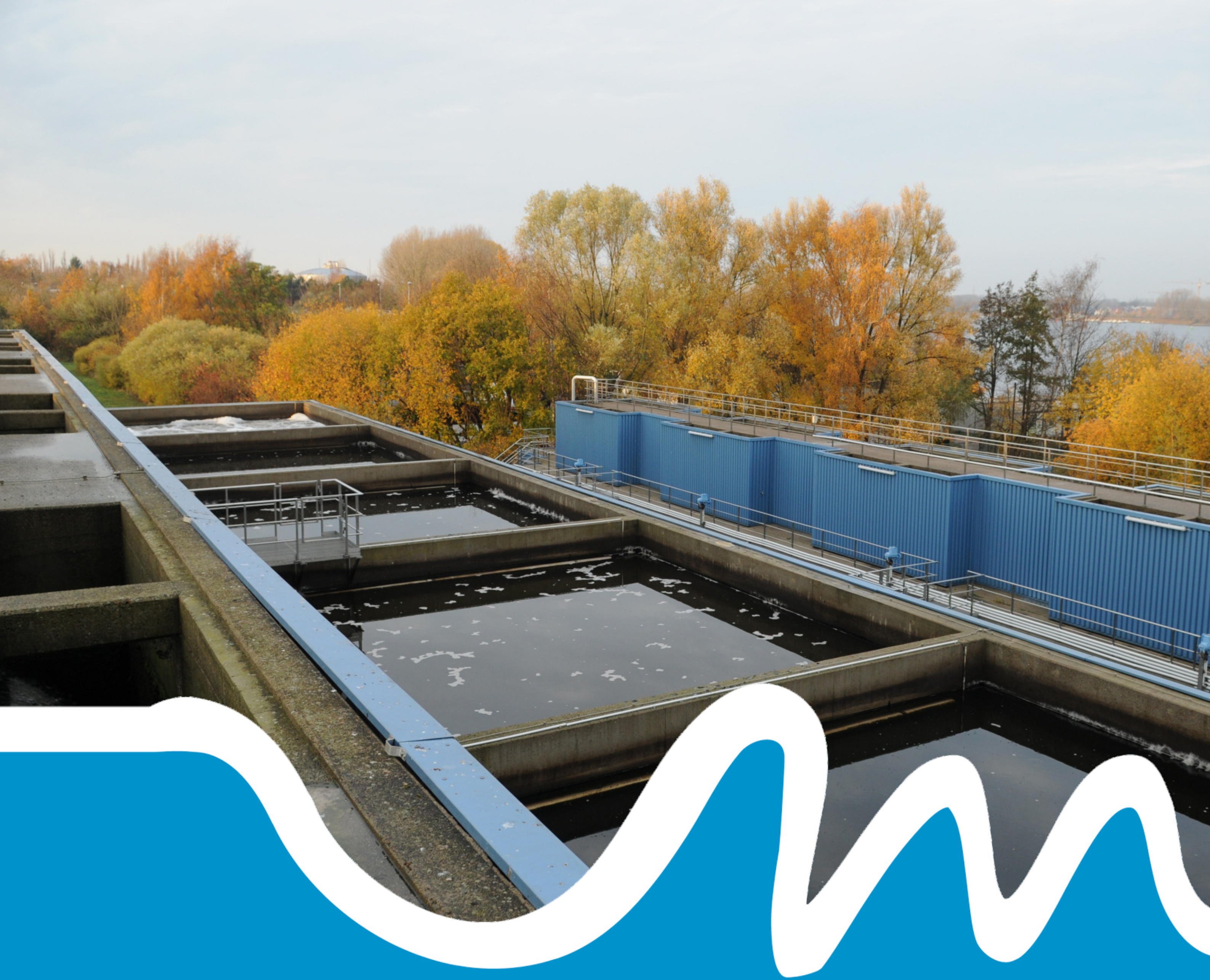

Advanced pharmaceuticals removal from wastewater - Roadmap for the model site Rostock wastewater treatment plant

Project MORPHEUS 2017 - 2019

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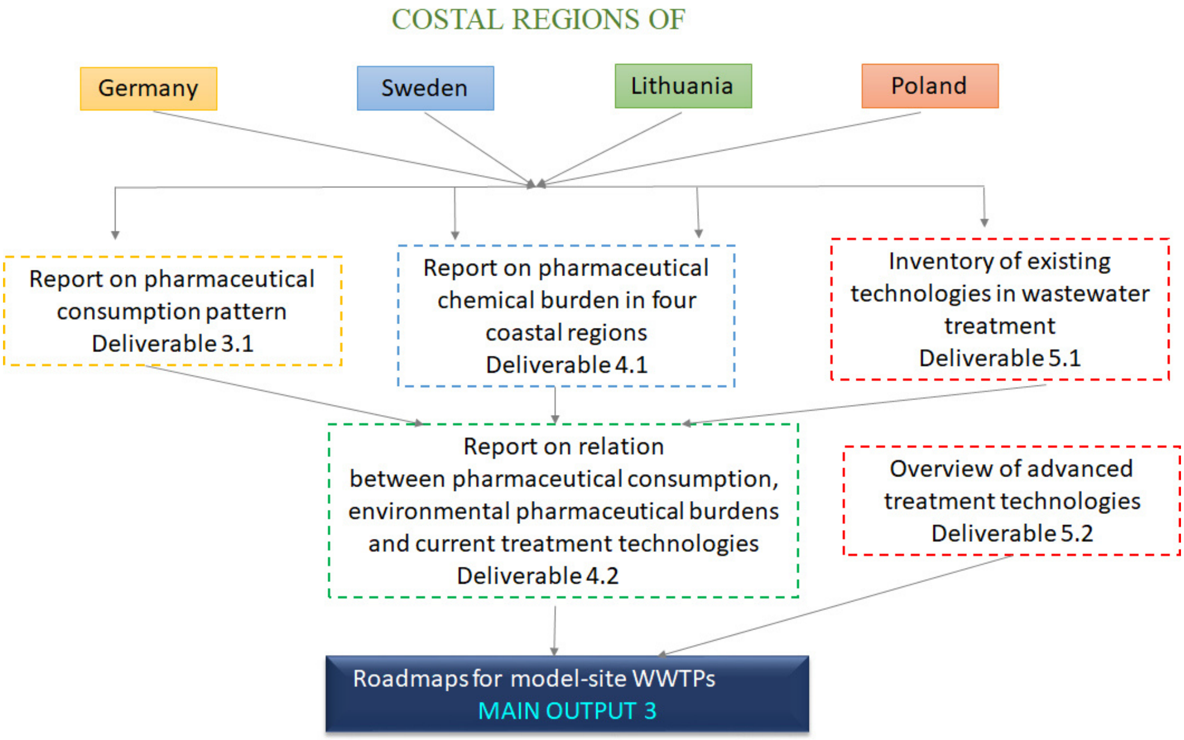
Cover photo

WWTP Rostock, BIOFOR® reactor basins. Photo: Franziska Stoll

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.

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 below).

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: Advanced pharmaceuticals removal from wastewater - roadmaps for model-site Degeberga wastewater treatment plant case study[7]).

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[8]).

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

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

MORPHEUS project as well as the pilot investments of advanced GAC treatment implemented in 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[9])

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.

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[9] 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).

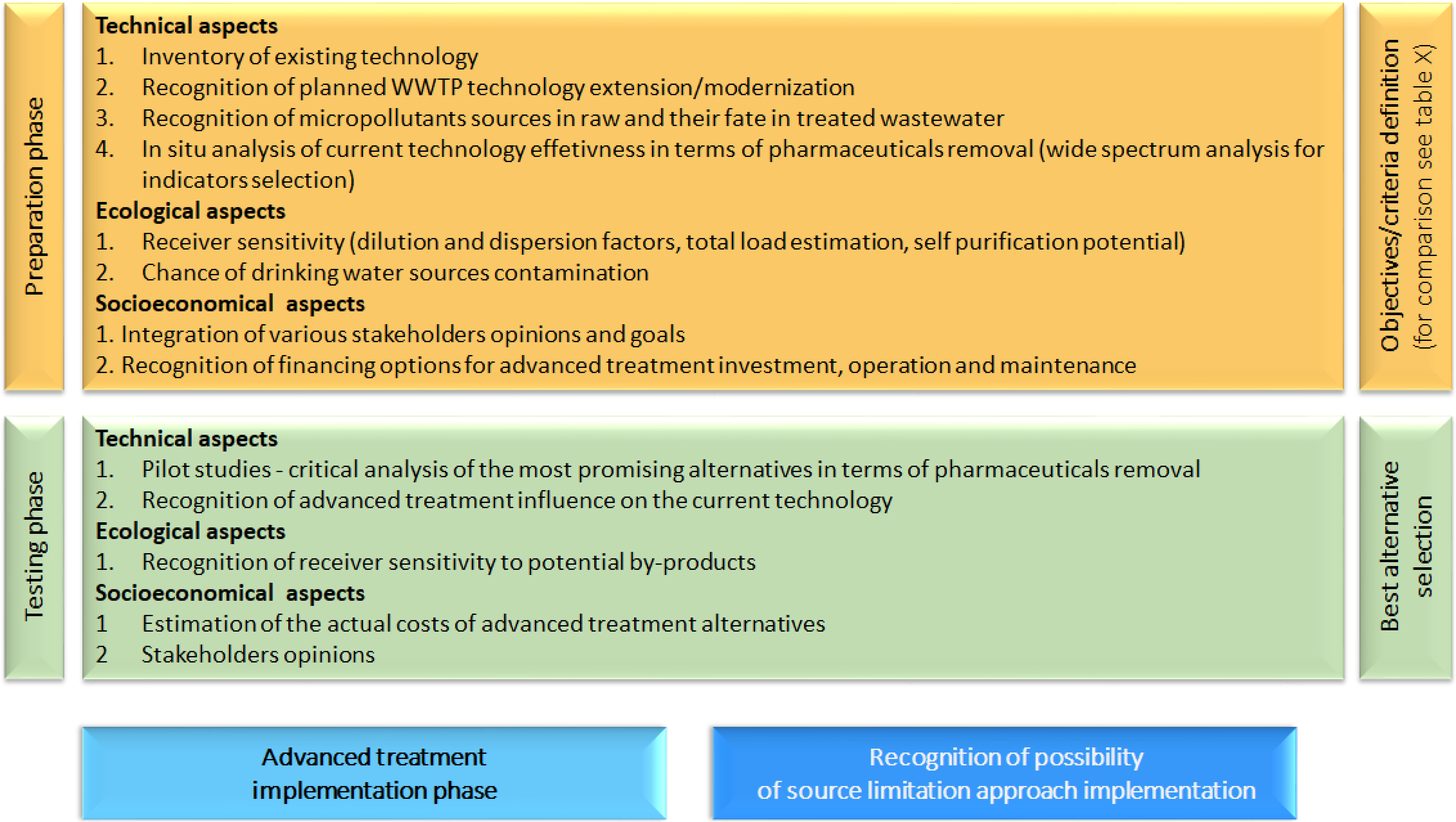
It can be concluded that the existing wastewater treatment systems, based on activated sludge, are not effective enough to remove most of the investigated pharmaceuticals and mixtures of pharmaceuticals are constantly discharged into the receiving water bodies. According to the obtained data, selected 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.



