

# Full scale advanced wastewater treatment at Herlev Hospital

Treatment performance and evaluation



Grundfos BioBooster A/S

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## 1 Foreword

This report is the final reporting of the private-public innovation project: “Test and Adaptation of Treatment Technologies for Hospital Wastewater - Pilot Treatment Plant Herlev Hospital”. The project period has been from September 2012 to January 2016.

Herlev Hospital is the plant owner. Grundfos Biobooster A/S, represented by Director Jacob Søholm, has been responsible for the overall project, MBR technology and the plant operation. DHI, represented by Chief Planner Ulf Nielsen, has been responsible for tests, evaluation and reporting. The other project participants have been UltraAqua A/S (ozone, activated carbon and UV (Ultra Violet light)) and Neutralox (Air treatment).

The project was funded by Herlev Hospital, The Capital Region of Denmark, The Market Development Fund, City of Copenhagen, Biofos A/S and The Municipality of Herlev.

This report was prepared by DHI with Ulf Nielsen as project manager.



## 2 Summary

Significant quantities of specialized pharmaceuticals are used in hospitals. These pharmaceuticals are excreted by patients via urine and faeces and end up in the wastewater, which also contains a mix of chemicals, viruses and resistant bacteria.

Many pharmaceuticals, such as antibiotics and cancer drugs, are toxic to aquatic organisms. Municipal treatment plants are not designed to remove these types of substances, which results in discharges to the aquatic environment. Also, harmful bacteria and viruses from patients can be spread via combined sewer overflows and flooding during heavy rainfall. Sewage workers as well as bathing visitors in the water areas may be infected.

For these reasons, the Danish environmental authorities want wastewater from hospitals with significant discharges of harmful substances to be treated at the source. But hospitals as well as municipalities need documentation of how hospital wastewater can be treated and whether it is technically/economically feasible.

Pre-tests in laboratory scale on possible innovative technologies were carried out by DHI for the Danish Environmental Protection Agency (EPA) in 2010-2011. These pre-tests were followed by targeted pilot and laboratory tests of the wastewater from Herlev Hospital. The laboratory tests showed that membrane bio-reactor (MBR) technology combined with activated carbon, ozonation and UV was efficient in relation to the critical pharmaceuticals and pathogens in the hospital wastewater.

But the pre-tests did not show how the technologies should be combined and adjusted to the continuous flow of hospital wastewater in full scale. At the same time, the treatment efficiency needed to be tested for the removal of a large number of pharmaceuticals and xenobiotics, toxic effects on algae, daphnia, fish as well as hormone effects. Furthermore, the effectiveness of the treatment needed to be tested in relation to viruses and antibiotic-resistant bacteria.

On this background, the Capital Region of Denmark and Herlev Hospital decided in 2012 to initiate a private-public innovation project with the first full scale test of treatment of hospital wastewater in Denmark. Herlev Hospital is the plant owner and Grundfos BioBooster A/S has been responsible for the overall project. DHI has been responsible for the tests, development, evaluation, and reporting.

The overall objective of the project was to provide in-depth knowledge to Danish hospitals and environmental authorities, offering them a more solid basis for deciding whether hospital wastewater treatment is a viable solution in their local area. At the same time, the intention was to create a complete solution with treatment of wastewater, air emissions and sludge (drying) on site, having the potential to be exported worldwide.

Herlev Hospital is a large scale university hospital with 700 beds and a yearly wastewater volume of 150,000 m<sup>3</sup>. The hospital is now under expansion and in 2020, the hospital will have 900 beds and discharge 200,000 m<sup>3</sup> per year. The hospital serves 700,000 citizens within a large variety of medical specialities. Within cancer treatment, the hospital treats patients from all Zealand.

The Herlev Hospital wastewater treatment plant (WWTP) was constructed from 2013 to 2014 and has been operated since May 2014. The test period covered a period of 1.5 years from May 2014 to November 2015. The plant consists of a membrane bioreactor (MBR) with nitrogen and phosphorus removal, followed by a combination of polishing technologies. During the test period, the polishing step was split into two separate lines with different configurations, Line 1 and Line 2, which were operated in parallel. Line 1 consisted of granular activated carbon (GAC) treatment, followed by ozone and UV. Line 2 consisted of ozone, followed by GAC treatment and UV. Operation of the different setup of the two lines allowed for comparison of GAC and ozone treatment. After the test period, Line 1 was reconstructed to the same setup as Line 2

because the evaluation showed that Line 2 performed most efficiently on removal efficiency as well as on GAC consumption.

All solid waste streams (screenings, sludge and spent GAC) are sent to incineration at the local household waste incineration plant (850-1,200 °C), where 80% of the energy produced is turned into district heating while 20% is used for power supply.

A central air treatment unit with a photoionization process based on UV-light treats all vent air from the plant (vacuum in the building). Microbiological risk investigations of the air emissions showed that treatment worked efficiently. No complaints from neighbors or others concerning odour problems were registered.

The wastewater treatment performance was evaluated in-depth through a monitoring and testing programme. 118 samples were analysed for active pharmaceutical substances and in total, 122 substances were analysed. In addition, tests were performed for bacteria, virus and toxicity on water living organisms. An overview of analyse and test results from raw wastewater to final treated effluent is presented in Table 2-1.

Table 2-1 Overview of treatment performance. From raw hospital wastewater to final treated effluent

Parameters	Raw untreated wastewater	Treated wastewater
Toxic and persistent antibiotics (e.g. ciprofloxacin, clarithromycin and sulfamethoxazole), painkillers (diclofenac) and cytostatics (e.g. capecitabine)	Factor 10-300 exceeding of effect limits ( $PNEC_{Freshwater}$ ) for water living organisms	99.9% removal and no exceeding of effect limits ( $PNEC_{Freshwater}$ ) for water living organisms
Contrast media (e.g. iomeprol)	High concentration (2,5-7 mg/l)	99% removal
Antibiotic resistant bacteria	High occurrence of antibiotic resistant bacteria	No fecal or antibiotic resistant bacteria
Water born viruses (norovirus)	High concentration ( $1.7 \cdot 10^5$ )	Under limit of detection (<26 GC/l)
Fish fry (zebra fish)	100 % mortality within 96 hours	0 % mortality within 96 hours
Crustacean (daphnies)	No offspring (all test animals died)	Offspring survives as in clean control water
Estrogenic activity (A-YES)	Estrogen effects	No estrogen effects

Table 2-1 shows that the load of pharmaceutical substances were removed by 99.9% and that the substances still measurable in the effluent were below the effect concentrations for freshwater living organisms ( $PNEC_{Freshwater}$ ) without dilution. The highly persistent, but less toxic, contrast media were removed by 99%. Fecal and antibiotic resistant bacteria were removed and viruses, represented by norovirus, could not be detected. Ecotoxicity effects on fish and daphnies as well as estrogenic effects could not be measured in the final treated effluent.

Treatment performance in relation to general organic substances and nutrients was high compared to typical emission requirements. At the end of the test period, where the biological and chemical processes were optimized, COD, Total-N and Total-P were measured to respectively 10-20, 2-3 and 0.2 mg/l in the effluent.

The evaluation of the treatment setup showed that the MBR-ozone-GAC setup was the most efficient setup compared to MBR-GAC-ozone. The tests showed that the ozonation had a higher pharmaceutical removal efficiency when it was applied before GAC and at the same time, it made the GAC more efficient. The MBR-ozone-GAC was also observed to result in less GAC



usage, most likely because the general organic matter is transformed into more water soluble compounds by the ozonation. No critical formation of ozone by-products, such as bromate or NDMA, was observed.

The assessment of the overall economy was based on a registration of all operational expenditures. This included consumption of energy, chemicals, GAC and the costs for handling of by-products as well as man-hours for service. In addition, there is also maintenance costs for general maintenance of the plant. This was calculated as 2-3% of the investment cost per year.

The investment cost of a fully operational WWTP is assumed to 25-35 million DKK. The investment depends highly on the construction of the building for the WWTP. The actual investment at Herlev Hospital was high due to a wish to construct a building for the WWTP with special architectural features. The economical key figures are presented in Table 2-2.

Table 2-2 Overall economical key figures for the Herlev Hospital WWTP.

Type of cost	DKK	EUR
Investment cost	25 - 35 mill. DKK	3.3 - 4.7 mill EUR
Operation & Maintenance costs	10.87 DKK/m <sup>3</sup>	1.45 EUR/m <sup>3</sup>
Fee for discharge to public sewer	25.54 DKK/m <sup>3</sup>	3.41 EUR/m <sup>3</sup>

Herlev Hospital is presently paying a discharge fee of 25.54 DKK/m<sup>3</sup> for discharge of wastewater to public sewer. If, in the future, the wastewater is discharged directly to the nearby local stream (Kagså), this fee will no longer be applied, which will result in possible savings of running costs of 15 DKK/m<sup>3</sup> (25.54 - 10.87 = 15 DKK/m<sup>3</sup>). A win-win situation can be achieved, where pollutants are removed, the treated water is used for conservation of the local stream and overall wastewater costs are saved. It should be noted that depending on the specific future solution, there will be other costs related to the direct discharge, such as construction of a dedicated pipeline or costs for using the rainwater pipeline of the water company.

If the treated water is released directly to the local stream (Kagså) and from here further on to the marine bathing water area (Lodsparken), possible environmental and health risks have to be assessed. Therefore, risk assessments were carried out in the local water areas based on hydrodynamic modelling of spreading and fate of chemical and microbiological parameters. The results showed that the estimated risks were negligible during normal operation of the WWTP.

The high water quality of the final effluent opens up many options for reuse. Presently, reuse of the treated water in the existing cooling towers at the hospital is planned. Around 10,000 m<sup>3</sup>/y are expected to be reused here. Practical planning for the implementation of the direct release to Kagså is being carried out at the time of writing.



### 3 Introduction

Significant quantities of specialized pharmaceuticals are used in hospitals. These pharmaceuticals are excreted by patients via urine and faeces and therefore end up in the wastewater, which also contains a mix of chemicals and resistant bacteria and viruses.

Many pharmaceuticals, such as antibiotics and cancer drugs, are toxic to aquatic organisms. Municipal treatment plants are not designed to remove these types of substances, which results in discharges to the aquatic environment. Also, harmful bacteria and viruses from patients can be spread via combined sewer overflows and flooding during heavy rainfall and may also infect sewage workers.

For these reasons, the Danish environmental authorities want wastewater from hospitals with significant discharges of harmful substances to be treated at the source. But hospitals as well as municipalities need documentation of how hospital wastewater can be treated and whether it is technically/economically feasible.

Pre-tests in laboratory scale on possible innovative technologies have been carried out by DHI for the Danish Environmental Protection Agency (EPA) in 2010-2011 [1]. These pre-tests were followed up by targeted pilot and laboratory tests of the wastewater from Herlev Hospital [2]. The laboratory tests showed that membrane bio-reactor (MBR) technology combined with activated carbon, ozonation and UV was efficient in relation to the critical pharmaceuticals and pathogens in the hospital wastewater.

But the pre-tests did not show how the technologies should be combined and adjusted to the continuous flow of hospital wastewater in full scale. And at the same time the treatment efficiency needed to be tested for a large number of pharmaceuticals and xenobiotics, toxic effects on algae, daphnia and fish, hormone effects and mutagenic effects. Furthermore, the effectiveness of the treatment needed to be tested in relation to viruses and antibiotic-resistant bacteria.

With this background, it was decided in 2012 to initiate this private-public innovation project with the first full scale test of treatment of hospital wastewater in Denmark. The technical/environmental objectives of the project are to:

- Test and adjust a concept for treatment of hospital wastewater in full scale. The concept is a complete solution including treatment of wastewater, air emissions and sludge (drying) on site
- Monitor and evaluate the efficiency of the treatment concept in relation to wastewater discharges of hazardous substances and pathogens, air emissions and sludge production
- Carry out a performance evaluation of the technical concept as well as operational and energy costs
- Carry out a health and environmental risk assessment of the discharge of wastewater to the local water stream (Kagså) in three scenarios: 1) Discharge of non-treated raw wastewater, 2) discharge of treated wastewater and 3) discharge of treated wastewater during reduced treatment efficiency

Thus, the overall objective is to provide in-depth knowledge to hospitals and environmental authorities, offering them a more solid basis for deciding whether hospital wastewater treatment is a viable solution in their local area.

Currently, Herlev Hospital's wastewater is discharged to the municipal sewer system, but after the present test of the treatment plant, the treated water is planned to be released into the local small stream (Kagså). Here, the treated water will contribute to a more stable water flow in the stream during the summer months. At the same time, it is planned that part of the treated water will be reused as cooling water in the existing cooling tower at the hospital.

### 3.1 About Herlev Hospital

Herlev Hospital is the second largest hospital on Zealand (after Rigshospitalet) with one of the widest professional profiles in the Capital Region and around 4,000 employees. The hospital has a 24-hour emergency reception and a large variety of specialized medical departments. The emergency reception serves 425,000 citizens from nine municipalities. Within certain medical specialties, Herlev Hospital serves 700,000 citizens throughout the region. Within cancer treatments, Herlev Hospital also serves citizens outside the capital region.

Herlev Hospital was constructed between 1966 and 1976. The hospital is now under reconstruction and expansion, including construction of a new regional sterilization as well as emergency, mother-child and diabetes centers. The main part of the reconstruction is planned to be completed in 2018 and the diabetes center in 2020.

Table 3-1 Key figures for Herlev Hospital in 2015 and expected key figures in 2020 after the planned expansion of the hospital.

Herlev Hospital	Today	2020
Number of beds	691	900
Floor area (m <sup>2</sup> )	180,000	250,000 (+ Diabetes center)
Wastewater volume (m <sup>3</sup> /year)	150,000	200,000
Production value (mill. Euro)	520	Not known
Main activities	Oncology, nuclear medicine and therapy, neurology, medicine, nephrology (dialysis), woman-child diseases, surgery, cardiology, radiology, hematology and anesthesiology, incl. a multidisciplinary pain center, etc.	New regional sterilization center New Diabetes center New Mother-child center Largest emergency hospital in the Region

## 4 Danish authority regulation of hospital wastewater

Since 2009, several Danish ministers for the environment have stressed the need for wastewater regulation of hospitals. The need for reduction of pharmaceutical residues in the water areas as well as a limitation of the spreading of antibiotic resistant bacteria have been the primary arguments for the need of action.

The Danish municipalities are the authorities responsible for the discharge of wastewater from hospitals as well as wastewater from industries and other point sources. Back in May 2009, the Ministry of Environment issued an action plan for hospital wastewater, which makes it clear that when it comes to discharge of wastewater, hospitals need to be regulated like industries by the municipalities. Hospitals discharge hazardous compounds and pathogens and can therefore be considered as point sources just like industries.

The municipalities follow the principles from the general Danish EPA guideline for municipal regulation of industrial wastewater [3]. This means that all hospitals need a permit to discharge wastewater to the sewer. This permit regulates the discharges of both pharmaceutical substances and hazardous pathogens like resistant bacteria.

In 2009, none of the Danish hospitals had permits regulating the discharge of pharmaceuticals. Today, the municipalities have issued permits for around 1/4 of the Danish hospitals. The rest of the permits are planned to be issued in 2016 and 2017.

The Danish municipalities have been challenged by this task because of the complexity of hospital wastewater and consequently, in 2011, they formed a task group to find common solutions on how to carry out the regulation. In 2013, the task group composed a guideline, through a nationwide collaboration, outlining the procedure for regulation of hospital wastewater discharges to public sewers [1]. The main elements of the guideline are:

- A list of guiding limit values for pharmaceuticals
- A method to rank hospitals in larger and smaller point sources

### Guiding limit values

Limit values have been set for 40 pharmaceuticals. The limit values set the maximum acceptable concentrations in wastewater from a hospital being discharged to public sewers. The limit values are based on ecotoxicological data as well as measured/calculated removal rates in conventional activated sludge wastewater treatment plants. Limit values for selected indicator substances are shown in Table 4.1. The ABC-score indicates the inherent hazardous properties of the substances. The Danish ABC-system is briefly described below.

Table 4-1 Guiding limit values for selected indicator substances. A list of 40 substances is published in DK [2].

Substance	ATC code	ABC-score	PNEC <sub>Freshwater</sub> [µg/l]	Guiding limit value [µg/l]
Azithromycin	J01FA10	A	0.09	0.12
Capecitabine	L01BC06	A	0.2	0.34
Carbamazepine	N03AF01	B	0.5	5.0
Ciprofloxacin	J01MA02	A	0.089	0.17
Citalopram	N06AB04	B	8	99
Clarithromycin	J01FA09	A	0.06	0.095
Diclofenac	M01AB05; M01AB55; S01BC03	A	0.1	0.13

Substance	ATC code	ABC-score	PNEC <sub>Freshwater</sub> [µg/l]	Guiding limit value [µg/l]
Erythromycin	J01FA01	A	0.2	0.9
Ibuprofen	C01EB16; M01AE01; M02AA13	B	4	1.7x10 <sup>2</sup>
Paracetamol	N02BE01	B	9.2	4.2x10 <sup>2</sup>
Sulfamethoxazole	J01EE01	A	0.12	0.31
Venlafaxine	N06AX16	B	0.1	1.0

### Ranking hospitals after size as point sources

In addition to the limit values, the municipal guideline includes a tool for ranking hospitals as point sources after their importance, in order to focus regulation on the most significant sources. The ranking is based on several criteria, taking the total pharmaceutical consumptions into account as well as the hospital's antibiotics consumption compared to the consumption in the rest of the catchment area. Table 4-2 shows the criteria for ranking the hospitals.

The ranking system is based on the fact that we still have limited knowledge about the environmental impact of the pharmaceuticals. The ranking assessment is therefore based on different relatively easily accessible data - like building a puzzle with different "method bricks" and getting stepwise an idea of the whole picture.

The two first criteria are based on consumption of hazardous pharmaceuticals. The Danish EPA guideline has a prioritization system – the ABC-system – for organic substances, which categorizes the substances into three groups (the criteria for categorizing in ABC is described in [3]):

*A: Highly hazardous substances*, are unwanted in wastewater because of their inherent properties (not biodegradable at aerobic conditions, potential for bioaccumulation and/or high toxicity), and should be substituted or reduced to a minimum

*B: Hazardous substances* are to be reduced so that environmental quality standards are not exceeded. At the same time the B-substances should be reduced according to the principle of using best available techniques (BAT)

*C: Unproblematic substances* are substances that under normal conditions are unproblematic to discharge to a municipal wastewater treatment plant (easily biodegradable and/or low toxicity to water living organisms)

The next ranking criteria is the sum of exceeding the guiding limit values for pharmaceuticals. Here, "the toxic unit" principle is used to set a comparable number for the amount of toxicity in the wastewater. The sum of exceeding is based on specific sampling and chemical analyzes of the wastewater.

The last criteria is the contribution of antibiotics (excl. the less hazardous penicillins) to the wastewater treatment plant (WWTP) as a percentage of the total load discharged to the WWTP from the whole catchment area (sum of households and institutions/hospitals/industries). This criteria indicates whether or not the hospital is a dominant source of the local WWTP.

Table 4-2 Criteria to rank hospitals in Denmark based on their classification as point sources [3]

Hospital-source	A: Highly hazardous pharmaceuticals (kg/yr)	B: Hazardous pharmaceuticals (kg/yr)	Sum of exceedance of guiding limit values (Measured conc./ guiding limit value)	Antibiotics contribution (excl. penicillins to WWTP) (%)
Minor	<50	<300	< 5	<5
Medium	50-100	300-500	5 - 20	5-20
Major	>100	>500	> 20	>20

Table 4.3 is an overview of the hospitals in the Capital Region of Denmark and shows how they are ranked. The data in the table is a result of the extensive mapping and risk assessment effort by the Region to understand the scale and impact of hospital wastewater discharges. The Region is, depending on the success of the full scale test of treatment technologies at Herlev Hospital, considering whole effluent wastewater treatment at the major sources identified in Table 4-3.

Table 4-3 Ranking of hospitals in the Capital Region of Denmark based on their classification as point sources.

Hospital/ Psychiatric Center	No. of beds	Catchment/ Water area	A: Highly hazardous pharmaceuticals [kg/yr]	B: Hazardous pharmaceuticals [kg/yr]	Sum of exceedance of limit values [MC/GLM]*	Antibiotics contribution (excl. penicillins) [%]	Classification as point source
Bornholm	100	Small/ marine	15	134		58	Medium source
Amager	120	Large/ marine	25	205		2	Minor source
Psych. Ballerup	140	Large/ marine	9	33		< 0,1	Minor source
Psych. St. Hans	180	Large/ marine	68	50		< 0,1	Minor source
Gentofte	280	Large/marine	52	337		2	Medium source
Glostrup	310	Large/ marine	50	286		13	Medium source
New Northern Zealand	670	Small/ fresh	130	989		79	Major source
Hvidovre	800	Large/ marine	111	818	25	27	Major source
New Bispebjerg	860	Large/ marine	108	708	48 and 172	8	Major source
Rigshospitalet	1,100	Large/ marine	436	1,381	105	28	Major source
New Herlev	950	Large/ marine	181	700	104	18	Major source

\* [Ratio between Measured Concentration and Guiding Limit Values]

As described above, the Danish municipalities are now in a process of issuing wastewater permits to all the Danish hospitals for discharges to sewer. These permits contain requirements for ongoing mapping and measuring of pharmaceuticals in the wastewater as well as requirements for action plans for implementation of best available techniques to reduce the discharges of pharmaceuticals.

When assessing whether it is fair that a hospital should invest in pollution reduction, the municipality carries out a proportionality assessment. The implementation costs of the reduction measures are to be proportional with the reduction of the environmental impacts. The municipalities take the following considerations into account when they assess the proportionality:

- The size of the hospital as a point source (as exemplified in Table 4.3)
- The need for investments in the solutions
- Size economic issues (super hospitals compared to small specialized clinics)
- Plans for construction/renovation activities (it's much easier for hospitals in a construction phase to implement new wastewater treatment solutions)

Based on this, the municipalities decide whether solutions like full wastewater treatment, batch treatments of specific sewage streams, collection of urine from specific patients etc. are to be implemented.

If a full scale wastewater treatment plant is implemented, a direct discharge of the treated wastewater to the local water area will normally be the most suitable solution. In these situations, the permit for discharge to sewer will be replaced –after proper assessment of effectiveness and robustness of the new treatment solution– by a municipal permit for discharge to the local water area.



## 5 Raw hospital wastewater characterisation and discharge

Significant amounts of specialised pharmaceuticals are excreted from the patients at hospitals. Pharmaceuticals are active substances that are excreted to a large extent from the body, mainly by the urine and to a lesser extent by faeces.

The pharmaceuticals end up in the hospital wastewater and form a complex mixture containing hazardous pharmaceuticals, resistant pathogens, disinfection chemicals and radioactive isotopes. In Denmark, and in most urban areas worldwide, raw hospital wastewater is discharged to the sewer and further transported to the municipal treatment plant. This conventional discharge situation is illustrated in Figure 5.1.

Municipal treatment plants are not designed to remove these types of substances, which results in discharges to the aquatic environment. During heavy rain events, the sewers will overflow and a mixture of sewage and rainwater will end up in local water bodies and, in extreme situations, also overflow in living areas. Bypass of the municipal treatment plant will also cause discharges of non-treated diluted raw wastewater. These overflow situations represent a risk of infections from waterborne pathogens that are present in the hospital wastewater (see Figure 5-1).

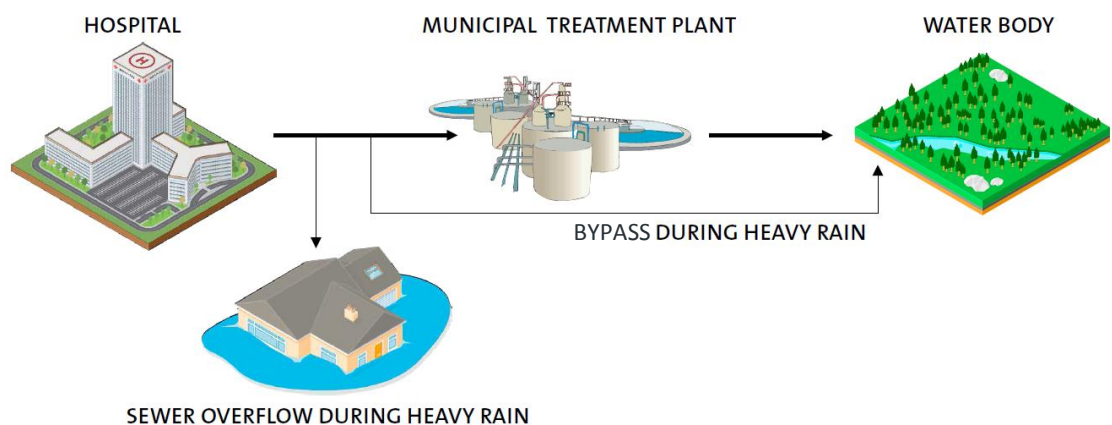


Figure 5-1 Conventional discharge of hospital wastewater to sewer.

Pharmaceuticals are biological active substances and they can impact water living organisms even in very low concentrations. Human pharmaceuticals like hormones, pain killers and anti-depressants can have adverse effects in fish, crustaceans and algae because these organisms have the same type of receptors (drug targets) as humans.

The effects on animals and plants can be very different from the intended pharmacological effects in humans, and knowledge is still lacking on possible adverse effects of the majority of pharmaceuticals. One of the most famous examples of unpredictable adverse effects is the almost total extinction of white-rumped vultures in India through carcasses of livestock treated with painkiller diclofenac. Another example is the feminisation of fish in freshwater areas due to the presence of contraceptive hormones from municipal wastewater.

In Denmark, selected pharmaceuticals have been monitored in freshwater streams in Zealand. Regarding pharmaceuticals that are used in large quantities in hospitals, the monitoring programs showed that antibiotics like sulfamethoxazole and clarithromycin are measured in freshwater in concentrations above the Predicted No Effect Concentrations (PNEC) [4]. Furthermore, sulfamethoxazole has been measured in sediments in the Baltic Sea, where also antifungal agent miconazole has been measured in blue mussels [7]. Herlev Hospital stands for 58%, 13% and 15% respectively of the total consumptions of sulfamethoxazole, clarithromycin

and miconazole observed in the large municipal wastewater treatment plant of the catchment area (Avedøre WWTP with 232,000 inhabitants), where Herlev Hospital is situated.

In a broader European context, selected pharmaceuticals have been chosen as “indicator substances” for monitoring programs or substances of particular concern. This is the case in the EU watch list, where six specific pharmaceuticals have been selected for monitoring in the member states [16]. In Switzerland, a list of 12 hazardous substances (10 pharmaceuticals) is used to evaluate WWTP polishing technologies [17], and in Sweden, 17 pharmaceuticals have been selected in addition to the EU watch list [7]. In North Rhine-Westphalia, a list of 17 pharmaceuticals is being discussed for WWTP monitoring programs [18]. These selected substances can be seen as representatives of the complex pharmaceutical consumption, which is seen in urban areas in general. At the same time, it is crucial from a treatment point of view to have the possibility of comparing the treatment performance across different matrices and techniques with the same substances.

