Sustainable treatment systems for removal of pharmaceutical residues and other priority persistent substances


ABSTRACT

Pharmaceutical residues and other emerging substances commonly summarised as micropollutants pass through wastewater treatment plants (WWTPs) and end up in the receiving waters and sludge. Many studies have investigated the removal efficiency of various techniques but a holistic evaluation of various relevant treatment alternatives regarding both the removal efficiency for various micropollutants, investment and operating costs, environmental impacts and future comprehensiveness is still lacking. This paper provides the results from a large 3-year project about the evaluation of sustainable treatment systems for removal of various micropollutants or disruptive effects at Swedish WWTPs and their environmental, economic and future sustainability. The presented results are based on our own pilot tests and related assessment and modelling efforts and provide a holistic view on advanced treatment of wastewater for removal of micropollutants.

Key words | GAC-biofilter, micropollutants, ozonation, pharmaceuticals, wastewater treatment

INTRODUCTION

Pharmaceutical residues and other emerging substances commonly summarised as micropollutants pass through modern wastewater treatment plants (WWTPs) and end up in the receiving waters and sludge. Already in the first studies, recipient concentrations detected with expected effects on aquatic organisms have raised concerns (Kim et al. 2007; Deblonde et al. 2011; Fick et al. 2011; Brodin et al. 2013; Vasquez et al. 2014). Chemicals released via WWTPs may also enter the aquatic food web and cause effects in higher organisms such as fish-eating birds or mammals including humans. Studies have also shown that antibiotics in the environment may contribute to the increase of antibiotic resistant genes in bacteria, which is a serious threat to our possibility to cure life-threatening diseases on the global scale (Gullberg et al. 2011; WHO 2014).

As current WWTPs are usually not able to remove microbial stable chemical pollutants, several various treatment technologies have been proposed and evaluated in several large projects, such as REMPHAWATER (2003), POSEIDON (2004), RiSKWa (2013) and the Swedish MistraPharma. Especially in Germany and Switzerland, advanced treatment technologies have been tested on a large scale (e.g. Abegglen & Siegrist 2012; ARGE 2013). Also in Sweden, technologies have been tested (e.g. Ek et al. 2014; Magdeburg et al. 2014; Maus et al. 2014).
However, few studies have included micropollutants other than pharmaceuticals and personal care products (PPCPs). Furthermore, previous studies on specific techniques or summarizing review studies (e.g. Margot et al. 2015; Grandclément et al. 2017; Bourgin et al. 2018) mainly focused on removal efficiency of PPCPs, while a more holistic review including costs, environmental impacts, and comprehensiveness towards future potential adaptation for an increased resource efficiency and sustainability, is not provided.

This paper presents the results from a Swedish project about testing, evaluating and comparing sustainable treatment systems for removal of a wide range of micropollutants. The presented results, that are based on own pilot tests and related assessment and modelling efforts, will give a holistic view on how available and applicable treatment techniques/systems at WWTPs should be selected for a sustainable removal of pharmaceuticals and other prioritized compounds.

**MATERIAL AND METHODS**

The project started in 2014 with a thorough review of emerging contaminants of concern in Sweden and detected by various screenings in the effluent of WWTPs, available technologies for their removal and aspects not covered yet related to these technologies. This resulted in a list of micropollutants to be considered, their effects on recipients, sampling analyses strategies, selection of treatment technologies, and combinations to be further investigated. Selected micropollutants for further investigation, including pharmaceutical residues, microplastic, etc., are shown in Table S1 (Supporting Material, available with the online version of this paper) and the complete review is provided by Baresel et al. (2015).

Several pilot studies on specific technologies/combinations were performed including ozonation (O3), granulated activated carbon (GAC) biofilter, the combination of O3 and GAC-biofilter, and the combination ultrafiltration (UF) and GAC biofilter. Besides these techniques, there exist others such as advanced oxidation processes (AOP) applying UV-light in different combinations with, for example, hydrogen peroxide (H2O2) and titanium dioxide (TiO2). The initial evaluation of available technologies done in the project concluded that ozone alone can be considered at least as good as these combinations to remove a broad spectrum of compounds in a relatively simple process (Baresel et al. 2015). Therefore, techniques with a significantly higher cost and resource consumption have been excluded from further studies.

