COASTAL & MARINE

Estimating and reducing the discharge of pharmaceutical substances into the South Baltic

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Editorial

Dear Reader,
Pharmaceutical residues in the Baltic Sea are a cause for concern particularly as the full impact on the marine environment is still unknown but a wide range of therapeutic classes can be detected in both water and sediment samples from most Baltic Sea areas. To reduce emissions, (non) regulatory measures are needed on both national and regional level. The Baltic Sea Pharma platform is an EUSBSR flagship process under PA Hazards running over the period 2015-2025 which brings together projects and stakeholders from the whole region to boost knowledge-sharing, increase effectiveness of measures, streamline activities and support regional policy development.
One of the projects that is part of the BSR pharma platform and that has made an important contribution to close the knowledge gap on the consumption of pharmaceutical substances and the loads of pharmaceuticals that enter the South Baltic Sea via wastewater treatment plants, is the EU South Baltic Programme project MORPHEUS – Model Areas for Removal of Pharmaceuticals in the South Baltic. In this issue of the Coastal & Marine magazine, the MORPHEUS project presents exemplary results from consumption patterns to analytical results of a broad range of pharmaceuticals monitored in four regions of the South Baltic region. It also presents available advanced treatment technologies that can help to reduce the emission of pharmaceutical substances from WWTPs and hence improve the water quality of the South Baltic Sea.

I wish you an interesting and informative read,
Maxi Nachtigall

Maxi Nachtigall
Swedish EPA,
EUSBSR PA Hazards Coordinator

Content

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Colophon

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MORPHEUS Model Areas for Removal of Pharmaceutical Substances in the South Baltic
Interreg South Baltic
EUROPEAN UNION European Regional Development Fund

Introduction

A major problem in Europe is the immense amount of chemicals being released into our water bodies via wastewater treatment plants (WWTPs). Of special concern are the pharmaceuticals specified in two recent EU watch lists of substances for Union-wide monitoring (COMMISSION IMPLEMENTING DECISION (EU) 2015/495 and 2018/840). Concerning human medicines, the consumption phase is considered to be the biggest contributor to the discharge of medicinal products into the environment, notably through excretions and incorrect disposal of unused medicines through sinks and toilets that then reach the environment via wastewater treatment effluents. A challenge is how to remove pharmaceuticals from wastewater in a cost-efficient way since current WWTPs operate using technologies that were not designed for this task. Presently it is very hard for key target groups, namely WWTP operators and local/regional decision makers, to make decisions or even ask critical questions concerning both analytical chemical data and advanced treatment technologies entering the European market.

The MORPHEUS project investigated pharmaceutical consumption, existing wastewater treatment technologies and environmental occurrence of 15 selected pharmaceutical substances (e.g. heart medicines, painkillers and antibiotics) in four model areas in the South Baltic: Skåne (Sweden), Mecklenburg (Germany), Pomerania (Poland) and Klaipeda (Lithuania). Consumption data gave information about the chemical load to be expected at individual WWTPs, as well as the main similarities and differences in consumption in different regions. Chemical analysis of water samples was performed by solid-phase extraction (SPE) combined with liquid chromatography tandem mass spectrometry (LS-MS/MS) of the 15 pharmaceuticals. This included incoming and outgoing wastewater at 15 WWTPs in the four regions, thereby a comparison of the removal efficiency of different pharmaceuticals and also differences between WWTPs could be made. The incoming wastewater concentrations additionally allowed for the calculation of the incoming chemical load, which in turn could be compared to the chemical load estimated from consumption data to reveal a possible link between consumption and occurrence data. Additionally, water samples from rivers and lakes upstream and downstream the WWTPs gave information on the occurrence of pharmaceuticals in the environment as a result of released wastewater. Knowledge of the ecological sensitivity of the receiving water, in relation to the chemical concentrations and loads, allowed for an easier prioritization of where advanced treatment initially should be installed. An overview of advanced treatment technologies was provided by the project to help understand both possibilities and limitations of the technologies available on today's market.

WWTP outlet Kristianstad

Altogether, the knowledge obtained in the MORPHEUS project intends to aid wastewater treatment plants and authorities with the future implementation of the most suitable advanced treatment technology at the most appropriate sites with the overall goal that they can then make as wise and knowledge-based investments as possible. This will help improve the water quality of the South Baltic Sea.

For detailed information on the project results that are presented in this issue, please find the full reports on the project's website at www.morpheus-project.eu.

Project Coordinator MORPHEUS
Professor Erland Björklund
Kristianstad University, Sweden

Pharmaceutical consumption patterns in the South Baltic Region – Comparing Sweden, Germany, Poland and Lithuania

Modelling of regional pharmaceutical consumption patterns is a method to quantify the loads of pharmaceuticals that enter wastewater treatment plants (WWTPs) via household sewage systems. In MORPHEUS, the regional consumption data from four coastal regions in Sweden, Germany, Poland and Lithuania was collected wherein different formats of yearly statistics are available. The regional statistics are either based on sales data of wholesalers/ pharmacies or on data from health care institutions.

In Poland and Lithuania, wholesale data is represented in numbers of reimbursed packages according to so-called EAN codes (bar codes). Combined with specific product information such as pharmaceutical content (milligram per pill and number of pills), the total consumed mass load can be calculated. In contrast, German and Swedish data are published by health care institutions and administrative authorities based on prescriptions in the statistical unit DDD (daily defined doses). For such a format, the WHO Collaborating Centre for Drug Statistics Methodology (WHOCC), provides a list of conversion factors to determine the corresponding total consumed mass loads. Finally, the loads are divided by the considered number of inhabitants in order to generate comparative but region-specific values as yearly intake loads per inhabitant [mg/ inh.].

Comparison of the four model areas

The calculations provide an initial idea of the main differences and similarities in consumption. Germany and Sweden are more comparable with one another, as are Lithuania and Poland where the health care systems result in similar data formats and availabilities; prescriptions vs. refunding, respectively. Nevertheless, in all four countries, similar trends were found for e.g. the antidiabetic pharmaceutical Metformin with intake loads ranging from 9,000 mg/inh. (Lithuania) to almost 30,000 mg/inh. (Germany) in 2015. On the contrary, the antibiotic Clarithromycin is used much more in Germany and Poland than in Sweden and Lithuania. It is important to mention here that the project focused on selected pharmaceuticals. The potential for replacing pharmaceuticals, such as antibiotics, is not necessarily covered by this analysis. Nevertheless, the main regional differences can be summarized in four key findings.

