Removal of APIs and bacteria from hospital wastewater by MBR plus O_3, $O_3 + H_2O_2$, PAC or ClO_2

U. Nielsen, C. Hastrup, M. M. Klausen, B. M. Pedersen, G. H. Kristensen, J. L. C. Jansen, S. N. Bak and J. Tuerk

ABSTRACT

The objective of this study has been to develop technologies that can reduce the content of active pharmaceutical ingredients (APIs) and bacteria from hospital wastewater. The results from the laboratory- and pilot-scale testings showed that efficient removal of the vast majority of APIs could be achieved by a membrane bioreactor (MBR) followed by ozone, ozone + hydrogen peroxide or powdered activated carbon (PAC). Chlorine dioxide (ClO_2) was significantly less effective. MBR + PAC (450 mg/l) was the most efficient technology, while the most cost-efficient technology was MBR + ozone (156 mg O_3/l applied over 20 min). With MBR an efficient removal of *Escherichia coli* and enterococci was measured, and no antibiotic resistant bacteria were detected in the effluent. With MBR + ozone and MBR + PAC also the measured effluent concentrations of APIs (e.g. ciprofloxacin, sulfamethoxazole and sulfamethizole) were below available predicted no-effect concentrations (PNEC) for the marine environment without dilution. Iodinated contrast media were also reduced significantly (80–99% for iohexol, iopromide and ioversol and 40–99% for amidotrizoateacid). A full-scale MBR treatment plant with ozone at a hospital with 900 beds is estimated to require an investment cost of €1.6 mill. and an operating cost of €1/m^3 of treated water.

Key words | Advanced oxidation processes, bacteria, hospital wastewater, MBR, pharmaceuticals

U. Nielsen (corresponding author)
C. Hastrup
M. M. Klausen
B. M. Pedersen
G. H. Kristensen
DHI, Urban & Industry,
Agern Allé 5 DK-2970 Hørsholm,
Denmark
E-mail: *uln@dhigroup.com*

J. L. C. Jansen
Water and Environmental Engineering,
Dept of Chemical Engineering,
Lund University,
P.O. Box 124, SE-221 00 Lund,
Sweden

S. N. Bak
Grundfos Biobooster A/S,
Randersvej 22a, DK8870 Langaa,
Denmark

J. Tuerk
Research Analysis,
Institute of Energy and Environmental Technology (IUTA), Bliersheimer Str. 60,
D-47229 Duisburg,
Germany

INTRODUCTION

Pharmaceutical consumption at hospitals is high compared to the consumption in other parts of society. High pharmaceutical consumption and excretion by patients means that hospital wastewater has a high concentration of many different active pharmaceutical ingredients (APIs). Also, hospital wastewater contains microorganisms, some of which are pathogenic. Hospital wastewater is almost always released to a municipal drainage network via which the hospital wastewater is mixed with other municipal wastewater (and stormwater in combined systems) and transported to a municipal wastewater treatment plant (WWTP) for treatment. Municipal WWTPs are often not designed to remove micropollutants such as APIs, and therefore removal at municipal WWTPs is often poor, especially for some APIs. Numerous studies have focused on the occurrence and fate of APIs in the environment. In many of these studies, APIs have been detected in WWTP effluents and in receiving water bodies (Fredskilde & Nielsen 2008; Bartelt-Hunt *et al.* 2009; Richardson & Ternes 2011) illustrating that APIs are released to the aquatic environment where they may have an effect, depending on concentration (Escher *et al.* 2011; Jean *et al.* 2012). Also, hospital wastewater can be released untreated to recipients when combined sewers overflow during heavy rains. During combined sewer overflows, there is also a risk of human exposure to hospital wastewater, which can pose a public health threat.

