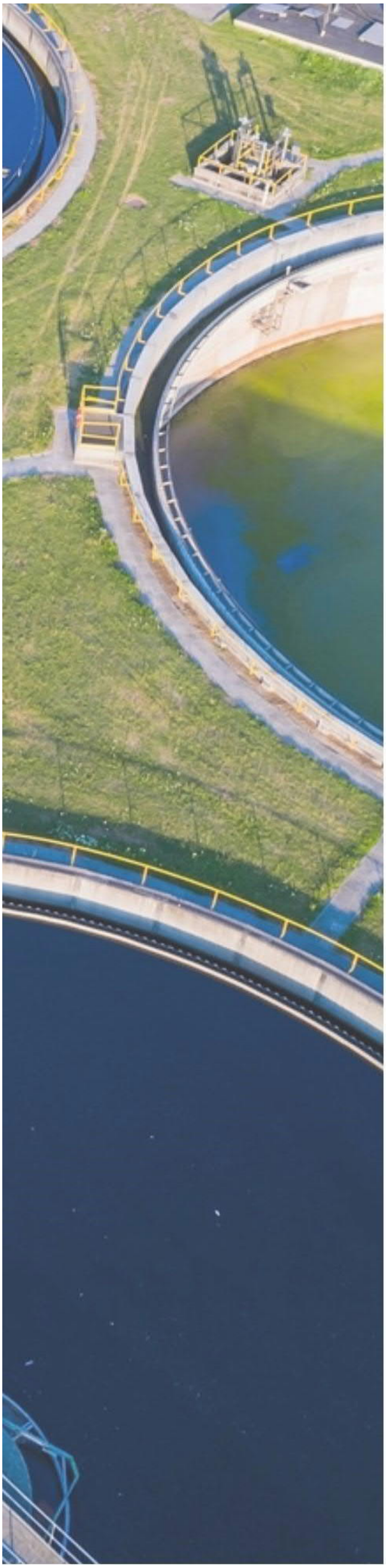

Interreg
South Baltic

EUROPEAN UNION

European Regional Development Fund

Less is More

less is MORE

Energy-efficient technologies for removal of pharmaceuticals and other contaminants of emerging concern

Deliverable 4.1 - part IV

Consolidated summary on treatment efficiency results

Foreword

This report is part of the project LESS IS MORE - Energy-efficient technologies for removal of pharmaceuticals and other contaminants of emerging concern. The project has been financed by the Interreg South Baltic Programme 2014-2020 through the European Regional Development Fund. The Swedish partners' participation in the project was co-financed by the Swedish Agency for Marine and Water Management.

Partners in the project are: Lund University, Department of Chemical Engineering; Sweden Water Research AB, Kristianstad University, Slagelse Utility, Slagelse Municipality, JSC "Kretinga Water" and Gdansk Water Fund.

The project started 1st of January 2018 and completion date is 30th of June 2021.

The specific project objective is to pilot -demonstrate, test and validate - new technological solutions for removing pharmaceuticals and other CECs as well as antibiotic-resistant bacteria that are suitable for small and middle sized WWTPs and to disseminate information on new technologies to the end-users.

This consolidated summary covers the treatment technology/efficiency results from three national reports, Part I-III, also within Deliverable 4.1.

This report was written by Michael Cimbritz, Åsa Davidsson, Ola Svahn, Ulf Nielsen and Kasparas Grazinskas

The contents of this report are the sole responsibility of the author[s] and can in no way be taken to reflect the views of the European Union, the Managing Authority or the Joint Secretariat of the Interreg South Baltic Programme 2014-2020.

Lund, June 2021

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Introduction

Large amounts of micro-pollutants are constantly introduced into the Baltic Sea. Pharmaceutical residues and other compounds of emerging concern (CECs) discharged from our society to the environment adversely affect aquatic ecosystems. 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 and bacterias. The negative effects of pharmaceuticals on the ecosystem include for example hormonal disorders, behavioral changes and various combinatory effects. Pharmaceutical residues and other CECs are spread through WWTPs effluents to receiving waters and action is thus needed at the WWTPs. A key problem, however, is the lack of energy efficient solutions for removal of micropollutants, without generation of potentially harmful transformation products.

