Advanced pharmaceuticals removal from wastewater - Roadmap for the model site Degeberga wastewater treatment plant

Project MORPHEUS 2017 - 2019

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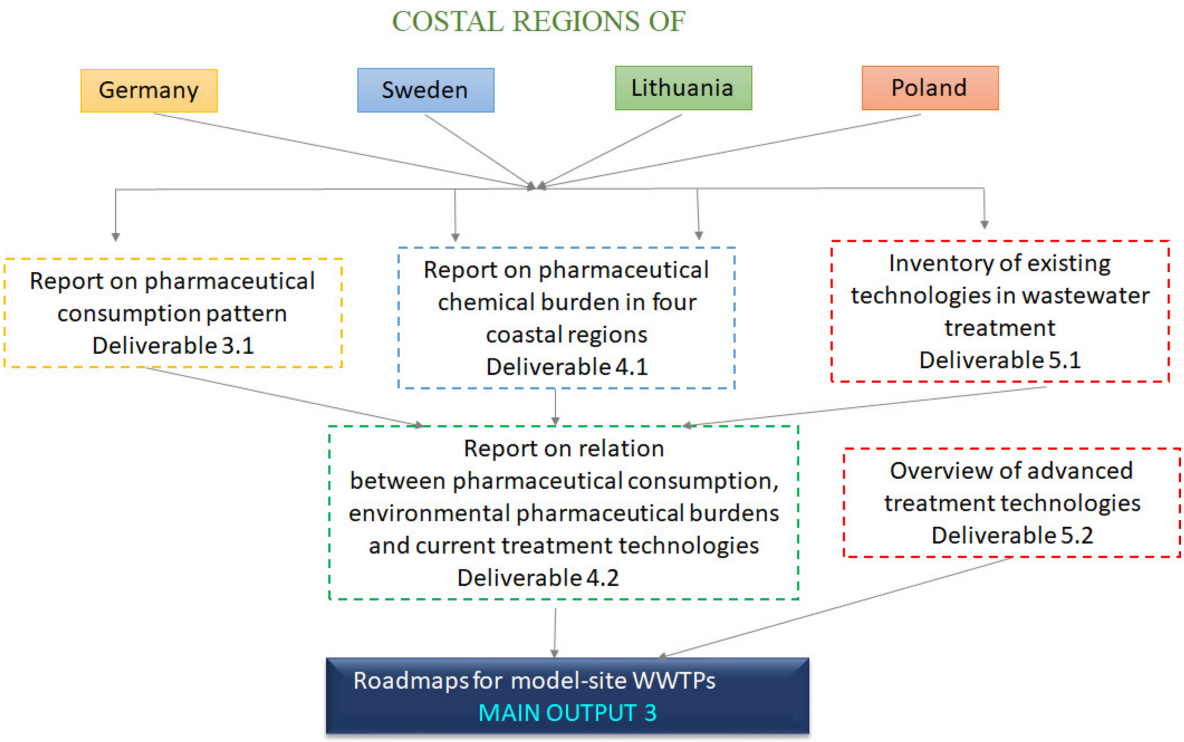
Cover photo

WWTP Degeberga. Photo: Erland Björklund

Key facts of the MORPHEUS project

MORPHEUS (Model Areas for Removal of Pharmaceutical Substances in the South Baltic) is a project financed by the European Union Interreg South Baltic Programme. The project duration is January 2017 – December 2019, with a total budget of EUR 1.6 million with a contribution from the European Regional Development Fund of EUR 1.3 million. The project has a total of 7 partners from four countries; Sweden, Germany, Poland and Lithuania: Kristianstad University (Lead Partner) – Sweden, EUCC – The Coastal Union Germany – Germany, University of Rostock – Germany, Gdansk Water Foundation – Poland, Gdansk University of Technology – Poland, Environmental Protection Agency – Lithuania and Klaipeda University – Lithuania. The project also has a total of 11 associated partners from these countries. For additional information on the project and activities please visit the MORPHEUS homepage at: www.morpheus-project.eu

The aim of this report is to provide Main Output 3, which is Roadmaps for investment in advanced treatment technologies at one selected WWTP in each region of the project; Poland, Sweden, Germany and Lithuania. Based on existing plant configurations and taking under consideration the current effectiveness in micropollutants removal, the analysis of possible upgrading and/or optimizing of existing technologies will be compiled with the information on e.g. feasibility, costs, and good practices connected with the suggested changes. These Roadmaps are provided for information purposes only and does not prejudge the final decision of the WWTPs operators, local authorities and other stakeholders.



Visualization of Main Output 3 in the context of MORPHEUS.

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Summary

Presence of human and veterinary pharmaceutical substances in our surrounding waterbodies is an emerging problem[1 2 3 4]. Thus, several pieces of European Union legislation, directly or indirectly and in different sectors rise the need of a strategic approach addressing pharmaceuticals and other emerging micropollutants (MPs) in the environment[5 6]. Currently two approaches are suggested to be developed simultaneously: (1) source and user measures - substitute critical MPs production and usage and (2) end-of-pipe measures - mitigate the dissemination of MPs by wastewater treatment plants (WWTPs). Since not all pharmaceuticals can be replaced with harmless (green) alternatives, end-of-pipe technologies seem to be essential to reduce the burden they pose on environment. Thus, wastewater sector's work is essential to protect the water resources, but need to be supported by the local society and authorities, as well as by reliable monitoring data on the current situation and information about the possible remedial actions.

Thus, in the model areas of Germany (Mecklenburg), Sweden (Skåne), Lithuania (Klaipeda) and Poland (Pomerania), the MORPHEUS project integrates crucial information on pharmaceutical consumption (Del. 3.1) and their release rates (Del. 4.1) by the existing WWTP technologies (Del. 5.1). This knowledge was combined with the environmental occurrence of pharmaceuticals (Del. 4.1, Del. 4.2). Additionally, the advanced treatment technologies that are already implemented in Sweden, Germany and Switzerland were presented and discussed in terms of: pharmaceutical removal efficiency, decision-making processes and the financing programmes (Del. 5.2). The above information is essential to reach the main objective of the MORPHEUS project - to inform stakeholders about the essence of the problem and solutions, possible to be undertaken at the local level in the wastewater sector.

Such efforts have already been undertaken by several countries, mainly Switzerland, but also, e.g., Germany and Sweden, and it is clear that the goals of the end-of-pipe strategy have to be clearly defined at national or even regional levels.

For this reason, four Roadmaps addressing the investment of advanced treatment technologies in selected regional WWTPs in Sweden, Germany, Lithuania and Poland were prepared. The proposed solutions were consulted with the key target groups of the MORPHEUS project:

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[1] Regulation (EU) No 1235/2010 OJ L 348, 31.12.2010, http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32010R1235&qid=1493205869407&from=EN

[2] Directive 2010/84/EU OJ L 348, 31.12.2010, http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32010L0084&qid=1493205642429&from=EN

[3] Communication from the Commission to the European Parliament and the Council: Action plan against the rising threats from Antimicrobial Resistance, COM/2011/0748 final. https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A52011DC0748

[4] http://ec.europa.eu/health/human-use/environment-medicines/index_en.htm

[5] Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council, https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:02008L0105-20130913

[6] Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy. https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A32013L0039

personnel at WWTPs and regional/national authorities. But to justify the economical aspect, besides the wastewater utilities and governmental bodies, also the local society needs to be attracted by this idea of a non-toxic environment.

Therefore, the process of advanced treatment implementation at WWTPs is suggested to be divided into two phases: (1) a preparation phase and (2) a testing phase. Both include the technical, ecological and socio-economical aspects needed to properly evaluate the inventory data, pharmaceuticals burden, stakeholders' opinions and financing options. Especially the pilot-scale experiments are critical to choose the most promising option of advanced treatment and its influence on current technology.

Sweden - Degeberga WWTP case study

Degeberga WWTP (Degeberga Avloppsreningsverk) is a well-functioning small object, which serves 1,350 inhabitants (PE= 950; Qav.= 9 m^3/h in 2016), and fulfills the current discharge requirements. Degeberga WWTP discharge the treated wastewater to the Segesholmsån River, and is the major source of pharmaceuticals to this recipient. Thus, Degeberga seems to be a good example of how to upgrade a small size WWTP. Additionally, Degeberga WWTP is already equipped with a final polishing step of sand-filtration, which is feasible for two advanced treatment technologies: ozonation and granulated activated carbon (GAC). Since ozonation technology would require some additional investment costs connected with the post-treatment step (e.g. sand filter or a pond of water), a GAC unit application is preferred. GAC filters are proven to be efficient in micropollutants removal, easy to use and maintain. Additionally, it should not cause any disturbance of existing processes. Besides the investment costs of GAC filters, additional costs (operation costs) seems to be connected only with the replacement of the used GAC, since it is a rather low-maintenance technology (for details please see below).