Based on this, the present project has selected 16 indicator substances from the total number of 122 analyzed pharmaceuticals, which are highlighted in the report. The indicator substances represent the most environmentally critical substances measured in the wastewater from Herlev Hospital as well as the substances, which have been most difficult to remove in the treatment processes. At the same time, we also included the antidepressants citalopram and venlafaxine, plus the blood pressure medicine metoprolol, although they have not been really critical in this wastewater. They are included because of intense international attention. The indicator substances are presented in Table 5-1, where the measurements in raw wastewater are compared to the PNEC values for freshwaters. A comparison to  $PNEC_{\text{Freshwater}}$  without dilution is relevant since the treated wastewater is planned to be discharged directly to the small local stream (Kagså).

Today, a total of approx. 1,100 pharmaceutical substances are used in Denmark. In Herlev Hospital alone, around 850 different active substances are used and 509 (2012-data) are defined as relevant from a water environment point of view. 235 of these substances contribute by more than 2% of the total estimated load discharged to the municipal WWTP (Avedøre WWTP).

Table 5-1 shows that in average, all indicator substances are measured above the  $PNEC_{\text{Freshwater}}$  values except for citalopram, venlafaxine and metoprolol, which are included because of international attention as described above. All measured pharmaceutical substances are presented in Section 7.1.1.

The raw hospital wastewater contains significant amounts of antibiotic resistant bacteria. Critical resistant bacteria such as vancomycin resistant enterococci (VRE), cephalosporin-resistant coli bacteria (ESBL) and carbapenem resistant bacteria (CPE) are measured in the raw wastewater from Herlev Hospital. The spreading of resistant bacteria and resistance genes by wastewater represents a possible health risk. The magnitude of this risk has not yet been sufficiently studied or fully understood.

An environmental hazard profile of the raw wastewater from Herlev Hospital is summed up in Table 5-2. The profile is based on the comprehensive monitoring programme performed through the project period. Each parameter is elaborated in Chapters 7 and 8.

Table 5-1: Measured indicator pharmaceuticals in raw wastewater from Herlev Hospital (July 2014 – November 2015). Values exceeding PNEC freshwater values are highlighted.

Therapeutics	Substance	Samples No.	Average ng/l	Min. ng/l	Max. ng/l	PNEC <sub>Freshwater</sub> [17]
Antibiotic	Azithromycin	13	893	250	1,900	90
Cancer treatment	Capecitabine	14	805	14	2,300	200
Epileptic treatment	Carbamazepine	14	390	110	1,000	500
Antibiotic	Ciprofloxacin	14	13,486	1,800	27,000	89
Antidepressant	Citalopram	13	299	120	620	8,000
Antibiotic	Clarithromycin	14	2,650	100	7,800	60
Painkiller	Diclofenac	14	646	300	1,100	100
Antibiotic	Erythromycin	13	1,005	63	5,200	200
Cancer treatment	Ifosfamide	14	1,987	< 10	7,600	162,000
Painkiller	Ibuprofen	13	22,131	6,100	52,000	4,000
Contrast media	Iomeprol	14	2,889,286	150,000	5,000,000	1,000,000
High blood pressure	Metoprolol	14	2,450	1,200	3,900	62,000
Painkiller	Paracetamol	14	352,143	60,000	800,000	9,200
Antibiotic	Sulfamethoxazole	14	5,336	2,500	16,000	120
Antibiotic metabolite	N4-Acetyl-Sulfamethoxazole	14	5,107	1,800	13,000	120
Antidepressant	Venlafaxine	14	484	250	1,100	900

Table 5-2 Hazard profile of raw wastewater from Herlev Hospital

Parameters	Raw untreated wastewater
Toxic and persistent antibiotics (e.g. ciprofloxacin, clarithromycin and sulfamethoxazole), painkillers (diclofenac) and cytostatics (e.g. capecitabine)	Factor 10-300 exceeding of effect limits (PNEC <sub>Freshwater</sub> ) for water living organisms
Contrast media (e.g. iomeprol)	High concentration (2,5-7 mg/l)
Antibiotic resistant bacteria	High occurrence of antibiotic resistant bacteria
Norovirus	High concentration ( $1.7 \cdot 10^5$ )
Fish fry (zebra fish)	100 % mortality within 96 hours
Crustacean (daphnies)	No offspring (all test animals died)
Estrogenic activity (A-YES)	Estrogen effects



## 6 Layout of the Herlev Hospital WWTP and overall performance

### 6.1 Plant layout, process train and sampling points

A full scale wastewater treatment plant has been established at Herlev Hospital. The location of the WWTP at the hospital site is shown in Figure 6-1 (red circle at the lower right corner of the figure).



Figure 6-1 Location of the WWTP at the Herlev Hospital site.

From the outside, the WWTP consists of a 550 m<sup>2</sup> building and two biological process tanks of 6 m height, covering another 200 m<sup>2</sup> area. The WWTP building is shown in Figure 6-2.



Figure 6-2 WWTP building at Herlev Hospital. The two biological reactors are placed behind the building.

The plant is dimensioned to treat all wastewater from the hospital, which in terms of flow corresponds to an average inflow of 500 m<sup>3</sup>/d. The plant consists of an MBR stage for biological treatment followed by a polishing stage including adsorption by granular activated carbon (GAC), ozonation and UV-radiation. Critical air discharges as well as general vent air is treated in an air treatment system.

The layout of the plant is illustrated in Figure 6-3.

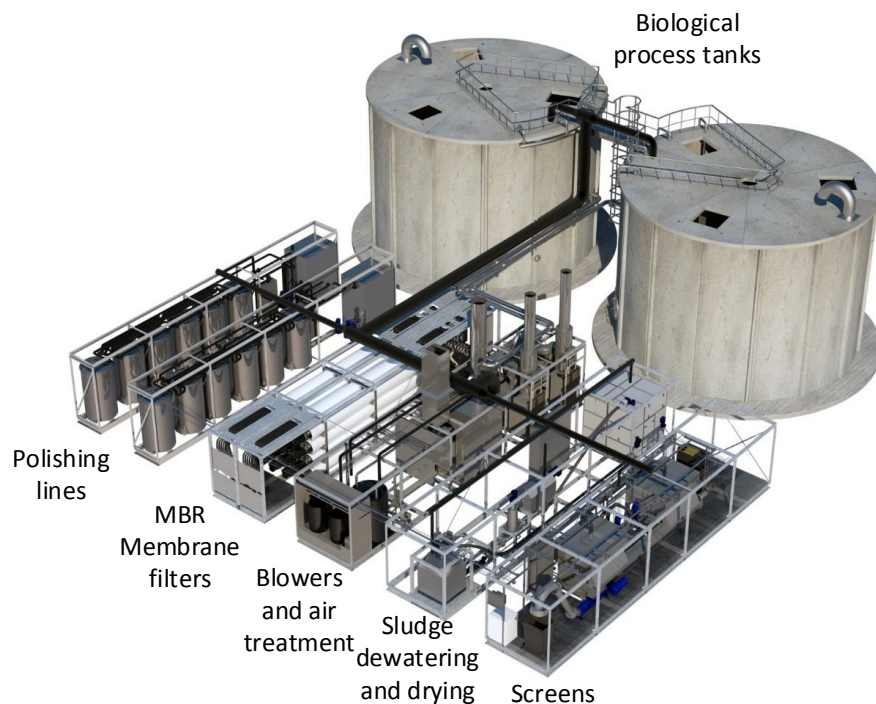


Figure 6-3 Layout of the Herlev Hospital WWTP.

As it appears from Figure 6-3, the process equipment is placed on separate frames, except for the biological process tanks. The frames are placed in a building (see Figure 6-2), whereas the process tanks are located outdoor.

Figure 6-4 shows a schematic process diagram of the plant. All wastewater from the hospital is collected through a separate sewer system, which discharges to a pump pit at the WWTP site. A grinder pump delivers the raw wastewater to a 1.5 mm screening facility. The screenings are dumped in big-bags, which are trucked to an incineration plant, and the screened wastewater is pumped to the biological process tanks.

The two process tanks are operated in parallel with intermittent aeration for nitrogen removal. Phosphorous is removed together with the surplus sludge through addition of aluminum coagulant. The biological sludge is separated from the treated wastewater through membrane filtration according to the MBR principle, using ceramic membranes with a nominal pore size of 0.2 µm. The membranes are configured as discs stacked on a central hollow shaft and placed horizontally in a cylindrical fiber glass housing. Membrane scouring is accomplished by rotation of the discs, and the permeate is collected in the central shaft. The plant is equipped with a total of 16 membrane filter units (MFU), which are connected to a permeate buffer tank through a manifold-system.

The solids retention time (SRT) can be estimated to 30 days. This estimate is based on an average amount of 5,000 kg TS in the reactors and a daily treated amount of surplus sludge of 165 kg TS.

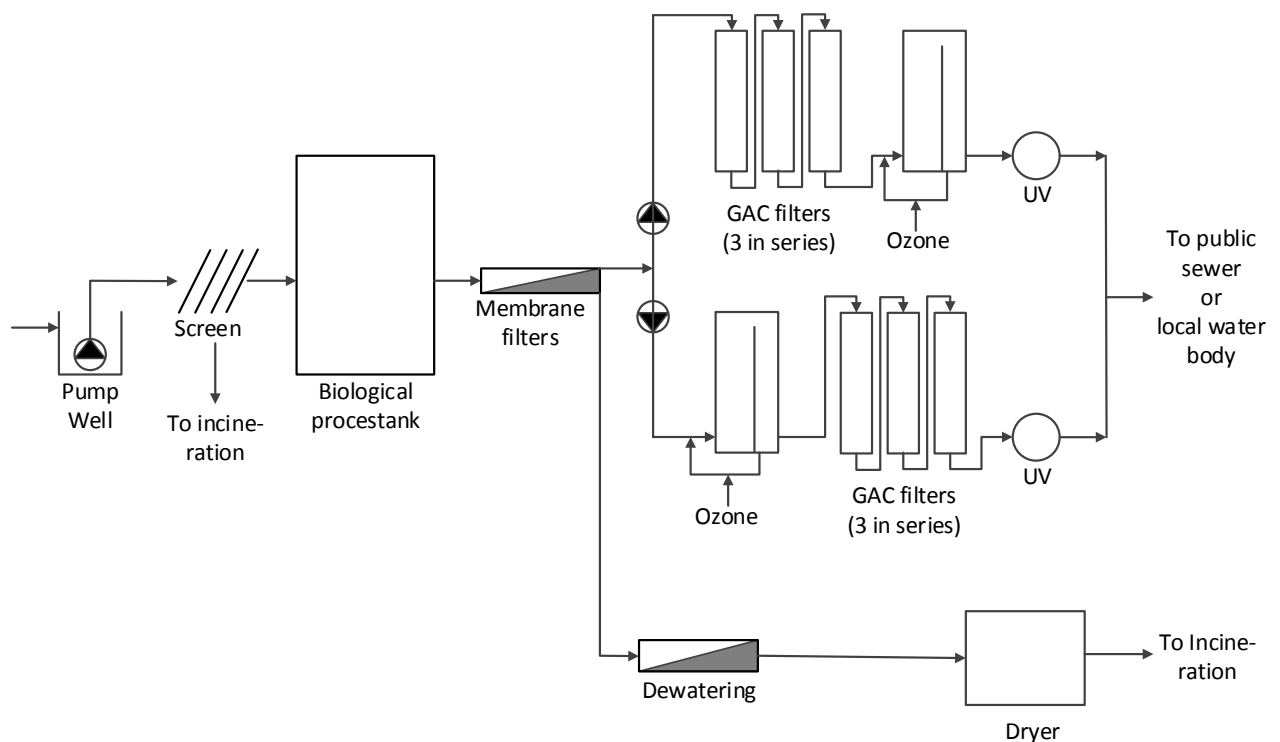


Figure 6-4 Schematic process diagram of Herlev WWTP

From the permeate buffer tank, the permeate is split into two flows leading to two separate polishing lines. Both lines include GAC, ozonation and UV. The only difference between the two lines is the order of the activated carbon and ozone.

The GAC stages are configured with 3 filter columns in series. Each GAC filter consists of two columns operated in parallel. Each column has an empty bed volume of 1.55 m<sup>3</sup>.

The reactors of the ozone stages have an active volume of 2.7 m<sup>3</sup>. The reactors are divided by an overflow weir into two chambers of 1.8 m<sup>3</sup> and 0.9 m<sup>3</sup>, respectively. At the maximum design flow of 13.5 m<sup>3</sup>/h, this corresponds to hydraulic retention times of 8 and 4 minutes, respectively. Ozone from an ozone generator is injected in a side stream loop of ozonated water, which is then mixed with permeate in a static mixer. The ozone rich water is led to the bottom of the first chamber of the ozone reactor (reaction chamber) and then flows over the weir to the second chamber (ozone decay chamber). Ozone is measured online in the off gas from the reactors in order to monitor the surplus concentration of ozone and control its dosage.

In the first seven months (June - December 2014) of the monitored period, the polishing lines were both operated with relatively low ozone doses of approximately 6 and 15 mg O<sub>3</sub>/l for Line 1 and Line 2, respectively. In this period, the typical level of Dissolved Organic Carbon (DOC) in the influent to the ozone reactor of Line 1 (GAC effluent) was 5 mg/l, and for the influent to the ozone reactor of Line 2 (MBR permeate) it was 10 mg/l. This corresponds to specific doses of 1.2 and 1.5 mg O<sub>3</sub>/mg DOC for Line 1 and Line 2, respectively, which is at a level often seen in international literature for polishing processes at municipal and hospital WWTPs.

However, it was noticed that the degree of removal of pharmaceuticals over the ozone stages was lower than expected, and the ozone doses were therefore increased to 15 and 24 mg O<sub>3</sub>/l for Line 1 and 2, respectively, in the remaining part of the monitoring period. In this second period, the general level of DOC in the effluent of the GAC filters of Line 1 increased to 6 mg/l, whereas the level in the permeate decreased to 7 mg/l. The specific doses in the period from January to November 2015 were therefore at a level of 2.5 and 3.4 mg O<sub>3</sub>/mg DOC for Line 1 and Line 2, respectively.

Each of the UV installations has one 220 W UV lamp. The inner diameter of the reactors is 150 mm and the length is 1,150 mm, corresponding to a reactor volume of 20 l. At the typical UV transmission level for polished water at Herlev WWTP (70%) and the typical flow interval of 10-15 m<sup>3</sup>/h, the obtained minimum doses (at the reactor wall) were in the interval of 5-10 mJ/cm<sup>2</sup>.

The effluent from the two polishing lines is discharged through a common outfall cascade connected to the public sewer.

The surplus sludge from the biological stage is dewatered in a screw press to approximately 20% Dry Matter (DM) and the dewatered sludge is then dried to 70-80% DM in a sludge dryer. The dried sludge is collected in big bags for transport to incineration.

Vent air as well as critical process air discharges are treated by a photoionization process based on UV-light in combination with catalytic converters creating strong oxidants that degrade the contaminants in the air.

The overall performance of the treatment plant is monitored through a routine sampling and analysis program covering traditional wastewater parameters such as COD, TN and TP (Chemical Oxygen Demand, Total Nitrogen and Total Phosphorous). In addition to this, but with a much lower frequency, a number of sampling and analysis rounds have been performed with pharmaceuticals, and occasionally other hazardous compounds, as the main analysis parameters. Figure 6-5 shows a simple flow diagram with indication of main sampling points.



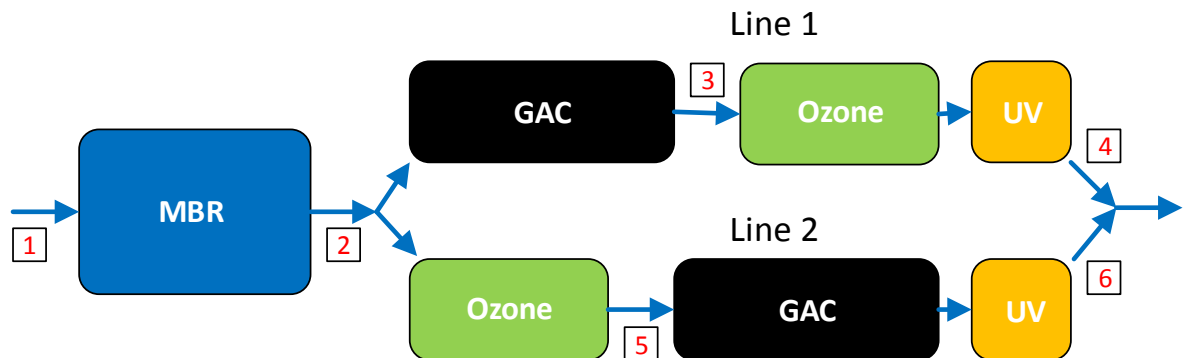


Figure 6-5 Simple flow diagram with main sampling points: 1) Influent to the MBR-plant (same as screened raw wastewater); 2) Effluent from the MBR-plant (same as permeate); 3) Effluent from the GAC-filters of Polishing Line 1; 4) Final effluent from Polishing Line 1; 5) Effluent from the Ozone-stage of Polishing Line 2; 6) Final effluent from Polishing Line 2.

Sampling points 1, 2, 4 and 6 are equipped with flow proportional sampling equipment, whereas samples from sampling points 3 and 5 are collected as grab samples

## 6.2 Operational incidents and change of GAC

Herlev Hospital WWTP was started up in the beginning of June 2014. During the period covered by this report, which is from the start up to the end of November 2015, a few unintended operational incidents took place. These incidents were primarily related to problems with the integrity of the membrane filters, resulting in transfer of biological sludge to the polishing lines, where it caused problems especially in terms of pressure build-up of the upstream GAC-filters. In the following, an overview of the disturbances, including their consequences in terms of change of GAC, is given:

November-December 2014: During November, a pressure increase was observed in Filter 1 of Line 1. On December 1, it was furthermore observed that *E. coli* was present in the permeate and in the final effluent after polishing. The membrane units were checked for their integrity and it was found that one of the discs had been hit by a small stone causing a piece of the membrane to break off. As a consequence of this, the membrane filtration unit in question was repaired on December 5. Additionally, Tank 1 in Line 1 was opened and the first 20 cm of GAC were substituted by fresh GAC. As this did not help to reduce the pressure, all GAC of the said tank was replaced on December 16.

May 2015: After the November-December incident, increasing pressure was observed for Filter 1 in Line 2. On May 5, this led to a complete change of the GAC for Filter 1 in Line 2.

June 2015: An operational incident took place during the night between June 29 and 30. The incident happened when one of the biological reactors was emptied in connection with exchange of the aeration diffusers. Due to maintenance tasks, the filter modules were operated in manual mode over the night of June 29, which caused some of the discs to crack. This resulted in sludge passing the membrane barrier and polluting the permeate, which again contaminated the upstream GAC filter of both polishing lines with activated sludge. During the following weeks, the cracked membrane discs were replaced by new discs and the GAC of Filter 1 of both Lines were replaced by fresh GAC. As a consequence of this incident, there was a downtime period for the polishing lines lasting from July 1 to July 10.

From the beginning of the period and until March 2015, there were recurring problems with elevated temperatures of the cooling water for the ozone generator. Too high temperature in the ozone generator results in reduced ozone production capacity, and this may lead to lower ozone doses than the operational set point. To ensure a sufficient ozone dose, it was necessary to

lower the flow of permeate to the polishing lines. Consequently, the polishing lines were unable to treat all the permeate produced until March 2015, when a new cooling system was installed.

### 6.3 Consumption of GAC

Consumption of GAC is a major operational cost and it is therefore important to keep track of the GAC change history. Table 6-1 shows the GAC changes carried out during the period covered by the report.

Table 6-1 GAC change history and bed volumes obtained for the changed GAC

Date	Filters changed	Quantity of GAC changed (kg)	Water received while in service (m <sup>3</sup> )	Corresponding no. of bed volumes
December 16, 2014	Filter 1, Line 1	1,457	23,331	7,526
May 5, 2015:	Filter 1, Line 2	1,457	43,588	14,061
July 8-9, 2015:	Filter 1, Line 1	1,457	53,566	17,279
July 8-9, 2015:	Filter 1 Line 2	1,457	55,077	17,767

Each GAC filter consists of two columns operated in parallel. As each column has an empty bed volume of 1.55 m<sup>3</sup>, each filter has a bed volume of 2 x 1.55 = 3.1 m<sup>3</sup>, and each polishing line therefore has a total empty bed volume of 3 x 3.1 = 9.3 m<sup>3</sup>. The density of the carbon is approximately 470 kg/m<sup>3</sup>, which means that each filter contains 3.1 m<sup>3</sup> x 470 kg/m<sup>3</sup> = 1,457 kg GAC.

An overall number of bed volumes realized for the two lines cannot be calculated, as the three filters of each Line have had their GAC content changed at different times (see Table 6-1). For every change of GAC, it is always the first filter in the direction of flow that is changed (the most polluted filter) and after a GAC change, the filter has its location changed in the flow order so that it is now the last filter in the direction of flow. The two other filters are then correspondingly moved backwards in the direction of flow. The number of bed volumes realized for the three filters of Line 2, as per November 2015, is shown in Figure 6-6.

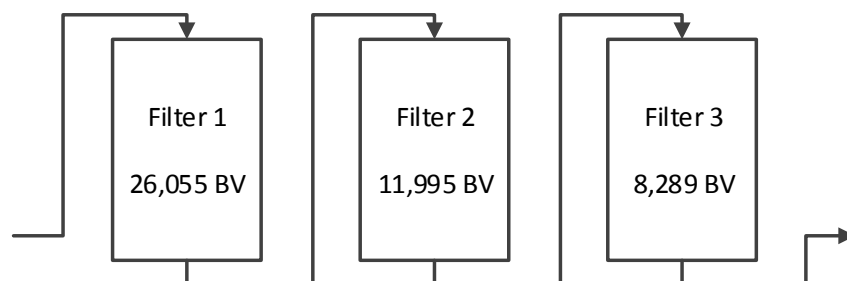


Figure 6-6 Number of bed volumes (BV) realized for the three filters of Line 2 as per November 2015.

The GAC of Filter 1 was never changed during the period June 2014 to November 2015. Its location in the direction of flow was, however, changed from being the last filter in June 2014 to being the first filter in November 2015. Filter 1 has therefore received all permeate led to Line 2 during the whole period, corresponding to 80,772 m<sup>3</sup>. With a volume of 3.1 m<sup>3</sup> for each of the filters, this gives 26,055 bed volumes as per November 2015. Filter 2 and 3 had their GAC

changed in May and July 2015, respectively (see Table 6-1), resulting in considerably lower bed volumes as shown in Figure 6-6.

When the Herlev Hospital WWTP is running in an optimized and routine mode, the general quantity of substituted GAC is assumed to be much less than indicated in Table 6-1, as the change in July of one filter in each line was solely due to an operational accident as explained in section 6.2. A best guess for the future quantity of GAC to be replaced, is one filter (1,457 kg) in each line per year, corresponding to a total quantity of approximately 3.000 kg/year for the WWTP.

## 6.4 Influent flow and flow through the polishing lines

Figure 6-7 shows the influent flow to Herlev Hospital WWTP. The flow pattern is due to weekly variations in the generation of wastewater, with the peaks representing the flow level on working days and the drops representing the flow level on weekends. As it appears from the figure, the flow level on working days is in the range 400-600 m<sup>3</sup>/d, whereas the flow level during weekends decreases to 200-300 m<sup>3</sup>/d. The average daily flow for the whole period (early June 2014 to end November 2015) was 420 m<sup>3</sup>/d.

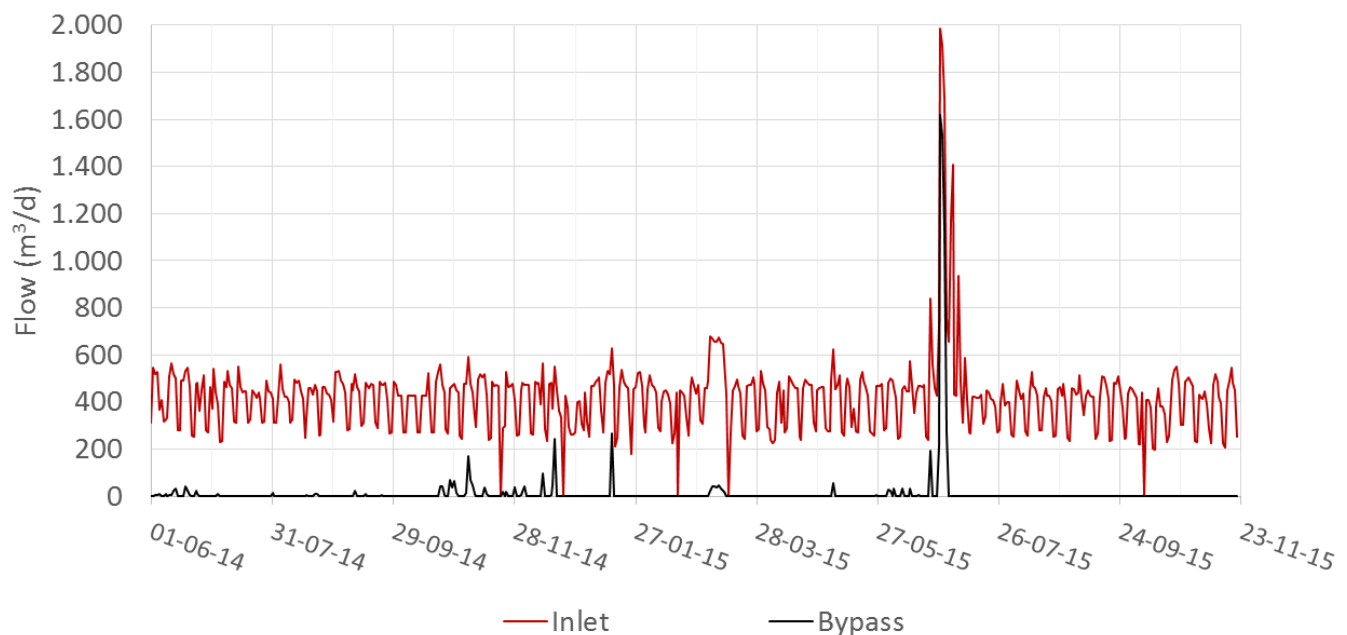


Figure 6-7 Influent flow and bypass flow at the Herlev Hospital WWTP from early June 2014 to end November 2015.

Figure 6-7 also shows the bypass flow, i.e. wastewater that bypasses the WWTP at the influent pumping station. As it appears from the figure, there are only a few incidences, where wastewater has been bypassed, and these incidences are primarily caused by influent flows exceeding the design capacity of 650 m<sup>3</sup>/d, typically caused by rainwater flowing into the wastewater collection system. This happens in spite of the fact that the wastewater collection system is a separate wastewater system, in principle without connection to the rainwater collectors. The very high peaks in June-July 2015 correspond to a period with heavy rains, where major overflows of rainwater collectors in the area affected the wastewater collection system. In these periods, it was necessary to bypass high quantities of wastewater/rainwater at the WWTP influent pumping station. The total quantity of permeate produced from June 2014 to November 2015 was 223,133 m<sup>3</sup>.

Figure 6-8 shows the flows to the two polishing lines (data on daily flow were not logged in the three first months of the period).