Degradation of micropollutants by ozonation is possible but may cause formation of potentially harmful transformation products, known and unknown. By applying technologies based on separation, rather than degradation, effects of transformation products can possibly be avoided. Such technologies include certain membrane processes and the application of activated carbon, either in the form of PAC (powdered activated carbon) or GAC (granular activated carbon). GAC is typically applied in the form of filters following biological wastewater treatment and provides opportunities for removal of a broad spectrum pharmaceutical residues and other CECs. Operation is easy and the foot-print relatively low. GAC filtration technology is therefore a strong candidate for plant upgrades to remove CECs.

Objective and implementation

The specific project objective has been to test and validate new technological solutions for removing pharmaceuticals and other CECs as well as antibiotic-resistant bacteria.

The focus has been on different removal technologies focusing on separation processes and the technology development process has been anchored in two innovative approaches: i) process-driven innovation by introducing the membrane early in the process line to improve the treatment process and enhance potential for reuse of energy and water; and ii) technology-driven innovation by developing and testing novel polishing technologies including GAC-filtration as a fourth treatment step.

A key part of the project has been to develop and test removal technologies focusing on membrane and activated carbon processes used in various combinations in pilot scale in three countries, Lithuania, Denmark and Sweden, see locations in Figure 1. Full-scale design was the focus in Denmark and Lithuania where GAC filters were installed as a fourth and final polishing step. Besides removal of organic micropollutants, reuse of treated wastewater was evaluated in the Danish plant where the GAC filter was preceded by a microsieve and proceded by in-line UV-treatment. In addition to having GAC as a polishing step, the pilot plant in Sweden also had a different innovative configuration of applying chemical precipitation with microsieving and Direct Membrane Filtration (DMF) on primary wastewater as pre-treatment to GAC filtration.

A project-wide analysis capacity of organic micro-pollutants, through MoLab, Kristianstad University, has made it possible for all samples to be analyzed in the same way. MoLab is a state-of-the-art advanced organic trace-analysis laboratory equipped with a high-end UPLC-MS/MS. The innovative sample preparation techniques and developed analytical methods enable whole water analysis at low limits of quantification (LOQs) with high precision for a wide range of CECs. In this project about 35 micropollutants were selected for analysis.

This report is a consolidated summary of the results also reported in detail in the three national reports.

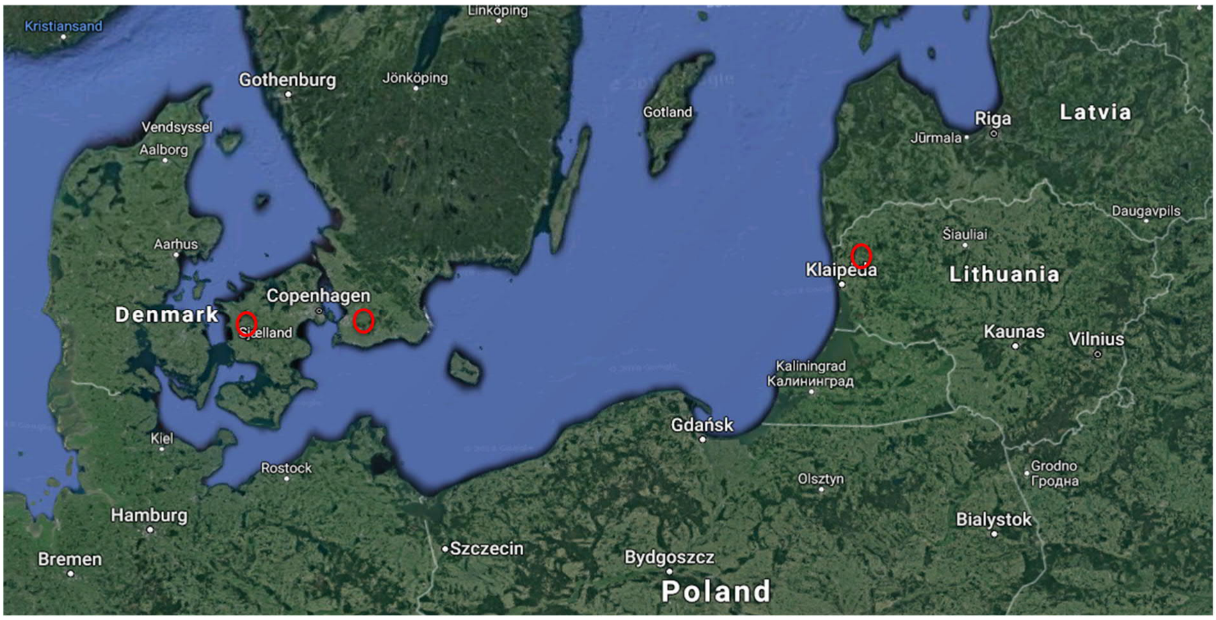
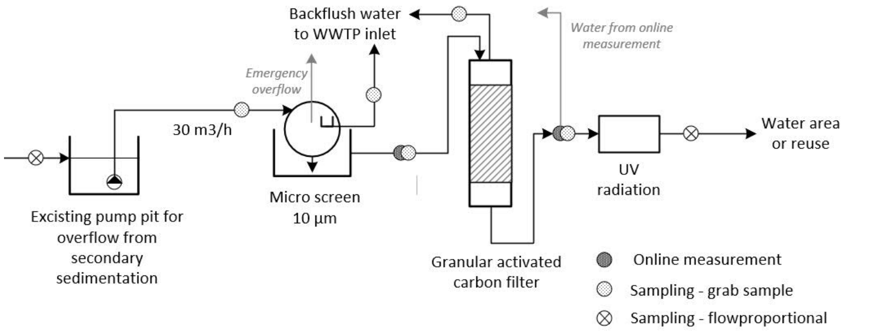