Germany - Rostock WWTP case study

The WWTP Rostock is the largest plant in the Federal State Mecklenburg-Vorpommern (235,645 inhabitants, PE=335.000), and discharges the highest total load of the investigated pharmaceuticals within the German model area (Del. 4.1). Thus, the WWTP Rostock represents the highest priority for introducing an advanced treatment technology to increase the removal rate of MPs (including pharmaceuticals).

For WWTP Rostock 6 different options of advanced treatment integration with the existing technology were discussed. For all, a sufficient elimination rate of pharmaceuticals can be presumed. In this case, the additional investments connected with modernisation, operation and maintenance expenses seem to be critical measures. Thus, two options were regarded as the most promising: (1) conversion of the BIOFOR-N into GAC; and (2) Ozone + BIOFOR-N + conversion of BIOFOR-DN into GAC. Among them, the first option appears to be the most cost-efficient solution with low impact on the existing treatment steps. The second option is suspected to provide the best elimination of micropollutants but causes additional efforts for the conventional nutrient removal (for details please see: Advanced pharmaceuticals removal from wastewater - roadmaps for model-site Rostock wastewater treatment plant case study).

Poland - Gdynia-Debogorze WWTP case study

In Poland Gdynia-Debogorze WWTP was selected as a model plant for the Roadmap. It is the second largest WWTP facility in the Polish model area, which in 2015 served 360 000 inhabitants (PE=476 000, Qav. = 55 294 m^3/d). Currently Gdynia-Debogorze WWTP is regarded as a modern, large object with a well-designed treatment process, fulfilling the discharge requirements in terms of macropollutants. Pharmaceuticals are, however, removed with limited efficiency (Del. 4.1). Importantly, the treated wastewater from Gdynia-Debogorze WWTP is directed into the Puck Bay (2.3 km from the coastline), which is an area protected by Natura 2000. Since some of the pharmaceuticals studied within the MORPHEUS project were detected in marine water at the discharge point (Erythromycin, Azithromycin, Clarithromycin, Sulfamethoxazole, Carbamazepine, Diclofenac, Metoprolol), the implementation of advanced treatment seems to be essential to protect this shallow western branch of the Bay of Gdansk.

Average effluent parameters, such as low total suspended solids and organic matter predispose this plant to ozonation and/or activated carbon technology, however powdered activated carbon (PAC) was excluded from consideration, due to requirements of Gdynia-Debogorze WWTP operators.

Note, in Poland, the main obstacles for advanced treatment implementations in the wastewater sector, besides the lack of legal basis, are the lack of data on pharmaceuticals fate in treated wastewater and receiving water bodies and a limited experience among the WWTPs exploiters. Thus, lab-, and pilot-scale studies are highly suggested to evaluate on-site the effectiveness of advanced treatment as well as to estimate the maintenance conditions and costs. But despite the pilot investments, a discussion on political and multi-stakeholder level is needed. It should be supported by monitoring data showing the pharmaceuticals fate and burden posed on the local aquatic environment. Fulfilling this knowledge gaps will probably attract also attention of local society and acceptance to share the cost bearing (for details please see: Advanced pharmaceuticals removal from wastewater - roadmaps for model-site. – Gdynia-Debogorze wastewater treatment plant case study[7]).

Lithuania – Klaipėda city WWTP case study

Klaipėda city WWTP is the largest WWTPs in the Lithuanian model area (Qav. = 41256 m^3/day) and discharges wastewater to the receiver Klaipėda Strait. As in Poland, also in Lithuania there is limited knowledge about the fate of pharmaceuticals in WWTPs, the effectiveness of their removal and the load discharged to the receiving water bodies. However, data provided by the MORPHEUS project as well as the pilot investments of advanced GAC treatment implemented in

[7] http://www.morpheus-project.eu/downloads/

Kretinga town WWTP (co-supported by the EU Interreg South Baltic programme) can give valuable information and serve as a guide for local stakeholders to plan future projects (for details please see: Advanced pharmaceuticals removal from wastewater - roadmaps for model-site. Klaipėda city wastewater treatment plant case study[8])

It can be concluded that water pollution is a trans-boundary problem, thus joint actions should be undertaken at the EU level. EU-level guidance or the EU-wide provision of information could be more efficient than action taken separately by individual Member States. However, the national/regional goals and obstacles as well as the wastewater sectoral specificity should always be considered in this process. For this reason, the information and data already available about pharmaceutical consumption, their pattern in wastewater and removal efficiency by WWTPs, as well as their fate in the water resources should be gathered, shared and complemented at national levels. There is also a need to disseminate those outcomes for public consultation to get a broad societal and political acceptance. The involvement of a wide a range relevant stakeholders can stimulate voluntary national initiatives during pharmaceuticals production, their consumption and at the disposal stage.

8 http://www.morpheus-project.eu/downloads/

1 Introduction

The MORPHEUS project aimed to combine the information on pharmaceuticals consumption (Del. 3.1) with their patterns observed in raw wastewater (Del. 4.1) to properly selected advanced treatment for model WWTPs located in the Baltic Sea coastal regions: Skåne (Sweden, SE), Mecklenburg (Germany, DE), Klaipeda (Lithuania. LT) and Pomerania (Poland, PL). The existing treatment technologies were investigated in terms of micropollutants removal efficiency, to estimate the release of pharmaceuticals via WWTPs discharges (Del. 4.1, Del. 4.2, and Del. 5.1). Additionally, strategies to reduce the release of micropollutants to the aquatic environment by advanced treatment technologies, already adopted in Switzerland, Germany and to some extent in Sweden were presented in Del. 5.2. That integrated information is essential for WWTP operators, regional/national authorities, and other target groups interested to reduce the environmental stress posed on the costal ecosystem of the Baltic Sea.

Based on the above-mentioned reports key facts about the environmental risks of pharmaceuticals within the MORPHEUS model area are as follow:

1. EU members are important consumer of medicinal products, however the level of pharmaceutical consumption as well as the consumption pattern significantly differs and depends on many factors including medical and socio-economical habits (Del. 3.1)
2. Data on pharmaceuticals consumption is scattered, especially for over-the-counter medicines (Del. 3.1)
3. An unknown share of unused or expired pharmaceuticals is not properly collected and disposed, mainly due to unclear waste management, especially inadequate implementation of take back schemes
4. Consumed pharmaceuticals are partly excreted via urine and feaces, thus the consumption is the main contributing step of pharmaceuticals presence in wastewater (Del. 4.1)
5. There is limited monitoring data on pharmaceuticals presence in WWTP inlets, outlets and receivers, mainly due to relatively high costs of analysis and lack of standardized methods for pharmaceuticals detection (Del. 4.1)
6. In the MORPHEUS model area, pharmaceuticals were detected in all tested compartments: raw and treated wastewater as well as wastewater receivers (Del. 4.1)
7. The effectiveness of biological wastewater treatment (mainly based on activated sludge) is high in terms of macropollutants, but varied strongly in terms micropollutants, most likely/potentially due to usually limited sorption to sludge flocks and biodegradation rates of pharmaceutical compounds (Del. 4.1)
8. Negative removal rates, obtained for some pharmaceuticals such as Carbamazepine, indicated the importance of other patterns such as sewage sludge management in cycling and balance of micropollutants within the WWTPs (Del. 4.1)
9. Numerous pharmaceuticals are usually detected in ecosystems, while the risk assessment is usually evaluated for a single compound; this does not reflect the combined hazard posed by multi-compounds mixture (Del. 4.1).

10. The precise knowledge of environmental behavior of most pharmaceuticals, their ecotoxicology and mixture effects are still limited.

Despite the lack of a comprehensive knowledge about the behavior and the effect, which pharmaceuticals pose to the environment, there is no doubt that their presence in the water bodies can be regarded as an emerging problem. This problem is expected to grow in the years ahead, mainly due to population aging and growth.

To reduce the environmental impact of pharmaceuticals and other micropollutants, a complex strategy is required, which includes mitigation at both the source and the user side, as well as the introduction of more advanced end-of-pipe technologies. In the case of pharmaceuticals, which are used in medical applications and are absolutely essential in our healthcare systems, they cannot easily be replaced or limited. Thus, advanced wastewater treatment is urgently needed to limit pharmaceuticals dissemination via the discharge of WWTPs' effluents.