Key finding 1: Country-specific consumption

Consumption of individual pharmaceuticals is country-specific. In all model areas, heart medicines are prescribed or refunded. Metoprolol is the highest consumed beta-blocking substance out of three investigated. The yearly intake in Sweden and Germany is distinctly higher than 1400 mg/inh., while Poland and Lithuania do not exceed 100 and 620 mg/inh., respectively (Fig. 1). This will affect the burden on the WWTPs and finally the environment.

Key finding 2: Seasonal variation of consumption

Consumption of some medicines varies with season, while others do not. The intake load of Metformin for treating diabetes is nearly steady, with a variation of about 5.5% between the monthly intake loads. The intake loads of the antibiotic Amoxicillin is much higher in the winter/spring season than in the summer season. The results seem to be reasonable since the risk of bacterial infections, colds etc. is much higher in winter and spring than in summer.

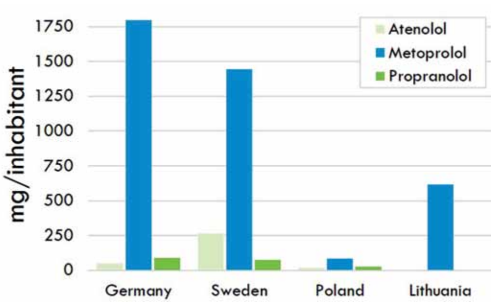


Figure 1. Total yearly intake loads of the heart medicines Atenolol, Metoprolol and Propranolol in mg/inh., data from 2015.

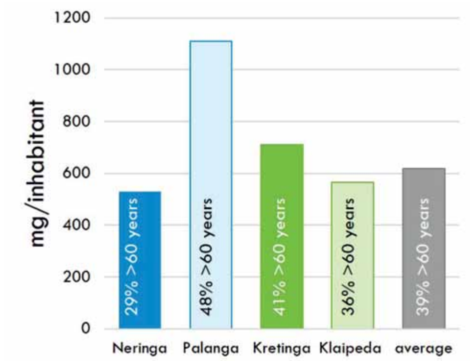


Figure 2. Total monthly intake loads of the heart medicine Metoprolol in mg/inh., data from Lithuania, 2015.

Key finding 3: Age-dependency of consumption
Consumption of some compounds differs between cities and rural areas probably due to differing age structures. For example, Metoprolol consumption per inhabitant is highest in Palanga, a seaside resort in Lithuania. Metoprolol is a beta-blocking substance (heart medicine) which is mostly consumed by elderly people. This correlates with the demographic structure of Palanga, which has a higher share of elderly inhabitants (>60 years) than other investigated municipalities. (Fig. 2)

Key-finding 4: Various distribution sites of pharmaceuticals
For Swedish and German data, a comparison of different distributing sites of pharmaceuticals was possible. This included prescriptions, hospitals and private purchases without prescription, namely "over the counter sales" (OTC). Overall, a relatively low contribution from hospitals was indicated for 15 out of more than 2,300 pharmaceuticals. The antibiotic Ciprofloxacin is frequently used in hospitals but is still usually distributed by pharmacies/prescriptions in both countries. Hence, a removal of pharmaceuticals only from hospital wastewater would not sufficiently reduce the load in wastewater. Due to outpatient intake, actions at municipal WWTPs are required to counteract increasing antibiotic resistance. In general, painkillers such as Ibuprofen are largely consumed via OTC. This means that a reduction of loads and resulting burden on the environment requires a fundamental rethink of individual applications of pharmaceuticals. Colds, headaches and feelings of discomfort might in some cases be treated with proven home remedies causing no side effects to the environment (Fig. 3).

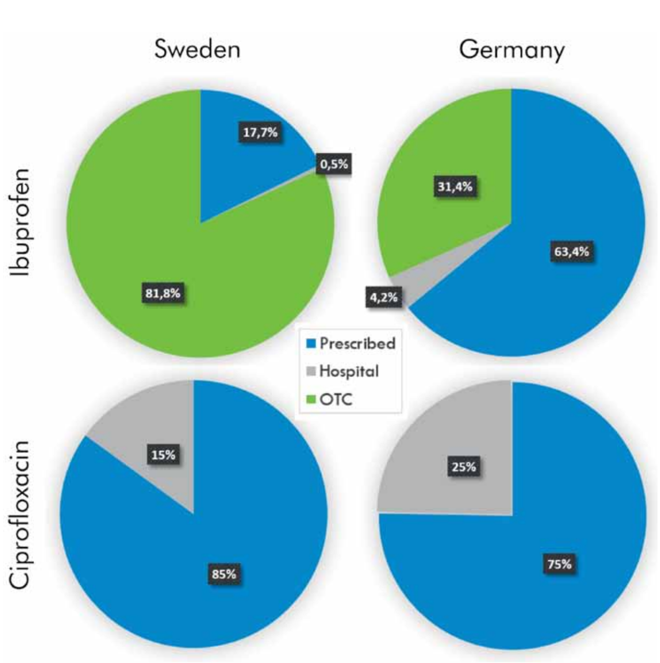
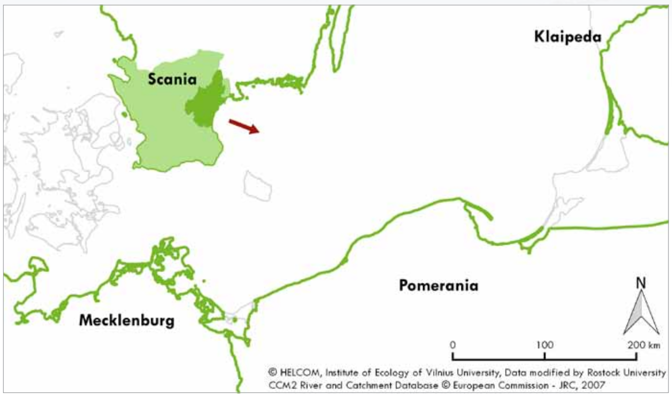


Figure 3 Comparison of different distributing sites of pharmaceuticals (prescriptions, hospitals and over the counter sales (OTC)) for Ibuprofen (top) and Ciprofloxacin (bottom) in Sweden (left) and Germany).