All treatment systems have been evaluated at the R&D-facility Hammarby Sjöstadsverk in Stockholm with municipal sewage from Stockholm city as the same wastewater characteristics for comparison. The main sewage treatment for the pilot test were conventional active sludge (CAS) and membrane bioreactor (MBR)-systems. For CAS the effluent of Stockholm’s existing WWTP Henriksdal and for MBR a pilot representing an exact copy of the future WWTP for Stockholm have been used, respectively. The only considered technology not investigated with pilot studies at the test site was the combination of powder activated carbon with UF. For this technology, results from pilot tests within the DEMEAU-project (www.demeau-fp7.eu) and pilot tests in Locle, Lausanne (RIBI SA Ingénieurs hydrauliciens, 2014) were used. Table 1 provides main applied operational conditions for the pilots, which agree with commonly reported conditions in the literature.

Based on the pilot test, several assessment studies for the various treatment options were carried out. These assessments included both environmental performance quantification, life cycle costs assessment, removal efficiency, evaluation, implementation aspects, and flexibility towards a more sustainable adaption in future. Here, also removal efficiencies that could not be measured in the actual test have been included in the assessments if relevant data from other studies were available. Finally, these different actions were combined in a holistic assessment of available treatment alternatives based either on single techniques or on combinations to facilitate implementation decisions made on sound and holistic ground for a short- and long-term sustainable choice.

Environmental impact assessment was evaluated by a life cycle assessment (LCA) according to the ISO standard (ISO 2006). All relevant resources needed to construct, operate and dismantle a plant including materials, energy and chemicals used throughout the plant’s life cycle were considered. Scale-up effects were considered by performing all analyses on two typical plant sizes: 20,000, 100,000 pe (personal equivalents corresponding to a theoretical biochemical oxygen demand (BOD)-load of 70 g/(p, d) and 150 m3/ (pe yr)) over 20 years of assumed plant life. The performed

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nominal pore size final UF [μm]</td>
<td>0.2</td>
</tr>
<tr>
<td>Nominal pore size UF as MBR [μm]</td>
<td>0.04</td>
</tr>
<tr>
<td>Ozone dosage [mg O3/g DOC]</td>
<td>0.5</td>
</tr>
<tr>
<td>Contact time ozonation [min]</td>
<td>15</td>
</tr>
<tr>
<td>Contact time GAC filter bed [min]</td>
<td>15</td>
</tr>
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LCA evaluated the most common environmental KPIs: global warming potential (GWP) [kg CO₂-Eq./m³], acidification potential (AP) [kg SO₂-Eq./m³], eutrophication potential (EP) [kg Phosphate-Eq./m³] and depletion of abiotic resources (AD) due to consumption of elements [kg Sb-Eq./m³] and fossil fuels [MJ/m³]. More details on methods can be found in the technical project description (Baresel et al. 2017a).

The economic evaluation was mainly based on five different plant sizes (2,000, 10,000, 20,000, 100,000, and 500,000) and comprised the calculation of the total annual treatment costs, including both capital expenses (CAPEX) and operating expenses (OPEX). CAPEX consists of civil, mechanical and electrical costs, including the cost of installation. It further accounts for replacing consumables. OPEX includes energy and chemical costs for operation, manpower and maintenance costs. CAPEX and OPEX were based on actual designs of full-scale plants of the various plant sizes and from a number of different technology and consultancy providers. These technology providers included most companies active on the Swedish market such as SUEZ/GE WATER, Xylem Inc., WEDECO, Christian Berner, WSP, Purac, Veolia, Ozonia (Veolia), Desotec, Primozone, Ramböll, etc. to obtain as true cost calculations as possible. Treatment costs per cubic meter of treated water (SEK/m³) were calculated by the sum of annual investment and operating costs divided by the total annual treated water flow in the wastewater treatment plant. More details on methods can be found in Baresel et al. (2017a).

RESULTS AND DISCUSSION

The result of the technology evaluation, including the review and actual testing activities, resulted in an overview of relevant treatment technologies (Figure 1). Some of these are ready to be implemented at WWTPs and have been chosen for further investigation, i.e. removal efficiency tests, LCA and life cycle costing (LCC) (dark coloured). The other technologies need some substantial improvement in terms of cost reduction (e.g. NF, RO and AOP) or further technology development (e.g. enzymes). UF and pulverised activated carbon (PAC) are not considered for further studies as they require an additional treatment step to achieve a significant
removal of micropollutants. This is for example the case in the combination PAC-UF and UF/GAC-biofilter. GAC is only considered as material in a biofilter as GAC-filter applied in wastewater treatment will act as such biofilter.

The initial review also revealed analytical problems with today’s methods, especially for the quantification of pharmaceuticals in wastewater. This has a significant impact on our understanding of actual concentrations in wastewater (which are underquantified to a significant extent) and thus on various removal efficiency evaluations as in the studies mentioned above. The knowledge gained and new methods developed are not presented here and the reader is referred to Magnér et al. (2017).