Estimation of pharmaceutical load discharged by WWTPs effluents

In total 15 WWTPs, located in the MORPHEUS model area, were selected in the project to analyze the dissemination of 15 pharmaceuticals in wastewater receivers. The pharmaceutical concentrations detected in treated wastewater during the summer (2017) and winter (2018) sampling campaign were used to estimate the pharmaceutical loads discharged by WWTPs effluents into the recipients. Additionally, in each sampling point the total load of all tested pharmaceuticals was calculated and is presented in Table 1 (detailed information is also provided in Del. 4.1 and 4.2).

According to the obtained data, the highest load was discharged directly to the Baltic sea by the WWTPs located in the Polish model area: Gdansk-Wschod WWTP and Gdynia Debogorze WWTP (average annual load: 216.16 kg and 146.66 kg, respectively). However, per 1000 residents the highest load was discharged by WWTP Palanga, located in the Lithuanian model area (0,84 kg per year). Additionally, probably due to more infections in the winter season and consequently increased consumption, antimicrobials, anti-inflammatory drugs and pain killers were observed at elevated concentrations both in raw and in treated wastewater (winter sampling campaign).

It can be concluded that the existing wastewater treatment systems, based on activated sludge, are not effective enough to remove most of the investigated pharmaceuticals and mixtures of pharmaceuticals are constantly discharged into the receiving water bodies. According to the obtained data, selected pharmaceuticals were detected in each WWTP's receiver. Beside the load of pharmaceuticals in treated wastewater, other parameters are also important, such as treated wastewater share and dispersion rate in the receiving water body. Nonetheless, selected pharmaceuticals were still detected, even when the treated wastewater was discharged by marine outflow located far from the coast (> 2km), as in case of the WWTPs in the Polish model area. It should also be considered that not only the investigated WWTPs may discharge pharmaceutical loads into the receiver but also upstream and downstream WWTPs, which can contribute to the measured concentrations within a water body.

Table 1. "Total" load (kg/year) of 15 pharmaceuticals discharged by WWTPs into the receiver bodies. Estimation based on the influent/effluent concentrations and the total volume of treated wastewater data obtained during summer 2017 and winter 2018 campaigns, for details see Del. 4.1 and 4.2.

SWEDISH MODEL AREA					
WWTP	**Kristianstad**	**Tollarp**	**Degeberga**	**-**	**Total**
Aver. inlet load, kg/year	598.68	25.23	23.43	-	647,3
Aver. inlet load kg per year per 1000 residents	11.51	8.41	24.66	-	-
Aver. outlet load, kg/year	33.27	2.03	0.53	-	35,8
Aver. outlet load kg per year per 1000 residents	0.64	0.68	0.56	-	-
Recipient	Hammarsjön lake /Helge Å river/ Hanöbukten bay	Vramsån river/ Helge Å river/Hanöbukten bay	Segeholmsån/Baltic Sea/Hanöbukten bay	-	
GERMAN MODEL AREA					
WWTP	**Rostock**	**Laage**	**Krakow**	**Satow**	**Total**
Aver. inlet load, kg/year	10079.06	91.15	437.87	85.46	10693.5
Aver. inlet load kg per year per 1000 residents	42.77	20.18	110.46	65.59	-
Aver. outlet load, kg/year	84.85	1.27	2.6	0.39	89.1
Aver. outlet load kg per year per 1000 residents	0.36	0.28	0.66	0.30	-
Recipient	**Unterwarnow**	**River Recknitz**	**River Nebel**	**River Mühlenbach**	
POLISH MODEL AREA					
WWTP	**Gdansk-Wschod**	**Gdynia-Debogorze**	**Swarzewo**	**Jastrzebia Gora**	**Total**
Aver. inlet load, kg/year	18840.65	18234.11	2423.13	421.8	39919.7
Aver. inlet load kg per year per 1000 residents	32.98	50.65	67.94	42.18	-
Aver. outlet load, kg/year	216.16	146.66	9.85	3.81	376.5
Aver. outlet load kg per year per 1000 residents	0.38	0.41	0.28	0.38	-
Recipient	**Gdansk Bay**	**Puck Bay**	**Baltic Sea**	**Czarna Wda river**	
LITHUANIAN MODEL AREA					
WWTP	**Klaipeda**	**Palanga**	**Kretinga**	**Nida**	**Total**
Aver. inlet load, kg/year	2459.8	235.08	433.22	11.5	3139.6
Aver. inlet load kg per year per 1000 residents	14.47	18.08	22.62	6.71	-
Aver. outlet load, kg/year	76.6	10.97	6.32	0.65	94.5
Aver. outlet load kg per year per 1000 residents	0.45	0.84	0.33	0.38	-
Recipient	**Klaipėda Strait**	**Baltic Sea**	**River Tenžė (drainage ditch)**	**Curonian Lagoon**	

As mentioned above, the Roadmaps aim to inform stakeholders about the problems and possible implementation of the best-suited, advanced treatment, which will be effective in the removal of pharmaceuticals and micropollutants. Decision making criteria for implementation of advanced treatment are given in Figure 1. They were divided in the two phases: a preparation phase and a testing phase, which both should include technical, socio-economic and ecological aspects, with special attention given to the environmental burden. In the preparation phase the crucial step is to define the local objectives and criteria for advanced treatment, while at the testing phase critical analysis of the most promising alternatives should be conducted by lab-, and or pilot-scale studies. It is important to correctly estimate the on-site effectiveness of the tested advance treatment as well as the costs of implementation and maintenance.

In the MORPHEUS project the following 4 WWTPs were selected for the roadmaps: Degeberga WWTP in Sweden, Rostock WWTP in Germany, Gdynia-Debogorze WWTP in Poland and Klaipeda WWTP in Lithuania.

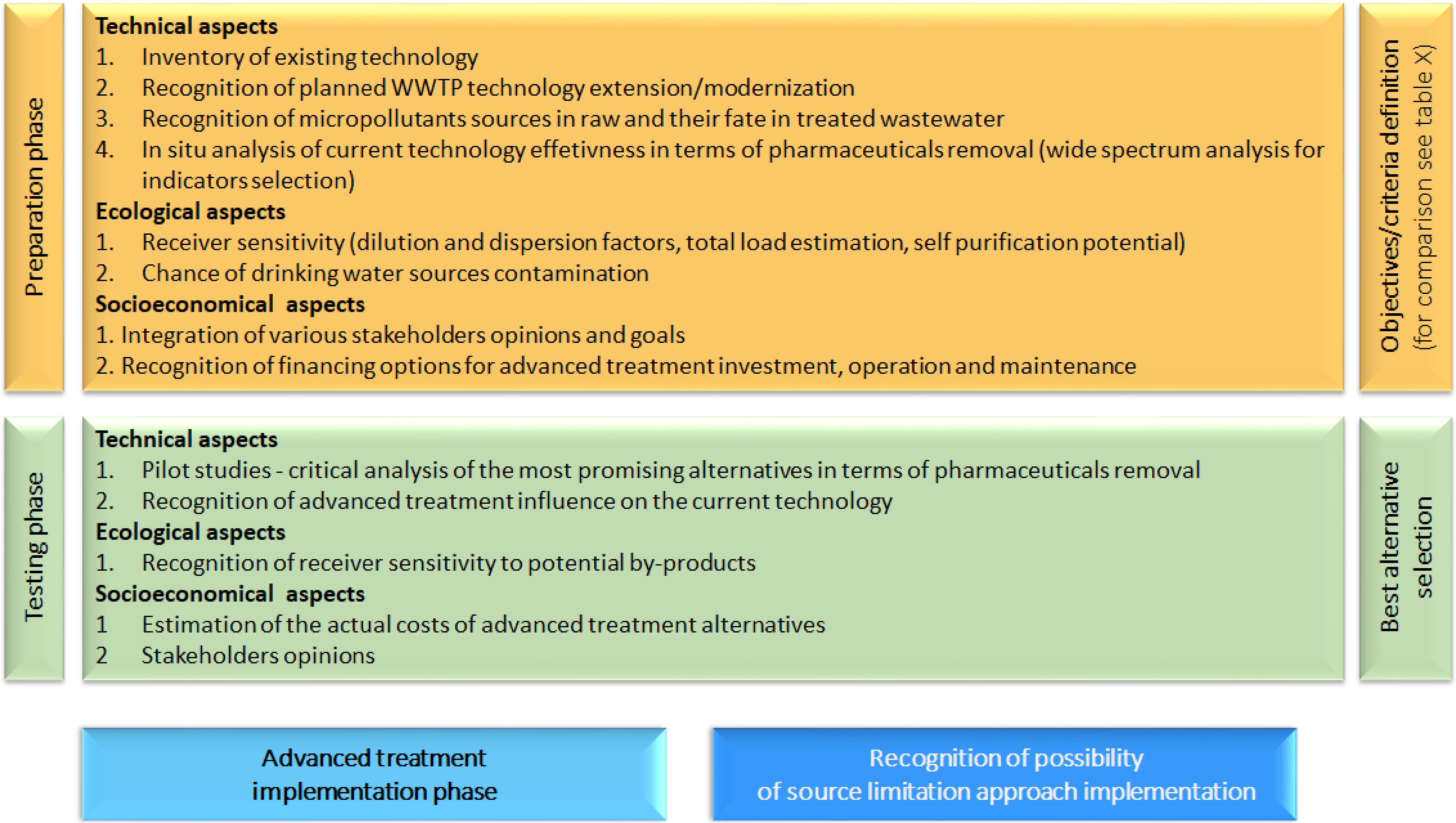

Fig. 1. Decision making criteria for implementation of advanced treatment at wastewater treatment plants.