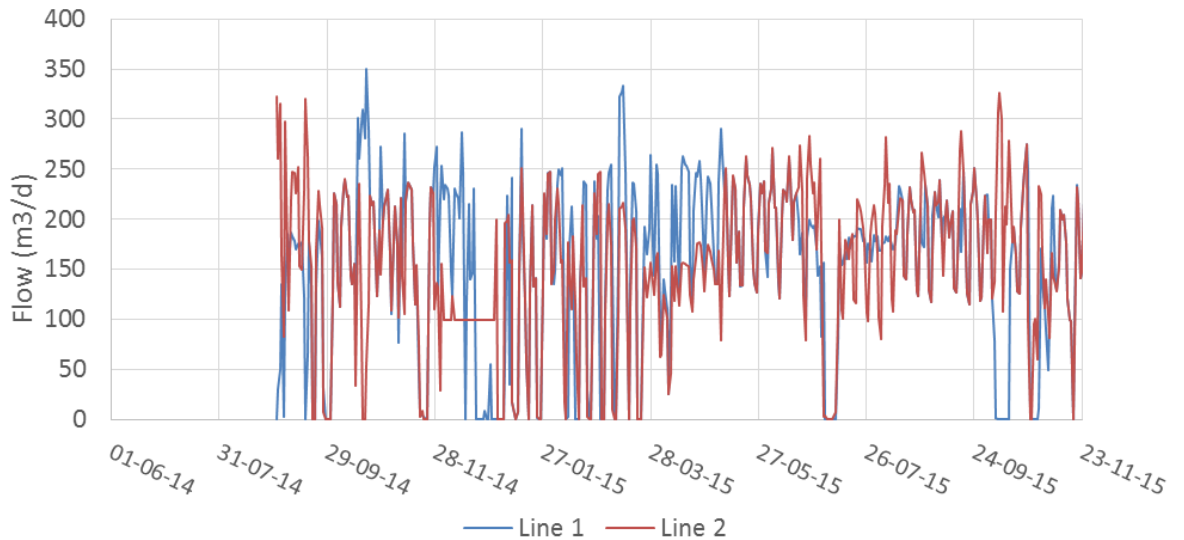


Figure 6-8 Flow to polishing Line 1 and 2

As it appears from Figure 6-8, the general flow level to the two polishing lines is in the range 200-250 m<sup>3</sup>/d on working days and 100-150 m<sup>3</sup>/d during weekends, corresponding to approximately half of the influent flow (see Figure 6-7). However, it also appears that there are regular periods of considerable length, where the flow goes to zero for both lines (e.g. January to March 2015). This has mainly been due to problems with the cooling system of the ozone generator, resulting in stops in the generation of ozone. In addition to this, it can be seen that there are periods where the flow of one of the lines is considerably smaller than half of the total influent (e.g. April 2015). This has been caused by pressure built up in the first GAC filter of the line in question, making it necessary to reduce the flow. During such periods with no flow or reduced flow to the polishing lines, it has been necessary to bypass part of the permeate directly to the outlet without polishing.

The total quantity of polished water in the period from June 2014 to November 2015 was 157,542 m<sup>3</sup>, of which the effluent from Line 1 and Line 2 amounted to 76,770 m<sup>3</sup> and 80,772 m<sup>3</sup>, respectively.

## 6.5 Overall Performance

The overall performance of the plant in the period of June 2014 to November 2015 is evaluated based on the results from monitoring of traditional wastewater parameters COD, TN and TP.

Figure 6-9 a) shows the COD concentrations in the inlet, in the permeate, as well as in Line 1 and Line 2. Figure 6-9 b) shows the same data for permeate, Line 1 and Line 2, but the scale of the Y-axis is smaller in order to allow for a more detailed analysis of COD concentrations after the biological stage. As it appears from Figure 6-9 a), the concentration of COD in the inlet water typically varies between 400 and 1,100 mg/l, and it further appears that the vast majority of the COD is removed in the biological stage. Figure 6-9 b) shows that COD in the permeate is relatively high in the beginning of the period, but then decreases and stabilizes at a level of 20-30 mg/l. It further appears that the polishing stage reduces the concentration of COD by another 5-10 mg/l so that the final effluent from the WWTP has a COD-level of 10-20 mg/l. The very low COD concentrations seen in September 2014 and May and July 2015 is due to the change of GAC (in August 2014 one filter was changed in Line 1, whereas in May and June 2015 one filter was changed in each Line 1 and 2).

Figure 6-9 c) shows the concentrations of TN in the inlet, permeate, Line 1 and Line 2.

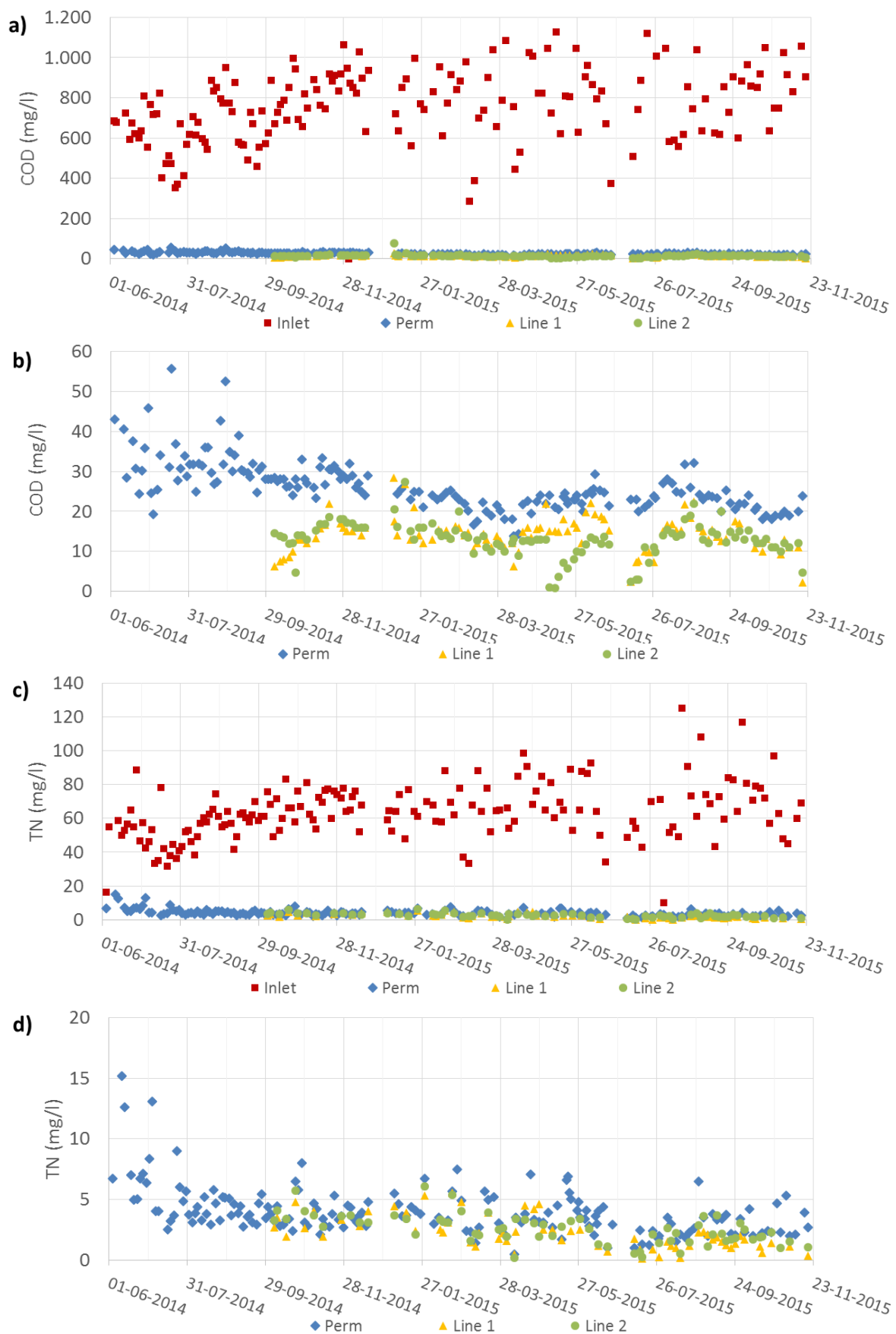


Figure 6-9 COD (a and b) and TN (c and d) concentrations in the inlet, permeate, Line 1 and Line 2.

Figure 6-9 d) shows the same data for permeate, Line 1 and Line 2, but the scale of the Y-axis is smaller in order to allow for a more detailed analysis of TN concentrations after the biological stage. From Figure 6-9 c) it appears that the concentration of TN in the inlet, typically varies between 40 and 100 mg/l and that the major part of TN is removed in the biological stage. It also appears that there is an increasing variation in the concentration of TN towards the end of the shown period. As expected, the polishing stage does not contribute much to the removal of TN (Figure 6-9 d).

Figure 6-10 shows the concentrations of TP in the inlet, permeate, Line 1 and Line 2. It appears from the figure that the typical inlet level of TP is in the range 10-20 mg/l, and a considerable part of TP is removed in the biological stage through assimilation by the bacteria. As expected, the polishing stage does not contribute much to the removal of TP, as the concentrations in the permeate and the polishing lines are largely the same.

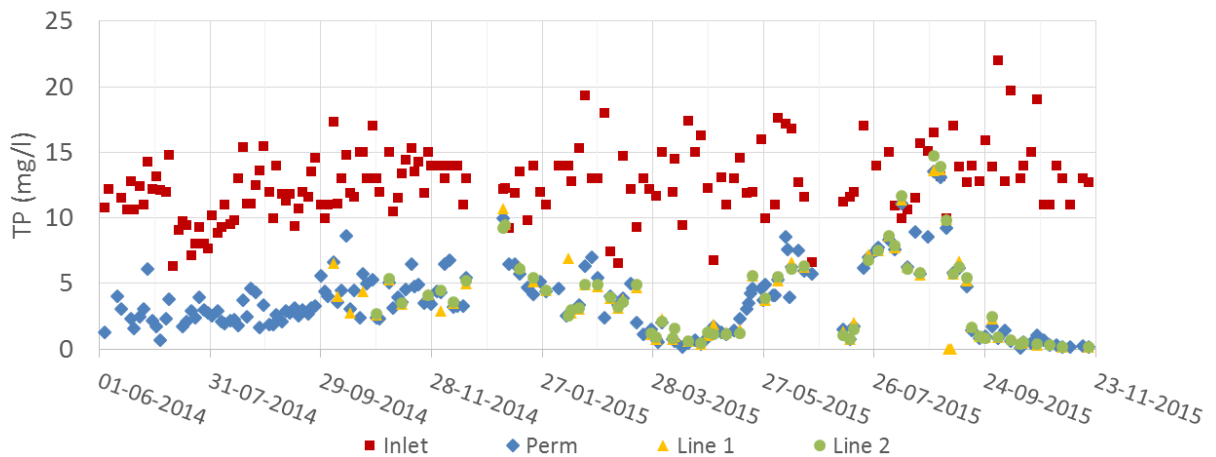


Figure 6-10 Concentrations of TP in inlet, permeate, Line 1 and Line 2.

In order to further remove TP, chemical precipitation is needed. At Herlev Hospital WWTP, precipitation of chemicals takes place by adding coagulant in the form of Polyaluminum Chloride (PAX) just before the MFUs. PAX was first added in April-May 2015, then again in July 2015 and finally from September 2015 and during the rest of the monitoring periods. This is reflected in Figure 6-10, where the concentrations of TP are reduced in these periods. The first two periods with dosing of PAX were decided in order to find an efficient procedure including dosing points and specific doses. The chosen procedure established in the last period resulted in an average TP concentration for November of 0.22 mg/l in the outlet.

## 6.6 Performance monitoring through online absorbance measurements

An online optical sensor manufactured by s::can has been installed at Herlev Hospital WWTP in order to test whether it is able to contribute to the efficiency of the monitoring program. The sensor used is a so-called spectrometer probe, which is able to measure absorbance for all wavelengths in the interval of 200-740 nm. The sensor is able to generate a so-called absorbance fingerprint, i.e. the absorbance for all wavelengths in the interval, but can also produce equivalent data to a number of traditional wastewater parameters, including TOC (Total Organic Carbon) and turbidity. To obtain these equivalent data, the absorbance at one or more specific wavelength(s) is transformed into concentrations through algorithms and calibration against data obtained by chemical analysis. The wavelengths and algorithms used for the different parameters are regarded as business secrets and only known by the manufacturer.

### Absorbance fingerprints

Initially, it was considered whether specific pharmaceutical compounds could be detected through absorbance fingerprints. Two pharmaceuticals, lomeprol and Ciprofloxacin, were tested. The compounds were added to distilled water in concentrations corresponding to the expected level in the wastewater as well as in concentrations 1-3 times higher in order to ensure a fingerprint, where the compounds can be clearly recognized. For both compounds it turned out that a clear peak indicating the presence of the compound could only be seen for concentrations higher than the expected concentrations in wastewater. When the two pharmaceuticals were added to actual polished water from Herlev Hospital WWTP, the peaks could still be distinguished in the basic fingerprint of the wastewater for the high concentrations of the added pharmaceuticals. As expected, the pharmaceuticals could not be detected for the low concentrations, corresponding to the expected concentration level in the wastewater (since they were not detected in distilled water without interfering absorbance from the wastewater). It was therefore concluded that the absorbance fingerprint cannot be used to monitor the presence of specific pharmaceuticals in wastewater, as their concentration in wastewater is too low to produce distinct peaks in a wastewater absorbance fingerprint.

### TOC equivalents

TOC equivalents (TOCeq) were measured online throughout the monitoring period. The idea was to use the TOCeq data as a substitution or supplement for chemical COD analysis carried out as part of the self-monitoring at the WWTP. Typically, the system was operated by alternating between longer periods of measurements on the permeate and the polished water.

Figure 6-11 shows COD measured by chemical analysis and against TOCeq measured by the s::can probe. The s::can probe was set up with a system of tubes and valves enabling the probe to receive water from different positions of the plant, one at a time, including permeate and polished water. In graphs showing long time series of s::can data, this is reflected as periodic missing data.

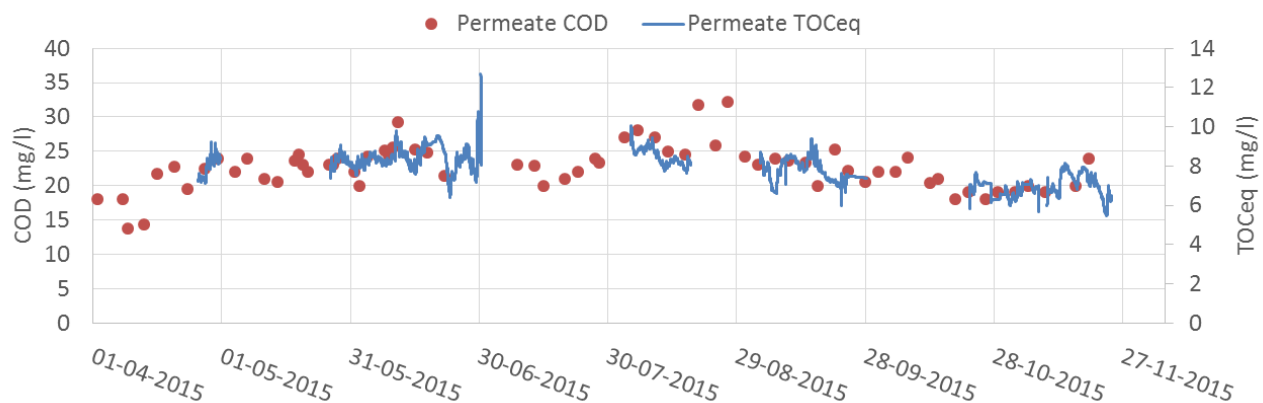


Figure 6-11 COD (chemical analysis) and TOCeq (absorbance measurement) in permeate.

As it appears from Figure 6-11, it seems that TOCeq reflects the concentration of COD very well with a COD/TOCeq ratio of a little less than 3. The peaks observed by the end of June coincide with a crack of a membrane in one of the membrane filtration units of the MBR stage and are caused by the presence of sludge in the permeate.

Based on these results, it is concluded that TOCeq can indeed be considered for use as an alternative or supplement for chemical COD analysis carried out as part of the self-monitoring at the WWTP. This could decrease time consumption for monitoring activities, since chemical COD analysis could be left out or measured at much lower frequency and at the same time provide much more detail as to the variation of concentration of organics in the monitored water

streams. The latter is illustrated in Figure 5-1 showing daily and weekly variation patterns for TOCeq.

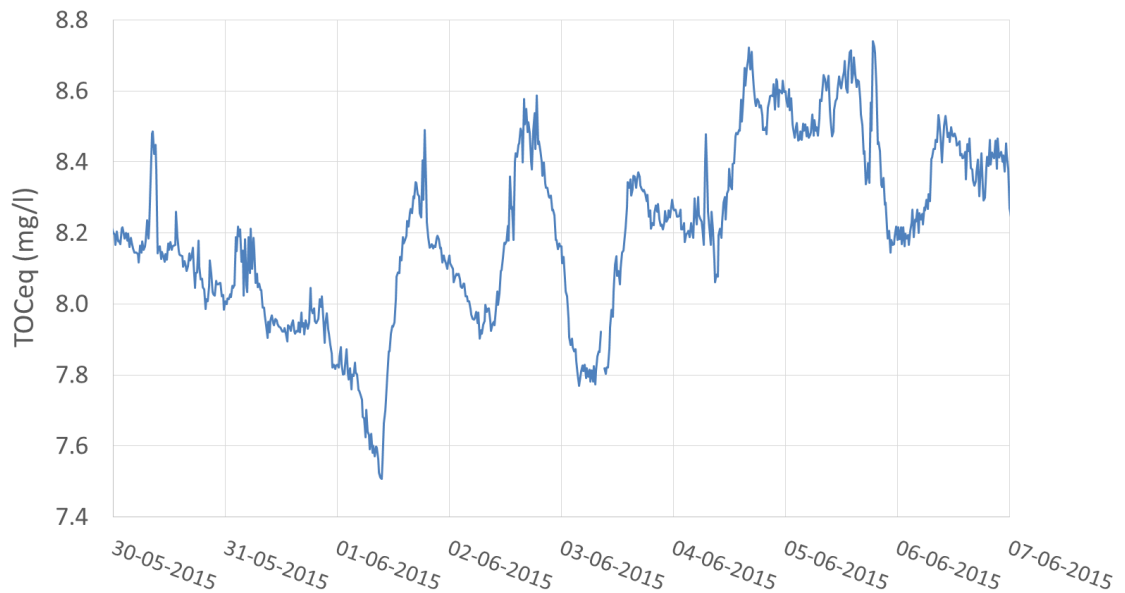


Figure 6-12 Daily and weekly variations in TOCeq.

As it appears from the figure, TOCeq decreases during the weekend of May 30-31 and then increases on Monday, following a more or less clear daily variation with low TOCeq values during the night.

### Turbidity equivalents

Monitoring of the presence of particles in the permeate is of major importance, as the presence of particles indicates potential problems with the integrity of the membrane filters. Traditionally, particles are measured as turbidity based on determination of the degree of light scattering, but light absorbance can also be used and the parameter is then called turbidity equivalents (TUReq) in order to distinguish it from traditional turbidity data.

Figure 6-13 shows the variation in TUReq in the permeate for the last 10 days of November 2015. The period shown covers two weekends and the five working days in between. As it appears from the figure, there is a clear daily and weekly variation with very low TUReq in the weekends and daily variations during working days with peaks in the afternoon, corresponding to flow variations.

Figure 6-14 shows data from turbidity measurements in the permeate for the same period, based on a traditional turbidity meter. As can be seen from the two figures, TUReq and turbidity show the same trend to a certain extent. In particular, it can be seen that while TUReq is stable and low during the first weekend, the traditional turbidity meter shows a couple of peaks, and furthermore that the TUReq peak observed between the 24<sup>th</sup> and 25<sup>th</sup> is less significant for the traditional turbidity measurement. There is no obvious explanation to these variations, but apart from the fact that the principle of measurement is different, it should also be noted that whereas the traditional turbidity meter is submersed in the buffer tank, which receives permeate directly from the membrane filters, the s::can probe receives permeate, which is pumped from the buffer tank through a tube system. If e.g. the peaks measured by the turbidity meter are actually due to gas bubbles (the permeate is depressurized in the permeate tank), these bubbles will probably have disappeared by the time that the permeate reaches the s::can probe.



It should be mentioned that the level of turbidity equivalents and turbidity reached on working days is not interpreted as loss of membrane integrity, since broken membranes or seals are known to result in considerably higher levels than what is observed here. It is not entirely clear what causes the increased levels of TUReq and turbidity at high flows, but very small particles (less than the nominal pore size of the membranes of 0.2 μm) and/or gas bubbles may play a role.

Based on the results, it is concluded that TUReq could be considered as a supplement to traditional turbidity meters for monitoring of membrane integrity.

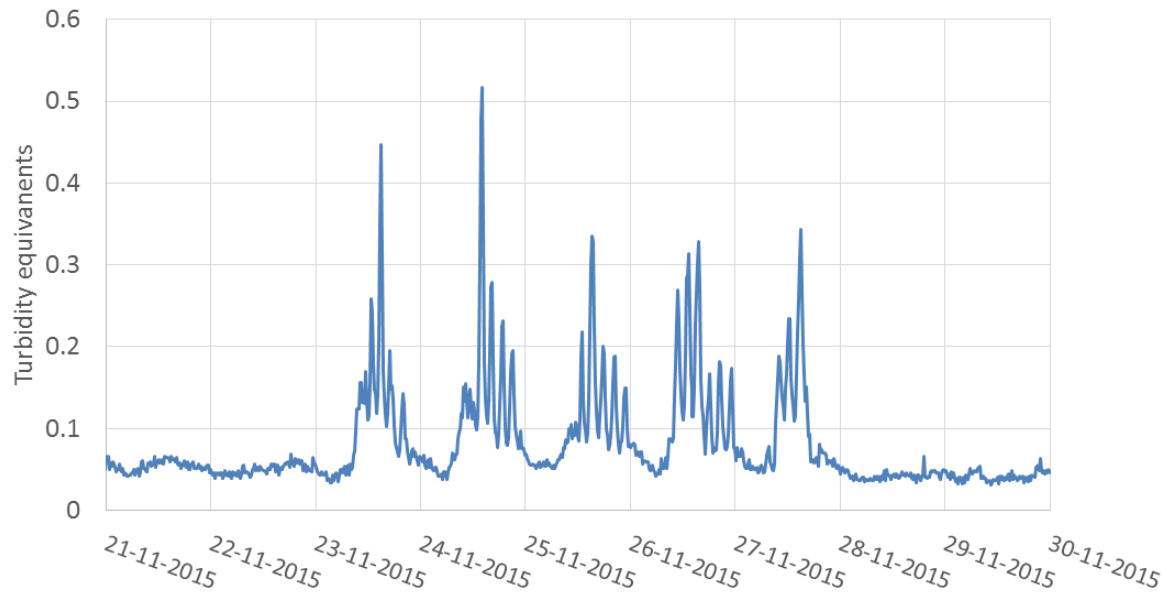


Figure 6-13 Turbidity equivalents (TUReq) in the permeate based on absorbance

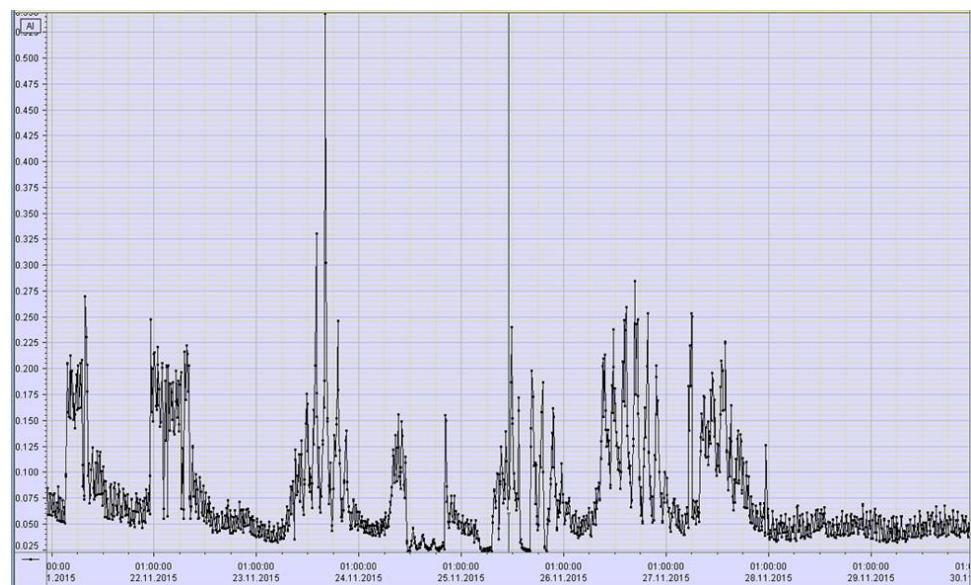


Figure 6-14 Turbidity (FNU) in the permeate measured by a turbidity meter. The period shown is the same as in Figure 6-13.

## 6.7 Chemical Enhanced Backwash

As part of the operational regime of the membrane filter units, a special membrane cleaning procedure is applied in order to reduce membrane fouling to an acceptable level. This procedure is called Chemical Enhanced Backwash (CEB) and involves addition of cleaning chemicals, including chlorine, to the filter discs. The chemicals will end up in the biological sludge, where it may result in the generation of chlorinated organic compounds (AOX). As AOX compounds are considered to pose a risk to human health, a campaign was conducted with the aim of assessing the AOX concentration levels in the permeate immediately after a CEB procedure of one of the filter units.

A CEB sequence includes the following steps:

1. Back-flush without chemical cleaning solution for 2 x 10 seconds.
2. Back-flow chemical cleaning solution until the filter discs are full. Then back-pulses once per minute with a back-pulse duration of 3 seconds. This is continued for approximately 40 minutes.
3. Recirculation of MLSS to the process tanks for 10 minutes without addition of chemical cleaning solution.
4. Permeation is resumed. Total time from start of the CEB-sequence to resume of permeation is 50 minutes.

Immediately after completion of a CEB procedure on one of the membrane filtration units, a permeate sample was taken directly from the relevant filter unit, and a couple of minutes later from the permeate tank, too. The results from AOX analysis of the samples are shown in Table 6-2.

Table 6-2 Concentration of AOX in permeate after a CEB-sequence, compared to the AOX background level of the permeate, and in the final effluent.

Sample	AOX (mg/l)
Permeate outlet directly from MFU immediately after completion of CEB	0.98
Permeate tank immediately after completion of CEB	0.60
Typical level in permeate tank	0.57
Monitored level in final effluent (after the polishing stage)	0.12-0.21

From earlier measurements, a background level of AOX in the permeate was found to be 0.57 mg/l. Based on this, it seems that a CEB sequence will slightly increase the AOX concentration in the permeate from the relevant filter unit, but after being mixed in the permeate tank with permeate from the other filter units, the AOX level decreases again to the background level. Furthermore, it can be seen in Table 6-2 that AOX is reduced over the polishing stage. As described in Section 7.5.1, the monitored level of AOX of 0.12-0.21 mg/l in the final effluent is not expected to be environmentally critical, as no toxic effects are seen from the effluent (see Section 7.2) and none of the typical critical compounds in AOX (e.g. chloroform) are measured in the effluent.

