 2013 (Local Government Denmark (KL), 2013). Pharmaceuticals present on this list were selected according to their toxic effect on bacteria, algae, crustacean, and fish etc., and since 2015, the list has expanded to include 40 pharmaceuticals in total (AMK, 2015). Present regulation for discharge of municipal wastewater treatment plant (WWTP) or the environment does not include pharmaceuticals.

Different strategies for removal of pharmaceuticals from wastewater have been applied, and several large projects have been conducted such as Pills, MistraPharma. The primary choice of technology applied in those projects was MBR technology (activated sludge systems coupled with ultrafiltration membranes) and/or chemical oxidation methods like ozonation s well as removal by absorption on activated carbon to reduce discharge to environment. The first full scale MBR plants were commissioned in Germany in 2011 (EU Project Pills, Marienhospital Gelsenkirchen, Germany) and the Netherlands (EU Project SLIK/Pills, http://www.pills-project.eu/). In Denmark, one MBR plant was built in 2014 with ozone and activated carbon as post treatment (Grundfos Biobooster, 2016). In Switzerland, the 100 biggest municipal WWTPs will be equipped with an effluent polishing step with ozone/activated carbon treatment to reduce discharge to the environment (Adriano Joss, EAWAG, Implementation period 2016 to 2040).

The present project investigated the potential of the MBBR technology in detail; MBBR alone or in combination with activated sludge termed HYBAS™ (a type of IFAS= `Integrated fixed film in activated sludge´); and by examining different types of wastewater containing pharmaceuticals, e.g. hospital wastewater as well as conventional municipal wastewater. Bench scale plants (liter scale) and pilot scale plants (m^3) have been operated at the above-mentioned test sites with continuous feeding for six to nine months at each location to ensure fully adapted biology in relation to the actual wastewater. Besides the optimized biological treatment step, chemical polishing by ozonation was also applied in pilot scale.

Summary

The aim of this project was to assess a biofilm-based technology for removal of pharmaceuticals from different wastewater types, e.g. toxic side streams, the entire hospital wastewater stream and municipal wastewater. The biofilm technology applied was the Moving Bed Biofilm Reactor (MBBR) principle, where biofilm formation is favored on plastic carrier material, which promotes slow growing microorganisms that are maintained in the treatment system. Both MBBR alone and in combination with activated sludge, termed HYBAS™, were investigated. HYBAS ™ is a type of Integrated Fixed Film and Activated Sludge Process developed by Krüger, Veolia. The figure below provides an overview of bench scale (liter scale) and pilot scale experiments (m^3) conducted on site for the different tests. Bench scale and pilot scale test sites were operated between 6-11 months on site on the locations illustrated below. The design and operation of the different bench-scale and pilot-scale plants is described in chapter 1.



Overview of the different test locations where wastewater discharge has been treated for pharmaceuticals

During the project, a comprehensive investigation on actual pharmaceutical consumption in one of larger hospitals was carried out, for details please see chapter 3. The outcome of the investigation confirmed that the majority of the environmentally critical pharmaceuticals defined by the Danish task force were in fact consumed in the private sector rather than at the hospital (Møller, Environmental Report, Aarhus University Hospital, 2014). Six out of all investigated compounds with the biggest environmental impact (accounting for more than 84%) could be linked to the private sector. These data emphasize that most of the pharmaceuticals are excreted from private homes and are therefore present in the wastewater to the local municipal WWTP.

In the project, the emphasis was put on promoting as high removal of medium-biodegradable and hardly-degradable pharmaceuticals as possible. The plants had also a high removal of nitrogen; in the bench scale plants nitrification and in the pilot scale plants both nitrification and denitrification. No chemical phosphorous removal was conducted and the technology for complete removal is well known and therefore not of interest in this project. Different wastewater parameters were investigated in bench-scale and pilot-scale plants, which are listed in chapter 1. Technology performance is described in chapter 4 and 5.

To evaluate the performance of the different treatment technologies: Hybas™ (IFAS type process), MBBR and CAS, two types of pharmaceutical degradation experiments were carried out. The first degradation experiment documented the removal potential of the system due to spiking of low concentrations of selected pharmaceuticals. The degradation was then monitored over a 24-hour period. The second type of experiment revealed the actual removal capacity at a given day by studying the removal of native pharmaceuticals in the treated wastewater, i.e. bulk water was followed through the pilot plant according to actual HRT (Escolar et al., 2015 a,b, Tang et al., 2017). This is further elaborated in chapter 2.

For each experiment, the presence of different pharmaceuticals was analyzed, and profiles of measured removals by biological and chemical treatment were established. The results were divided into biological removal as the main project goal was to develop and enhanced biological removal and secondly the results obtained from chemical oxidation by ozone. Ozone dose could be minimized as a consequence of efficient biological removal. More than 27 pharmaceutical compounds were investigated, and many of these compounds are present on the Danish guiding limit value list for pharmaceuticals in wastewater discharged from hospitals (Local Government, 2013, AMK, 2015). To improve readability of this report, only a selection of the most important pharmaceuticals is presented. The comprehensive investigations either have been published (Casas et al., 2015a, b and Tang et al., 2017) or are expected to be submitted during the year under Tang et al., 2018 a, b and Ooi et al., 2018 a, b.

The outcome of this project emphasized the superiority of biofilms for degrading pharmaceuticals, regardless of treatment location. It was documented that promoting and maintaining bacteria specialized in degradation of especially medium-degradable and hardly degradable compounds like diclofenac, were possible and even to degrees not observed before (Tang et al., 2017). The efficient biological removal significantly influenced the subsequent ozone dose required for residual pharmaceutical concentrations. By investigating several different locations, a benchmarking could be carried out including degradation capabilities and operation and maintaining costs, see Table below. In the table, no costs for ozonation of the wastewater have been included in the alternative `Treatment at municipal WWTP´. Please see chapter 6 and 7 for a more detailed calculation and estimates for the MBBR polishing.

Overview of financial costs for removal of pharmaceuticals directly at the hospital and at the municipal WWTP.

Location	**Treatment at hospital**		**Treatment*** at municipal WWTP**	
		DKK/m^3 wastewater		**DKK/m^3 wastewater**
Water volume/year	150,000 m^3		12-15 million m^3	
Plant Costs	21-26 million DKK		26-30 million DKK (excl. ozonation)	
Total consumables*	405,000 DKK/ year	2.70 - 3.25	900,000** DKK/ year	
Analysis	100,000 DKK/ year	0.67	150,000 DKK/ year	
Maintenance plant	575,000 DKK/ year	4.35	600,000 DKK/ year	
Operation	150,000 DKK/ year	1.00	200,000 DKK/ year	
Total OPEX	1,230,000 DKK/ year	**8.20 - 8.70**	**1,850,000 DKK/ year**	**0.21**

* Electricity (0.75 DKK/kWh), Chemicals, Ozone, Screenings and Sludge

** MBBR (mainly electricity)

*** MBBR post treatment of municipal WWTP effluent

The costs of a decentralized solution at a Danish medium-sized hospital are 8.2-8.7 DKK/m^3, which is significantly less per m^3 treated wastewater compared to the MBR solution built in Denmark (Grundfos Biobooster, 2016).

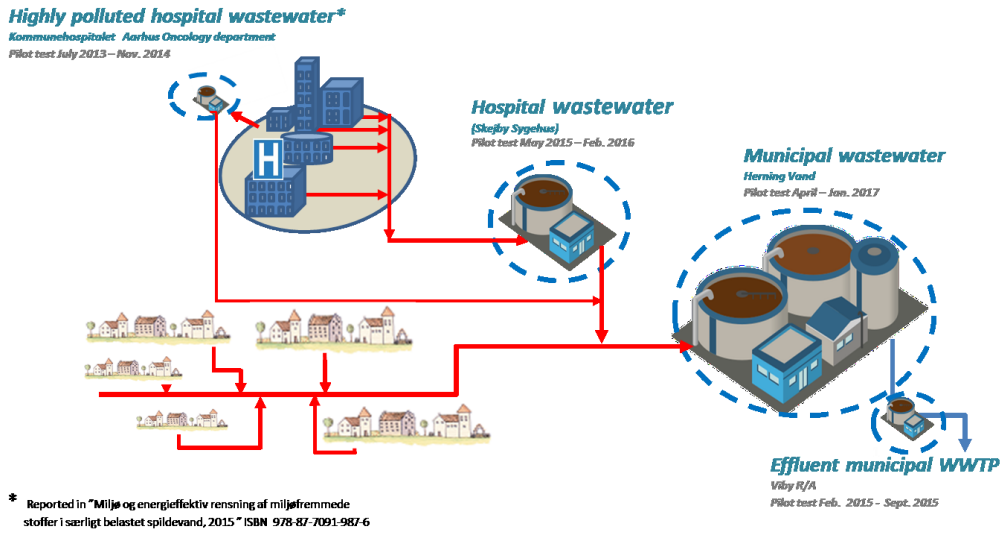
However, only a minor fraction of the consumed pharmaceuticals in Denmark are targeted by this decentralized treatment solution (estimates between 1-4 %, Mose-Pedersen et al., 2007). In addition, the pharmaceuticals identified as problematic for the environment (Local Government Denmark (KL), 2013, AMK 2015) are in fact discharged from private homes rather than from the hospitals (Møller, Environmental Report, 2014, this report). Therefore, it is necessary to rethink how to remove pharmaceuticals from wastewater where the majorities of the pharmaceuticals are present. Here the most logical treatment site would be as posttreatments at the municipal wastewater treatment plants.

The new innovative polishing MBBR solution tested at the municipal WWTP in Viby showed promising perspectives, as the costs for treatment were rather low, and a very efficient biological removal of pharmaceuticals was achieved. By treating both hospital wastewater and household wastewater with the above-mentioned polishing technology, the pharmaceuticals discharged in the wastewater will be degraded regardless origin. However, much larger water volumes are treated by the municipal polishing solution and thereby much larger amounts of pharmaceuticals (as concentrations of pharmaceuticals do not considerably differ) are prevented from reaching the aquatic environment. The costs pr. m^3 for treating wastewater with the MBBR polishing technology were low, so it would still be a feasible solution even with a larger wastewater volume. It is expected that the MBBR polishing can be further optimized (in terms of feeding regimes, HRT etc.), and there is a need for upscaling the process from bench-scale to at least pilot-scale considering the size of potential full scale facilities. Therefore, from an environmental point of view, degradation of pharmaceuticals should be carried out centrally at municipal WWTP rather than at the point sources alone. Centralized removal of micropollutants is conducted in Switzerland, which is considered to be the leader within this area.

Resume

Målet med dette projekt har været at anvende en biofilmbaseret teknologi til fjernelse af lægemiddelstoffer fra forskellige typer af spildevand (både ubehandlet spildevand og som polering af behandlet spildevand), herunder toksiske delstrømme fra hospitaler, samt råspildevand fra hospitaler og forsyningernes rensningsanlæg. Til formålet er anvendt den biofilmbaserede teknologi MBBR (*Moving Bed Biofilm Reactor*), som er baseret på dannelsen af biofilm på bæremedier (såkaldte carriers), hvilket fordrer at langsomt voksende mikroorganismer kan forblive i systemet.

Projektet har fokuseret på dels at teste MBBR teknologien alene og dels at kombinere teknologien med aktivt slam i den såkaldte HYBAS^{TM} teknologi, som er udviklet af Krüger, Veolia, og er baseret på den såkaldte fastfilm og aktiv slamproces. Figuren neden for giver et overblik over testlokaliteterne for de gennemførte laboratorie- (liter) og pilotskalatests (m3). Begge testtyper var af 6-11 måneders varighed og foregik på de testlokaliteter, som er illustreret på figuren nedenfor. Design, anlægskonfiguration og drift af laboratorie- og pilotanlæg er beskrevet i kapitel 1.



Overblik over testlokaliteterne for de gennemførte laboratorie- og pilotskalatests.

Gennem projektet er der blevet foretaget en grundig undersøgelse af det faktiske forbrug af lægemiddelstoffer på et af de større hospitaler i Danmark (se i øvrigt kapitel 3). Resultatet af denne undersøgelse bekræftede formodningen om, at størstedelen af de miljøkritiske lægemiddelstoffer defineret af den danske arbejdsgruppe under KL, rent faktisk indtages i de private hjem fremfor på hospitalerne (Møller, Environmental Report, Aarhus University Hospital, 2014). Ud af alle de undersøgte lægemidler, kunne seks lægemidler kobles til den private sektor svarende til mere end 84% af den samlede miljøbelastning oprindeligt tilskrevet hospitaler. Disse data viser med al tydelighed at størstedelen af lægemidlerne udskilles fra private hjem og dermed ledes til de kommunale renseanlæg, hvor behandlingen oftest ikke er målrettet denne type komponenter.

I projektet har der været fokus på at opnå så høj en fjernelsesgrad som muligt af de middelsvært-nedbrydelige og svært-nedbrydelige lægemiddelstoffer. I de testede anlæg blev der samtidig opnået en god reduktion af kvælstof ved hhv. nitrifikation i laboratorieskala og nitrifi-

kation samt denitrifikation i pilotanlæg. Kemisk fosforfjernelse blev ikke benyttet i testopstillingerne, og idet metoder til komplet fosfor fjernelse allerede er velkendte, blev der ikke fokuseret her på i nærværende projekt. Både i laboratorie- og pilotskala blev der analyseret for en vifte af spildevandsparametre. Disse fremgår af kapitel 1, mens effekten af renseteknologiernes er beskrevet i kapitel 4 og 5.

Med udgangspunkt i to typer af test, der begge var baseret på omsætningen af lægemidler, blev effekten af de forskellige vandbehandlingsteknologier - HYBAS^{TM} (baseret på IFAS), MBBR og CAS (konventionel aktiv slam anlæg) – evalueret. Den første type af test havde til formål at dokumentere omsætningspotentialet af udvalgte lægemiddelstoffer, som i testforløbet blev tilført i lave koncentrationer, hvorefter fjernelsesgraden blev undersøgt over en 24 timers periode. Den anden type test fokuserede på den faktiske fjernelsesgrad (dvs. aktuelle lægemidler tilstede i spildevand på den pågældende dag) baseret på den observerede fjernelse af disse stoffer fra behandlet spildevand. Begge typer af test er yderligere beskrevet i kapitel 2.

Projektet havde først og fremmest til formål at udvikle og optimere den biologiske omsætning af lægemiddelstoffer og dernæst også optimere den kemiske efterpolering ved brug af ozon. Tilstedeværelsen af forskellige lægemiddelstoffer blev således analyseret i hver test og fjernelsesgrader ved hhv. biologisk og kemisk behandling blev bestemt. Resultaterne viste, at ved at forbedre den biologiske rensning ift. lægemiddelstoffer kunne doseringen af ozon reduceres i den efterfølgende kemiske behandling, så at det rensede spildevand kunne overholde de foreslåede grænseværdier (AMK, 2015).

Mere end 27 lægemiddelstoffer blev undersøgt og mange af disse lægemidler er at finde på den danske liste over lægemidler som ønskes begrænset i spildevand fra hospitaler (Local Government, 2013, AMK, 2015), men i denne rapport er der dog kun præsenteret et udvalg af disse. Resultater fra alle undersøgte lægemidler i dette projekt er enten blevet publiceret (Casas et al., 2015a, b and Tang et al., 2017) eller forventes publiceret inden for det næste år (Tang et al., 2018 a, b og Ooi et al., 2018 a, b).

Resultaterne af projektet understreger gevindsten ved at benytte biofilm til nedbrydning af lægemidler uanset, hvor rensningen finder sted. Endvidere dokumenterede projektet, at det var muligt at favorisere væksten og bibeholde tilstedeværelsen af bakterier, som er specialiserede i omsætningen af middelsvært-nedbrydelige og svært-nedbrydelige komponenter som eksempelvis diclofenac (Tang et al., 2017).

Den effektive biologiske rensning havde desuden en signifikant effekt ift. at reducere det nødvendige forbrug af ozon til fjernelse af de sidste medicinrester. Ved at undersøge flere forskellige spildevandsstrømme var det muligt at lave en benchmarking af omsætningsgraderne samt omkostninger til drift og vedligehold af denne type anlæg, se tabel 1 nedenfor. I tabellen er omkostningerne til ozonering ikke inkluderet i ”rensning på kommunalt renseanlæg”. Se i øvrigt kapitel 6 og 7 for flere detaljer.

Oversigt over omkostningerne forbundet med fjernelsen af lægemidler hhv. direkte ved hospitalet og på det kommunale rensningsanlæg.

Location	Rensning direkte ved hospital		Rensning*** på kommunalt rensningsanlæg	
		DKK/m^3 spildevand		DKK/m^3 spildevand
Vandvolumen pr. år	150.000 m^3		12-15 millioner m^3	
Etablerings-omkostninger	21-26 millioner DKK		26-30 millioner DKK (u/ ozonering)	
Drifts-omkostninger*	405.000 DKK/ år	2.70 - 3.25	900.000** DKK/ år	
Analyser	100.000 DKK/ år	0.67	150.000 DKK/ år	
Vedligeholdelse af renseanlæg	575.000 DKK/ år	4.35	600.000 DKK/ år	
Mandetimer	150.000 DKK/ år	1.00	200.000 DKK/ år	
Total OPEX	1.230.000 DKK/ år	**8.20 - 8.70**	**1.850.000 DKK/ år**	**0.21**

* Elektricitet (0.75 DKK/kWh), Kemikalier, Ozon, Screeninger and Slam

** MBBR (primært elektricitet)

*** MBBR som efterpolering på kommunalt rensningsanlæg

Omkostningerne forbundet med en decentral renseløsning til behandling af spildevand fra et dansk mellemstort hospital beløber sig til 8,2-8,7 DKK/m^3, hvilket er betydeligt mindre per m^3 behandlet spildevand sammenlignet med et dansk MBR anlæg i Danmark (Grundfos BioBooster, 2016).

Dog fanges kun en mindre fraktion af lægemiddelforbruget der indtages i Danmark, ved denne decentrale behandlingsløsning (estimeret medicinforbrug på hospitaler 1-4 %, Mose-Pedersen et al.,2007). Dertil kommer at de lægemidler der er identificeret som værende problematiske for miljøet (Local Government Denmark (KL), 2013, AMK 2015) udskilles fra de private hjem fremfor på hospitalerne (Møller, Environmental Report, 2014, this report). Derfor er det nødvendigt at gentænke hvor lægemidler skal fjernes fra vores spildevand, nemlig de steder hvor der er det største forbrug, her vil det mest logiske sted være central behandling på renseanlæggene som efterpolering.
Den innovative MBBR poleringsteknologi, er blevet testet ved Viby Renseanlæg og viste spændende perspektiver, da kostprisen for behandling var lav, grundet den meget effektive biologiske fjernelse af lægemidler. Ved at behandle både hospitalsspildevand og husholdningsspildevand med førnævnte teknologi, vil lægemidlerne nedbrydes uanset om de stammer fra hospitalets spildevand eller fra forsyningens spildevand. Dog behandles større volumener af spildevand ved poleringsteknologien hos en forsyning, og derved undgås større koncentrationer af lægemidler havner i det akvatiske miljø (da koncentrationen af visse miljøkritiske lægemidler stort set ikke varierer). Prisen pr. m3 behandlet spildevand med MBBR poleringsløsningen var så lav, at det stadig skønnes en mulig løsning selvom det kræver behandling af større mængder af spildevand. Det forventes at MBBR løsningen kan optimeres yderligere (ift. regenereringstider og HRT) og der er behov for at hele processen opskaleres fra laboratorieskala, til mindst pilot skala størrelse der kan relateres til en fuldskala løsning. Set fra et miljømæssigt perspektiv skal fjernelsen af lægemidler fra spildevandet derfor foregå centralt på forsyningernes renseanlæg fremfor hos punktkilderne alene. Centrale løsninger til fjernelse af mikroforureningsstoffer håndteres centralt i Schweiz, som betragtes som et af de førende lande inden for fjernelse af lægemidler fra spildevandet.

1. Identification of test-sites and design of bench-scale and pilot-scale treatment

1.1 Development of treatment concept

For degradation of difficult biodegradable organic compounds biofilm offer an important advantage compared to activated sludge systems, as the process train can be designed for isolated growth of bacteria mainly proliferating on difficult degradable compounds. The MBBR (Moving Bed Bio Reactor) technology is based on the biofilm principle with an active biofilm growing on small specially designed plastic carriers that are kept suspended in the reactor. The carriers are designed both to provide a large protected surface area for the biofilm to grow as well as optimal conditions for the bacteria community when the carriers are suspended in water. The process configuration with reactors in series allows each reactor to maintain its own bacteria community. The carriers with biofilm are prevented from leaving the reactor by the presence of a physical barrier installed in the tank outlet, and each reactor has therefore unique properties, e.g. the used source of substrate and sludge age. At the same time, the system allows development of fast and slow growing bacteria. This can be an advantage when the aim is to degrade difficult degradable compounds present in low concentrations, which is the case for many of the pharmaceutical compounds in domestic wastewater. On the contrary, activated sludge systems have one bacteria community with same sludge age, as sludge is circulated from outlet to inlet section providing the bacteria access to easy degradable compounds on a regular basis. Furthermore, a portion of the bulk biomass in an activated sludge system is continuously withdrawn as excess sludge not allowing maintenance of slower growing bacteria in the system. However, an advantage with an activated sludge system could be the larger amount of biomass present in the system, which could be a gain in case adsorption plays a role in the removal of pharmaceuticals.

The results from the first Mermiss project (reported in Kragelund Rickers et al., 2015) indicated that it was possible, in a MBBR system with three reactors in series, to develop biofilm with specific capabilities in each reactor in relation to degradation of pharmaceuticals. The initial test was in bench-scale (3L glass vials) with the focus of validating the MBBR process with respect to pharmaceutical degradation in larger scale, and tested on different wastewater types. Also to investigate the possibility of maximizing the global removal of pharmaceuticals with a system capitalizing on the benefits of both biofilm and suspended communities. This was realized by combining the pure biofilm MBBR with an IFAS system (Integrated Fixed Film Activated Sludge) called HYBAS™.

1.2 Test sites identified

For sidestream treatment of hospital wastewater, wastewater from the Dept. of Oncology at Aarhus University Hospital was selected, and which contained probably some of the most toxic compounds discharged from a hospital. Estimated flow from this department is 15,000 – 20,000 m^3/year. The tests were carried out in bench-scale.

For testing MBBR as a polishing technology, Viby wastewater treatment plant in Aarhus was selected also because no hospital wastewater is received at the WWTP. Viby WWTP is designed for a capacity of 84,000 PE, and currently it is treating wastewater with a load of 79,386 PE and in total approx. 10 million m^3/year. The tests were carried out in bench-scale.

For testing the treatment of the entire wastewater stream from a hospital, the DNU hospital was selected. DNU hospital is currently under construction and will be the largest hospital in the central region of Denmark by 2020. Today the number of beds is approx. 1150, and the wastewater volume corresponds to 180,000 m^3/year and have 10,200 employees. In 2020, the rebuilding of DNU will be finished and will thus have approx. 1080 beds and the same wastewater volume as today. The tests were carried out in pilot-scale.

HYBAS™ treatment of the municipal wastewater was carried out at Herning municipality. The WWTP for Herning municipality is designed for 175,000 PE, and is at present receiving wastewater corresponding to 120,000 PE, which is in total up to 11-12 million m^3/year. The tests were carried out in pilot-scale. The new hospital in Herning, currently under construction, is expecting to discharge the combined wastewater in a separate pipe to Herning Municipality´s wastewater treatment plant. The estimated volume of hospital wastewater is 150,000 m^3/year in 2020.

1.2.1 Bench-scale plant for side-stream treatment at Dept. of Oncology

The purpose of the treatment simulated in this experimental system was to perform full treatment, including removal of micropollutants, of wastewater from a hospital ward with an IFAS system (HYBAS™) before discharge to the municipal sewer. The bench-scale plant designed for continuous feed was placed in a 40´container. The container was located outside the Dept. of Oncology at Aarhus University Hospital for side-stream treatment of wastewater withdrawn from the department´s sewer. The bench-scale treatment included removal of organic carbon, nitrification as well as micro pollutants.

The wastewater was pumped from the sewer into a mixing tank, from where it was fed to the HYBAS™ system. The process line consisted of four 3-liter reactors in series (H1, H2, H3 and P, Figure 1). H1 contained only activated sludge, H2 and H3 had both activated sludge and carriers with biofilm (filling ratio of 50%), and P was a polishing step with only biofilm carriers (MBBR). After the HYBAS™ reactor H3, a settling tank was installed and from here, the activated sludge was recycled back to reactor H1. The activated sludge was frequently replaced with sludge from the local municipal plant Viby WWTP. The pilot plant was operated in six months.

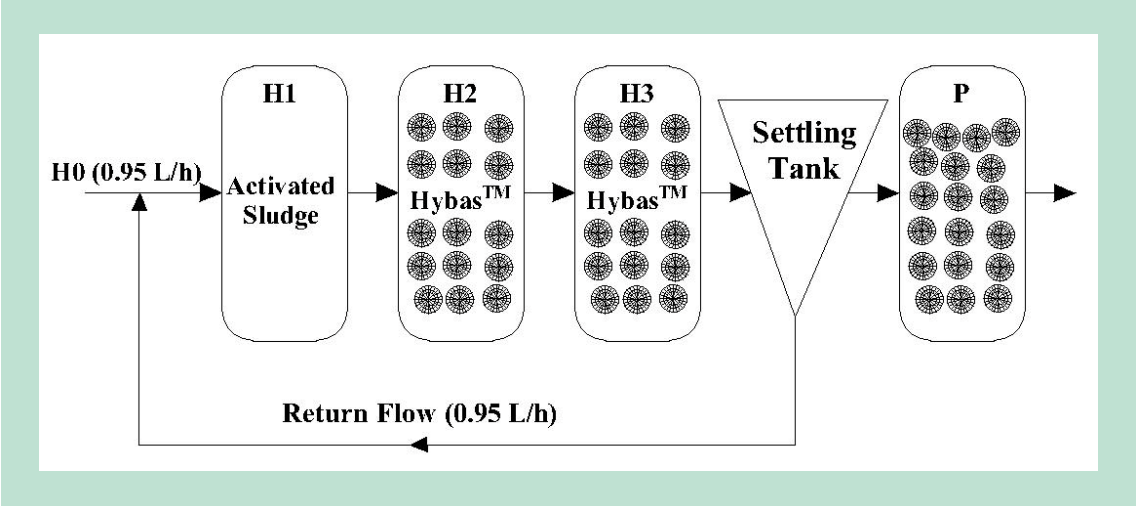


Figure 1. Overview of the Hybas™ configuration.

1.2.2 Bench scale plant for treatment of municipal WWTP effluent

The bench-scale plant was set up at the municipal wastewater, Viby WWTP, in Aarhus. The purpose of the treatment simulated by this experimental system was to obtain removal of mi-

cro pollutants in typical effluent waters from a municipal activated sludge wastewater treatment plant with nitrogen and phosphorous removal, before discharge to a recipient. The container with the bench-scale plant was placed next to the WTTP outlet, from where effluent subject to treatment was extracted. The configuration of the plant was as shown in Figure 2, with three reactors, containing AnoxKaldnes™ K5 carriers (AnoxKaldnes, Lund, Sweden) and with filling ratio of 50%. The bench-scale treatment included removal of organic carbon, nitrification as well as micro pollutants.

Two reactors operated in line receiving effluent wastewater and working as the polishing reactors. The third reactor (position C) was fed with primary settled wastewater, in order to enhance biofilm growth on the carriers (regeneration). After approx. two days of operation in this mode, the flow was changed, putting the lowest loaded polishing reactor (B) in regeneration position and the reactor just regenerated in first position in the main treatment line.

This rotation was carried out in order to let all reactors get a period of regeneration before being placed in polishing position. The purpose of the rotation was to ensure that the microbial community was able to regenerate after periods with very limited organic material available simultaneously preventing overgrowth of the dedicated biomass by fast growing heterotrophic microorganisms. Patent application is filed for this operation mode (Swedish patent application no 1650321-1). The pilot plant was operated for seven months.

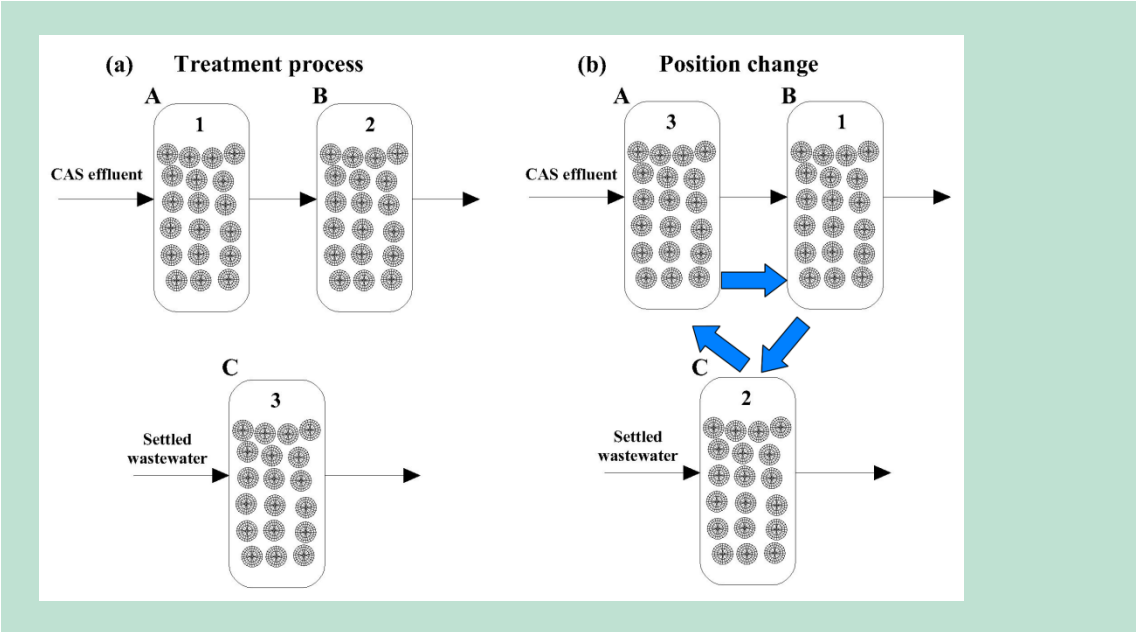


Figure 2. Operation of two MBBR tanks polishing effluent from the Viby WWTP (positions A and B), while the growth/regeneration of biofilm was stimulated in a third MBBR reactor (position C).

1.2.3 Pilot-scale treatment of hospital wastewater

In the vicinity of the Aarhus University Hospital and close to the main sewer near the hospital a suitable location for the pilot-scale treatment plant was identified. The pilot plant was housed in two 40´containers. The pilot plant was fed with wastewater drawn from the main hospital sewer and the treatment included removal of organic carbon, nitrogen as well as micro pollutants. Pilot plant effluent was discharged to the municipal sewer.

The pilot plant was designed as a six-staged MBBR (4 reactors M1 – M3, each1 m^3, 2 reactors M4-M5, each 0.5 m^3), i.e. the plant was designed as a pure biofilm system. A holding tank for raw wastewater was placed upstream the MBBR. The purpose of this holding tank was to

stabilize the feed flow. The holding tank had an overflow pipe allowing a higher flow through the tank compared to the pilot plant capacity preventing septic conditions. Raw hospital wastewater passed a 100-micron filter before entering the first reactor, M1. All the MBBRs were operated with a carrier filling ratio of 50% of AnoxKaldnes™ K5 carriers (AnoxKaldnes, Lund, Sweden). The set-up is illustrated in Figure 3. M1 and M4 were operated as denitrifying (DN) reactors, while the remaining four reactors were aerated and performed COD removal, nitrification (N) processes and removal of micropollutants. The treatment line was operated at temperature of approx. 20 °C.

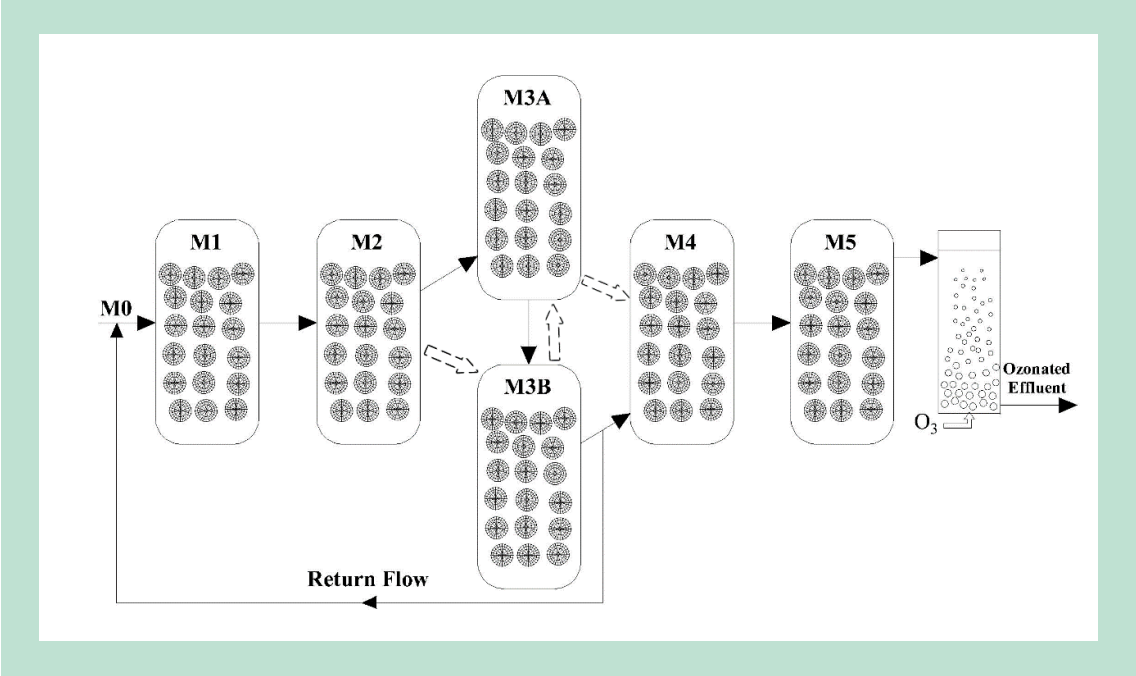


Figure 3. Overview of the pilot-scale MBBR treatment plant in Aarhus University Hospital, Skejby. Denitrification (DN) processes occur in M1 and M4 while M2, M3A, M3B and M5 perform nitrification (N) processes.