2 The general designed criteria, measures and decision-making criteria for implementation of advanced treatment in WWTPs

Up to now, a number of studies have indicated and recommended the two technologies ozonation and activated carbon as effective in removal of pharmaceuticals (and other micropollutants) from wastewater at reasonable costs (Table 1-5, for details see Del. 5.2). A schematic overview of the advanced treatment units placement within the steps of conventional wastewater treatment technology are suggested and can be seen in Figures 2-4

Additionally, the presence of micropollutants in the treated wastewater as well as the removal effectiveness should be controlled. It is therefore recommended to monitor the presence of indicator substances in the WWTP's influent and effluent. The indicators need to be chosen according to the following criteria:

- be present in sufficiently high concentrations in influent of targeted WWTPs with small load variation.
- their removal by conventional (biological) WWTPs should be little or non-existent.
- their removal by advanced treatment should be specific (high or low) to the method
- they can be assessed simply, during a single run with LC/MS/MS.

Table 2. General design criteria in Germany and Switzerland for removal of micropollutants from municipal WWTP effluent using ozonation [9]

Subject	Unit	Value
Dosage ozone	g O_3 / g DOC	0.6–0.9
Dosage ozone	mg O_3/L*	4–14
Hydraulic Retention Time Contact Tank	minutes	15–30 (reactor 10–25 min; Removing remaining ozone 5 min)
Power consumption	kWh/kg O_3 * h	10
Power consumption	W/treated m^3	45

Based on Dissolved Organic Carbon in WWTP effluent of 7 - 15 mg/L

[9] Mulder et al. (2015) Costs of Removal of Micropollutants from Effluents of Municipal Wastewater Treatment Plants - General Cost Estimates for the Netherlands based on Implemented Full Scale Post Treatments of Effluents of Wastewater Treatment Plants in Germany and Switzerland. STOWA and Waterboard the Dommel, The Netherlands

Table 3. General design criteria in Germany and Switzerland for removal of micropollutants from municipal WWTP effluent using PAC[10]

Subject	Unit	Value
Dosage PAC	g PAC / g DOC	0.7–1.4
Dosage PAC	mg PAC /l*	10–20
Dosage coagulant	mg/l	4–6
Dosage polymer	mg 100% active /l	0,2–0,3
Hydraulic Retention Time Contact Tank	Minutes	30–40
Surface load settler	m/h	2.0
Recycle factor PAC	-	0.5–1.0
Power consumption	W/treated m^3	45

Based on Dissolved Organic Carbon in WWTP effluent of 7 - 15 mg/L

Table 4. General design criteria in Germany and Switzerland for sand filtration after ozonation or PAC[11]

Subject	Unit	Value
Upflow velocity	m/h	12
Backwash water	% of incoming flow	5–10
Power consumption	W/treated m3	15

Table 5. General design criteria for removal of MPs from biologically treated wastewater by GAC units in Germany and Switzerland[12]

Subject	Unit	Value
Empty Bed Contact Time	minutes	20–40
Upflow velocity	m/h	6–10
Backwash water	% of incoming flow	5–15
Power consumption	W/treated m^3	40
Replacement coal	-	After 7.000–15.000 bed volumes (standing time 4 months to 1 year)

[10] as in [9] Mulder et al. (2015)
[11] as in [9] Mulder et al. (2015)
[12] as in [9] Mulder et al. (2015)

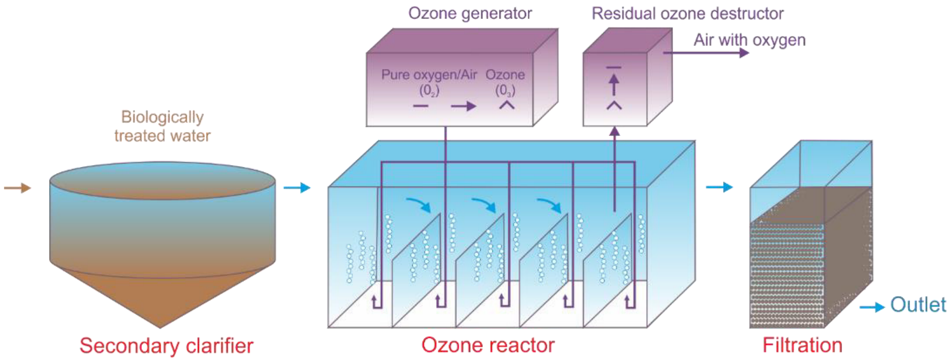

Figure 2. Schematic overview of the ozone unit suggested location in the conventional wastewater treatment technology (modified from[13])

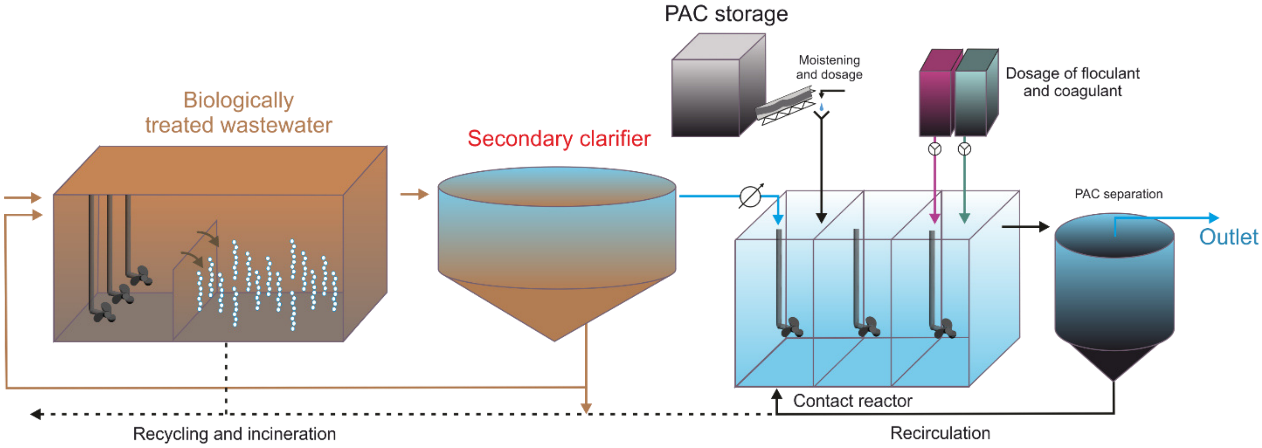

Figure 3. Schematic overview of the PAC unit suggested location in the conventional wastewater treatment technology (modified from[14])

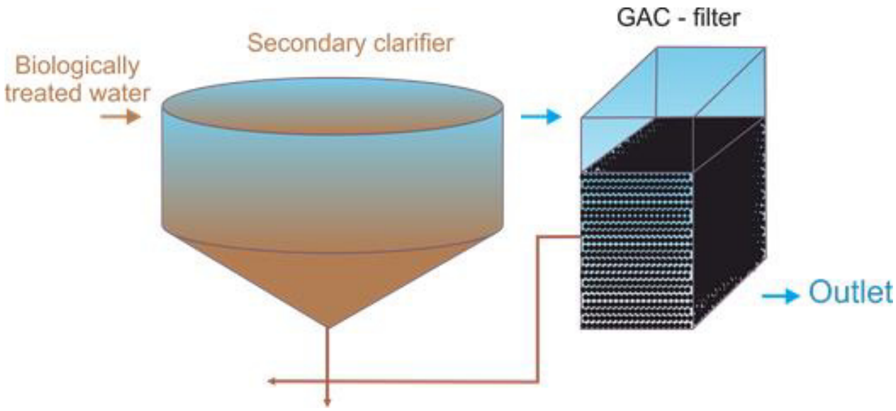

Figure 4. Schematic overview of the GAC unit suggested location in the conventional wastewater treatment technology (modified from[15])

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[13] Abegglen C. & Siegrist H. (2012): Mikroverunreinigungen aus kommunalem Abwasser. Verfahren zur weitergehenden Elimination auf Kläranlagen. Bundesamt fur Umwelt, Bern, Umwelt-Wissen Nr.1214: 210 S.
[14] as in [13] Abegglen & Siegrist (2012)
[15] as in [13] Abegglen & Siegrist (2012)

3 Roadmap for Sweden

3.1 Justification of the selection of Degeberga WWTP for the Roadmap

In Sweden, Degeberga WWTP was selected as a model WWTP for the Roadmap. Degeberga WWTP is a small WWTP on the east side of Region Skåne releasing its wastewater into a the small Segesholmsån River which ends in the Baltic Sea in the Hanöbukten Bay. The geographical position of Segesholmsån River area in Region Skåne, Sweden is seen in Figure 1.