The objectives of the study were to test, on a pilot and laboratory scale, different treatment technologies for on-site treatment of hospital wastewater. Urine separation at hospitals and the possibility of treatment of urine alone to remove APIs were also studied in depth. However, urine separation and treatment were found to be problematic due to frequent demands for maintenance and high construction/retrofit costs associated with the additional plumbing requirements, and urine separation is therefore

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not covered further in this article. Furthermore, the objectives were to assess the efficiency of the tested treatment technologies as well as to assess the construction and operating costs of each technology, allowing for a final evaluation of the tested technologies based on removal efficiencies and costs. The pre-treatment activities were carried out in a membrane bioreactor (MBR) pilot plant followed by laboratory testing of four polishing technologies: ozone (O_3), ozone + hydrogen peroxide ($O_3 + H_2O_2$), powdered activated carbon (PAC) and chlorine dioxide (ClO_2). MBR technology was chosen because of a lower footprint and more efficient removal of micropollutants than conventional treatment plants and because it provides a significant hygienization of sewage water (Ottoson *et al.* 2006; Radjenovic *et al.* 2009; Beier *et al.* 2012). The chosen polishing technologies are from a mechanistic point of view potentially effective on micropollutants such as APIs.

METHODOLOGY

Collection and transport of hospital wastewater

The hospital wastewater used for the study was collected at Denmark's largest hospital, Copenhagen University Hospital, which was selected for wastewater collection due to the many functions performed and thereby pharmaceuticals consumed at the hospital. The wastewater from the hospital was collected from a central sewage well containing wastewater from all wings of the hospital, ensuring a representative API composition. Wastewater was collected on two separate occasions to ensure sample freshness. The wastewater was pumped from the well into a tanker truck of 33 m^3 capacity, which was rinsed internally prior to transport to prevent contamination from previous contents, and then driven from the hospital to the MBR pilot plant.

MBR setup

The MBR pilot plant, designed and produced by Grundfos BioBooster A/S, was set up in Bjerringbro, Jutland approximately 340 km from Copenhagen University Hospital where the wastewater samples were collected. The MBR pilot plant consisted of a 1 mm sieve to remove larger particles and a pipe of 0.37 m^3 volume; the area of the ceramic ultrafiltration membrane was 3.75 m^2 with a pore diameter of 0.06 μm. Backwash was performed every other minute with 4 l permeate. The flow through the MBR pilot plant was 2.2 m^3/day and 24.6 l/m^2/h. The sludge age was approximately 35 days but is only an estimate due to the relatively short period of operation.

Three test runs were performed with the MBR. The first run (April 24 – May 6, 2011) was a commissioning period where municipal wastewater from Bjerringbro was treated. The second run (May 6 – May 19, 2011) was made using the first batch of hospital wastewater from Copenhagen University Hospital in order to allow the microorganisms to adjust to the contents of hospital wastewater. The third and final run (May 19 – May 21, 2011) was also made using wastewater from Copenhagen University Hospital, but here the second batch of wastewater was used.

Polishing technologies setup

The four polishing technologies tested in this study were tested at laboratory scale on MBR-treated hospital wastewater. The treated wastewater used in the experiments with the polishing technologies was sampled during the third round of MBR testing.

Ozone

The ozone was produced in a LAB2B ozone-generator from Triogen Ltd with a capacity of up to 10 g O_3/h and a built-in gas flow meter and regulator valve for gas flow. Ozone concentration in the gas phase was monitored using UV-spectrophotometry. Dissolved ozone was monitored amperometrically using a membrane ozone sensor (0–10 mg/l range) with a membrane insensitive to chemicals and tensides. A ceramic diffuser (VitraPOR® glassfilter from ROBU® with porosity class 4 (10–16 μm)) mounted at the bottom of the glass reaction column was used for ozone entry. The wastewater sample was continuously circulated through the reaction column at a minimum flow of 30 l/h, and samples were taken from the column via a two-way glass valve throughout the treatment process for analysis of the removal process. Four reaction times were tested in order to compare API removal and treatment costs associated with each reaction time. The tested reaction times were 3, 6, 10 and 20 min corresponding to ozone doses of 26, 52, 82 and 156 mg O_3/l (or 1.3, 2.6, 4.1 and 7.8 g O_3/g TOC), respectively. The doses were chosen based on experimental data for the ozone consumption monitored continuously in both gas and liquid phase during the experiment as well as the corresponding decline in dissolved organic carbon and UV light absorbance at 254 nm measured in grab samples taken at specified time intervals during the experiment.

