Membrane Bioreactor Processes to Meet Todays and Future Municipal Sewage Treatment Requirements?

Christian Baresel*, Klara Westling1, Oscar Samuelsson1, Sofia Andersson1, Hugo Royen1, Sofia Andersson2 and Niklas Dahlén2

1IVL Swedish Environmental Research Institute AB, Valhallavägen 81, 100 31 Stockholm, Sweden
2Stockholm Vatten och Avfall VA AB, 106 36 Stockholm, Sweden

*Corresponding author: Christian Baresel, IVL Swedish Environmental Research Institute AB, Valhallavägen 81, 100 31 Stockholm, Sweden, E-mail: christian.baresel@ivl.se

Abstract

The Membrane Bioreactor (MBR) technology is evaluated concerning central municipal sewage treatment aspects including nutrient removal, removal of micropollutants (MP) and emissions of greenhouse gases (GHG). Pilot-scale experiments in preparation for the world largest MBR process in Stockholm, Sweden show that

- Targeted effluent qualities could be achieved under various loads,
- Lower direct GHG emissions from the treatment compared to traditional treatment processes were observed,
- The investigated MBR process provided an efficient removal of microplastics and bacteria, and
- If combined with additional complementary treatment an effective removal of pharmaceutical residues and other micropollutants.

Keywords: Membrane Bioreactor; Sewage treatment; Nutrient removal; Ultrafiltration; Greenhouse gas emissions; Emerging pollutant; Micropollutants; Pharmaceutical residues; Ozonation; Activated carbon; Microplastics

Introduction

Several municipal wastewater treatment plants (WWTPs) in Stockholm, Sweden, will within the near future face both an increased load due to a growing population as well as more stringent effluent quality requirements. The latter mainly regarding nutrients due to Sweden's commitment to the Baltic Sea Action Plan and the implementation of the European water framework directive (WFD). In addition, removal of emerging substances such as pharmaceutical residues, micro plastics and antibiotic resistance are gaining more attention since WWTP effluent is the most or one of the most significant sources of such loads to the environment [1-4]. Pharmaceutical residues and other emerging substances are generally not efficiently removed in conventional WWTPs [5]. The WFD has defined a list of prioritized substances including pesticides, biocides, flame-retardants and metals [6], which already today require monitoring and treatment. Several other substances, including some pharmaceuticals, are on the 'watch list' of emerging pollutants that may be placed on the WFD priority list. Requirements for additional treatment, in larger WWTPs, for the reduction of some pharmaceutical residues and other micropollutants (MP) could thus be expected, also in other countries than Switzerland, where such a regulation is already in place. The potential negative effects on aquatic organisms, the aquatic food-web and higher organisms, as well as the risk of increased numbers of antibiotic resistant genes in bacteria, all present a threat to our environment, health and society [7-9]. Another increasing concern for wastewater treatment are emissions of greenhouse gases (GHGs). At WWTPs, special attention is given to nitrous oxide (N₂O), which is a highly potent GHG (298 times more potent than carbon dioxide (CO₂) [10]). At incomplete nitrification and denitrification N₂O can be emitted, which may cause a significant negative overall environmental impact of the treatment process [11,12].

Even though regulations may earliest come in place in some years from now, many WWTPs actively work on reducing GHG emissions from wastewater treatment processes. Besides the requirements to increase capacity, improve treatment efficiency and reduce GHG emissions, many WWTPs also face the problem that they cannot expand spatially as they are located in densely populated areas or underground.

New solutions for space-efficient, high-capacity and flexible municipal wastewater treatment processes are thus required. Stockholm Water and Waste Company (Stockholm Vatten och Avfall), Sweden's largest water service organization, is directly facing the above problems of space limitation, increased capacity need and stricter effluent requirements at the Henriksdal WWTP in Stockholm. As a result, the existing conventional activated sludge process (CAS) will be converted to a Membrane Bioreactor (MBR), doubling the capacity by using existing process volumes only. The new process will be the world's largest MBR facility with a capacity of 1.6 million PE (predicted load year 2040).