The flow of the pilot plant followed either the black path or the dotted path, each lasting for 12 hours. Feed flow was 300 l/h and return flow 500 l/h (M1, M2, M3A and M3B). The pilot plant was in operation for 11 months.

1.2.4 Pilot-scale treatment for municipal wastewater

The purpose of the treatment simulated by this experimental system was to perform complete treatment of municipal wastewater including removal of micro-pollutants before being discharged to a nearby creek. The pilot-scale treatment included removal of organic carbon, nitrogen as well as micro pollutants.

The MBBR pilot plant used for the tests at the Aarhus University Hospital was modified to fit the treatment objectives for wastewater from Herning municipality. The volumes of the reactors were not changed. However, instead of a pure biofilm MBBR process, the HYBAS™ process was selected combining activated sludge and biofilm. The process change required a settling tank after the H3A and H3B reactors for return of activated sludge to reactor H1. Reactors H1 and H2 operated only with activated sludge, H3A and H3B worked with activated sludge and biofilm on carriers, and reactors H4 and H5 worked as MBBR i.e. without activated sludge, see Figure 4.

The raw wastewater was first pumped through a 100-micron filter before entering H1 (pre-denitrification reactor). Recirculation for pre-denitrification was taken from H3 outlet and return sludge was pumped from the settling tank back to H1. The pilot plant was operated at the temperature of the incoming wastewater varied in between 12 to 19 °C depending on season.

Reactors H1 and H4 were denitrifying reactors (pre- and post- denitrification), and the remaining reactors were aerated for removal of organic matter and nitrogen. Ethanol for post-denitrification was added as required.

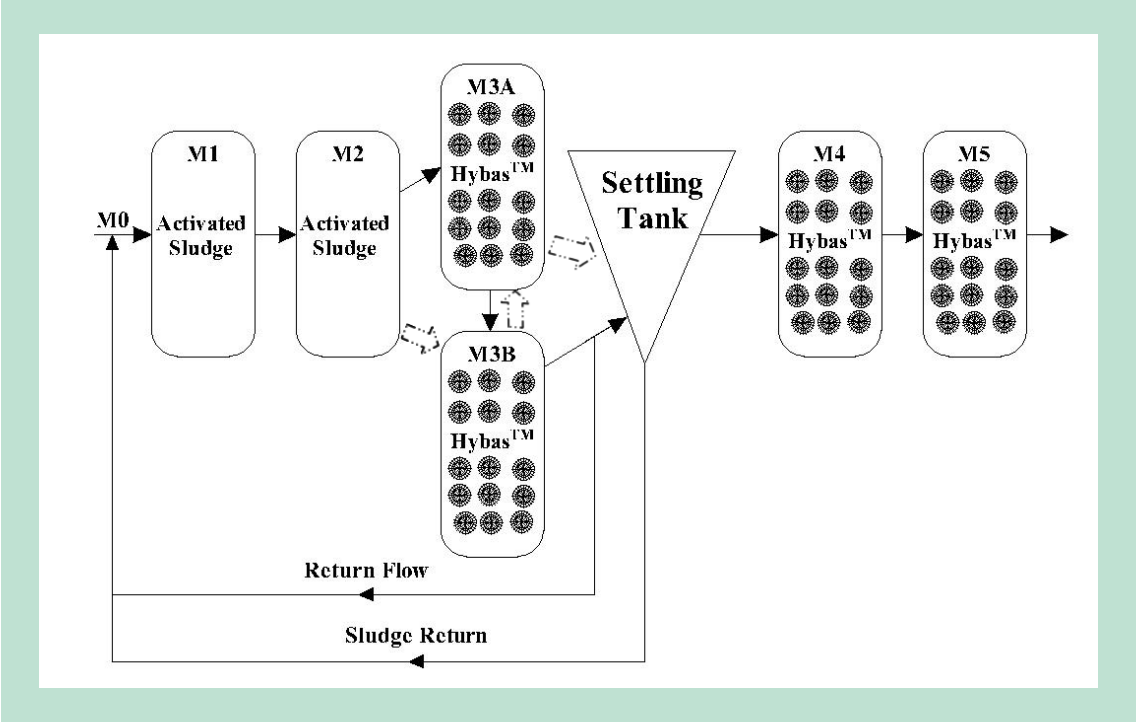


Figure 4. Overview of pilot-scale treatment as HYBAS™ system at Herning municipality.

The pilot plant was in operation for nine months.

1.3 Wastewater parameters measured during operation

A number of parameters were monitored to ensure stable operation of both the bench-scale and pilot-scale treatment plants. Inlet flow, pH, dissolved oxygen and temperature were recorded several times a week for the bench-scale and weekly or continuously online for the pilot-scale treatment plant. Recirculation was monitored in pilot-scale.

Analysis was performed on a weekly or biweekly basis for COD, TOC, TN, TP, NO_3-N, NO_2-N, NH_4-N (both filtered and non-filtered samples), suspended solids (SS) and for HYBAS™ systems sludge volume. The amount of biomass in the system (TS/VS) including biomass amount on carriers (biofilm) were determined regularly.

2. Pharmaceuticals and analytical methods

2.1 Selection of pharmaceuticals investigated

2.1.1 Pharmaceuticals of interest in Europe

Several attempts have been made all over Europe to identify pharmaceuticals representing a potential risk for recipient waters. In 2013 in Denmark, a guideline has been proposed to regulate hospital wastewater to sewer, as point sources and therefore, discharge of pharmaceuticals will have to be regulated as industries (Local Government Denmark, 2013). The list comprising of 40 pharmaceuticals present in hospital wastewater as maximum concentrations when discharged into the sewer, and their proposed guiding limit values were listed (Table 1, AMK 2015). The pharmaceuticals present on the list were selected based on their usage i.e. predicted effluent concentrations in hospital wastewater, the stability score (persistence) and potential hazardous impact, i.e., predicted environmental no effect concentrations (PNEC).

Table 1. The Danish proposed guiding limit values in hospital wastewater discharge.

Pharmaceutical	ATC code	PNEC [µg/L]
Dronedaron	C01BD07	0.4
Ibuprofen	C01EB16; M01AE01; M02AA13	4
Furosemid	C03CA01	31
Propranolol	C07AA05	0.1
Amlodipin	C08CA01	1
Candesartan	C09CA06	0.12
Cyproteron	G03HA01	0.3
Prednisolon	H02AB06; C05AA04; A07EA01; S01BA04	
Ceftazidim	J01DD02	0.13
Sulfamethoxazol	J01EE01	0.12
Erythromycin	J01FA01	0.2
Clarithromycin	J01FA09	0.06
Azithromycin	J01FA10	0.09
Ofloxacin	J01MA01	0.1
Ciprofloxacin	J01MA02; S01AX13; S02AA15	0.089
Efavirenz	J05AG03	1.2
Capecitabin	L01BC06	0.2
Nilotinib	L01XE08	0.26
Fulvestrant	L02BA03	0.00057
Bicalutamid	L02BB03	0.1
Mycophenolic acid	L04AA06	0.1

Pharmaceutical	ATC code	PNEC [µg/L]
Diclofenac	M01AB05; M01AB55; S01BC03	0.1
Naproxen	M01AE02	6.4
Propofol	N01AX10	2.3
Tramadol	N02AX02	
Paracetamol	N02BE01	9.2
Carbamazepin	N03AF01	0.5
Olanzapin	N05AH03	1.1
Quetiapin	N05AH04	10
Fluoxetin	N06AB03	0.11
Citalopram	N06AB04	8
Duloxetin	N06AX21	0.43
Disulfiram	N07BB01	0.46
Buprenorphin	N07BC01; N07BC51	14
Deferasirox	V03AC03	0.53
Lanthanum	V03AE03	10
Ciprofloxacin	J01MA02; S01AX13; S02AA15	0.089
Prednisolon	H02AB06; C05AA04; A07EA01; S01BA04	
Tramadol	N02AX02	

In Switzerland, different lists have been created for two purposes: a) to describe quality of surface waters and b) to document the efficiencies of newly established treatment options using ozonation and activated carbon (Table 2).

Table 2. Swiss list for surface waters including pesticides, personal care compounds and industrial chemicals (BAFU: 2012, Mikroverunreinigungen aus kommunalem Abwasser).

Substances	Chronic quality criterion (LZ-UQN) [µg/L]
Compounds	
Atenolol	150
Azithromycin	0.09
Bezafibrat	0.46
Carbamazepine	0.5
Carbamazepine-10, 11-dihydro-10, 11-dihydroxy	0.1
Clarithromycin	0.06
Diclofenac	0.05
Erythromycin	0.04
Ethinylestradiol	0.000037
Ibuprofen	0.3
Mefenamic acid	4

Substances	Chronic quality criterion (LZ-UQN) [µg/L]
Metoprolol	64
Naproxen	1.7
Sotalol	0.1
Sulfamethoxazole	0.6
Trimethoprim	60
Estradiol	0.0004
Estrone	0.0036
Nonylphenol	0.013
Benzotrazol	30
EDTA	2200
Methylbenzotriazol	75
NTA	190

Depending on the aim of the list and the legislation it is embedded in, the list is based on PEC/PNEC considerations which give a toxicology or effect driven priorisation, or as a list of compounds used for benchmarking one treatment plant against others. However, often the PEC/PNEC assessments underlie high insecurities on both predicted environmental concentration (PEC) and well as the predicted no effect concentration (PNEC) are uncertain by orders of magnitude. The Danish 33+3 list is a typical example for such priorisation (Local Government Denmark, 2013). Priorisations used for benchmarking are generated to compare the performance of a newly established treatment plant e.g. to a standard well-functioning treatment technology e.g. ozonation plant. Here the target is solely to establish that the new plant is working as well as a comparative one.

As such lists are generated with respect to local usage, technical possibilities and political objectives, the original purpose of such lists needs to be kept in mind when using them, to prevent excessive overspending in projects and in investments. However, there are some compounds that are often used in assessments and in lists: They usually contain a combination of compounds extremely difficult to degrade (e.g. Diclofenac, Carbamazepine, X ray contrast media) several antibiotics (sulfonamides, macrolides, fluorquinolones) to cover the issue on antibiotic resistance and blood pressure regulators (beta blockers) as well as some antidepressants and analgetics as the latter two groups are ubiquitous.

Within research and development projects, the resources for chemical analysis are usually limited as they were in this case. It was thus aimed for analyzing as many compounds as possible in one chromatographic run on HPLC-MS/MS. Other compounds that would have required a second run, e.g. using a different HPLC- column, other ionization method, etc., would demand doubling these efforts. Such compounds were thus excluded from the project to gain most data related to the processes, i.e. most value for money.

Based on these above-mentioned assumptions, the following pharmaceuticals were identified within this project, and guiding limit concentrations are those proposed in AMK, 2015, see Table 3 .

2.2 Selection of compounds investigated in this study

The list of compounds selected in the Mermiss project is comprised of pharmaceuticals with all relevant functionalities
though deliberately avoiding pharmaceuticals that are difficult to quantify as ethinylestradiol, or specialized cancer drugs.

The list is thus comprised of a selection of some of the pharmaceuticals present on the AMK, 2015 as well as additional antibiotics not present on the AMK list. By selecting these different pharmaceuticals representing different biodegradability, sufficient data for documenting the performance of individual reactors as well as the entire treatment concept would be possible. Selection of different types of antibiotics were included in the analyses as sulfonamides, macrolides and a fluoroquinolone. Sulfadiazine, sulfamethizole, sulfamethoxazole are typical sulfonamides, which are fully synthetic antibiotics. One conjugation product is also included as this acetyl metabolite is typically excreted after human metabolism and also the sulfonamide booster trimethoprim is also of interest. The macrolide antibiotics azithromycine, clarithromycine, erythromycin, roxythromycin and clindamycine are antibiotics with complex structures originating from fermentation processes and ciprofloxacine is the only fluoroquinolone included in this study.

Blood pressure agents as beta-blockers exemplified as atenolol, metoprolol, propranolol and sotalol are present on the Mermiss list. The pharmaceuticals are primarily used in private homes, and the required daily doses are in the g range, as opposed to more modern blood pressure regulators so expected concentrations in wastewater are in the µg/L range.

A selection of different hardly biodegradable compounds as carbamazepine, citalopram, diclofenac, venlafaxine were also included as they are consumed in large quantities. Ibuprofen is relatively easy to degrade, but it is one of the most used compounds reaching the highest concentrations of pharmaceuticals in wastewater, similar to phenazone.
The X-ray contrast media are very difficult to remove with ozonation, sorption and biodegradation, and they were included as worst case.

Table 3. Compounds included in the determination of MERMISS process control.

	Standard LOQ*	Expected in municipal wastewater*,**	Included in the 33 list [PNEC]	Included in the Swiss list [with target]	Comment
Unit	**ng/L**	**ng/L**	**ng/L**	**ng/L**	
Antibiotics (also metabolites)					
ac-sulfadiazin	70	1000	-	-	
azithromycin	1000	1000	90	90]	
ciprofloxacin	10000	1000	5	-	
clarithromycin	1000	1000	60	60	
clindamycin	1000	1000	-	-	
erythromycin	1000	1000	40	40	
roxithromycin	700	1000	-	-	
sulfadiazin	30	1000	-	-	
sulfamethizol	78	1000	-	+	
sulfamethoxazol	33	1000	120	600	
trimethoprim	22	1000		-	
Blood pressure					
atenolol	70	1000	-	150 000	
metoprolol	3,8	1000	-	64 000	
propranolol	50	1000	+[100]	-	
sotalol	260	1000		100	

	Standard LOQ*	Expected in municipal wastewater*,**	Included in the 33 list [PNEC]	Included in the Swiss list [with target]	Comment
Unit	**ng/L**	**ng/L**	**ng/L**	**ng/L**	
Antidepressant/ analgesic					
carbamazepine	70	1000	+ [500]	500	
citalopram	43	1000	+[8000]	-	
diclofenac	57	1000	+[100]	50	
ibuprofen	5300	1000	+[4000]	500	
phenanzon	130	-	-	-	
tramadol	800	1000	+[2250]	-	
venlafaxin	200	1000	-	-	
X-ray contrast media					
iohexol	1000	5000	-	-	
iopromid	10000	50000	-	mentioned, but no target	
Iomeprol	1000	5000	-	mentioned, but no target	
Iopamidol	1000	5000	-	mentioned, but no target	

* Danish Nature Agency: Hospital Wastewater – BAT and development of treatment technologies. June 2011. Report prepared by DHI (in Danish)

** Danish Nature Agency: NOVANA - Screening for human pharmaceuticals in water bodies. 2015 (in Danish)

2.3 Analysis of compounds

The single pharmaceuticals and in some cases their transformation products were analyzed by HPLC-MS/MS, as all compounds are relatively hydrophilic and have low volatility. HPLC-MS/MS is based on a separation of the compounds in the liquid phase in this case a Phenomenex Synergi (150x 2mm) column was used and a gradient of acidified (0.5% formic acid) water/methanol was utilized. The eluent started with 100% water to wash of ionic compounds. Following that, the eluent was gradually increased in methanol content until it reached 100%. Over this period the eluent was directed into the ion source of the mass spectrometer. In the ion source all incoming compounds were ionized and directed to the triple quadrupole mass analyzer. In the first quadrupole, the respective compounds were isolated, in the second one fragmented with nitrogen gas while in the third quadrupole the respective fragments were analyzed quantitatively- this operation is called multi reaction monitoring (MRM). Each compound is quantified using two MRM transitions to eliminate false positives or false negatives. Samples from the experiments were analyzed directly after removing particulate matter by centrifugation using high volume injections. Each sample was injected twice to eliminate possible problems with injections, and compared via an internal standard calibration to a multilevel calibration typically containing 15 data points. Compounds labeled with stable isotopes (deuterium or ^{13}C) were used as internal standards.

The described method is very sensitive meaning low limits of quantifications could be achieved, and it is very selective, meaning there are very few signals disturbing those that

were used for quantitation. The used method is usually producing standard deviations of less than 10%; thus the primary assessment of data was performed considering a difference exceeding the SD of 10% was considered as different: So if the difference between 2 samples exceeded 20% differences was considered as established. In cases of doubt, more sophisticated considerations were performed.

2.4 Methodology for determining the degradation of pharmaceuticals

Two types of methods were applied to determine the degradation of pharmaceuticals regardless of treatment scale (bench-scale or pilot scale). The first approach involved spiking experiments where a known concentration of selected compounds was added and samples were taken over a period of 24 h. The spiking experiments reveal the potential removal capacity and is described in further details below. In the second approach, a bulk of water was followed throughout the treatment system according to the designed hydraulic retention time (HRT), and samplings were conducted in the different reactors accordingly. By combining these approaches, information on actual removal capability of the treatment system at a particular day was investigated as well as the overall potential of the system.

2.4.1 Characterizing the potential removal capacity of treatment plants

The first method of evaluating the degradation of pharmaceuticals, investigates the potential removal of the analyzed treatment technology (MBBR or HYBAS™) in a bench-scale reactor. The number of investigated reactors, corresponds to reactors in the treatment setup in either bench-scale (3 aerobic reactors) or pilot scale (six reactors of which two are anaerobic).

To determine the potential removal capacity by piking experiments, samples of wastewater (and carries) were collected at the different treatment technology setups and brought back to laboratory. Here lab-scale experiments were carried out in 3 L reactors, simulating the treatment train (bench-scale/ pilot scale). The wastewater and carrier ratio were kept in the same order of magnitude as in the treatment trains (bench-scale/ pilot scale).

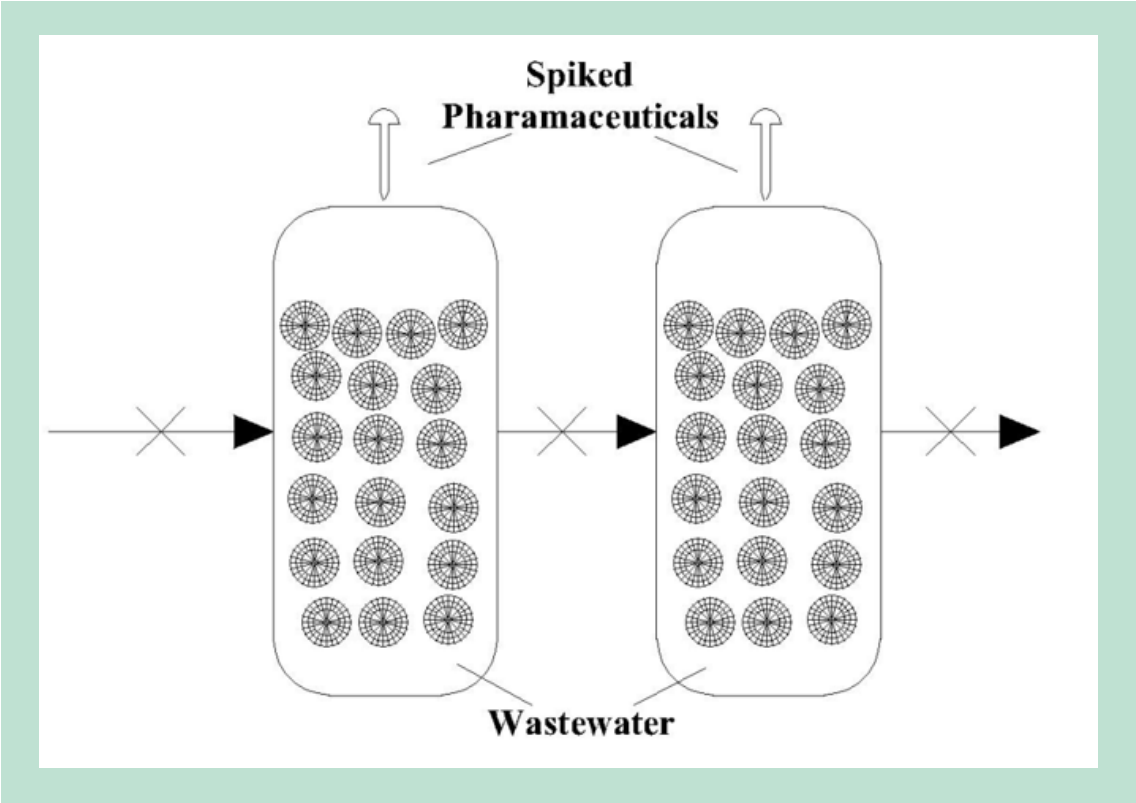


Figure 5. Overview of the experimental setup for determining the potential capacity of the treatment technology, investigated in batch scale. The flow between reactors is discontinued while concentrations of spiked pharmaceuticals were measured over a 24 hour time-period.

To evaluate the potential capacity for pharmaceutical degradation by each reactor's biofilm, the flow was stopped by shutting down all pumps and interrupting all flow paths, as described in Figure 5. A mix of 33 compounds (in 500 µl of methanol) was added to each reactor, which resulted in compound-specific starting concentrations between 3 µg·L-1 and 20 µg·L-1. After the initiation of the spiking experiment, 10 mL samples were taken from each reactor using a glass pipette at 1 min, 20 min, 1 h, 2 h, 3 h, 4 h, 6 h, 10.5 h, 21.5 h, 24 h and 25.5 h.

In the pilot scale experiments, water from reactors 3A and 3B or 3A/3B formed a single reactor (M3 or H3) because they were assumed to be at similar conditions. At the same time, the reactors were maintained at their normal operating conditions, for examples, sparging of nitrogen (N2) gas through the denitrifying reactors (in pilot scale M1 or H1 and M4 or H4) and maintaining the pH.

2.4.1.1 Calculation for potential removal capacity from batch experiment

To evaluate degradation the potential removal of pharmaceuticals, first-order degradation equation (Equation 1) was fitted using GraphPad Prism, with no weighting:

Equation 1: $C = C_0 \cdot e^{-kt}$

Thus in Graphpad, the concentrations of the given pharmaceutical over 24 hours for each reactor were plotted and mathematically compared and fitted to equation 1.

To calculate and predict removal from the batch experiments, equation (2) was used

Equation 2: $Removal = 1 - \frac{1}{(1+k_A\, HRT_A)(1+k_B\, HRT_B)}$

Where k symbolises the reaction rate constants (as obtained from equation (1)) of each reactor (A and B) and HRT is the hydraulic retention time.

2.4.2 Actual removal of pharmaceuticals during treatment (continuous flow experiment)

The second method of evaluating the degradation of pharmaceuticals, investigates the actual removal of the analyzed treatment technology (MBBR or Hybas™) at a given scale (lab scale or pilot scale) see Figure 6. The number of investigated reactors, corresponds to reactors in the treatment setup in either lab scale (3 aerobic reactors) or pilot scale (six reactors of which two are anaerobic)

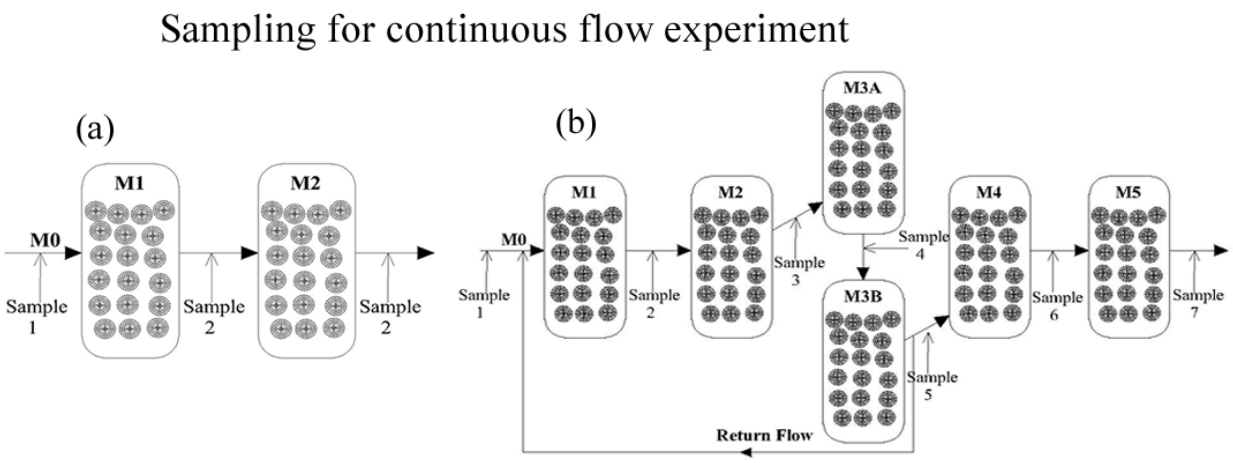


Figure 6. Overview of the experimental setup for determining the actual capacity of the treatment technology, investigated in the corresponding treatment scale ((a): lab-scale sampling; (b): pilot-scale sampling). Samplings between the reactors are carried out based on the HRT of the system.

The principle behind the sampling was to follow the water flow through the system. Different HRT´s were applied depending on scale of system and treatment type.

Table 4. Overview of applied HRTs at the different treatment locations

Scale of treatment	Place of treatment	Number of reactors	Total HRT (hours)	Reactor HRT (hours)
Lab scale	Sidestream treatment	3		
Lab scale	Polishing at municipality	3	1	0.5
			4	2
Pilot scale	MBBR at hospital	6	12	2
Pilot scale	Hybas™ at municipality	6	8	1-1.5

2.4.2.1 Calculations for actual removal capacity from continuous flow experiments

The concentrations measured follow the hydraulic retention time (HRT) in each reactor. The results are shown as native concentrations of the selected pharmaceuticals present for the different reactors. Potential impact of recirculation (MBBR pilot scale) or recirculation and sludge (Hybas™ pilot plant) is depicted.
The removal rate is calculated by comparing concentrations between influent (Inf.) and the last reactor (M3 in lab scale or M5/H5 in pilot scale) using Equation 3:

Equation 3: $$Removal = \left(1 - \frac{Concentration\ last\ reactor\ (\boldsymbol{M5})}{Concentration\ initial\ (\boldsymbol{Inf.})}\right)$$

Results are shown as overall actual removal of pharmaceuticals detected at native concentrations at a given day.

However, some pharmaceutical can be designated either rather low or even negative removal. This can occur with pharmaceuticals that are excreted as conjugates. The excreted conjugates can be de-conjugated by bacterial enzymes while transported in the sewer or during the wastewater treatment. Another reason of increasing level of a parent compound could be transformation of metabolites from other parent compounds (Kovalova et al., 2012). An increased concentration of a pharmaceutical in the effluent or at an intermittent treatment stage has formerly been observed in WWTPs (Ternes, 1998, Onesios et al., 2009; Falås et al., 2012) and in hospital waste water treatment plants (Kovalova et al., 2012; Cruz-Morató et al., 2014). This effect is expected to be even stronger in source treatment systems as hospital waste water because there will be shorter time for de-conjugation in the sewer in the case of source treatment as the travel time in the sewers is much shorter than to the WWTP.

2.5 Mapping of consumption of pharmaceuticals at DNU hospital

A mapping was carried out on one of the major wastewater treatment plants Lynetten where several hospitals discharge their wastewater, among which Rigshospitalet is a large contributor (Mose Pedersen et al., 2007). Use of pharmaceuticals at the involved hospitals and medicine used in private homes were characterized by using the public available database where all medicine used in Denmark is registered (http://www.medstat.dk/en). Here it is possible to differentiate between medicine used in the private sector and the hospital pharmacies. The study revealed that most medicine was in fact consumed in private sector (96-99%) and only approx. 1-4% at the hospital (Mose-Pedersen, 2007).

Based on this information, an additional mapping has been carried out to estimate the discharge of toxic pharmaceuticals from Aarhus University Hospital (AUH) to the wastewater based on data from 2011 up to 2015. The number of hospitalized patients and patients receiving pharmaceuticals during ambulatory treatment was recorded at the different years. Comparison of pharmaceuticals distributed by the hospital pharmacy was compared to pharmaceuticals listed in the Electronic Patient Journal (EPJ). Based on this information it has been possible to determine whether the toxic pharmaceuticals used in the treatment of patients is discharged directly from the hospital or from the patient's own home.

Mapping was carried out on pharmaceuticals present on the guiding limit list (Local Government Denmark, 2013). Only pharmaceuticals, where more than 2% of the national consumption was consumed at the hospitals were included. Currently this form a list of 42 pharmaceuticals, which can be expected to be discharged in larger quantities from the Danish hospitals leading to unwanted environmental impact.

The calculated environmental impact is based on the PNEC values set by the Danish authorities (Local Government Denmark, 2013). A precondition in the mapping is that all prescribed pharmaceuticals are discharged directly to the toilet/sewer, which is a conservative assumption. Thus, neither metabolization of pharmaceuticals nor potential cocktail effects of pharmaceuticals and half times of pharmaceuticals are taken into consideration.

3. Mapping of pharmaceuticals

3.1 Mapping of pharmaceuticals discharged from Danish hospitals

Comprehensive characterizations of the pharmaceuticals used in different Danish hospitals have been investigated for more than a decade (Mose-Pedersen et al., 2007) by using the available public database (http://www.medstat.dk/en) where all consumption of pharmaceuticals is registered. This database allows differentiating between pharmaceuticals consumed at the hospital (registered as the hospital pharmacy) or in the private sector (homes). Mappings were carried out on several hospitals in Zealand, Denmark. The study revealed that approx. 1 - 4% of the entire consumption of pharmaceuticals took place at the hospital and the remaining part was consumed in the private sector and thus discharged here (Mose-Pedersen, 2007). Based on these comprehensive studies on hospitals´ impact on the environment, a work group was established whose task was to define which pharmaceuticals should be minimized in the discharge of hospital wastewater. The outcome of this work was the creation of a list with guiding limit values for 36 pharmaceuticals in 2013, with pharmaceuticals known to have toxic effect on bacteria, algae, Crustacea, and fish etc., Later, the list was expanded to include 40 pharmaceuticals (AMK, 2013). Today, guiding limits exist for recommended maximal concentrations for hospital wastewater discharged to a municipal wastewater treatment plant, and also guiding values for direct discharge. However, at present no hospitals have been allowed to discharge treated wastewater directly to recipients, even though this is the main focus for the Herlev Hospital wastewater treatment plant (Grundfos Biobooster, 2016).

3.2 Mapping of pharmaceuticals discharged from DNU in wastewater

One of the reasons why this mapping is highly relevant is that the treatment of patients today to a much larger extent than before is carried out as outpatient treatment. This means that the patients are sent home directly after treatment, hence the medication that they are given is discharged from their homes and not from the hospital. Data from Aarhus University Hospital (AUH) shows that from 2007 to 2015 the number of ambulant patients at AUH has increased from around 600.000 in 2007 to 800.000 in 2015, e.g. more than 33% increase (Møller, Environmental report, AUH, 2014), see Figure 7.

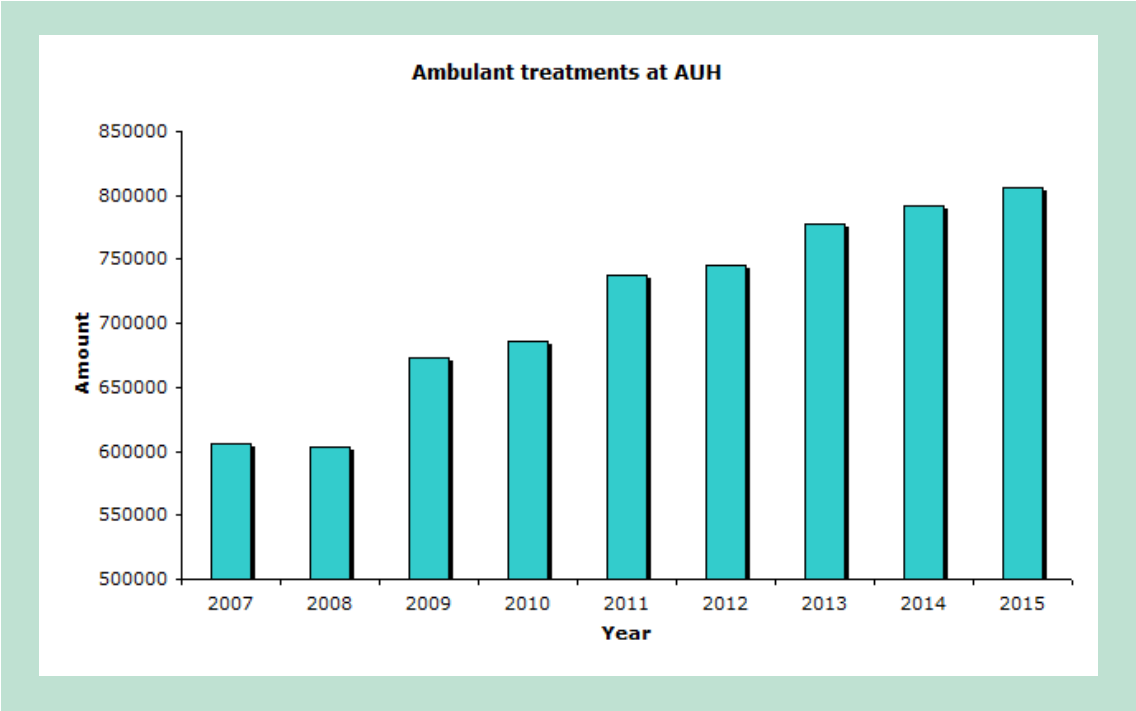


Figure 7. Development of ambulatory treatments at DNU hospital from 2007-2015.

Great effort has been used for implementing an Electronic Patient Journal (EPJ) in the Danish healthcare sector. The EPJ has made it possible to extract data from the hospital database showing whether medication is for treatment of an ambulant or a hospitalized patient. This makes it possible to determine whether a toxic pharmaceutical used in the treatment of patients, is discharged into the hospital´s wastewater or instead discharged from the patient's own home. This provides important knowledge as to where the discharge of toxic pharmaceuticals in fact is taking place and where the largest benefit of wastewater treatment for pharmaceutical reduction can be obtained.