Ozone + hydrogen peroxide

The test procedure for ozonation with addition of hydrogen peroxide was very similar to that of ozonation only. Hydrogen peroxide was dosed using a pump and adjusted to the ozonation rate. A hydrogen peroxide to ozone ratio of 0.45 g/g was used to ensure a slight surplus of hydrogen peroxide. Four reaction times were tested: 5, 15, 25 and 40 min corresponding to ozone doses of 130, 450, 780 and 1,300 mg O_3/l and hydrogen peroxide doses of 60, 200, 350 and 585 mg H_2O_2/l, respectively. The doses were chosen based on experimental data equalling the ozone tests.

Chlorine dioxide

The test setup for oxidation with chlorine dioxide consisted of a constantly stirred tank. The chlorine dioxide concentration was monitored at constant intervals and regulated, if necessary, by adding more chlorine dioxide to the tank. The chlorine dioxide consumption was 60 mg/l MBR-treated hospital wastewater. Two reaction times were tested in order to compare API removal and treatment costs: 15 and 120 min. The doses were chosen just above the consumption of the liquids. The concentration was followed during the course of the experiment and dosing stopped when no more chlorine dioxide was consumed.

Powdered activated carbon

The laboratory tests with activated carbon were performed in batches in blue-cap glass bottles. Two types of PAC were used, Norit 830W and Calgon Carbon Filtrasorb F400. Each of the two PAC types were tested in three concentrations: 150, 300 and 450 mg/l. The chosen PAC concentrations were based on a Swedish study of APIs in the aquatic environment (Wahlberg *et al.* 2010). MBR-treated hospital wastewater and PAC at the designated concentrations were added to the blue-cap bottles, placed on a shaking table and allowed to stand for 48 h. Afterwards the PAC was removed through filtration using a 1 μm glass fiber filter, and the sample was frozen and sent for analysis.

Analytical procedures

Analyses were performed both before and after MBR-treatment and treatment with the four polishing technologies. Central to the study were the analyses of API concentrations in the hospital wastewater both before and after treatment, which allowed for assessment of the efficiency of the different treatment technologies in removing APIs. Analyses were performed at two laboratories, the Institute of Energy and Environmental Technology (IUTA) in Germany and Umeå University in Sweden. The IUTA analyses screened for more APIs than Umeå's analyses, and thus the IUTA methods are described here. Water analyses were performed after solid phase extraction (SPE) by liquid chromatography tandem mass spectrophotometry (LC-MS/MS). Three different SPE and separation methods were used depending on the type of API. 93 APIs were measured.

Analyses of the microbial contents of the hospital wastewater were performed on the influent and effluent of the MBR pilot plant. The analyses were performed at DHI's laboratories in Denmark by incubation on agar plates. The plates were incubated at 37 °C for 22–26 h.

RESULTS

Results of the pilot-scale MBR treatment and laboratory-scale testing of polishing technologies are presented in the following section.

Results of MBR treatment

Three pairs of samples were taken of influent and effluent from the MBR pilot plant in order to evaluate the efficiency of the plant in removing APIs. The samples were taken from the third test run of the pilot plant. Samples 1 and 2 were taken on the morning and afternoon of May 20, 2011, respectively, and sample 3 was taken on the morning of May 21, 2011. Table 1 shows the concentrations of 60 different APIs in the influent and effluent of the MBR pilot plant. Removal is also shown. The treatment efficiency of the plant varies greatly from substance to substance. For some substances the concentrations are below the limit of detection in the influent, and therefore it is not possible to calculate removal for these substances.