Figure 1. Locations of the three pilot plants in the Less is More project, Svedala in Sweden, Kretinga (outside Klaipeda) in Lithuania and Slagelse (on the west side of Sjaelland) in Denmark.

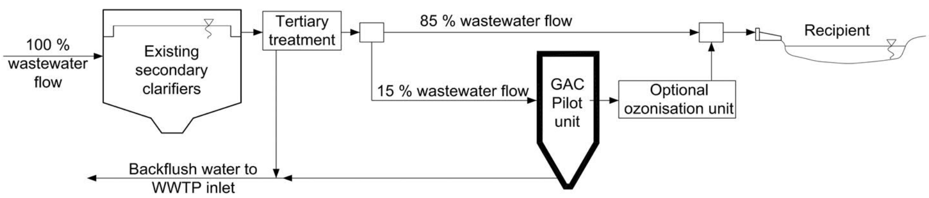
Treatment technologies

The treatment technologies, which were evaluated by pilot plant construction and operation, all included GAC-filtration. The pilot plant concepts are shown in Figure 2.

Concept for pilot plant in Slagelse, Denmark



Concept for pilot plant in Kretinga, Lithuania



Concept for pilot plant in Svedala, Sweden

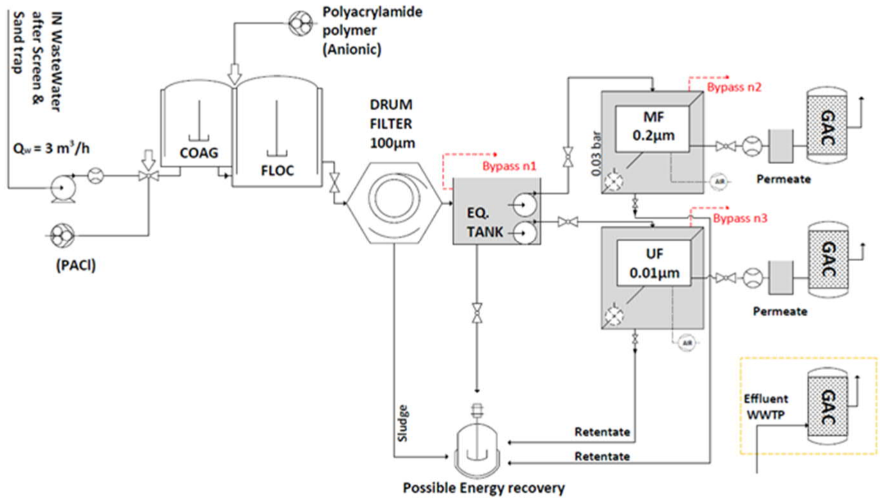


Figure 2. Concept charts for the three pilot plants in Denmark, Lithuania and Sweden.