The results from the mapping show three vital conditions:

1. There is a continuous and significant rise in the environmental impact due to the activities related to AUH.
2. The greatest part of the environmental impact originates from pharmaceuticals discharged from the private sector (homes) due to ambulant treatments.
3. The environmental impact from the pharmaceuticals discharge from the private sector is increasing more rapidly than the impact that can be traced back to wastewater discharged from the hospital.
4. Almost the entire environmental impact (98%) originates from only six out of 42 investigated pharmaceuticals. These are:
    a. Mycophenolic acid (represents 71% of the environmental impact)
    b. Clarithromyzine
    c. Sulfamethoxazole
    d. Sertraline
    e. Ciprofloxacine
    f. Capecitabine

The importance of the environmental impact from pharmaceuticals consumed in private homes, and thus being discharged to the local municipalities wastewater, is shown in Figure 8. In 2015, approx. 84% of the entire environmental impact previously assigned to the hospital pharmacy could be traced to ambulant patient treatments. There are no indications in Den-

mark that the trends observed within ambulant treatment at Aarhus University Hospital (AUH) will change, and therefore deeper understanding of the exact environmental impact of hospitals has been desired. These newer data show that the environmental impact from AUH, based on the guiding limit values for selected pharmaceuticals, is much lower than previously anticipated due to the importance of ambulant treatments.

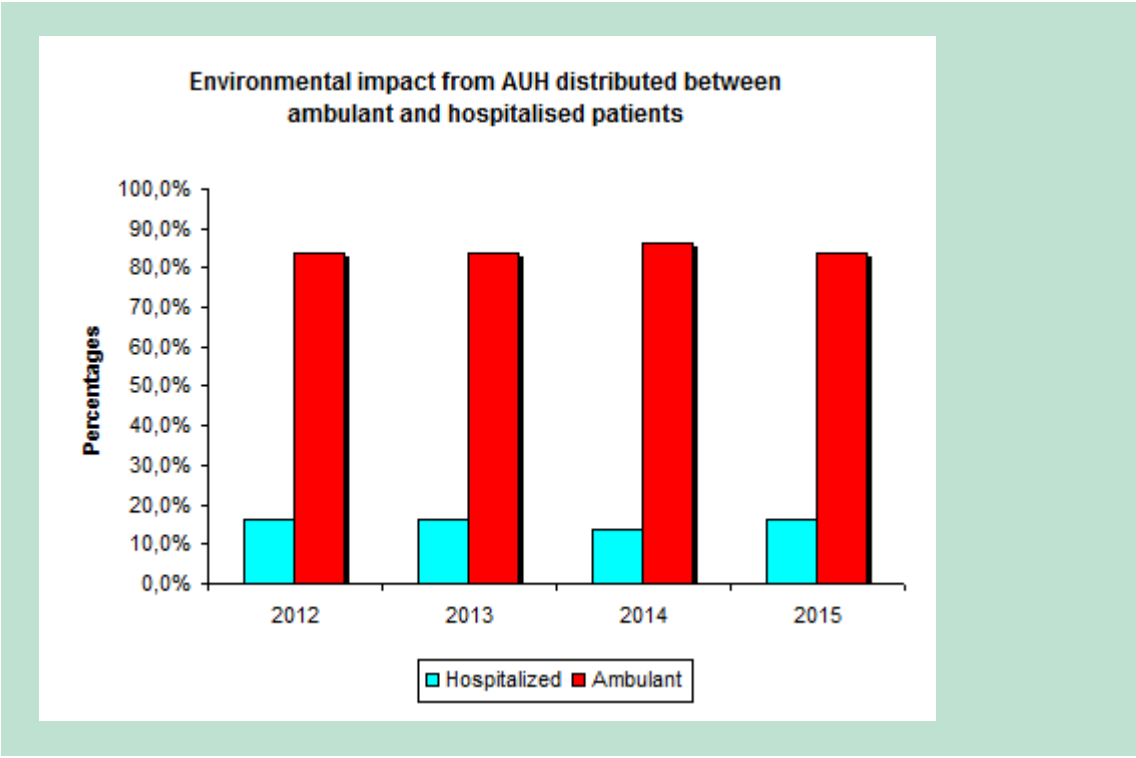


Figure 8. Overview of environmental impact from 2011-2015 of the medicine consumed either by hospitalized patients or ambulant patients.

In order to evaluate the relevance of the mapping and whether it actually reflects the reality, the calculated results have been compared with measurements from the MBBR pilot plant at AUH. In Table 5, the calculated and measured concentrations for a number of pharmaceuticals are compared. The predicted calculated concentrations represent a theoretical maximum concentration in the wastewater from AUH, when medication only given to hospitalized patients is included in the calculation. Hence, the measured concentrations should be lower than the calculated concentration, since the calculated concentrations are theoretical maximum concentrations. In seven out of nine cases, the measured concentrations in wastewater supports (are lower than) the calculated concentrations from the mapping. Based on the measurements, it can be concluded that the calculated numbers regarding the environmental impact of hospital discharge do reflect the reality. The two measured concentrations that are (10-20%) higher than the calculated concentrations can be explained by the daily variations in pharmaceutical consumption at AUH.

Table 5. Predicted and measured concentrations of selected pharmaceuticals at AUH. Green marking indicates concordance.

Pharmaceutical	Raw wastewater from hospital (inlet concentration to pilot MBBR plant)	Predicted concentrations due to hospital wastewater discharge
	µg/l	µg/l
Azithromycine	1.090	1.71
Carbamazepine	0.129	2.53
Ciprofloxacine	2.783	46.50
Clarithromycine	1.603	13.78
Diclofenac	0.212	0.19
Ibuprofen	16.300	87.71
Propranolol	0.243	0.52
Sulfamethoxazole	1.830	135.51
Venlafaxine	2.700	2.23

Only between 1 and 4% of the total amount of pharmaceuticals consumed in Denmark are consumed within the healthcare sector (hospitals, Mose- Pedersen et al., 2007). When analysing only a small number pharmaceuticals, i.e. 42 out of more than 1000 different pharmaceuticals used in the healthcare sector, it was expected to find that the environmental impact from discharge directly from the hospital would be dramatically higher than from peoples´ homes. However, the numbers showed that the majority of the environmental impact (84%) was associated to wastewater discharged from the private sector. This leads to the conclusion that wastewater treatment locally at the source (hospital) may fail to materialize the expected environmental effect. Before investing in treatment of wastewater for pharmaceuticals, it is crucial to evaluate both the environmental effect of treatment and the related financial costs for treatment depending on where treatment is applied. Therefore, it is recommended to make more comprehensive investigations between the assumed/assigned pharmaceuticals consumption at hospitals (database) and compare these data with data from EPJ, to validation whether the data from AUH represent an artefact or a general trend.

4. Performance of sidestream- and polishing treatment in bench-scale

4.1 Sidestream treatment at Dept. of Oncology, Aarhus, Denmark

4.1.1 Daily operation of sidestream HYBAS™ bench-scale operation

The overall performance of the bench-scale treatment sidestream, was evaluated according to conventional wastewater parameters. The HYBAS™ treatment was the choice of technology for treating wastewater originating from the Dept. of Oncology, Aarhus University Hospital, denoted NBG. Process set up was: reactor H1 working with only activated sludge and the following reactors H2 and H3 with both activated sludge and carriers with biofilm and finally H4 only with carriers, located after the sedimentation tank and separation of activated sludge. The concentration of chemical oxygen demand, COD, in the period of operation is shown in Figure 9.

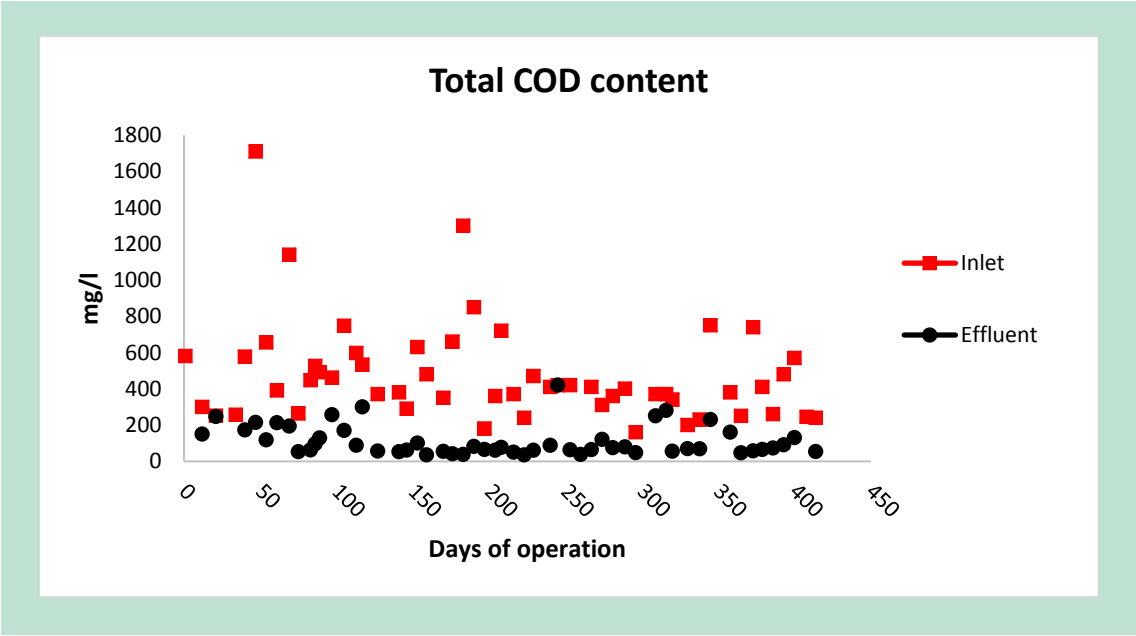


Figure 9. The content of COD in un-filtrated inlet- and effluent samples of the HYBAS™ bench-scale plant.

The unfiltered COD (COD total) was on average 480 mg/l in inlet and 80 mg/l in effluent samples, with variations due to different workload at the hospital and/or periods with rainwater mixing into the sewer. The dissolved fraction of COD in the wastewater was also monitored. Average inlet values of 280 mg COD/l and effluent values of 60 mg COD/l, see Figure 10.

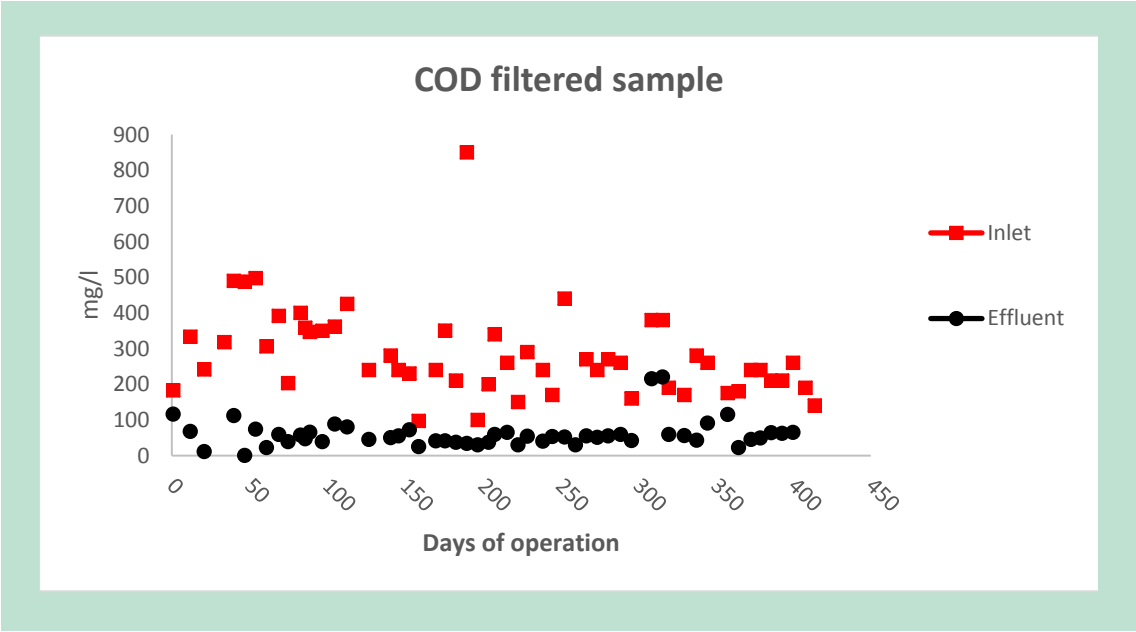


Figure 10. Chemical oxygen demand COD in dissolved inlet and effluent samples from the HYBAS™bench-scale plant.

Ammonium-N, and nitrate-N were also monitored during the period of operation, and values were used to adjust the operation of the HYBAS™-bench-scale treatment plant, see Figure 11. It should be noted that denitrification reactor was not included in this bench-scale set up.

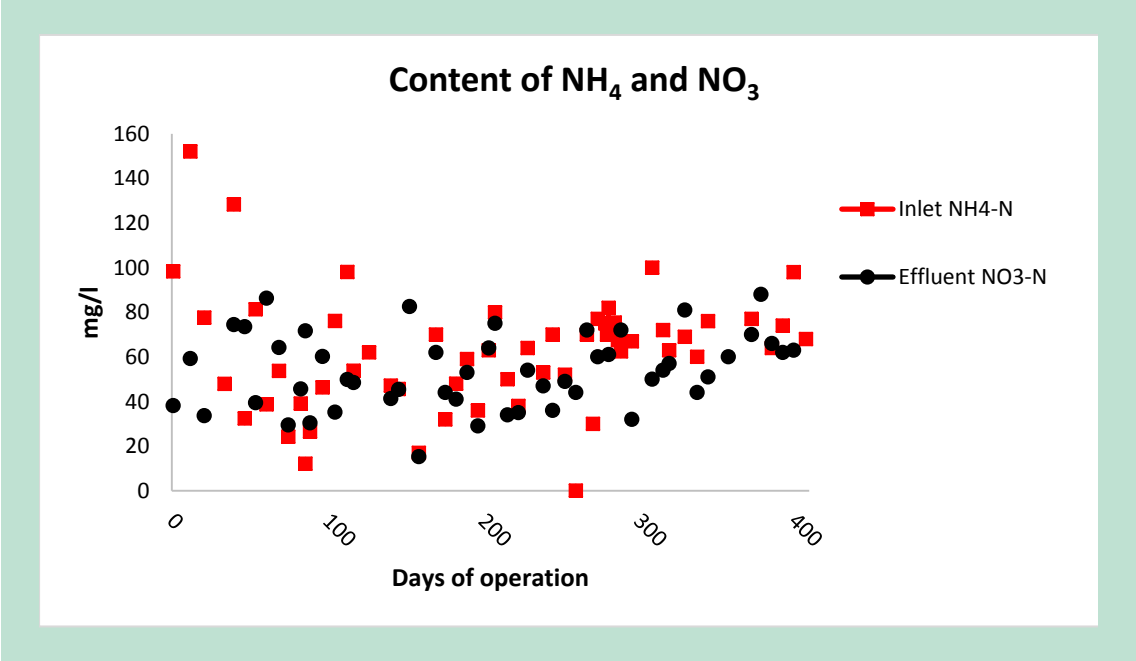


Figure 11. Concentrations of NH_4^+-N and NO_3-N in inlet and effluent samples from the HYBAS™ bench-scale plant.

As can be seen in Figure 11, the inlet concentration of ammonium varies. The concentration of nitrate in the outlet of HYBAS™ bench-plant follows the concentration of ammonium in the inlet. This indicates an effective nitrification where ammonium is oxidized to nitrate.

The amount of biomass on carriers is shown in Table 6. Biomass on carriers was highest in the first HYBAS™ reactor (with both activated sludge and carriers) and decreased in the fol-

lowing HYBAS™ reactor, and the lowest amount of biomass on carriers, were seen on carriers from the polishing MBBR reactor H4. Generally, the biomass content on individual carriers was low.

Table 6. Overview of biomass on carriers from different HYBAS™ reactors from bench-scale treatment.

Biomass in mg/carrier	H2	H3	P
Day 26	-	4.3	2.4
Day 69	8.7	7.9	0.2
Day 155	6.9	5.8	-

4.2 Biological and chemical degradation of pharmaceuticals in the HYBAS™ and Conventional Activated Sludge, CAS systems

The degradation of pharmaceuticals in the HYBAS™ bench-scale plant located at the sewer outlet from Oncology Department at Aarhus University Hospital (NBG) was tested on several occasions. The experimental setup is described in Chapter 1, and further details are available in (Escolà Casas et al., 2015a). Two different approaches enabled a deeper understanding of the removal capacity of the system. The spiking experiments show the potential of the system whereas the concentration profiles show actual removal of pharmaceuticals present at a certain time.

In Figure 12one of the results for biological degradation of eight selected compounds is shown. As evident, the presence of pharmaceuticals prior to spiking was observed and all compounds were degraded at least to some extent in the different reactors of the bench-scale set-up. For comparison, the degradation of the same compounds in activated sludge is also shown (Figure 13).

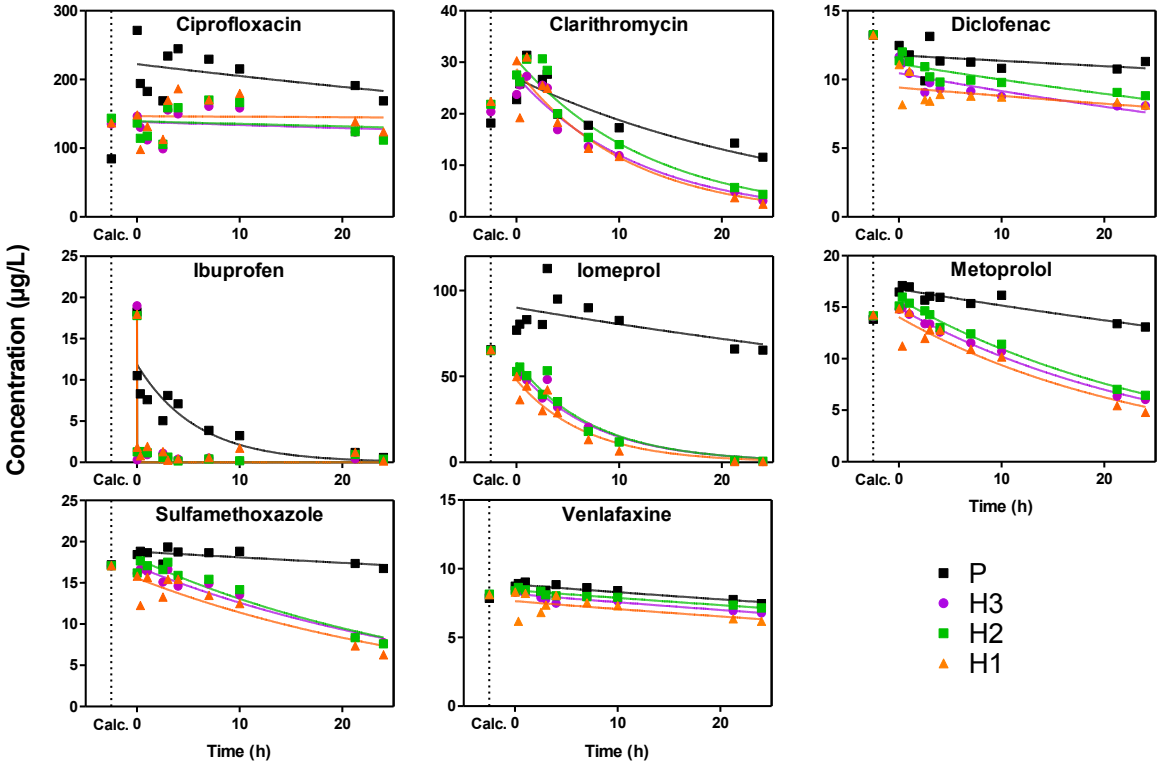


Figure 12. Concentration of selected pharmaceuticals in reactor H1, H2, H3 and P in the batch experiment. Filled lines correspond to a first-order kinetics fitting. Concentrations shown on the vertical dotted-line denote the theoretical concentration based on spiking.

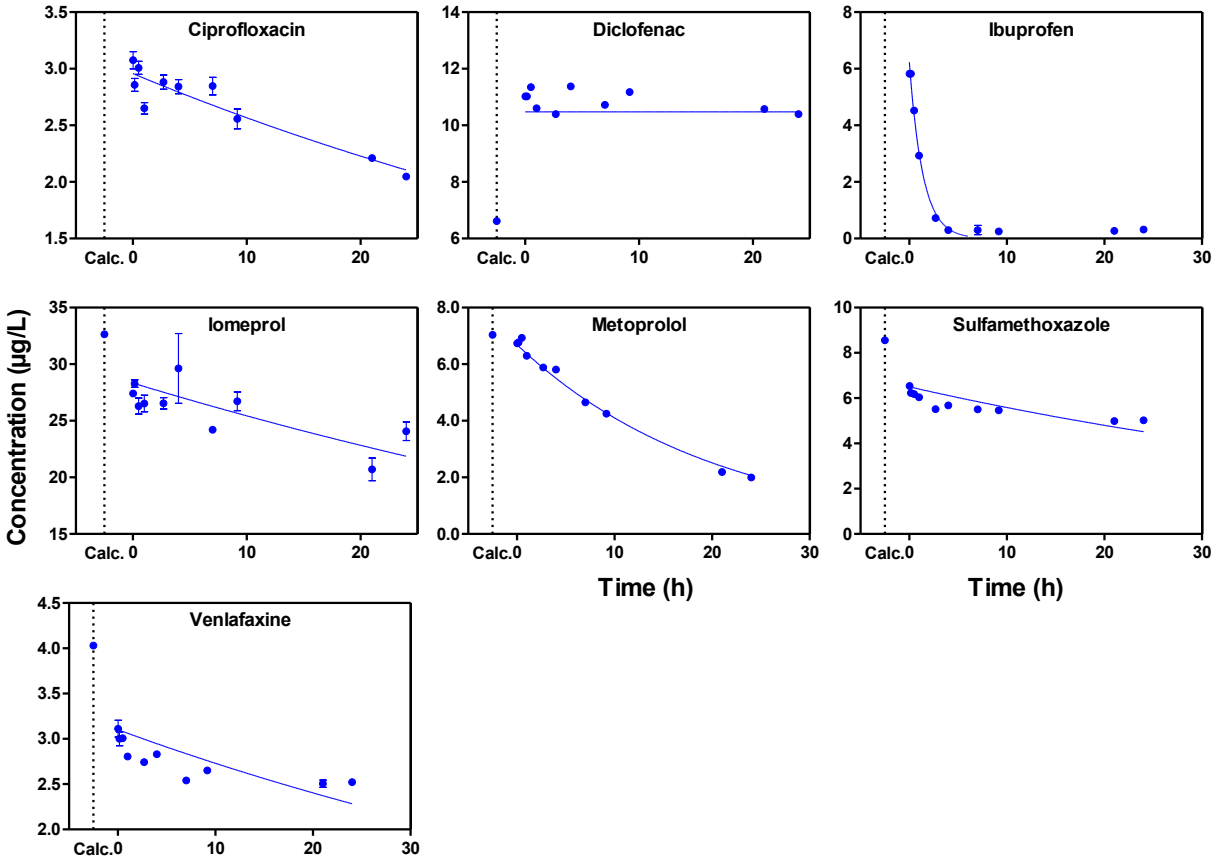


Figure 13. Concentration of selected pharmaceuticals in CAS in the batch experiment. Filled lines correspond to a first-order kinetics fitting while concentrations shown on the vertical dotted-line denote the theoretical concentration based on spiking. Due to analytical complications, clarithromycin was not shown.

The potential removal in HYBAS™ was for most of the pharmaceuticals higher than in CAS. Especially, for the X-ray contrast compounds, the HYBAS™ treatment had the potential to remove 60-70% of iohexol, iomeprol and iopromide, whereas less than 10% removal was observed in CAS.

In general, the potential removal of the pharmaceuticals in the HYBAS™ including the polishing MBBR step was more or less at the same level as in only the HYBAS™ bench-scale system. This indicates that the benefit of having biofilm fixed on carriers is fully utilized in the three reactor systems and the addition of a MBBR polishing step only further reduces some pharmaceuticals.

The removal capacity in actual pharmaceutical concentrations in the wastewater for the individual HYBAS™ reactors at a given day, was also investigated, see Figure 14. Only a selection of data is shown below and all data can be found in the published papers on MBBR and HYBAS system (Casas et al., 2015 a,b).

The continuous flow experiments were conducted at two different time points for HYBAS™ and for CAS, hence direct comparison is not possible. In the HYBAS™ experiment, only 17 out of 27 selected pharmaceuticals were detected. The concentrations in the influent water (H0), the three HYBAS™ reactors (H1, H2 and H3), and in the polishing reactor (P) are

shown. Some of the compounds were removed to a certain extent (ibuprofen and metoprolol) whereas for the other compounds, only minor reduction in concentration was observed.

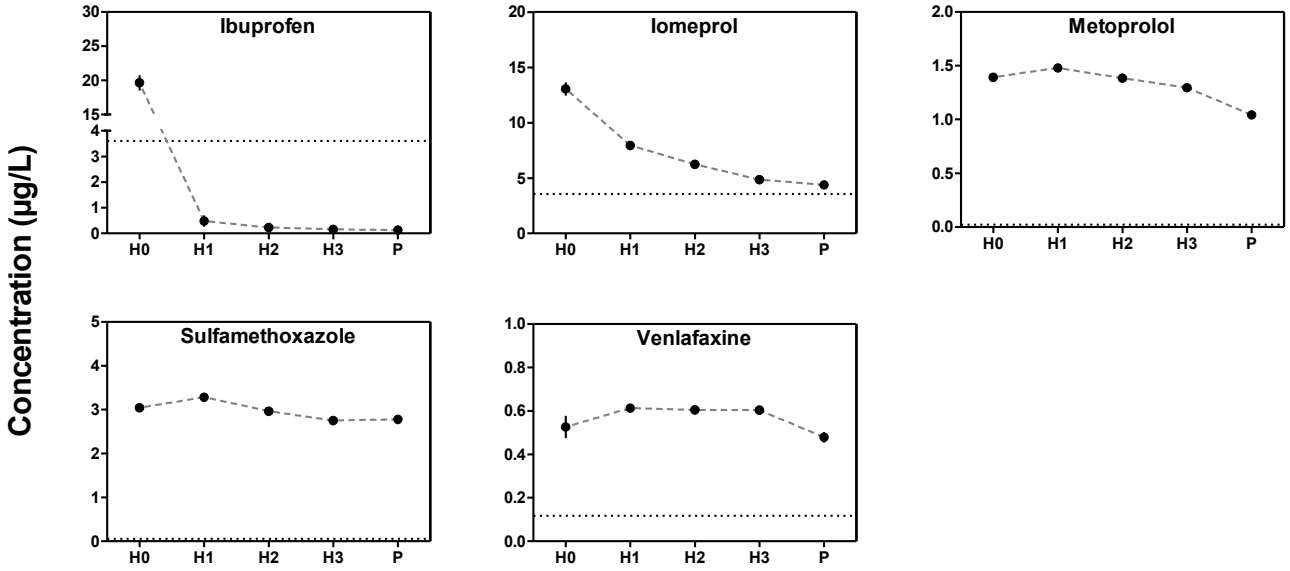


Figure 14. Overview of the concentration of the selected pharmaceuticals through the HYBAS™ with polishing, (due to analytical complications and detection of indigenous concentration below LOQ, 5 out of 8 compounds were shown).

In the CAS experiment, 12 compounds were detected above limit of quantification in the inlet. The concentrations in the inlet and outlet of the CAS are shown in Figure 15 and all compounds detected are available in Casas et al . (2015 a, b). The removal of ibuprofen, iomeprol and metoprolol was observed in the CAS system. Large differences of initial concentrations of the selected compounds (up to factor 2) between the different sampling days were observed.

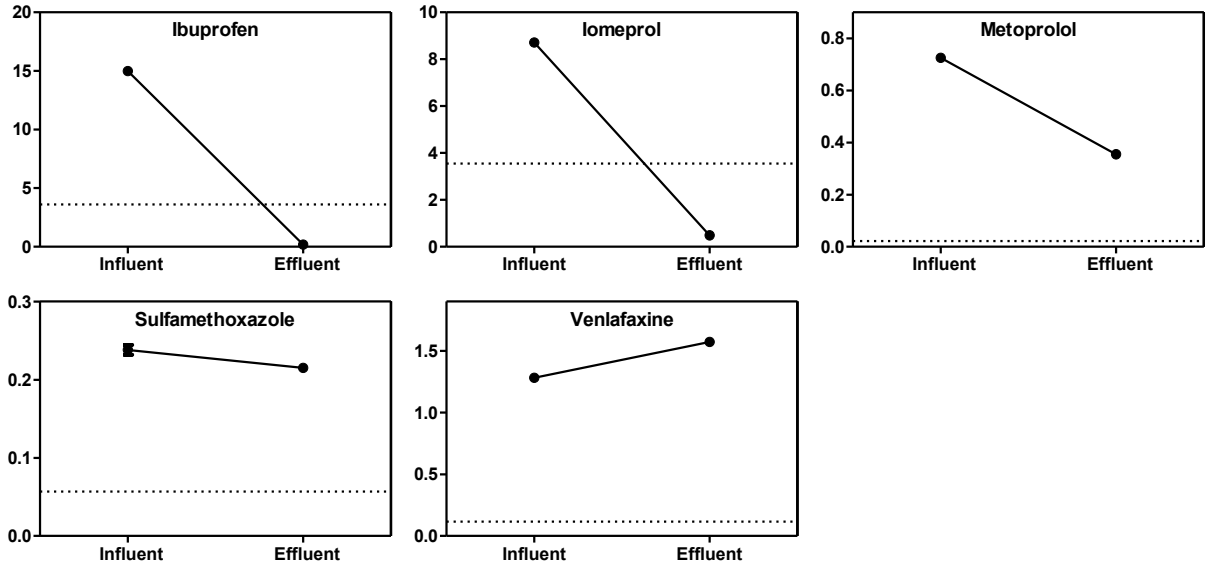


Figure 15. Overview of the concentration of the selected pharmaceuticals in CAS. The dotted line indicates LOQ, (due to analytical complications and detection of indigenous concentration below LOQ, only 5 out of 8 compounds were shown).

In HYBAS™ system, most of the pharmaceuticals were removed (Figure 14). For the following pharmaceuticals, atenolol, clindamycin, ibuprofen, iohexol, and iomeprol all had decreasing concentrations through HYBAS™. The pharmaceuticals propranolol, sulfadiazine and sulfamethoxazole are known to be excreted as conjugates whereas acetyl-sulfadiazine is a known metabolite from other compounds. Of these pharmaceuticals, an increased concentration of acetyl-sulfadiazine and propranolol was observed in the HYBAS™ system compared to inlet concentrations (Figure 14), whereas the concentration of sulfadiazine and sulfamethoxazole seems to be unaffected through HYBAS™. Carbamazepine, citalopram, erythromycin, and trimethoprim have all an increase in concentration in H1 compared to the inlet and then a decrease again in H3 or in the polishing reactor.

In CAS, removal of six out of the 12 detected pharmaceuticals (atenolol, ibuprofen, iohexol, iomeprol, metoprolol, sulfamethizole) was observed. Two compounds (citalopram and propranolol) had an increased level in the outlet compared to the inlet concentration.

Out of the 17 detected pharmaceuticals, 9 pharmaceuticals were found to have a negative removal in the HYBAS™ (H0-H3). This may be due to de-conjugation or formation from metabolites. After the polishing step/reactor (P) only 2 of the pharmaceuticals (acetyl-sulfadiazine and sulfadiazine) still showed a negative removal. When the removal was positive for HYBAS™ (H0-H3) then in most cases no further or only very little increase in the removal was found after polishing reactor (P).

The actual removal capacity of the HYBAS™ system alone and with a polishing step, was identified and compared to CAS, see (Figure 16).

In the HYBAS™ system without polishing, the predicted removal fitted well with the observed for four compounds (sulfamethizole, iomeprol, iohexol and ibuprofen). For clindamycin, the observed removal in the continuous flow experiment was much higher than predicted based on the batch experiment. For the other pharmaceuticals, a much lower removal was observed than predicted. A higher predicted removal than observed indicates that the HYBAS™ system had the potential to remove pharmaceuticals, which are not occurring due to de-conjugation or formation from metabolites. In cases where a polishing MBBR step was connected after the HYBAS™ treatment system, the observed and predicted removal had in general a better fit, as the polishing reactor was hypothesized to remove the de-conjugated pharmaceuticals.

In CAS, a higher removal than predicted was observed in three compounds (sulfamethizole, iomeprol, iohexol). For compounds with a predicted removal higher than 30%, the observed and predicted removal is similar.