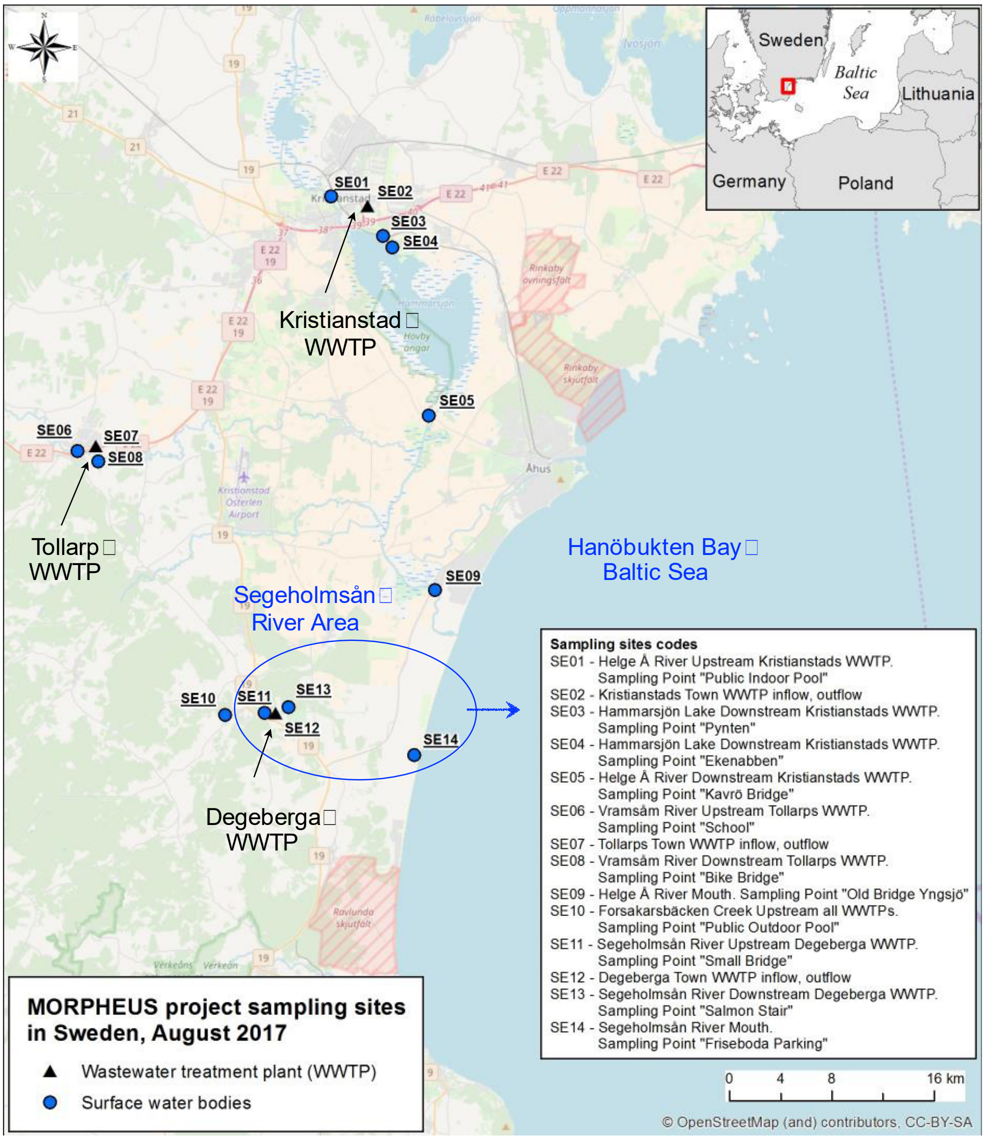

Figure 1. The geographical position of the Degeberga WWTP in the Swedish Model Area.

Degeberga WWTP (Degeberga Avloppsreningsverk) serves the population of the small town of Degeberga (1,350 inhabitants 2016-12-31). Degeberga WWTP is the only WWTP releasing wastewater to the river since the river does not pass by any other major towns. Consequently, Degeberga WWTP is the major source of pharmaceuticals to this recipient. No major industry is connected to the WWTP. The treatment technology at Degeberga WWTP consists of mechanical, biological and chemical treatment as outlined in more detail below. The requirements for treatment of water is an average yearly value of 10 mg/L of BOD_7 and 0.3 mg/L of P_{tot}. According to the Environmental Report 2017 these outlet concentrations were < 3mg/L BOD_7 and 0.05 mg/L P_{tot} giving treatment efficiencies of 99.5% and 99.3%, respectively. The treatment efficiency the past 9 years is shown in Figure 2.

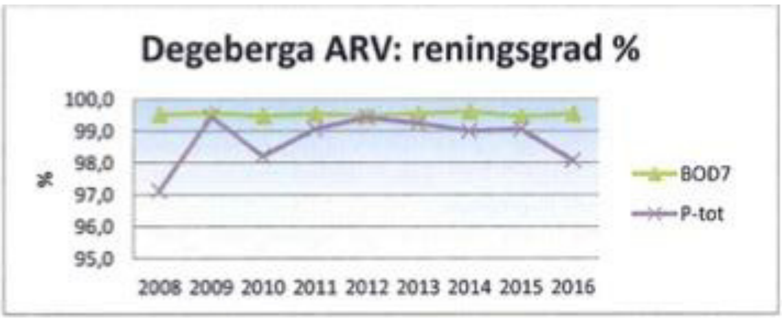

Figure 2. Treatment efficiency of BOD_7 and P_{tot} at Degeberga WWTP 2008-2016. Graph from Degeberga WWTP Environmental Report 2016.

Degeberga WWTP fulfills the requested discharge requirements and is a well-functioning WWTP. In 2016 the organic matter discharged by Degeberga WWTP, expressed as BOD_7 and COD, equaled about 119 kg/year and 1186 kg/year, respectively. Additionally, 1039 kg of N_{tot} and 13 kg of P_{tot} was released to the river. A more detailed overview of the WWTP and its placement along the Segesholmsån River is shown in Figure 3.

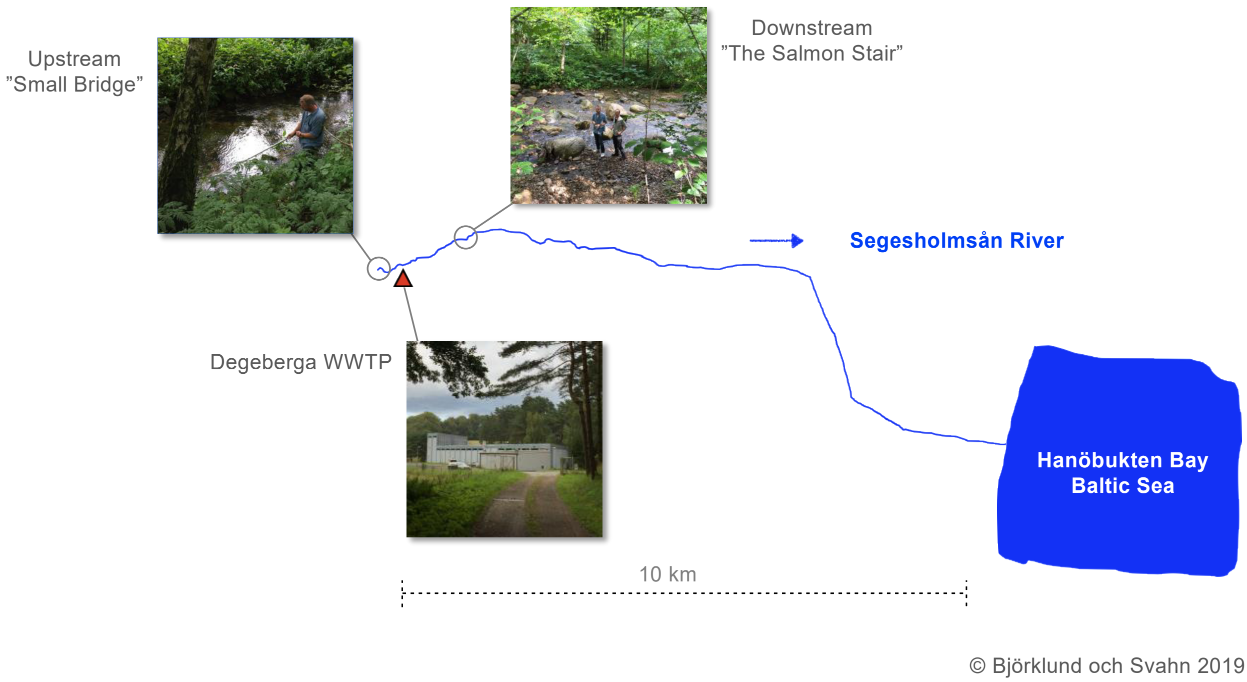

Figure 3. Expanded view of Degeberga WWTP and the Segesholmsån River with upstream and downstream sampling points.