Preparation phase

Technical aspects

1. Inventory of existing technology
2. Recognition of planned WWTP technology extension/modernization
3. Recognition of micropollutants sources in raw and their fate in treated wastewater
4. In situ analysis of current technology effetivness in terms of pharmaceuticals removal (wide spectrum analysis for indicators selection)

Ecological aspects

1. Receiver sensitivity (dilution and dispersion factors, total load estimation, self purification potential)
2. Chance of drinking water sources contamination

Socioeconomical aspects

1. Integration of various stakeholders opinions and goals
2. Recognition of financing options for advanced treatment investment, operation and maintenance

Objectives/criteria definition
(for comparison see table X)

Testing phase

Technical aspects

1. Pilot studies - critical analysis of the most promising alternatives in terms of pharmaceuticals removal
2. Recognition of advanced treatment influence on the current technology

Ecological aspects

1. Recognition of receiver sensitivity to potential by-products

Socioeconomical aspects

1 Estimation of the actual costs of advanced treatment alternatives
2 Stakeholders opinions

Best alternative selection

Advanced treatment implementation phase

Recognition of possibility of source limitation approach implementation

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 [10]

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

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[10] 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[11]

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[12]

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[13]

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)

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

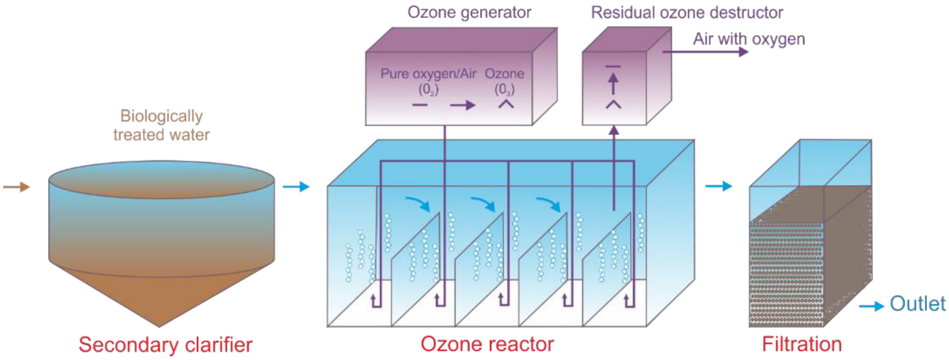

Figure 2. Schematic overview of the ozone unit suggested location in the conventional wastewater treatment technology (modified from[14])

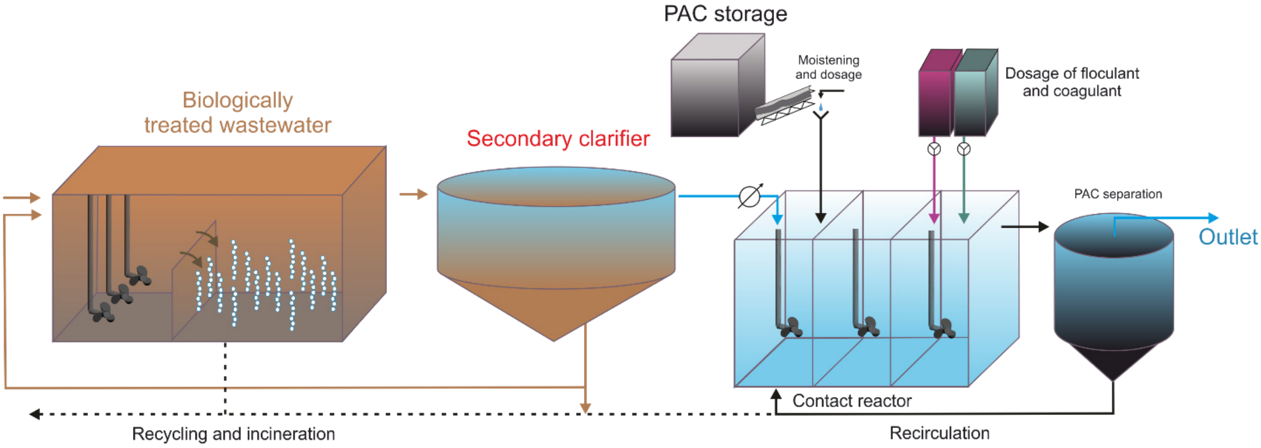

Figure 3. Schematic overview of the PAC unit suggested location in the conventional wastewater treatment technology (modified from[15])

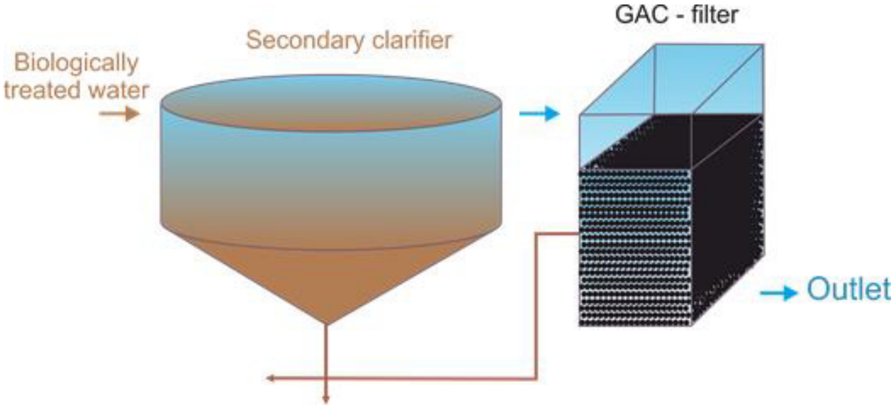

Figure 4. Schematic overview of the GAC unit suggested location in the conventional wastewater treatment technology (modified from[16])

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[14] 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.

[15] as in [13] Abegglen & Siegrist (2012)

[16] as in [13] Abegglen & Siegrist (2012)

3 Roadmap for WWTP Rostock

3.1 Relevance of WWTP Rostock for emission of human pharmaceuticals to the South Baltic

Rostock is located directly at the coast. The discharge point of the WWTP is in the estuary of the river Warnow, called Unterwarnow, which is about 15 km south of its mouth into Baltic (see Figure 1). Within the selected German model area (Baltic Sea Catchment within the Federal State Mecklenburg-Vorpommern, which is mainly rural area), WWTP, Rostock is the largest plant and treats a yearly average wastewater load about 335.000 PE. Besides industrial effluents, domestic wastewater of 235,645 inhabitants (data 2015) equal to nearly 15% of the population within the federal state (or 19% of population within Baltic Sea Catchment/model area Mecklenburg, respectively) is treated here. Additionally, two of the largest hospitals in the federal state and several other health and care facilities are located in the city and discharge into this plant. Consequently, the highest loads of pharmaceuticals causing a burden to the environment of the South Baltic Sea are emitted by WWTP Rostock which has also been investigated in WP4 (Del. 4.1). In WP3, this was also confirmed by analysis of consumption patterns depending largely on the number of connected inhabitants (Del. 3.1). Not surprisingly, the highest total loads of investigated pharmaceuticals were discharged via WWTP Rostock, namely up to 17 kg/a Carbamazepine, 36.1 kg/a Diclofenac and 24.6 kg/a Metoprolol. Among the four model WWTPs in the German model area, the corresponding removal rates of these pharmaceuticals showed the lowest level (-19%, 40% and 90%, respectively). Based on the mass-flow analysis for a selection of four pharmaceuticals (see Del. 4.2), both measured and predicted loads in the influent (MEC/PEC) confirmed that according to discharged loads into the South Baltic Sea WWTP Rostock represents the highest priority for introducing an advanced treatment technology for removal of micropollutants including pharmaceuticals.