MBRs combine the biological activated sludge process with membrane separation, which provide distinct advantages over the CAS. Advantages include a significantly better effluent (permeate) quality regarding particles, disinfection capabilities due to the membrane pore size, higher volumetric loading due to higher sludge concentrations in the biology, reduced footprint and process flexibility towards influent changes. Even the treatment of MP may be more efficient using MBRs compared to traditional treatment systems. This is partly explained by the fact that MP attached to particles can effectively be removed by filtration whereas dissolved MP can be degraded more effectively because of the higher biological activity in a MBR process. In addition, a more efficient polishing treatment compared to CAS can be achieved [11,13-18].

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Drawbacks of the process are the high energy use for aeration and the use of cleaning chemicals in the filtration step to curb fouling and scaling on the membrane surface, which reduces the permeability of the membranes.

MBRs have been used for a number of decades but only in the last decade, MBRs gained more attention for the treatment of both municipal and industrial wastewater. This is mainly due to a significant cost reduction of membranes and process development decreasing energy requirements [19-23].

The aim of this research work is to investigate the MBR technology concerning the overall holism and resource efficiency towards some of the most central treatment aspects including nutrient removal, removal of micropollutants and minimizing of GHG emissions. Through actual pilot-scale experiments, the paper describes the performance of the studied system under various test periods defined to present and future requirements of the growing region of Stockholm, Sweden.

Methods
Pilot characteristics

IVL Swedish Environmental Research Institute and Stockholm Water and Waste Company have together setup and since September 2013 operated a pilot-scale treatment line with a capacity corresponding to 0.015% of the total Henriksdal WWTP facility (design year 2040). The pilot-scale treatment line (Figure 1) was constructed as a copy of the future treatment line at Henriksdal WWTP and located at the R&D facility Hammarby Sjöstadsverk (www.hammarbysjostadsverk.se, part of the Swedish Water Innovation Center in Stockholm).

The influent to the pilot is taken from the untreated inflow to the Henriksdal WWTP and filtered through a 3 mm strainer. The flow into the pilot is proportional to the flow to the main WWTP. The pilot consists of an aerated pre-pretreatment tank, a primary clarifier, a biological reactor with a total volume of about 29m³ that is divided into anoxic and aerobic zones, followed by an ultra-filtration (UF) tank of 13.2m². Nitrate is recirculated from the beginning of the pre-denitrification zone, and sludge is recirculated from the UF tank to the beginning of the pre-denitrification zone. A separate de-aeration tank aiming to reduce the oxygen concentration in the return sludge by nitrification/respiration is further used (RAS DeOx). Supernatant from sludge dewatering is added to this step. The ultrafiltration consists of two modules with Flat Sheet membrane type MFM 100 from Alfa Laval (Denmark). The UF units are operated intermittently with relaxation times of 2 minutes after 10 minutes of operation. The nominal pore size is 0.2 microns with a minimum and maximum pore size of 0.17 microns and 0.26 microns, respectively. The total membrane area per module is 79.64m² spread over 44 membrane sheets.

The TransMembrane Pressure (TMP) control strategy was applied for membrane operation. When the permeability decreased about 30% from its initial value, cleaning of the membranes (Clean-In-Place, CIP) was performed according to supplier requirements. Sodium hypochlorite was used for removal of organic coatings and oxalic acid for the removal of inorganic coatings.

Sampling

Sampling varied during the pilot study depending on evaluation focus. However, daily and weekly composite samples were collected in the influent, after the primary clarifier and in the effluent. Grab samples were collected in the biological reactors and the UF tank. Standard parameters analyzed included total organic carbon (TOC), biological oxygen demand after 7 days (BOD₇), total phosphorous (TP), phosphorus-phosphorous (PO₄-P), suspended solids (SS), volatile suspended solids (VSS), total dissolved solids (TDS), ammonium-nitrogen (NH₄-N), nitrate-nitrogen (NO₃-N), nitrite-nitrogen (NO₂-N), total nitrogen (TN), digested iron (Fe), digested P in sludge and temperature (T). Online measurements for PO₄-P, NO₃-N, NH₄-N, dissolved oxygen (DO), SS, pH, redox, water and air flow, temperature, pressure and water level were used at several locations in the pilot for both process monitoring and control.