Segesholmsån River is one of the best-preserved rivers in Region Skåne. It has a relatively undisturbed stream with clean, cold and oxygen-rich water, which contains many sensitive species. The river houses several fish species such as Trout, Common minnow, Eel, European bullhead and rare species of Caddisflies. Segesholmsån River ends in a nature preservation area at the Baltic Sea called Friseboda. Both Segesholmsån River and Friseboda are part of a unique wetland called "Vattenriket" which was given the status of a UNESCO Biosphere Reserve in 2005. The area holds a great variety of species of which many are red listed. Finally, it could be noted that the part of the Baltic Sea where Segesholmsån River ends is called the Hanöbukten Bay (Figure 3). This bay has for at least 10 years suffered from problems with declining number of fishes and fishes showing signs of poor health. The reason behind this is not known despite several investigations. One suggestion has been that a large number of chemicals (a cocktail of chemicals) released to the recipient may have negative impact on this sensitive ecosystem. Part of the solution to these problems may therefore be to decrease the chemical burden from WWTPs such as the one in Degeberga. Degeberga WWTP will thereby serve as a good first example of how WWTPs in this region can be upgraded without great costs since Degeberga WWTP is a small size WWTP.

3.2 Pharmaceuticals in the Degeberga WWTP's inflow, outflow and receiver

Despite that the occurrence of pharmaceuticals in the environment has been investigated for decades, there is most often a lack of knowledge about the amount pharmaceuticals received and released by specific WWTPs in Sweden. Within the MORPHEUS project two sampling campaigns were carried out; September 2017 and February 2018. The result obtained for Degeberga WWTP are presented in Table 1 (for details see MORPHEUS project's Deliverable 4.1).

a) Pharmaceuticals found in inlet water (see Table 1):

- Some of the highest concentrations were observed for Ibuprofen with a value of up to 307 µg/L, Paracetamol up to 47 µg/L, Ciprofloxacin up to 8.8 µg/L, Carbamazepine up to 5.7 µg/L and Naproxen up to 5.3 µg/L, Sulfamethoxazole on the other hand could not be detected in one sample while in the other it was as low as 2.3 ng/L. Several other compounds never exceeded 155 ng/L, including Azithromycin, Clarithromycin, Erythromycin, Estrone and Propranolol.
- For some pharmaceuticals the concentrations differed somewhat between seasons, the most pronounced being Ciprofloxacin (highest in winter), Clarithromycin (highest in summer) and Erythromycin (highest in summer)

b) Pharmaceuticals found in outlet water (see Table 1):

- In the outlet water the highest concentrations were observed for Carbamazepine up to 5.0 µg/L, Diclofenac up to 1.4 µg/L, Oxazepam up to 866 ng/L and Metoprolol up to 304 ng/L.

- In outflow several compounds were at very low concentrations or could not be detected at all such as Atenolol, Estrone, Ibuprofen, Paracetamol and Sulfamethoxazole.

c) Based on the inlet and outlet concentrations the removal rates were calculated (see Table 1).

- The removal rates varied largely, from -167% for Sulfamethoxazole (winter season) up to 100% for Atenolol, Estrone, Ibuprofen and Paracetamol. Removal efficiencies >90% were also observed for Azithromycin, Ciprofloxacin, Metoprolol and Naproxen.
- Poor removal efficiency was observed for Carbamazepine 11% (summer), Diclofenac -35% (winter), Erythromycin -136% (winter) and Oxazepam 19% (winter).
- Substantial differences in removal rates between season were observed for Diclofenac (67% summer and -35% winter) and Erythromycin (21% summer and -136% winter).

d) Pharmaceuticals found in the Segesholmsån River area, catchment area of treated wastewater (Table 1).

- In the river water upstream Degeberga WWTP only traces of one pharmaceutical could be detected; 12 ng/L of Naproxen in the winter sample. This supports that no other major source of pharmaceuticals exists in the Segesholmsån River upstream Degeberga WWTP.
- Downstream Degeberga WWTP a number of pharmaceuticals were detected. The highest concentrations were Carbamazepine up to 52 ng/L, Diclofenac up to 7.8 ng/L, and Oxazepam up to 8.6 ng/L
- Based on the outlet concentrations and information about volume treated wastewater at Degeberga WWTP the yearly amounts of the investigated pharmaceuticals release could be calculated to be 345 g Carbamazepine, 89 g Diclofenac, 67 g Oxazepam and 17 g Metoprolol. All other pharmaceuticals were below 3 g yearly.

Table 1. Concentration of selected pharmaceuticals in inlet and outlet water from Degeberga WWTP as well as upstream and downstream the WWTP in the Segesholmsån River, which is the catchment area for the released wastewater. Removal efficiency (%) for the various pharmaceuticals is also shown in the table along with the yearly load to the recipient (g/year). Data collected from Deliverable 4.1 of the MORPHEUS project.

Pharmaceutical compound	Upstream WWTP Summer/winter		Concentration in WWTP				Removal efficiency		Outlet chemical load from WWTP to recipient	Downstream WWTP summer/winter	
			inlet		outlet						
			summer/winter		summer/winter		summer/winter				
	ng/L		ng/L		ng/L		%		g/year	ng/L	
Atenolol	-	-	3,701	2,955	-	2.1	100	100	0.1	-	-
Azithromycin	-	-	34	155	0.7	12	100	92	0.5	-	-
Carbamazepine	-	-	5,663	4,589	5,052	3,673	11	20	345	52	15
Ciprofloxacin	-	-	918	8,816	7.0	66	99	99	2.9	0.6	-
Clarithromycin	-	-	128	0.4	7.2	0.8	94	No data	0.3	-	-
Diclofenac	-	-	2,515	1,070	821	1,442	67	-35	89	7.8	5.7
Erythromycin	-	-	67	3.1	53	7.4	21	-136	2.4	0.6	-
Estrone	-	-	75	109	-	0.1	100	100	0.0	0.3	0.2
Ibuprofen	-	-	307,278	153,666	-	3.2	100	100	0.1	-	-
Metoprolol	-	-	3,469	3,456	304	128	91	96	17	2.6	-
Naproxen	-	12	1,893	5,301	21	13	99	100	1.4	-	-
Oxazepam	-	-	1,236	1,075	825	866	33	19	67	8.6	3.7
Paracetamol	-	-	38,018	46,936	-	3.0	100	100	0.1	-	-
Propranolol	-	-	55	98	11	36	80	63	1.9	-	-
Sulfamethoxazole	-	-	-	2.3	-	6.2	No data	-167	0.2	-	-

3.3 The general measures and decision-making criteria for Degeberga WWTP

According to point 1.1, the decision-making criteria for implementation of advanced treatment in Degeberga WWTP are given in Table 2.

Table 2. Decision making criteria for implementation of advanced treatment in Degeberga WWTP. Data collected from Deliverable 5.1 of the MORPHEUS Project and Degeberga WWTP Environmental Report 2016.