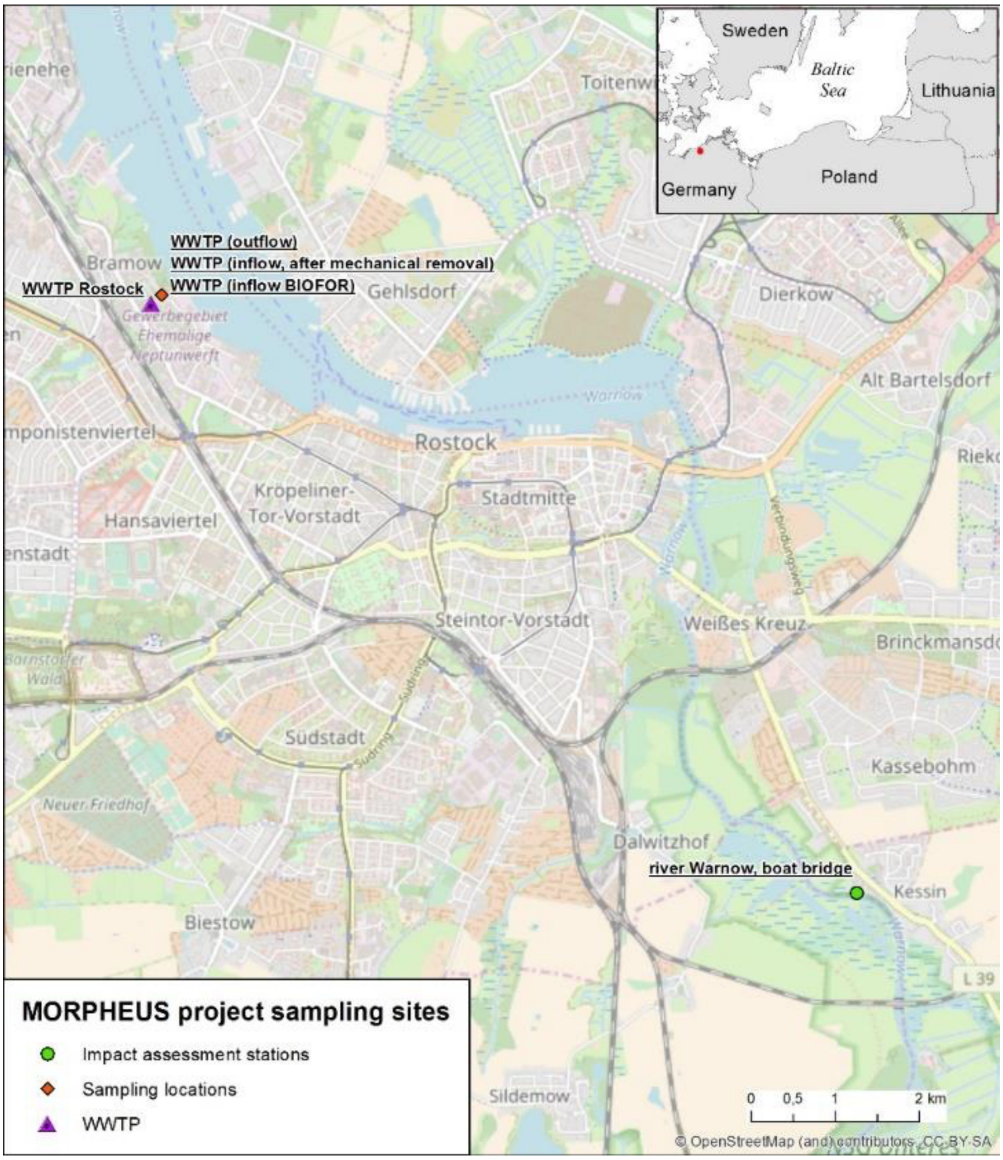

Figure 1: Map of Rostock with WWTP Rostock

3.2 Technology and operational parameters of the WWTP Rostock

Fehler! Verweisquelle konnte nicht gefunden werden. gives a technological overview of the WWTP Rostock. Following a conventional mechanical treatment (consisting of rake, aerated sand trap and primary clarifier), the biological treatment is performed in two technological units in series. The main treatment is performed in an activated sludge unit with pre-denitrification and enhanced biological P-removal (according to the Johannesburg procedure). In the so-called BIOFOR® reactor basins, the effluent of the secondary clarifiers is post-treated in two biological filter units: (1) nitrification and (2) denitrification with Methanol dosage. With the current operation conditions the filters rather provide a polishing function (N, P and VSS) than an intense biological treatment.

The primary and excess sludge are thickened separately and anaerobically digested. The digested sludge is dewatered with centrifuges and incinerated in external incineration plants.

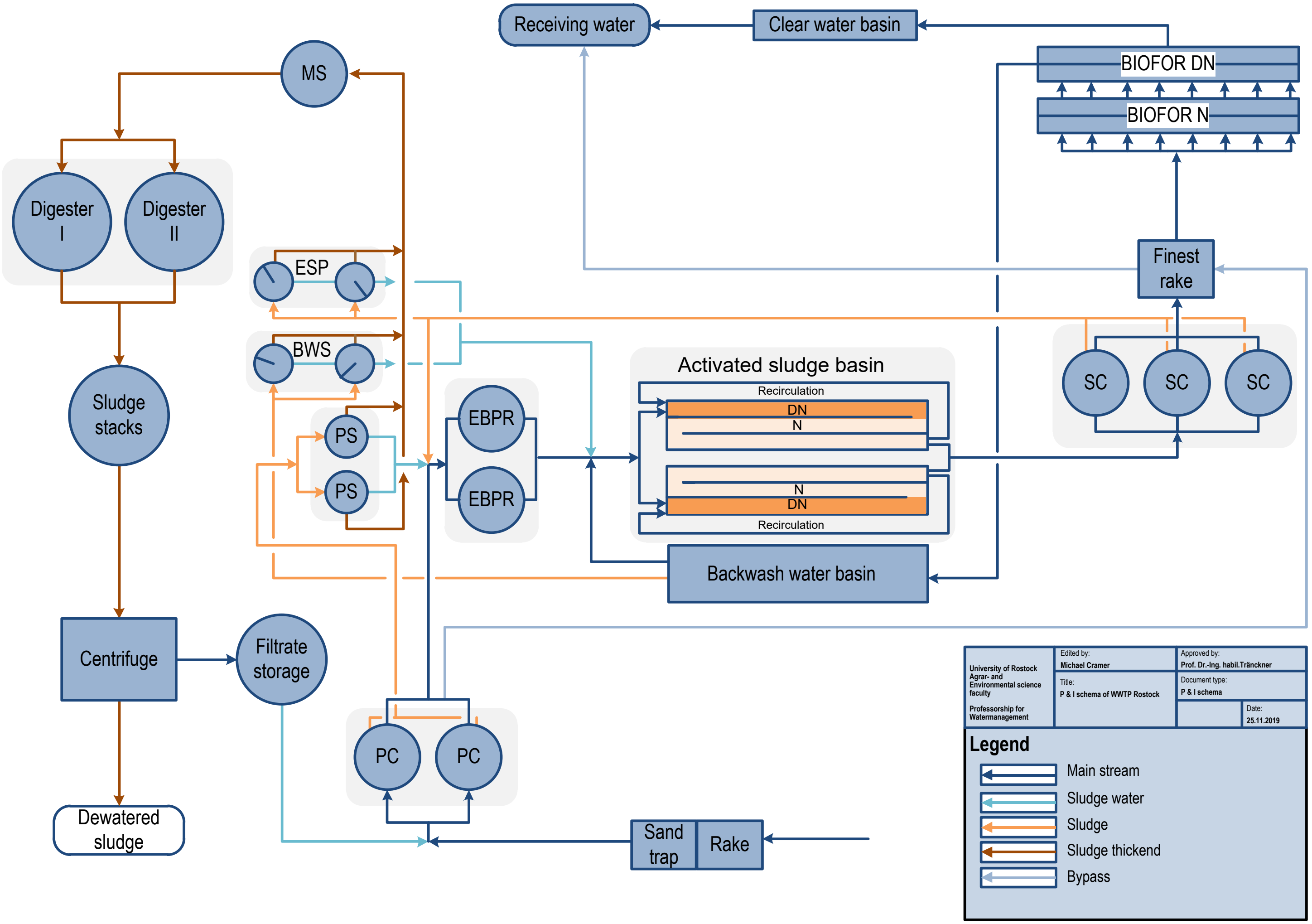

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Figure 2: Map of the WWTP

In the next years, an incineration plant for about 25,000 tons sludge (dry matter), will be installed next to the WWTP Rostock. The vapours of the sludge drying will then be treated at WWTP Rostock, too.