Tests periods

During the first year of operation (Oct 2013-Aug 2014), the study focus was to reach targeted effluent concentrations of nitrogen (6 mg TN/L) and phosphorus (0.2 mg TP/L) at different loading and dosing conditions divided into four different test periods (P1-P4, see Table 1). During the second year of operation (Sep 2014-Nov 2015), a larger focus was given to optimizing the overall treatment efficiency of the system and specifically the phosphorous removal. The test periods of the second year (P5-P17) were based on different control strategies for dosing of precipitation chemical. During P1-P9, Sodium Acetate (NaOAc) was used as external carbon source for post-denitrification. From test period P10 and onwards, the proprietary blend Brenntaplus was used. Table 1 below and Table S1 in the supporting information provide detailed information about the test periods.

Oflgas emissions

To investigate N₂O emissions from the MBR process, two screening campaigns of the various process steps including primary clarifier, biological reactors and UF tank were conducted; the first without and the second with addition of reject water from sludge dewatering to the RAS DeOx. The pilot was operated with the same conditions under both campaigns (test period P13). Each reactor was covered and all process off gas was measured and analyzed by Teledyne analytical instrument (Model GFC-7002E). Figure S1 in the supporting information provides a general
### Table 1: Overview of the 17 test periods (P) and their characteristics (mean values)

<table>
<thead>
<tr>
<th>Test period (P)</th>
<th>Week/Year</th>
<th>Flow (^1) [m(^3)/h]</th>
<th>Organic load (^1) [mg/L]</th>
<th>Flux [L/(m(^2)*h)]</th>
<th>Sludge content [mg/L]</th>
<th>Dosing Carbon (^2)</th>
<th>Dosing P-removal Amount [mg/L]</th>
<th>Location (^2)</th>
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<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>P1</td>
<td>50/13-13/14</td>
<td>2.5 (C)</td>
<td>3.2</td>
<td>15.7</td>
<td>3500-6000</td>
<td>5-15</td>
<td>6-12 FeSO(_4)</td>
<td>1Q</td>
</tr>
<tr>
<td>P2</td>
<td>14-21/14</td>
<td>2.5 (D)</td>
<td>3.2</td>
<td>15.7</td>
<td>4500-6000</td>
<td>15</td>
<td>12 FeSO(_4)</td>
<td>1Q</td>
</tr>
<tr>
<td>P3</td>
<td>22-27/14</td>
<td>4.3 (D)</td>
<td>5.5</td>
<td>27.0</td>
<td>8000</td>
<td>30</td>
<td>20 FeSO(_4)</td>
<td>1Q</td>
</tr>
<tr>
<td>P4</td>
<td>28-36/14</td>
<td>2.75 (D)</td>
<td>3.5</td>
<td>17.2</td>
<td>6000</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>P5</td>
<td>39-44/14</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>18.1</td>
<td>5000</td>
<td>-</td>
<td>20 FeCl(_3)</td>
<td>1Q</td>
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<tr>
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<td>45-50/14</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>16.0</td>
<td>5000</td>
<td>50</td>
<td>30 FeSO(_4)</td>
<td>1F</td>
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<tr>
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<td>51/14-03/15</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>16.2</td>
<td>5000</td>
<td>45</td>
<td>20 FeSO(_4)</td>
<td>4Q</td>
</tr>
<tr>
<td>P8</td>
<td>04-09/15</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>17.8</td>
<td>5500</td>
<td>55</td>
<td>12 FeSO(_4)</td>
<td>1 &amp; 3Q</td>
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<td>P9</td>
<td>10-13/15</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>15.1</td>
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<td>1Q</td>
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<tr>
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<td>14-15/15</td>
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<td>3.6</td>
<td>16.5</td>
<td>5500</td>
<td>80</td>
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<tr>
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<td>50</td>
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<tr>
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<td>19-23/15</td>
<td>2.8 (D)</td>
<td>3.6</td>
<td>17.5</td>
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<td>55</td>
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<td>2F</td>
</tr>
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<td>3.6</td>
<td>15.4</td>
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<td>30</td>
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<td>3P</td>
</tr>
<tr>
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<td>2.8 (D)</td>
<td>3.6</td>
<td>12.5</td>
<td>5500</td>
<td>8</td>
<td>14 FeSO(_4)</td>
<td>2F &amp; 3P</td>
</tr>
<tr>
<td>P15(^a)</td>
<td>34-36/15</td>
<td>3.2 (D)</td>
<td>4.0</td>
<td>13.7</td>
<td>5500</td>
<td>55</td>
<td>10 FeSO(_4)</td>
<td>1F</td>
</tr>
<tr>
<td>P16</td>
<td>37-38/15</td>
<td>3.2 (D)</td>
<td>4.0</td>
<td>18.4</td>
<td>5500</td>
<td>60</td>
<td>8 FeSO(_4)</td>
<td>2F &amp; 2P</td>
</tr>
<tr>
<td>P17</td>
<td>39-45/15</td>
<td>3.2 (D)</td>
<td>4.0</td>
<td>18.0</td>
<td>5500</td>
<td>-</td>
<td>18 FeSO(_4)</td>
<td>1F &amp; 2P</td>
</tr>
</tbody>
</table>