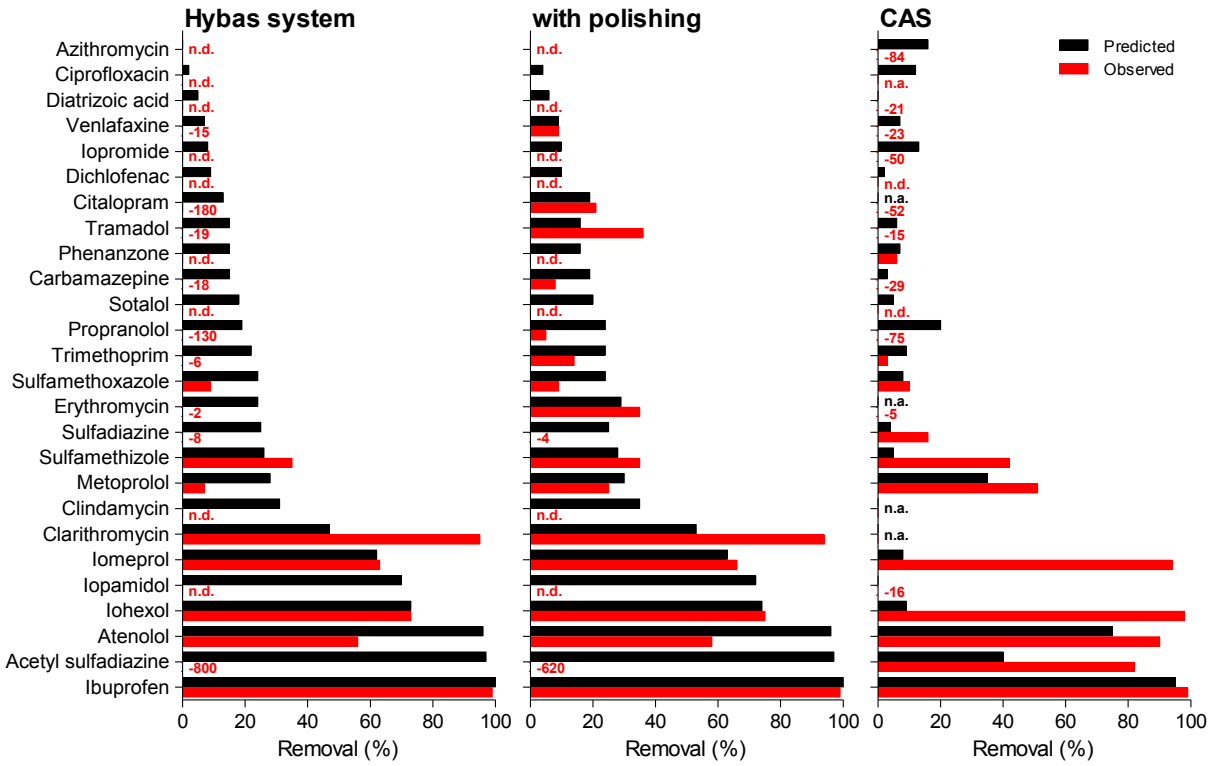


Figure 16. Comparison of the predicted removal and the observed removal of selected pharmaceuticals for HYBAS™, HYBAS™ with polishing and CAS.

In the HYBAS™ system, with and without the MBBR polishing, the removal of the pharmaceuticals was above 20 % whereas in the CAS only few compounds were removed with more than 20%. Acetyl sulfadiazine appeared to be removed to a greater extent in CAS compared to HYBAS™ system. However, the negative removal obtained in HYBAS™ system was a result of a very active system forming acetyl sulfadiazine as a metabolite during degrading of other pharmaceuticals. Thus, in general, the HYBAS™ system performed better in removal of pharmaceuticals than the CAS.

4.3 Results from bench-scale polishing of effluent at Viby municipal WWTP

4.3.1 Daily operation of the polishing MBBR at Viby

Polishing of effluent from a municipal plant with N and P removal with MBBR was carried out at Viby municipal WWTP, Aarhus. The bench-scale test unit (in 40´ container) was placed at the WWTP outlet. The configuration of MBBR treatment was the rotating operation mode as described in Chapter 1.2.2., i.e. an operation principle with the purpose of ensuring access to sufficient organic material to maintain the biofilm but to prohibit overgrowth of slow growing bacteria with specific capabilities. The operation period with access to COD is called the regeneration phase. (Swedish patent application no 1650321-1).

As evident from Figure 17, a very low content of organic matter was present in the effluent of the Viby WWTP corresponding to inlet concentrations to the MBBR polishing step. Only minor differences could be noted in between inlet concentrations and concentrations present in the different MBBR reactors. The reactor in regeneration position, receiving primary clarifier effluent, received a higher content of organic material than the in-line reactors.

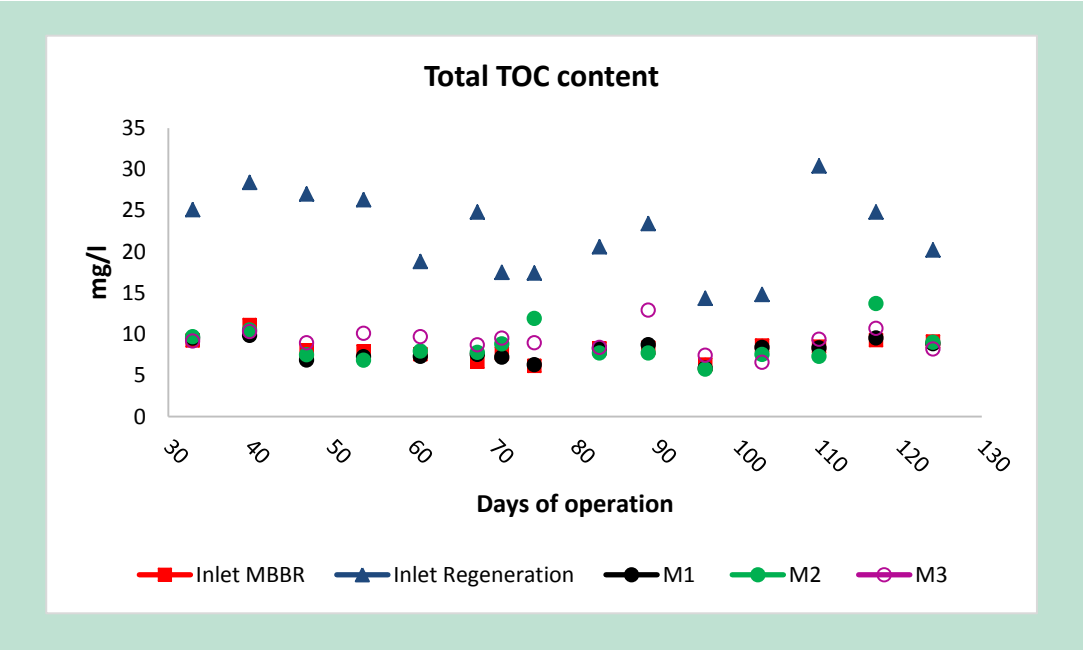


Figure 17. Overview of TOC content in inlet and effluent reactor samples.

The content of ammonia was also analyzed for, as shown in Figure 18. Quite low concentrations were detected in inlet MBBR samples and the concentrations were further reduced throughout the system.

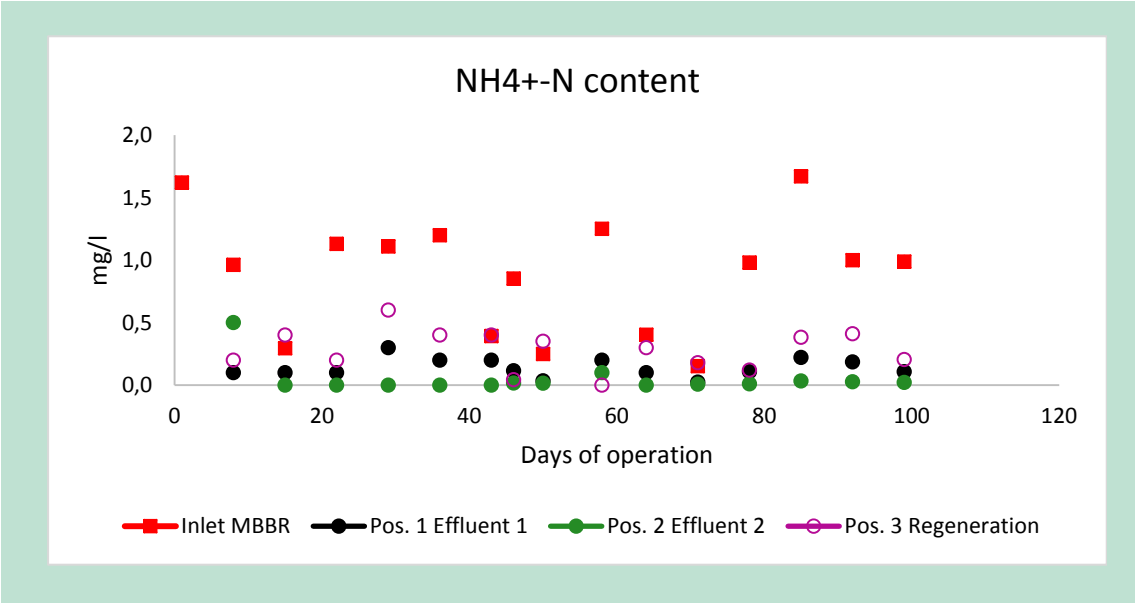


Figure 18. Ammonia content of inlet MBBR and of the different MBBR reactor positions (A, B and C).

The amount of biomass on the carriers in the different MBBR reactors was also quantified, see Table 7. The biomass content was very low reflecting the organic scarcity.

Table 7. Overview of biomass present on carriers from the different reactors.

M1	M2	M3
7.4 mg/carrier	6.2 mg/carrier	5.9 mg/carrier

4.3.2 Biological and chemical degradation of pharmaceuticals

In Figure 19, the biological degradation of the 8 selected compounds is shown. Two pharmaceuticals could not be detected (clarithromycin and Iomeprol) but the remaining selected pharmaceuticals were present in different concentrations prior to experiments. All pharmaceuticals were degraded during the experiment. Based on these results, no differences in degradation capability can be observed in the different positions (A, B and C). Position A is fed by treated effluent, Position B receives effluent from Position A and Position C is in regeneration position and receives primary clarifier effluent.

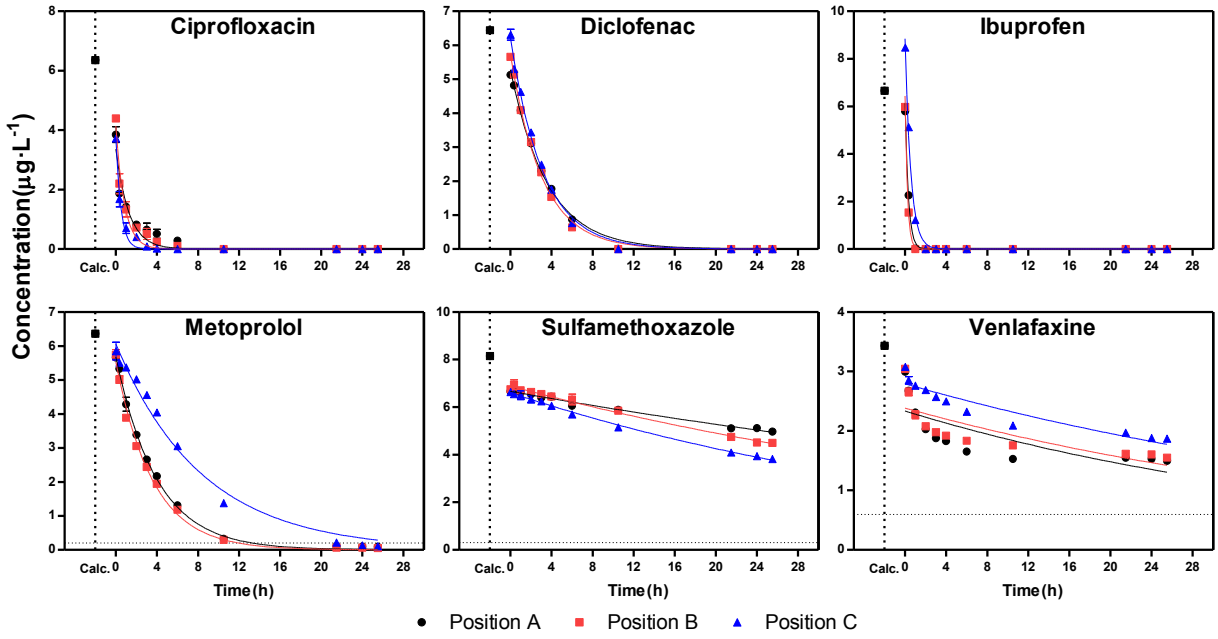


Figure 19. Concentrations of selected pharmaceuticals during spiking experiment showing potential removal capacity in each reactor (Position A, B and C). Filled lines correspond to a first-order kinetics fitting. Black squares represent the theoretical concentration of individual pharmaceuticals based on stock solutions, (due to analytical complications, e.g. a shift in the retention time, clarithromycin and iomeprol were left out).

The potential removal capacity of each reactor is depicted in Figure 20. Only positions A and B are shown, and both positions are equally capable to degrade the selected pharmaceuticals with a minor superiority of position B for degrading sulfamethoxazole.

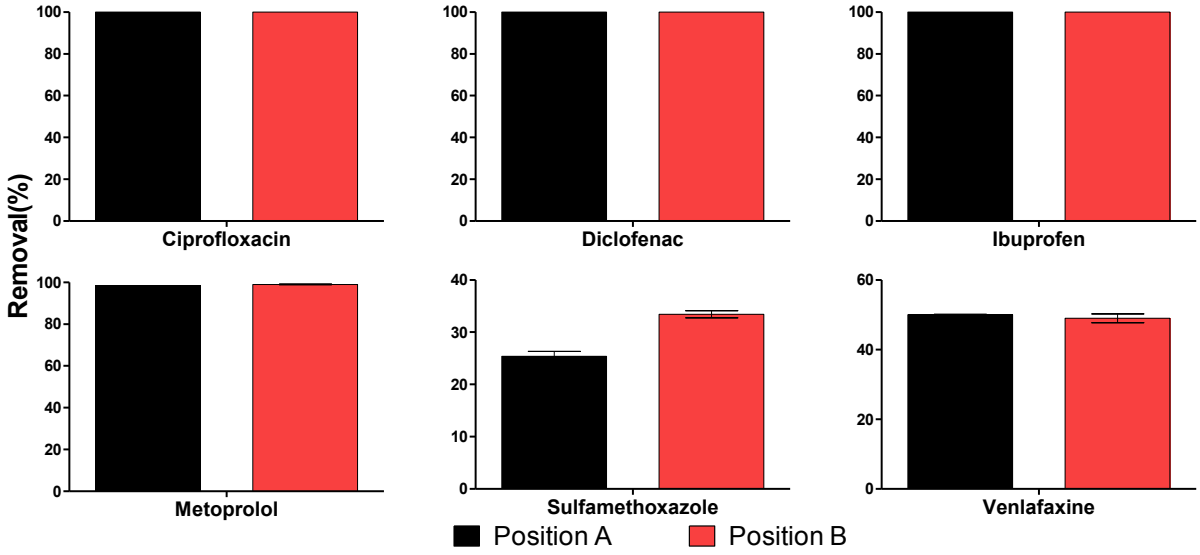


Figure 20. Potential capacity of the two positions, for removal pharmaceuticals over a period of 24 hours. Position A is fed from effluent waters, and position B from the effluent of position A, (due to analytical complications, e.g. a shift in the retention time, clarithromycin and iomeprol were left out).

The removal capacity at actual concentrations of the individual positions at a given day was also investigated, see Figure 21. As evident, very low concentrations of the selected pharmaceuticals were in most cases present in the inlet to the reactors. The pharmaceuticals were removed to concentrations below LOQ. Clarithromycin was not detected during the experiment.

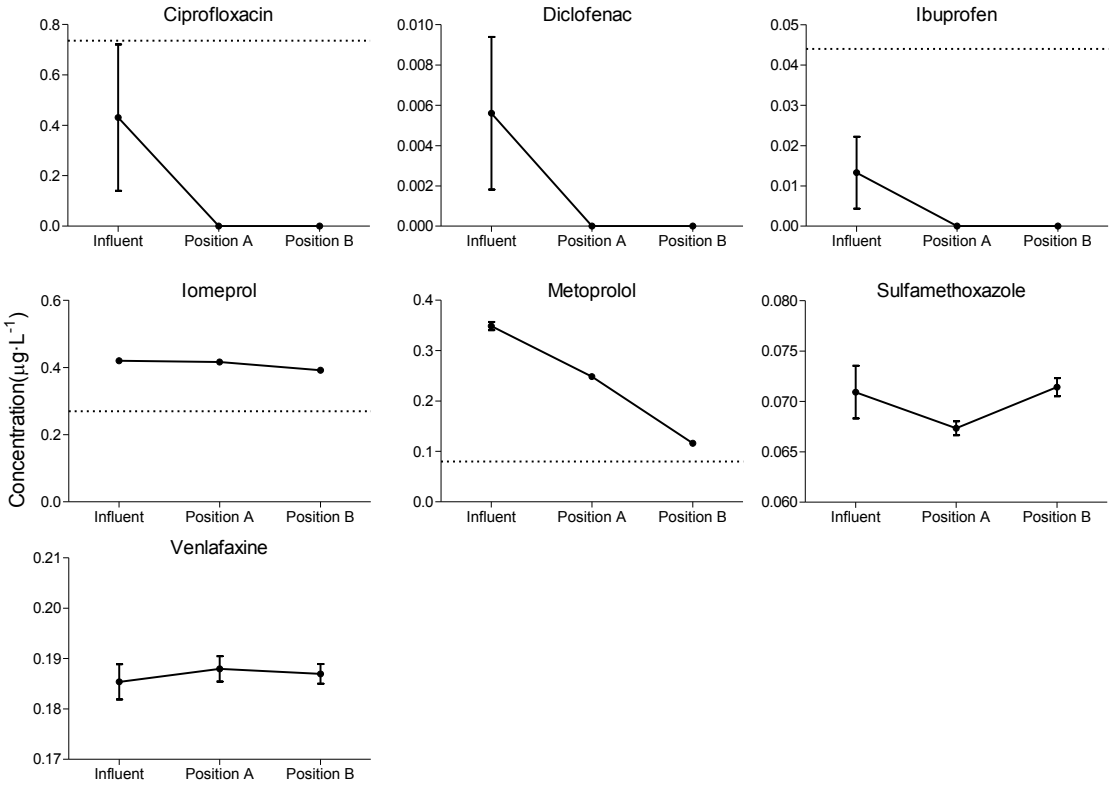


Figure 21. Average concentrations and SD (n=2, each sample was analyzed twice) in the polishing reactors during the continuous flow experiment. The dotted lines indicate the limit of quantification (LOQ), and Clarithromycin was detected below LOQ).

Calculation of the actual biological removal of tested pharmaceuticals in percentage at different retention times was also performed, see Figure 22.

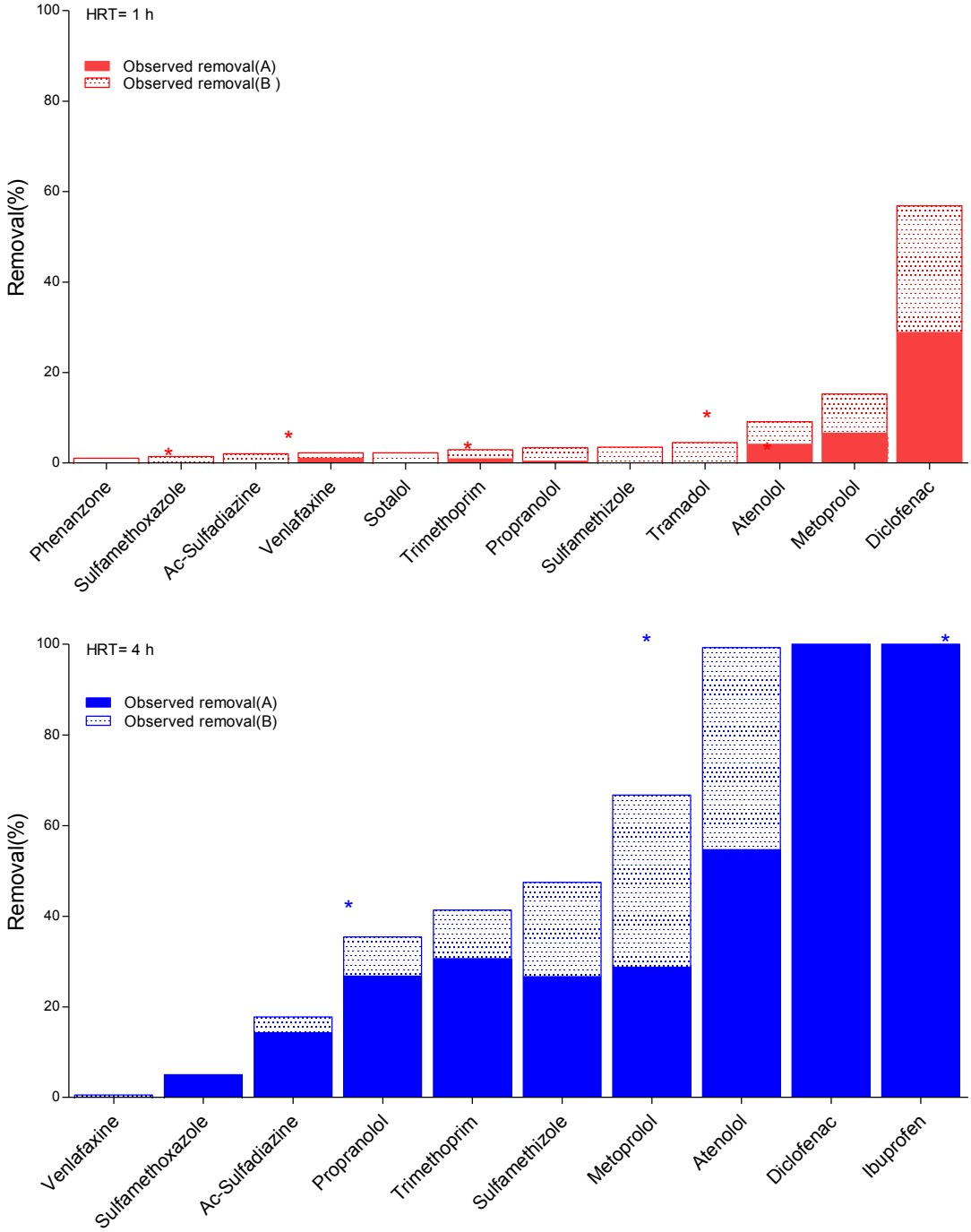


Figure 22. Measured removal from the continuous flow experiments at two different HRT (1 h and 4 h) over the polishing reactors (position A and B), shown as percentage degradation. A star indicates that compound concentration was below LOQ.

The treatment capability was also evaluated according to the proposed guiding limit values. Indigenous influent concentration (without spiking) and effluent concentration (after the treatment system) were analyzed (Figure 23). As evident, all of the selected pharmaceuticals are removed to concentrations lower that the guiding limit values using only biological treatment. In addition, very high removal rates were observed for diclofenac.

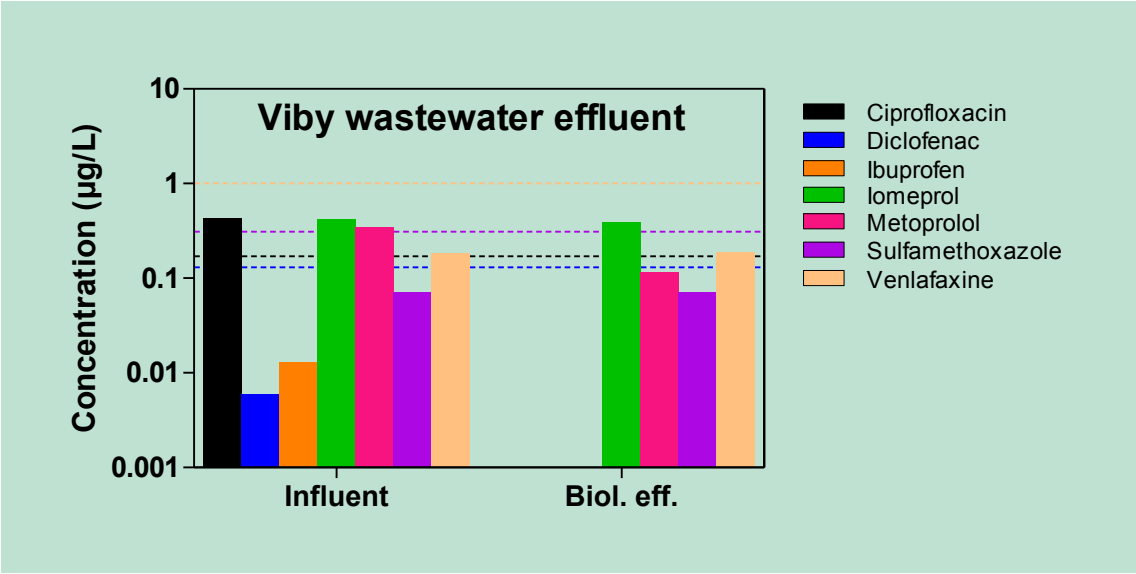


Figure 23. Analysis of concentration of the continuous flow experiment showing the indigenous influent concentration (without spiking) and the effluent concentration after the MBBR treatment (the horizontal lines represent the proposed guiding limit values for each compound).

4.3.3 Comparison of removal efficiency in sidestream treatment of hospital wastewater with polishing MBBRs in bench-scale reactors

In order to compare the efficiency of pharmaceutical degradation in different systems, the results obtained for sidestream treatment of hospital wastewater using different treatment technology; MBBR (results available in Casas et al., 2015b), CAS, and HYBAS™ are compared with the results obtained with polishing MBBR reactors operated according to the rotating principle (see Figure 24). All tests performed in the bench-scale test unit. As evident from the figure, many of the compounds were not detected and therefore comparison is difficult.

In the polishing MBBR (for WWTP effluent) step, efficient removal of ciprofloxacin, diclofenac, ibuprofen and metoprolol was achieved. However, no removal of venfalaxine or sulfamethoxazole was observed. For the MBBR treating hospital wastewater, no removal was observed for ciprofloxacin, diclofenac, metoprolol and sulfamethoxazole (data from Casas et al., 2015). HYBAS™ treatment of hospital wastewater removed ibuprofen, and iomeprol and none of the remaining compounds. CAS removed ibuprofen, iomeprol and metoprolol but none of the remaining compounds. All in all, the most efficient treatment technology investigated in bench-scale was identified as polishing MBBR reactors with alternating operation, and the least efficient treatment was obtained using conventional activated sludge.

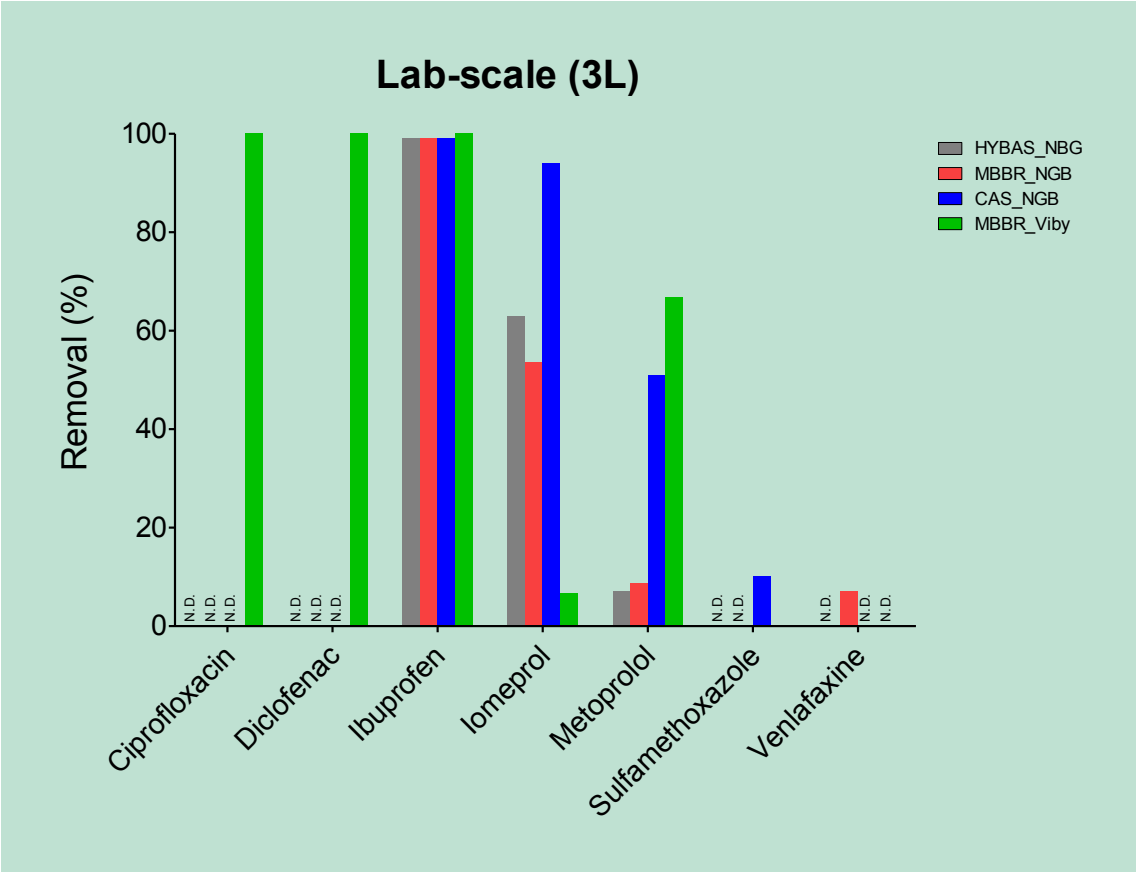


Figure 24. Comparison of pharmaceutical degradation using different treatment technologies (HYBAS™, MBBR, CAS and Polishing MBBR) shown as percentage in bench-scale.

The removal capacity was also compared between the different bench-scale treatments in accordance to the proposed guiding limit values (Figure 25). The inlet concentration of the selected pharmaceutical varied to a great extent and many of them were present in concentrations below the proposed guiding limit values for individual treatment technologies. These differences can be explained solely by the location for treatment (e.g. hospital wastewater and municipal effluent wastewater). Interestingly, the presence of iomeprol (X-ray contrast media) is observed in all investigated bench-scale experiments and not at very different concentrations, supporting the findings described in Chapter 3.2 that people after treatment at the hospital discharge the pharmaceuticals in their own homes and thus the compounds are seen in municipal wastewater as well as in hospital wastewater.

The concentration of the compounds varies, which emphasizes the fact that comparison between different treatment technologies is difficult when experiments are not conducted at the same time.

Only some of the experiments are depicted in this report, and in depth analysis of the different treatment performances can be found in Casas et al., 2015a, b, and Tang et al., 2017.

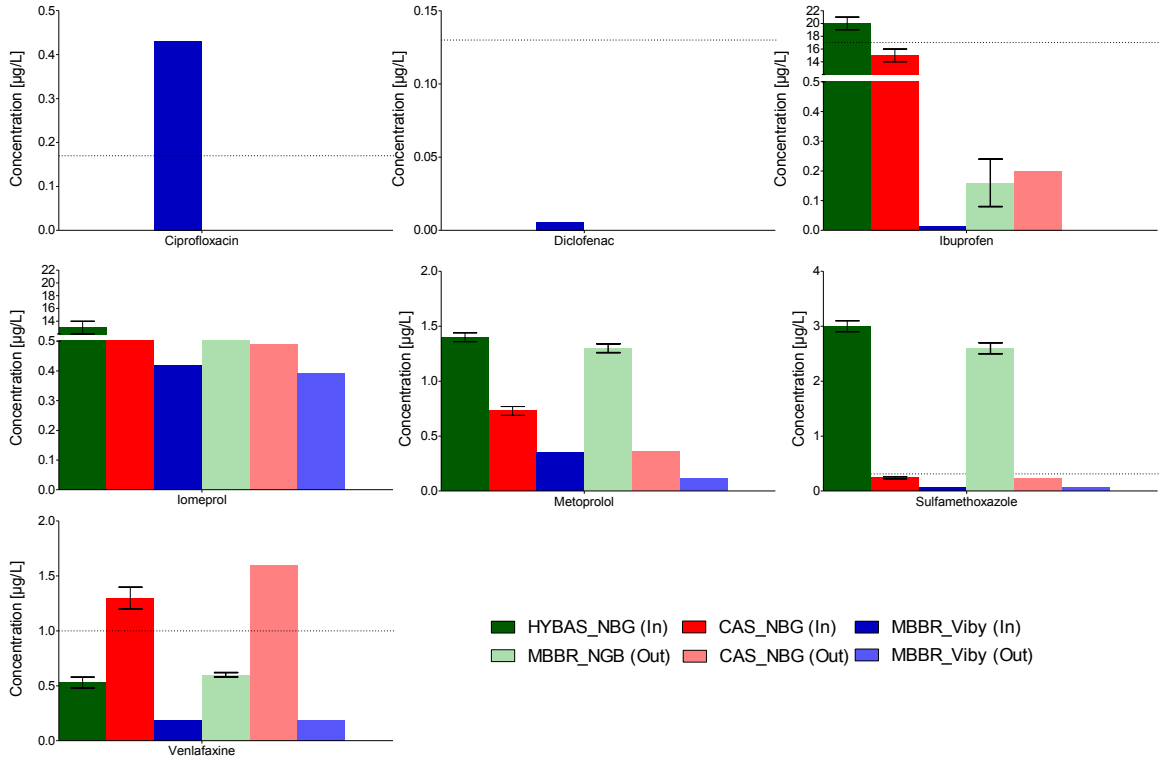


Figure 25. Native concentration of pharmaceuticals present in influent and effluent after biological treatment at different bench-scale treatment sites. MBBR data originated from Casas et al., 2015a. The horizontal lines represent the Danish proposed guiding limit values for each compound. No limit yet proposed for metoprolol and iomeprol.

4.4 Conclusion

Both a train of reactors including activated sludge and biofilm (HYBAS^{TM}) and a pure biofilm system, MBBR, reached higher removal efficiencies of pharmaceuticals, compared to conventional low-loaded activated sludge CAS, particularly when considering specific difficult degradable compounds (e.g. diclofenac).

Additionally, the staged treatment systems reached a low content of organic matter through a consistent and small stepwise removal throughout the entire treatment train. Furthermore, effluent from this process resulting in low DOC makes it more suitable for a cost effective polishing ozonation in case further reduction of pharmaceuticals is required.

Removal of ibuprofen reached as predicted approximately 100% removal in HYBAS^{TM}, MBBR as well as CAS in the treatment systems. It shall however be noted, that 100% removal of diclofenac, one of the pharmaceuticals considered as very difficult to biodegrade, was obtained in the polishing MBBR operating according to the rotating principle. This polishing MBBR system was intermittently fed by biological untreated wastewater resulting in a more adapted biofilm for degradation diclofenac.

Degradation in small pilot-scale treatment systems cannot necessary directly be mirrored in full-scale treatment. Therefore, a larger pilot-scale plant with 1 m^3 reactors was built in order to verify the removals before applying these treatment systems in full-scale.

5. Development and performance of the pilot-scale plant treating hospital and municipal wastewater

5.1 Treatment of entire wastewater from hospital (DNU)

5.1.1 Daily operation of MBBR pilot-scale operation at DNU

The overall performance of the biological MBBR pilot plant was evaluated based on conventional wastewater parameters. The MBBR technology was applied for treating wastewater from AUH hospital. Two 40´ containers with the entire pilot plant inside were located at the sewer outlet from the hospital. Wastewater was pumped from a well on the sewer line into pilot plant. The design of the pilot plant is shown in Chapter 1.2.3.