Modelled consumption patterns can predict the concentrations of pharmaceutical substances entering WWTPs
The results from the MORPHEUS project have shown that the estimated loads derived from modelled consumption patterns are comparable to the loads derived from measured concentrations for many of the monitored pharmaceutical substances.
In summary, it can be stated that consumption of pharmaceuticals differs between countries, regions and even cities. A comparative analysis conducted in the MORPHEUS project confirmed that distribution patterns depend on doctors' prescriptions, the season, and age of the population. A proportion of each consumed medicine end up in WWTPs and, since many of them are not removed sufficiently with current treatment technologies, they eventually find their way into rivers, lakes and the South Baltic Sea. Investigating the local consumption patterns is recommended to understand which pharmaceuticals are most relevant in which region. Combining this knowledge with chemical analysis of pharmaceuticals in WWTPs and receiving water bodies, will aid prioritization processes and ensure wiser investments in advanced treatment technologies to remove relevant pharmaceuticals from the local wastewater and the aquatic environment. The first step we can all make however, is to use and dispose of pharmaceuticals wisely for our own sake and the sake of the environment.

Alena Kaiser, Jens Tränckner & Erland Björklund
University of Rostock, Germany & Kristianstad University, Sweden

Estimating the local chemical pharmaceutical burden using chemical analysis of wastewater and surface water – The example of Diclofenac in Kristianstad Municipality, Region Skåne



In the MORPHEUS project we estimated the occurrence and load of pharmaceuticals in four areas of the South Baltic Sea. This included the released loads from selected WWTPs as well as the environmental occurrence of pharmaceutical substances downstream of the WWTPs in the coastal regions Skåne (Sweden), Mecklenburg (Germany), Pomerania (Poland) and Klaipėda (Lithuania).

In this article, we report on some of the findings from the Swedish model area Skåne and Kristianstad Municipality in the north east part of Skåne (Scania) as an example of a local assessment.

Figure 1. Sampling points of treated wastewater at the outlet of Kristianstad WWTP (1a, left) and Tollarp WWTP (1b, right).

In total 15 pharmaceuticals were evaluated in MORPHEUS, and here we present data for one of these which is much used in Sweden; Diclofenac.

In Kristianstad municipality there are several WWTPs. Here we focus on two, which both release their treated wastewater from the two cities Kristianstad and Tollarp into the Helge Å river system. The two WWTPs differ in size with the largest being Kristianstad WWTP (Figure 1a) treating a yearly wastewater volume of 8,186,000 m^3 from roughly 52,000 people and large food industries, while Tollarp WWTP (Figure 1b) treats 361,000 m^3 from 4,790 people and food industries.

Samples were taken in the outlet water of both WWTPs and the Diclofenac concentrations obtained were 579 ng/l and 646 ng/l for Kristianstad and Tollarp WWTP, respectively (average of a summer and a winter sample). By multiplying this concentration by the yearly volume of treated wastewater, the total released burden of Diclofenac was calculated to be 4.7 kg from Kristianstad WWTP and 0.23 kg from Tollarp WWTP (Figure 2).

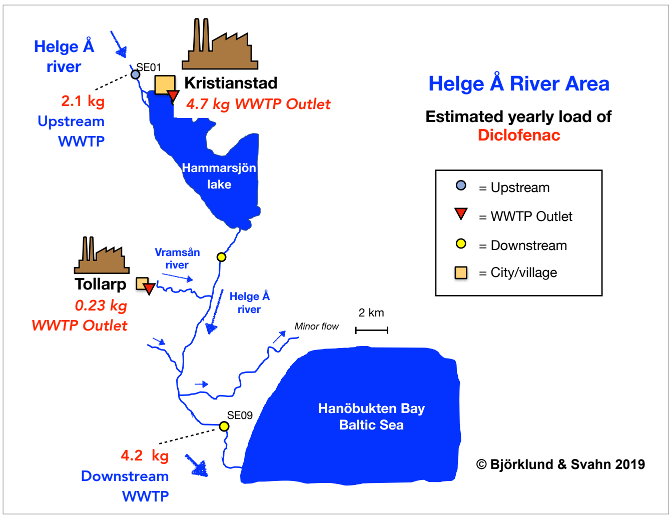


Figure 2. Schematic overview of the Helge Å river area within the UNESCO Biosphere Reserve "Vattenriket". In the picture, four sampling points are indicated; the upstream sampling in Helge Å river (SE01, Fig. 3), Kristianstad WWTP outlet (Figure 1a), Tollarp WWTP outlet (Fig. 1b) and the downstream sampling point in Helge Å river. (SE09, Header).

The lower part of Helge Å river, including the Hammarsjön Lake, is a unique wetland and was given the status of a UNESCO Biosphere Reserve in 2005 with the name "Vattenriket". The area holds a great variety of species of which many are red listed. The Helge Å river has a length of almost 200 km and an average yearly flow of around 37 m^3/s (2016). The Helge Å river enters Lake Hammarsjön in the northwest corner and flows through the lake to the south. Kristianstad WWTP releases its wastewater into the northeast corner of Lake Hammarsjön.

The Vramsån river is a tributary of the main Helge Å river and is one of Europe's finest places for a number of bivalves. The Vramsån river has a length of 55 km and an average flow of 3.4 m^3/s (2016), and enters the Helge Å river downstream from Tollarp WWTP which releases its wastewater into the Vramsån river. All together the Helge Å river and the Vramsån river represent very different types of dilution scenarios. Additionally, the UNESCO Biosphere reserve, including the two rivers and Lake Hammarsjön, is a great example of an area with high ecological value and sensitivity.

In order to study the presence of pharmaceuticals in the UNESCO Biosphere Reserve "Vattenriket", as a consequence of the release of pharmaceuticals from WWTPs in the area, two sampling points were chosen. One of these was a sampling point called SE01 upstream from Kristianstad WWTP (Figure 2 and Figure 3). Here the background concentration of Diclofenac was ca. 1 ng/l. By multiplying this value by the average yearly flow of the Helge Å river, it shows that roughly 2.1 kg of Diclofenac enters the UNESCO Biosphere Reserve "Vattenriket" yearly. This load comes from upstream WWTPs. There are more than 30 WWTPs that discharge along the Helge Å river, which are a major source of pharmaceuticals entering the UNESCO Biosphere Reserve "Vattenriket".

Figure 3. Sampling point SE01 in Helge Å river, upstream of Kristianstad wastewater treatment plant, see Figure 2.