Table 1 shows that the removal of common pain killers like ibuprofen and paracetamol was very high with over 99% removal for both APIs. Hormones (17-α-estradiol and 17-β-estradiol) also showed a high removal. The MBR pilot plant also showed efficient treatment for β-lactam antibiotics with a mean removal rate of 97% for amoxicillin and 73% for cefuroxime. The removal rates for sulfamethoxazole and sulfamethizole were also high at 97 and 99%, respectively, while the removal rate for N^4-Acetylsulfamethoxazole, a metabolite of sulfamethoxazole, was only 38%.

Table 1 | Influent and effluent concentrations in ng/l from the MBR pilot plant along with removal in percent. The analyses were performed at the IUTA laboratories. Mean values are shaded grey. Values are rounded

	Influent			Effluent			Removal			
API	Sample 1 [ng/l]	Sample 2 [ng/l]	Sample 3 [ng/l]	Sample 1 [ng/l]	Sample 2 [ng/l]	Sample 3 [ng/l]	Sample 1 [%]	Sample 2 [%]	Sample 3 [%]	Mean [%]
Acetylsalicylic acid	<10	<10	<10	<10	<10	<10				
ac-sulfadiazine	110	130	150	30	38	45	73	71	70	71
ac-sulfamethoxazole	63	59	79	43	32	50	32	46	37	38
Amoxicillin	33	37	43	1	1	1	97	97	98	97
Atenolol	170	180	250	50	57	73	71	68	71	70
Azithromycin	1,600	1,900	2,500	590	700	770	63	63	69	65
Bendroflumethiazid	<25	<25	<25	<25	<25	<25				
Bezafibrat	<10	<10	<10	<10	<10	<10				
Bisoprolol	30	27	32	13	9	20	57	65	38	53
Capecitabine	45	41	40	10	12	11	78	71	73	74
Carbamazepine	2,300	2,500	3,200	2,600	2,200	3,100	−13	12	3	1
Cefuroxime	210	200	150	52	52	44	75	74	71	73
Chloramphenicol	<10	<10	<10	<10	<10	<10				
Ciprofloxacin	6,000	6,400	7,600	4,900	3,300	4,500	18	48	41	36
Citalopram	300	300	400	320	300	340	−7	0	15	3
Clarithromycin	1,300	1,400	1,800	470	540	590	64	61	67	64
Clindamycin	31	27	24	99	80	110	−219	−196	−358	−258
Clofribic acid	<10	<10	<10	<10	<10	<10				
Cyclophosphamide	14	12	16	12	11	14	14	8	13	12
Diclofenac	170	150	130	91	130	130	46	13	0	20
Erythromycin	330	370	520	230	220	310	30	41	40	37
Erythromycin dehyd.	610	660	840	460	420	570	25	36	32	31
Fenofibrat	31	28	<12.5	22	<12.5	<12.5	29	>55		
Fenofibrinsäure	<5	<5	<5	<5	<5	<5				
Furosemide	4,400	4,600	6,100	5,000	5,000	5,000	−14	−9	18	−1
Gemcitabine	<5	<5	<5	<5	<5	<5				
Ibuprofen	2,400	2,200	1,900	1	1	1	>99	>99	>99	>99
Ifosfamide	62	70	91	69	62	94	−11	11	−3	−1
Ketoprofen	<10	<10	<10	<10	<10	<10				
Megastrol	<5	<5	<5	<5	<5	<5				
Metoprolol	3,700	3,600	2,900	2,500	2,100	2,800	32	42	3	26
Metronidazol	<125	<125	<125	<125	<125	<125				
MTX	290	310	190	1	1	1	>99	>99	99	99
Naproxen	<10	<10	<10	<10	<10	<10				
Ofloxacin	<5	<5	<5	<5	<5	<5				
Oxcarbazepine	300	380	450	1	2	1	>99	99	>99	>99
Paracetamol	26,000	23,000	32,000	1	1	1	>99	>99	>99	>99
Phenanzon	84	83	89	110	86	120	−31	−4	−35	−23
Propranolol	260	280	360	220	190	270	15	32	25	24