## 7 Treatment performance for pharmaceuticals and other hazardous substances

During the test period from 01.06.2014 to 30.11.2015, the following analyses and tests have been carried out in relation to wastewater samples taken from the sampling points described in section 6.1:

- Pharmaceuticals
- Ecotoxicological tests
- Tests of estrogenic effects
- Metals
- Hazardous substances (organic)
- VOC
- Radioactivity
- Oxidation by-products
- Bacteria and viruses (Chapter 8)

This chapter presents analytical data and evaluation of the performance during treatment in MBR, ozonation and GAC.

### 7.1 Pharmaceuticals

An overview of the sampling points is presented in Figure 6-3. The wastewater sampling included 24-hours automatic flow proportional sampling of the influent (Point 1), the permeate after the MBR plant (Point 2) and after Line 1 (Point 4) and 2 (Point 6), respectively. After GAC in Line 1 (Point 3) and after ozone in Line 2 (Point 5), the sampling was carried out as spot sampling.

Altogether 122 different pharmaceuticals were analysed in 118 samples (see Table 7-1). A total number of 80 parameters (including four metabolites) were detected in quantities above the limits of detection (LOD) in the raw wastewater. The sampling frequency varied during the test period. Institut für Energi- und Umwelttechnik e. V. (IUTA) analysed 67 different pharmaceuticals and six contrast media. In addition, five metabolites and six other hazardous substances (corrosion inhibitors, fungicides and pesticides) were analysed. The analyses performed by Eurofins comprised 93 pharmaceuticals, of which 67 are different from those analysed by IUTA.

A total number of 509 (2013 data) environmentally relevant pharmaceuticals (without vitamins, proteins etc. according to the EMA guideline [32]) are being used at Herlev Hospital [33]. This highlights the fact that the pharmaceuticals analysed in this project only comprise a limited number of the total number of pharmaceuticals used at the hospital due to a general international lack of commercially accessible chemical analysing methods.

Table 7-1: Analyses for pharmaceuticals. Total number of samples analysed and analyses carried out by IUTA and Eurofins

	Number
Total samples analysed	118
Total number analysed parameters	122
Parameters >LOD in raw wastewater (Point 1)	80
IUTA analysed parameters	78
Eurofins analysed parameters	93

The treatment performance concerning pharmaceuticals is predominantly evaluated based on analyses carried out by IUTA (113 samples). In total, Eurofins analysed five samples.

### 7.1.1 Raw wastewater characteristics (Influent)

The measured influent quality – raw wastewater – is presented in Table 7-2. A total of 80 parameters were measured in concentrations higher than LOD (limit of detection). The measured pharmaceutical concentrations are compared to PNECs for freshwater ( $PNEC_{Freshwater}$ ). Numbers higher than  $PNEC_{Freshwater}$  are marked. For 26 substances, the average were higher than the  $PNEC_{Freshwater}$  and for five substances only the maximum concentration was higher than  $PNEC_{Freshwater}$ .

Substances analysed by Eurofins are marked with \* in Table 7-2. All the other substances were analysed by IUTA.

Table 7-2: Concentrations of pharmaceuticals higher than LOD in raw wastewater (influent). Flow proportional 24-hours samples from 17.07.2014 to 16.11.2015.

ng/l	Number	Average*	Min.	Max.	Std. dev.*	PNEC <sub>Fresh water</sub> [19]
Amidotrizoic Acid	14	31,000	85	120,000	42,000	
Amiloride	14	22	<10	83	25	1,700
Amlodipin*	2	43	34	51	12	1,000
Atenolol*	2	600	450	750	210	150,000
Atorvastatin	14	630	110	1,300	410	200
Azithromycin	13	890	250	1,900	550	90
Bendroflumetiazid*	2	58	53	62	6	
Bisoprolol	14	81	32	140	37	35,600
Capecitabine	14	800	14	2,300	580	200
Carbamazepine	14	390	110	1,000	250	500
Carvedilol*	2	25	11	38	19	290
Cefalexin	12	50	<10	420	120	50
Cefotaxime	14	390	<10	3,700	980	500,000
Cetirizine*	2	730	680	770	64	
Ciprofloxacin	14	13,500	1,800	27,000	8,600	89
Citalopram	13	300	120	620	140	8,000
Clarithromycin	14	2,700	100	7,800	2,500	60
Clindamycin	14	300	28	1,200	360	3,700
Clozapin*	2	200	130	260	92	180
Codein*	2	6,600	5,100	8,100	2,100	75,000
Cyclophosphamide	13	450	15	1,200	360	65,000
Cyklofosamid*	2	560	160	950	560	65,000
Diazepam*	2	11	<10	16	8	12,000
Diclofenac	14	650	300	1,100	250	100
Enalapril	14	820	<10	1,600	410	180,000
Erythromycindehydrato	13	1,000	63	5,200	1,300	200
Estriol*	2	1,100	630	1,500	620	7.5
Estron*	2	160	140	170	21	0.8
Estradiol*	2	42	32	52	14	0.1#
Fluvastatin*	2	9	<10	13	6	
Furosemid *	2	11,000	8,900	13,000	2,900	31,000
Gemfibrozil*	2	170	150	180	21	150
Hydrocortison	12	500	<10	1,700	460	100,000
Hydroklortiazid *	2	1,200	1,200	1,200	0	143,000
Ibuprofen	13	22,000	6,100	52,000	14,000	4,000
Ifosfamide	14	2,000	<10	7,600	2,700	162,000
Iohexol	14	400,000	160,000	760,000	170,000	
Iomeprol	14	2,900,000	150,000	5,000,000	1,400,000	1,000,000
Iopamidol	14	1,400,000	250	2,500,000	850,000	
Iopromid	10	1,100	<10	4,200	1,300	1,360,000
Ioversol	9	270	<10	1,000	310	
Isosorbidmononitrat*	2	210	160	260	71	

ng/l	Number	Average*	Min.	Max.	Std. dev.*	PNEC <sub>Fresh water</sub> [19]
Losartan	14	4,100	2,100	8,300	1,7000	245,000
Mefenamic acid	13	94	<10	340	110	4,000
Metoprolol	14	2,500	1,200	3,900	800	62,000
Metronidazole	13	8,500	<10	26,000	6,800	12,500
Mianserin*	2	21	10	31	15	640
Mirtazapine	13	82	<10	160	61	6,900
N4-Acetyl-Sulfadiazin	14	140	<10	780	230	20,000
N4-Acetyl-Sulfamerazin	14	14	<10	82	23	
N4-Acetyl-Sulfamethazin	14	15	<10	93	26	30,000
N4-Acetyl-Sulfamethoxazole	14	5,100	1,800	13,000	3,400	120
Naproxen*	2	2,400	2,200	2,500	210	6,400
Norfloxacin	14	10	<10	74	18	32
Ofloxacin	13	69	<10	330	95	100
Omeprazol*	2	10	<10	14	6	100,000
Oxazepam*	2	620	410	820	290	500
Paclitaxel	7	1,700	<10	12,000	4,500	740
Paracetamol	14	350,000	60,000	800,000	190,000	9,200
Phenazone	14	90	<10	560	150	800,000
Prednisolon	11	590	<10	3,600	1,100	230
Propranolol*	2	6,500	3,000	10,000	5,000	100
Ramipril*	2	21	20	22	1	100,000
Ranitidine	14	850	100	3,900	1,200	31,000
Ritalinic acid	14	610	130	1,400	430	77,000
Roxithromycin	14	830	<10	5,100	1,500	3,400
Sertralin*	2	46	40	52	8	0.52
Simvastatin	13	45	<10	310	87	21,000
Sulfadiazine	14	330	<10	1,800	630	20,000
Sulfadimethoxine	14	55	<10	700	190	
Sulfamethazine	14	36	<10	310	82	30,000
Sulfamethoxazole	14	5,300	2,500	16,000	3,900	120
Sulfapyridine	14	1,100	94	3,700	920	10,000
Tamoxifen	13	25	<10	150	46	400
Tetracyclin*	2	98	<10	190	130	10,000#
Tramadol	14	4,300	<10	9,500	2,300	100
Trimethoprim	14	2,100	1,000	5,900	1,400	62,000
Venlafaxine	14	480	250	1,100	250	100
Warfarin	14	21	<10	80	20	5.900
Zopiclone	14	100	<10	230	77	43

# Environmental Quality Standard (EQS) [14]; \* Substances analysed by Eurofins; \* Half of the LOD is used for calculations

15 pharmaceuticals have been selected as indicator substances (see Chapter 5). The measured concentrations of the indicator substances are illustrated in Figure 7-1.

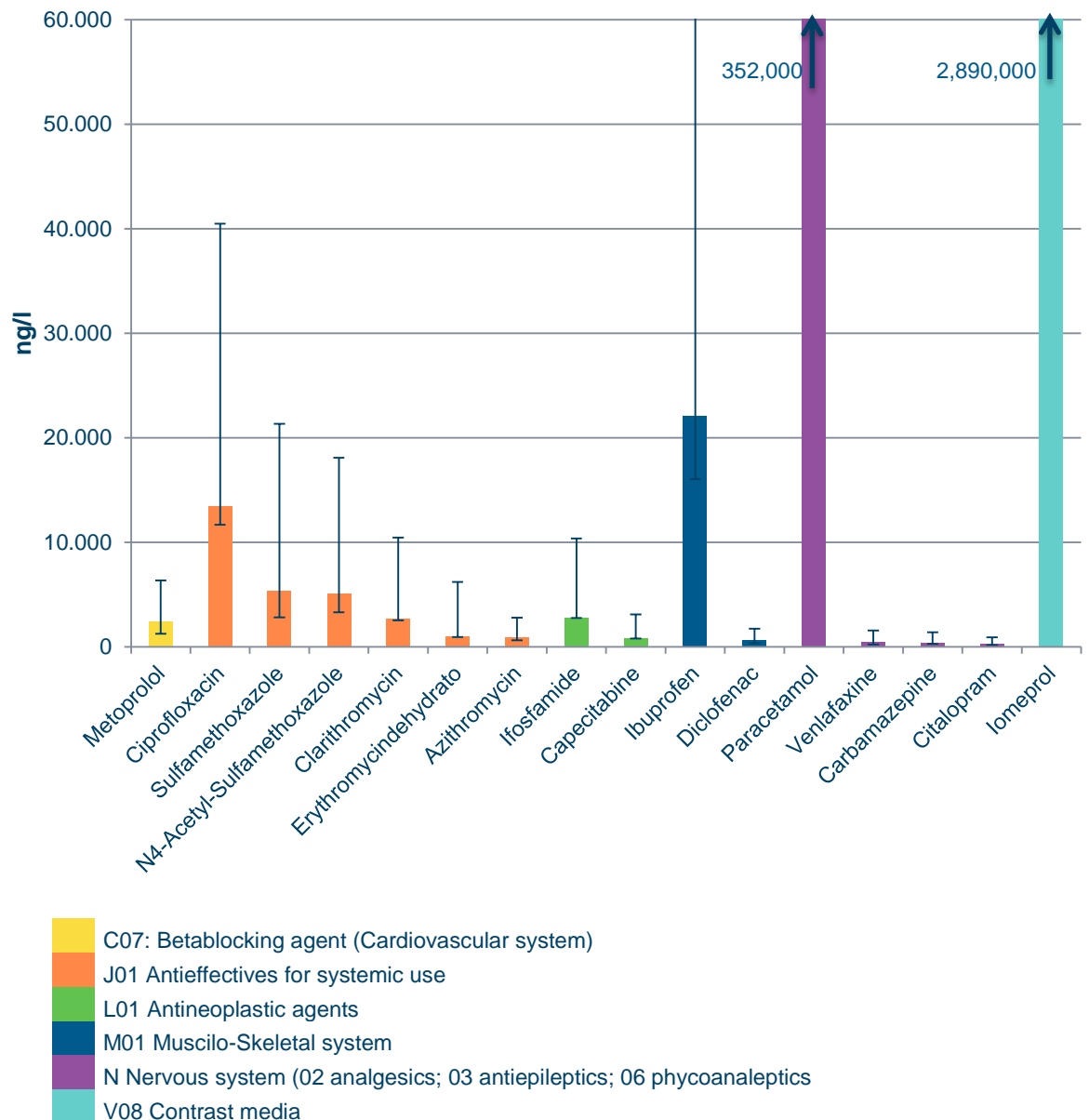


Figure 7-1: Concentration of indicator substances measured in the influent (raw wastewater). The dataset represents four sampling campaigns in May, June and November 2015.

The average influent concentrations of the indicator substances in four samples from May to November 2015 are presented in Figure 7-1. The metabolite N4-Acetyl-sulfamethoxazole, which originates from sulfamethoxazole, is also included. Contrast media (represented by iomeprol) is measured in the highest concentrations followed by painkiller paracetamol. The summarized concentrations of the different therapeutic groups measured in the raw wastewater (influent) are presented below in Figure 7-3.

### 7.1.2 Effluent quality

The analyses showed that the setup of Line 2 (MBR-Ozone-GAC) was the most efficient (see Section 7.1.3) and the plant was reconstructed by the end of the test period so that both lines were operated with the configuration MBR-Ozone-GAC. Therefore, only data from Line 2 are described in this section. Effluent data from the period from May to November 2015 are presented in Table 7-3, as this period represents the optimised setup, which corresponds to the

future normal operation of the WWTP. The table only includes pharmaceuticals with concentrations above limits of detection (LOD).

From Table 7-3 it appears that all measured concentrations in the effluent were below  $PNEC_{Freshwater}$ .

Table 7-3: Measured pharmaceuticals above LOD in effluent from Line 2. Flow proportional 24-hours samples from May, June and November 2015.

ng/l	N	Average	Min.	Max.	Std.dev.	$PNEC_{Fresh}$
Amidotrizoic Acid	4	6,983	230	12,000	5,213	
Azithromycin	4	7	<10	11	3	90
Ciprofloxacin	4	39	<10	66	25	89
Ifosfamide	4	14	<10	25	11	162,000
Iohexol	4	2,151	<10	4,900	2,032	
Iomeprol	4	21,008	30	45,000	19,704	1,000,000
Iopamidol	4	8,003	11	19,000	8,289	
Iopromid	4	62	<10	150	62	1,360,000
Ioversol	4	43	<10	150	72	
Losartan	4	7	<10	12	4	245,000
Ritalinic acid/Methylphenidat	4	16	<10	27	13	77,000
Sulfamethoxazole	4	42	<10	72	30	120

Six indicator substances were measured in concentrations higher than the LOD (see Figure 7-2). The indicator substances included three antibiotics (azithromycin, ciprofloxacin and sulfamethoxazole), one antineoplastic agent (ifosfamid), and one contrast media (iomeprol).



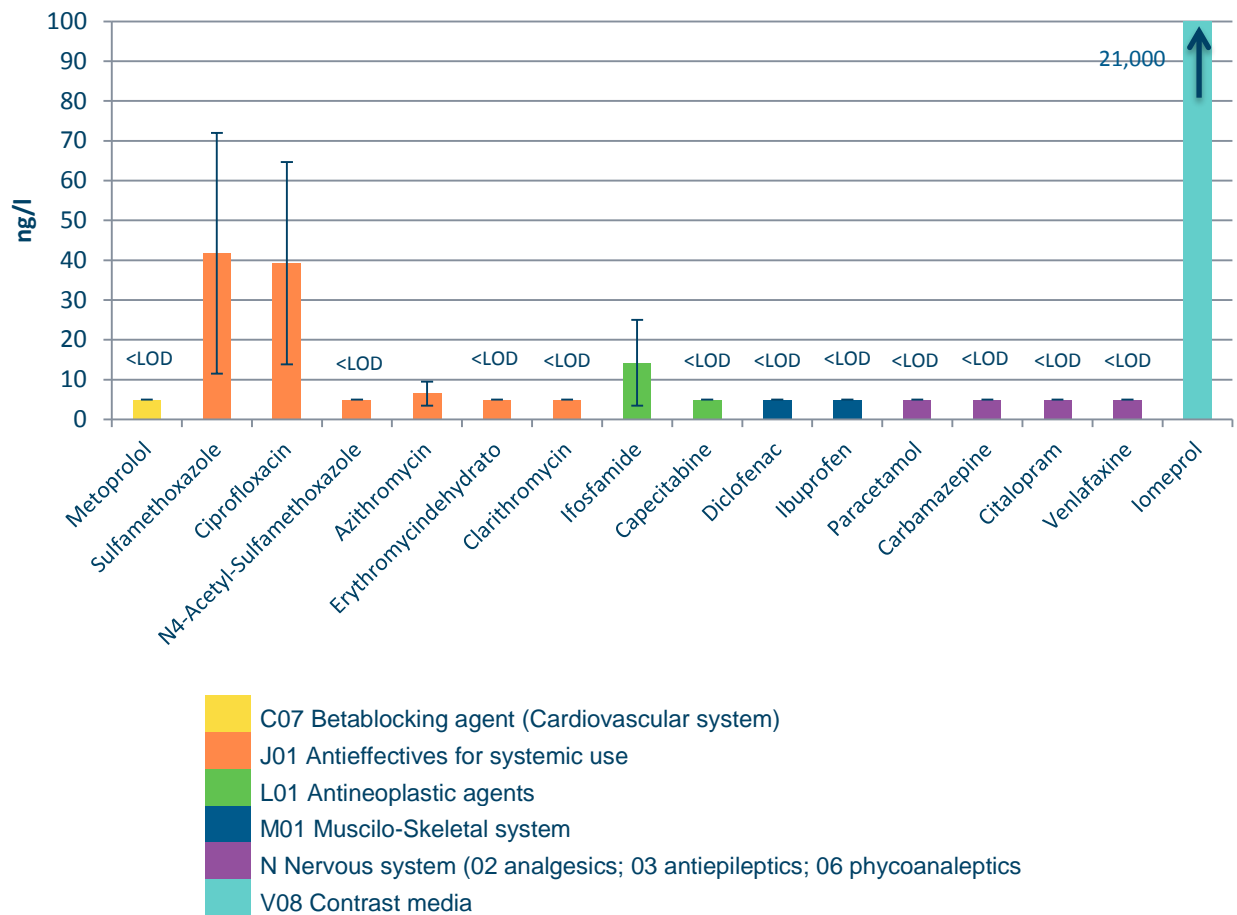


Figure 7-2: Concentration of indicator substances measured in effluent from Line 2. The dataset represents four sampling campaigns in May, June and November 2015.

### 7.1.3 Removal efficiency

The overall removal efficiency is illustrated by the sum of concentrations of measured pharmaceuticals in the influent, in the permeate after MBR, and in the effluent (see Figure 7-3). Pharmaceuticals measured below the limit of detection enter into calculations as half of LOD.

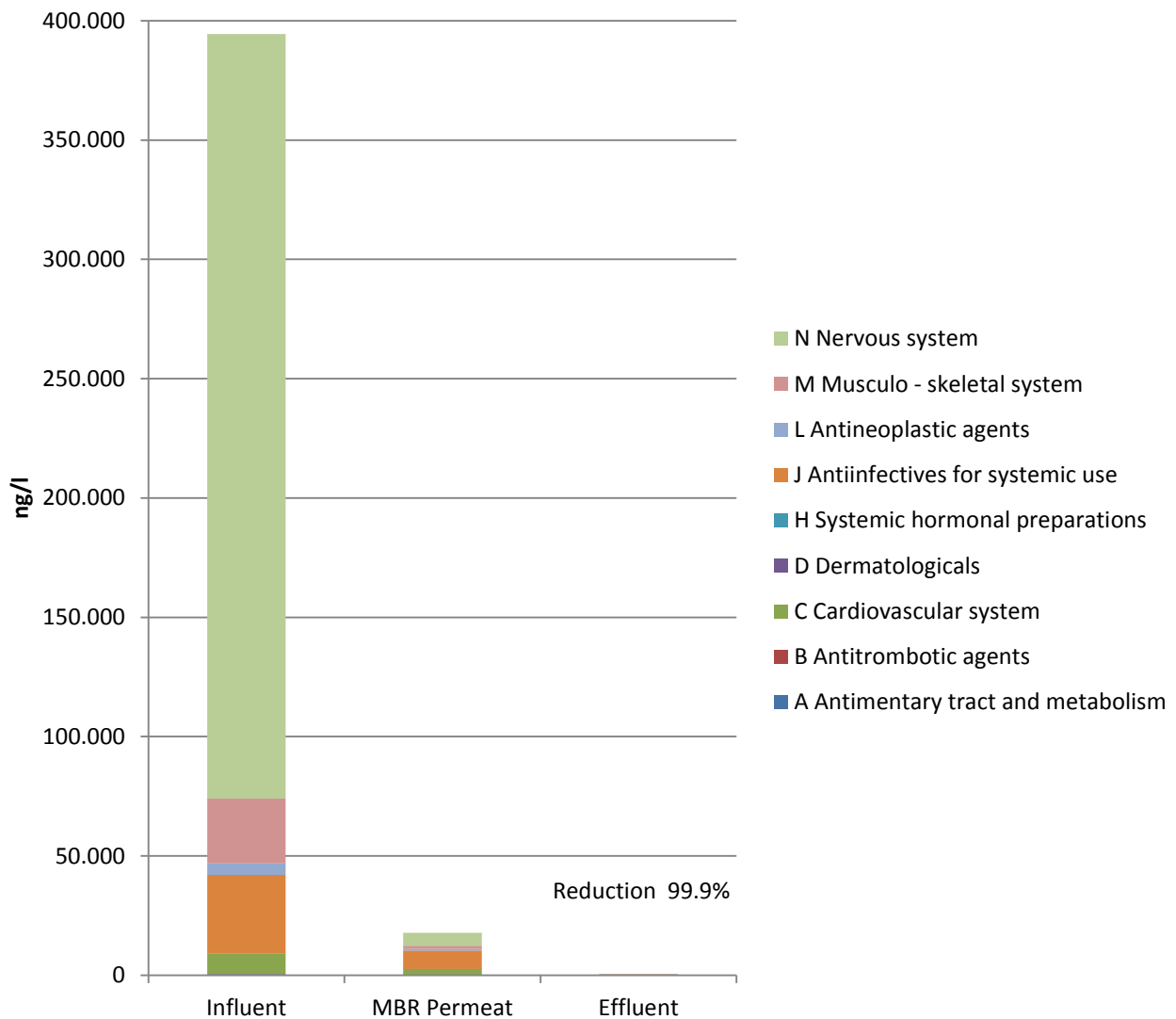


Figure 7-3: Total concentration of pharmaceuticals – without contrast media – in influent, after MBR (permeate) and effluent (Line 2). Data refers to sampling in May, June and November 2015.

Figure 7-3 shows that influent concentrations are dominated by ATC group N (Nervous system), including painkiller paracetamol (315,000 ng/l), ATC group M (Muscular-skeletal system), including ibuprofen (26,200 ng/l) and ATC group J (Antiinfectives), including sulfamethoxazole (8,050 ng/l). A likewise proportional picture can be observed after MBR in the permeate, where the Antiinfectives (dominated by sulfamethoxazole with 3,450 ng/l) show the highest amount followed by nervous system substances (dominated by tramadol with 3.350 ng/l).

Compared to the influent and without considering contrast media, 95% of the measured pharmaceuticals are reduced in the MBR and 99,9% are removed in the effluent after the final polishing (ozone and GAC). Although 95% of the pharmaceuticals are removed in the MBR, a number of substances are still above the  $PNEC_{Freshwater}$ . Table 7-4 shows that 21 substances are measured above  $PNEC_{Freshwater}$  in the raw wastewater and 14 substances are still above after MBR treatment (results from analysis by IUTA). In the effluent after polishing with ozone and GAC, no pharmaceuticals can be measured above  $PNEC_{Freshwater}$ .

Table 7-4 Pharmaceuticals measured above  $PNEC_{Freshwater}$  in the influent (raw wastewater), MBR permeate, and final effluent (Line 2). Measured concentrations (MEC) /  $PNEC_{Freshwater}$  ratio are shown in brackets with min – max ratios. Data refer to sampling from June 2014 to November 2015 for effluent and to sampling in May, June and November 2015 in MBR permeate and effluent. Analysis performed by IUTA.

Influent	MBR permeate	Effluent (Line 2)
Atorvastatin (0.6 - 6.5)	Azithromycin (2.3 - 8.1)	No pharmaceuticals exceeding $PNEC_{Freshwater}$
Azithromycin (2.8 - 21)	Carbamazepine (0.7 - 1.3)	
Capecitabine (0.1 - 11)	Cefalexin (LOD - 3.2)	
Carbamazepine (LOD - 2.0)	Ciprofloxacin (4.6 - 93)	
Cefalexin (LOD - 8.4)	Clarithromycin (2.2 - 10)	
Ciprofloxacin (20 - 303)	Diclofenac (4.2 - 11)	
Clarithromycin (1.7 - 130)	Erythromycin (0.3 - 1.1)	
Diclofenac ((3.0 - 11)	Iomeprol (LOD - 1.0)	
Erythromycin (0.3 - 26)	N4-Acetyl-Sulfamethoxazole (0.4 - 1.9)	
Ibuprofen (1.5 - 13)	Ofloxacin (LOD - 1.5)	
Iomeprol (0.2 - 5)	Sulfamethoxazole (11 - 64)	
Metonidazole (LOD - 2.1)	Tramadol (26 - 44)	
N4-Acetyl-Sulfamethoxazole (15 - 108)	Venlafaxine (5.2 - 7.7)	
Ofloxacin (LOD - 3.3)	Zopiclone (0.6 - 4.7)	
Paracetamol (6.5 - 87)		
Prednisolon (LOD - 16)		
Sulfamethoxazole (21 - 133)		
Sulfapyridin (0.1 - 3.7)		
Tramadol (LOD - 95)		
Venlafaxine (2.5 - 11)		
Zopiclone (LOD - 5.3)		

LOD: Limit of detection.

Due to the differences of concentration levels, the treatment efficiency of pharmaceuticals and contrast media is presented in a separate figure (Figure 7-4). A total of 900 kg iodinated contrast media is used for clinical purposes in Herlev Hospital per year (2012-2013 data). Around 82% of the consumed amounts can be measured in the raw wastewater/influent (yearly average 4,927,000 ng/l and 150,000 m<sup>3</sup> wastewater = 739 kg/y).

Table 7-4 shows that iomeprol was found at the highest concentration in the influent and iopamidol as the second highest concentration. For all the contrast media, the MBR reduced the concentration by 75%, and after the ozonation and GAC in Line 2, removal efficiency reached 99% compared to that of the influent concentration.