If the purpose is to decrease the total amount of pharmaceuticals that enter the entire river system, Kristianstad WWTP should be equipped with advanced treatment technologies to remove 4.7 kg of Diclofenac. The next step would be to contact adjacent municipalities, upstream from the Helge Å river system, to ask them to take actions to reduce the extra 2 kg of Diclofenac that is entering at the upstream point of Lake Hammarsjön. Thirdly, Tollarp WWTP should implement advanced treatment technology to take out 0.23 kg of Diclofenac.

From a concentration perspective, Kristianstad WWTP should once again be equipped with advanced treatment technology, since the concentration of Diclofenac in the north-eastern bay of Lake Hammarsjön is almost 400 ng/l. This is 4 times higher than the concentration stated by the Swedish Agency for Marine and Water Management of 100 ng/l which must not be exceeded to classify the environmental status of the surface water as good. The reason for this high concentration is poor dilution since the Helge Å river does not pass directly through this more stagnant part of the lake. Secondly Tollarp WWTP should be upgraded since the concentration here is roughly 10 ng/l which is relatively high compared to the upstream point in Helge Å river which is in the order of 1 ng/l.

Finally, a point downstream of all WWTPs, called SE09 (Figure 2), was analysed and shown to contain ca 2 ng/l of Diclofenac. This showed that roughly 4.2 kg of Diclofenac will enter the Baltic Sea annually from the Helge Å river system.

All together the work conducted in Kristianstad municipality and the UNESCO Biosphere Reserve "Vattenriket" is an example of how the monitoring of released pharmaceuticals in a specific area can be used to make better-informed and knowledge-based decisions on where to take action to start reducing the chemical burden on the South Baltic Sea.

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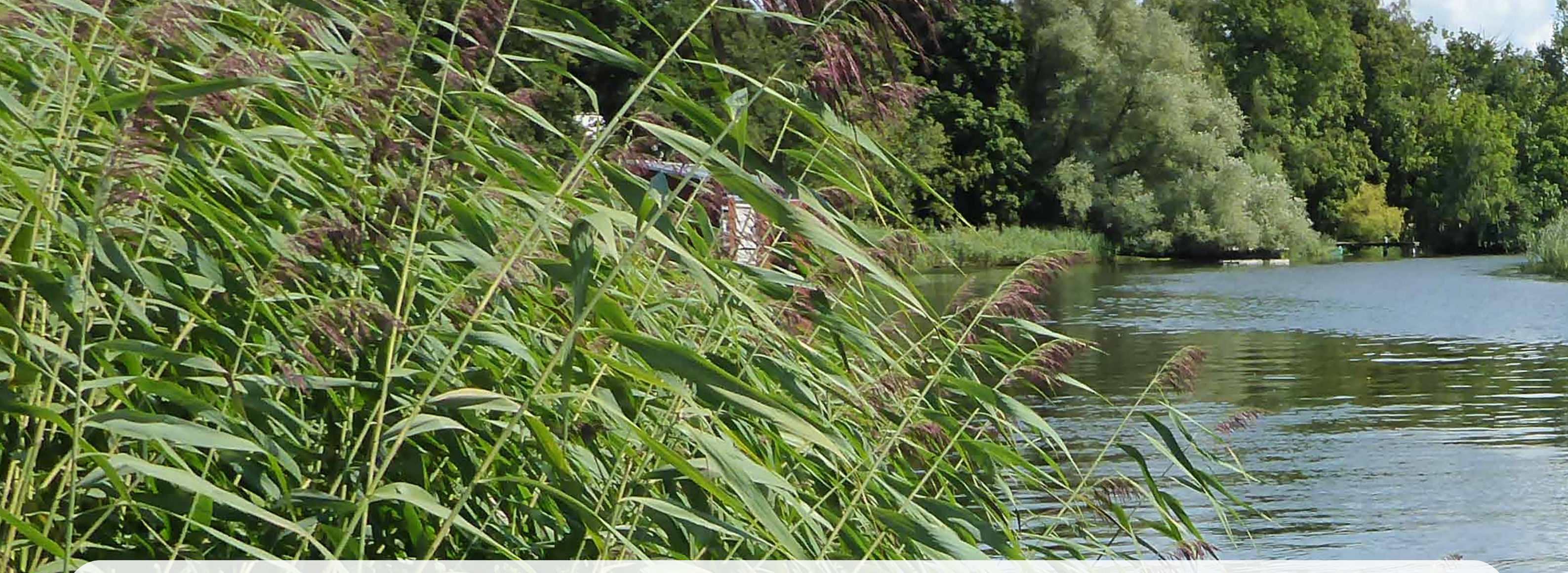
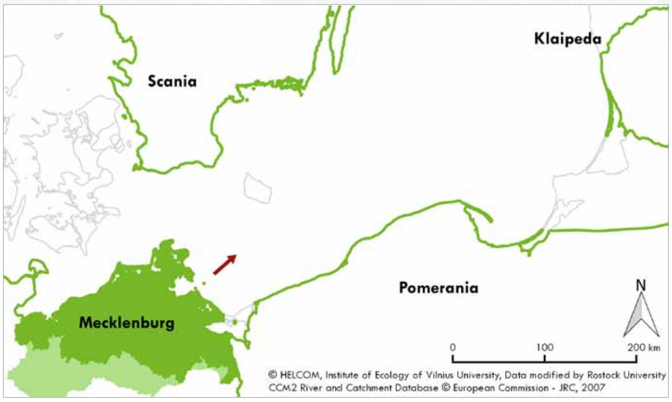


Monitoring and Modelling Pharmaceutical Substances in WWTPs and Rivers within the German Model Area Mecklenburg



Most of the German Baltic Sea catchment area is located within the federal state Mecklenburg-Western Pomerania (German: Mecklenburg-Vorpommern; MV), covering app. 23,000 km² (see map). Around 75% of the Federal State is discharging into the Baltic Sea, and the remaining south-western part discharges via the river Elbe into the North Sea.

In the model area, large wetlands, rivers and streams are fed mainly by groundwater inflow. The river systems are characterized by a small-spatially structured network with numerous small water bodies and ditches. MV is the least densely populated and least industrially developed state in Germany with 69 persons/km² compared to the German average of about 232 persons/km². With its low population density and good quality of primary clayey soil, the landscape is dominated by rural areas with more than 60% land use in agriculture and app. 22% covered by forest. MV is also rich in water with more than 2,000 lakes and about 2,000 km of Baltic Sea coastline which both provide valuable natural habitats and numerous opportunities for the strong tourism sector in this region.