Table 1: Operational parameters of the WWTP Rostock

Parameter		Value	Remarks
Q_{max}		7,000 m^3/h	Design value
Q_{DW}		39,000 m^3/d	yearly average
$B_{COD,in}$ (DW)		40,800 kg/d	40,756
$BT_{KN,in}$ (DW)		3,220 kg/d	3,220
$B_{P,in}$ (DW)		420 kg/d	419
V_{AST} (total) thereof:			
	$V_{anaerob}$	8,800 m^3	
	V_{DN} (Winter)	10,000 m^3	can varied from 0 m^3 to 4.000 m^3
	V_N (Winter)	20,000 m^3	can varied from 16.000 m^3 to 30.000 m^3
TSS_{AST}	Winter	4.2 kg/m^3	
	Summer	3.3 kg/m^3	
ESP	max.	11,160 kg/d	
	min.	8,930 kg/d	
SRT	min.	12.4 d	Seasonal and operational variation. Minima occur generally end of winter.
	max.	13.7 d	
Aerobic SRT	min.	4.9 d	
	max.	5.9 d	
Effluent SC = Influent BIOFOR			as load:
COD		48 mg/L	$B_{COD,in}$ = 2,270 kg/d
BOD_5		7.5 mg/L	B_{BOD5} = 373 kg/d
TSS		8.0 mg/L	B_{AFS} = 390 kg/d
NO_2-N		0.3 mg/L	B_{NO2-N} = 15.5 kg/d
NH_4-N		1.2 mg/L	B_{NH4} = 62.3 kg/d
total P		0.4 mg/L	B_P = 17.8 kg/d
BIOFOR-N			12 filter cells
A_F		876 m^2	Filter area
h_F		4.0 m	filter height
V_F		3,504 m^3	filter volume
BIOFOR-DN			12 filter cells
A_F		876 m^2	Filter area
h_F		2.5 m	filter height
V_F		2,628 m^3	filter volume

4 Options for upgrading with a treatment step for removal of organic micropollutants ("4th treatment step")

4.1 Generally available options

Del. 5.2 "Overview of advanced technologies in wastewater treatment for removal of pharmaceuticals and other micropollutants" gives a detailed overview of existing treatment options for the removal pharmaceuticals in domestic WWTPs. Generally, it can be distinguished into oxidation (using Ozone) and adsorption processes (using activated carbon). Both have their special application conditions and require in most cases a post-treatment. Their introduction into an existing plant has to be therefore well reflected. Relevant criteria for the choice of the best suited technology are summarized as follows:

- Elimination of relevant pharmaceuticals
- Existing and potentially usable technology and built infrastructure
- Space demand and availability
- Sludge disposal
- Synergy effects for improving other effluent parameters (COD, VSS, P) and for disinfection
- Holistic energy balance (including external energy demand, e.g. for activated carbon production/reactivation)
- additional manpower requirement
- only for Ozone: Potential of Bromate formation if relevant for the water body or downstream water usages

The elimination/transformation rate of ozone oxidation and activated carbon differs depending on the regarded substance (see Del. 5.2). However, both technologies have proved to achieve a high reduction above 80% for a broad spectrum of compounds. A detailed weighing depending on the wastewater composition and the treatment objectives is currently difficult, since Germany is still lacking a list of agreed indicator substances. Therefore, an equality of both technologies with regard to elimination is postulated, here, and the discussion is focused on the technological feasibility and sustainability.

In the following the technologies and design rules are characterized. The next chapter will refer to this, discussing an upgrade for WWTP Rostock.

4.2 Design data: flow

The design flow for an advanced treatment step has to be agreed upon with the environmental agency (here Staatliches Amt für Landwirtschaft und Umwelt Mittleres Mecklenburg, StALU-MM) since it mainly depends on the ambient water conditions. If no restriction exists with regard to the sensitivity of the water bodies, the advanced treatment step for removing micropollutants should at least treat the yearly dry weather flow Q_{DW} [17]. Since the recipient Unterwarnow and this part of the South Baltic Sea are no protected habitat and provide a rather efficient and high dilution, a treatment design for yearly dry weather flow should be a reasonable choice. Higher flows under stormwater conditions have to be bypassed without advanced treatment.

Hence, according to KOM-M_NRW [18] the design flow for the treatment step shall be equal to the yearly average of the maximum hourly dry weather flow ($Q_{DW,hmax}$). Higher resolved flow data to derive this value directly were not available for this study. However, past time series analysis show that $Q_{DW,hmax}$ does not exceed 2 times $Q_{DW,hmean}$. For further discussion, a reasonable design flow is estimated with the following parameters:

Yearly average dry weather flow: 39,000 m³/d

Peak flow:

$$Q_{Design} = 2 \cdot \frac{Q_{d,DW}}{24} = 2 \cdot \frac{39\,000\ \text{m}^3/\text{d}}{24\ \text{h/m}^3} = 3250\ \text{m}^3/\text{h}$$

4.3 Oxidation with Ozone

4.3.1 Function and general application conditions

Ozone (O_3) has a high oxidation potential and reacts fast and selectively with a multitude of functional groups by splitting of one oxygen atom. In parallel, Ozone decays in water by forming $OH^{\bullet}$- radicals. These have an even higher oxidation potential and react unselectively with nearly all compounds. Accordingly, they are caught also by those “harmless” compounds (scavengers). In wastewater these are mainly carbonate and bicarbonate compounds, organic background matter (rest COD, i.e. DOC). Ozone itself can also oxidise nitrite but not ammonia. So, prerequisite for an efficient ozone application is a far-reaching biological wastewater treatment. Therefore, ozonation shall be installed at least at the effluent of a secondary clarifier of a fully nitrifying activated sludge system. An additional filtration unit before ozonation can further reduce the VSS and DOC background concentration and thus the required ozone dosage. High sludge age is advantageous far going COD/DOC reduction.

The specific ozone demand is given in a range of 0.6 to 0.9 mg O_3/mg DOC [19] [20]. In most cases ozone is generated from liquid oxygen (LOX). In this technology, about 10% of the oxygen will be transformed into ozone. For a fairly complete decay, the ozone reactor shall be designed for a detention time of 15 to 30 minutes. Plug-flow type reactors provide the most efficient use of reactor volume. They can be approximated by a cascade of completely stirred tank reactors or zic-zac reactors with guiding walls. Gas-liquid-exchange increases nearly proportionally with the depth of ozone diffusers. For an efficient ozone dispersion, a minimum reactor depth of 5 meters is

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[17] KOM-M_NRW (2016). *Anleitung zur Planung und Dimensionierung von Anlagen zur Mikroschadstoffelimination, 2. Auflage*, Kompetenzzentrum Mikroschadstoffe NRW

[18] KOM-M_NRW (2016). *Anleitung zur Planung und Dimensionierung von Anlagen zur Mikroschadstoffelimination, 2. Auflage*, Kompetenzzentrum Mikroschadstoffe NRW

[19] Abbeglen C. and Siegrist H. (2012). *Mikroverunreinigungen aus kommunalem Abwasser; Verfahren zur weitergehenden Elimination auf Kläranlagen*, Hrsg.: Bundesamt für Umwelt (BAFU), Schweiz, Umwelt-Wissen Nr.1214.

[20] Barjenbruch M. and Firk W. (2014). Möglichkeiten der Elimination von Spurenstoffen auf kommunalen Kläranlagen. Korrespondenz Abwasser 61(10), 861-75.

recommended. The reactor basin must be covered gas seal and the gas space has to be exhausted continuously. The off-gas must be completely decomposed in an "ozone destroyer".

Since ozone only reacts with certain functional groups, the pharmaceuticals are not completely mineralized but transformed into metabolites. To further degrade these transformation products, a downstream biological treatment is required. In most cases, biofilm systems (biofilter, moving/fixed bed systems) are proposed. But also polishing ponds (e.g. WWTP Bad Sassendorf in NRW, Germany) are successfully applied. The combination of ozonation and biological post treatment has an advantageous effect on COD removal (10-60% depending on the ozone dosage and the post treatment).

4.3.2 Integration of Ozone at WWTP Rostock

At first sight, the best location to introduce ozonation at WWTP Rostock is between secondary clarifier and BIOFOR-N (see Figure 3). This way, the biofilter can be used for post biological treatment of transformation products and decay of possibly remaining ozone. A proximate design of ozonation at WWTP Rostock is summarized in Table 2

So far DOC is not monitored in the effluent of the secondary clarifier, but can be roughly estimated from the COD or BOD_5 value. Metcalf.&.Eddy [21] propose a ratio BOD_5/DOC of 0.2 to 0.5.

For the design of the ozone generator this yields to a required ozone dosage of 10 to 31 mg/L.

$$\begin{aligned} c_{O3} &= \left(0.6...0{,}8\ \mathrm{mgO_3/mg\ DOC}\right)\cdot\left(15...37.5\ \mathrm{mg/L\ DOC}\right) + 3{,}43\ \mathrm{mgO_3/mg\ NO_2\text{-}N}\cdot 0{,}33\ \mathrm{mg/L\ NO_2\text{-}N} \\ &= 10...31\ \mathrm{mg/L} \end{aligned}$$

With the determined design flow of 3,250 m³/h, this would require an ozone mass transfer of 33 to 100 kg/h. This wide range is mainly due to the high uncertainty concerning DOC. To reduce this range, a DOC monitoring campaign is currently prepared. The ozone reactor requires a volume about 1600 m³.

The built transformation products and left ozone could be treated at the BIOFOR-N. As side effect, an additional COD removal between 5 to 25 mg/l can be expected.