(*continued*)

Table 1 | continued

	Influent			Effluent			Removal			
API	Sample 1 [ng/l]	Sample 2 [ng/l]	Sample 3 [ng/l]	Sample 1 [ng/l]	Sample 2 [ng/l]	Sample 3 [ng/l]	Sample 1 [%]	Sample 2 [%]	Sample 3 [%]	Mean [%]
Roxithromycin	130	130	160	110	110	130	15	15	19	17
Simvastatin	<10	<10	<10	<10	<10	<10				
Sotalol	16	17	28	19	17	28	−19	0	0	−6
Sulfadiazine	620	630	380	7	12	7	99	98	98	98
Sulfamethazine	<10	<10	<10	<10	<10	<10				
Sulfamethizole	1,500	1,500	1,600	11	15	17	99	99	99	99
Sulfamethoxazole	12,000	12,000	16,000	270	320	450	98	97	97	97
Tamoxifen	<5	<5	<5	<5	<5	<5				
Tramadol	1,500	1,800	2,000	1,400	1,300	2,000	7	28	0	11
Trimethoprim	3,800	4,100	4,900	3,700	3,300	4,000	3	20	18	14
Venlafaxin	470	540	670	510	390	590		28	12	20
17α-Estradiol	23	12	20	<1	<1	6	>96	>92	70	70
Estron	<1	<1	<1	<1	<1	<1				
17β Estradiol	<1	9	7	<1	<1	<1		>89	>86	
17α Ethinyl-Estradiol	<5	<5	<5	<5	<5	<5				
Amidotrizoeacid	47,000	22,000	30,000	39,000	22,000	41,000	17	0	−37	−7
Iohexol	640,000	400,000	540,000	340,000	430,000	390,000	47	−8	28	22
Iomeprol	<50	<50	<50	12,000	<50	<50				
Iopamidol	<50	<50	<50	<50	<50	<50				
Iopromide	750,000	530,000	870,000	470,000	660,000	490,000	37	−25	44	19
Ioversol	450,000	290,000	430,000	290,000	340,000	340,000	36	−17,24	21	13

Removal of iodinated contrast media was low, and these contrast media were the APIs present in highest concentrations in the pilot plant effluent with concentrations up to 540 μg/l. The concentrations of clindamycin, phenanzon, amidotrizoeacid, sotalol, furosemide and ifosfamide are higher in the effluent than in the influent. This is probably due to deconjugation during treatment.

Overall, the results of the analyses for API removal show that MBR treatment is sufficient to remove some substances, while further treatment is required in order to achieve sufficient removal of other substances. Table 2 shows the effect of the MBR treatment on the microbial content of hospital wastewater. Data are shown for *Escherichia coli*, total coliforms and total enterococci in both influent

Table 2 | Colony forming units (CFU) of *E. coli*, total coliforms and total enterococci per ml in three samples of influent and effluent of the MBR pilot plant operating with hospital wastewater taken May 20, 2011

	E. coli		Total coliforms		Total enterococci	
	Influent [CFU/ml]	Effluent [CFU/ml]	Influent [CFU/ml]	Effluent [CFU/ml]	Influent [CFU/ml]	Effluent [CFU/ml]
Sample 1	1,500	<5	150,000	27	43,000	180
Sample 2	4,500	<5	110,000	10	43,000	170
Sample 3	2,600	<5	100,000	18	37,000	63
Avg. reduction [%]	> 99.83		99.98		99.66	

and effluent of the MBR pilot plant. It can be seen that the removal rates are all above 99%. These removal rates are comparable to those achieved at well-functioning activated sludge wastewater treatment plants (Ottoson *et al.* 2006). Also, testing showed that the treated wastewater had no antibiotic effect on *E. coli* or *Staphylococcus aureus*.