Both removal efficiencies for different types of micropollutants or disruptive effects including pharmaceuticals, antibiotic resistance, disinfection, estrogenic effects, microplastics and PFAS/PFOS (perfluorinated alkylated substances/perfluorooctanesulfonate) have been evaluated for the selected techniques/combinations as presented in Figure 1. The analytical techniques used to quantify micropollutants and detailed results from these actual tests, and associated assessment of the environmental and economic sustainability of the various treatment solutions and implementation aspects can be studied in the project reports. Baresel et al. (2017a, 2017b, 2017c) also summarised in a briefing report to the Swedish EPA for new regulations on micropollutant removal at Swedish WWTPs (Baresel et al. 2017d).

Figure 2 shows a condensed summary of the main findings of the comprehensive study. Results for all investigated plant sizes are combined but scale effects have to be considered. Both costs and environmental impacts decrease with increasing plant size. Energy and chemical use have been identified as main parameters determining the environmental impact. However, this impact may be generated onsite, e.g. during ozone production or membrane operation at WWTP, or remotely, e.g. during production and regeneration of activated carbon. Treatment cost per treated cubic meter of wastewater, as shown in the table below, indicates that additional treatment for removal of micropollutants would only imply minor extra cost for WWTPs compared
to current treatment costs of wastewater. While cost for alternatives including membranes are still more expensive than other techniques, both investment and operation cost for ultrafiltration have been decreasing significantly the past 10 years and are expected by the authors to decrease further.

One of the most significant results of the project is that the current focus on PPCPs may result in technology implementations at WWTPs that are only efficient in the short term. Ozonation has been identified as less efficient as activated carbon for some micropollutants (Table S1, available with the online version of this paper) but has long been pointed out as the most feasible solution for extra treatment of wastewater. This is mainly explained because ozonation at relevant ozone doses (0.5–1 mg O\textsubscript{3}/g DOC) can be achieved at relatively low costs and space requirements and the removal efficiency of most pharmaceutical residues is satisfactory. However, considering other sustainability aspects such as formation of toxic decomposition products and the removal of other micropollutants than just pharmaceuticals, other technologies appear to be more suitable (see Figure 2). Regarding the treatment cost, it also has to be considered that ozonation requires a biological polishing afterwards to remove the risk of the formation of toxic by products. The cost and environmental impacts of this extra treatment step are normally not included in evaluation of ozonation. From Figure 2 and Table S1 it becomes obvious that only the combination of several techniques can accomplish a broad removal of micropollutants. This, however, may imply higher treatment cost and environmental impacts that have to be considered in relation to reduced emission of micropollutants.

Another important outcome of the project is the evaluation of adaptability of various treatment techniques towards more sustainable operation considering technology advancements currently going on in order to reduce cost and environmental impacts of the additional treatment. While there is little room for improvements for ozonation except a better dosing control coupled to actual loads, other technologies/combinations have a significant potential for further advancements. For technologies comprising activated carbon (both GAC and PAC), advances in producing biochar from different organic wastes can significantly reduce both costs and environmental impacts. The project used for example sludge from the WWTPs to produce biochar as filter material, which would reduce the environmental impact and cost of this treatment system significantly. New membrane advancements that reduce both production costs and energy/chemical consumption, alternative system configurations, less harmful cleaning chemicals and an overall increased removal efficiency that also reduces subsequent polishing steps can further improve filtration technologies. Even though the improvement potential is more difficult to quantify, it is an important sustainability aspect for the further development and should be considered in the choice of treatment technology.

CONCLUSIONS

It is clearly shown that a sole focus on only one group of micropollutants may result in implementation of technologies that are not resource-efficient in the long term. Even so, much focus is currently on pharmaceutical residues in WWTPs, different types of micropollutants or disruptive effects, including antibiotic resistance, disinfection, estrogenic effects, microplastics and PFAS/PFOS, have already been identified as potential problems. As WWTPs represent a collection point for society’s discharge of contaminants, focus on removal of these micropollutants and requirements on their removal from WWTP-effluent is just a matter of time. The approach of adding different treatment steps one after another to follow societies focus on various micropollutants over time is not a sustainable way in terms of economic and environmental sustainability.

Besides a broader range of micropollutants, other aspects than cost should be included in order to provide a holistic and long-term sustainable implementation of treatment technologies. Here the improvement potential of a treatment option to become more sustainable in terms of environmental impact and costs should be considered.

The choice of which technology/combination that should be implemented at WWTPs is a complex issue and requires careful consideration of the various aspects presented here and preconditions at individual WWTPs. An effective and inclusive treatment of micropollutants can only be achieved by combining different technologies.

ACKNOWLEDGEMENTS

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REFERENCES


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