In total, 586 WWTPs are operated in MV. They are categorized by size class according to their designed wastewater capacities using the unit population equivalents (PE). It can be seen that despite the high number of smaller WWTPs, the largest share of volume is connected to size class 4 and 5 (Table 1.). Regarding the German Baltic Sea catchment, around 1.2 million inhabitants are connected to more than 400 WWTPs which are mostly small, except those in larger coastal cities such as Rostock and Wismar. All WWTPs operate both mechanical and biological treatment techniques, even those of classes 1 and 2. WWTPs larger than class 2 do also include special treatment technologies for removal of the nutrients nitrogen and phosphorus. Advanced treatment for micro-pollutants is not yet implemented in any WWTPs in this region but is gaining increasing amounts of attention from public authorities.

On a national level, the so called "Federal Stakeholder Dialogue on Trace Substances" was launched in 2016. With a joint approach, the stakeholders shall develop a mutual strategy to reduce pharmaceuticals and other micro-pollutants in the water system. This process is still ongoing. In parallel to these developments, MV has been performing a ten year monitoring campaign (2008-2017) to assess the environmental pharmaceutical concentrations in surface water bodies. In total, 51 substances have been analyzed at 187 monitoring points. The recently published results indicate clearly that pharmaceuticals are omnipresent in inland and coastal waters. One of the highest detections, found in 75% of all samples, was reported for Carbamazepine, a persistent substance applied as an antiepileptic medicine. Additionally, Carbamazepine exceeded the proposed environmental quality standards of the German federal EPA (UBA) at several locations.

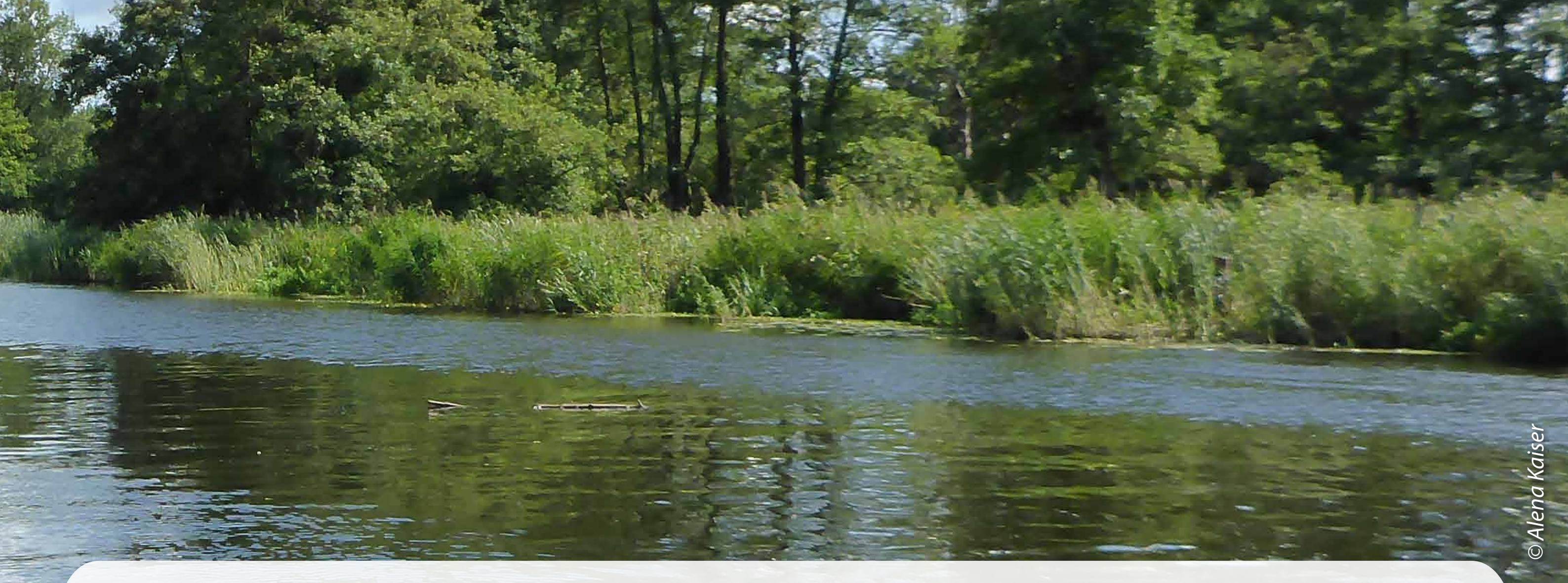
Within the framework of MORPHEUS, a comprehensive sampling campaign for estimating the pharmaceutical burden in the Model Area was conducted too. In order to cover the variability of WWTP types in Mecklenburg, four WWTPs of different size classes were chosen for analyzing the consumption and occurrence of selected pharmaceuticals, namely WWTP Rostock, WWTP Laage, WWTP Krakow, and WWTP Satow (Table 2).

Size class (design size in PE)	Number of WWTP	Total capacity (PE)	Share (%) of treated wastewater
5 (>100,000)	4	940 000	28.6
4 (10 001 -100 000)	47	1 820 250	55.4
3 (5 001-10 000)	21	166 892	5.1
2 (1 000-5 000)	86	214 870	6.5
1 (<1 000)	428	144 939	4.4

Table 1. Treated wastewater according to WWTP size classes in the Model Area of the Federal State Mecklenburg-Western-Pomerania, Germany (2014)

Size class (PE)	Name WWTP	Total capacity (PE)	Actual (PE)
5(>100000)	Rostock	400000	342483
4(10001-100000)	Laage	20000	12658
3(5001-10000)	Krakow	7500	6209
2(1000-5000)	Satow	2500	2300

Table 2. Selected German WWTPs for sampling in MORPHEUS and their sizes expressed as personal equivalents (PE)



For indicating the direct influence of wastewater effluents in the receiving water bodies, mainly small rivers and ditches, both upstream and downstream of the treated wastewater discharge point were sampled with a minimum distance of 200 m to ensure total mixing within the water bodies.