Results of polishing technologies

The results of the laboratory tests with the four polishing technologies, ozonation, ozonation + hydrogen peroxide, oxidation with chlorine dioxide and activated carbon, applied on MBR-treated hospital wastewater are presented in Table 3.

The results presented for PAC are the results obtained using Calgon Carbon Filtrasorb F400. The results with Norit 830 W PAC were comparable or only marginally different from those obtained for Filtrasorb F400. For ozone, only results from the 10 and 20 min reaction times (82 and 156 mg O_3/l) are presented, and for ozone + H_2O_2, only results from the 5 and 15 min reaction times (130 mg O_3/l; 60 mg H_2O_2/l and 450 mg O_3/l; 200 mg H_2O_2/l) are presented. For activated carbon, the results of the batch experiments with 150 and 450 mg/l PAC are presented.

Predicted no-effect concentration (PNEC) and Environmental Quality Standard (EQS) values found from literature are listed for most of the APIs that were analyzed. Measured concentrations exceeding PNEC and EQS values are marked with bold text. The results for chlorine dioxide are based on analyses made at Umeå University, and therefore the results are limited to a smaller number of APIs.

As seen in Table 3, ozone (10 min/82 mg O_3/l), ozone + H_2O_2 (5 min (130 mg O_3/l; 60 mg H_2O_2/l) and 15 min (450 mg O_3/l; 200 mg H_2O_2/l)) and chlorine dioxide (15 and 120 min) all have effluent concentrations above PNEC/EQS values for at least two APIs. Ozone (20 min/156 mg O_3/l) and PAC (150 and 450 mg/l) do not have any effluent API concentrations above the listed PNEC/EQS values. In general, the results for chlorine dioxide show poor removal compared to the other polishing technologies. It is not possible to assess the removal of iodinated contrast media by chlorine dioxide due to limited data, but removal of iodinated contrast media is generally poor for the three other polishing technologies. The largest reduction in iodinated contrast media concentrations is seen for PAC.

The estimated costs for the polishing technologies are presented in Table 4. At €0.22/m^3, the least expensive polishing option is ozonation for 10 min/82 mg O_3/l. The most expensive polishing options tested are PAC (450 mg/l) and ozone plus hydrogen peroxide with estimated costs of €1.06 and €1.08/m^3, respectively.

DISCUSSION

For many APIs, MBR treatment is an effective treatment that can reduce concentrations to below PNEC/EQS values. However, the results from the MBR pilot plant showed that for some APIs such as azithromycin, ciprofloxacine, ibuprofen, sulfamethoxazole and tramadol, concentrations in MBR-treated wastewater were still above existing PNEC values. Also, MBR removal of iodinated contrast media was low, resulting in high effluent concentrations of these persistent APIs.

The four tested polishing technologies can be used in combination with MBR treatment in order to further reduce the concentrations of those APIs that were not removed sufficiently during MBR treatment. Results from the laboratory-scale experiments with ozone, ozone + H_2O_2, PAC and chlorine dioxide allow for the selection of a best available polishing technology. It is, however, important to keep the uncertainties of the results in mind, especially because many of the studied API concentrations are in the low ng/l range. Also, there are APIs that were not analyzed for and for which removal is therefore unknown. The fact that the experiments were performed at laboratory scale may also mean that slightly different results may be achieved if the polishing technologies are scaled up to full-scale implementation.

The comparison of the available API removal data and treatment costs showed that ozone (20 min/156 mg O_3/l) was the best available polishing technology. PAC (450 mg/l) provided lower effluent concentrations than ozone (20 min/156 mg O_3/l) for a number of APIs, but with a treatment cost of €1.06/m^{3}compared to €0.40/m^3 it was decided that the improved performance did not outweigh the increased costs. It is estimated that MBR treatment combined with ozonation at 156 mg O_3/l will have an investment cost of approximately €1.6 mill. and an operating cost of approximately €1/m^3 for a hospital with around 900 beds.