All three pilot plants tested polishing technologies including GAC-filtration as an additional treatment step, i.e. GAC filtration was used after conventional biological treatment. The Danish plant used microsieving as pre-treatment, and also included a UV-radiation step for post-treatment. The Kretinga WWTP in Lithuania already included microsieving, as a tertiary treatment step after biological treatment. Therefore no extra pre-treatment before the GAC-filter was applied.

The Swedish pilot plant included a GAC-filter treatment line (with sand filtration as pre-treatment) as a polishing step after biological treatment but another concept, Direct Membrane Filtration, DMF, followed by GAC-filtration was also tested in a parallel treatment line. The DMF + GAC-line differed from the pilots using GAC as polishing step since the GAC-filter was loaded with water that had not passed biological treatment, which influences the TOC (DOC) content and the characteristics of the organic matter. Both microfiltration (MF) and Ultrafiltration (UF) was tested in parallel.

GAC as a polishing step

Selected design parameters for the GAC-filter processes are shown in Table 1.

Table 1. Design parameters for pilot-scale GAC processes in the project.

	Slagelse	Kretinga	Svedala WWTP effluent
Pre-treatment	Microsieve 10 µm	Microsieve 50 µm	Sand filter
Pilot flow (m^3/h)	30	22	0.114
Bed height (m)	3.5	2	0.54
Cross section area (m^2)	4.9	3.14 (d=2 m)	(d=0.22)
EBCT (min)	34	17	10
Filtration rate (m/h)	6.1	7	3
Type of backwash water (BW)	Effluent from WWTP	Effluent from tertiary	GAC-filtrate
Treatment of BW	Sent to inlet of WWTP	Sent to inlet of WWTP	To biological treatment
Mass of carbon (kg)	7 480		9
Volume of carbon (m^3)	15.6	6.3	
Carbon type	Desotec Organosorb 20[4]	Aquasorb 5000 or Cyclecarb	Aquasorb 5000

The GAC filters were all operated following activated sludge processes with similar and relatively low DOC-content in the order of 10 mg/L. The GAC filter at Svedala was a small-scale column with reduced contact time to enable prolonged operation in terms of total number of filtrated bed volumes. Longer and even shorter contact times were also evaluated. The full-scale filters at Slagelse and Kretinga were operated with filtration rates and empty bed contact times corresponding to state-of-the-art guidelines for GAC filtration of biologically treated wastewater.

Figure 3. GAC filter pilot plant in Kretinga, Lithuania.

GAC after DMF

The treatment steps proceeding GAC for this treatment line included coagulation, flocculation, microsieving and membrane filtration, see Figure 4. Coagulation and flocculation was applied to precipitate phosphorous and enhance particle separation in the microsieve (MS; 100 µm) prior to membrane filtration. Microfiltration (MF; 0.2 µm) and ultrafiltration (UF; 0.01 µm nominal pore opening) were operated in parallel for comparison reasons, using MS filtrate as feed. After membrane filtration, each generated permeate was filtered through a GAC filter for MP abatement.

Figure 4. DMF pilot plant including coagulation/flocculation tank (left side), drum sieve, retention tank and membrane unit (middle, black)

Treatment efficiency results

Removal of organic micropollutants in existing wastewater treatment as well as removal by addition of GAC as a polishing step were evaluated for the three wastewater treatment plants. As an alternative to existing treatment, the DMF concept followed by GAC was tested and evaluated in addition at Svedala WWTP.

Initial screening without GAC

Initial analyses in 2018 of influent and effluent samples from all three WWTPs confirmed that existing wastewater treatment (conventional biological and chemical treatment) is only removing organic micropollutants to a limited extent, see Figure 5.