\(^1\) C - constant flow, D - dynamic flow, controlled by flow signal to full-scale WWTP Henriksdal
\(^2\) See Figure 1 for location, F = fixed dose, Q = flow proportional, P = proportional to effluent phosphorus concentration
\(^a\) Tank modification of the RAS/DeOx allowed for a higher load

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The pilot study included investigation of the removal efficiency of various micropollutants in the MBR process with or without the following complementary steps: ozonation and BAf(GAC), a biological active filter with granulated active carbon. Ozone tests were performed during a series of 3-4 days with sampling after three retention times at each configuration change. BAf(GAC) tests were carried out during 20 months with weekly composite sampling. Some of the daily composite samples for process operation of the pilot were collected during various periods during one year for analyses of microplastics. Investigated micropollutants included a wide range of relevant pharmaceuticals and other emerging substances, estrogen effect, bacteria, and microplastics (see Table S1 for details about investigated substances). Standards and analytical methods have been developed for analyses except for microplastics that were analyzed according to a method described by Magnusson et al. [25]. The supporting information provides more details including a schematic layout of the pilot setup for the ozonation and BAf(GAC) tests.

**Other tests**

The pilot study included a number of other activities related to the overall treatment performance of the MBR process. This included mapping of the sludge microflora in the UF tank and comparison with the conventional active sludge process in the full-scale Henriksdal WWTP using the MiDAS protocol [26,27]. Further, a simulation model of the MBR-pilot based on the activated sludge Model No. 1, ASM1 [28] was established and used to test and evaluate various operational changes before implementation.

**Results and discussions**

The results show that the process configuration was capable to meet targeted removal requirements for both nitrogen and phosphorus (Figures 2 and 3). Due to various tests plans, load cases but also occasional disruptions in the operation, the reduction as presented in the Figures was not below desired target levels 100% of the time. Average effluent concentrations were 4.2 mg TN/L and 0.24 mg TP/L for year 2013, 4.1 mg TN/L and 0.24 mg TP/L for year 2014 and 4.6 mg TN/L and 0.26 mg TP/L for year 2015.

Between weeks 44-49/2013, no precipitation chemical was added with elevated TP-effluent concentrations as a result. Addition of precipitation chemical started with low doses and the dose was successively increased until satisfactory effluent concentrations were reached. Increased TP-effluent concentrations between weeks 03-09/2013 were due to trials with low chemical dosage.

Addition of reject water to the treatment line was started from week 46/2014 and caused an approximately 10% increase in the total nitrogen load and temporarily increased effluent concentrations. Influent samples were collected prior to reject side-stream addition. After week 49/2014, adjustment in the carbon dosing control to consider the addition nitrogen load caused by the reject water achieved again lower effluent concentrations. Increased effluent concentrations during weeks 15-21/2015 were due to trials with no addition of carbon source.

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with reject water addition, respectively. Emissions were also high in the second aerated zone and nonylphenol was removed as well by Membrane B. It is also lower than internationally has been evaluated for the same substances Membrane permeability during the whole project period. Limit by both technologies. Most investigated substances could be removed by more than 90% over the size at the R&D-facility and several full-scale nitrogen removing WWTPs in the Stockholm region that were measured to emit between 0.13-1.2% of the influent nitrogen as N\textsubscript{2}O. It is also lower than internationally reported N\textsubscript{2}O emissions varying between 0.8% and 6.5% [30-32]. An explanation for these low emissions compared to conventional process configuration remains yet to be determined and for this reason, new measuring campaigns are planned. However, the increased biological activity in the MBR process compared to the other studied systems may be responsible to some part for the lower emissions.