Criteria	Degeberga WWTP
WWTP Catchment measures	
Number inhabitants connected to WWTP	Not stated, but Degeberga Population was 1,350 persons 2016
WWTP catchment area	Mainly households, no industrial wastewater
WWTP receiver	Segesholmsån River and Hanöbukten Bay in the Baltic Sea
Receiver protection	Nature preservation area and UNESCO Biosphere reserve “Vattenriket”
Fraction of wastewater in the receiver flow	Average yearly dilution factor is ca 0.006 but varies greatly over the year. Dilution factor is much smaller in the summer.
Possible impact on water resources	Possible impact on the sensitive aquatic species of the river
Technological measures	
WWTP flow designed; average flow 2016	23 m^3/h; 9 m^3/h
WWTP load PE designed; current	2000 PE; 950 PE
WWTP total treated yearly volume 2016	79 000 m^3
WWTP size	Small
WWTP mechanical treatment	mechanical screens and aerated grit chamber, then the waste water is pumped to an activated sludge process.
WWTP biological step	traditional activated sludge process with denitrification basin, where the wastewater is mixed with sludge from the chemical treatment step, followed by aerated basin (nitrification step). In this step the water is mixed with sludge from the clarifier (sedimentation step right after the biological treatment).
WWTP chemical step	PIX (ferrous chloride) dosing system is used for phosphorus removal; flocculation process is followed by sedimentation basins. The sludge from chemical treatment process is returned to the denitrification step.
WWTP post-treatment	open sand filter for a final polish
WWTP effluent parameters (in 2016)	TSS = -, COD = 15 mg O_2/dm^3, BOD_7 = 1.5 mg O_2/dm^3, N_{tot} = 13.2 mg N/dm^3, P_{tot} = 0.16 mg P/dm^3

WWTP sewage sludge management	The excess sludge from the sedimentation step after the biological treatment step is stored in a sludge storage magazine, and thereafter pumped manually out into reed beds. In the reed beds the sludge is going through dewatering, mineralization and hygienization processes.
Available space	Yes
Presence of qualified personnel	Yes
Pharmaceutical burden	
Pharmaceuticals in WWTP inflow	Ibuprofen up to 307 µg/L, Paracetamol up to 47 µg/L, Ciprofloxacin up to 8.8 µg/L, Carbamazepine up to 5.7 µg/L and Naproxen up to 5.3 µg/L
Pharmaceuticals in WWTP outflow	Carbamazepine up to 5.0 µg/L, Diclofenac up to 1.4 µg/L, Oxazepam up to 866 ng/L and Metoprolol up to 304 ng/L
Effectiveness of current technology in pharmaceuticals removal	The lowest (<33%) or even negative removal was observed for Carbamazepine, Erythromycin, Oxazepam and Sulfamethoxazole. Large seasonal variations in removal was detected for Diclofenac (67% summer and -35% winter) and Erythromycin (21, -136%).
Pharmaceutical indicators	
Pharmaceuticals of high (>90%) and stable removal rate at both seasons	Atenolol, Azithromycin, Ciprofloxacin, Estrone, Ibuprofen, Metoprolol, Naproxen and Paracetamol.
Pharmaceuticals of low (<33%) removal rate during at least one season	Carbamazepine, Diclofenac, Erythromycin, Oxazepam and Sulfamethoxazole
Additional relevant measures	
presence of bromide	Not tested
presence of chrome	Not tested
presence of N-nitrosodimethylamine	Not tested
presence of DOC	Not tested
Other measures	-
Stakeholder perspective	The method should be robust, easy to use and it should not cause any disturbance of existing processes. Consequently, the possibility of using existing infrastructure without changes is highly beneficial since it lowers cost of investments. This means that technologies that can be added as an extra fourth treatment step are preferable. The technology should be proven to reduce pharmaceutical efficiently over time.

According to the decision-making criteria, Degeberga WWTP is a modern WWTP, with a well-functioning treatment process, which fulfills the current discharge requirements. Two technologies could be considered; ozonation and granulated activated carbon (GAC). GAC differs from ozonation in the sense that it physically removes organic contaminants from the wastewater as compared to ozone that transforms compounds to new compounds by chemical reactions. In this respect one of the benefits with GAC is that no potentially harmful new compounds are produced when filtering through GAC. The implementation of ozonation would also require some kind of extra step such as a sand filter or a pond of water after the ozonation step before releasing the water to the recipient. This is to assure that there are no additional reactive chemicals left after the ozonation process. Based on this, and the below discussion, GAC was considered the most suitable choice of technology at Degeberga WWTP and the sensitive Segesholmsån River.

3.4 GAC unit for Degeberga WWTP – full-scale study at a small-scale WWTP

Kristianstad Municipality has previously made available its largest WWTP (Centrala Reningsverket Kristianstad, treating water corresponding to 118,000 PE) for a now completed experiment where researchers Ola Svahn and Erland Björklund from Kristianstad University together with personnel at the WWTP conducted full scale long-term advanced treatment study with granulated activated carbon (GAC), using sand as a pre-filtering step. This was part of a large governmental funding (via the Swedish Agency for Marine and Water Management) for testing various types of advanced wastewater treatment systems between 2014-2019. For 3 years personnel at Kristianstad WWTP operated the large pilot-scale GAC unit. In this project nearly 50.000 m^3 of wastewater was treated, corresponding to 50.000 BV. The pilot plant had a filter size of 1 m^3 GAC preceded by 1 m^3 sand. The flow rate in the pilot-plant was 2m^3/h. This combination of sand and GAC in a serial filter showed very good purification results over time for a huge majority of the 22 investigated pharmaceutical residues. Among others, diclofenac could be removed by more than 90% over a time span of 1 year with this set-up. It was also shown to be very robust, with a minimum of maintenance. The sand-filter was backflushed a few times a week, while the GAC filter never required any kind of backflushing. As a consequence of this project, Kristianstad municipality now also has gained knowledge and access to qualified personnel for running a GAC-filter.

Degeberga WWTP is already equipped with suitable existing treatment processes as well as a final polishing step of sand-filtration, which is feasible for a GAC unit application. There is also plenty of space for placement of the GAC filter at the WWTP site.

To cover the costs for constructing the GAC filter at Degeberga WWTP, Kristianstad municipality applied for governmental funding (via the Swedish Environmental Protection Agency) that were made available in 2018. In the application for funding Kristianstad WWTP expressed the following purpose and expected benefits:

"The purpose of carrying out drug treatment at Degeberga ARV is partly to improve the environment, and thus dramatically reduce the ecological stress caused by pharmaceutical residues, in Segesholmsån River downstream of the treatment plant and in the sea where the river flows into Hanöbukten Bay in Friseboda nature preservation area on the east coast. On the one hand, the purpose is to gain experience for the industry as a whole about drug purification at

smaller treatment plants. But also, to some extent, for Kristianstad Municipality's own part, to gain their own experience of investing in and operating such a facility before an inquiry into a possible pharmaceutical treatment at Centrala Reningsverket i Kristianstad (CRV)."

Kristianstad Municipality also describes why they would like to start with a smaller WWTP before working at a larger scale by the following arguments:

"Investing in pharmaceutical treatment at a small treatment plant becomes more expensive per purified cubic meter of water compared to investing in a larger treatment plant. At the same time, the cost in absolute terms is considerably lower, and it can therefore be cost-effective for the state, overall, to first gain experience at smaller plants, before investing in large treatment plants. The project can also contribute to future decisions regarding whether and when drug treatment may become relevant for smaller treatment plants, <2000 PE more generally."

The cost of investment for the large pilot-plant described above, running 2m^3/h through 1 m^3 GAC was roughly 100.000 EUR. The overview and principle of this large pilot-scale system is shown in Figure 4. A photo of the pilot-scale plant in operation is seen in Figure 5.

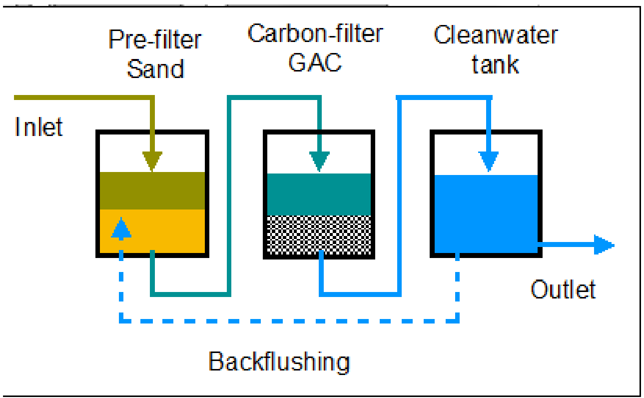

Figure 4. Design of the large pilot-scale treatment plant that was evaluated at Kristianstad Municipality with 50.000 m^3 waste water at a flow of 2m^3/h, using a Triton® bottom filter for good throughput. The system consists of 2 filter units that can be connected serially or in parallel for increased flexibility, followed by a clean water tank. The pilot-scale plant was designed by Måns Hansson at Malmberg Water AB.