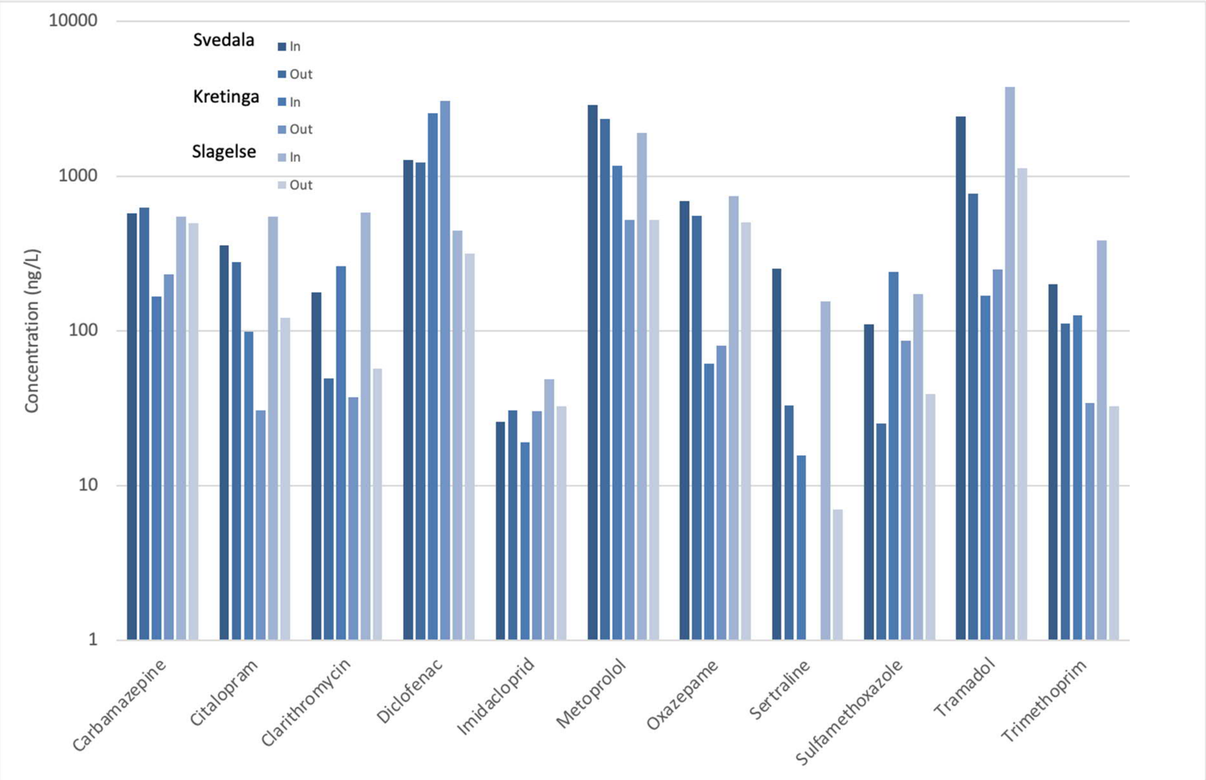


Figure 5. Influent and effluent concentrations of selected organic micropollutants in samples from the wastewater treatment plants in Svedala, Kretinga and Slagelse.

Most substances are only partially removed and some substances can be considered as persistent, for example carbamazepine and diclofenac. Thus, advanced treatment is necessary to accomplish removal of these and other persistent organic micropollutants. Some contaminants, such as sertraline and sulfamethoxazole, are on the contrary considerably reduced in the wastewater treatment plants but as a result of different mechanisms. Sertraline is sorbing to organic compounds and will therefore end up in the waste sludge from the plants whereas sulfamethoxazole is degraded in the biological treatment. Overall the inlet OMP concentration levels were twice as high in Svedala and Slagelse compared to Kretinga at the screening occasion. The reason might be lower pharmaceutical consumption. Another possible explanation is differences in the sewage network and various degrees of dilution. Inlet concentration levels also indicate different pharmaceutical consumption patterns. Concentration of diclofenac was almost twice as high in Kretinga compared to Svedala, and more than five times compared to Slagelse. Usage of tramadol appears to be lower in Kretinga than in Svedala and Slagelse.

GAC as polishing step

In Figure 6 the removal of six OMPs are presented together with the number of processed bed volumes (BV) at the three pilot plants. Given the smaller volume of the pilot in Svedala, together with a relatively low contact time, it ran the largest number of BVs (22 500), followed by Slagelse (15 000) and Kretinga (10 000). The low contact time is reflected in a minor column bleed for all OMPs even at a very low number of BVs, except for citalopram. Removal of OMPs were high in general up until about 7000 BV for all three pilots, when breakthrough of weak adsorbers such as sulfamethoxazole became apparent. Both pilots in Kretinga and Slagelse showed steep breakthroughs for the majority of OMPs, whereas the pilot in Svedala had a more prolonged removal, regardless of OMP studied. The carbon type used in Svedala had previously been used in a similar project, which showed very high removal and durability. Interestingly, the removal of OMPs in the Slagelse pilot increased at roughly ten thousand BV, which coincided with an increased contact time and a more frequent back wash schedule. Unfortunately, the long-term effect of this adjustment could not be further evaluated within in the scope of this project.