**Treatment of micropollutants**

The MBR process provides a high-quality, particle-free effluent compared to traditional activated sludge processes. Bacteria, including multiresistant bacteria, of all sizes larger than the membrane pore size are efficiently removed from the wastewater by the MBR process. However, very low concentrations (<65 cfu/100 mL) of bacteria were still detected in the MBR effluent but it could not be determined if these bacteria originated from sample contamination or contact of the permeate with the atmosphere. Both aspects are almost impossible to avoid in sewage treatment environments. Not a single microplastic particle was detected in the MBR effluent (removal efficiency 100%), whereas effluent water from the full-scale CAS process including a final sand filtration contained both plastic fibres and plastic fragments (removal efficiency 90.7%). Non-synthetic fibres were found in both MBR and CAS effluents. Analyses of pharmaceutical residues in the MBR effluent showed similar levels as in the full-scale CAS effluent (except for amlodipine and sertraline that were reduced to a somewhat higher extent in the MBR process). This indicated no increased removal effect of pharmaceuticals by the MBR process compared to the CAS process as suggested in other studies [14,16].

The complementary treatment of MBR effluent with ozonation or a biological active filter has been evaluated for the same substances and estrogen effect (see Table S1 in the supporting information). All investigated substances could be removed by more than 90% over the complementary treatment step only. In addition, the studied phenolic compounds triclosan and bisphenol A were reduced to below the detection limit by both technologies. Most nonylphenol was removed as well by both ozonation and BAF(GAC), while only the BAF(GAC) worked well for octylphenol. Total coliforms in the treated MBR effluent were reduced with about 80% by ozonation and >85% by the BAF(GAC). Interestingly, Article 12 recommended N\textsubscript{2}O to be measured in waste streams with a high flow rate and high N\textsubscript{2}O concentration. However, this is not always possible. N\textsubscript{2}O emissions vary between 0.8% and 6.5% [30-32]. An explanation for these low emissions compared to conventional process configuration remains yet to be determined and for this reason, new measuring campaigns are planned. However, the increased biological activity in the MBR process compared to the other studied systems may be responsible to some part for the lower emissions.

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fecal coliform removal was absent in the BAF(GAC) during the first weeks of operation while a reduction of more than 90% was achieved after 3 months of operation. This might be explained by the establishment of a biology in the filter that outcompetes fecal coliforms. Ozonation could only reduce fecal coliforms at higher ozone doses of >9 g O₂/m³.

Compared to similar complimentary treatment of CAS effluent (i.e. the same influent water, see e.g. Baresel et al. [33] and Ek et al. [34]), lower ozone doses were required to achieve a high reduction of persistent substances during ozonation, and a significant reduction of clogging and backwash frequency was achieved in the BAF(GAC) when treating MBR effluent. Both aspects have a direct impact on the operational cost of the advanced treatment of effluents. Both aspects are also related to the higher quality of MBR effluent compared to traditional CAS effluent, even with sand filtration. The long-term evaluation (2 years) of the biological filter is still ongoing and the final evaluation remains to be done. However, the filter capacity was maintained even after 20 000 Empty bed volumes without the need of GAC replacements. The removal efficiencies of ozonation and BAF(GAC) were also compared with reverse osmosis (RO) which was performed in parallel experiments at the R&D-facility. More details are provided by Baresel et al. [13].

Other relevant tests

The microbial population composition and dynamics in activated sludge from the MBR process was significantly different compared to the full-scale CAS process at Henriksdal WWTP [35]. The mapping showed that it might be a useful tool to understand process changes and for operation control in the future.

Model simulations showed a decrease in carbon source consumption and overall improvement of nitrogen removal if the nitrate recirculation was moved to a location earlier in the post-denitrification (before the dosing point for external carbon). Based on the simulation results the pilot operation was changed.

Conclusions

The evaluation of the MBR process shows that targeted effluent qualities of <0.2 mg TP/L and <6 mg TN/L could be achieved under various loads. However, this may require relatively high precipitation chemical and external carbon doses at maximum load conditions.