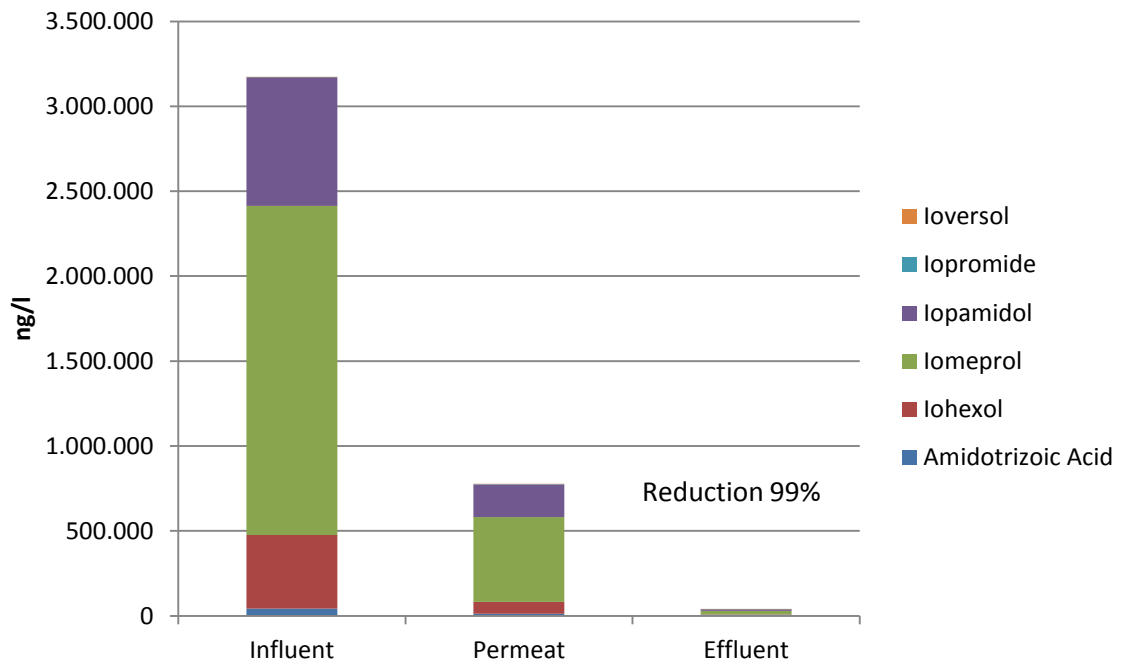


Figure 7-4: Total concentration of contrast media in influent, MBR permeate and effluent (Line 2). Data refer to sampling in May, June and November 2015.

Figure 7-5 shows the percentage of individual pharmaceuticals removed during MBR treatment. Substances below limits of detection as well as substances only measured once in influent or permeate during the four measuring campaigns have been left out.

For some pharmaceuticals, a negative reduction was observed. This is likely to be explained by conjugation and deconjugation. Conjugation is a process by which a compound is made water soluble by the binding of a functional group to the parent compound. Conjugation occurs in the human body, primarily in the liver, and is one of the ways in which the human body excretes pharmaceuticals. When a pharmaceutical is conjugated, it is not detected and identified as the parent compound in chemical analyses because of the functional group. During MBR treatment, the pharmaceutical can deconjugate, meaning that the functional group is removed. Without the functional group, the pharmaceutical is instead detected and identified via chemical analysis.

From Figure 7-5 it can also be observed that the painkillers ibuprofen and paracetamol presented removal rates near 100%, as expected. Paracetamol was reduced below LOD and ibuprofen presented a 98.8% reduction. As for the macrolides antibiotics (which are in the EU watch list), there is only a very limited removal concerning azithromycin (17%) and clarithromycin (45%), while erythromycin is removed by 85%. The painkiller diclofenac was measured in higher concentrations in the permeate from the MBR than in the influent, which is likely to be explained by the conjugation-deconjugation process described above.

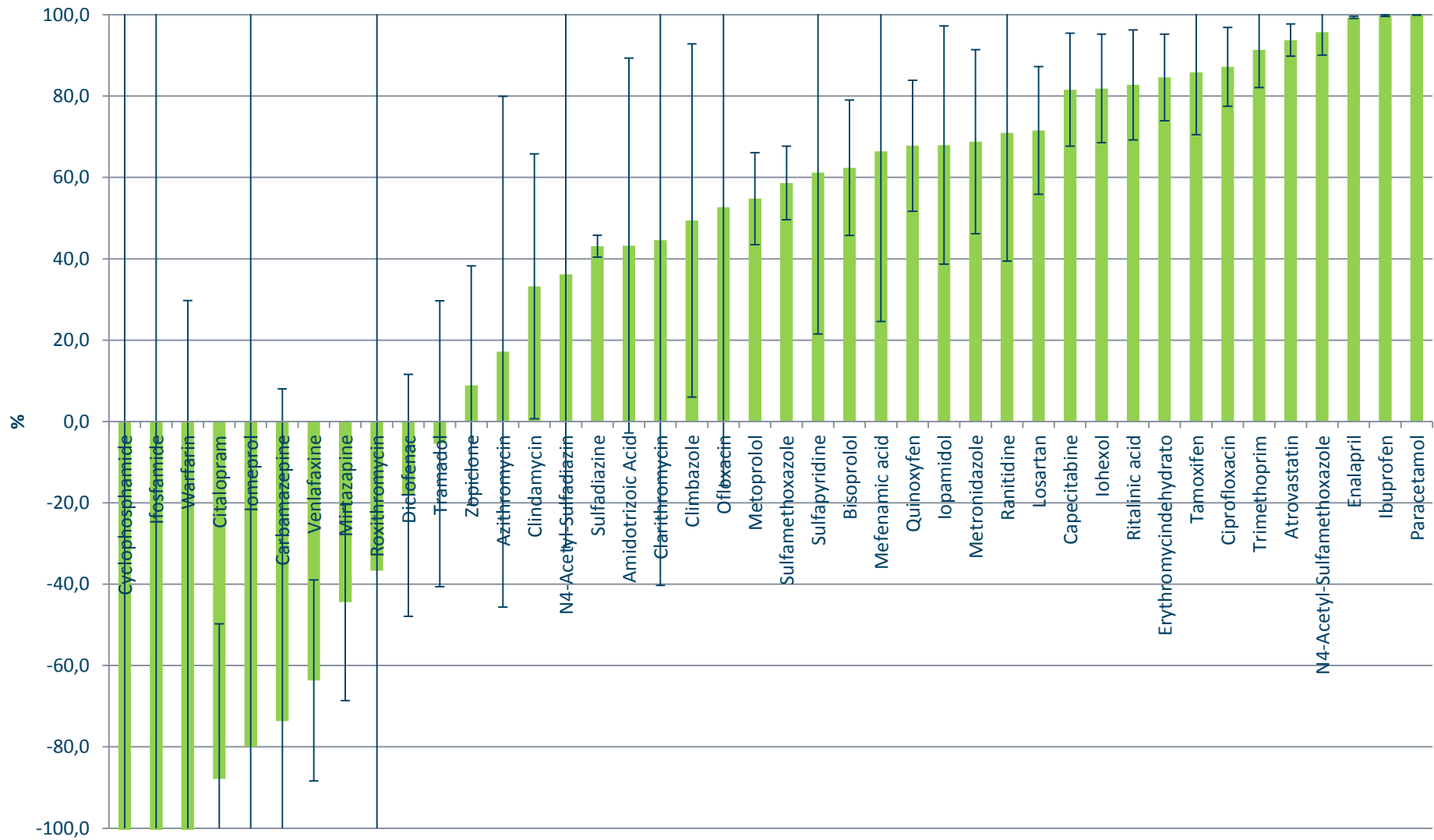


Figure 7-5: Removal of pharmaceuticals in MBR treatment. Data represents the average of samplings in May, June and November 2015. Standard deviations are marked on the columns.

### 7.1.4 Operational incidents

In Section 6.2, major operation disturbances are described. Figure 7-6 illustrates the effect on treatment performance from the incident with the cracked membrane disc in November 2014 (Incident 1). The concentration of ciprofloxacin increased in November and December 2014 and exceeded the  $PNEC_{Freshwater}$  values. In all the other samples from the effluent of Line 2, the ciprofloxacin concentration was below  $PNEC_{Freshwater}$ , which is 89 ng/l [19]. Incident 2 took place in June 2015 during exchange of the aeration diffusers and resulted in another cracked membrane. This time no exceeding of  $PNEC_{Freshwater}$  values were observed in the final effluent.

The change of GAC filters described in Section 6.2 are indicated in Figure 7-6. The changes of GAC filters were due to pressure build up in the first filter (Filter 1) in both Line 1 and Line 2. The change of filters did not impact the effluent quality significantly. The filters are installed in series and when one filter is changed, the new filter is placed at the end (Filter 3) of the line of filters. Because of this “serial” operation of the filters, it is not possible to calculate simple amounts of loads of bed volumes for the operation of GAC filters (see Section 6.2).

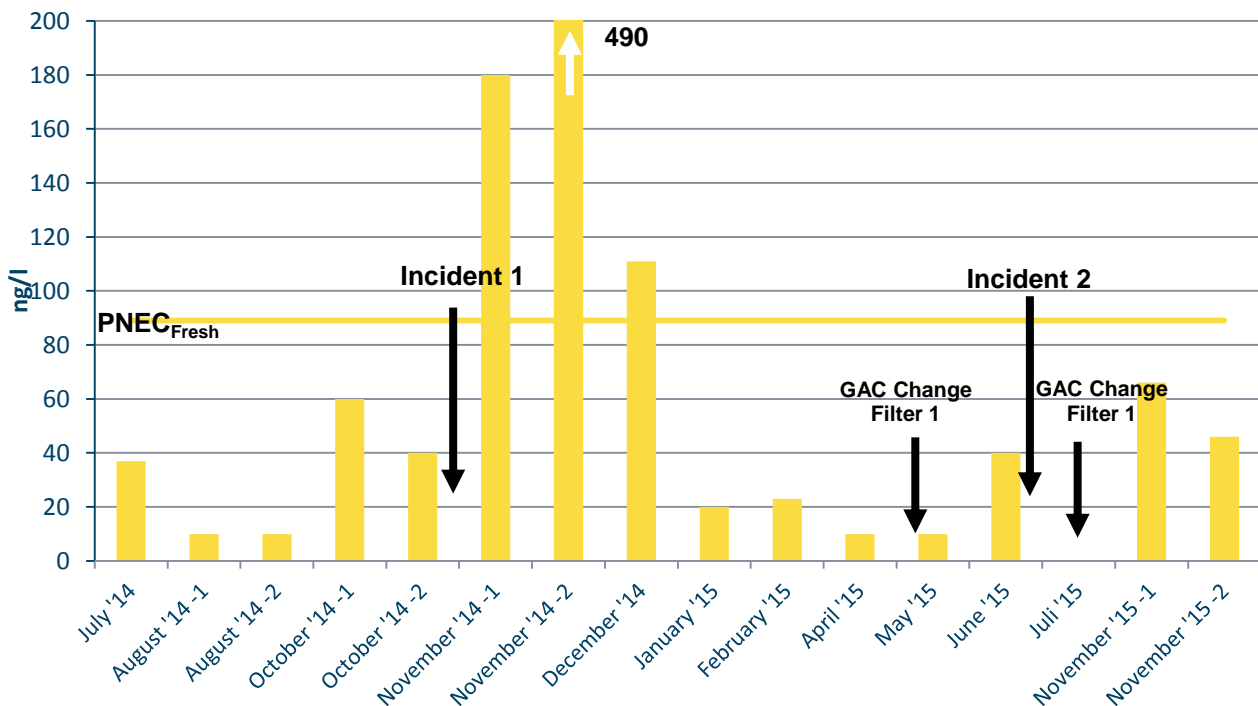


Figure 7-6: Development in ciprofloxacin concentration in the effluent (Line 2). The  $PNEC_{Freshwater}$  for ciprofloxacin is 89 ng/l. Operational incidents and change of GAC filters are indicated

Other pharmaceutical concentrations in the effluent were also influenced by Incident 1. The same pattern, but in a minor degree, was observed for sulfamethoxazole, and even smaller changes in concentration effluent were found for clarithromycin and diclofenac (see Figure 7-7). Additionally, smaller increases in effluent concentrations (below  $PNEC_{Freshwater}$ ) were observed for losartan (max 98 ng/l) and roxythromycin (max 82 ng/l) during Incident 1.

It can be concluded that ciprofloxacin and sulfamethoxazole were measured above the  $PNEC_{Freshwater}$  values during the MBR incidents (only during Incident 1). This indicates that the polishing sections with ozone and GAC can handle incidents with leaking MBR membranes to some extent and still keep the influents below the  $PNEC_{Freshwater}$  values. But these incidents also highlight the importance of preventing them with leaking membranes. The incidents result in biosludge pollution of the GAC filters, which again leads to a need for premature change of the

GAC filters. Surveillance and control of non-leaking membranes is crucial for an overall stable and cost efficient treatment performance.

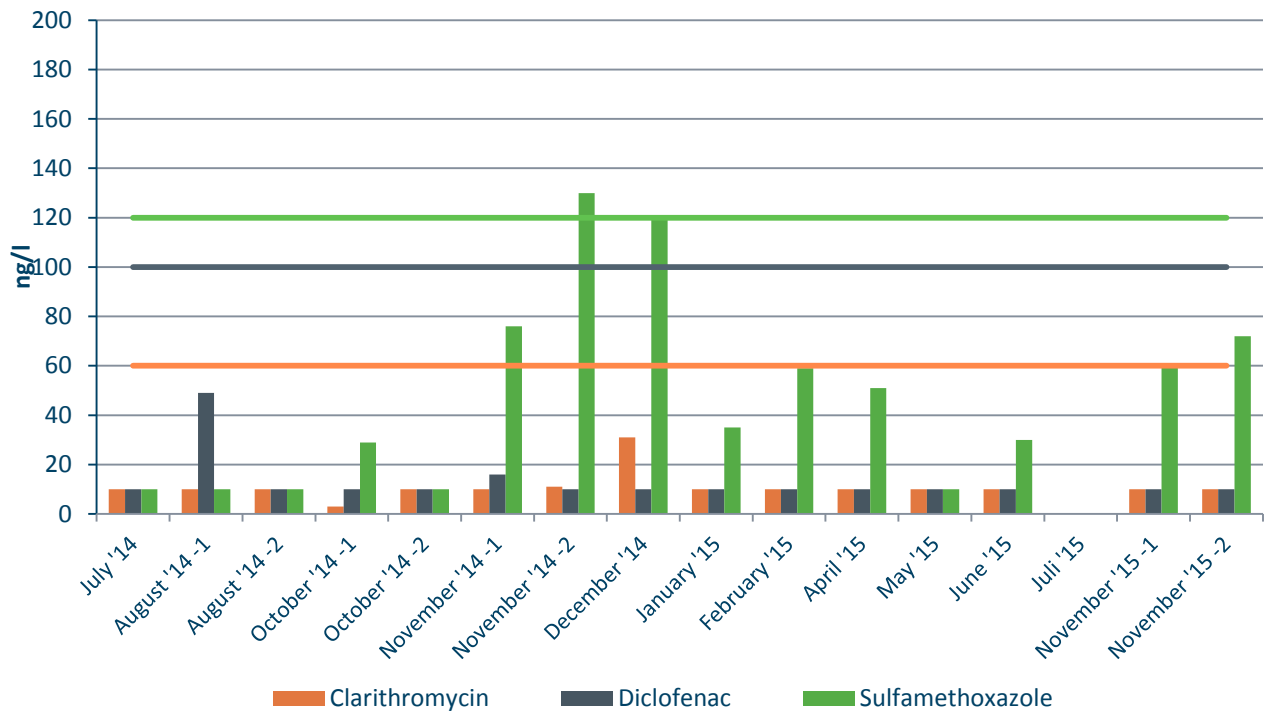


Figure 7-7: Development of clarithromycin, diclofenac and sulfamethoxazole concentrations in the effluent (Line 2). The PNEC<sub>Freshwater</sub> values for clarithromycin 60 ng/l, diclofenac 100 ng/l and sulfamethoxazole 120 ng/l. PNEC<sub>Freshwater</sub> values are shown as horizontal lines.

### 7.1.5 Effect of polishing line configuration and different ozone dosages

The two polishing lines of the WWTP were set up in different orders as described in Chapter 6 and Chapter 0. Both lines were feeded with the same MBR treated permeate. The question was whether Line 1 setup with MBR-GAC-Ozone or Line 2 with MBR-Ozone-GAC was the most efficient. Both lines had a final UV treatment as an extra microbiological barrier.

The overall technical concept is evaluated in Chapter 12. Differences in the efficiency of removal/transformation of pharmaceutical substances are evaluated in the following. In general, both lines showed efficient removal rates of the substances. However, it became clear during the last part of the test period that Line 2 with MBR-Ozone-GAC had a steadily better removal of the pharmaceuticals than Line 1.

Removal rates (in percentage) of the indicator pharmaceuticals for Line 1 and 2 are illustrated in Figure 7-8. Figure 7-8 shows the efficiency with the optimized ozone dosages, 2.5 and 3.4 mg ozone/mg DOC for Line 1 and 2, respectively. These rather high dosages were used because lower dosages (1.2 and 1.5 mg ozone/mg DOC, respectively) used at the beginning of the test period had proven to be less efficient (see description in Section 6.1).

The reduced efficiency from lower dosages can also be observed in the results of the low dosage tests, which were carried out in April and June 2014. Here, the dosages were reduced to 1.0 mg ozone/mg DOC in both lines. Results from the June low-dosage-test are illustrated in Figure 7-9. The figure shows that the removal rates from ozonation were lower in both lines. Especially for carbamazepine, citalopram and diclofenac in Line 2, it is obvious that the removal efficiency from the ozonation decreases, when the dosages are reduced from 3.4 (see Figure 7-8) to 1.0 mg ozone/mg DOC (see Figure 7-9).

From Figure 7-8 it can also be seen that carbamazepine, citalopram, diclofenac and venlafaxine are efficiently removed/transformed by GAC as well as ozone, depending on which treatment method comes first. Particularly for carbamazepine and diclofenac, similar observations have been made in other studies (see e.g. [37]). For contrast media iomeprol and antibiotic sulfamethoxazol it is observed that the combination of ozonation followed by GAC makes the GAC removal more efficient compared to GAC-Ozone. This can most likely be explained by the change of composition of the effluent organic matter (DOC) by the ozonation. The ozonation transforms large molecular-weight DOC into smaller compounds without reducing the DOC concentration. The ozonation of DOC reduces its aromaticity and hydrophobicity leading to a decreased adsorbability and in turn to less adsorption competition against the organic micropollutants (such as the pharmaceuticals) [38]. The reduced competition against micropollutants leads to more efficient removal rates and less GAC usage because of less “filling” of the GAC (for similar experiences see [37] and [38]).

The increased efficiency of Line 2 (MBR-Ozone-GAC) can also be illustrated by the measured concentrations of the most critical indicator substances (see Figure 7-10 and Figure 7-11). The figures show that the sulfamethoxazole and iomeprol (representative for all contrast media) are more efficiently removed in the final effluent in Line 2 than in Line 1.



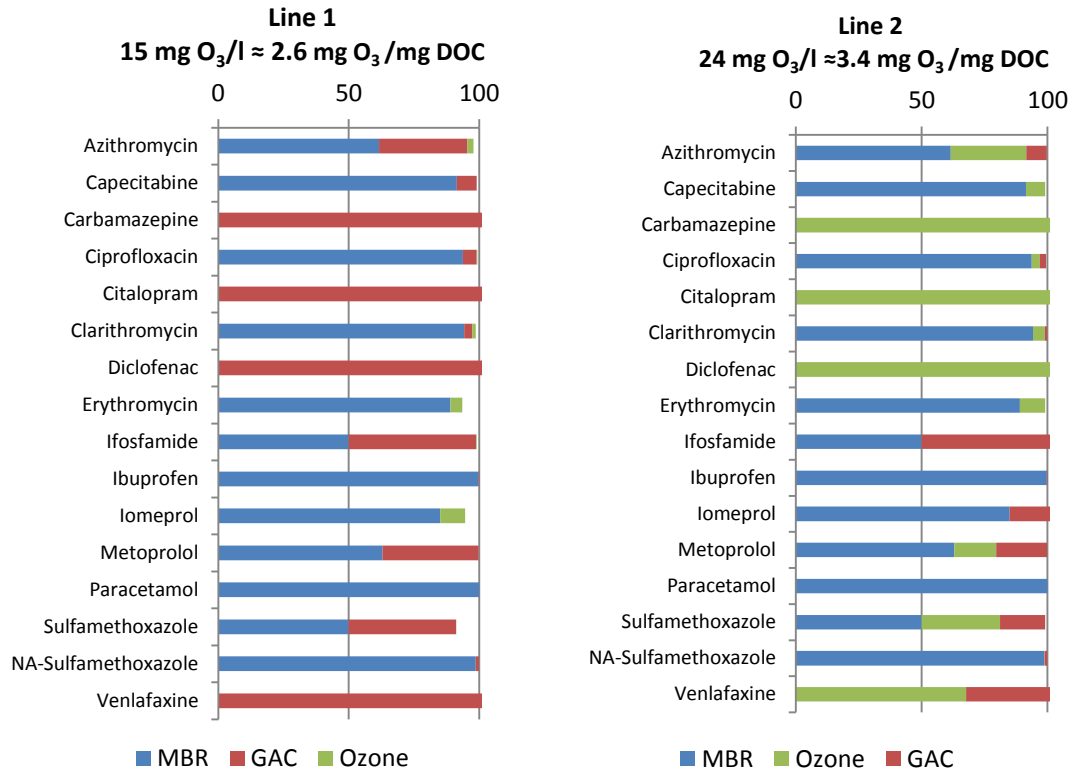


Figure 7-8: Indicator substances removed (%) by MBR, GAC and ozone for polishing Line 1 and 2 on 10.06.2015 with high ozone concentration.

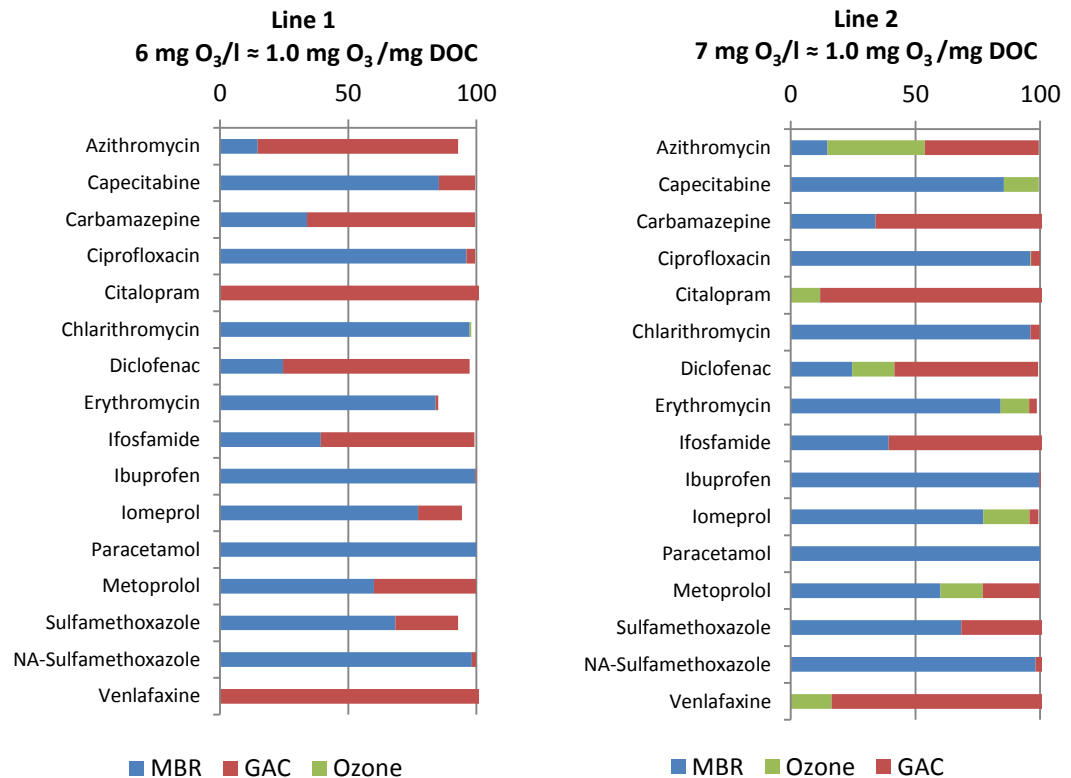


Figure 7-9: Indicator substances removed (%) by MBR, GAC and ozone for polishing Line 1 and 2 on 11.06.2015 with low ozone concentration.

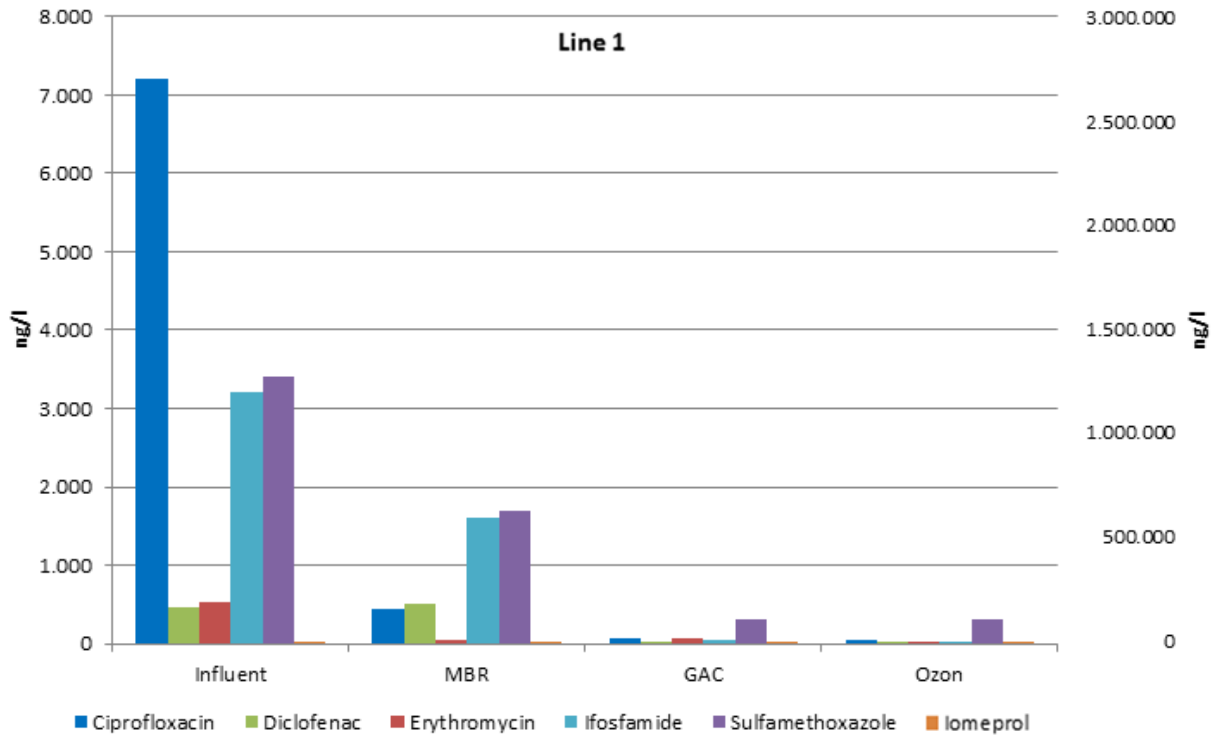


Figure 7-10: Concentrations of ciprofloxacin, diclofenac, erythromycin, ifosfamide, sulfamethoxazole and iomeprol in the influent, after MBR, after GAC and after ozonation in Line 1. Iomeprol refers to the secondary axes, while the other substances refer to the primary axes. All pharmaceuticals were analysed in samples taken on 10.06.2015.