The analysis results for the selected water bodies have shown similar results to the ten-year monitoring campaign in MV. Besides Diclofenac and Metoprolol, Carbamazepine revealed the highest concentrations of up to 2.17 µg/L downstream of WWTP's discharge points. The detection rate of Carbamazepine was 100% both in summer and winter samplings. Of special interest was the concentration found in the river Warnow upstream of the WWTP Rostock. The high level of Carbamazepine proves that small WWTPs, discharging into the river catchment upstream, contribute significantly to the concentration measured. Therefore, an accumulation model within the complete catchment area has been developed to identify hotspots of pharmaceutical burden on the environment. Herein, the highest concentration of Carbamazepine measured during our sampling campaign matched one of the predicted hot spots: the receiving water body of WWTP Krakow downstream. Even though a need for action is thereby revealed, this does not make a statement on the WWTP's removal efficiencies but on the ability of the rivers and streams to dilute the wastewater effluents.

Figure. Sampling in Krakow, WWTP outflow and downstream

The removal efficiency was investigated by analyzing both the WWTP's inflow and outflow concentrations and calculating the corresponding load balances. The analysis of 24-hours-mixing samples of the four WWTPs showed that the removal rates of Carbamazepine is fluctuating at a comparatively low level from -19 to 38%. Hence, mechanical and biological treatment techniques are not yet sufficient to remove Carbamazepine from wastewater.

However, it is not feasible to estimate the total burden of pharmaceuticals for all receiving waters and discharging WWTPs by expensive sampling and analyzing procedures. To gain a holistic view, a regional mass-flow model was developed in MORPHEUS using regional pharmaceutical consumption statistics of health insurances and sale companies. The model was successfully validated by the sampling results of the wastewater inflow and now gives a good estimate for the whole area.

Finally, the MORPHEUS project supported and brought attention to the processes that handle pharmaceutical burden on the model area Mecklenburg. The project drew the attention of WWTP operators to this topic and was able to present a first assessment of the actual pharmaceutical discharges by domestic WWTPs into the surface water system of MV. The conducted monitoring campaigns, in line with the findings of the regional authorities, highlight an existing pharmaceutical burden on the environment. The general outcome emphasizes that rural areas also contribute to pharmaceutical discharge into the Baltic Sea. However, regarding the fragmented wastewater treatment in rural areas, a technological reduction will become challenging and requires a sound cost-benefit analysis. Each of us should therefore take responsibility for the conscious consumption and disposal of pharmaceuticals.

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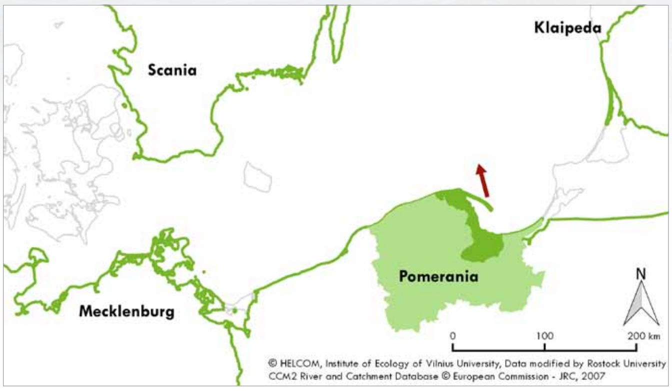
Exemplary results from the Polish model area in the Pomeranian Voivodeship



The Pomeranian Voivodeship is situated in northern Poland, bordering the shore of the Baltic Sea. It is one of the most developed regions in Poland, combining recreational, agricultural and industrial areas with historical traditions (including solidarity). In the Pomeranian Voivodeship 83% of the population are connected to 160 wastewater treatment plants (WWTPs), which serve about 1,920,400 inhabitants and treat a total load of 3,146,300 PE (population equivalent). A rapid development of sewage networks and WWTPs started after the Polish accession to the European Union in 2003. The requirements of the Accession Treaty in the wastewater sector successfully forced the implementation of environmental directives, e.g. limitation of nutrient discharge into the Baltic Sea. However, micropollutants (including pharmaceutical substances) released from the Polish catchment area are still an essential challenge since there is neither a legal basis nor other documents related to their analysis or removal from wastewater.

Within the MORPHEUS project, four model WWTPs were chosen in the Pomeranian Voivodeship for analyses of the presence and elimination of pharmaceutical substances: Gdansk-Wschod WWTP, Gdynia-Debogorze WWTP, Jastrzebia-Gora and Swarzewo WWTP. For wastewater treatment, each of the model WWTPs uses the activated sludge method with advanced nutrient removal. At large WWTPs, such as Gdansk-Wschod and Gdynia-Debogorze (> 100,000 PE) serving urban areas, no technological problems have yet been observed. However, a gradual increase of the inert COD (Chemical Oxygen Demand, organic matter of limited biodegradability) in the incoming wastewater is observed. This might be connected with an increasing amount of chemicals used by households, as well as with the discharge of hospital and industrial wastewater to the municipal sewer. The two other WWTPs, in the seaside locations of Jastrzebia Gora and in Swarzewo face an increasing amount of wastewater from May to August during the tourist season. They also have to accept wastewater collected from surrounding septic tanks, which is usually highly condensed and thereby influences the treatment efficiency.

Concentrations of selected pharmaceutical substances in the inflow and outflow of the model WWTPs

Within MORPHEUS the presence of pharmaceutical substances was monitored in the inlet and outlet of the selected WWTPs as well as in the receiving waters (examples of the results are given in Fig. 1). Among the tested pharmaceuticals in the influent, the highest concentration was measured for Ibuprofen from 200 µg/l up to 1.6 mg/l. For some pharmaceuticals, belonging to antimicrobials (Azithromycin, Clarithromycin, Ciprofloxacin) and anti-inflammatory drugs (Ibuprofen, Diclofenac), enhanced seasonal consumption might cause their elevated concentrations (above 1 µg/l) in the winter compared to summer values. In the WWTP effluents, the highest concentrations were found for Azithromycin (up to 4 µg/l), Clarithromycin (up to 2.9 µg/l), Metoprolol (up to 1.3 µg/l), Diclofenac (up to 3.7 µg/l) and Carbamazepine (up to 2.3 µg/l). Note that in the studied WWTPs, the removal efficiencies of pharmaceutical substances varied widely, from - 266% up to 100%. It can be linked, for instance, to the difference in the sludge age between summer and winter time.

With the highest efficiency, of over 90%, Ciprofloxacin, Estrone, Estradiol, Naproxen, Paracetamol, and Ibuprofen were removed. Azithromycin, Diclofenac and the beta-blockers Metoprolol and Propranolol did not exceed an average removal rate of 30%.