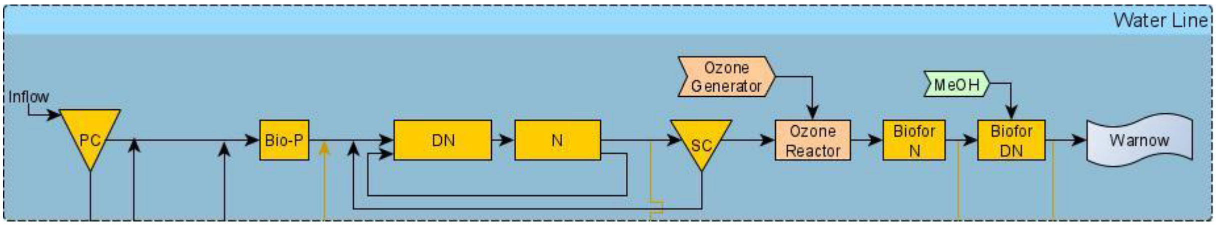

Figure 3: Integration of Ozonation into the WWTP Rostock

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[21] Metcalf.&.Eddy (2014). *Wastewater Engineering - Treatment and Resource Recovery*. Mac Graw Hill Education. ISBN 978-0-07-340118-8

Table 2: Approximate design parameters for ozonation at WWTP Rostock

Parameter	Value	Remarks
Specific Ozone concentration	0.6…0.8 g O_3/g DOC	according to Barjenbruch and Firk (2014)
Ozone demand for NO_2-N oxidation	3.43 mg O_3/mg NO_2-N	
Contact time at Ozone reactor	15…30 min.	
Reactor depth	5.0 m	
Ozone concentration at the product gas	10% (≙ 148 g/Nm^3)	
BOD_5/DOC	0.2…0.5	
Design flow	3,250 m^3/h	
BOD_5 at dosage point	7.5 mg/L	
NO_2-N at dosage point		
Estimated DOC	15…37.5 mg/L	
design Ozone concentration	10…31 mg/L	for DOC and nitrite
design Ozone mass flow	33…101 kg/h	
Required design Ozone Flow	222…684 Nm^3/h	
design Oxygene demand	2,220…6,840 Nm^3/h	
Volume of ozone reactor	813…1,625 m^3	
Required area	163…325 m^2	

However, the greatest constraint for an ozonation at this point of the WWTP is the required post-denitrification at the BIOFOR-DN. Since about 90% of the produced gas is oxygen, the oxygen concentration at the inflow to the BIOFOR-N will exceed by far 10 mg/l and may reach even values of 30 mg/l. These concentrations will be reduced only to a very limited extent at the BIOFOR-N. The theoretical methanol demand can be expressed by

$$B_{MeOH} = 2{,}47 \cdot NO_3\text{-}N + 1{,}53 \cdot NO_2\text{-}N + 0{,}87 \cdot O_2$$

The consumption of those high oxygen concentrations using methanol, in order to subsequently denitrify 1.5 mg/L nitrate would be rather uneconomic if ever feasible with the available reaction time. So before installing ozonation in front of the BIOFOR filters, alternative solutions for the nitrogen removal have to be developed. Options are here the further optimization of the first treatment step in combination with process water treatment in the sludge line.

If the BIOFOR-DN is given up for nutrient removal, a conversion into a granulated activated carbon filter (GAC) could be considered. This would combine both basic advanced treatment processes, oxidation and adsorption with an expected high elimination efficiency. The positioning of the GAC behind ozonation and biological depth filtration would provide an advanced pre-treatment for DOC and solid matter, resulting in long Empty Bed Contact Time (EBCT) and backwash intervals. The process combination of ozonation and GAC is currently intensively investigated. The partial fractioning of large molecules by ozone can lead to significant better adsorption performance of the GAC [22]. It may be advantageous to work with rather low ozone

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[22] Reungoat J., Macovca M., Escher B. I., Carswell S., Mueller J. F. and Keller J. (2010). Removal of micropollutants and reduction of biological activity in full scale reclamation plant using ozonation and activated carbon filtration. *Water Research* **44**(2), 625-37.

doses for an optimal adsorption process. For the special situation of two filters in series, a fairly efficient biological mineralization of transformation products can be expected.

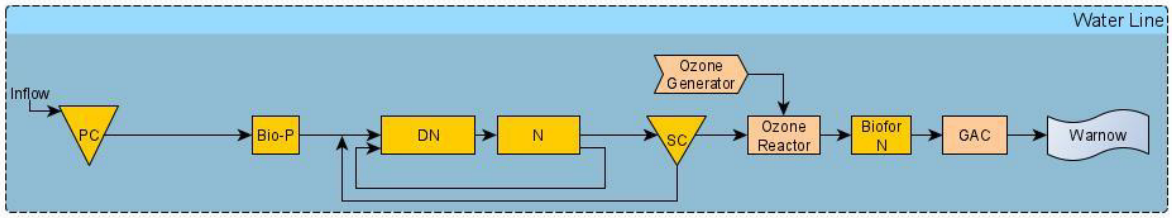

Figure 4: Optional combination of Ozonation and GAC (scheme without sludge line)

4.4 Powdered Activated Carbon (PAC)

4.4.1 Function and general application conditions

Dissolved micropollutants can be adsorbed onto the surface of activated carbon. The intensity of adsorption depends strongly on the characteristics of the compound: biologically well degradable, polar compounds with low molecular weight are generally poorly adsorbed, while large nonpolar molecules are preferably adsorbed. Other important influencing parameters are the concentration of the pollutant, the contact time and the DOC background concentration. In wastewater treatment, both technologies powdered activated carbon (PAC) and granulated activated carbon are applied (GAC).

PAC is directly dosed into the wastewater. The overall technological objective is an efficient loading with short contact time and a subsequent complete separation of the PAC from the cleaned wastewater. This can be implemented at a WWTP in different ways. Generally, it can be distinguished into:

- Direct dosing of PAC in the activated sludge tank (AST) + rest filtration behind the secondary clarifier
- PAC dosing in the effluent of the secondary clarifier
    - Two step separation: Sedimentation tank + filtration
    - One step separation filtration

The direct dosing of PAC in the AST is hardly applied so far. The potential advantage is the low demand for additional infrastructure. If a tertiary sand filtration exists, the WWTP has to be expanded only by the PAC dosage. However, due to the mixture into the sludge liquor the PAC loading efficiency is rather low, leading to higher specific PAC demand (20 to 60 mg/l), compared to dosing into the effluent of a biological treatment stage.

The most common PAC technology at WWTP is the dosage into the effluent of a secondary clarifier. Depending on existing infrastructure, there are different technological options. One option is the installation of a contact basin with sedimentation chamber and recirculation of PAC. The PAC recirculation decouples the hydraulic retention time from the contact with the recirculated PAC. The hydraulic retention time should be minimum 30 minutes. Excess PAC is often recirculated into the biological stage for rest loading and removed with the excess sludge. Due to the insufficient PAC separation by sedimentation, a final filtration is required. Flocculation followed by sand filtration is often used. Cloth filters are also applicable. Alternatively, the pore volume of a depth filter can be used as contact volume for the PAC. This requires a good adjustment of the filter media to the PAC grain/floc size in order to completely use the available filter bed volume. The hydraulic contact time should be in same range as for contact basins (30 min.). The backwash water can then be recirculated to the biological treatment stage for further adsorption and removed with the excess sludge. The dosage in the effluent of the secondary clarifier requires significantly less PAC. According to operational experience a good removal of pharmaceuticals can be achieved with a PAC dosage of 10 to 20 mg/L[23]. These data also show a parallel COD reduction of 25 to 55%.

At WWTP Rostock, PAC dosing in the effluent of the secondary clarifier with an additional sedimentation tank and filtration is not further considered due to economic reasons. The application of the present BIOFOR-N filter is recommended in order to save resources and design a sustainable concept for WWTP Rostock.

A special and very compact solution is PAC dosage into a membrane bioreactor. This combines a particle free effluent with the option of operating with high sludge age to maximize biological degradation of organic micropollutants. Due to the infrastructural conditions of WWTP Rostock, this is not an option.

4.4.2 Integration of PAC at WWTP Rostock

Direct dosage at AST

When dosing PAC directly at the AST, the dosage station point should be installed at the end of the nitrification volume, with lowest concentrations of COD. The final filtration could be performed by the BIOFOR-N. However, this would require the dosage of flocculants in front of the filter. Probably the existing dosage point for chemical P removal could be used for this purpose.

Table 3 gives a summarized overview of the design and potential constraints. Operational information on direct PAC dosing at an AST are rare. The widely varying information on required dosage (see above) was slightly reduced for the design (20…40 mg/L). Dosage in the AST will lead to accumulation of PAC at the sludge liquor. As well-known from simultaneous Phosphorous precipitation, this leads to very delayed response of the whole system, which is rather controlled by the total accumulated mass of reactant than by the current dosage. Designing such a system for peak conditions is rather uneconomic. Therefore, the average dry weather flow is chosen here as design flow. With the specific dosage this leads to a daily PAC demand of 780…1560 kg/d which directly contributes to sludge production. Without adapting the total suspended solids (TSS) concentration at the AST, this would reduce the sludge age by 0.8 to 1.5 days and the aerobic sludge age by 0.4 to 0.8 days, respectively. Since PAC improves settle ability, a slight increase of TSS_{AST} could be feasible to mitigate this effect. The sludge of WWTP Rostock is already incinerated. The adsorbed pharmaceuticals will be safely destroyed via this disposal. The disposal costs will increase proportionally to the additional sludge productions by 7 to 14%.

Summarised, PAC dosage at the AST should be feasible without large infrastructural and operational changes but is related with a high PAC demand. Since PAC is the decisive item of

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[23] Metzger S., Tjoeng I., Rößler A., Schwentner G. and Rölle R. (2014). Kosten der Pulveraktivkohleanwendung zur Spurenstoffelimination am Beispiel ausgeführter und in Bau befindlicher Anlagen. *Korrespondenz Abwasser, Abfall* **61**(11), 1029-37

consumable expenditures, this option will be related rather high operational (OPEX) but comparably low capital expenditures (CAPEX).