The high API and bacteria removal rates that can be achieved by combining MBR and ozone treatment mean that there is a potential for utilizing the treated hospital wastewater. In Denmark advanced local treatment of hospital wastewater is now starting to be recognized as a win-win

Table 3 | Concentrations in ng/l of different APIs in the effluent of the MBR pilot plant as well as after application of ozone, ozone + H_2O_2, PAC and chlorine dioxide. Predicted no-effect concentrations (PNEC) and Environmental Quality Standards (EQS) are presented for comparison

		Ozone		Ozone/H_2O_2		PAC		Chlorine dioxide			
API	Outlet from MBR	82 mg O_3/l 10 min	156 mg O_3/l 20 min	130 mg O_3/l; 60 mg H_2O_2/l 5 min	450 mg O_3/l; 200 mg H_2O_2/l 15 min	150 mg/l	450 mg/l	60 mg/l 15 min	60 mg/l 120 min	PNEC[a]& EQS[b]	PNEC & EQS Reference
ac-sulfadiazine	80	<5	<5	55	16	<5	<5				
ac-sulfamethoxaz.	**1,100**	**780**	67	**1,900**	**990**	<5	<5			220[a]	DHI (2010)
Amoxicillin	<5	<5	<5	<5	<5	<5	<5				
Atenolol	1,500	<10	<10	250	27	<10	<10	15	16	77,700[a]	MST (2007)
Azithromycin	**1,100**	<5	<5	**37**	<5	<5	<5	430	220	9.4[a]	FASS (2011)
Bisoprolol	34	7.9	6.3	11	8.7	<5	6	20	19	35,600[a]	FASS (2011)
Capecitabine	16		<10	<10	<10	<10	<10			580[a]	FASS (2011)
Carbamazepine	2,200	<5	<5	14	<5	<5	5.8	2,100	1,800	2,500[a]	MST (2007)
Cefuroxime	<25	<25	<25	<25	<25	<25	<25				
Ciprofloxacine	**4,800**	**650**	250	**1,400**	230	36	<5	**113,000**	**32,000**	300[a]	DHI (2010)
Citalopram	**750**	45	27	36	<5	<5	<5	220	210	510[a]	MST (2007)
Clarithromycin	920	**51**	12	30	5.3	<5	<5	410	390	18,700[a]	FASS (2011)
Clindamycin	130	16	<5	<5	<5	<5	<5	4	3	3,600[a]	SFT (2009)
Cyclophosphamide	19	9.7	<5	6.6	<5	<5	<5			1,120,000[a]	SFT (2006)
Diclofenac	<5	51	<5	<5	<5	<5	<5	1	5	10[b]	EU (2012)
Erythromycin	540	<20	<20	<20	<20	<20	<20			2,000[a]	MST (2007)
Erythromycin deh.	780	<20	<20	<20	<20	<20	<20			2,000[a]	MST (2007)
Fenofibrat	13	<12.5	<12.5	<12.5	<12.5	<12.5	<12.5				
Furosemide	5,400	<25	<25	30	<25	<25	<25			45,140[a]	SFT (2009)
Ibuprofen	**1,100**	190	12	550	50	<1	<1			700[a]	DHI (2010)
Ifosfamide	120	35	8.6	14	<5	<5	<5			162,000[a]	FASS (2011)
Metoprolol	1,200	31	17	440	20	<5	8.7	1,900	1,900	8,800[a]	MST (2007)
MTX	<2	<2	<2	<2	<2	<2	<2				
Oxcarbazepine	<1	<1	<1	<1	<1	<1	<1				
Paracetamol	<1	<1	<1	<1	<1	<1	<1				
Phenanzon	190	<10	<10	<10	<10	<10	<10				
Propranolol	**300**	<2	<2	<2	<2	<2	<2			5[a]	FASS (2011)
Roxithromycin	180	16	7.9	6.1	<5	<5	<5	28	22	100,000[a]	FASS (2011)

(*continued*)