Figure 26 and Figure 27, show the total COD and filtered COD (COD in solution) in inlet samples (total COD average 767 mg/l and filtered sample COD average 428 mg/l). Total COD concentration in effluent from the biological treatment in the pilot plant was on average 60 mg/l and in filtered sample 44 mg/l. The inlet TOC level also varied with an average concentration of 48 mg/l and effluent concentration of 23 mg/l, see Figure 28.

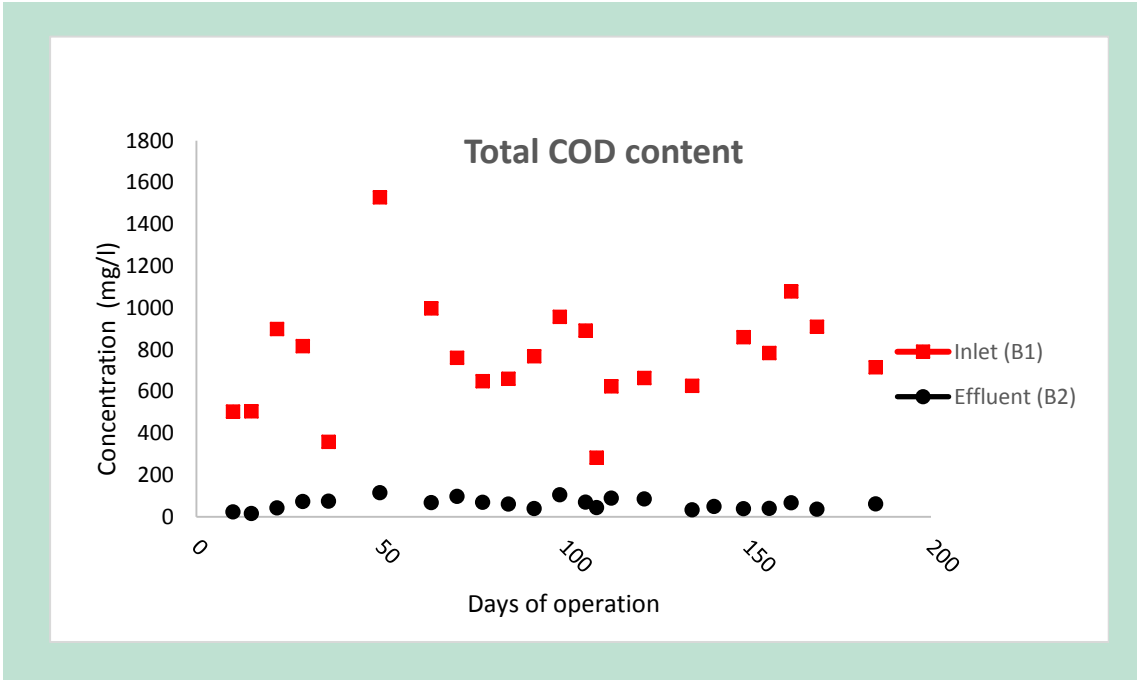


Figure 26. Overview of the total COD concentration in inlet and outlet samples during MBBR pilot plant operation.

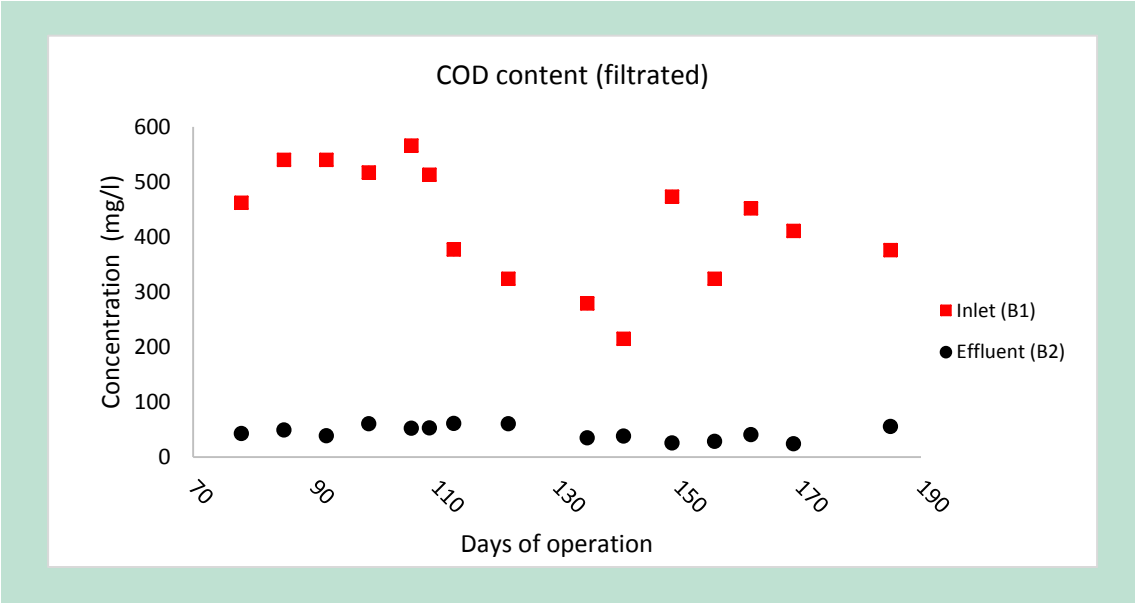


Figure 27. Overview of the filtrated COD concentration in inlet and outlet samples during MBBR pilot plant operation.

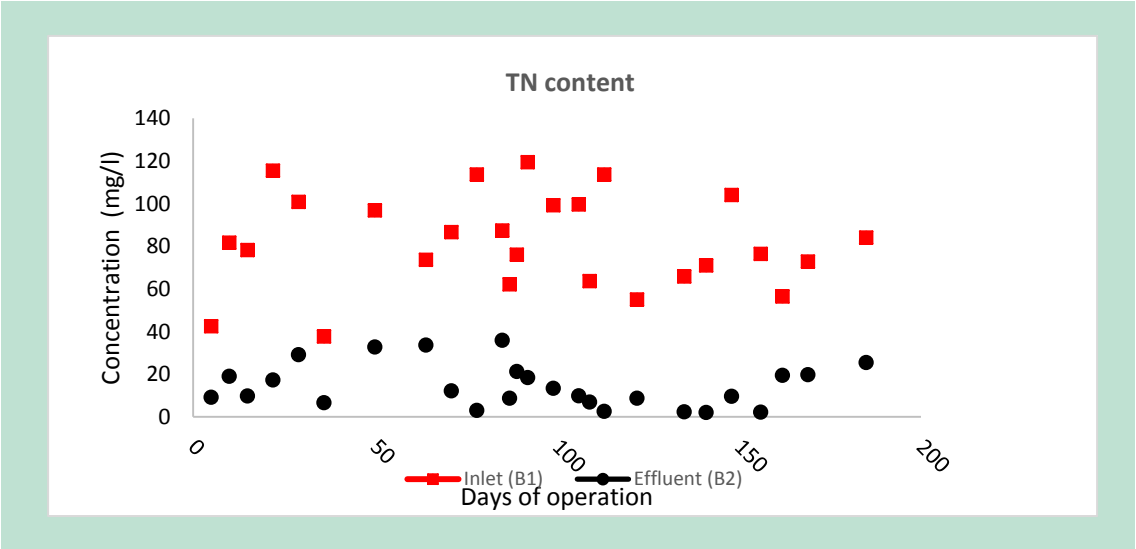


Figure 28. Overview of total TOD content in inlet and effluent samples during MBBR pilot plant operation.

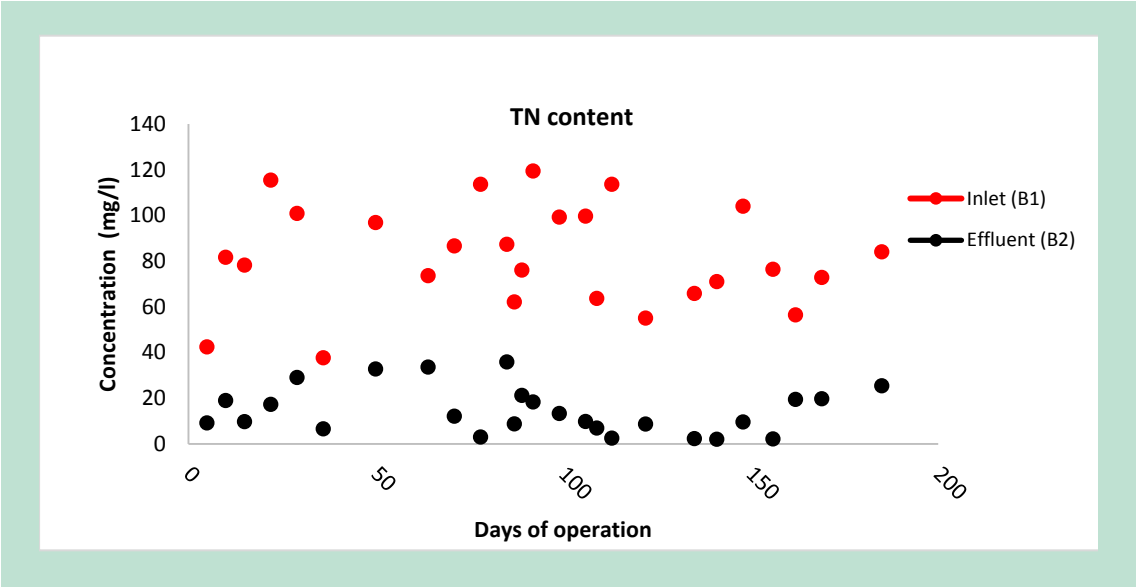


Figure 29. Overview of TN content in inlet and effluent during MBBR pilot plant operation.

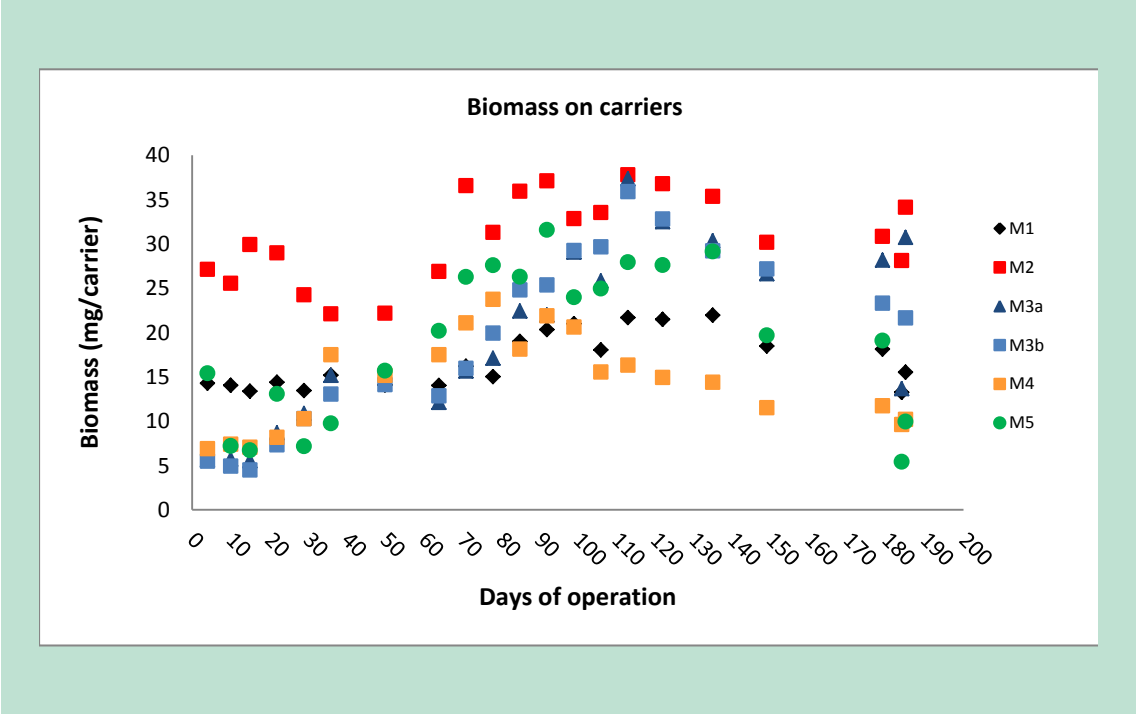


Figure 30. Development of biomass on carriers from the different reactors. Black and orange represent anaerobic reactors.

The level of total nitrogen (TN) and total phosphorus (TP) were also monitored throughout the test period; Figure 29 and Figure 30. Total inlet TN concentrations was average 82 mg/l and the TN in effluent was average 14.8 mg/l, where the limit value of Denmark is set at 8 mg/l. It should be noted that the pilot was operated with full nitrification and the main part of nitrogen in effluent was in form of nitrate. As the pilot plant was operated with focus on removal of pharmaceuticals and not in particular the nitrate removal, which is well known, no further attempts were made to reduce nitrate.

The development of biomass on carriers in the different MBBR reactors were determined on a weekly basis, see Figure 30. In general, the biofilm thickness on carries from the anaerobic reactors (M1 and M4) were smaller, compared to the biomass on carriers in aerobic reactors.

5.1.2 Biological and chemical degradation of pharmaceuticals

The curves for all compounds achieved from the batch experiment (spiking tests Chapter 2.4.1) were plotted as a function of time, showing the potential removal capacity of the system. Results for selected pharmaceuticals (ciprofloxacin, clarithromycin, diclofenac, ibuprofen, iomeprol, metoprolol, sulfamethoxazole and venlafaxine) are represented in Figure 31. Higher concentrations observed than the calculated one, confirm the presence of the pharmaceutical in the wastewater. All compounds were degraded to some extent, except for ciprofloxacin. For diclofenac and venlafaxine, known as recalcitrant compounds, better biodegradation were observed under nitrifying conditions (M2, M3A/M3B and M5) compared to denitrifying conditions (M1 and M4).

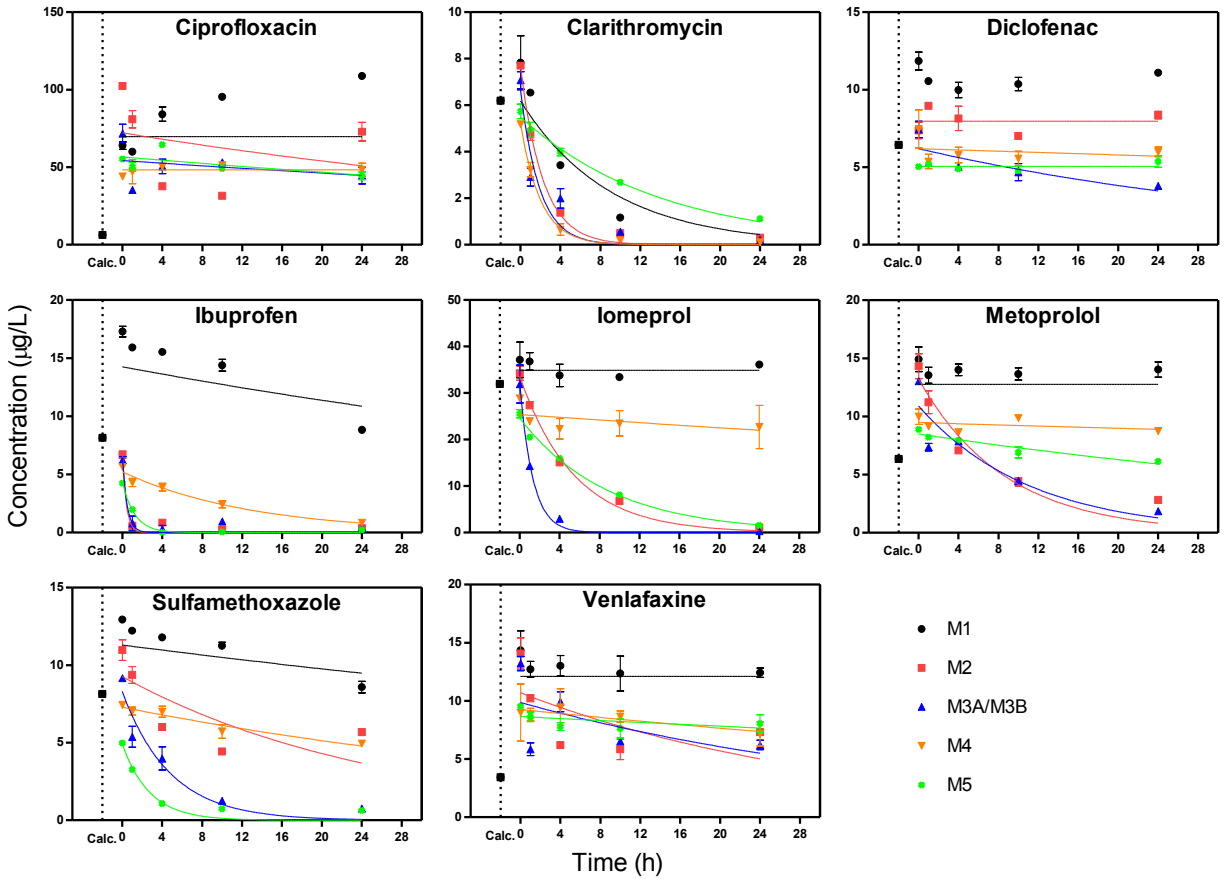


Figure 31. Concentrations of selected pharmaceuticals during spiking batch experiment in each reactor (M1, M2, M3A/M3B, M4 and M5). Filled lines correspond to a first-order kinetics fitting. M 1 and M4 are denitrifying, the remaining reactors are aerobic. Black closed square symbols in the figure denotes the theoretical calculation of individual pharmaceutical concentrations according to stock concentrations of pharmaceuticals.

The potential removal capacity of each reactor step is depicted in Figure 32 for selected compounds. As evident, degradation of pharmaceuticals was occurring in all reactors of the MBBR treatment train on hospital wastewater. In general, the two anoxic reactors (M1 and M4 showed in black and orange) showed a lower ability to degrade pharmaceutical than the aerobic ones.

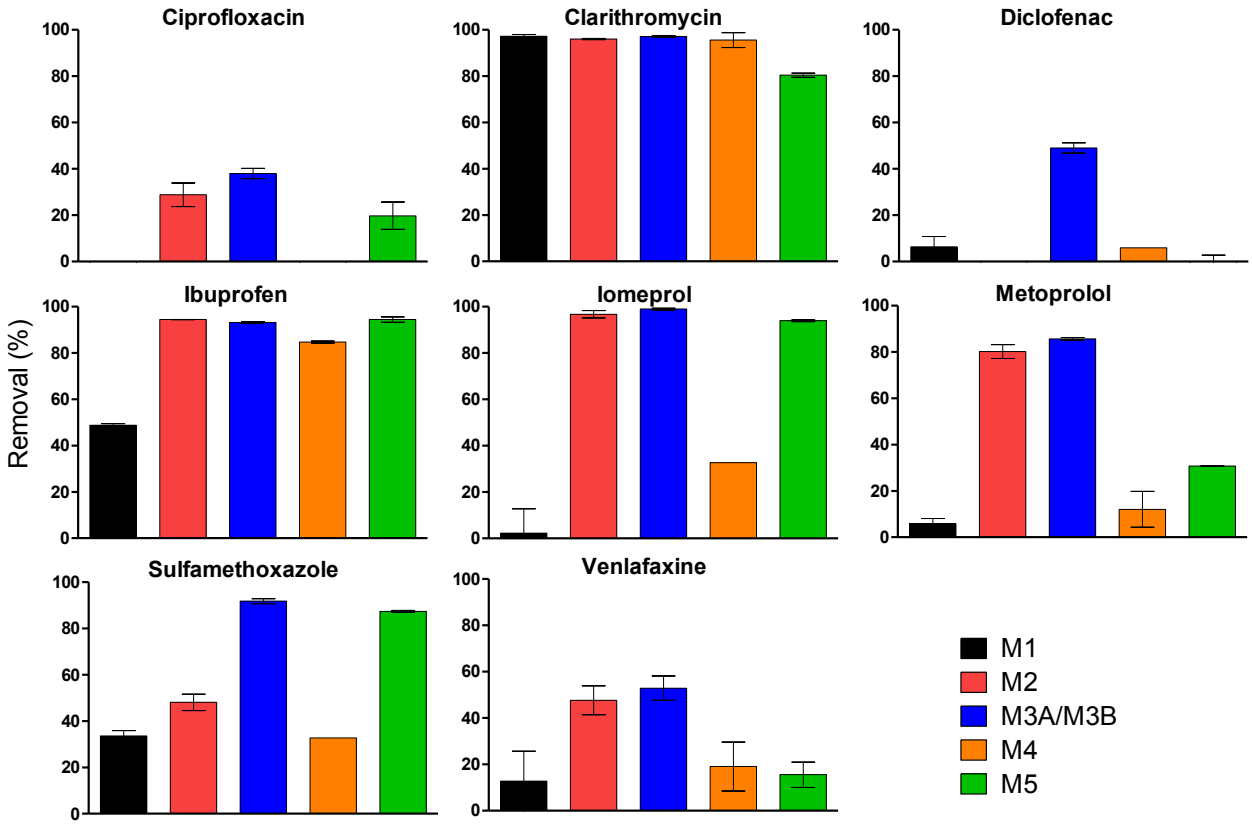


Figure 32. The potential capability of each reactor to remove pharmaceuticals over a period of 24 hours. Reactors M1 and M4 (black and orange) were denitrifying whereas the remaining reactors were aerobic.

The actual removal capacity of the individual MBBR reactors at a given day was also investigated, see Figure 33. Very different inlet concentrations of the selected compounds were seen. For most pharmaceuticals, a clear degradation could be observed, however, metoprolol only to a minor extent. For some compounds, higher concentrations were observed in the different reactors compared to inlet concentrations. This could be due to deconjugation where a biological or chemical transformation of a compound results in detection, as observed for iomeprol and sulfamethoxazole.

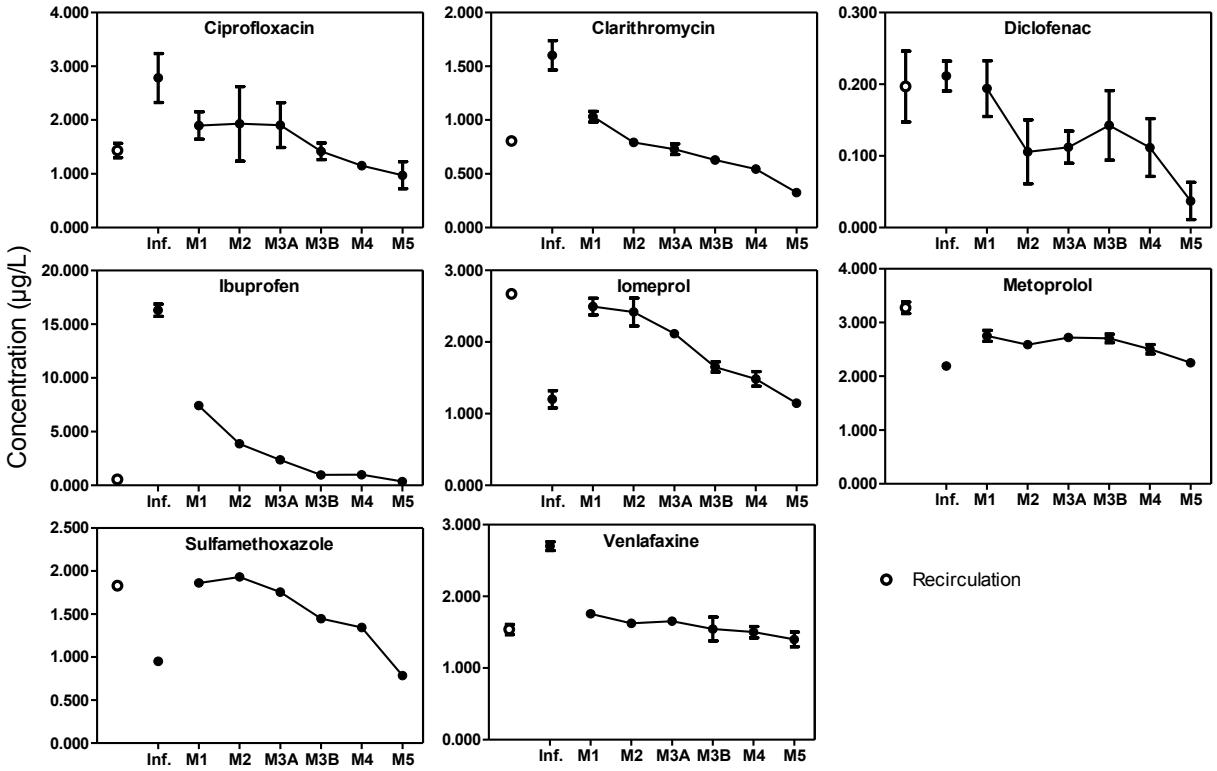


Figure 33. Average concentrations and standard deviations (n=3, each sample was analyzed twice) in the different MBBR reactors during the continuous flow experiment. The impact on pharmaceutical concentration was measures as recirculation concentration in which concentrations in M1 include both recirculated wastewater from M3B and influent (inf.).

An example of the actual removal capacity of the entire biological treatment is depicted in Figure 34. Three of the 22 tested compounds were removed to lesser extent than 20% and thus, not considered to be degraded. For 11 compounds, 50% or higher removal were observed including several contrast media compounds, carbamazepine and diclofenac.

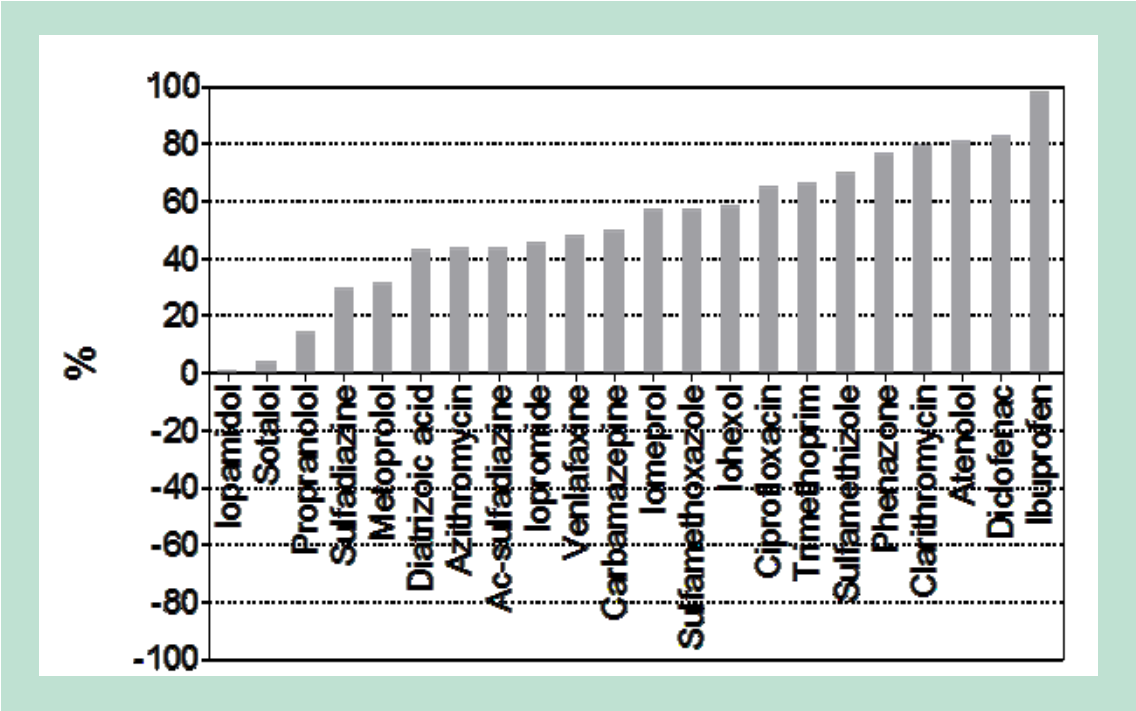


Figure 34. Measured removal from the continuous flow experiment over the pilot-scale MBBR treatment plant, shown as percentage degradation.

These data reflect actual measured removal by the biological system on the particular sampling day. The concentration of pharmaceuticals was also compared to proposed Danish guiding limit values. Native influent concentration (without spiking) and effluent concentration after the biological treatment system were recorded (Figure 35). Diclofenac, in particular, was degraded to a level where the residual concentration was below the proposed guiding limits and this by only passing biological treatment.

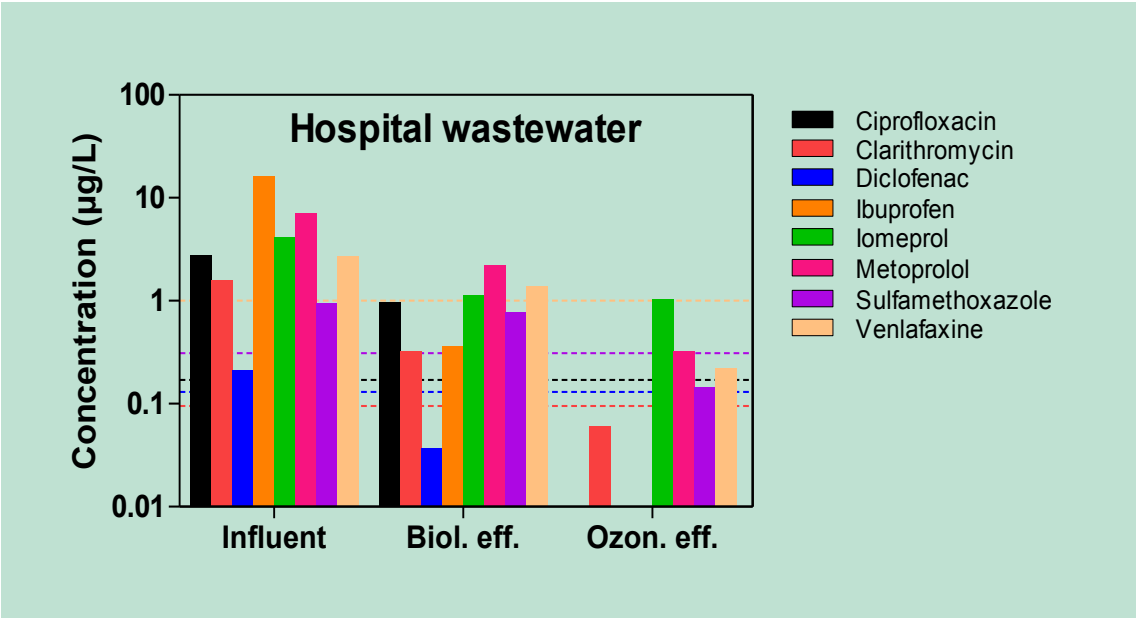


Figure 35. Concentration of pharmaceuticals present in the native influent, in the effluent after biological treatment and after ozonation. The horizontal lines represent the Danish proposed guiding limit value for each compound except for metoprolol and iomeprol where no limit exists.

The biological treatment in the MBBR pilot plant efficiently reduced the concentrations of pharmaceuticals compared to inlet. Pharmaceuticals still above the guiding limit values after biological treatment (ciprofloxacin, clarithromycin, venlafaxine and sulfamethoxazole), are efficiently removed during the ozonation. After ozonation treatment, all the selected compounds fell below the guiding limit values.

5.1.3 Ozonation experiments at DNU

In this experiment, ozonation treatment conducted in lab-scale was compared to ozonation in pilot-scale. As evident in Figure 36, the pilot-scale ozone treatment had slightly higher removal efficiency compared compared to lab-scale ozone treatment, which could be explained by a better mixing of the ozone microbubbles in pilot-scale ozone reactor compared to lab-scale reactor. Diclofenac was not detected in the sample.

Reduction of residual concentrations of pharmaceuticals present after the staged MBBR treatment is shown in Figure 36. Selected pharmaceuticals were present in the wastewater and the dosage (DDO_3) required for 90% removal of the individual pharmaceuticals was identified. The concentrations of all investigated compounds decreased with increasing ozone dosage. The required ozone dosage to reach 50% of pharmaceutical removal was around 8 mg O_3/L in the pilot-scale plant and 10 mg O_3/L in lab-scale plant, respectively.

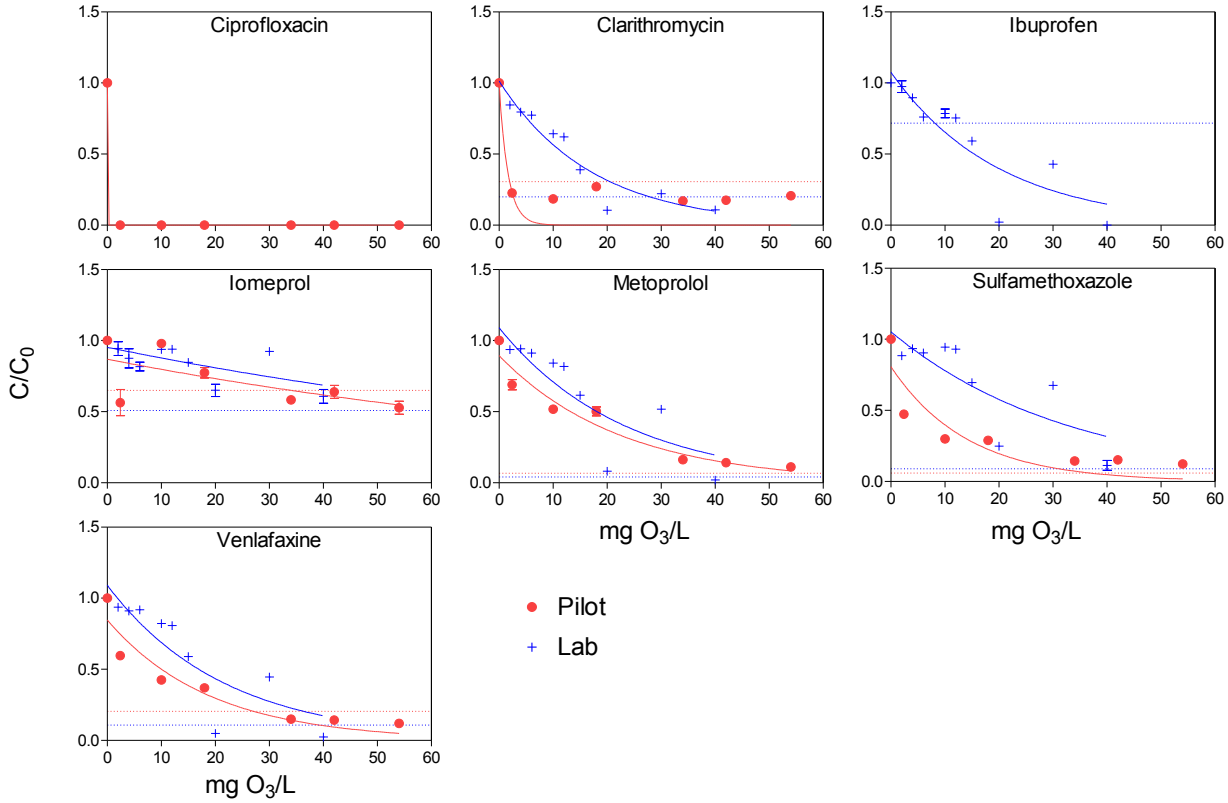


Figure 36. Pharmaceutical removal by ozonation in the effluent of the staged MBBR pilot plant using either the onsite continuous pilot ozonation system or batch treatment in bench scale. Dashed lines correspond to limit of quantification (LOQ) by HPLC-MS/MS for each experiment. Error bars represent standard deviations.