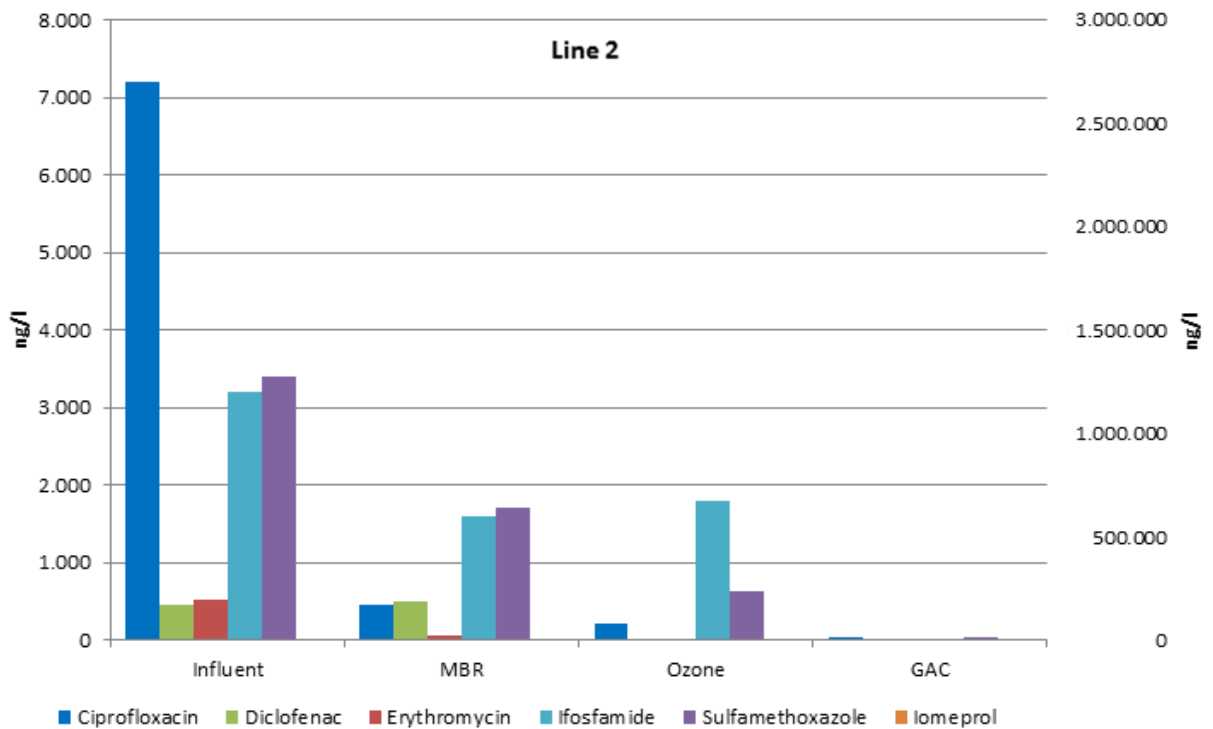


Figure 7-11: Concentrations of ciprofloxacin, diclofenac, erythromycin, ifosfamide, sulfamethoxazole and iomeprol in the influent, after MBR, after GAC and after ozonation in Line 2. Iomeprol refers to the secondary axes, while the other substances refer to the primary axes. All pharmaceuticals were analysed in samples taken on 10.06.2015.

## 7.2 Ecotoxicology

Ecotoxicological bioassays measure the effect of a range of concentrations of a sample on different endpoints in an organism and can be used as a supplement to analytical detection of pollutants in wastewater and effluent. The bioassays integrate the effects of all compounds in the sample and therefore give an indication of possible effects on aquatic organisms, even if the concentration of all measured pollutants is below the detection limit or the Environmental Quality Standards (EQS).

The toxicity of the influent and the effluent from Line 1 and 2 towards different aquatic organisms was tested in a range of bioassays (Table 7-5), carried out by the Research Institute for Ecosystem Analysis and Assessment (gaiac) [15]. The bioassays included a short-term reproduction test with water flea (*Daphnia magna*) and a fish embryo test with the zebrafish *Danio rerio* for evaluation of potential effects in the environment, as well as a micronucleus test for evaluation of genotoxicity using V79 cells from Chinese hamsters. Samples for testing were collected in sampling campaigns carried out in October 2014 and May 2015.

The bioassay with water flea, tested the effect of the undiluted influent and effluent from Line 1 and Line 2 on survival of the females, the cumulative offspring number, and the body length of females. The influent caused all females to die within two days and therefore no offspring was produced and no increase in body length occurred. Only slight effects were observed in the tests with the two effluent samples. In the second tests with effluent from Line 1, a 20% mortality was observed, while the other treatments with effluent showed no effects. The cumulative offspring number was slightly affected by the effluent from both Line 1 and Line 2 in the sampling campaign of May 2015, where the number of offspring was reduced by 23 and 32%, respectively. However, the difference compared to the controls was not statistically significant. The body length of females was not affected by the effluent from either of the polishing lines.

The bioassay with zebrafish embryos tested the mortality measured as either embryo coagulation, non-detachment of tail from yolk-sac, or lack of heartbeat. Embryos were exposed to wastewater concentrations of either 12.5%, 25%, 50%, or 100% for 96 hours. The undiluted influent sample from both sampling campaigns caused 100% mortality, and for the more diluted samples, toxicity decreased with decreasing concentration of wastewater. The effluent samples indicated no fish embryo toxicity at any of the dilution levels tested.

The micronucleus test was performed according to the ISO guideline 21427-2 (2004), which allows for the determination of genotoxicity of water and wastewater samples. The assay uses V79 cells from Chinese hamsters and measures the increase in the frequency of micronucleated cells, which are cells with damage to the chromosomes or the mitotic apparatus induced by water-soluble substances. The test showed no indications of genotoxic effects in the effluent from Line 1 and Line 2. For the influent sample, minor genotoxic effects cannot be excluded.

Table 7-5 Results of bioassays with samples of influent and effluent from the two polishing lines. Green color indicates no negative effects (<20 % inhibition). Yellow color indicates weak or moderate effects. Red color indicates strong effect (<80 % inhibition).

Bioassay	Endpoint	No of samples	Influent		Effluent Line 1		Effluent Line 2	
			SC1	SC3	SC1	SC3	SC1	SC3
Water flea ( <i>Daphnia magna</i> ) 8-9 days reproduction test	Survival	2						
	Cumulative offspring number		*	*		**		**
	Body length of females		*	*				
Zebrafish embryos ( <i>Danio rerio</i> ) 96 hours mortality test	Mortality measured as either embryo coagulation, non-detachment of tail from yolk-sac, or lack of heartbeat	2						
Micronucleus test (V79 cells from Chinese hamsters )	Genotoxic effects	2						

\* All females exposed to the influent samples were dead within two days, and therefore no offspring was produced and no increase in body length occurred.

\*\* The cumulative offspring number was slightly affected by the effluent from both Line 1 and Line 2 in the sampling campaign of May 2015, where the number of offspring was reduced by 23 and 32 %, respectively. However, the difference compared to the controls was not statistically significant.

SC: Sampling Campaign

In addition to the bioassays described above, an algal growth inhibition test with *Pseudokirchneriella subcapitata* was also performed on samples from the two above sampling campaigns and a third campaign carried out in January 2015. In all three sampling campaigns, a marked effect on the algae growth rate was observed for both influent and effluent samples, which was not expected considering the results from the bioassays with other organisms. Therefore, in addition to the influent and effluent samples, the second sampling campaign also included a sample of tap water and a grab sample from Kagså. These two samples were included in order to examine, whether clean water would also affect the algae growth rate. The effluent samples reduced the algae growth rate by up to 80%, and the same levels of inhibition were observed for the tap water and the sample Kagså. These results indicate that the observed inhibition of algae growth rate is more likely a result of the sample matrix than of the presence of toxic compounds.

Overall, the results of the different bioassays indicate that while the influent is highly toxic to all organisms tested and possibly genotoxic, the effluent is non-toxic and no significant differences are observed between the two treatment lines.

### 7.3 Estrogen activity

As a supplement to the analyses of hormones in the wastewater, a yeast estrogen screen assay with the yeast *Arxula adenivorans* (A-YES) was performed to determine estrogenic effects of the total matrix. The assay measures the response of a yeast cell to compounds with an estrogenic effect, and the result is expressed as 17 $\beta$ -estradiol equivalents (EEQ). The A-YES assay was made with samples of influent and effluent from polishing Line 1 and Line 2, collected on October 8, 2014 and on June 10, 2015.

The estrogenic activity in the influent samples was in the range 0.16 – 44 ng EEQ/L and in the effluent, the estrogenic effects were significantly reduced to the range of <0.076 – 1.1 ng EEQ/L. In Figure 7-12, the EEQ concentrations of the effluent samples are compared to the Danish EQS for 17 $\beta$ -estradiol [14], which is 0.1 ng/l for inland waters. Only the effluent sample from line 1 in June had concentrations slightly above the EQS, while all other samples were well below the EQS.

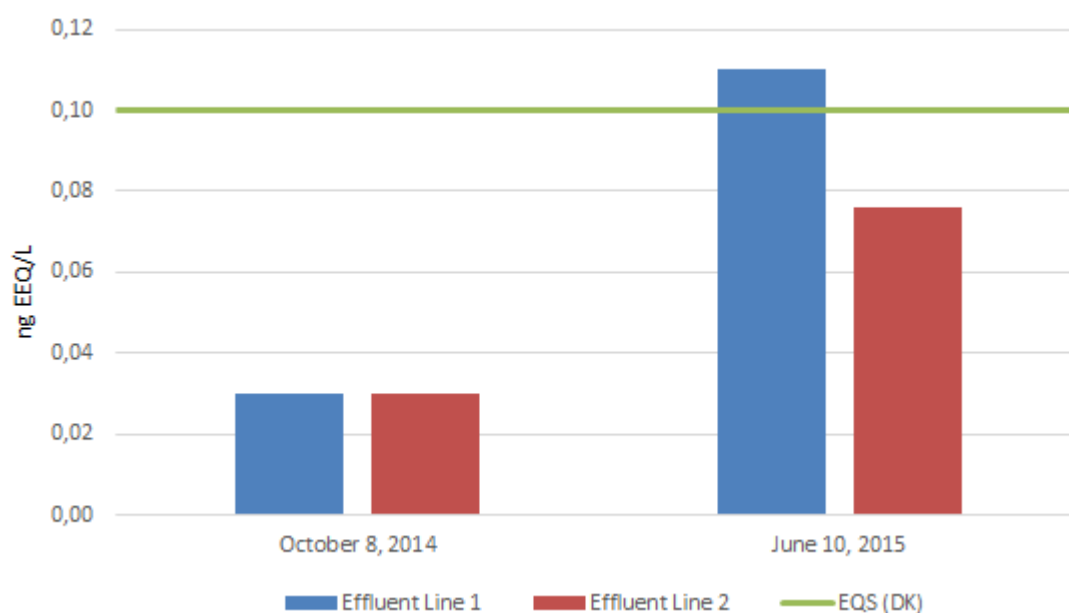


Figure 7-12. The concentration of 17 $\beta$ -estradiol equivalents in effluent of polishing Line 1 and 2 collected on October 8, 2014 and June 10, 2015. The concentrations were determined by use of a yeast estrogen screen assay with the yeast *Arxula adenivorans* (A-YES).

Although the EQS is only slightly exceeded in the sample from Line 1 from June 2015, the results of the A-YES assays still show that the current setup of polishing Line 2 is the most efficient for removal of estrogenic activity from the wastewater.

## 7.4 Other hazardous substances

### 7.4.1 Metals

Table 7-6 shows the measured concentrations of metals in the influent to the wastewater treatment plant, the effluent from polishing Line 2, and from two grab samples from the nearby watercourse Kagså. Concentrations exceeding EQS are highlighted.

Table 7-6 Concentration of metals ( $\mu\text{g/L}$ ) measured in the influent, the effluent from Line 2 at WWTP Herlev Hospital, and in the watercourse Kagså in 2015. The Environmental Quality Standards (EQS) for inland surface waters are shown for comparison.

$\mu\text{g/L}$	Influent	Effluent Line 2	Kagså watercourse	EQS <sup>1)</sup>
No of samples	1	3	2	
Sampling dates	11/5	11/5, 25/8, 27/8	25/8, 27/8	
Lead (Pb)	4.0	< 0.5-0.24	0.38-39	1.2 <sup>2)</sup>
Cadmium (Cd)	0.079	< 0.003-0.016	0.0089-0.39	$\leq 0.08-0.25^{3)4)5)}$
Chromium (Cr)	2.1	0.10-4.2	1.6-5.2	CrVI 3.4 <sup>3)</sup> / CrIII 4.9 <sup>3)</sup>
Copper (Cu)	110	< 1.0-2.5	10-89	1 (12) <sup>3)4)</sup>
Mercury (Hg)	< 0.05	< 0.05	< 0.05	0.05 <sup>3)4)</sup>
Nickel (Ni)	4.8	0.92-5.7	1.2-6.3	4.0 <sup>2)</sup>
Zink (Zn)	100	7.3-36	29-490	7.8 <sup>3)4)5)</sup>

1) The EQS expressed as an annual average value (AA-EQS). The EQS refers to Danish or European EQS given in Statutory Order No. 1070 of 09/09/2015.

2) The EQS applies to the bioavailable concentration.

3) EQS refers to the dissolved concentration.

4) In assessing the monitoring results or calculated concentrations in a water body, the natural background concentration must be taken into account. A maximum value is indicated in parentheses if available.

5) The EQS depends on the water hardness.

The variation in the measured concentrations of metals in the watercourse Kagså is due to the influence of precipitation in the sample from August 27, 2015. During heavy rain, Kagså receives urban run-off from many different areas as well as diluted wastewater from combined sewer overflows, which is likely to cause the higher concentrations of metals in Kagså on this date.

Generally, the measured concentrations of metals in the effluent from Polishing Line 2 were below the EQS for inland surface waters.

In one sample, the total concentration of nickel was above (5.7  $\mu\text{g/L}$ ) the EQS value of 4  $\mu\text{g/L}$ , but the EQS only applies to the bioavailable concentration of nickel. Further analysis and assessments must establish the bioavailable fraction of nickel in the effluent from Herlev Hospital WWTP and in Kagså in order to compare the effluent concentration with the EQS.

The concentration of zinc in the effluent from polishing Line 2 was approximately 4.5 times above the EQS (7,8  $\mu\text{g/L}$ ) in two samples from august 2015 (35  $\mu\text{g/L}$  and 36  $\mu\text{g/L}$ , respectively). But the concentrations were on the same level as the concentration measured in Kagså on August 25, 2015 (29  $\mu\text{g/L}$ ) and 13 times lower than the concentration measured in Kagså on August 27, 2015 (490  $\mu\text{g/L}$ ), where the watercourse was influenced by precipitation, run-off and possible wastewater. In effluents from municipal WWTPs and from rooftop run-off, the maximum concentrations of zinc have been measured to 110  $\mu\text{g/L}$  and 700  $\mu\text{g/L}$ , respectively.

It is necessary to take the dilution of the wastewater in the local water body into account too, when comparing with the EQS for inland surface waters.

## 7.4.2 Organic hazardous compounds

A number of other hazardous compounds were measured in the influent and effluent of WWTP Herlev Hospital in 2015.

EDTA (EthyleneDiamineTetraacetic Acid), LAS (Linear Alkylbenzene Sulfonate), DEHP (DiEthyl Hexyl Phthalate), nonylphenoles and bisphenol A are often present in hospital wastewater. The benzotriazoles and the pesticides diuron, quinoxifen, terbutryn and climbazole were all

analysed in the same analysis package as the pharmaceuticals. Benzotriazol has wide applications such as dishwasher detergents, corrosion inhibitor in cooling systems, and benzotriazole derivatives are also used as chemical precursors in medicine.

Table 7-7 shows the results of the analysis.

Table 7-7 Concentration of hazardous compounds measured in the influent and effluent from Line 2 at WWTP Herlev Hospital, compared to Environmental Quality Standards (EQS) or Predicted No Effect Concentrations (PNEC) for inland surface waters. Concentrations above EQS or PNEC are highlighted.

µg/L	Parameter	Influent	Effluent Line 2	EQS1/PNEC
1 sample (11.05.2015)	EDTA	< 3,000	< 100	100 <sup>2)</sup>
	LAS	1,600	< 100	54
	DEHP #	23	< 0.1	1.3
	Nonylphenol #	0.63	< 0.05	0.3
	Nonylphenol monoethoxylates	1.2	< 0.05	
	Nonylphenol diethoxylates	< 1	< 0.1	
	Sum af Nonylphenol+ethoxylates	1.8	-	
	Bisphenol A	3.1	0.01	0.1
4 samples (18.05, 10.06, 04.11, 16.11 2015)	1H-Benzotriazol	76-300	0.015-0.17	0.9 <sup>3)</sup>
	5-Methyl Benzotriazol	<0.01-1.1	<0.01	1.0 <sup>3)</sup>
	DimethylBenzotriazol	<0.01	<0.01	0.1 <sup>3)</sup>
	Climbazole	0.01-0.34	<0.01	0.52
	Diuron #	<0.01	<0.01	0.2
	Quinoxifen #	<0.01-0.034	<0.01	0.15
	Terbutryn #	<0.01	<0.01	0.065

1) The EQS expressed as an annual average value (AA-EQS). The EQS refers to Danish or European EQS set down in Statutory Order No. 1070 of 09/09/2015 [14]

2) Danish EPA EQS from 2002 published in [5]

3) Predicted No Effect Concentration (PNEC) [19]

# Priority substances under the Water Framework Directive [14]

All of the measured concentrations of hazardous compounds were below the Environmental Quality Standards (EQS) and Predicted No Effect Concentrations (PNEC).

### 7.4.3 VOC

Grab samples were collected on October 14, 2014 from:

- Permeate (sampling point 2, see Figure 6-5)
- After Ozone (sampling point 5)
- Effluent from Line 2 (sampling point 6)

The samples were analysed for Volatile Organic Compounds (VOC). No Volatile Organic Compounds (VOC) were detected above the detection limit (<0.5 mg/L).

### 7.4.4 Radioactivity

Table 7-8 shows the results of the analysis of gamma emitting isotopes in influent, permeate (after MBR treatment) and effluent from WWTP Herlev. DTU Nutech (Centre for Nuclear

Technologies at Technical University of Denmark) conducted the analysis with gamma spectrometric equipment and lead shielded germanium detectors calibrated with a certified standard. The analysis were conducted with sample volumes of one litre and measuring times of one hour.

Only I-131 and Cr-51 were detected in the influent, permeate and effluent. Naturally occurring radon (Ra-226) was detected in very low concentrations. No other gamma emitting isotopes<sup>1</sup> were detected by the analysis in the two sampling rounds.

Before the sampling on April 24, three patients each received a dose of 3,7 GBq I-131 in the morning at Herlev Hospital, which resulted in the higher concentrations of I-131 in the samples from April 24 and 25 compared to the concentrations measured in the samples from May 13.

Table 7-8 Concentrations of gamma radioactive isotopes in the influent, permeate (after MBR treatment) and the effluent from Line 2.

Bq/L	April 24 and 25, 2015			May 13, 2015	
	I-131	Cr-51	Ra-226	I-131	Cr-51
Influent	48,000	400	< 500	12	7
Permeate	3,300	< 40	20	150	20
Effluent Line 2	1,900	40	< 50	120	< 6

According to the Danish Statutory Order 954 of 23<sup>rd</sup> of October 2000, liquid radioactive waste can be discharged to sewer, if the radioactive concentration is below 100,000 Bq/L. The sum of the measured radioactive isotopes in both influent, permeate and effluent was below this limit.

Because of the limited sampling (grab samples) and measuring time (one hour), it is possible that other isotopes can be present in the wastewater. However, the other isotopes used at Herlev Hospital have a shorter half-life (110 min – 2.8 days) than I-131 (8 days) and Cr-51 (28 days) and the hospital uses them in smaller doses. Therefore, they are not likely to cause exceeding of the discharge limit.

With the coming implementation of Council Directive 2013/59/Euratom of 5 December 2013 [9], the Danish Health Authorities may reduce the limits for discharge of radioactive substances to sewer in the future. In some member states in the EU, the discharge limits for radioactive substances are much lower than the current limit in Denmark. In Germany, all facilities are required to have holding tanks installed and discharges from facilities must remain below a limit of 5 Bq/L at the point of discharge into the public wastewater network [10].

Because of the uncertainties regarding the future discharge requirements (also regarding direct discharge of the wastewater to the stream Kagså) along with the challenges of handling the radioactive sludge from WWTP Herlev Hospital (see Section 9.1.1), Herlev Hospital is now working on a delay and decay solution (holding tanks) for the specific wastewater stream from the Oncology Department containing I-131.

<sup>1</sup> Besides I-131 and Cr-51, Herlev Hospital also uses the gamma emitting isotopes: Tc-99m, F-18, I-123, In-111 and Sm-153 for nuclear medicine therapy, examination and research.



## 7.5 Ozonation by-products

### 7.5.1 Bromate

During ozonation, chloride and bromide might be transformed to hypochlorite and hypobromite, and be further oxidized to chlorate and bromate. Bromate is the by-product that causes the most concern due to its potential carcinogenicity and it is therefore highly undesirable. A provisional WHO guideline value for drinking water has been set at 10 µg/l [30].

A measuring campaign of bromide-bromate was carried out in June 2015. Bromide-bromate was measured in the permeate as well as after ozonation and GAC in both Line 1 and Line 2. No bromate above the detection limit (< 2 µg/l) was measured before or after ozonation or in the final effluent. Bromide concentrations varied between 250 and 270 µg/l.

AOX (absorbable organic halogens) was measured in the same campaign and the concentration was 0.57 mg/l in the permeate, which is close to the earlier measured concentration in the raw wastewater from Herlev Hospital of 0.47 mg/l measured in February 2013. This is also within the range of other AOX reported in wastewater from German hospitals (0.13-0.94 mg/l) [31]. It is not clear what the primary sources for the elevated levels of AOX detected in hospital wastewater is but chlorinated disinfection agents and ionidated contrast media may contribute [31]. The latter could explain the AOX concentrations in the permeate, as the total concentration and ionidated contrast media are measured to around 0.5 mg/l in the permeate (see Section 7.1).

AOX in the final effluent from Line 2 (MBR-Ozone-GAC) was measured to 0.12-0.21 mg/l. This level is difficult to explain, as the measured chlorinated compounds, which could be expected to be part of AOX, are under the detection limits (e.g. chloroform) and the total amount of ionidated contrast media is below 0.06 mg/l in the effluent of Line 2. Conclusively, the measured AOX in the effluent is not expected to be environmentally critical, as no toxic effects are seen from the effluent (see Section 7.2) and none of the typical critical compounds in AOX (e.g. chloroform) are measured in the effluent. More studies are needed, if the composition of the AOX in the final effluent should be clarified.

### 7.5.2 NDMA

N-nitrosodimethylamine (NDMA) can be formed during ozonation in wastewater from effluent organic matter. WHO has issued a guideline value for drinking water of 100 ng/l for NDMA [40].

Effluent from Line 2 was analyzed for NDMA and seven other nitro-compounds (NDEA, NDPA, NDPA, NDBA, NMEA, NMOR, NPIP, NPYR). NDMA was measured below the limit of detection (LOD: <8 ng/l). All other nitro-compounds were also measured below the LOD.



## 8 Treatment performance in relation to bacteria and virus

### 8.1.1 Concentrations of *E. coli*, enterococci and norovirus

Every week from November 2014 to November 2015, concentrations of *E. coli* were measured in the permeate (Sampling point, see Figure 6-5) by Eurofins using Colilert®, as a control to ensure non-leaking membrane filtration. *E. coli* were only detected in two weeks as a result of the incident with the cracked membrane discs in November 2014 (see section 6.2). In the rest of the test period, no *E. coli* were detected in the permeate or effluent.

Concurrently, measuring for *E. coli*, enterococci and norovirus in both influent and effluent was carried out in a 10 week measuring campaign from October 2014 to January 2015. *E. coli* and enterococci were measured by DHI using Colilert® and Enterolert® [11]. Norovirus was measured by DTU Food using qPCR [12].

Table 8-1 shows the detected concentrations in raw wastewater (Influent), the insufficiently treated effluent during the incident with leakage due to the cracked MBR membrane in November 2014 (Effluent, Line 1 and 2), and concentrations in the effluent under normal operation (Effluent, Line 1 and 2).

Table 8-1 Measured concentrations of *E. coli*, enterococci and norovirus during 10 weeks measuring campaign. Minimum, maximum and average values are shown. Analysis for *E. coli* and enterococci were carried out by DHI and for norovirus by National Food Institute, DTU DK.

	Measuring period	N	<i>E. coli</i>	<i>Enterococci</i>	<i>Norovirus</i>
			MPN/100 ML	MPN/100 ML	Genome copies/L
Raw wastewater (Influent)	27.10.14 - 12.01.15	18	3.1·10 <sup>6</sup> - 1.0·10 <sup>7</sup> 5.5·10 <sup>6</sup>	6.8·10 <sup>6</sup> - 1.5·10 <sup>7</sup> 9.5·10 <sup>6</sup>	211 - 6.2·10 <sup>5</sup> 1.7·10 <sup>5</sup>
Effluent during incident with reduced treatment efficiency due to cracked MBR membrane	10.11.14 - 01.12.14	8	< 1 - 3 0.75*	< 1 - 8 2.8*	< 26** - 861 170*
Fully treated effluent during normal operation	15.12.14 - 12-01.15	3	< 1	< 1	< 26** - < 26** < 26**

\* Half of Limit of detection (LOD) is used for calculation

\*\* Lowest detected concentration is used as LOD

Table 8-1 shows that low concentrations of *E. coli*, enterococci and norovirus were present in the effluent during the malfunctioning MBR membrane in November 2014. Under normal operation, these potential pathogens are not present in the effluent.

The measured concentrations from the 10 weeks-campaign shown in Table 8-1 are used to estimate health risks in three different discharge situations/scenarios with direct discharge to the local stream (Kagså) and bathing area (Lodsparken). The three situations/scenarios are:

1. Untreated raw wastewater is discharged
2. Insufficiently treated wastewater with an effluent quality corresponding the quality during the incident with the cracked membrane in November 2014
3. Fully treated waste under normal operation

The health risk assessments are described in Chapter 13.

### 8.1.2 Antibiotic resistant bacteria

Selected antibiotic resistant bacteria were analyzed as part of the 10-weeks monitoring campaign (See Section 8.1.1). The resistant bacteria were analyzed by DHI and Clinical Microbial Department (KMA) at Herlev Hospital using modified Colilert® and Enterolert® [11]. The measuring programme included analysis for cefotaxime (cephalosporin), ciprofloxacin (fluoroquinolone) and meropenem (carbapenem) resistant *E. coli* as well as vancomycin resistant enterococci. The detected meropenem resistant *E. coli* could not be fully confirmed as *E. coli* and therefore these results are not reported.

Table 8-2 shows the detected concentrations in raw wastewater (Influent), the insufficiently treated effluent during the incident with leakage due to the cracked MBR membrane in November 2014 (Effluent, Line 1 and 2), and concentrations in the effluent under normal operation (Effluent, Line 1 and 2).