Surprisingly, Oxazepam, Erythromycin and, only in the winter season, Carbamazepine were found in higher concentrations in the outflow then in the inflow. It is suspected that rejected water, produced from sludge treatment processes, may contribute to the increase of pharmaceutical loads.

Presence and fate of pharmaceutical substances in the receiving water bodies

The data obtained in the MORPHEUS project confirmed the previous assumptions that the existing treatment technology, based on activated sludge, is not efficient in the removal of a number of pharmaceutical substances.

According to the measured concentrations of pharmaceuticals in marine water samples, this reflects their fate and presence in the WWTPs effluents, and despite the high dilution rate in the receiving water bodies they are still present at detectable concentrations. Thus Diclofenac and Carbamazepine were found with the highest concentrations (up to 22 ng/l and up to 37 ng/l, respectively), followed by Metoprolol, Azithromycin, Clarithromycin and Sulfamethoxazole (< 10 ng/l). A similar pattern was found in both sampled rivers: Vistula and Czarna Wda. In Czarna Wda, however, the concentrations were much higher (e.g. Diclofenac up to 126 ng/l and Carbamazepine up to 175 ng/l), due to the large proportion of WWTP effluent in the river water.

Interestingly, in both marine and river water samples, higher concentrations of pharmaceuticals were in general found in the summer season compared to the winter season. This phenomenon should be further studied in detail. Likely explanations might be increased population due to heavy tourism and less water in the rivers during summer.

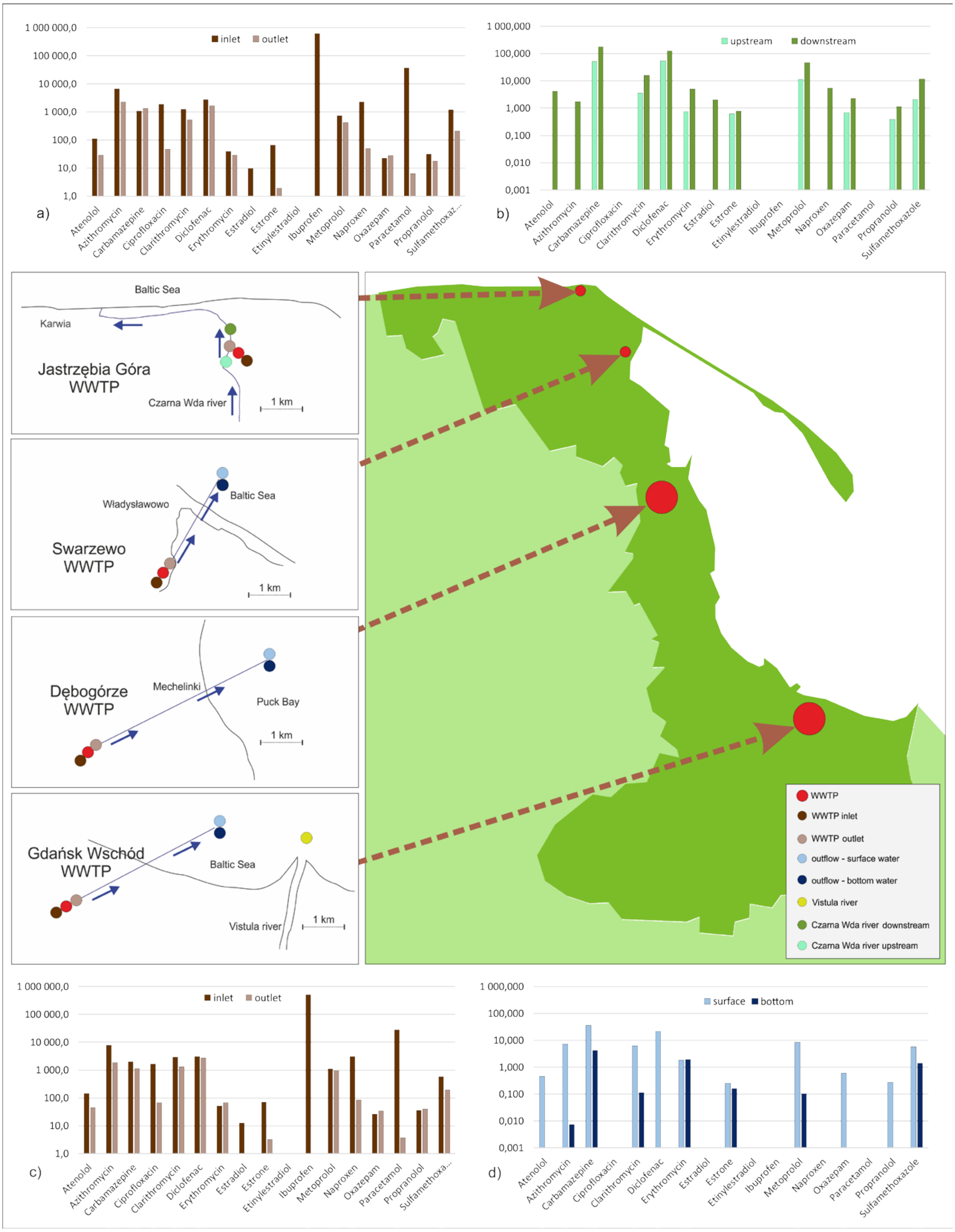


Figure 1. Model WWTPs in the MORPHEUS project: Gdansk-Wschod (Q=92 958 m³/d, PE=742 521); Gdynia-Debogorze WWTP (Q=55 294 m³/d, PE=476 000); Swarzewo WWTP (Q=6 164 m³/d, PE=149 000); Jastrzebia-Gora WWTP (Q=1 678 m³/d, PE=12 540). Examples of pharmaceutical substances concentration [ng/l in logarithmic scale] are presented for the influent and effluent of Jastrzebia-Gora WWTP (a) and Gdansk-Wschod WWTP (c) as well as in their receivers: Czarna Wda river (b; upstream and downstream the treated wastewater discharge) and Gdansk Bay (d; 2.3 km from the coastline, bottom and surface water samples nearby the diffuser systems of submarine collector), respectively.