A more simple method was applied to detect humic-like and protein-like substances present in wastewater, based on fluorescence. By differentiating between different emission and excitation spectra, humic substances (Ex335&Em450) and protein-like substances (Ex 275&

Em340) could be detected. Based on this, deducing the protein content out of the humic content as observed by the correlation between the substances, (R^2 = 0.97).

Experiments were carried out on M5 effluent (outlet of biological MBBR treatment). Natural fluorescence in ozone treatments in pilot-scale and in bench-scale treatment revealed that the pilot-scale ozonation seemed more efficient compared to the lab-scale treatment (Figure 37). As observed, the fluorescence intensity decreased with increasing ozone dosages in both pilot and bench scale experiments.

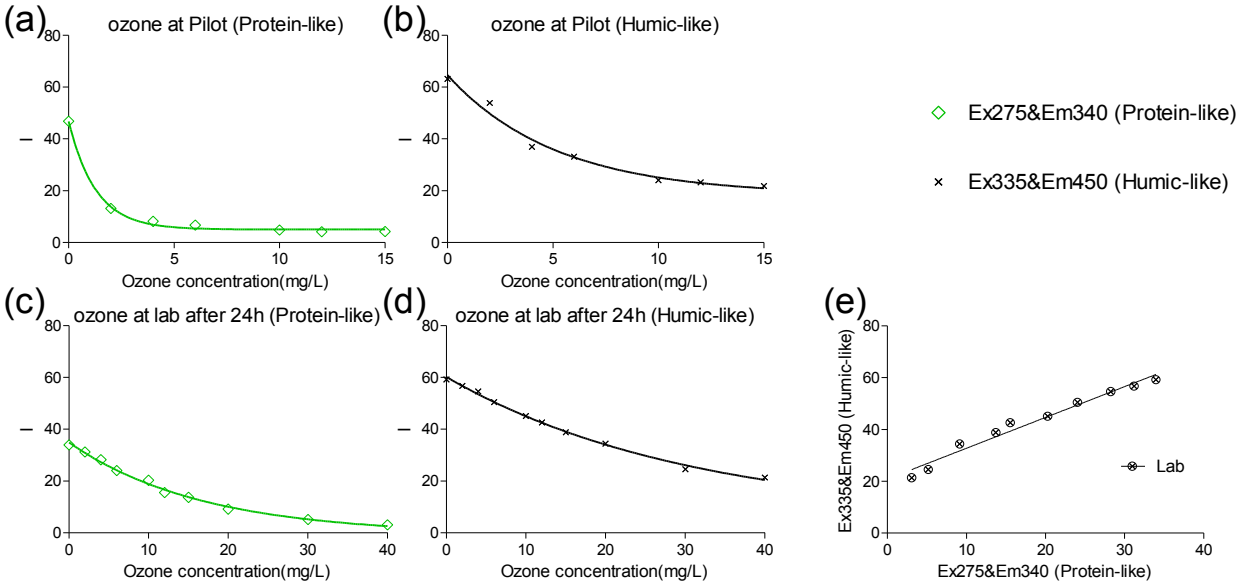


Figure 37. Comparison of remaining natural fluorescence of M5 effluent treated by ozone using the pilot or bench method. The correlation of the two fluorescence wavelengths ($\lambda_{275, 340}$) and ($\lambda_{335, 450}$) was fitted by a straight line so correlations between lab scale and pilot-scale experiments were possible.

The MicroTox effect is shown in Figure 38, where the inhibition was determined as a reduction in bioluminescence. The inhibition of bioluminescence was generally increased with increasing contact time (up to 15 min). The bioluminescence generally decreased through the MBBR pilot plant (except for M4), indicating that toxic substances were removed during these MBBR treatment processes. However, the inhibition was increased between M3B to M4, which could be explained by overdosing of ethanol, used as supplementary carbon source to M4 to improve denitrification.

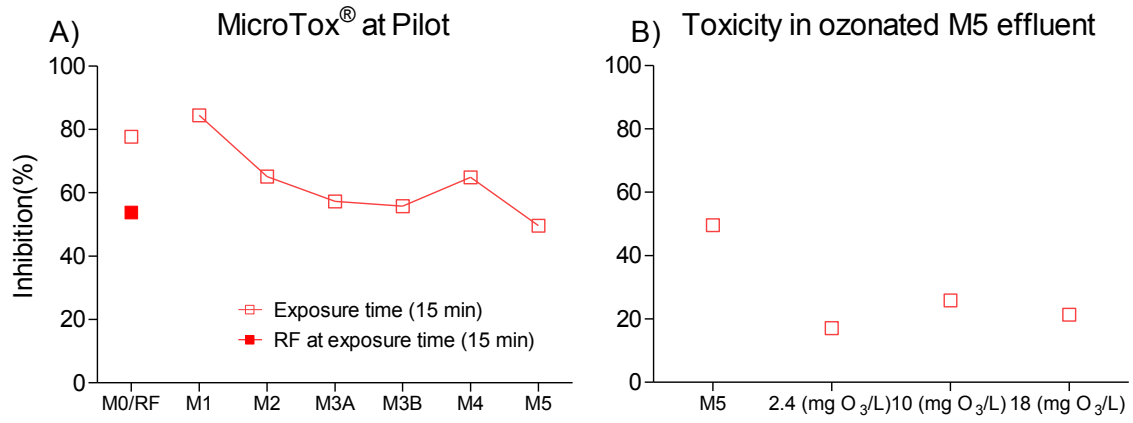


Figure 38. MicroTox tests based on bioluminescence with 15 min exposure time in A) pilot-scale MBBR treatment train. RF: return flow from M3B. B) toxicity in M5 effluent after ozone treatment with ozone dosages: 2.4, 10 and 18 mg O_3/L.

5.2 Treatment of municipal wastewater at Herning municipality

5.2.1 Daily operation of pilot-scale operation

The choice of technology for this pilot test was HYBAS™ for traditional advance wastewater treatment including biological nitrogen removal, but also for improved removal of pharmaceuticals present in municipal wastewater. The containers with the pilot plant received untreated wastewater from the main inlet to the municipal plant.

The overall performance of the HYBAS ™ pilot plant was evaluated based on conventional wastewater parameters. Average content of total COD in inlet samples was ~ 444 mg/l and average effluent content ~38mg/l, see Figure 39. Average content of soluble COD in inlet was ~137 mg/l and effluent concentration ~27 mg/l, see Figure 40. The TOC content in inlet samples was on average ~44 mg/l and effluent concentration ~11 mg/l (Figure 41).

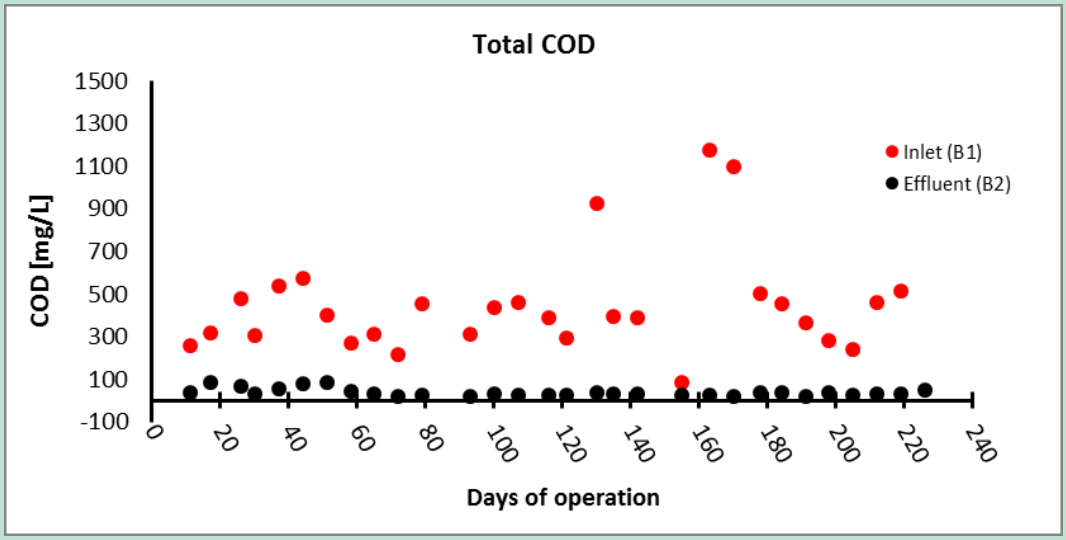


Figure 39. Overview of total COD content in inlet and effluent samples during HYBAS™ pilot plant operation.

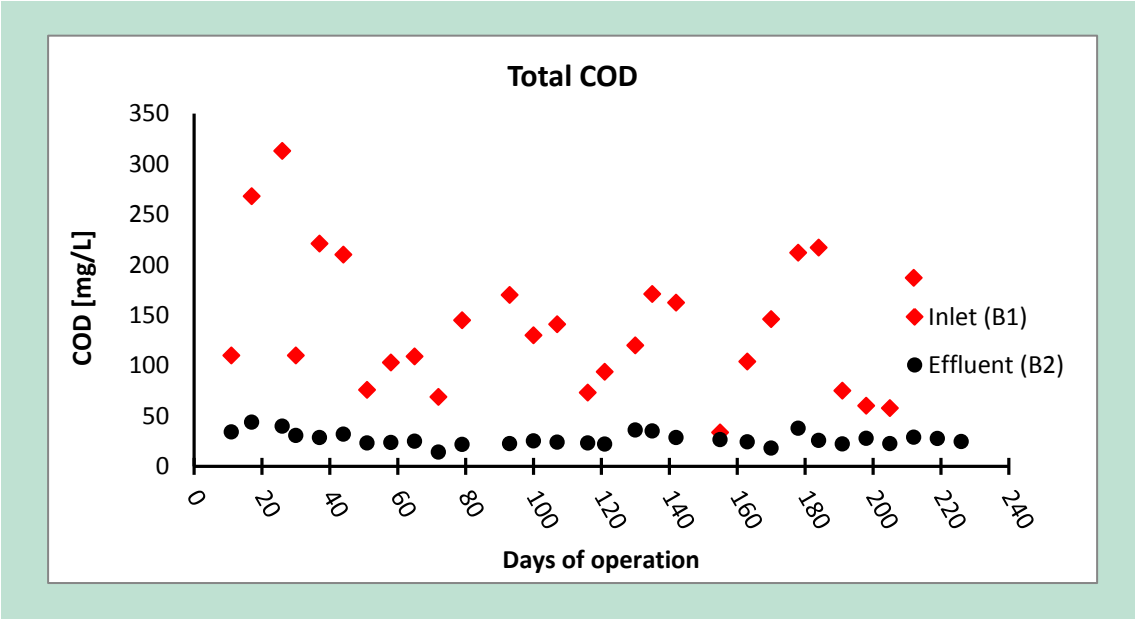


Figure 40. Overview of filtrated COD content in inlet and effluent samples during HY-BAS™ pilot plant operation.

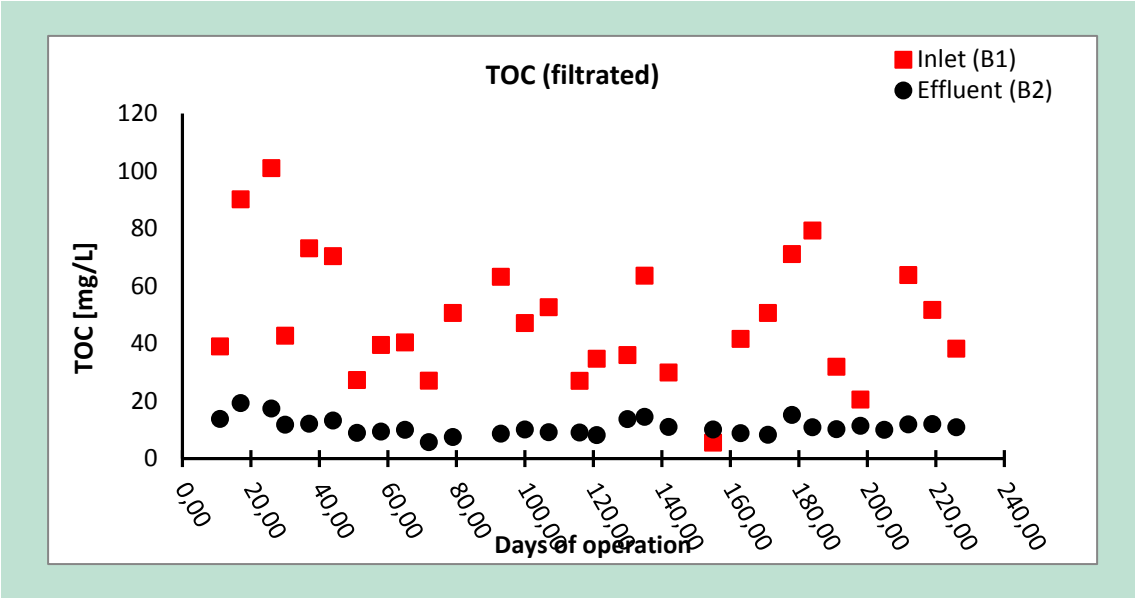


Figure 41. Overview of TOC content in inlet and effluent samples during HYBAS™ pilot plant operation.

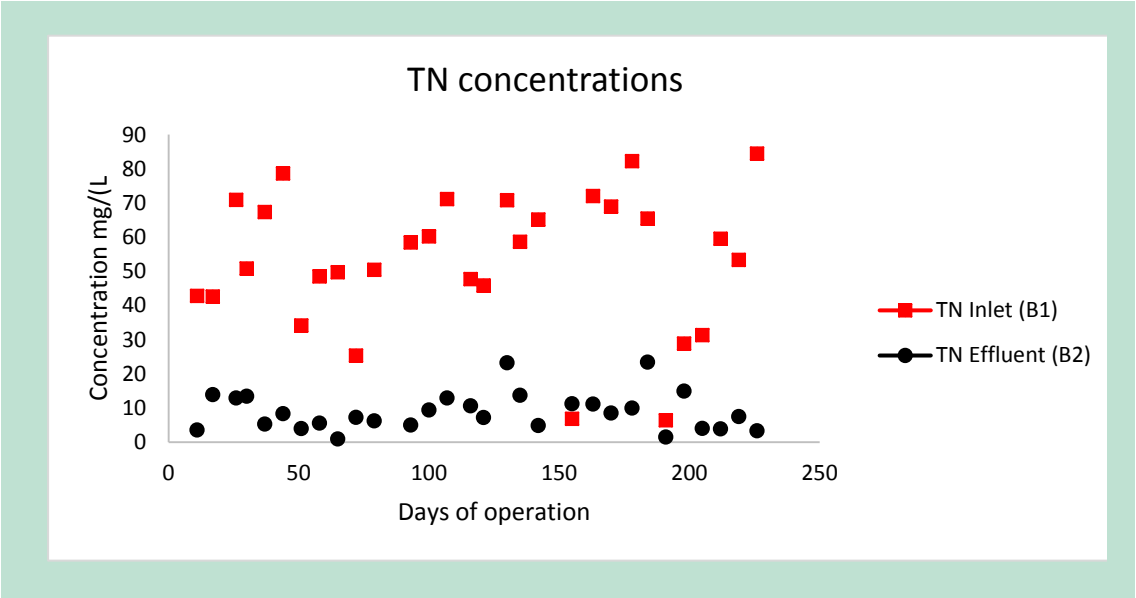


Figure 42. Overview of the TN concentration in inlet and effluent samples during the HYBAS™ pilot plant operation.

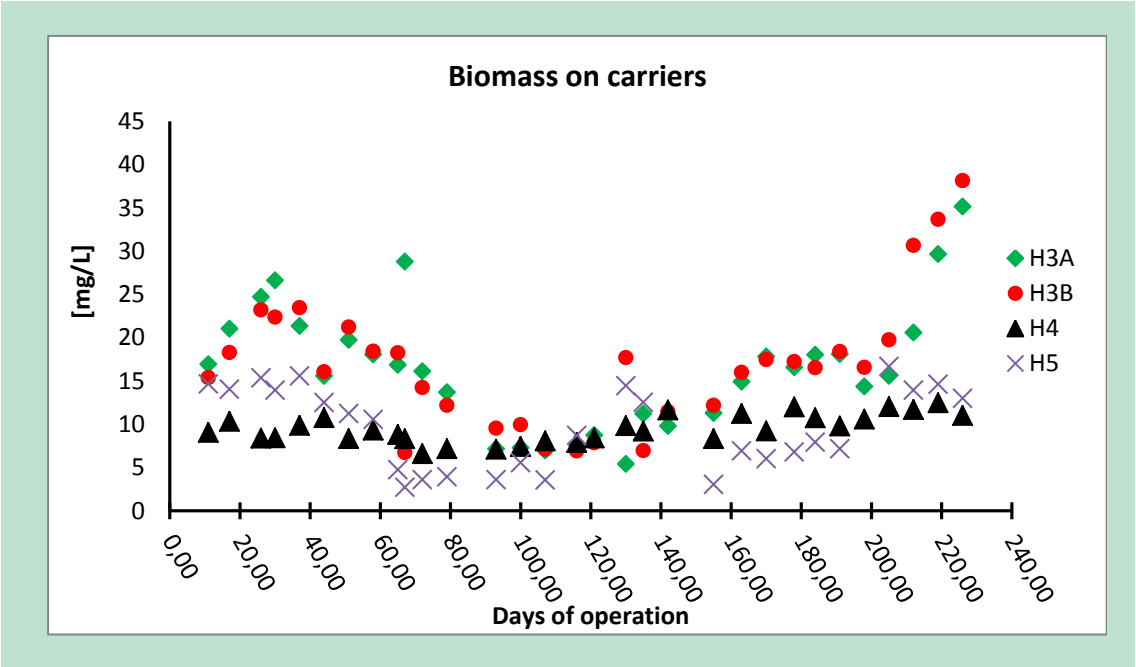


Figure 43. Development of biomass on carriers from the different reactors. M4 is an anaerobic reactor, the rest aerobic ones.

The level of TN in inlet samples was ~52 mg/l and effluent content ~8.9 mg/l (Figure 42). The effluent was slightly above the limit values for direct discharge (8 mg/l) though as earlier mentioned main part of nitrogen was nitrate.

The development of biomass on carriers is shown in Figure 43. The carriers used in the HYBAS™ pilot plant originated from the MBBR at AUH and the biofilm growth declined as the wastewater from Herning municipality was less concentrated compared to hospital wastewater. Activated sludge used in start of HYBAS™ was taken from the municipal plant. As can be seen from Figure 43 the biomass content on carries declined during the first couple months, probably because the activated sludge present in the first reactors of the pilot plant,

consumed some of the organic material present in the wastewater, leaving less to bacteria growing in biofilm. The average biomass content on carriers is lower in the final reactors of the HYBAS™ pilot plant (average H4: 9 mg/l, H5: 10 mg/l) compared to the MBBR pilot plant operated at the hospital (average M4: 19 mg/l and M5: 20 mg/l). It should be noted that reactors M4 and M5 are pure MBBR reactors, i.e. no activated sludge present.

5.2.2 Biological and chemical degradation of pharmaceuticals

Concentration curves for all investigated pharmaceuticals achieved from the batch experiments (data not shown), were plotted over time and data can be found in Tang et al., in prep. Figure 44 shows the biological degradation of the selected eight compounds in the different reactors. For a few compounds, higher concentrations were detected in the wastewater than the theoretically calculated concentration based on spiking dosage, showing the presence of pharmaceuticals as ciprofloxacin and diclofenac in the municipal wastewater.

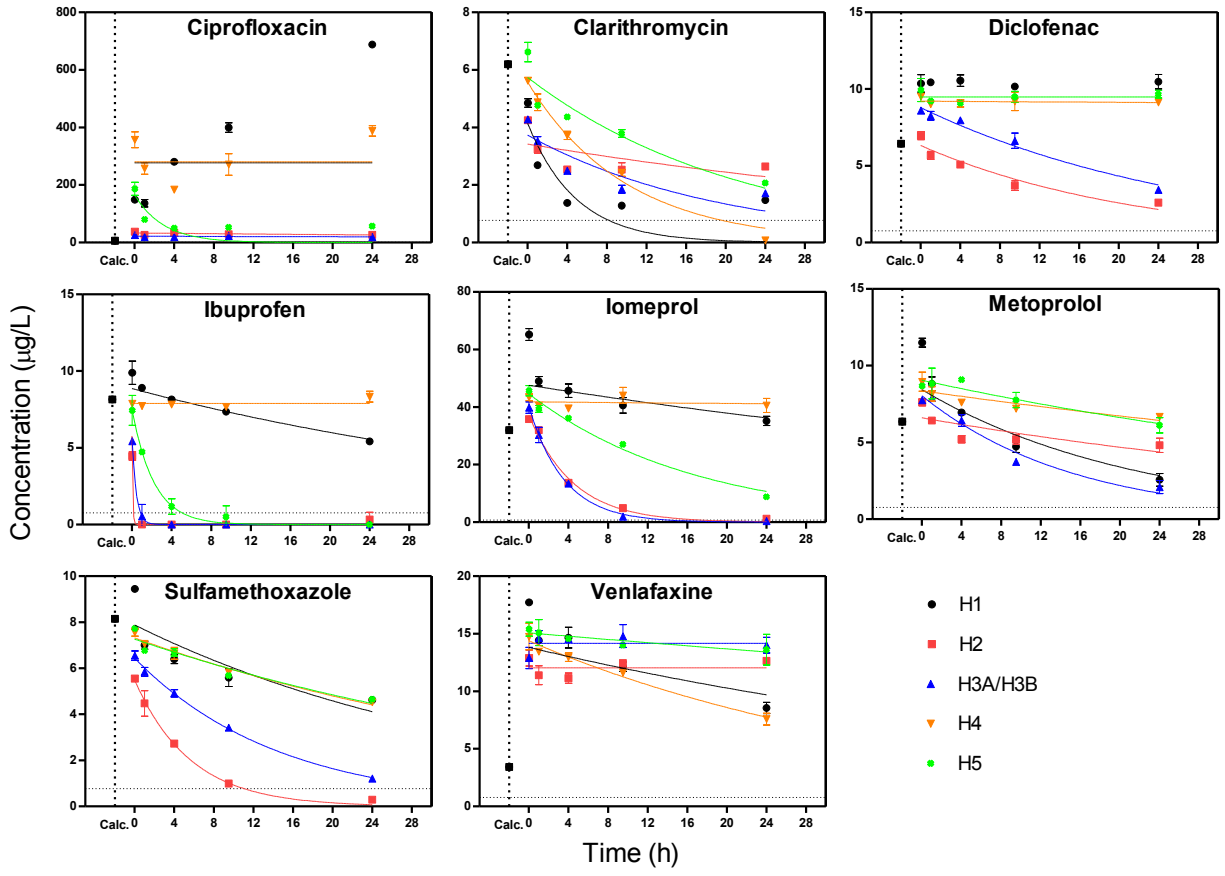


Figure 44. Concentrations of selected pharmaceuticals during batch experiment in each reactor (H1, H2, H3, H4 and H5). Filled lines correspond to a first-order kinetics fitting. Black squares are theoretically calculated dosages of each compound based on added stock solution. Dashed lines represent LOQ.

All compounds were biological degraded and in most cases, the highest degradation capacity was observed in the aerobic reactors except for venlafaxine and clarithromycin where degradation mainly took place under anoxic conditions.

The removal potential in percentage for the selected compounds for each reactor in the HYBAS™ pilot plant is shown in Figure 45.

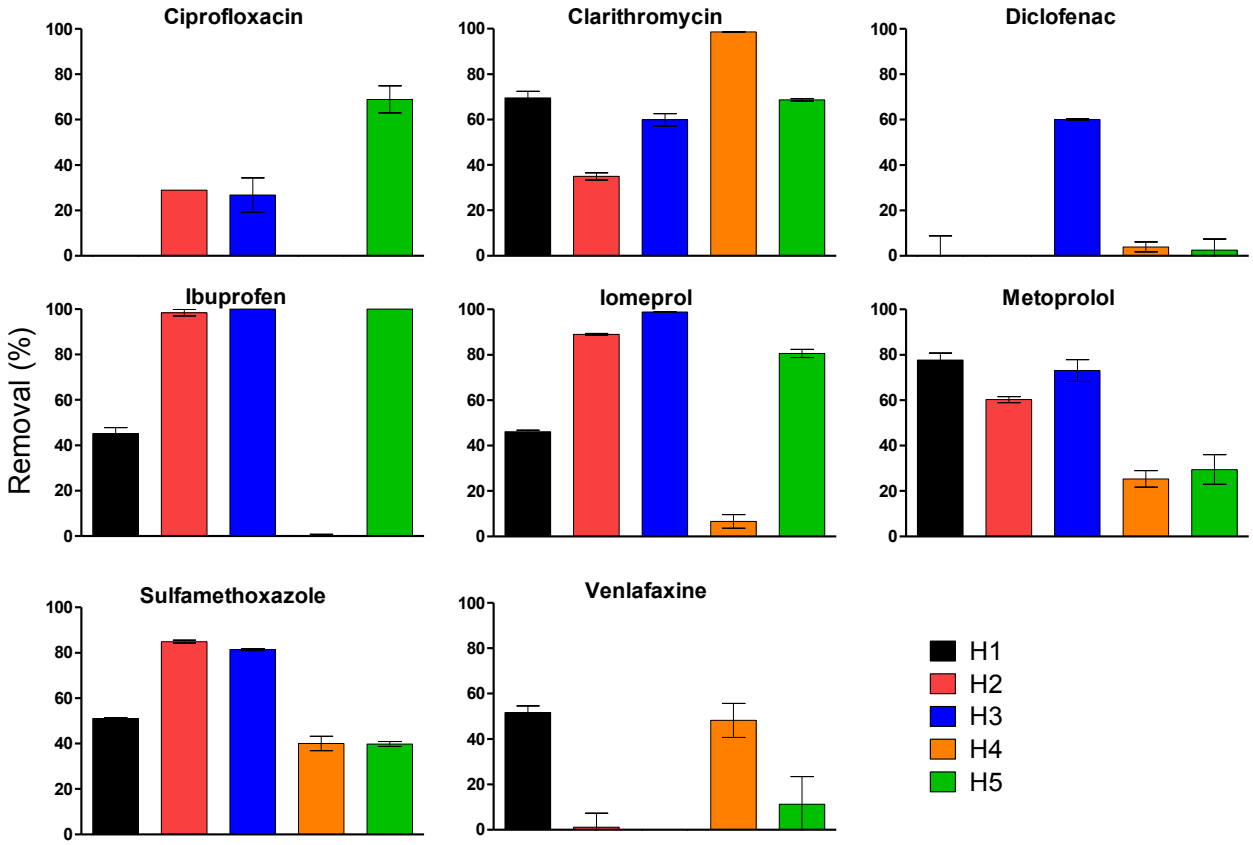


Figure 45. The potential capacity (%) of each reactor to remove pharmaceuticals over a period of 24 hours. Reactors H1 and H4 were denitrifying (black and orange) where the remaining reactors were aerobic. Carriers were present in H3, H4 and H5.

All compounds were removed to a great extent biologically (between 15% and 100%) in the different reactors. In general, the denitrifying (anoxic) reactors removed the selected compounds to a lesser extent than the aerated ones (oxic). However, very high removal capacity was observed in the anoxic reactors H1 and H4 for venlafaxine, metoprolol and clarithromycin.

The actual removal capacity of the individual reactors in the HYBAS™ pilot plant at a given day, was also investigated, see Figure 46. The concentration of the pharmaceuticals present were analyzed in inlet samples and also in the recycled water (recirculation for pre-denitrification in H1) and from clarifier after H3A/B to H1. For all the selected compounds, a reduction in concentration was observed throughout the HYBAS™ pilot-scale treatment plant. No deconjugation was observed.

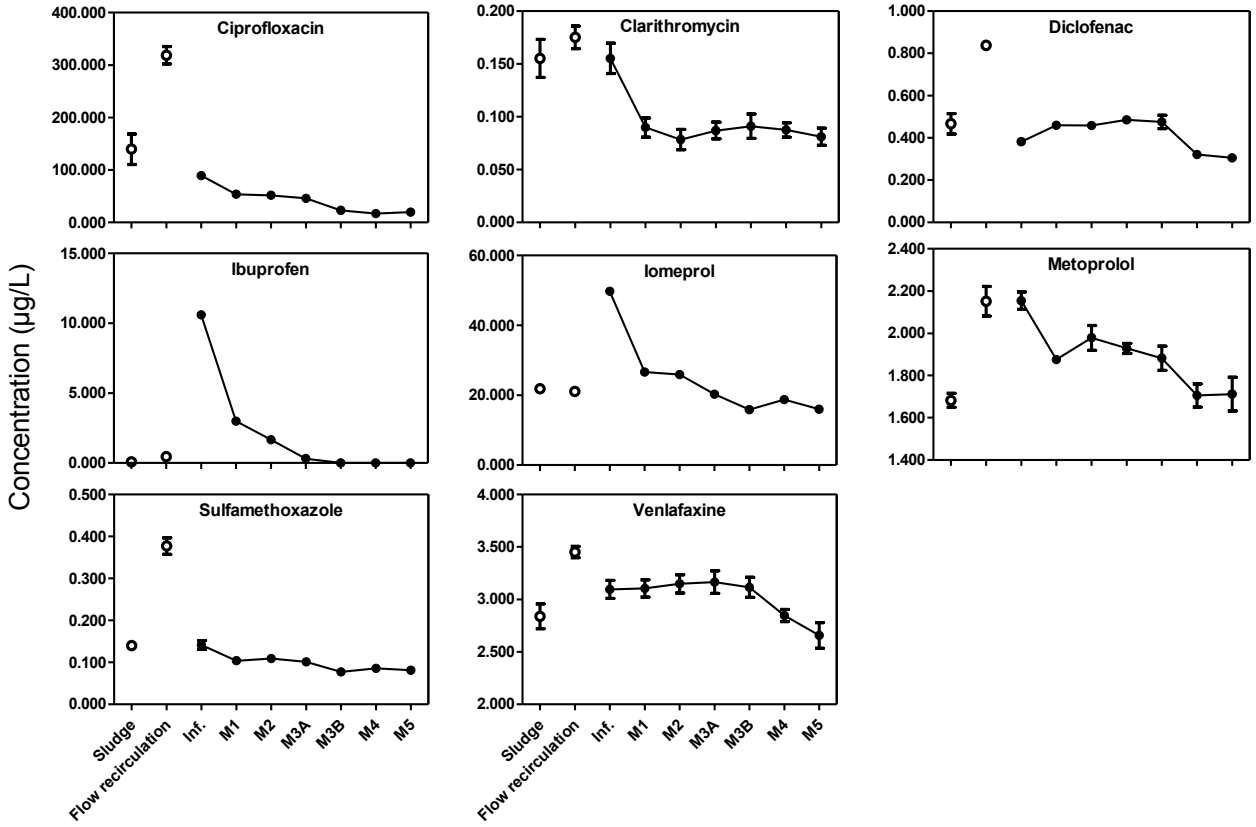


Figure 46. Average concentrations and SD (n=3, each sample was analyzed twice) in the HYBAS™ pilot reactors during the continuous flow experiment. The impact on pharmaceutical concentration was measures as recirculation concentration and sludge.

Calculating the actual biological removal of tested pharmaceuticals in percentage was performed. An example of the actual removal capacity of the entire biological treatment is depicted in Figure 47. Eight of the investigated compounds were degraded less than 20%, three compounds were identified as contrast media for which no guiding limit values has been proposed. For the remaining sixteen compounds, clear degradation capacities in the HYBAS™ pilot plant were observed.