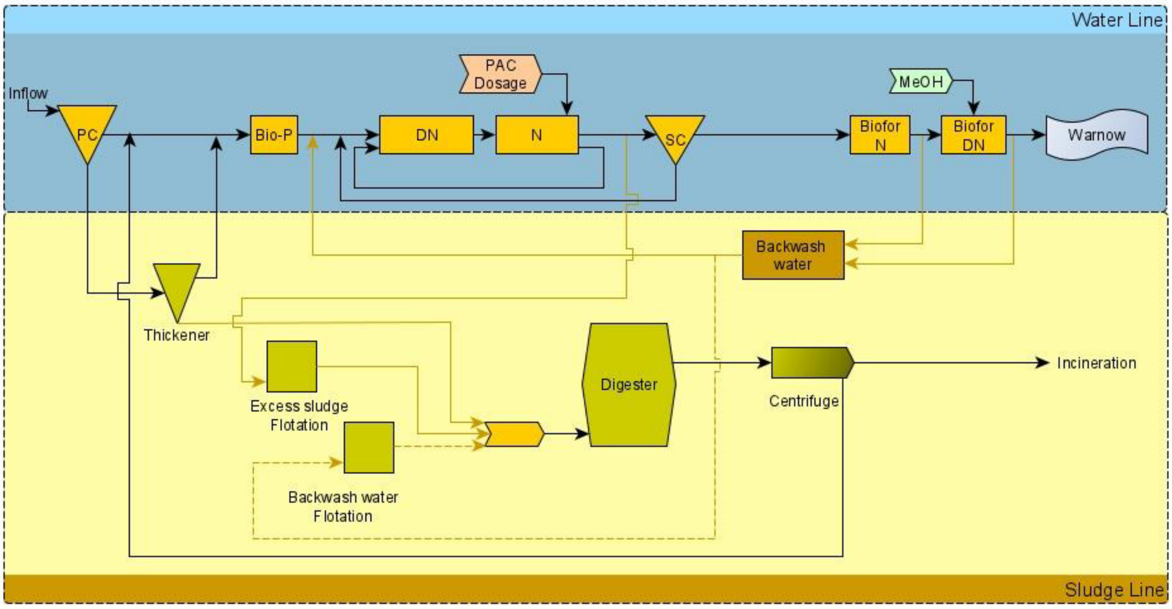

Figure 5 Direct dosage of PAC into AST

Table 3: Approximate design of direct PAC dosage at the activated sludge tank

Parameter	Value	Remarks
Specific PAC dosage	20…40 mg/L	Estimated from literature
Design flow	39,000 m^3/d	
Current max sludge production	11,940 kg/	
Current min sludge age	12.4 d	
Current min aerobic sludge age	5.9 d	
Specific PAC dosage	20…40 mg/L	Estimated from literature
Required daily PAC dosage	780…1,560 kg/d	
Max. sludge production with PAC dosage	11,940…12,720 kg/d	PAC dosage = additional sludge production
Min. SRT with PAC dosage	10.9…11.6 d	Proportional reduction due to increased sludge production, without adaptation of TSS_{AST}
Min. aerobic SRD with sludge dosage	5.1…5.5 d	

Dosage between secondary clarifier and BIOFOR-N

The more common way of PAC is the dosage at the effluent of the secondary clarifier. For the WWTP Rostock, generally two options are feasible:

- Direct dosage in front of the BIOFOR-N
- Dosage into a contact basin with sedimentation and PAC-recirculation

Figure 6 gives a scheme of the first option. Here, the BIOFOR-N serves as both, the contact volume and the PAC separation from the treated wastewater. The required PAC dosage is estimated here with 10 mg/L. In contrast to the dosage at the AST, the peak flow is decisive here. For peak conditions 32.5 kg/h PAC have to be dosed. The daily demand is about 390 kg/d.

The BIOFOR-N has a media height of 4.0 m. To provide an EBCT of 30 minutes the filter velocity should not be faster than 8 m/h. For the design flow, this value is reached, when 6 cells are in operation. Assessing the efficiency of PAC separation and the provision of contact volume requires technological investigation. But even if a rather efficient usage of the pore volume for solid matter retention can be achieved, the required backwash interval will significantly increase. The backwash water can either be conveyed to the sludge treatment directly or recirculated to the AST. This would reduce the current minimal sludge retention time (SRT) to about 11 days and the aerobic SRT to 5.3 days, respectively. The effluent COD will be reduced from currently about 50 mg/L to about 25-35 mg/L.

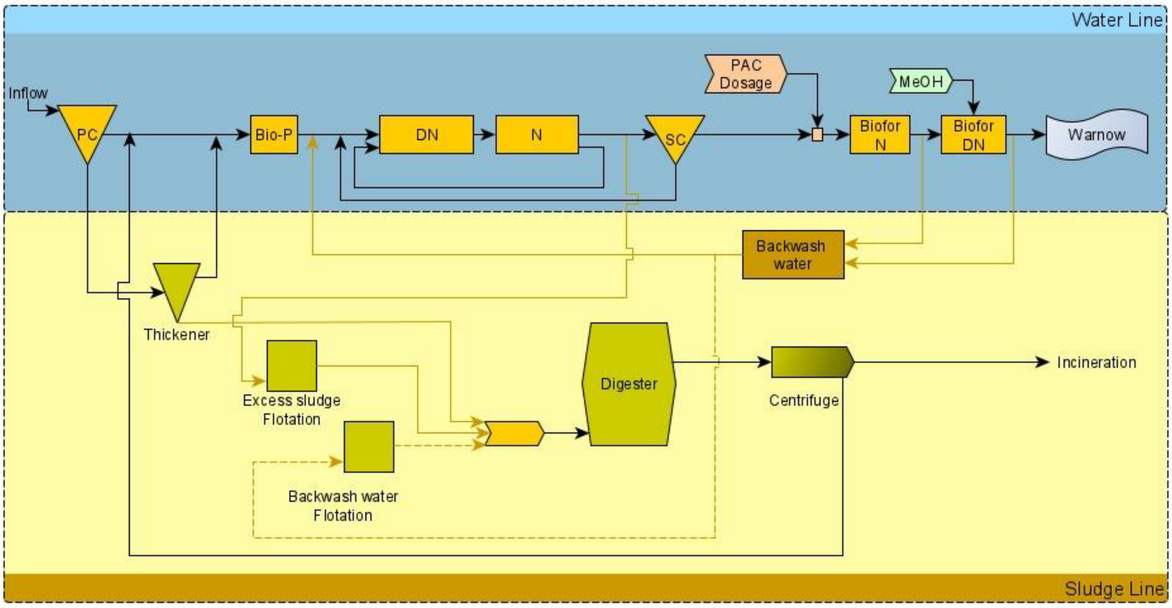

Figure 6: PAC dosage at the influent of BIOFOR-N

Parameter	Value	Remarks
Specific PAC dosage	10 mg/L	
Design flow (peak)	3,250 m³/h	
Design flow (average)	39,000 m³/d	
PAC dosage	10 mg/L	
Hydraulic retention time at contact basin (option 1: separate contact reactor with PAC recirculation)	30 min.	
Volume of contact basin	1,625 m³	
Hydraulic retention time at the BIOFOR-N (option 2: direct dosing)	30 min.	
Required v_{max} to achieve HRT	8 m/h	
Specific PAC dosage	10 mg/L	
Required daily PAC dosage	390 kg/d	
Max. sludge production with PAC dosage	11,550 kg/d	PAC dosage = additional sludge production
Min. SRT with PAC dosage	5.3 d	Proportional reduction due to increased sludge production, without adaptation of TSS_{AST}

4.5 Granulated Activated Carbon (GAC)

4.5.1 Function and general application conditions

Dissolved micro-pollutants can be adsorbed onto the surface of activated carbon. The intensity of adsorption depends strongly on the characteristics of the compound: biologically well degradable, polar compounds with low molecular weight are generally poorly adsorbed, while large nonpolar molecules are preferably adsorbed. Other important influencing parameters are the concentration of the pollutant, the contact time and the DOC background concentration. In wastewater treatment, both technologies powdered activated carbon (PAC) and granulated activated carbon are applied (GAC).

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The most common PAC technology at WWTP is the dosage into the effluent of a secondary clarifier. Depending on existing infrastructure, there are different technological options. One option is the installation of a contact basin with sedimentation chamber and recirculation of PAC. The PAC recirculation decouples the hydraulic retention time from the contact with the recirculated PAC. The hydraulic retention time should be minimum 30 minutes. Excess PAC is often recirculated into the biological stage for rest loading and removed with the excess sludge. Due to the insufficient PAC separation by sedimentation, a final filtration is required. Flocculation followed by sand filtration is often used. Cloth filters are also applicable. Alternatively, the pore volume of a depth filter can be used as contact volume for the PAC. This requires a good adjustment of the filter media to the PAC grain/floc size in order to completely use the available filter bed volume. The hydraulic contact time should be in same range as for contact basins (30 min.). The backwash water can then be recirculated to the biological treatment stage for further adsorption and removed with the excess sludge. The dosage in the effluent of the secondary clarifier requires significantly less PAC. According to operational experience a good removal of pharmaceuticals can be achieved with a PAC dosage of 10 to 20 mg/L [24]. These data also show a parallel COD reduction of 25 to 55%.