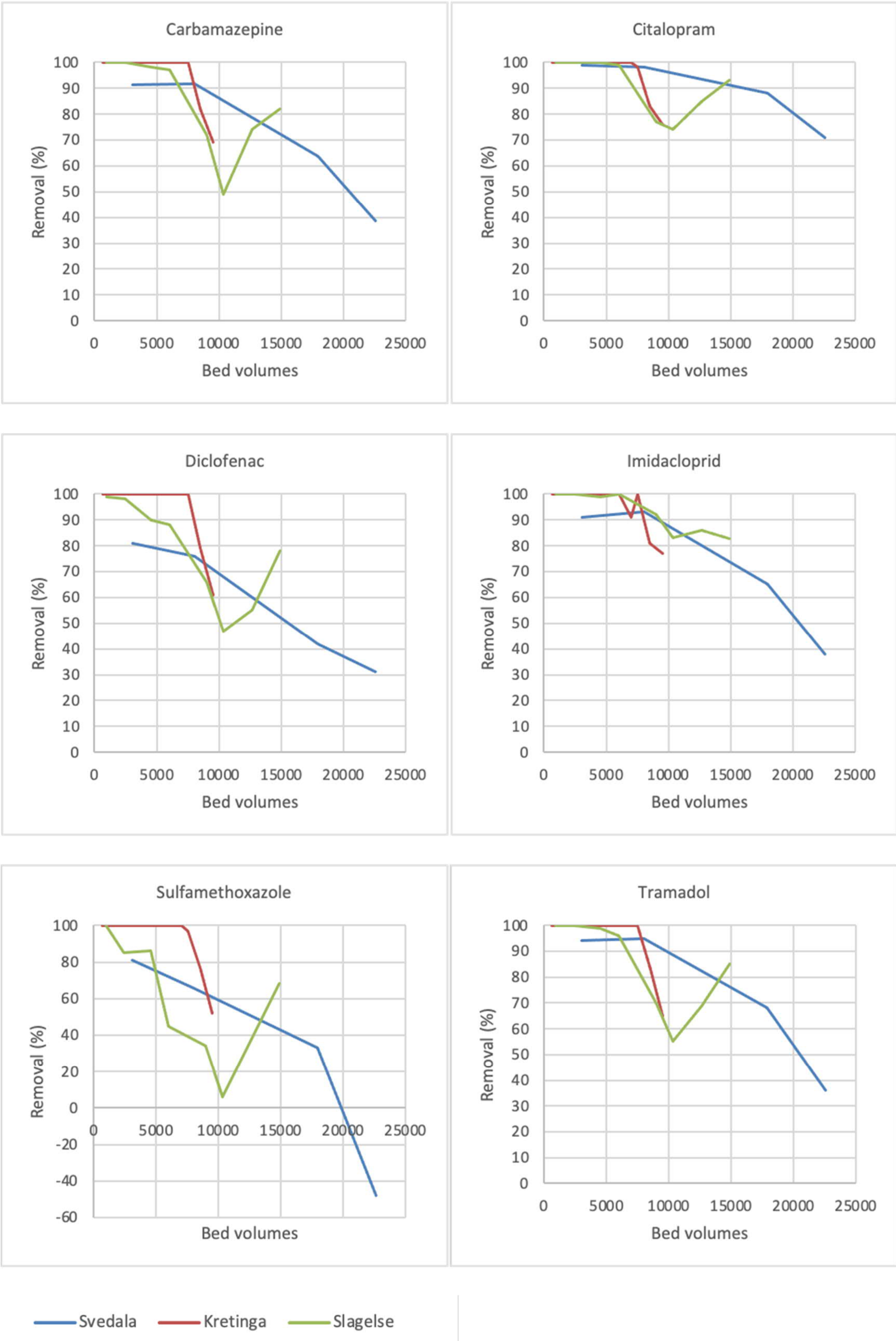


Figure 6. Removal (%) of six OMPs plotted against the number of processed bed volumes at the three pilot plants.