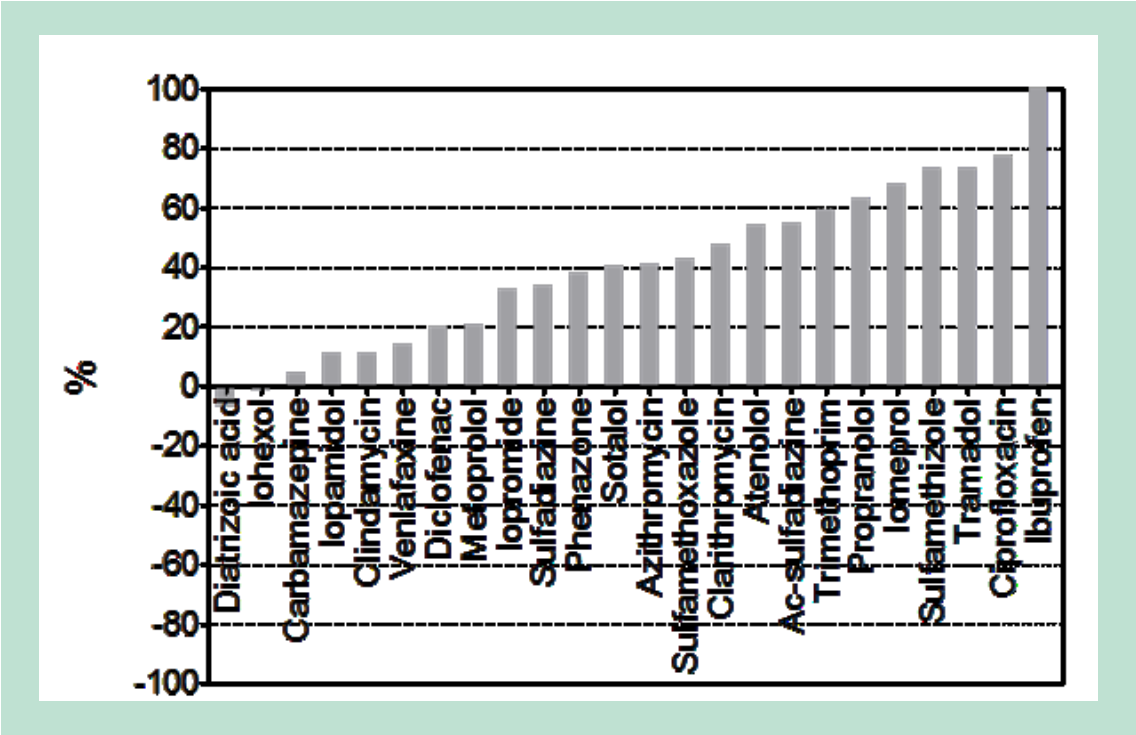


Figure 47. Measured removal from the continuous flow experiment over the pilot-scale HYBAS™ treatment plant, shown as percentage degradation.

The removal capacity of the HYBAS™ pilot plant, was also analyzed in accordance to the guiding limit values. Indigenous influent concentration (without spiking) and effluent concentration (after the treatment system) were analyzed. All selected compounds were present in concentrations above the guiding limit values in the inlet sample (see Figure 48). The biological treatment reduced the number of compounds above the guiding limit values and only the concentrations of ciprofloxacin, diclofenac and venlafaxine were still above the limit. None of the selected pharmaceuticals were observed after the ozonation treatment. No guiding limit values exist for metoprolol and iomeprol.

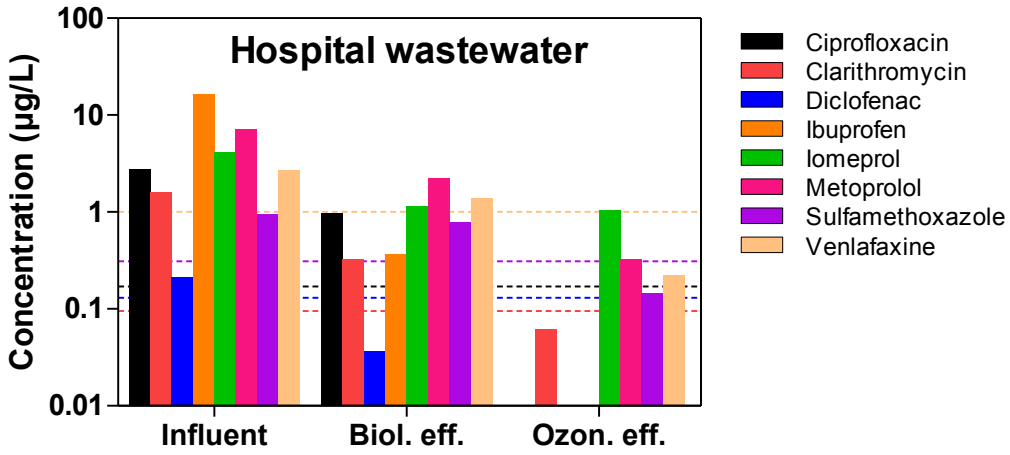


Figure 48. Concentration of pharmaceuticals present in the native influent and in the effluent after biological treatment and ozonation (the horizontal lines represent the Danish proposed guiding limit value for each compound except for metoprolol and iomeprol).

5.2.3 Antibiotic resistant bacteria

Samples from the MBBR pilot plant operating on hospital wastewater, and samples from the HYBAS™ pilot plant operating on municipal wastewater were collected. For comparison, inlet and effluent samples from Viby municipal WWTP (No hospital wastewater), Egaa (municipal plant receiving hospital wastewater) as well as effluent samples from Herning municipal wastewater treatment plant were investigated. The total number of bacteria present was determined in inlet and effluent samples using Compact Dry. In addition, antibiotic resistant bacteria *E. coli* was analyzed for ciprofloxacin, cefuroxime, gentamicin, and sulfamethoxazole resistance applying a newly developed ComPact dry method. Literature values for added antibiotics could not be applied, and therefore, an investigation of the required antibiotic concentration needed to differentiate between resistant and non-resistant *E. coli* was carried out. Investigated antibiotic resistant *E. coli* strains were provided from the department of clinical microbiology at the Aarhus University Hospital, Aalborg.

The number of antibiotic resistant *E. coli* were investigated in several samples originating from the MBBR pilot plant treating hospital wastewater. Inlet samples, and effluent samples were quantified in terms of antibiotic resistant *E. coli* (Figure 49) and compared to the receiving WWTP Egaa.

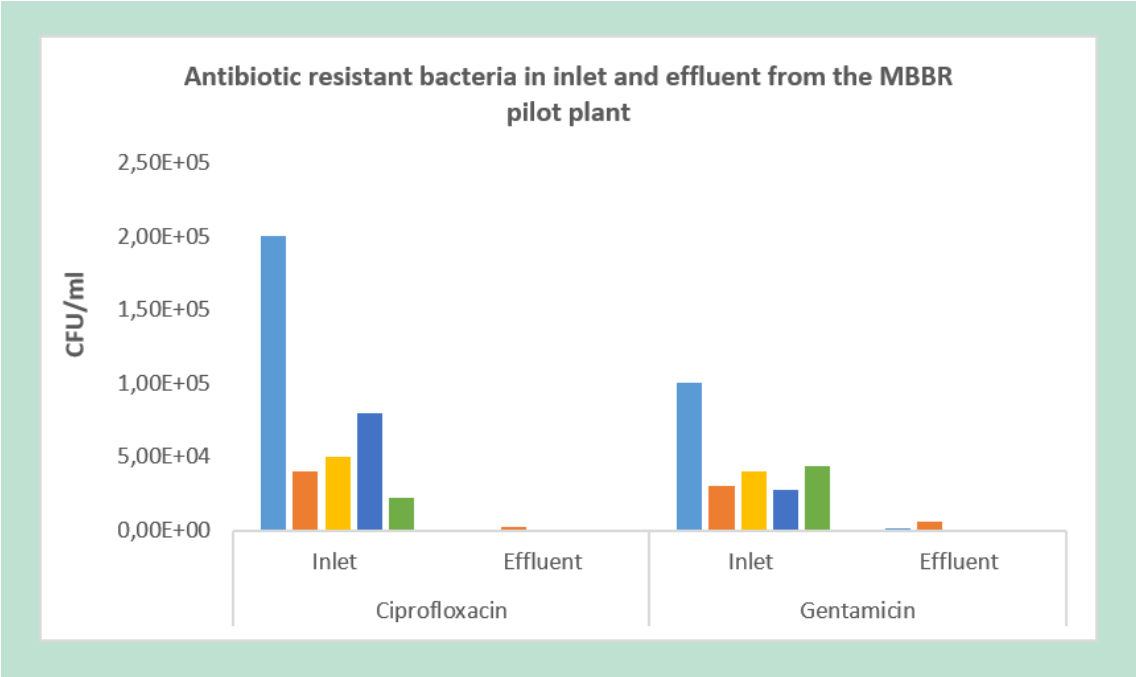


Figure 49. Overview of antibiotic resistant bacteria detected in inlet and effluent samples from the MBBR pilot plant.

The number of antibiotic resistant bacteria in the inlet were by biological treatment in the MBBR pilot plant reduced to low numbers in the effluent samples, as investigated for two common types of antibiotics, namely ciprofloxacin and gentamicin.

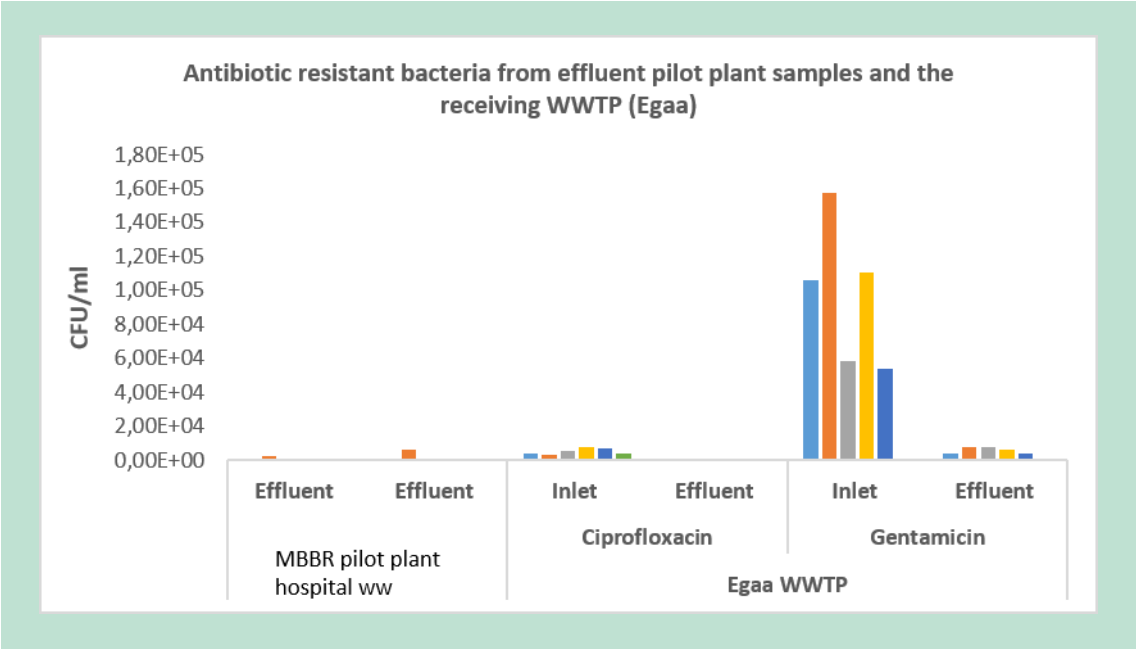


Figure 50. Comparison of antibiotic resistant bacteria in effluent from MBBR pilot plant and presence of antibiotic bacteria in inlet and effluent samples from the WWTP (Egaa) receiving wastewater from the hospital AUH.

As evident, the number of antibiotic resistant bacteria present in inlet samples of Egaa could not solely be ascribed to hospital wastewater influence but also considerable contribution from private households (Figure 50).

The number of bacteria were investigated in the raw wastewater from the hospital AUH As evident, a majority of the bacteria detected in the raw wastewater from hospital were resistant to one or more antibiotics, see Figure 51.

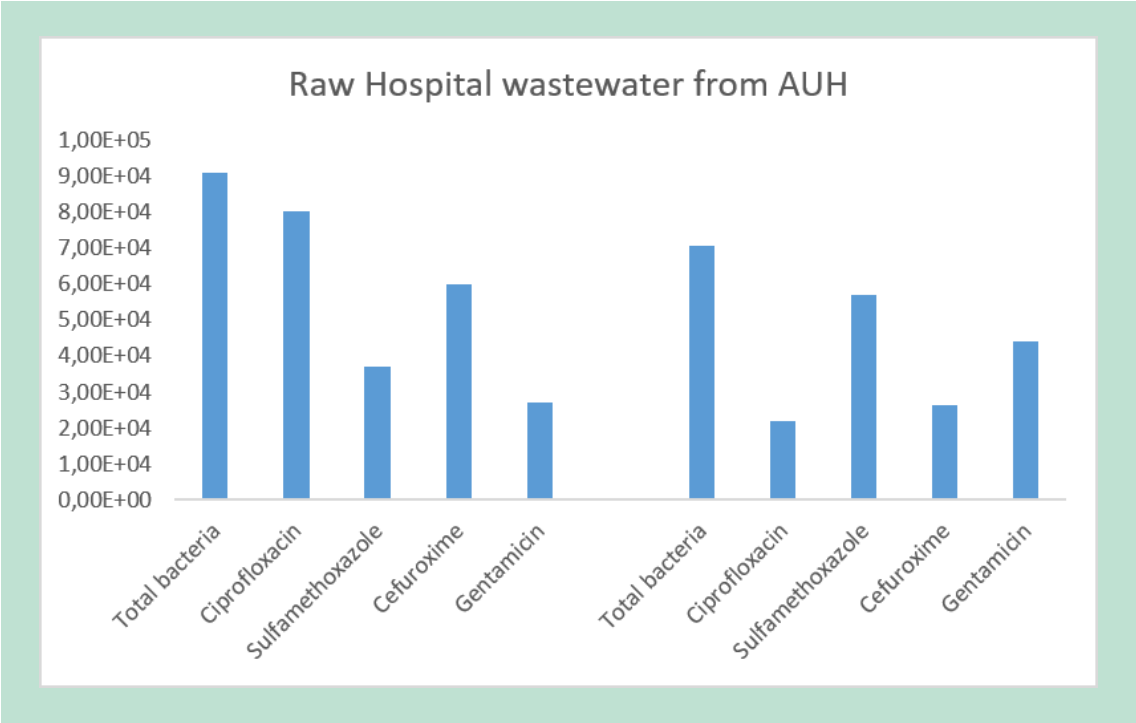


Figure 51. Overview of antibiotic resistant bacteria detected in raw wastewater from hospital at two sample dates.

Inlet and effluent samples from Egaa WWTP were also investigated. The main reason is that all hospital wastewater from AUH is discharge to public sewer connected to the WWTP at Egaa municipality. Inlet and effluent samples from Viby WWTP were also included for comparison, as no hospital wastewater is discharged to the WWTP in this municipality. There is a evident difference in the presence of bacteria detected in inlet samples at Viby WWTP and at Egå WWTP, see Figure 52. Also in terms of antibiotic resistant, *E. coli* differs between these two WWTPs.

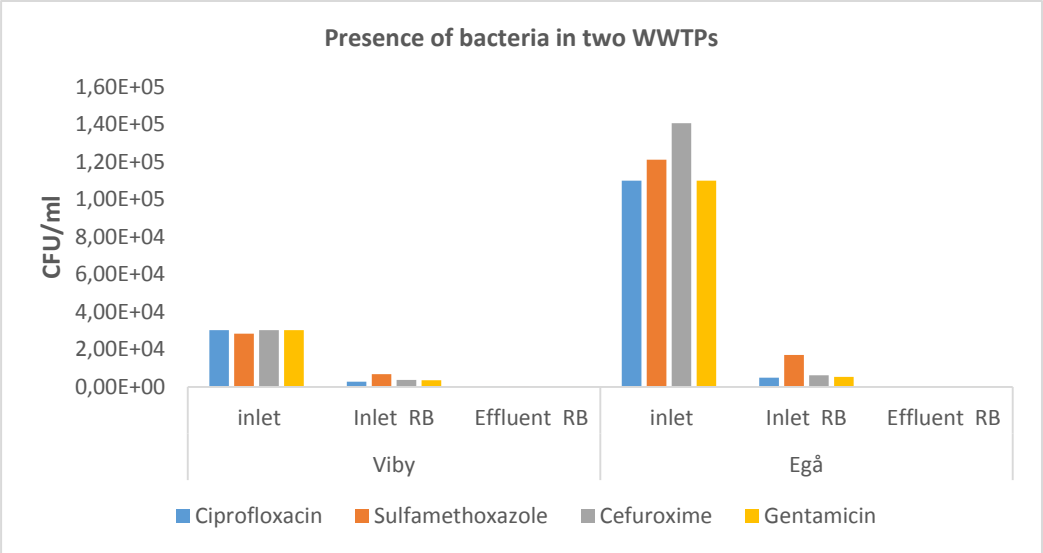


Figure 52. Overview of antibiotic resistant bacteria detected in inlet and effluent samples from Viby (no hospital) and Egaa (with hospital) municipalities. RB denotes resistant bacteria.

Based on these data, a ratio between the two municipal WWTPs were calculated, see Table 8 Egaa wastewater exceeds by far Viby wastewater both in terms of total bacteria in inlet, and inlet resistant bacteria. However, the Egaa treatment plant is much more efficient at reducing the presence of antibiotic resistant *E. coli*, i.e. fewer resistant bacteria are present in effluent samples from Egå compared to Viby effluent. This is also the case when investigating the actual numbers of resistant bacteria in effluent samples (data not shown).

Table 8. Overview of ratio between bacteria present in Egaa WWTP (with hospital) and without hospital (Viby) WWTP.

	Egaa/Viby bacteria ratio		
Antibiotic	Inlet samples	Inlet resistant bacteria	Effluent resistant bacteria
Ciprofloxacin	3.6	1.8	0.1
Sulfamethoxazole	4.3	2.5	0.3
Cefuroxime	4.6	1.7	0.5
Gentamicine	3.6	1.5	0.4

5.2.4 FrogBox®

A new method for real-time monitoring monitoring effect of micropollutants/ Endocrine disruptive compounds (EDC) present in wastewater was set up in the Hybas™ pilot plant at Herning municipality. The technology is termed WatchFrog and is based on changes in genetic expression of the green fluorescent protein (GFP), present in genetically modified larval amphibians responding to EDCs (Fini et al. 2009). The fluorescence of the larvae increases or decreases due to presence of EDC compounds influencing the activity of the thyroid axis. The changes in GFP expression is quantitative; e.g. the more endocrine effect is present in the water, the greater the change in fluorescence (Fini 2007).

Briefly, 50 larvae were exposed to a continuous flow (approx. 100ml/min) of the inlet or outlet samples from the Hybas™ pilot plant. Wastewater was filtered (100 microns) and heated at 21

°C prior to entering the FrogBox®. Six times a day, the fluorescence of larvae was quantified using a flow cell passing a camera and an LED illuminator. Specific wavelength filters allowed the expression of eGFP to be quantified in each larva. At each time point, the 50 larvae passed six times in front of the camera and an image analysis algorithm selected images representing in-focus well orientated full larvae, typically a minimum of 100 good larvae images is selected at this step. The mean of the larvae fluorescence was calculated for each time point. Every Tuesday, a new cartridge containing 50 larvae was replaced in the FrogBox®. The old cartridge was returned to Watchfrog for proper handling of the genetically modified tadpoles. Inlet and outlet samples from the HYBAS™ pilot plant were assessed for potential thyroid disrupting activities. The percentage of measurement points with fluorescence induction over 12% was determined for all samples. Points below the 12% threshold were considered not to have any thyroid disrupting activities. Watchfrog determined this threshold during the validation of the model using reference active or inactive compounds.

The concentration of triiodothyronine (thyroid hormone, T3) used for the positive control in the thyroid assay was 3.25μg/l corresponding to concentration of T3 presents in the plasma of tadpoles during metamorphosis. Higher concentrations above this lead to adverse physiological effects. To recalculate fluorescence intensities measured, a conversion factor between fluorescence and hormonal equivalent was applied (EQH =10^((Value of normalized data point-0,977)/0,99). This conversion factor has been previously determined by a calibration curve obtained with several dosages of T3 on this FrogBox®.

The following results were obtained during the period when the FrogBox was installed at the HYBAS™ pilot plant at Herning municipality. Different setups at the pilot plant were tested to assess the effect of biological treatment and the effect of different ozonation concentrations (see Figure 54). Data from the pilot plant were compared to the full scale Herning WWTP effluent. An overview of fluorescence in effluent wastewater from the full scale Herning WWTP at different time point is shown (Figure 53).

Correlation between hormonal equivalents is depicted on the left y-axis of the graphs. Fluorescence intensities are grouped within limits as lower limit represents point of detection and upper limit, where induction of potential effects can be observed is shown on the y-axis to the right. The level of potential thyroidal effects corresponds to 3.25 µg/l.

As evidenced by Figure 53, most of the wastewater samples investigated during the one week trial, showed no increase in fluorescence (values were between upper and lower limits for detection), and no thyroidal disruptive concentrations were detected, either (threshold for thyroidal disruptive concentration 3.25 µg/l).

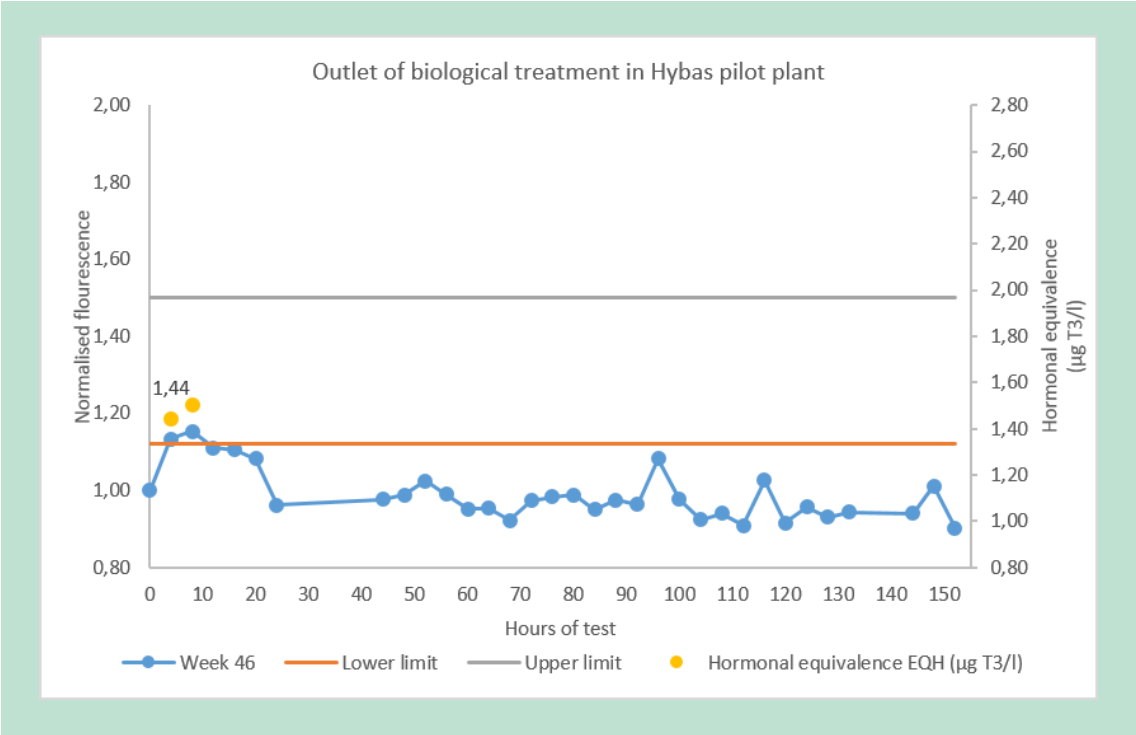


Figure 53. Overview of fluorescence in effluent wastewater from the Hybas pilot plant (biological treatment) at different time point. Correlation between hormonal equivalents is depicted on the left y-axis. Lower limit represents point of detection and upper limit, fluorescence intensities where induction of potential thyroidal effects can be observed.

Effect of different ozone doses were also investigated see Figure 54. Increased dose of ozone did not result in increased toxicity measured as fluorescence neither were problematic thyroidal concentrations detected.

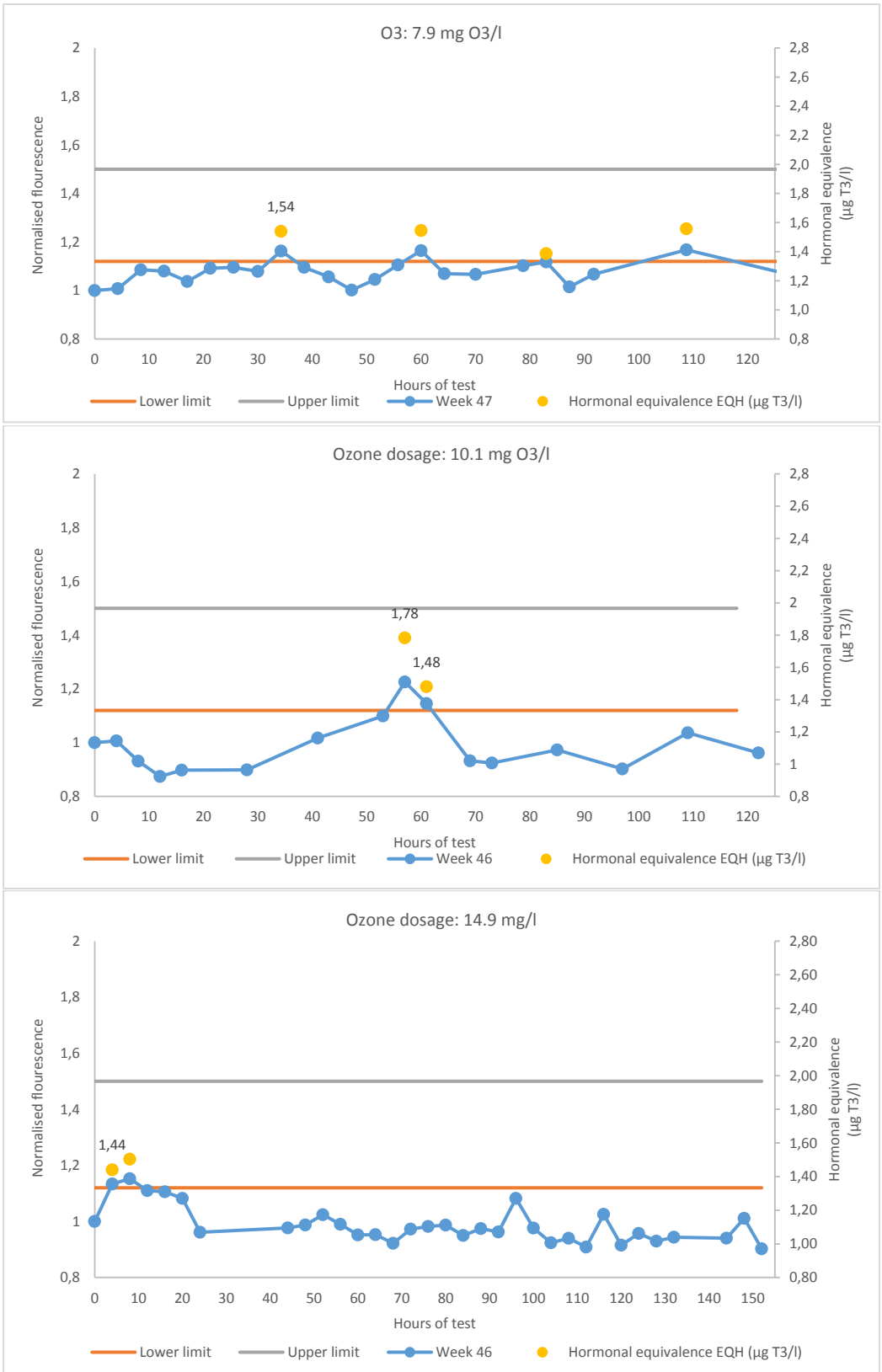


Figure 54. Effect of different ozone doses (7.9 mg/l 10.1 mg/l and 14.4 mg/l) on the thyroid disrupting effect measured as fluorescence and hormonal equivalence. Lower limit represents point of detection, and upper limit the fluorescence intensities where induction of potential thyroidal effects can be observed

Comparison of FrogBox® data obtained in the Hybas™ pilot plant (with and without ozonation), with data for effluent wastewater from the full scale municipal WWTP at Herning was performed, see Overview of fluorescence in effluent wastewater from the full scale Herning WWTP at different time point. Correlation between hormonal equivalents is depicted on the left y-axis. Lower limit represents point of detection and upper limit, fluorescence intensities where induction of potential thyroidal effects can be observed. No difference in neither fluorescence nor thyroidal activity were observed when treatment in pilot scale coupled to ozonation was compared to conventional wastewater treatment, see Figure 55.

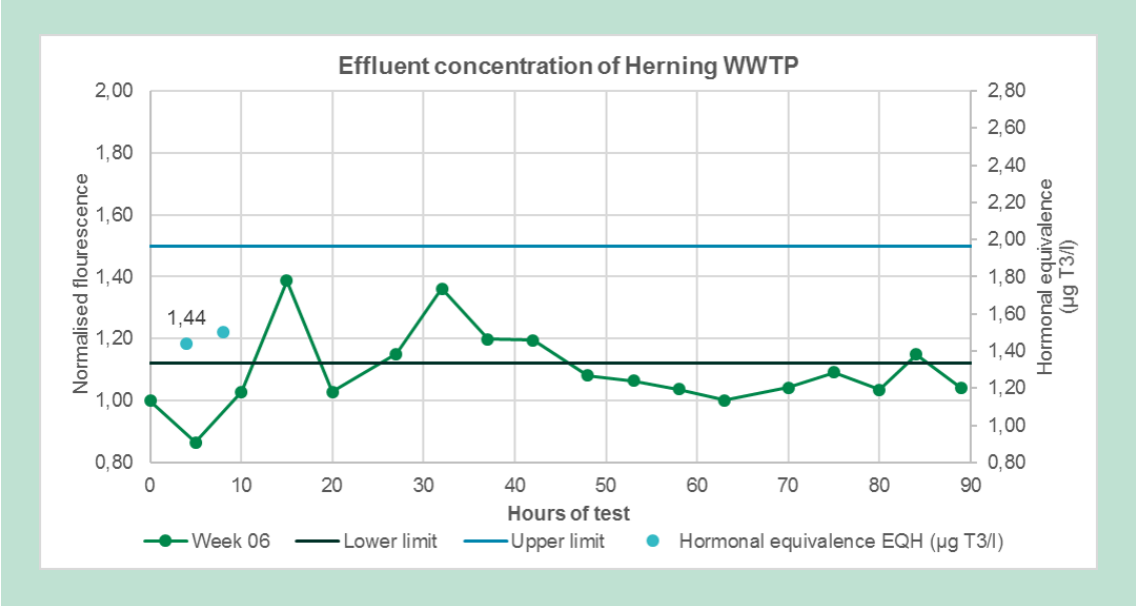


Figure 55. Overview of fluorescence in effluent wastewater from the full scale Herning WWTP at different time point. Correlation between hormonal equivalents is depicted on the left y-axis. Lower limit represents point of detection and upper limit, fluorescence intensities where induction of potential thyroidal effects can be observed.

6. Design of full scale treatment for hospital wastewater

6.1 Design of full scale treatment of hospital wastewater

The main target for the pilot tests was to obtain efficient biological removal of pharmaceuticals. Two different process concepts, MBBR and HYBAS, were tested at AUH, NBG (treating wastewater from Oncology Department) and only MBBR was tested for the combined effluent from Aarhus University Hospital AUH, Skejby. The pilot plants were designed to obtain an effluent quality equivalent to the requirement for direct discharge from municipal wastewater plants in Denmark. Biological nitrogen removal was fully implemented in the treatment schemes; however, no specific biological or chemical phosphorous removal step was included in the set up. The lack of operational data for phosphorous removal is not considered of concern as chemical precipitation of phosphorous in municipal wastewater is well known and related cost can easily be estimated.

In addition to the biological treatment, also continuous ozonation tests were performed on biologically treated effluent at the AUH pilot plant, to quantify the dose required to reach treatment targets. Following ozonation, the water passed a polishing bio-filtration step to remove easy biodegradable ozonation by-products.

The two pilot plants used had different capacities. The bench scale unit used at Kommmunehospitalet had capacity of 1 l/h and the pilot unit used at Skejby had a capacity of 300 l/h. Both the bench scale and the pilot plant were operated continuously for more than 9 months, so the developed biomass could be considered as representative of the systems and reliable for design.

The results from these pilot tests are described in Chapter 4 and 5.

6.1.1 Description of treatment concept for hospital wastewater.

The treatment results obtained in the pilot tests with hospital wastewater do not provide convincing proof for the HYBAS process (combination of activated sludge and biofilm) offering notable performance advantages to the pure biofilm process MBBR. Furthermore, when considering the complexity of operating an activated sludge type process in low capacity installations, it was concluded that the MBBR process was more suitable for the potential sizes of installations serving only hospitals and MBBR was therefore selected for the full scale hospital wastewater treatment concept.

The load from the hospital to the wastewater treatment plant is calculated as the example shown in Table 9.

Table 9. Calculation of load from hospital used for plant design.

Ambulant patients		**Number/year**	**325199**
Visit average duration hours			3
Ambulant patients treatment		days/week	5
When		day h	8
Load per patient corresponding to full time		Full time patient	0,125
		During day time	0,375
Net load Persons		average/day	156
		peak hours/day	469
Beds somatic patients			621
Number of hospitalization		per year	53421
Average days		per år	182000
Load factor (occupancy rate)		%	80,29%
Net load PE		average/day	499
Beds psychiatric patients			82
Number of hospitalization			53421
Occupancy rate		%	95,00%
Net load PE		average/day	78
Staff		number	3600
		Average number/shift	1200
		During day shift	1800
Total PE load	**Calculated**	**average**	**1933**
		Max	**2972**
	DESIGN	**Design average**	**2000**
		Design Max	**3000**

For design purposes, the average person load (PE) to the treatment plant is multiplied by the number of persons connected to the WWTP. The normalized numbers for PE is recalculated with the average effluent concentrations for Skejby Hospital, in order to estimate average and peak loads to be expected. Table 10 shows the design basis used for estimate of both CAPEX and OPEX for the hospital wastewater treatment plant.

Table 10. Design basis for wastewater treatment plant for hospital wastewater.