Table 3 | continued

API	Outlet from MBR	Ozone		Ozone/H_2O_2		PAC		Chlorine dioxide		PNEC[a] & EQS[b]	PNEC & EQS Reference
		82 mg O_3/l	156 mg O_3/l	130 mg O_3/l; 60 mg H_2O_2/l	450 mg O_3/l; 200 mg H_2O_2/l	150 mg/l	450 mg/l	60 mg/l	60 mg/l		
		10 min	20 min	5 min	15 min			15 min	120 min		
Sotalol	300	<5	<5	<5	<5	<5	<5	4	2		
Sulfadiazine	23	6.1	<5	54	24	5	<5				
Sulfamethizole	45	71	46	290	270	16	<10			800[a]	DHI (2010)
Sulfamethoxazole	**680**	**510**	190	**1,600**	**970**	200	<10	**520**	**660**	220[a]	DHI (2010)
Tramadol	**1,200**	38	22	390	7.9	<5	5.3	860	440	5,000[a]	MST (2011)
Trimethoprim	3,200	24	9.5	37	<5	11	<5	2,100	2,200	10,000[b]	MST (2010)
Venlafaxin	550	19	8.7	140	8.3	<5	<5	250	170	900[a]	SFT (2009)
Amidotrizoeacid	65,500	49,000	39,000	41,000	35,000	15,000	210				
Iohexol	550,000	620,000	110,000	500,000	130,000	51,000	520				
Iopromide	1,000,000	660,000	53,000	560,000	140,000	12,000	140				
Ioversol	330,000	420,000	50,000	270,000	67,000	16,000	310				

Table 4 | Estimated treatment costs per cubic meter in Euro for the polishing technologies tested following MBR treatment. The costs are additional to MBR treatment

Polishing technology		Operational costs [€/m^3]
Ozone	10 min (82 mg O_3/l)	0.22
	20 min (156 mg O_3/l)	0.40
Ozone + H_2O_2	5 min (130 mg O_3/l; 60 mg H_2O_2/l)	0.34
	15 min (450 mg O_3/l; 200 mg H_2O_2/l)	1.08
PAC	150 mg/l	0.31
	450 mg/l	1.06
Chlorine dioxide	120 min (60 mg ClO_2/l)	0.30

situation for future hospitals (Danish Nature Agency 2011). After effective treatment a pollution problem is solved, and at the same time the treated water could be reused for purposes such as irrigation and technical water or discharged to the local water area reducing the load on the drainage network and municipal treatment plant. Calculations on savings from not paying sewer drainage taxes (€2.7/m^3) shows that a MBR + O_3 plant in Copenhagen will have a payback period of 4–5 years.

CONCLUSIONS

The MBR pilot plant tested during the study showed effective removal of many APIs. Hormones and commonly used painkillers such as ibuprofen and paracetamol were removed to a large extent. MBR pilot plant results also showed that antibiotics, central nervous system APIs and cardiovascular APIs were removed to a lesser extent. The removal of iodinated contrast media was poor with high effluent concentrations (up to 540 µg/l). A significant reduction in the concentration of bacteria was recorded after MBR treatment, with removal rates for total coliforms and total enterococci of 99.98 and 99.66%, respectively.

Laboratory testing showed that effective removal of the vast majority of APIs can be achieved by MBR followed by polishing with ozone, ozone + H_2O_2 or PAC. Polishing with chlorine dioxide was significantly less effective. MBR treatment followed by ozonation at 156 mg O_3/l yielded effluent API concentrations below available PNEC and EQS values. Polishing with PAC also yielded effluent API concentrations below PNEC and EQS values, but due to

the higher cost of PAC (€1.06/m^3 compared to €0.40/m^3 at 156 mg O_3/l), ozonation at 156 mg O_3/l was selected as the best available polishing technology. It is estimated that the investment cost for an MBR and ozone treatment system designed for a hospital with 900 beds will be approximately €1.6 mill. and operating costs will be approximately €1/m^3.

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