Table 8-2 Measured concentrations of antibiotic resistant *E. coli* and enterococci during 10 weeks measuring campaign. Minimum, maximum and average values are shown. Analysis were carried out by DHI and KMA, Herlev Hospital

	Measuring period	N	Cefotaxime res. <i>E. coli</i>	Ciprofloxacin res. <i>E. coli</i>	Vancomycin res. enterococci
			MPN/100 ML	MPN/100 ML	MPN/100 ML
Raw wastewater (Influent)	27.10.14 - 12.01.15	18	1.0·10 <sup>5</sup> - 9.8·10 <sup>5</sup> 4.4·10 <sup>5</sup>	4.6·10 <sup>5</sup> - 1.5·10 <sup>7</sup> 7.1·10 <sup>5</sup>	1.5·10 <sup>5</sup> - 1.1·10 <sup>6</sup> 4.8·10 <sup>5</sup>
Effluent during incident with reduced treatment efficiency due to cracked MBR membrane	10.11.14 - 01.12.14	8	< 1 - 1 0.6*	< 1 - 1 0.6*	< 1 - 3 0.9*
Fully treated effluent during normal operation	15.12.14 - 12-01.15	3	< 1	< 1	< 1

\* Half of Limit of detection (LOD) is used for calculation

Table 8-2 points out that resistant *E. coli* and enterococci may appear in low concentrations during an incident with a cracked membrane like the one in November 2014. Under normal operation, no resistant *E. coli* and enterococci will be present in permeate or final effluent.

### 8.1.3 Effect of UV-treatment on bacteria

As described in Section 6.1, each of the polishing lines uses UV as the final treatment of the effluent. Each line has a 220 W UV lamp installed. Because of the relatively low UV-transmission rates of the polished wastewater in Herlev (typically 70%), the minimum applied dose is between 6 and 10 mJ/cm<sup>2</sup>. This is a low dose compared to the commonly applied doses of around 40 mJ/cm<sup>2</sup> in water works etc. Originally, 40 mJ/cm<sup>2</sup> was also the design goal for the Herlev installation.

Although the minimum applied UV-dose was rather low, the effect on heterotrophic plate counts (22 °C and 37 °C) was high (see Section 8.1.2). Compared to the Danish Drinking Water Act [22], the plate counts were almost as low as the requirement for water quality at consumer tap (only a slight exceeding at 37 °C plate count).

Table 8-3 Bacteria measured before and after UV treatment in Line 2. Sampling dates: 25.08.2015, 27.08.2015 and 28.09.2015.

Parameter	Unit	Before UV (N = 3)	After UV (N = 2)	DK Drinking Water Act [22] (user's tap)
Coliforms 37°C	MPN/100 ml	<1 - 1	<1	N,m,
<i>Escherichia coli</i>	MPN/100 ml	<1	<1	N,m,
Culturable bacteria 22 °C	CFU/ml	>3000	16-27	200
Culturable bacteria 37°C	CFU /ml	>3000	15-48	20

N.m.: Not measurable

As pointed out in Section 8.1.1, no fecal indicators (*E. coli*) can be measured after the 0.2 µm MBR filtration under normal operation. The high heterotrophic plate counts (> 3,000 CFU/ml) in the water before the UV are therefore assumed to be regrowth of bacteria in the polishing lines after the membrane filtration. And therefore, the high plate counts before the UV are not assumed to pose any critical pathogen health risks – and for now, the UV treatment is not considered as a critical treatment step.



## 9 Disposal of screenings, sludge and spent GAC

Wastewater treatment at the Herlev Hospital WWTP generates three types of solid residues:

- Screening material retained at the pretreatment screens.
- Surplus sludge from the biological stage (the sludge is dried to approximately 70% prior to disposal).
- Spent granular activated carbon from the activated carbon columns of the two polishing lines.

The final disposal method for all three types of residue is incineration.

Screenings and surplus sludge end up in big bags that are stored at the WWTP site until it is picked up by truck and transported to a nearby incineration plant (Vestforbrænding), where it is incinerated at a temperature between 850 and 1,200 °C.

Spent GAC is picked up by a vacuum tanker directly from the columns. The spent GAC is transported to the same incineration plant as the screenings and surplus sludge.

Table 9-1 shows the quantities of residues generated during a four month period (August to November 2015), in which focus was on stable operation after running-in of the whole plant.

Table 9-1 Quantities of solid residues from wastewater treatment at Herlev Hospital WWTP for the period August to November 2015.

Type of solid residues	Quantity for August to November 2015 (ton)	Number of pick ups
Screenings	5.6	4?
Dried surplus sludge	24.3	4
Spent GAC	1.0*	NA*

\* In reality there was no disposal of spent GAC in the period August to November 2015. The shown value is the estimated average consumption of GAC for a 4 month period.

### 9.1.1 Radioactivity

On May 13, 2015, gamma emitting isotopes were determined in sludge from Herlev Hospital WWTP. Concentrations of 600,000 Bq/kg I-131 and 8,000 Bq/kg Cr-51 were detected in the sludge. No other gamma emitting isotopes were detected during the one hour measuring with gamma spectrometric equipment and lead shielded germanium detectors performed by DTU Nutech.

In addition to this, the National Institute of Radiation Protection performed measurements of the ionising radiation inside Herlev WWTP on June 22, 2015. Around the filling and storage of bigbags with dried sludge, the ionising radiation was measured to:

- 30-35 µSv/hour on the surface of bigbags (filling)
- 19-50 µSv/hour on the surface of bigbags (storage)

The Danish Statutory Order 954 of 23<sup>rd</sup> of October 2000 [20] describes possible ways of disposal of radioactive solid waste, depending on the radioactivity of the waste. In order to send the sludge to incineration, the radioactivity for each big-bag must be less than:

- 5,000,000 Bq/bigbag for radionuclide group 2 (such as I-131)
- 50,000,000 Bq/bigbag for radionuclide group 3 (such as In-111)
- 500,000,000 Bq/bigbag for radionuclide group 4 (such as Cr-51)
- 5  $\mu$ Sv/hour on the surface of bigbag

Otherwise, the sludge needs special treatment or decay.

Each bigbag with dried sludge at Herlev WWTP contains app. 800 kg of sludge, corresponding to app. 480,000,000 Bq I-131 per bigbag if the concentration of the grab samples are representative for the whole bigbag. Furthermore, the radiation at the surface of the bigbags exceeded the limit of 5  $\mu$ Sv/hour.

In order to avoid storing of the sludge on site for decay because of the high concentrations of I-131 in the sludge, Herlev Hospital is working on a delay and decay solution (holding tanks) for the specific wastewater stream from the Oncology Department containing I-131.



## 10 Air treatment

Wastewater and sludge treatment may cause microorganisms, such as pathogenic bacteria and viruses in the wastewater, to be aerosolized and released to ambient air. To prevent the generated bioaerosols from posing a health threat to WWTP workers or habitants in the surroundings, the vent air and critical process air are treated as described in Chapter 6.1.

The assessment of the efficiency of the treatment of the air emissions from the WWTP to remove airborne virus and bacteria was performed by researchers from The National Research Centre for the Working Environment and DTU Food [23]. Air samples were collected 1.5 m above ground level from the following positions at the WWTP and examined for presence of potential harmful bacteria, viruses and endotoxins: Pre-treatment unit (indoor), bagging station (indoor), wastewater outlet (indoor), air outlet on the roof (outdoor), and 9 m downwind from the air outlet on the roof (outdoor). Reference measurements were taken upwind from the WWTP (outdoor). The sample collection was carried out on May 27 and June 23 2015, with sampling periods corresponding to approximately the length of a working day.

The investigations showed no significant differences in total particle and endotoxin concentrations between WWTP position measurements and upwind reference measurements. Total bacteria concentrations were found to be either comparable or significantly lower in the air emission than the upwind reference and inside the WWTP, indicating that the air treatment removes bacteria in the air to a level equal to or below what is found in the air at the reference point. Furthermore, there was no evidence of pathogenic bacteria being released in the air emission from the WWTP. Finally, traces of airborne Norovirus genomes were detected both at the WWTP air outlet and inside the WWTP, although in concentrations not likely to pose a health related risks to the WWTP surroundings. Hence, the risk of exposure to harmful pathogenic bacteria and viruses to the surroundings from air emission from Herlev Hospital WWTP is assessed to be very low.



## 11 Resource consumption and overall economy

### 11.1 Energy consumption

Energy consumption is a parameter of major concern in the assessment of the environmental effect and cost effectiveness of treatment processes. Energy consumption is therefore specifically addressed in this section.

During the reference period of August to November 2015, the total flow specific energy consumption of Herlev Hospital WWTP was found to be 2.3 kWh/m<sup>3</sup>. In Figure 11-1 is shown the energy consumption distributed on unit processes.

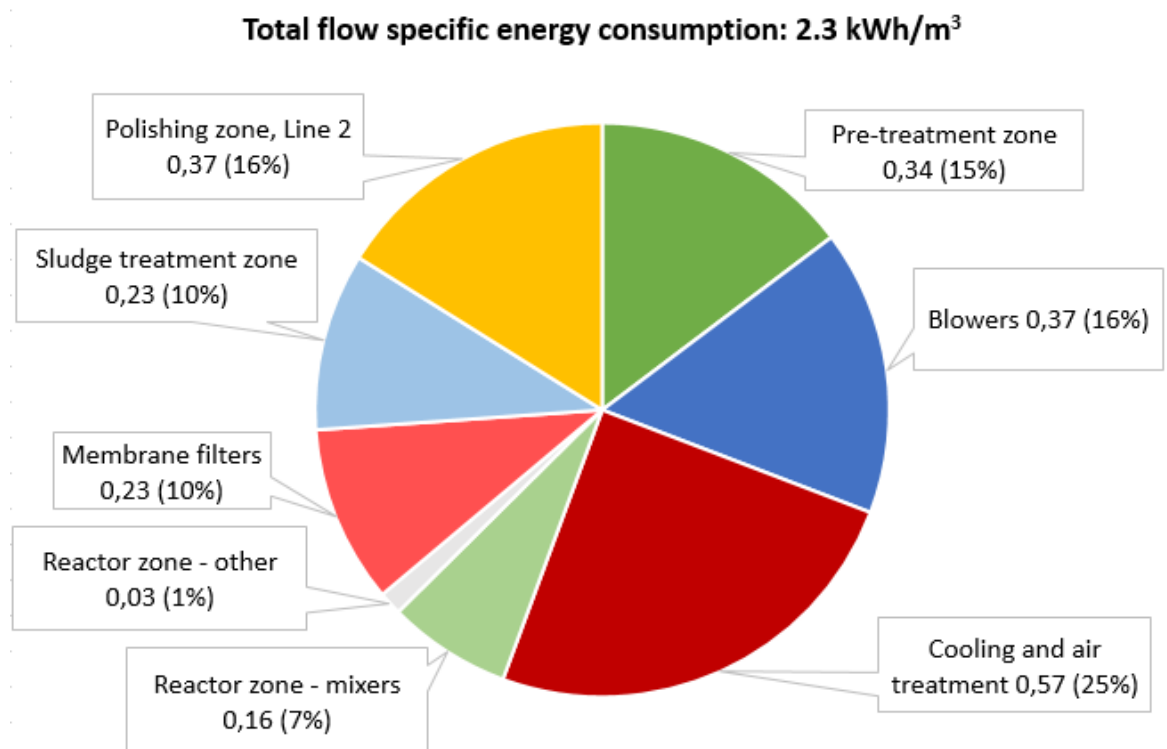


Figure 11-1 Flow specific energy consumption distributed on unit processes.

As it appears from Figure 11-1, the largest consumer of energy is the supply zone exclusive of blowers (air treatment and cooling water), followed by blowers (aeration of biology), polishing zone (pumps, ozone generation, UV-irradiation) and pre-treatment zone (pumps, screens, conveyor). The membrane filters (pressure pumps and rotation) make out 10% - the same as sludge treatment (dewatering and drying). The reactor zone (mainly mixers) makes out 8%.

### 11.2 Operational expenditures

In order to assess the operational expenditures for Herlev Hospital WWTP when operating in good and stable mode, it has been decided to look at the final four months (August to November 2015) of the total monitoring period covered by this report. In this period, focus has been on stable operation after completion of test campaigns and optimization of operational parameters for all unit processes. Regarding the polishing lines, the calculations are made for Line 2 only with ozonation followed by GAC-filtration, as this is the preferred configuration of the polishing

processes (see discussion in chapter 0). Flow specific costs are calculated based on a total influent flow of 48.586 m<sup>3</sup> during the period. There was no bypass at the inlet pumping station, so the permeate flow was the same as the inlet flow except for a very small difference, mainly caused by variations in the water level of the biological tanks, which function as a buffer between the inlet and the membrane filters. An inventory of all operational costs is given in Table 11-1.

Table 11-1 Flow specific operational expenditures for Herlev Hospital WWTP

	Aug-Nov 2015	Unit	Unit cost (DKK)	Cost (DKK)	Specific cost (DKK/m <sup>3</sup> )	
<b>Energy</b>						
Pre-treatment zone	16,432	kWh	1.29	21,197	0.44	
Supply zone - blowers	17,977	kWh	1.29	23,190	0.48	
Supply zone - other	27,694	kWh	1.29	35,725	0.74	
Reactor zone - mixers	7,774	kWh	1.29	10,028	0.21	
Reactor zone - other	1,458	kWh	1.29	1,880	0.04	
Membrane filters	11,175	kWh	1.29	14,415	0.30	
Sludge treatment zone	11,175	kWh	1.29	14,415	0.30	
Polishing zone, Line 2	8,988	kWh	1.29	11,595	0.48	
<b>Total Energy</b>						<b>2.97</b>
<b>Chemicals</b>						
PAX 15	3,741	l	2.4	8,945	0.18	
Ultrasil 25	772	l	21.0	16,208	0.33	
Ultrasil 78	82	l	24.0	1,974	0.04	
Polymer	329	kg	27.0	8,883	0.18	
Defoamer	0	kg	39.0	0	0.00	
O <sub>2</sub> for O <sub>3</sub> generator, Line 2	1,783	kg	2.0	3,565	0.15	
<b>Total chemicals</b>						<b>0.89</b>
<b>GAC</b>						
GAC - Line 2	486	kg	32	15,552	0.64	<b>0.64</b>
<b>By-products</b>						
Grit removal	5,560	kg	1.6	8,896	0.18	
Neutralox	0	m <sup>3</sup>	NA	0	0	
Sludge disposal fee	24.3	ton	435	10,571	0.22	
Sludge transport	4	Pick ups	59	236	0.005	
<b>Total by-products</b>						<b>0.41</b>
<b>Man hours</b>						
Time consumption	105	h	450	47,250	0.97	<b>0.97</b>
<b>Total operational expenditures</b>						<b>5.87</b>

As it appears from Table 11-1, the total operational expenditures are divided into consumption of energy, chemicals and GAC as well as costs related to disposal of solid residues (by-products) and time consumption for the operators.

Regarding change of spent GAC, it should be noted that in reality, there were no disposal of spent GAC in the period August to November 2015.

The shown values are based on the estimated yearly quantity of GAC to be changed (see discussion in section 6.3), divided by 3 to obtain a theoretical average for the 4 month period.

From Table 11-1 it can be seen that the total operational expenditures amount to 5.87 DKK/m<sup>3</sup> (0.78 EUR/m<sup>3</sup>), of which energy costs make out a little more than half of the total costs.

### 11.3 Overall economy of the Herlev Case

In the case of Herlev Hospital, the investment cost for a fully operational WWTP is assumed to be 25-35 million DKK. The investment is highly dependent on the construction of the building for the WWTP. The actual investment at Herlev was high due to a wish to construct a building for the WWTP with special architectural features.

As it appears from section 11.2, the operational expenses are found to be 5.87 DKK. In addition to this, there are also costs for general maintenance of the plant. These can be estimated based on the investment cost and in this case, a rate of 2-3% of the investment cost per year is used, corresponding to 750,000 DKK/year. With a yearly flow of 150,000 m<sup>3</sup>, as realized in 2015, this gives a flow specific maintenance cost of 5 DKK/m<sup>3</sup>. Total operation and maintenance cost can be assumed as 10.87 DKK/m<sup>3</sup>.

Herlev Hospital is presently paying a discharge fee of 25.54 DKK/m<sup>3</sup> for discharge of wastewater to the public sewer. This cost for discharge to the sewer should be compared with the O&M costs for Herlev Hospital WWTP, as it is expected that the treated wastewater from the WWTP can be discharged directly to the recipient (Kagsåen) in the future, in which case the sewer discharge fee would no longer apply.

The overall economic key figures for investment, operation & maintenance and sewer discharge fee are summarized in Table 11-2.

Table 11-2 Overall economic key figures for Herlev Hospital WWTP.

Type of cost	DKK	EUR
Investment cost	25 - 35 Mill. DKK	3.3 - 4.7 Mill EUR
Operation & Maintenance costs	10.87 DKK/m <sup>3</sup>	1.45 EUR/m <sup>3</sup>
Fee for discharge to public sewer	25.54 DKK/m <sup>3</sup>	3.41 EUR/m <sup>3</sup>

As it appears from Table 11-2, the fee for discharge to the public sewer is considerably higher than operation & maintenance cost, which means that the potential savings of running costs by treatment and direct discharge as compared to discharge to the public sewer is considerable (25.54 - 10.87 = 15 DKK/m<sup>3</sup>). It should be noted that depending on specific local conditions, there might be other costs related to direct discharge, such as establishment of a dedicated pipeline.

Investment costs are sometimes converted to running capital costs so that a total yearly cost, or total flow specific cost, can be compared with existing total yearly costs or existing total flow specific costs. In this case, however, this is omitted since the capital costs could vary a lot

depending on the local situation, and since the purpose in this context is to provide a general picture of the overall economy.

## 12 Evaluation of the technical concept

Different process trains for removal of micro pollutants in wastewater have been tested in a number of pilot and full scale projects, especially in Germany, Netherlands, Switzerland and France. Most of these projects used a sequence of treatment processes starting with biological treatment and followed by a number of polishing processes, including GAC-filtration and ozonation. There are different views as to the optimal order of GAC-filtration and ozonation, i.e. should GAC-filtration be the first step followed by ozonation or vice versa?

Prior to the start of the present project, a series of laboratory tests were made in order to provide indications as to the efficiency of the polishing processes (GAC, ozone and UV), including the effect of changes of operating parameters. The feed water used in these laboratory tests were permeate from a pilot MBR plant fed with wastewater from Herlev Hospital. The MBR technology was chosen for the biological stage, primarily because of the particle free effluent resulting from the use of membranes for separation of the biological sludge from the treated water, which is of the outmost importance for the efficiency of subsequent polishing processes.

Whereas the results of the laboratory tests gave good indications regarding the removal efficiency for pharmaceuticals, GAC adsorption capacity and ozone dose, they did not provide a final answer as to the optimal order of the GAC and ozone processes. To study this further, it was decided to test both sequences in full scale, and Herlev Hospital WWTP was therefore designed with two different polishing lines, where the sequence of the process steps in Line 1 was GAC + Ozone and the sequence in Line 2 was Ozone + GAC.

A comparison of the performance of the two polishing lines at Herlev Hospital WWTP indicates that ozonation followed by GAC-filtration is the most advantageous configuration considering best removal of pharmaceuticals as well as best operating economy.

In general, both configurations show excellent removal of pharmaceuticals, as pharmaceuticals not removed by the upstream process, are removed in the downstream process no matter which of the two processes comes first. However, as illustrated in Figure 7-7 and Figure 7-10, ozonation followed by GAC-filtration seems to have an even better removal efficiency than the opposite configuration. As discussed in section 7.1.5, this can most likely be explained by the change of composition of the effluent organic matter (DOC) by the ozonation, resulting in a reduced adsorbability of DOC. This leads to less adsorption competition against the organic micropollutants (such as the pharmaceuticals), resulting in more efficient removal rates.

Ozonation followed by GAC-filtration also seems to result in lower operational costs than the opposite configuration. This is indicated by the fact that pressure was building up faster in the first GAC filter of Line 1 (GAC + ozonation) than in the first GAC filter of Line 2 (ozonation + GAC), leading to the first change of GAC in Line 1 more than 4 months before the first change of GAC in Line 2 (see section 6.3). Please note that the second change in July was caused by a major leakage of sludge from the membrane filters resulting in immediate clogging of the GAC filters of both lines. This is in accordance with the effect of ozonation on the adsorbability of DOC as mentioned above, since reduced adsorbability would result in less usage of the GAC adsorption capacity and consequently lead to increased running time for the activated carbon filters, if ozonation is placed upstream from the GAC columns.

Although the configuration with ozone followed by GAC filtration results in a somewhat higher ozone consumption, as the concentration of organics in the influent to the ozonation reactor is higher than in the opposite configuration, this is more than compensated by less cost for change of GAC.

Based on this, it was decided to operate both polishing lines of the full scale plant with ozone followed by GAC (implemented from November 2015), Hence, the final overall process concept is as illustrated in Figure 12-1.

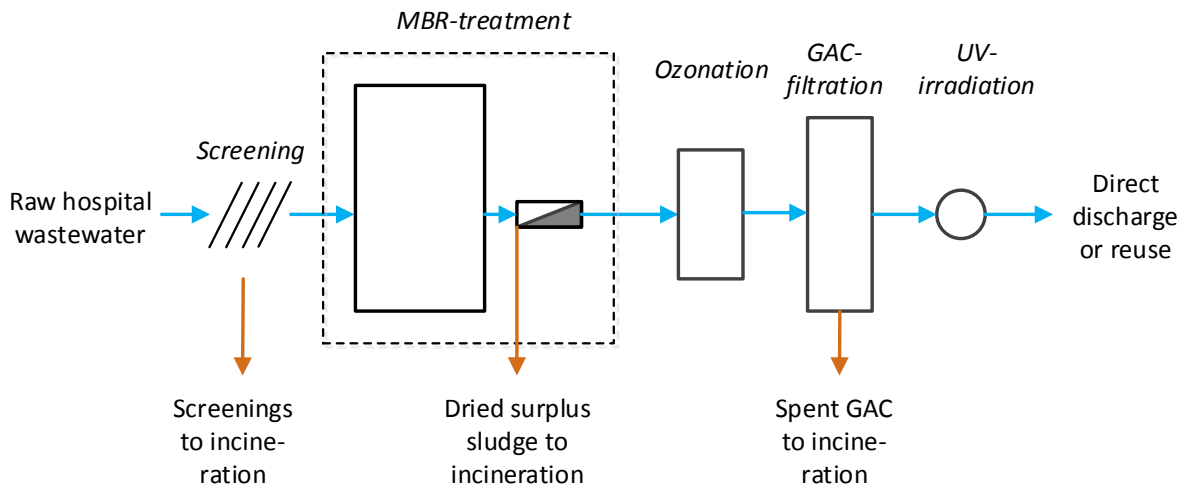


Figure 12-1 Final overall process concept for Herlev Hospital WWTP,

As indicated in Figure 12-1, the high quality of the WWTP effluent opens up for direct discharge to a recipient or for reuse purposes. Figure 12-2 illustrates how a dedicated hospital WWTP can be integrated in the overall water cycle of a hospital, including complete decoupling from the public sewer and rainwater collection systems. As it appears from the figure, the effluent from the hospital WWTP could be:

- infiltrated
- discharged directly to a local water body
- used for recreational purposes, i.e. to provide water to channels and ponds that are part of park landscapes inside the hospital area
- reused for technical purposes with cooling and irrigation water as the most obvious options

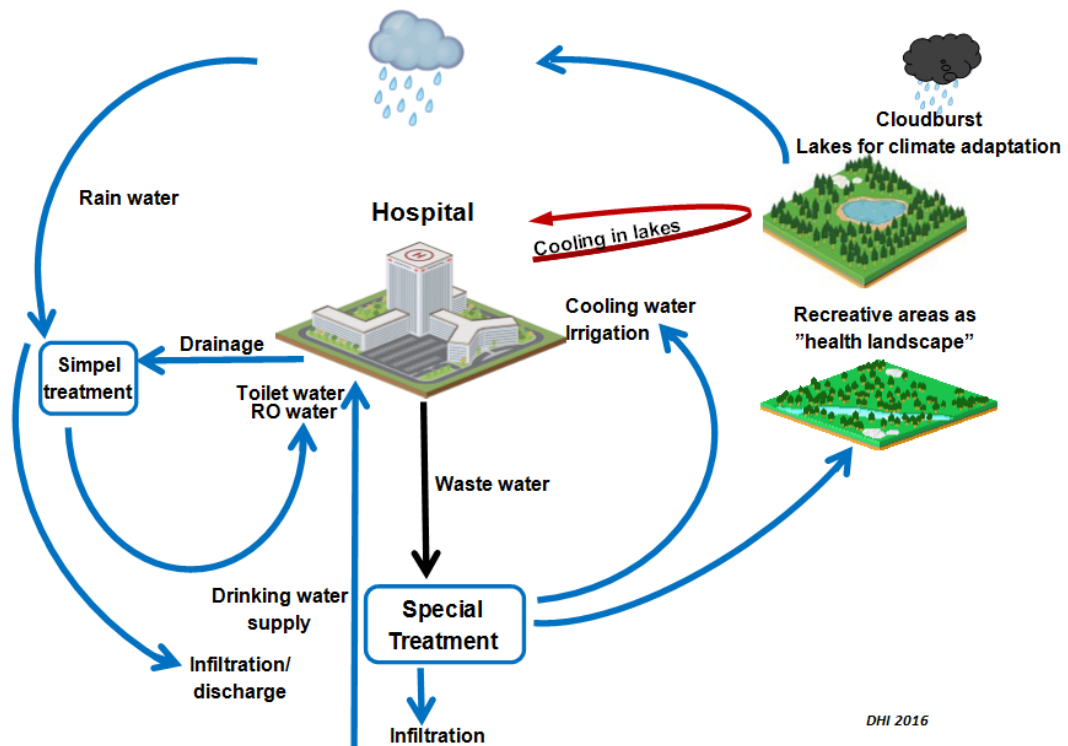


Figure 12-2 Scenarios for integration of a dedicated hospital WWTP in the overall water cycle for the hospital



Presently, specific reuse and recreational use of treated water from Herlev Hospital is planned. The plan is to:

- Release the treated wastewater to the nearby small stream named Kagså (see Figure 12-3 and Figure 13-1). Presently an amount of 140,000 m<sup>3</sup>/y, and in 2020 around 190,000 m<sup>3</sup>/y
- Reuse the treated wastewater in the existing cooling towers at the rooftop of Herlev Hospital (see picture in Figure 12-4). Around 10,000 m<sup>3</sup>/y will be reused here.

Practical planning for implementation is being carried out at the time of writing.



Figure 12-3 The local stream named Kagså situated next to Herlev Hospital



Figure 12-4 Cooling towers at the rooftop of Herlev Hospital



## 13 Health and environmental risk of direct discharge scenarios

The treated wastewater from Herlev WWTP is planned to be discharged into the local stream, Kagså. Kagså is a tributary to Harrestrup Å (Å = river in Danish), which flows into Kalveboderne, which is a marine water area see Figure 13-1.