A special and very compact solution is PAC dosage into a membrane bioreactor. This combines a particle free effluent with the option of operating with high sludge age to maximize biological degradation of organic micro-pollutants. Due to the infrastructural conditions of WWTP Rostock, this is not an option.

GAC filtration is performed in depth filters. The used GAC grain size varies between 0.5 and 4.0 mm. GAC should be applied for biologically well treated wastewater with low DOC. Generally, they are at least located at the effluent of the secondary clarifier. Since GAC provide an efficient solid matter retention, the inflow to the filter should be nearly suspense free, if possible TSS < 15 mg/L. Increased solid matter loads would result in frequent backwashing, which can damage the mechanically fragile activated carbon. Therefore, an advanced suspense/DOC removal before GAC can be advantageous. In WWTP generally three technological options are available:

- GAC without preceding solid matter removal (i.e. secondary clarifier + GAC)
- conventional solid matter retention before GAC (i.e. floc filter or biological filter + GAC)
- membrane filtration before GAC

As usual in depth bed filtration, the decisive design parameters are filter depth [m] and filter velocity [m/h]. Filter velocities from 2 to 8 m/h are recommended [25]. Required values of EBCT are in the range of 15 to 30 minutes with slight dependency on the DOC background concentration. The operational costs depend strongly on the achievable bed volumes (BV) before breakthrough

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[24] Metzger S., Tjoeng I., Rößler A., Schwentner G. and Rölle R. (2014). Kosten der Pulveraktivkohleanwendung zur Spurenstoffelimination am Beispiel ausgeführter und in Bau befindlicher Anlagen. *Korrespondenz Abwasser, Abfall* **61**(11), 1029-37

[25] KOM-M_NRW (2016). *Anleitung zur Planung und Dimensionierung von Anlagen zur Mikroschadstoffelimination, 2. Auflage*, Kompetenzzentrum Mikroschadstoffe NRW

of the GAC. The BV depend strongly on the regarded substance, the DOC background and the operational conditions. Values between 7,000 to more than 40,000 BV are reported in literature[26].

Backwash of GAC is delicate. It is required to remove retained solid matter and to reduce biofilm but can lead to abrasion of the GAC material. If periodic backwash is required, GAC with sufficient mechanical resistance should be selected. The backwash procedure has to be adapted accordingly.

Integration of GAC at the WWTP Rostock

For the WWTP Rostock, the most obvious integration of GAC would be the transformation of the BIOFOR-N into a GAC filter (see Figure 7). This would require to replace the current biolite media (burned clay material with a grain size of 1.0…2.5 mm) by GAC.

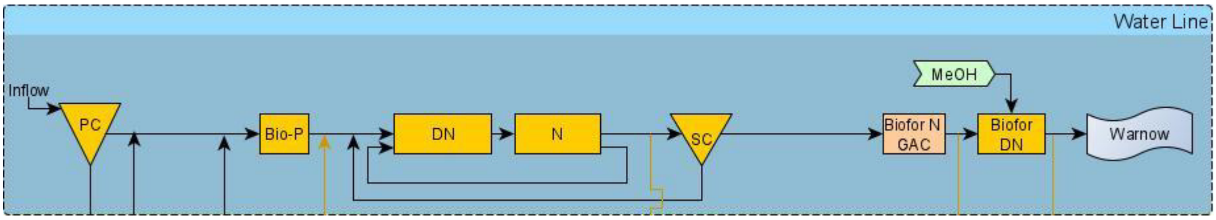

Figure 7: Transformation of BIOFOR-N to GAC filtration

An according "reference plant" is the WWTP Obere Lutter [27]. At this WWTP a macro-porous GAC (Aquasorb 5000) with a grain size of 0.63-2.36 mm on a GAC support layer of 2.0…4.75 mm was selected (see Figure 8). The filter candle was maintained. In large full-scale tests with varying operational conditions (filter velocity, continuous vs. discontinuous operation etc.) a generally adaptability of this filter type to GAC was proven. It could be shown that operation with low constant flow velocity of 2 m/h and according discontinuous operation of filter cells led to a better utilization of adsorption capacity (14,000-16,000 BV) than operation with varying filter velocity (2-8 m/h) (13,000 BV). Compared to other published data this is in a lower range. In parallel to the intended elimination of micropollutants, a COD removal of about 45% was observed. The backwash procedure was successfully changed from combined air-water flushing to pure water flushing. Losses of filter media were not observed. The operational costs were calculated with 0.09 €/m³.

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[26] Benström F., Nahrstedt A., Böhler M., Knopp G., Montag D., Siegrist H. and Pinnekamp J. (2016). Leistungsfähigkeit granulierter Aktivkohle zur Entfernung organischer Spurenstoffe aus Abläufen kommunaler Kläranlagen. *Korrespondenz Abwasser, Abfall* **63**(4), 276-88

[27] Nahrstedt A., Burbaum H., Mauer C., Alt K., Sürder T. and Fritzsche J. (2014). Einsatz granulierter Aktivkohle auf dem Verbandsklärwerk "Obere Lutter". *KA-Korrespondenz Abwasser* **61**(5), 408-26.

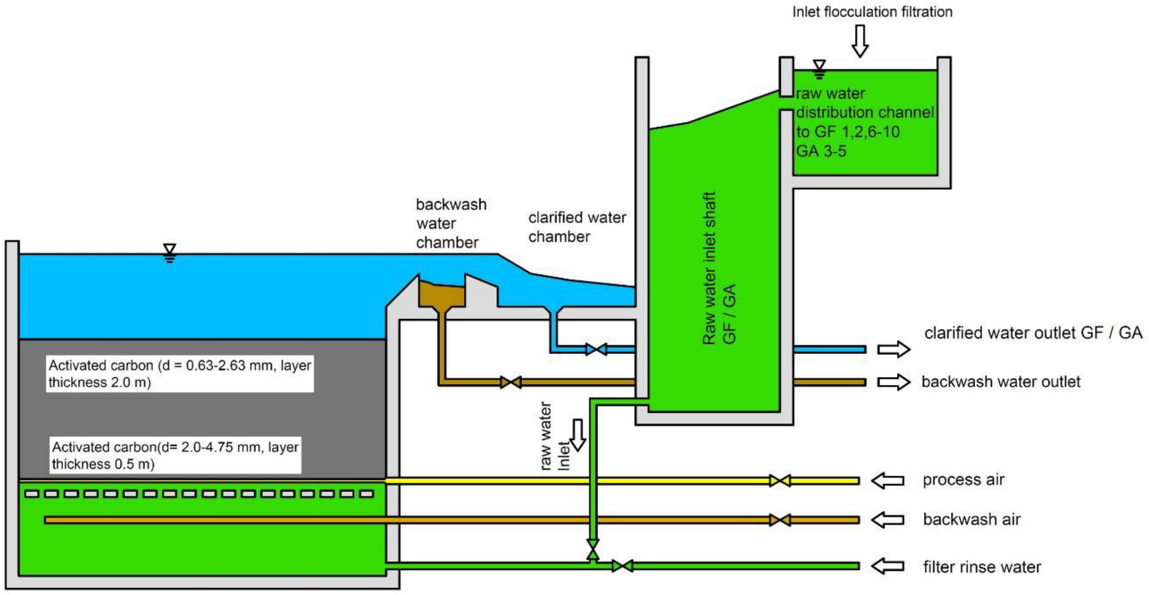

Figure 8: Scheme of a transformed BIOFOR-GAC filter [28]

Applying the characteristic design parameters and operational experience of other plants to WWTP Rostock shows that a conversion of the BIOFOR-N into GAC is generally feasible (Table 4). An EBCT of 30 minutes would require a maximum velocity of 8 m/h. For the design peak flow, this is assured if only half of the filter area is in operation. If all cells (without being in backwash) are in operation, a filter velocity of 2 m/h is achieved at average design flow. In a review of 34 full scale GAC filters (Benström *et al.* 2016), achievable BV treated were in range of 5,000 to 15,000 (with outliers in both directions). Taking this range, the service life of one GAC filling would be 4 to 11 months. The expected rather high DOC background concentration is adverse but can partly be compensated by a fairly long EBCT of in average more than 30 minutes. Based on the experience of Nahrstedt *et al.*[29], the COD concentration could be reduced by about 20 mg/l.

Currently the BIOFOR-N is only treating ammonia in peak load situations. This function could generally be maintained when operation as GAC. However, under extreme hydraulic load conditions, a raw water bypass around the activated sludge step is designated to avoid an overloading of the secondary clarifiers. In this case, raw wastewater would be directly conveyed to the GAC and impair significantly the adsorption capacity. To enable a sustainable GAC filtration, this operation would not be applicable further. Accordingly, the sedimentation step of the activated sludge step had to be enforced before converting the BIOFOR-N into GAC.