Based on analysis at Skejby hospital			Based on standard PE		Selected design basis	
Average concentrations Skejby hospital			g/Person equivalents (PE)	Load based on standard 3000 PE	Average load kg/d	Peak load kg/d
SS	mg/l	400	72	216	160	216
BOD5	mg/l	350	60	180	140	189
COD	mg/l	770	130	390	308	416
CODsol	mg/l	370	58	174	148	200
Ntot	mg/l	70	12	36	28	38
NH3-N	mg/l	60		0	24	32
Ptot	mg/l	14	2	6	6	8
Flow	m3/d				400	540
	m3/year				150	200
	m3/h average				17	23
	m3/h max hour					35
	m3/h peak					61

It should be noted that the calculated load is based on activities related to patients and staff. Any water discharged from utilities like blow down from cooling towers, chillers, boiler, water treatment and wastewater from laboratories etc. has not been included in the estimate. It is however assumed that the volumes are fairly limited and that any wastewater from these activities containing harmful substances is handled separately and not discharged into the sewer. Furthermore, any runoffs from roofs and consolidated surfaces are not supposed to be mixed with the sewage, i.e. not included in estimates.

Due to the proximity of wastewater treatment plant and wastewater source, large variations in flow and to some extent also composition can be expected. It is not feasible to design for extreme peak flows as the duration of peaks can be anticipated to be short. Certain hydraulic equalization capacity however needs to be incorporated in treatment concept.

In Figure 56 the process scheme for the hospital wastewater treatment plant, developed and designed based on the project results is shown. Below is a short description of the treatment steps.

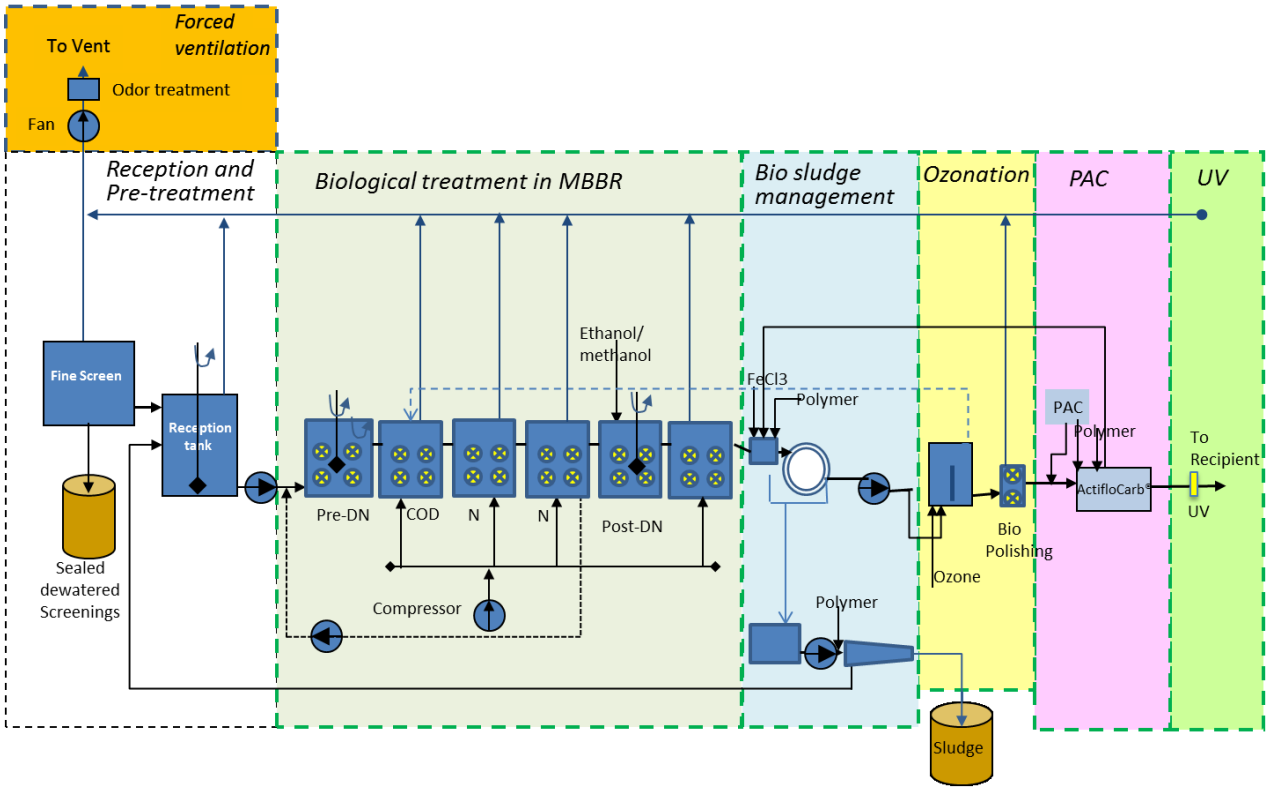


Figure 56. Process flow diagram for full scale MBBR wastewater treatment plant for hospital.

Wastewater is piped to the inlet structure of the treatment plant where it passes a fine screen connected to a screenings washer and compaction unit. Screenings are discharged from the encapsulated unit into an enclosed container.

The screened water is either pumped or gravitated to the reception tank. The reception tank can either be a separate tank or an integrated part of the first process tank. The reception tank is designed to handle sudden peak flows or few hours per day with above average inlet flow, in order to maintain a fairly stable load to the biological treatment.

The biological treatment is designed for nitrogen removal, i.e. nitrification (ammonia oxidation) and denitrification (nitrate removal). The main part of nitrate produced by oxidation of ammonia is removed in the pre-denitrification and in case there is too high nitrate level out from main process tanks, residual nitrate is removed in the post-denitrification step.

The biological treated effluent passes drum filters with 20 to 10 μm filter cloths. For precipitation of phosphorous and coagulation of fine material ferric chloride and polymer are added to filter feed. Suspended solids in filtered effluent is approx. ≤ 2 mg/l and P ≤ 0.5 mg/l. Collected solids are diverted to a sludge holding tank, from where it is pumped to a centrifuge for dewatering.

Filtered effluent is pumped to the tank for ozonation. The ozonation reactor is designed to prevent short circuiting of inlet to outlet. Ozone is produced based on pure oxygen. In general, 15 – 20% of oxygen is converted to ozone in the ozone generator. Off gas from the ozonation tank, which is depleted of ozone, is diverted to the first oxic MBBR tank, where residual oxygen is used by the bacteria performing aerobic degradation of organics. By this arrangement the residual oxygen is valorized and furthermore the off gas from ozonation is bio scrubbed.

After ozonation, water flows into a bio-polishing step, which is a small MBBR reactor, also called BioProtector®. In the bio-polisher the ozonation degradation products, which potentially could be toxic, are biodegraded

Treatment with activated carbon (PAC) is incorporated in the treatment concept as a final polishing step in case of final effluent discharge criteria call for further removal of pharmaceuticals. PAC is added to the water to absorb any residual pharmaceuticals and furthermore this process step removes any residual suspended solids. PAC is dosed to the inlet to Actiflo®Carb and gets in contact with the water in the PAC pre-contact tank before entering the ballasted flocculation system Actiflo® where a coagulant and polymer is added in order to trap the solids including PAC on micro sand. The particles will afterwards settle in the lamella sedimentation compartment. Settled sludge is recirculated and passes a hydro cyclone where the micro sand is separated and returned to process. The main part of the separated sludge, which also contains PAC, is returned to the PAC pre-contact tank and an amount corresponding to sludge production is returned to the coagulation tank upstream of the drum filter. Here it will be removed together with biological sludge. By returning the extracted loaded PAC to upstream of bio sludge separation any available absorption capacity on the PAC will be used.

As a final (and second) disinfection barrier, the water passes a UV system before discharge.

The plant is fully automated with a SCADA system controlling and recording performance. Operator intervention is after the plant has been commissioned not expected. However, normal surveillance is required for checking equipment, cleaning of on-line probes, checking supply of chemicals, maintenance, sampling, general cleaning etc.

6.1.2 CAPEX and OPEX estimate for wastewater treatment plant for hospital wastewater

As a representative example, a green field wastewater treatment plant at a hospital in Denmark with plant capacity 150,000 m^3 wastewater/year has been chosen. Design basis / loads are presented in Table 10 (chapter 6.1.1.) and estimated investment cost can be found in Table 11. Specific site conditions or specific demands to design might lead to additional investment cost. The numbers refer to a standard quality plant constructed at a site with soil conditions not requiring special foundation and cost for equipment and civil works based on Danish cost level.

Table 11. Capital expenditure for wastewater treatment plant for hospital capacity 150,000 m3/year.

	Million DKK
Total Equipment, Engineering, Erection, Commissioning etc.	18 - 20
Total Civil Works	3.5 - 6
Total Plant	**21 - 26**

Operation cost has been estimated for the same plant, see Table 12. The estimate covering expenses for laboratory analyses is uncertain and most likely overestimated, as the number of laboratories, carrying out the analyses will increase in the future and increased competition will reduce price. In addition, required sample frequency and number of compounds are not yet known. Cost of capital has not been included in OPEX estimate as depreciation period and interest rate on capital investment varies from entity to entity.

The maintenance cost is calculated based on 5 % /year for mechanical and electrical equipment and 2 % /year for civil works. The distribution of consumable costs is depicted in Figure 57.

Table 12. Operational expenditures for wastewater treatment plant for hospital capacity 150,000 m^3/year.

	DKK/Year	DKK/m^3 Wastewater
Total Consumables*	405,000	2.70 - 3.25
Analysis	100,000	0.67
Maintenance plant	575,000	4.35
Operation	150,000	1.00
Total OPEX	**1,230,000**	**8.20 - 8.70**

* Electricity (0.75 DKK/kWh), Chemicals, Ozone, Screenings and Sludge

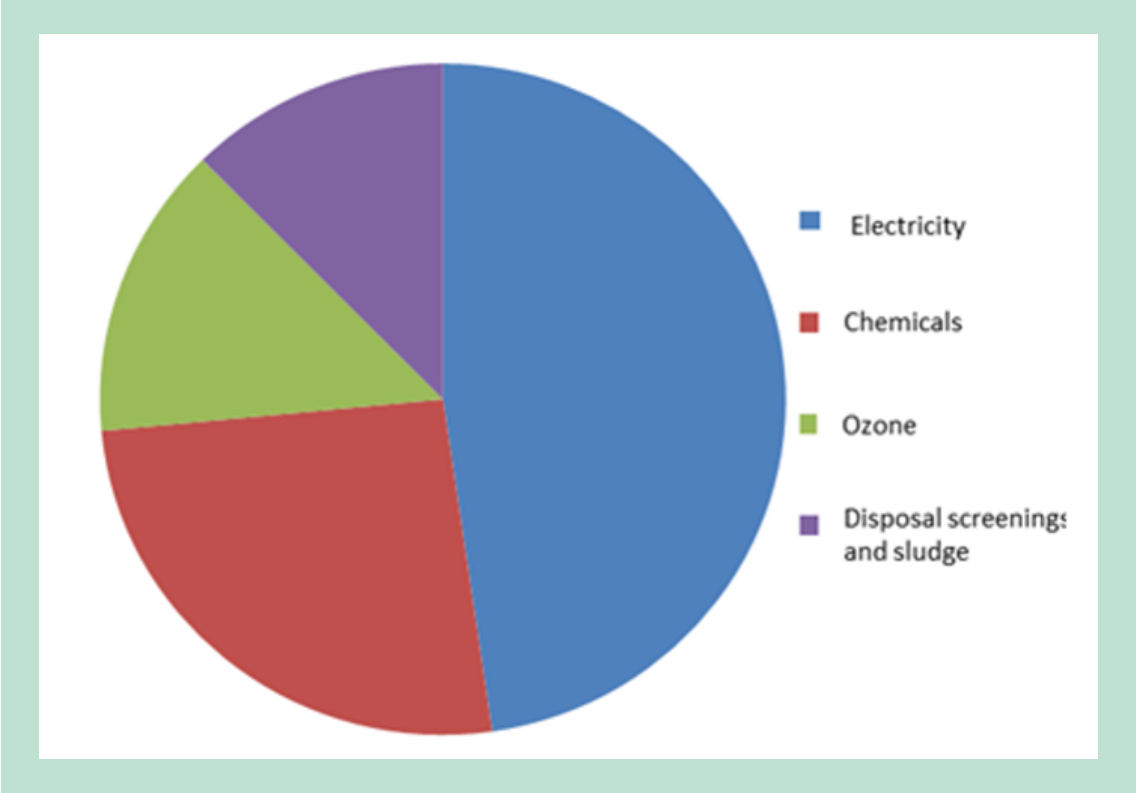


Figure 57. Distribution of cost of consumables.

7. Benchmarking of investigated solutions for removal of pharmaceuticals

7.1 Benchmarking of treatment solution for hospitals and alternative solutions for the municipality

One of the project goals was benchmarking of alternative locations for removal of pharmaceuticals in wastewater. Figure 58. shows an illustration of the locations which were included in the investigation.

Pilot plants in different sizes were operated with the following types of wastewater:

- Wastewater from a hospital department with highly toxic compounds were used (Dept. of Oncology).
- Combined effluent from a hospital.
- Normal municipal wastewater.
- Effluent from a typical municipal wastewater treatment plant with nitrogen and phosphorous removal.

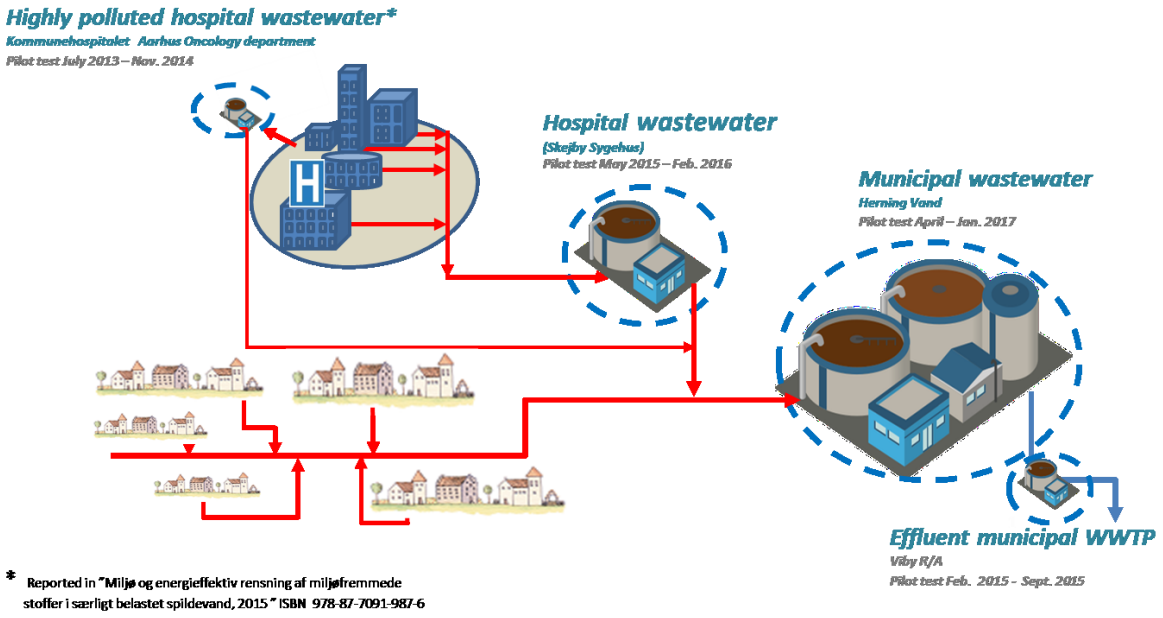


Figure 58. Test locations for removal of pharmaceuticals in wastewater.

The reason for wanting to bench mark alternative locations for treatment was the findings that only a limited part of the global discharge of pharmaceuticals in Denmark in fact happens in the hospitals (see Chapter 2 and 3). It was therefore important to evaluate process efficiency, but also the cost for dedicated treatment for removal of pharmaceuticals at other potential locations.

7.2 Wastewater from a hospital department where highly toxic substances are used

Pilot tests performed with MBBR on effluent from the dept. of Oncology has already been reported (Casas et al., 2015a and Kragelund et al., 2015). In present report, the HYBAS on wastewater from Oncology department are reported and can also be found in Casas et al., 2015 b.

For smaller hospitals, where it is possible to isolate a few toxic wastewater streams, a decentralized treatment could be an option. The remaining wastewater could then be discharged into the sewer without any treatment as composition is similar to normal municipal wastewater. Treatment of wastewater from an Oncology Department represents this case.

Costs for treatment of combined effluent from a hospital are reported in chapter 6.1. In Table 13. CAPEX (excluding cost for capital) and OPEX are estimated based on same principles for a MBBR plant of lower capacity (70,000 m^3/y), representing the case with treatment of selected more toxic wastewater from hospital.

Table 13. CAPEX estimate for wastewater treatment plant for hospital wastewater capacity 70,000 m^3/year.

	Million DKK
Total Equipment, Engineering, Erection, Commissioning etc.	12 - 15
Total Civil Works	3.5 - 5
Total Plant excluding ozone	15.5 - 20

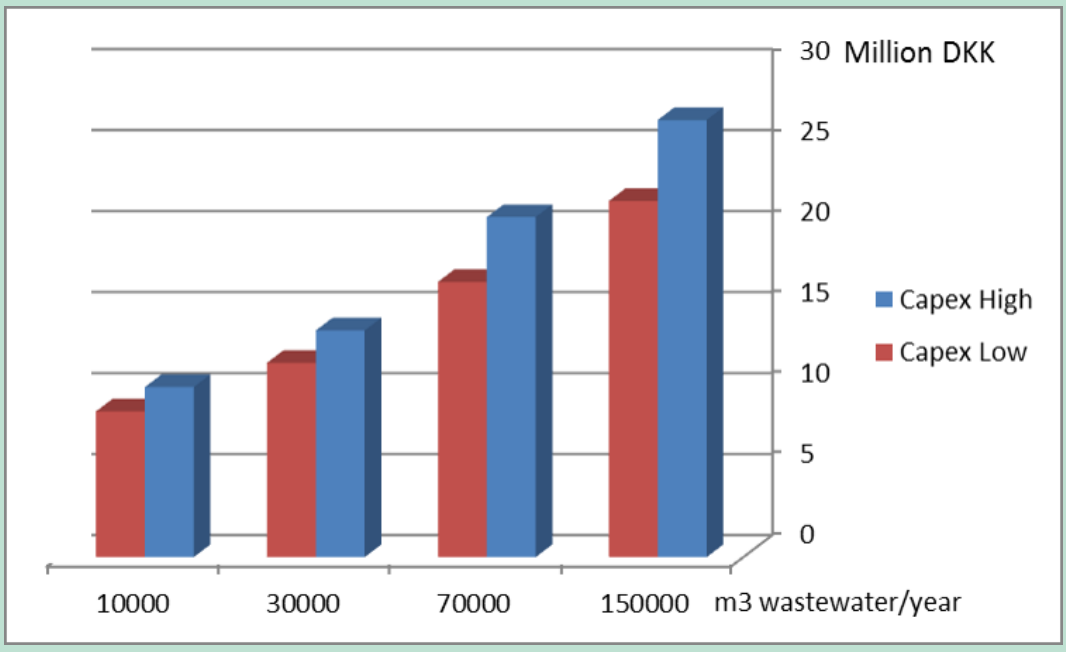


Figure 59. CAPEX estimate for MBBR hospital wastewater treatment plant at varying capacities/year.

Figure 59 shows the influence of plant capacity on CAPEX. The smaller capacity, the higher cost per m^3 capacity, which is not unique for this treatment concept. The same consequence of lower plant capacity is reflected on OPEX as well. It is not the cost for consumable which makes the big difference, but cost for analysis, plant surveillance etc. OPEX are to be covered by fewer m3 treated wastewater, hence cost per m3 treated water increases significantly.

Table 14 shows estimated OPEX (excl. capital cost) for a plant with capacity 70,000 m^3/year. Compared to a plant with approx. the double capacity (Table 12) the operation cost per m^3 for the lower capacity plant is approx. 50% higher.

Table 14. Operational Expenditures for wastewater treatment plant for hospital capacity 70,000 m^3/year

	DKK/Year	DKK/m^3 Wastewater
Total Consumables*	205,000	2.70 - 3.50
Analysis	100,000	1.43
Maintenance plant	430,000	6.10
Operation	150,000	2.14
Total OPEX	**885,000**	**12.40 - 13.20**

* Electricity (0.75 DKK/kWh), Chemicals, Ozone, Screenings and Sludge

7.3 Full stream treatment at the municipal wastewater treatment plant

Biological wastewater treatment is implemented in most of the developed countries and in Europe and North America also with nutrient (N and P) removal. Project focus has therefore been to study methods to upgrade existing facilities to accommodate pharmaceuticals removal. Two options have been tested to obtain improved removal of pharmaceuticals at the municipal wastewater treatment plant.

- Incorporating improved treatment in the existing biological wastewater treatment plant by modifying the activated sludge process to HYBAS™ (IFAS, Integrated Fixed-Film Activated Sludge type process)
- Incorporating a polishing step for the effluent from a traditional activated sludge WWTP (with N+P removal) for removal of pharmaceuticals based on a biofilm process (MBBR)

7.3.1 Modification of activated sludge process to HYBAS™

To promote growth of bacteria with capabilities to degrade pharmaceuticals in an activated sludge plant with nitrogen removal, carriers for biofilm growth are introduced in some of the process tanks. In the biological reactor where most of the degradable COD has been removed from the wastewater, there is a potential for a slow growing biofilm on carriers. These bacteria will, due to limited access to easy degradable organics, specialize in degradation of slowly degradable organic compounds, i.e. pharmaceuticals. The bacteria will be maintained in the same reactors corresponding to a long sludge age as opposed to conventional activated sludge plants.

It is difficult to make representative CAPEX and OPEX estimates for retrofitting existing plants with HYBAS™ as plants are very individually designed. Furthermore, there is a huge capacity range from small municipal to large scale wastewater treatment plants. Therefore, a brief description of the financial consequences of this option is listed below.

Modification of an existing WWTP with activated sludge to HYBAS™ would mean adding carriers to the process tanks (preferably two or more tanks in series) where the soluble COD is low and where the main nitrification process takes place. The tank(s) now with carriers, will need to be retrofitted with sieve(s) in order to prevent carriers from leaving the tank together with the tank effluent. Existing aeration system (diffused aeration system) will meet the mixing and aeration requirement, but in certain cases additional mixing and/or aeration could be required. The carrier filling rate in HYBAS™ tanks is between 40 – 50%. In addition, carriers provide additional biological treatment capacity for the tank. Under normal conditions, the treatment capacity (COD/N removal) for HYBAS™ tanks with 40 -50% carriers is approximate-

ly 50% higher than a tank with only activated sludge. However, increasing of plant load normally require hydraulic extensions, which in such case also need to be considered.

Retrofitting to HYBAS™ process for improved removal of pharmaceutical would normally not be associated with a load increase. However, a certain load increase can be allowed if there is hydraulic capacity available.

The plant will in addition to improved degradation of pharmaceuticals, also experience improved nitrification capacity and a nitrification process less susceptible to low water temperature regardless of treatment capacity. The advantage of HYBAS™, is no need for additional tank capacity, but during installation, a partially close down during retrofitting is expected. However, temporary close down could though be performed during hottest season sufficient nitrification capacity probably would be available, despite reduced tank volume.

7.3.2 Polishing of effluent from municipal WWTP with MBBR alternating operation

As described in Chapter 4.3, the most efficient biological treatment technology investigated in bench scale, was identified as polishing MBBR reactors with alternating operation, MBBR A/O. This would be the easiest option to implement in full scale at a municipal plant, as there is very limited interference with the existing plant as only additional space in the vicinity of the plant is required. Figure 60 show the process principle.

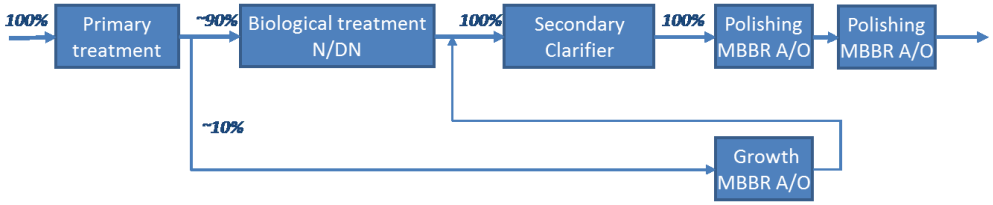


Figure 60. MBBR Alternating Operation incorporated in typical municipal WWTP scheme.

The polishing MBBR reactors operate in a rotating mode allowing bacteria at intervals to be `off line´ in order to get access to easy degradable carbon source to cover their demand for basic cell growth. When `in line´ the availability of carbon is extremely limited as COD in treated effluent, being the feed, is very low. The purpose of the `off line´ period is to provide cell growth to compensate for cell decay during their `in line´ operation, however without allowing for overgrowth of bacteria in the biofilm feeding on difficult degradable pharmaceuticals. The process is a new innovation and a patent application has been filed (Swedish patent application no 1650321-1). In principle, the process also offers a 10% increase in treatment capacity for COD and N removal, however no additional clarifier capacity is part of the concept, i.e. no additional hydraulic capacity is provided.

The design parameters for the process is not yet optimized and validated in semi industrial scale. Process optimization and validation is proposed as a process maturation development project, for which the Mermiss partners have applied for partial public funding. Process design for CAPEX and OPEX estimate for polishing treatment option is therefore based on results from the smaller scale continuous tests performed in the project and is therefore subject to some uncertainty.

Switzerland is the only country in Europe having a law regarding micro-pollutants discharged from municipal wastewater treatment plants. The Swiss law requires primarily 100 of the total 700 treatment plants in the country to implement the fourth step. The selected plants are larger ones and they are supposed to treat 50 % of the total wastewater. Their target is 80 % remov-

al of micro-pollutants in the upgraded plants, resulting in the 50 % elimination of micro-pollutants in the country (Mulder M. et al, 2015).

There is no decision yet on removal of pharmaceutical in municipal wastewater treatment plants in Denmark, hence there are no target effluent concentrations defined. It is therefore not possible to make an estimate for a dedicated treatment concept. For that reason, it has been decided to make an estimate for a biological post treatment solution with alternating MBBR and separately, an estimate for additional ozone treatment if further removal would be required. As can be seen in Chapter 4.3 the MBBR alternating polishing process has for many of the pharmaceuticals achieved 50 - 100% removal. In case ozonation is required to reach certain concentration targets, the efficient biological polishing has reduced the ozone dose requirement considerable.

Combined sewer systems still exist in many municipalities in Denmark, which means at times the WWTPs receive very high flows during rain events. At present, it has been decided to make a design for average daily flow, though depending on future regulation other design basis could be required. Contrary to activated sludge systems, the treatment capacity is not severely influenced by a considerably higher flow as the bacteria are attached to carriers and stay in the reactors regardless of flow. The effect of a larger flow might only be a reduced performance due to shorter retention time and quantity of removed compounds will be unchanged.

Estimate of capital expenditures (CAPEX) is shown in Table 15 and Table 16 show the operational expenses (OPEX), both based on the same principles as described in Chapter 6.1.2. Plant capacity used for design example is average flow of 1200 m^3/h and a yearly flow of 9 million m^3. The numbers indicated for ozone are given to indicate the cost for ozonation in case required.

Table 15. Capital Expenditures for Polishing MBBR.

	Million DKK
Total Equipment, Engineering, Erection, Commissioning etc.	20 - 24
Total Civil Works	6 - 8
Total Plant excluding ozone	26 - 30
Ozone Installation*	7 - 12

* In case required

Table 16. Operational Expenditures for polishing MBBR and cost for ozonation if required.

	DKK/Year	**DKK/m^3 Wastewater**
MBBR (mainly electricity)	900,000	
Analysis	150,000	
Maintenance	600,000	
Operation	200,000	
Total OPEX	**1,850,000**	**0.21**
OPEX Ozone 5 mg O3/l	**2,200,000**	**0.24**
OPEX Ozone 10 mg O3/l	**3,400,000**	**0.38**

7.4 Conclusion

The aims of this project were to test different process configurations (biofilm alone or in combination with activated sludge) as well as different locations for treatment e.g. toxic wastewater side-streams or whole wastewater stream from a hospital, or treatment at the municipal wastewater treatment plant. This work made the comparison of the treatment technologies and treatment locations possible, both in terms of efficiency of pharmaceutical degradation and the estimate of related costs (CAPEX and OPEX). To our knowledge, this is the first project that has provided comprehensive on-site treatment of pharmaceuticals in scalable systems, and has assessed the benefits at all levels of centralized and decentralized solutions for removal of pharmaceuticals from wastewater.

The outcome of this project emphasized the superiority of biofilms to degrade pharmaceuticals, regardless of treatment location. It was documented that promoting and maintaining bacteria specialized in especially medium-degradable and hardly degradable compounds like diclofenac, were possible (Casas et al., 2015 a, b) and even to degradation degrees not observed before (Tang et al., 2017). Substantial biological reduction of pharmaceuticals was documented also to higher degrees than observed in similar full-scale plants (Grundfos Biobooster, 2016). The efficient biological removal significantly influenced the subsequent ozone dose required for removal of residual pharmaceutical concentrations. By investigating several different treatment locations, a benchmarking could be carried out including degradation capabilities and operation and maintaining costs, see Table 17. More detailed calculations, assumptions and estimates for the different options, can be found in chapter 6 and 7.

Table 17. Overview of economical calculations for removal of pharmaceuticals directly at the hospital and at the municipal WWTP.

Location	Treatment at hospital		Treatment at municipal WWTP	
		DKK/m3 Wastewater		DKK/m3 Wastewater
Water volume	150,000 m3/year		12-15 million m3/year	
Plant costs	21-26 million DKK		26-30 million DKK /excl.ozonation equipment	
Total consumables*	405,000 DKK/ year	2.70 - 3.25	900,000** DKK/ year	
Analysis	100,000 DKK/ year	0.67	150,000 DKK/ year	
Maintenance plant	575,000 DKK/ year	4.35	600,000 DKK/ year	
Operation	150,000 DKK/ year	1.00	200,000 DKK/ year	
Total OPEX	1,230,000 DKK/ year	**8.20 - 8.70**	**1,850,000 DKK/ year**	**0.21**

* Electricity (0.75 DKK/kWh), Chemicals, Ozone, Screenings and Sludge

** MBBR (mainly electricity)

The costs of a decentralized solution at a Danish medium-sized hospital are 8.2-8.7 DKK/m^3, which is significantly less per m^3 treated wastewater than the MBR solution built in Denmark (Grundfos Biobooster, 2016).

However, only a minor fraction of the consumed pharmaceuticals in Denmark is targeted by this decentralized treatment solution (estimates between 1-4 %, Mose-Pedersen et al., 2007). In addition, the pharmaceuticals identified as problematic for the environment (Local Government Denmark (KL), 2013, AMK 2015) are in fact discharged from private homes rather than from the hospitals (Møller, Environmental report, 2014, this report). Therefore, it is necessary

to rethink how to remove pharmaceuticals from wastewater where the majorities of the pharmaceuticals are present. Here, the most logical treatment site would be as post treatments at municipal wastewater treatment plants.

The new innovative polishing MBBR solution tested at the Viby municipal WWTP showed promising perspectives, as the costs for treatment were rather low and a complete biological removal of pharmaceuticals was achieved. By treating both hospital wastewater and household wastewater with the mentioned polishing technology, the pharmaceuticals discharged in the wastewater will be targeted regardless of origin. However, much larger water volumes are treated by the municipal polishing solution and thereby the load in kgs of pharmaceuticals to the environment will be dramatically reduced. The costs pr. m^3 for treating wastewater with the MBBR polishing technology were low, so it would still be a feasible solution even with a larger wastewater volume. It is expected that the MBBR polishing can be further optimized (in terms of feeding regimes, HRT etc.), and there is a need for upscaling the process from bench-scale to at least pilot-scale. Therefore, from an environmental point of view, degradation of pharmaceuticals should be carried out centrally at municipal WWTP rather than at the point sources alone. Centralized removal of micropollutants is conducted in Switzerland, which is considered to be the leader within this area.

8. Abbreviations

Actiflo®	Ballasted sedimentation process
Actiflo®Carb	Ballasted sedimentation process where powdered activated carbon is added
Bioprotector®	Polishing MBBR for easy biodegradable dissolved organic compounds
CAPEX	Capital Expenditures
CAS	Conventional Activated Sludge
HYBAS™	Hybrid Activated Sludge (IFAS process)
IFAS	Integrated Fixed Film and Activated Sludge
KL	Kommunernes Landsforening, `Local Government Denmark´
MBBR	Moving Bed Bioreactor
OPEX	Operation Expenditures
PAC	Powdered Activated Carbon
AUH	Aarhus University Hospital

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Environmentally friendly treatment of highly potent pharmaceuticals in hospital wastewater

Teknologisk Institut har sammen med Aarhus Universitet, Danmarks Tekniske Universitet, Air Liquide, Krüger Veoilia, Herning Vand, Aarhus Vand og Det Nye Universitetshospital Aarhus gennemført MUDP-projektet "Miljøeffektiv rensning af højpotente lægemiddelstoffer i hospitalsspildevand", i daglig tale MERMISS. Projektet har til formål at udvikle en teknologi til rensning af lægemiddelstoffer fra hospitalsspildevand og fra centrale renseanlæg baseret på en biofilmløsning.

Den traditionelle metode til rensning af spildevand er baseret på aktivt slam. Metoden er effektiv overfor letnedbrydelige lægemidler, men ineffektiv overfor middelsvære og svært nedbrydelige lægemidler.

Teknologien med biofilm er testet i laboratorieskala og i pilot-skala på dels råspildevand med koncentreret indhold af lægemidler fra en kræftafdeling, dels blandet råspildevand fra et hospital, dels almindeligt råspildevand fra Herning Vand, og dels på udløbsvand fra Viby renseanlæg. Over 95% af den samlede belastning med lægemidler i miljøet kommer i dag fra almindeligt husspildevand, både fra håndkøbsmedicin og fra patienter i ambulant behandling.

Projektet gennemførte således en benchmarking af lægemiddelfjernelse på forskellige typer af spildevand, og kunne på den baggrund demonstrere, at en biofilmbaseret teknologi er langt mere effektiv end den konventionelle aktiv slam behandling, som bruges i dag. Bl.a. viser projektet, at teknologien med fordel kan anvendes til at efterpolere allerede renset spildevand, og at driftsomkostningerne til teknologien er relativt lave.

Resultaterne af projektet er så lovende, at de allerede er anvendt til at starte et nyt MUDP-projekt, MerEff, der tester teknologien til at efterpolere renset spildevand i større skala på Herning Vands renseanlæg.

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