The aim of the risk assessment is to establish acceptable limit-concentrations for selected pathogens and critical pharmaceuticals. Three discharge scenarios are analysed as described in Chapter 8:

- *Untreated raw wastewater.* This scenario simulates a complete breakdown of the WWTP, where untreated wastewater with concentrations equal to influent concentrations is discharged directly into Kagså during a period of several days
- *Reduced treatment efficiency.* This scenario simulates a realistic incident with an effluent quality corresponding to the observed effluent quality during the incident with the cracked membrane in November 2014. See Figure 7-6
- *Fully treated wastewater.* This scenario simulates normal operation

For the health risk assessment, two exposure scenarios are analysed:

- Indicator bacteria, norovirus and vancomycin resistant indicator bacteria at Lodsparken in Hvidovre, which is the bathing location in Kalveboderne nearest to Harrestrup Å
- A hypothetical worst case scenario, where children play in Kagså

For the environment, the effects of four critical pharmaceuticals are assessed in the marine recipient, Kalveboderne. The discharge of wastewater into Kalveboderne is estimated by hydraulic models and combined with measured influent and effluent concentrations to and from Herlev Hospital WWTP to yield estimations of concentrations in Kagså and in Kalveboderne.

For the indicator bacteria, the estimated concentrations are compared to the legal requirements to bathing water. Estimated Norovirus concentrations are compared to an acceptable health risk for infection of 3%, corresponding to an approximate health risk of excellent bathing water quality according to the methodology in the bathing water directive [13]. For the Vancomycin resistant enterococci, the ingested dose during bathing for children at Lodsparken is estimated and discussed.

The health and environmental risk assessments are described more in details in a separate report [39].

### 13.1 Hydrodynamic and process modelling

In Kalveboderne (marine area), the fresh water from Harrestrup Å will be mixed and diluted in the salt water. The dilution will vary due to variation in the flow in Harrestrup Å and the currents in Kalveboderne.

The hydraulic modelling was carried out using MIKE 3 FM. The MIKE 3 FM describes the spreading and transport of the contamination in three dimensions. The hydrodynamic model is coupled to an ecological equation solver, ECO Lab. The latter simulates the decay of the indicator bacteria and norovirus based on key forcing factors such as irradiance, temperature, salinity and the current delivered by MIKE 3 FM. The model is running in high resolution (flexible mesh), covering the bathing areas of the harbour. Hydrographical boundary conditions are delivered by the WaterForecast. This is an operational service by DHI providing daily updated data on current speed and direction, wave periods, heights and directions, salinity and temperature. Meteorological boundary conditions are acquired online from a Danish Weather Forecast supplier.

It is assumed that the treatment plant discharges a constant volume of 540 m<sup>3</sup>/day. Data regarding currents and flow in Harrestrup Å in the period from 27-5-2012 to 14-08-2012 have been used, based on catchment modelling and local rain gauged data.

Kagså is usually almost dry during the summer months. We have therefore assumed that the only water in Kagså during the model period is water from Herlev WWTP. This assumption does not significantly affect the dilution of water from the treatment plant in Kalveboderne, but may overestimate the concentration of wastewater from the Herlev treatment plant in Kagså during periods with heavy rain.

The reason why pharmaceuticals penetrate the wastewater treatment plant is that they have a low degradability and do not adsorb much. We therefore assume that the four selected pharmaceuticals do not degrade or adsorb on the way to Kalveboderne. The concentration of the pharmaceuticals in Kalveboderne is therefore calculated using the dilution of the water from the treatment plant. Consequently, the assessment of the pharmaceuticals is considered to be a worst case assessment.

The concentrations of norovirus, *E. coli* and enterococci were modelled by assuming a decay, once they reached Kalveboderne. Decay and sedimentation in Kagså and Harrestrup Å have not been included. The assessments of the concentration of norovirus, *E. coli* and enterococci is therefore considered to be a worst case assessment.



Figure 13-1: Map of Copenhagen showing the receiving waters, Kagså, Harrestrup Å and Kalveboderne, including the locations used for the risk assessment.

## 13.2 Environmental risk of pharmaceutical compounds

The exceeding of predicted no-effect concentrations (PNEC) has been modelled in Kalveboderne (marine area). Concentrations of four critical indicator substances (see Chapter 5) have been modelled. The substances are ciprofloxacin, clarithromycin, diclofenac and sulfamethoxazole.

The estimation of the average concentrations in the three scenarios (untreated, reduced and treated wastewater) is based on the dilution of the water discharged from Herlev Hospital WWTP and the concentration of the four compounds in the discharged water. The estimated concentrations were then compared to the marine PNEC<sub>Marine</sub>.

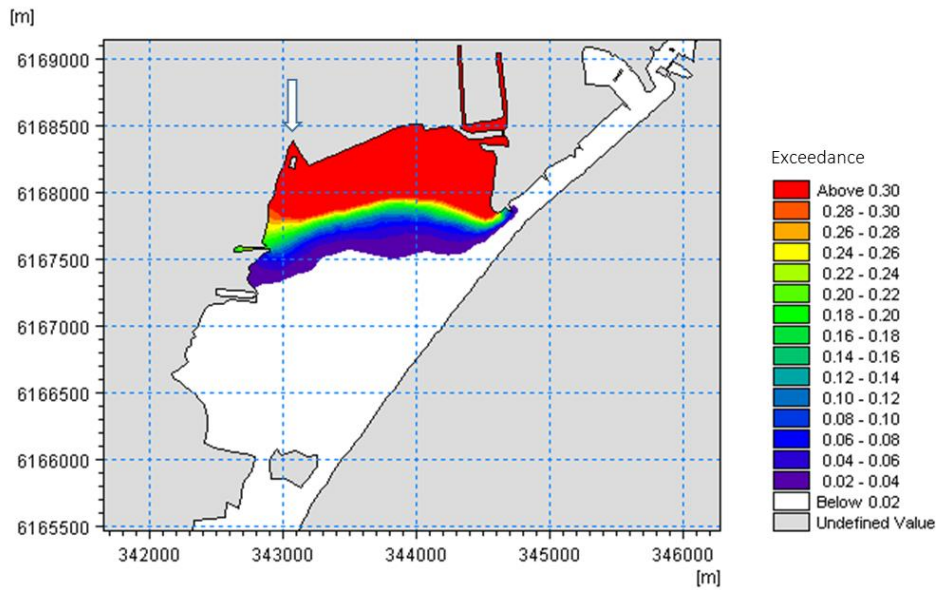


Figure 13-2: Fraction of time, where the concentration of ciprofloxacin is expected to exceed the PNEC<sub>Marine</sub> (9.8 ng/l), if untreated wastewater is discharged from Herlev Hospital WWTP. In red areas, the PNEC<sub>Marine</sub> will be exceeded in more than 30% of the time. Estimates are based on an average discharge concentration of 14,000 ng/l from Herlev Hospital WWTP. The arrow shows the outlet of Harrestrup Å.

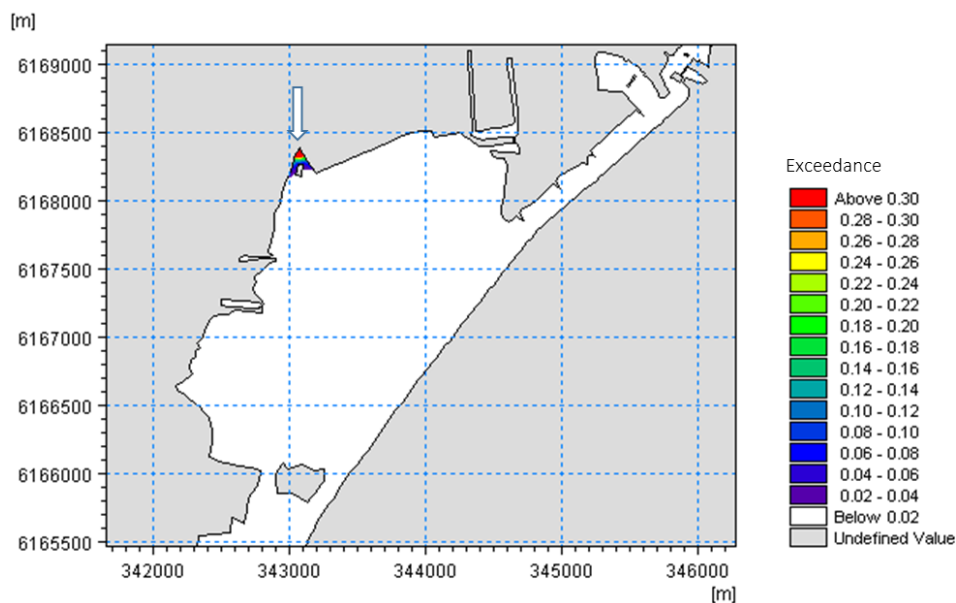


Figure 13-3: Fraction of time, where the concentration of Ciprofloxacin is expected to exceed the PNEC<sub>Marine</sub> (9.8 ng/l), if wastewater treated with reduced efficiency is discharged from Herlev Hospital WWTP. In red areas, the PNEC<sub>Marine</sub> will be exceeded in more than 30% of the time. Estimates are based on an average discharge concentration of 260 ng/l from Herlev Hospital WWTP. The arrow shows the outlet of Harrestrup Å.

The modelling shows that the concentration in Kalveboderne (the marine recipient) will not exceed the PNEC<sub>Marine</sub> at any time during a situation with a normal operation. The most critical compound is ciprofloxacin. If the treatment efficiency is reduced, e.g. due to cracked membranes, a small area at the outlet of Harrestrup Å runs a risk of being exposed to concentrations above PNEC<sub>Marine</sub>. If untreated wastewater is discharged over a prolonged period, a larger area will be exposed to concentrations above PNEC<sub>Marine</sub> for more than 30% of

the time. In order to stay below the  $PNEC_{\text{Marine}}$  in more than 95% of the time in a modelled point 200 m from the outlet point, the discharge concentrations should be below approximately 2,000 ng/l for the four compounds. These concentrations are considered to be a worst case scenario, since we have not included any degradation/decay from Herlev Hospital WWTP discharge to Kalveboderne.

### 13.3 Health risk assessment

This health risk assessment includes estimations of risk of infection with norovirus and estimations of faecal indicator bacteria concentration originating from water discharged from the Herlev WWTP to Kagså and further to the marine recipient, Kalveboderne. The three analysed scenarios are the same as described above (untreated, reduced and treated wastewater).

The concentration of the fecal indicator bacteria *E. coli* and enterococci was determined in the treated and untreated wastewater from Herlev Hospital WWTP. The transport to Kalveboderne and the dilution and decay of the bacteria (in Kalveboderne) were modelled in order to estimate the concentrations at the beach (Lodsparken). The modelled concentrations were compared to the EU bathing water requirements (see Figure 13-4 and Figure 13-5).

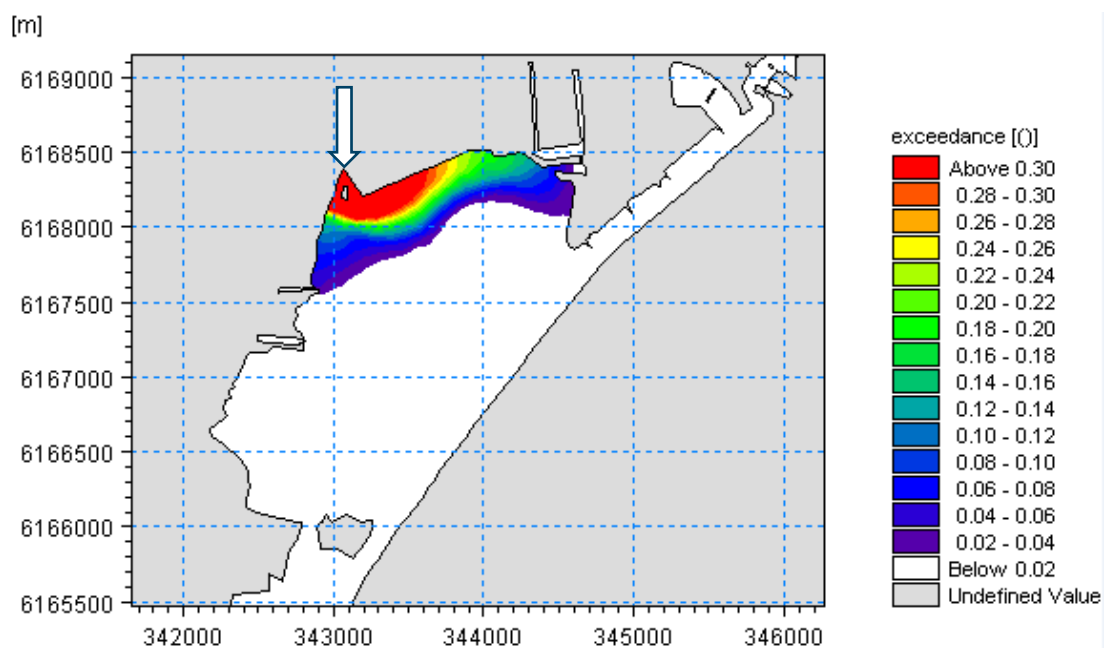


Figure 13-4 Fraction of time, where the concentration of *E. Coli* is expected to exceed the excellent bathing water limit (250 MPN per 100 ML), if untreated wastewater is discharged from Herlev Hospital WWTP. In red areas, the limit will be exceeded in more than 30% of the time. Estimates are based on an average discharge concentration of  $5.5 \cdot 10^6$  MPN per 100 ML from Herlev Hospital WWTP. The arrow shows the outlet of Harrestrup Å.

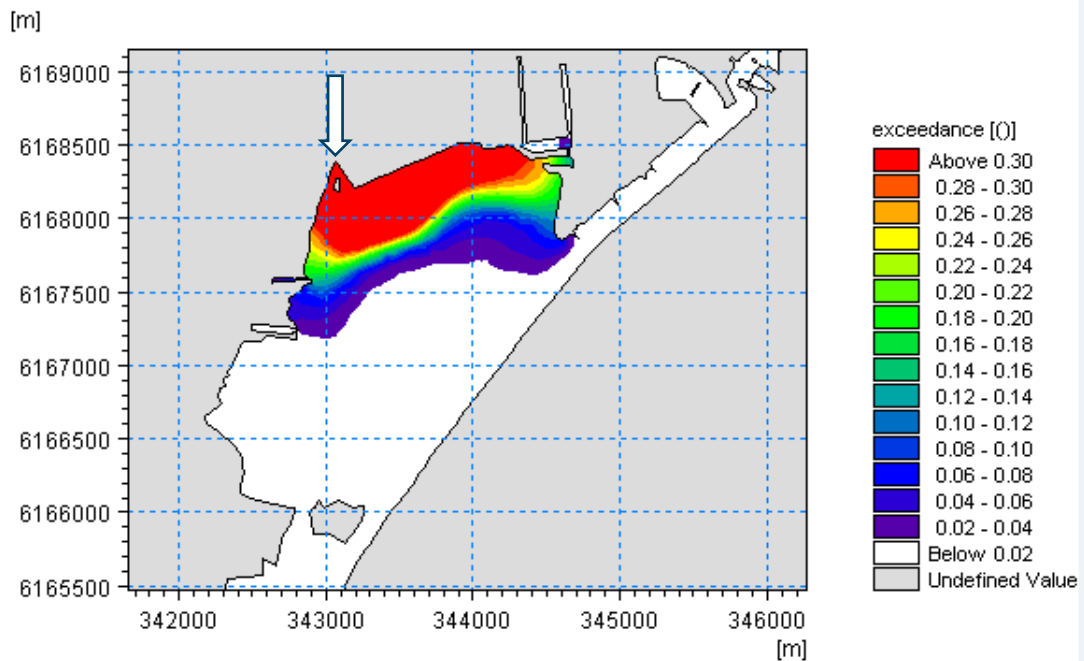


Figure 13-5 Fraction of time, where the concentration of enterococci is expected to exceed the excellent bathing water limit (100 MPN per 100 ML), if untreated wastewater is discharged from Herlev Hospital WWTP. In red areas, the limit will be exceeded in more than 30% of the time. Estimates are based on an average discharge concentration of  $9.5 \cdot 10^6$  per 100 ML from Herlev Hospital WWTP. The arrow shows the outlet of Harrestrup Å.

As was done for the pharmaceuticals (section 13.2), we have determined the distributions of concentrations using MonteCarlo simulation, which makes it possible to present the results as percentiles.

Table 13-1: Estimated mean concentrations and 95%-tiles of *E. coli* and enterococci at the beach Lodsparken after discharge of untreated, partially treated and treated wastewater, as well as the legal requirements to excellent water quality. Unit: pr. 100 ml

Discharge scenario:	Untreated wastewater	Reduced treatment efficiency	Fully treated wastewater
<b><i>E. coli</i> (per 100 ml)</b>			
Average	298	0.00006	< 0.00003
95%-tile	876	0.00016	< 0.00010
Requirement (95%-tile)*	250	250	250
<b>Enterococci. (per 100 ml)</b>			
Average	974	0.00033	< 0.00006
95%-tile	5234	0.00098	< 0.00036
Requirement (95%-tile)*	100	100	100

\*Required 95%-tile for achieving excellent bathing water quality according to the European Bathing Water Directive [8].

Table 13-1 shows the estimated average concentrations and the 95%-tiles for *E. coli* and enterococci. The table also shows the required 95%-tile for achieving “excellent bathing water quality” according to the European Bathing Water Directive [8]. Table 13-1 shows that if untreated wastewater from Herlev Hospital is discharged, it is likely to surpass the limit for “excellent bathing water quality” for both *E. coli* and enterococci.

The concentration of *E. coli* (<1 MPN/100 ML) and enterococci (<1 MPN/100 ML) in the treated water is so low (see Table 8-1) that it will comply with the requirements of the Bathing Water Directive, even during a direct undiluted release to Kagså.



The concentration of norovirus at the beach Lodsparken was modelled in the same way as it was done for *E. coli* and enterococci. The results are shown in Table 13-2. If untreated wastewater is discharged from Herlev Hospital WWTP, the average concentration of norovirus in the stream Kagså is estimated to be 160,000 gene copies/l and 22 gene copies/l at the beach Lodsparken. The treatment with reduced efficiency removes approximately 3 log<sub>10</sub>units of the noroviruses and > 4 log<sub>10</sub>units when the wastewater is fully treated, assuming a limit of detection of 26 gene copies/l.

Table 13-2: Estimated mean and 95%-tiles of estimated concentrations (Genome copies/L) and risks of norovirus infection (for children pr. bath in 1 hour) after discharge of untreated, reduced treatment and fully treated wastewater into the stream Kagså and at the beach Lodsparken.

Discharge scenario:	Untreated wastewater	Reduced treatment efficiency	Fully treated wastewater
<b>Kagså (receiving stream)</b>			
Average concentration (GC/L)	170,000	170	< 26
95%-tile concentration (GC/L)	625,000	861	< 26
Average Risk of infection	31%	0.4%	< 0.03%
95%-tile Risk of infection	58%	1.9%	< 0.11%
<b>Lodsparken (bathing water)</b>			
Average concentration (GC/L)	22	0.022	< 0.002
95%-tile concentration(GC/L)	72	0.051	< 0.012
Average Risk of infection	0.048%	5.1 · 10 <sup>-5</sup> %	< 3.9 · 10 <sup>-6</sup> %
95%-tile Risk of infection	0.096%	8.6 · 10 <sup>-5</sup> %	< 1.3 · 10 <sup>-5</sup> %

The risk of infection was calculated by applying the approximated β-Poisson norovirus dose-response model with immunity [25]. The dose-response model is shown in Figure 13-6. The Figure shows the probability of infection (a probability of 1 corresponds to 100%) as a function of the ingested dose of norovirus.

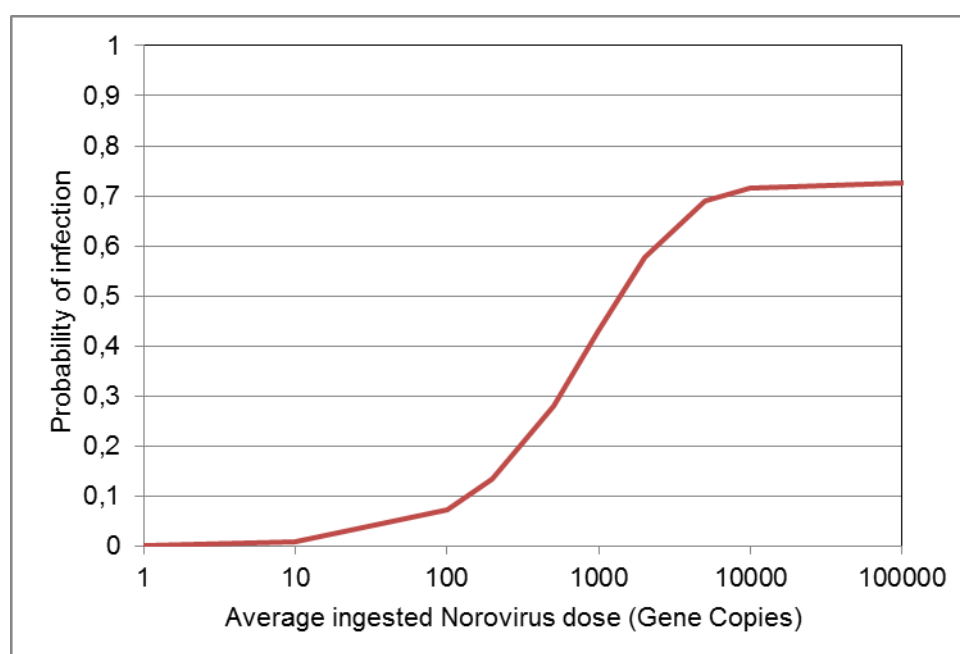


Figure 13-6: The approximated β-Poisson dose-response model with immunity ( $\alpha = 2.91$ ,  $\beta = 2734$ , [21]). The figure shows the relation between the ingested dose and the risk of infection for a population. A certain proportion of the population is immune. Therefore, the highest probability of infection is around 72%.

The ingested dose is determined by multiplying the concentration of norovirus in the ingested water with the ingested volume, which was assumed to 37 ml on average for children during 1 hour bathing (Gamma distributed  $r = 0.64$ ,  $\lambda = 58$ , [23]). The distribution of the risk of infection was estimated by using the MonteCarlo simulation.

The estimated risks of infection are shown in Table 13-2. A risk below 3% in average risk of infection corresponds to an excellent bathing water quality according to the methodology used in the EU Bathing Water Directive [13]. If untreated wastewater is discharged into stream Kagså and children play or bath in the water, the average estimate of the risk of infection is 31%, but there is a 5% probability that the risk of infection is higher than 58%. During the period, where the treatment efficiency was reduced, the risk of infection was below the acceptable 95%-tile of 3%. Considering that other pathogens may be present in the wastewater, the total health risk may anyway be higher than 3% in Kagså during an incident with reduced treatment. The risk of infection is negligible during normal treatment.

The risk of infection with norovirus at the beach Lodsparken is way below the 3% risk for infection accepted for bathing water.

A reversed calculation has been used to estimate the acceptable 95%-tile concentration of discharge in the Herlev Hospital WWTP wastewater. The 95%-tile discharge concentration for achieving an average risk of norovirus infection of 3% pr. bath [13] at the beach Lodsparken was estimated to be approximately  $3 \cdot 10^6$  NoV gene copies/L. The 95%-tile discharge concentration in wastewater from Herlev Hospital WWTP for achieving an average risk of norovirus infection of 3% pr. bath [13] in Kagså was estimated to be approximately 360 NoV gene copies/L.

It should be mentioned that a considerable uncertainty is associated to the estimation of the risk of infection by norovirus. Beside the uncertainty on the measured and calculated concentrations, the uncertainty is related to several other issues, such as:

- The data underlying the norovirus dose-response curve are relatively weak
- The dose response trials were executed on healthy adults, not children
- The norovirus strains used for the dose response trials may have been different from the strains detected in our study.
- The qPCR method detects the presence of norovirus RNA, which do not necessarily originates from infective norovirus particles.

The risk given in this report should therefore be seen as the best available information concerning the risks associated with the discharge of wastewater from Herlev Hospital WWTP.

### 13.3.1 Concentrations and ingested doses of vancomycin resistant enterococci

The distribution of the concentrations of vancomycin resistant enterococci (VRE) was estimated for the three scenarios. The concentrations have been used as input for estimation of doses of VRE using the method described for norovirus in the section above. The results are shown in Table 13-3. It is seen that the chance of ingesting a VRE during bathing at the beach Lodsparken is negligible, when the water is fully treated (concentration in treated wastewater is below limit of detection, see Table 8-2. In case of discharge of untreated water, a considerable number of VREs is likely to be ingested.

Table 13-3: Estimated average concentrations and 95%-tiles concentrations of vancomycin resistant enterococci at the beach Lodsparken after discharge of untreated, partially treated and treated wastewater, and the associated estimated doses children will be exposed to during 1 bath.

Discharge scenario:	Untreated wastewater	Reduced treatment efficiency	Fully treated wastewater
<b>Concentration of vancomycin resistant enterococci (pr. 100 ml)</b>			
Average	53	0.00011	< 0.00006
95%-tile	262	0.00047	< 0.00036
<b>Dose of vancomycin resistant enterococci ingested by children during 1 bath</b>			
Average	18	0.00004	< 0.00002
95%-tile	46	0.00008	< 0.00005

*Enterococcus faecium* and *Enterococcus faecalis* are inhabitants of the human intestine but they are also important pathogens. Typical clinical manifestations include bacteremia, urinary tract infections, and skin infections [29]. They are genetically capable of acquiring, conserving and disseminating genetic traits, including resistance determinants among enterococci and related Gram-positive bacteria [27]. Hence, the presence of VRE in bathing water may contribute to the spread of VRE or resistant properties to bathers and subsequently an increased risk of infections that are difficult to treat. However, transfer of VRE from environmental sources causing infections in humans have not yet been documented [28] and the risk is currently unknown. Our results suggest that a continuous discharge of untreated wastewater over days may expose bathers to an increased risk of infection, whereas the treatment makes the probability of ingesting a VRE negligible.

### 13.3.2 Conclusion on health risk assessments

The assessments have shown that even after a realistic incident with a decrease in the treatment efficiency, the treated wastewater will not have any influence on the hygienic bathing water quality at the beach Lodsparken. Only a total breakdown over a period of days will result in breaching of the legal requirements to bathing water quality. Similarly, a calculation of the risk of infection with norovirus during bathing at Lodsparken shows negligible risk during normal operational conditions. Even discharge of untreated wastewater will only have a small impact on the risk of norovirus infection at Lodsparken.

In Kagsåen, where the treated wastewater is assumed to be undiluted, the estimated risk of norovirus infection is negligible during normal operation. However, if children play in the water during incidences with reduced treatment efficiency, a small risk of infection, corresponding to the requirements to excellent bathing water quality, can be expected. As other pathogens may be present, the risk of infection may be larger. Concerning *E. coli* and enterococci, only a situation with discharge of untreated wastewater will pose a risk above excellent bathing water quality. It must be taken into account that health risk assessment is associated with a large uncertainty and should be regarded as the best available information concerning the risks associated with the discharge of wastewater from Herlev Hospital WWTP.

Vancomycin resistant enterococci may reach the beach Lodsparken and be ingested by the bathers only if untreated wastewater is discharged over a period of days. Only in this case there is an increased risk of transferring VRE or antibiotic resistant properties.



## 14 References

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