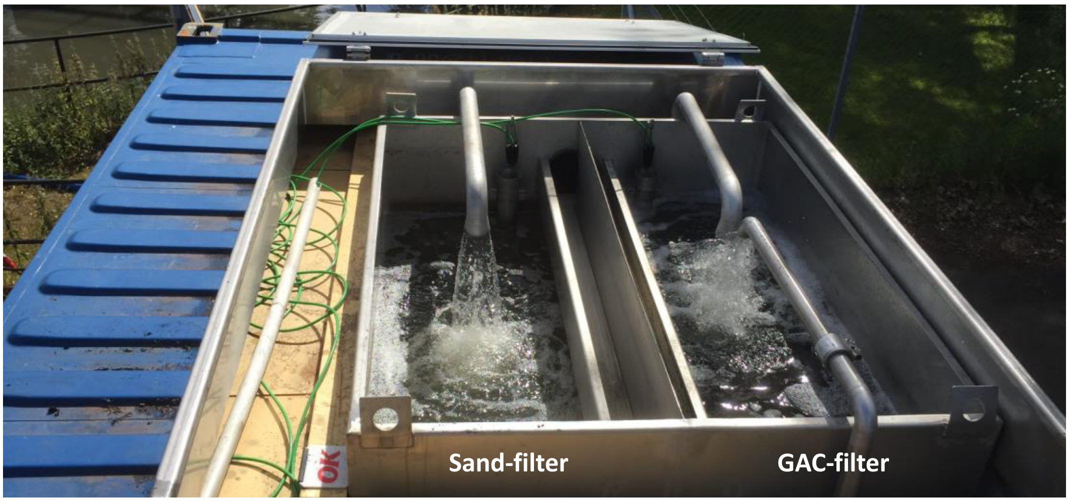

Figure 5. Pilot-scale plant in operation at a flow of 2m^3/h. To the left 1 m^3 of sand as a pre-filter step followed by 1 m^3 GAC to the right. Photo: Erland Björklund.

In Degeberga the size of the GAC-filter will be eleven times larger. Degeberga is dimensioned for 22 m^3/h even though it is only running 9 m^3/h on average. Yet, the fourth step will be designed to take nearly the full capacity of Degeberga, which means going from 2 m^3/h to ca. 22 m^3/h. The same type of filter as the one described above in Figure 4 and Figure 5 will be connect the existing WWTP effluent, right after the already existing sand filter at Degeberga WWTP. The investment costs of building a 22 m^3/h filter system on-site at Degeberga were stated in the application sent by Kristianstad Municipality to the Swedish EPA as shown in Table 3 below.

Table 3. Estimated costs of investment for a GAC-filter for treatment of 22 m^3/h of wastewater at Degeberga WWTP's effluent.

	Total WWTP effluent
Investment cost	**Approximatively**
Constructing GAC-filters on-site	925,000
Sand-filters for pre-treatment	Already existing
Civil work	160,000
Total	1,085,000

The investment costs are estimated to be between 1.0-1.1 million EUR.

When it comes to operation costs, they are to a large extent connected with the replacement or reactivating of the granulated activated carbon. Prices may differ as well as the time that a specific carbon can treat the wastewater from pharmaceuticals before breakthrough. Given prices of activated carbon given by supplier are in the order 800-1,000 EUR/m^3. Degeberga would require 11 m^3 to fill the GAC-filters. With a price of 900 EUR/m^3 this would mean 9,900 EUR. Previous investigations have shown that the carbon will last roughly 1 year. Assuming a flow of 22 m^3/h for a whole year would mean a total of 192,720 m^3/year. This would give a cost of 0.051 EUR/m^3 or ~ 0.05 EUR/m^3. At the moment the flow at Degeberga is 9 m^3/h which menas that if the carbon only is burdened with half the volume of wastewater per hour it will last longer which would reduce the cost. Additionally, there is a possibility that the carbon will last longer than one year, which would further decrease costs. A solid and sound micro pollutant residue analysis campaign during one year of operation will therefore be an important to tool to estimate the life span, and hence the costs, of the GAC. This procedure is highly recommended for all advanced treatment, no matter choice of technique.

Furthermore, the carbon can be reactivated several times, which might decrease the cost substantially. The price tag of 0.05 EUR/m^3 is therefore at the high end. The price per cubic meter is therefore more likely somewhere in the span of 0.025-0.050 EUR/m^3 (Table 4).

Apart from the activated carbon there might be increased costs of energy at the WWTP when using GAC. These costs are somewhat uncertain, but in a report from the Swedish EPA it was estimated that for larger WWTPs (<100,000 PE) the energy consumption would increase by 2-10 % (1-6 kWh/PE, year). Assuming 5 kWh/PE and that Degeberga has a population of 2,000 PE (which is the maximum capacity), this would mean a total of 10000 kWh extra per year. The price of 1 kWh is roughly 0.10 EUR, meaning extra costs per year of 1,000 EUR. This would add an extra 0.0052 EUR/m³ or ~ 0.005 EUR/m³ (Table 4). Energy demands estimated by the company building the GAC-filter showed that the primary consumption was related to pumping water. The costs for this was ca 1.1 EUR/day meaning a total cost per year of 402 EUR. This in turn would lead to a cost of only 0.0021 EUR/m³. The power consumption is therefore estimated to range between 0.0025-0.0050 EUR/m³ (Table 4).

Finally, there will be some extra costs associated to man power. From the previous large-scale pilot project there was basically no maintenance on the GAC-filter. The only thing to consider was back-flushing the sand-filter a few times a week. However, this work has to be done even without the GAC-filter in place. It is estimated that on average 1 h/week is sufficient, giving 52 extra working hours per year, which is around 0.33 month of work. Assuming that the costs for this 0.33 month is around 2,000 EUR (all costs included) it would add 0.010 EUR/m³ (Table 4).

Table 4. Estimated costs of operation for a GAC unit for treatment of 22 m³/h of wastewater at Degeberga WWTP's effluent.

	Total WWTP effluent
Operating costs	**Approximatively**
GAC filter replacement	~ 0.025-0.050 EUR per m³
Power consumption	~ 0.0025-0.0050 EUR per m³
Civil work	~ 0.01 EUR per m³
Total	~ 0.0375-0.0650 EUR per m³

The total costs of operation for treating 1 m³ waste water with GAC would add up to be in the order of 0.0375-0.0650 EUR/m³. However, this is an uncertain figure, and part of the installation of GAC at Degeberga WWTP is to get new information about the actual costs associated with advanced treatment. This will aid in the process of possibly upgrading the largest WWTP in Kristianstad (Centrala Reningsverket, CRV) with advanced treatment, thereby having better control of costs.

3.5 Pharmaceuticals suggested for Degeberga WWTP as indicators of advanced treatment effectiveness

To assess the removal efficiency of pharmaceuticals by GAC, some selected compounds (indicators) should to be tested in the influent of Degeberga WWTP and in the effluent of the GAC unit. For Degeberga WWTP a minimum could be Carbamazepine, Erythromycin, Diclofenac, Oxazepam and Metoprolol as indicators, based on the removal efficiency data in Table 1 above. These are present in measurable concentrations in raw wastewater, and are persistent to removal by conventional treatment based on activated sludge technology. However, the list of indicators is not limited these but others could be chosen based on other criteria.

4 Conclusion

Degeberga WWTP (Degeberga Avloppsreningsverk) is a well-functioning small object, which is the major source of pharmaceuticals to its recipient - Segesholmsån River. Thus, the implementation of advanced treatment technology at Degeberga WWTP is expected to nearly completely reduce the anthropogenic stress posed on this sensitive river as well as to reduce the chemical burden to the Hanöbukten bay of the Baltic Sea.

Currently, Degeberga WWTP is equipped with a sand-filtration as a final polishing step, and therefore feasible for implementation of a granulated activated carbon (GAC) filter as a fourth treatment step. This technology is appropriate for Degeberga WWTP because it is a module-like add-on technology which is relatively easy to install. As a result, the GAC filter does not require highly experienced staff for conducting the process and maintenance. The knowledge and experience in advanced treatment gained during implementation and exploitation of GAC can be used in the future, e.g., for scaling up the process. Additionally, GAC treatment effectiveness against pharmaceuticals can also be verified in situ, and provides a factor of human resources engagement. Furthermore, the large reduction of pharmaceuticals and other micropollutants release is expected to be beneficial on the recipient quality. Thus, the natural environment recovery should be carefully monitored to find out the timeframe of this process.

Project partners

Kristianstad University, SE
www.hkr.se

Gdansk Water Foundation, PL
www.gfw.pl

EUCC
The Coastal Union Germany, DE
www.eucc-d.de

Gdansk University of Technology, PL
www.pg.edu.pl

University of Rostock, DE
www.auf.uni-rostock.de

Environmental Protection Agency, LT
www.gamta.lt

Klaipeda University, LT
www.ku.lt

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