The tested membranes showed a high average permeability and complete removal of particles throughout the whole test period. The UF- membranes showed increased fouling at high doses of precipitation chemicals, especially if the dosing point was near to or in the UF tank. A precipitation chemical dosage control strategy based on the effluent phosphate concentration was identified as favorable to achieve stable low phosphorus concentrations in the effluent without risking excessive fouling of the membranes.

The results further indicate that the studied MBR process had a lower direct GHG impact than other traditional treatment processes as the observed amount of nitrous oxide emissions were lower than what has been reported for CAS processes.

The MBR process provides an efficient removal of microplastics and bacteria due to the integrated UF. A complementary treatment of the MBR effluent by ozonation or biological active filter with activated carbon as filter material can provide an effective removal of pharmaceutical residues and other micropollutants with less effort than efforts compared to comparable treatment of CAS effluent.

In general, the project results show that the MBR process provides a flexibility to meet various demands for efficient sewage treatment to low effluent concentrations of organics, nutrients, suspended solids and micropollutants. The advanced biological process configuration in combination with an effective membrane separation as the main treatment process produces a high-quality effluent that can effectively be upgraded by additional polishing steps.

Outlook

The project is still on-going. Investigating a number of issues including increased treatment and resource efficiency, reduced use of chemicals and energy and comparison between the flat-sheet membranes used in this study and hollow-fibre membranes. The formation and potential problems of chlorinated organic contaminants (measured as AOX (Adsorbable Organic Halides) and EOX (Extractable Organic Halides)) and other environmental impacts of membranes operation and cleaning will be investigated.

A longer nitrous oxide emissions measurement campaign is planned to further investigate the positive results presented here. This campaign will also include N₂O in the water phase with a novel online probe [36] in order to quantify the N₂O production and consumption dynamics.

Further, more tests with resource-efficient removal of micropollutants and a holistic integration of various complementary treatment steps into the MBR process will be investigated. This includes, for example, tailor-made enzyme filters for polishing of the MBR effluent.

The findings from the mapping of the microbial population indicate that the properties of the MBR sludge differ from the CAS sludge, which may affect the sludge handling, i.e. thickening, dewatering and digestion. To establish if such conclusions can be drawn, the sludge handling will be investigated in pilot-scale and linked to the microbial population composition in further studies.

References


Supporting Information

**Micropolutants**

**Table S1: Investigated micropolutants and effects.**

<table>
<thead>
<tr>
<th>No.</th>
<th>Substance</th>
<th>Pharmaceutical/ Characteristics</th>
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<td>Bisfenol A, triclosan, nonylfenol, oktfenol, etc.</td>
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The BAF(GAC) was tested in a pilot filter (diameter 62 cm) with automatic backwash when the filter bed started to clog (control level was set to 45 cm above the coal bed). The filter consisted of a 10 cm thick gravel/sand bed at the bottom and a 1 m layer of commercial granulated carbon (Filtrasorb 400, Chemviron Carbon, density ~ 0.5 kg/L). The contact time in the bed (EBCT) was about 14 minutes, which was based on earlier tests with various effluents [1,2]. The water passed through the filter was collected in an equalization tank for backwash. Backwash consisted of a sequence of pulses of pressurized air to terminate any pressed layers and backwash with water from the equalization tank.

**Figure S1:** General setup of the off-gas measurements in the various reactors.

**Figure S2:** Schematic illustration of the pilot setup with tests for ozonation and BAF(GAC) - biological active filter with activated carbon as filter material. Ozonation was tested at doses between 3 and 13 g O₃/m³ at a DOC of 10 mg/L using a Wedeco unit (Modular HC8) with oxygen production from air (pressure swing method), an ozone generator, a reactor and a degassing chamber. The ozone generator capacity was 8 g ozone/h (16 g ozone/m³) at an average residence time of 20 minutes). The reaction vessels consisted of two columns in series with upstream flow, where ozone was added via a diffuser at the bottom. Each column had a volume of 115 liters and a height of 4.2 meters. Degassing was accomplished via a simple overflow with air extractors. In some experiments, only the second column was used at a DOC of 3 mg/L. The column had a flow of 700 L/h, which gave a residence time (HRT) of about 10 minutes in the column. At the highest dose, 13 g of ozone/m³, the flow was reduced to 550 L/h for the production of ozone through the ozone generator would suffice. The contact time then became 13.6 minutes.

**References**