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[28] As above
[29] As above

Table 4: Approximate design of GAC at the current BIOFOR-N

Parameter	Value	Remarks
Design flow (peak)	3,250 m³/h	Relevant for v_{max} and minimum EBCT
Design flow (average)	39,000 m³/h	Relevant for BV and average EBCT
Estimated DOC	15…37.5 mg/L	
TSS	8 mg/L	
min. EBCT	30 min.	
required v_{max}	8.0 m/h	
number of cells for $v_F < v_{max}$	≥ 6	
Min. filter velocity at average design flow	2 m/h	11 cells in operation (1 cell in backwash)
Range of achievable bed volumina treated	5,000…15,000 BV	Characteristic range of full-scale operation data (review of (Benström *et al.* 2016)
Service life of one GAC filling at average design flow	112…337 d 3.7…11.2 months	

5 Comparative assessment

In total 6 different options to integrate an advanced treatment step for removal of micropollutants at WWTP Rostock were detected and approximately designed. For all, a sufficient elimination rate for pharmaceuticals can be presumed from existing operational experience. All of them use more or less existing technology for an according upgrade. However, the additional investments and operational expenses differ significantly. Some provoke conflicts the conventional biological treatment processes. Namely the PAC processes will also have impact on the sludge treatment. At the current state, a clear favourite option cannot be identified. However, the comparing assessment (see Table 5) indicates two interesting candidates:

1. Conversion of the BIOFOR-N into GAC
2. Ozone + BIOFOR-N + conversion of BIOFOR-DN into GAC

The first option seems to be the most cost-efficient solution with low impact on the other treatment steps. The second option will provide the best elimination of micropollutants but causes additional efforts for the conventional nutrient removal.

Table 5: Comparative assessment of different options to integrate advanced removal of pharmaceuticals at WWTP Rostock

Assessment criteria	**Ozone**	**Ozone + GAC**	**PAC dosage in AST**	**PAC dosage in front of BIOFOR**	**Conversion of BIOFOR-N into GAC**
Efficient use of existing infrastructure for integration of advanced treatment	Good BIOFOR-N for post treatment	Very good BIOFOR-N for biological post treatment and SM reduction BIOFOR-DN as GAC	Moderate BIOFOR-N for rest removal of PAC	Good BIOFOR-N for PAC removal and a contact volume	Very good Multipurpose use for adsorption and polishing filter (SM, ammonia)
Possible conflicts for integration into existing processes	Conflict with nitrogen removal at BIOFOR-DN		Reduces SRT	Significant increase of backwash intervals, reduces SRT	Existing raw water bypass would impair adsorption
Additional Infrastructural requirements	Medium		Medium	Medium	Medium
Additional operational expense Staff Consumables Energy (on WWTP)	Low high (oxygen) high		High very high (PAC) low	Medium High High (backwashing)	Low High (GAC) Low
Expected elimination efficiency	Good	Very good	Medium	Good	Good
Combined assessment	**Promising** nutrient removal needs be resolved, BIOFOR-DN cannot be used efficiently	**Very promising** nutrient removal needs be resolved, best elimination potential due to additional GAC	**Not efficient**	**Needs further assessment**, mainly with regard to PAC separation	**Very promising**, lowest additional investments

6 Further Steps for Implementation of Advanced Treatment

According to the selected potential designs of advanced treatment at WWTP Rostock, the techniques need to be tested at small scales in the laboratory and ideally also in pilot-scale for verification and clarification of remaining questions, e.g. operation, control and selection of material (GAC). On the one hand, available GAC was tested for wastewater of WWTP Rostock. On the other hand, pre-tests at laboratory scale were optimized in terms of costs and easy methods to apply.

6.1 Application of GAC used in drinking water treatment at WWTP

Since drinking water provision and wastewater treatment are managed by the same operator in Rostock (NORDWASSER), there are interesting opportunities regarding similar treatment techniques such as ozonation and GAC-filters. Two studies were compiled to investigate whether GAC applied in drinking water treatment can be used secondarily at the WWTP, too. Therefore, GAC was obtained from the local drinking water treatment facility and adsorption characteristics have been investigated in laboratory. In the first study it turned out that a direct application of used GAC (left granulated) is not suitable for further adsorption which was tested with the most relevant three pharmaceutical substances (Carbamazepine, Diclofenac, Metoprolol). In the second study, the used GAC has been pulverized to PAC and showed much better adsorption characteristics which corresponds to previous studies, particularly Rohn & Nahrstedt [30]. However, the assessment for introducing PAC at the WWTP Rostock revealed needs for further investigation regarding PAC separation or identified as not efficient.

6.2 Development of specific parameters in the laboratory

Performing the studies to investigate GAC characteristics in terms of adsorption, usually pharmaceutical concentrations need to be determined for numerous samples. In a laboratory with basic equipment, instruments like HPLC, GC and MS for analysis of trace substances are not available so that samples have to be sent out to certified laboratories. The corresponding costs for each sample (>50€) exceeded the project's budget so that more practicable parameters have been tested. One of the best known cumulative parameters especially for organic substances is the so-called SAC254. It describes the specific adsorption at ultraviolet wavelengths, here at 254 nm, to identify dissolved organic compounds. The application of UV-probes to control removal rates at WWTPs was broadly tested [31] and showed that UV adsorption correlates with removal rates. Making use of this correlation, the UV adsorption of investigated pharmaceuticals Carbamazepine, Diclofenac and Metoprolol have been calibrated to known concentrations. Instead of the SAC254 a specific wavelength related to the pharmaceutical was chosen to improve the visibility in corresponding adsorption spectra. As a result, the pharmaceutical

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[30] Rohn A. and Nahrstedt A. (2017). Verwendung gebrauchter Aktivkohlen aus der Trinkwasseraufbereitung zur Spurenstoffentfernung bei der Abwasserreinigung. *Korrespondenz Abwasser* **64** (10), 212-16.

[31] Abbeglen C. et al. (2018). *Erfahrungen mit UV/VIS-Sonden zur Überwachung der Spurenstoffelimination auf Kläranlagen*, Hrsg.: VSA Plattform „Verfahrenstechnik Mikroverunreinigungen“, Schweiz

concentrations of the samples could be identified only by spectroscopy and do not require expensive analysis.

6.3 Application of parameters in practice

After successful application of the specific adsorption spectra to measure the pharmaceutical concentration at laboratory scale, the method was tested under real wastewater conditions, too. Therefore, additional 24h-composite-samples have been collected at WWTP Rostock in December 2019 with a mobile sampler, in detail in the inflow of BIOFOR as well as in the outflow of BIOFOR which corresponds to outflow of WWTP. These sampling points were chosen in accordance to previous sampling campaigns in Summer 2017 and Winter 2018 where final removal efficiencies of the BIOFOR-treatment step were detected (e.g. 45% for Metoprolol and 54% for Diclofenac). With regard to the method, the signal noise of sampled wastewater was unfortunately too high to identify pharmaceutical pollution. There were no clear signals visible in the adsorption spectra, neither at 254 nm nor at pharmaceutical specific wavelengths. According to the signal noise of the wastewater, the limit of detection in this method was 5 mg/L, 7 mg/L, 7 mg/L for Carbamazepine, Diclofenac and Metoprolol, respectively. With this knowledge, we would not suggest to apply the detection method via UV adsorption in real conditions only, but supplement it with trace substance analysis in HPLC-MS/MS or similar. Nevertheless, UV adsorption is a proper detection method for (spiked) laboratory scale experiments where the limit of detection of these pharmaceutical can be lowered significantly and peaks of spectroscopy become evaluable.

7 References

Abbeglen C. and Siegrist H. (2012). Mikroverunreinigungen aus kommunalem Abwasser; Verfahren zur weitergehenden Elimination auf Kläranlagen, Hrsg.: Bundesamt für Umwelt (BAFU), Schweiz, Umwelt-Wissen Nr.1214.

Abbeglen C. et al. (2018). Erfahrungen mit UV/VIS-Sonden zur Überwachung der Spurenstoffelimination auf Kläranlagen, Hrsg.: VSA Plattform „Verfahrenstechnik Mikroverunreinigungen“, Schweiz.

Barjenbruch M. and Firk W. (2014). Möglichkeiten der Elimination von Spurenstoffen auf kommunalen Kläranlagen. Korrespondenz Abwasser 61(10), 861-75.

Benström F., Nahrstedt A., Böhler M., Knopp G., Montag D., Siegrist H. and Pinnekamp J. (2016). Leistungsfähigkeit granulierter Aktivkohle zur Entfernung organischer Spurenstoffe aus Abläufen kommunaler Kläranlagen. Korrespondenz Abwasser, Abfall 63(4), 276-88.

KOM-M_NRW (2016). Anleitung zur Planung und Dimensionierung von Anlagen zur Mikroschadstoffelimination, 2. Auflage, Kompetenzzentrum Mikroschadstoffe NRW.

Metcalf.&.Eddy (2014). Wastewater Engineering - Treatment and Resource Recovery. Mac Graw Hill Education. ISBN 978-0-07-340118-8

Metzger S., Tjoeng I., Rößler A., Schwentner G. and Rölle R. (2014). Kosten der Pulveraktivkohleanwendung zur Spurenstoffelimination am Beispiel ausgeführter und in Bau befindlicher Anlagen. Korrespondenz Abwasser, Abfall 61(11), 1029-37.

Nahrstedt A., Burbaum H., Mauer C., Alt K., Sürder T. and Fritzsche J. (2014). Einsatz granulierter Aktivkohle auf dem Verbandsklärwerk "Obere Lutter". KA-Korrespondenz Abwasser 61(5), 408-26.

Rohn A. and Nahrstedt A. (2017). Verwendung gebrauchter Aktivkohlen aus der Trinkwasseraufbereitung zur Spurenstoffentfernung bei der Abwasserreinigung. Korrespondenz Abwasser 64 (10), 212-16.

Reungoat J., Macovca M., Escher B. I., Carswell S., Mueller J. F. and Keller J. (2010). Removal of micropollutants and reduction of biological activity in full scale reclamation plant using ozonation and activated carbon filtration. Water Research 44(2), 625-37.

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