GAC after DMF

GAC filtration following DMF showed a very high removal potential of organic micropollutants. All substances studied were initially removed to at least 97%, see Figure 7. Long term effects and breakthrough behaviour could however not be analysed, since severe fouling of the membranes only permitted short time operation, in the range of a few days, before substantial cleaning was needed to continue operation. Further development of membrane operation on raw wastewater is needed in order to evaluate and potentially establish a full-scale concept based on DMF and GAC filtration. Tests in laboratory and pilot scale showed that approximately 90% of COD could be removed with DMF and coagulation/flocculation with microsieving. Most of the reduction was attributed to the microsieve with preceding coagulation/flocculation. Moreover, the DMF concept provides opportunities for reuse of wastewater and control of P-removal with potential for >95% phosphorus removal.

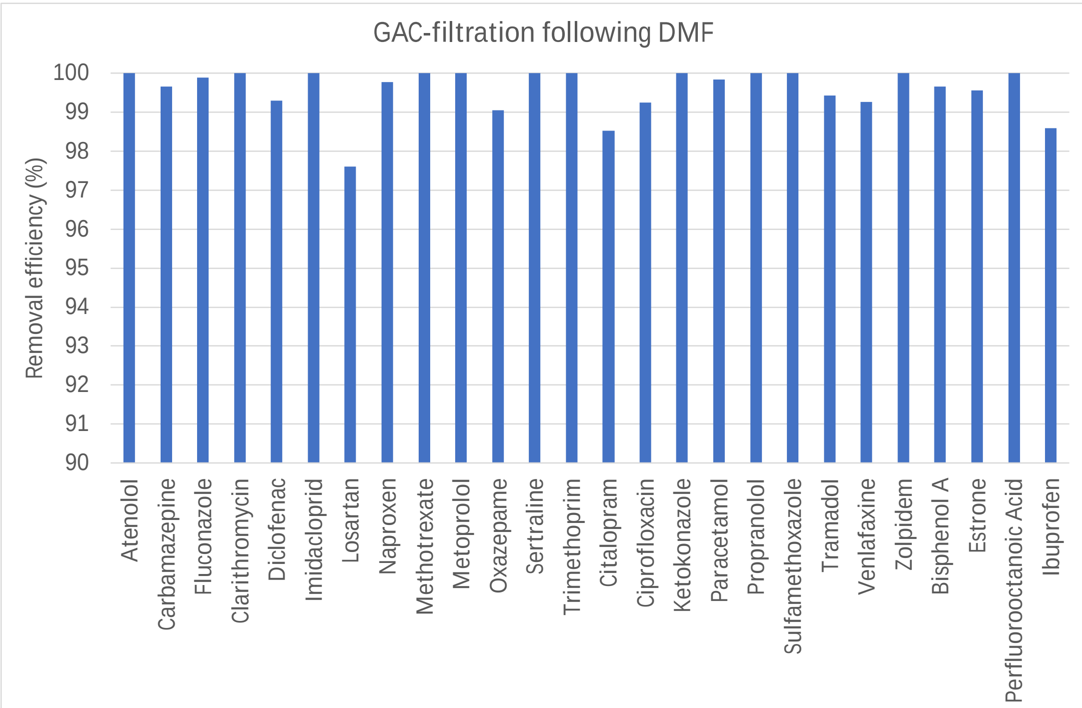


Figure 7. Initial removal efficiency over the GAC-filter following direct membrane filtration.

Final remarks

The results from the three pilot plants show that it is possible to achieve high removal efficiency of pharmaceuticals and other CECs by introduction of GAC-filtration after conventional wastewater treatment including biological treatment. The GAC-filtration polishing step is a rather simple concept to implement and operate. Costs are strongly related to exchange of filtration media, either in the form of new or regenerated granules. In order to extend the time between exchanges, pre-treatment is crucial to reduce the content of organic matter, which in turn reduces competition from dissolved organic carbon (DOC) for adsorption sites on the carbon. The carbon type and contact time also appears to be of vital importance, depending on which type of compound; weak or strong adsorbers, and what removal level that is required.