It can be concluded that the WWTPs in the Polish model area, based on activated sludge systems, are effective in macropollutant removal and in the removal of some pharmaceutical substances, as pain killers (Naproxen, Paracetamol, and Ibuprofen). However, relevant antimicrobial agents and other important pharmaceuticals, used for high blood pressure, inflammation and pain, depression and anxiety are still present in WWTPs' effluents and their receivers. Thus, in order to protect the aquatic environment, drinking water supplies as well as to reduce antimicrobial resistance dissemination, control strategies for pharmaceutical substances are required.

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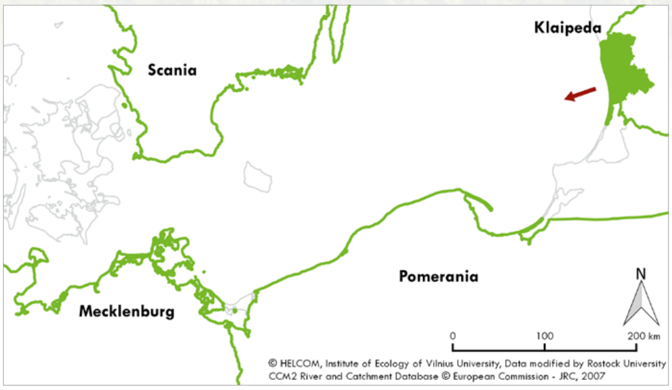
Main findings from the Lithuanian model area



There are four urban agglomerations situated in the Lithuanian coastal area which have a direct impact on the Curonian Lagoon and the Baltic Sea: the municipalities of Klaipėda city which is the third largest city in Lithuania, Palanga, Kretinga and Nida. Therefore, the WWTPs of the mentioned urban areas and their receiving water bodies were selected as most relevant for the implementation of the MORPHEUS project tasks. Pollution load of pharmaceutical substances in the Lithuanian coastal region was one of the main assessment criteria when selecting WWTP for upgrading with advanced treatment technology.

Wastewater treatment capacities and load estimation

The WWTP in Klaipėda is the largest among the four selected WWTPs by design capacity and annual volume of treated wastewater. The average daily volume of treated wastewater in 2018 varied from 848 m^3 in Nida, 3,478 m^3 in Kretinga, 7,993 m^3 in Palanga and 36,478 m^3 in Klaipėda.

An estimation of the pharmaceutical load from the WWTP's effluents shows that Klaipėda discharges the highest amount of pharmaceutical substances to coastal surface water bodies.

According to the average (in 2017 and 2018) pollution load calculations, Klaipėda contributes almost 77 kg or 81% of the total pharmaceutical substances load of 94.54 kg, while Palanga, Kretinga and Nida discharge 10.97 kg or 11.6 %, 6.32 kg or 6.7% and 0.66 or 0.7% respectively (Table 1).

Results of the chemical analysis in the receiving water bodies

Summer and winter water samples were taken in 2017 and 2018 in the following waterbodies/recipients of WWTP discharges: Baltic Sea (Lithuanian coast), Curonian Lagoon, Klaipėda Strait, river Tenžė (tributary of the Akmena-Danė), and in the Akmena-Danė river mouth. Figure 1 presents the average concentrations of pharmaceutical substances in those waterbodies.

Despite the dilution rate, several pharmaceuticals were detected in Baltic Sea water samples. Carbamazepine, Erythromycin, Estrone and Sulfamethoxazole were detected at low concentrations, above the method quantification limit (MQL). The same substances were also detected in slightly higher concentrations in the water samples from Klaipėda Strait. Additionally, Clarithromycin, Diclofenac, Paracetamol, Ibuprofen and Metoprolol were found in the Klaipėda Strait water. In the water of the Curonian Lagoon near Nida, only five pharmaceuticals at low concentrations were found: Carbamazepine, Clarithromycin, Diclofenac, Estrone and Paracetamol. All 15 pharmaceuticals were detected in river Tenžė. The highest concentrations were observed near the outlet of the Kretinga WWTP.

Figure 2 shows the comparison of average concentrations (near the outlet and downstream) in the Klaipėda Strait in summer and winter samples. Concentrations of Carbamazepine, Erythromycin and Sulfamethoxazole were higher in summer, concentrations of Clarithromycin, Diclofenac, Ibuprofen, Metoprolol and Paracetamol were detected in notably higher concentrations in winter, probably due to the flu season.

	Outlet load of pharmaceuticals				**Total, kg**
	Klaipėda	*Palanga*	*Kretinga*	*Nida*	
Load in 2017 (summer sampling), kg	63.14	11.23	8.22	0.62	83.21
Load in 2018 (winter sampling), kg	90.05	10.71	4.42	0.69	105.87
Average annual load (average of load in 2017 and 2018), kg	76.59	10.97	6.32	0.66	94.54
Percentage of total average load in 4 WWTPs in %	81.0	11.6	6.7	0.7	
	Recipient waterbody				
	Klaipėda Strait	Baltic Sea	River Tenžė (drainage ditch)	Curonian Lagoon	

Table 1. Outlet chemical load (kg/year) of 15 pharmaceuticals in 4 WWTPs. An estimate of the load of pharmaceuticals released to the recipient from each individual WWTP expressed as kg/year was calculated based on the effluent concentrations and the total volume of treated wastewater in 2017 and 2018, respectively.

Preconditions for the application of advanced treatment technologies

In Lithuania, discussions on a political and multi-stakeholder level have not yet started and knowledge gaps limit the opportunity to initiate the introduction of pilot or full-scale advanced treatment technologies. Despite that, pilot investments, partly supported by the EU Interreg South Baltic Programme, are being implemented for the application of technological solutions for removing pharmaceutical substances and other micropollutants in Kretinga WWTP. Recently, bilateral cooperation initiatives have been taken by Lithuanian and Latvian governmental and academic institutions to fill knowledge gaps by extended monitoring of WWTPs and surface waters to develop water protection policy/strategy-oriented recommendations and initiating prefeasibility studies for the application of advanced treatment technologies in selected WWTPs.

Preliminary findings

An upgrade of the Nida settlement WWTP would not significantly reduce the impact on the quality of the Curonian Lagoon aquatic environment due to the small amount of wastewater and low pollution loads. In addition, the introduction of advanced technology would further increase the existing high water service prices.

Although the Tenžė river contains the highest concentrations of pharmaceuticals among the four wastewater receivers, priority for the implementation of advanced treatment in Kretinga WWTP could not be given for several reasons. Firstly, there is the ongoing implementation of GAC (granulated activated carbon) treatment as a pilot test. Secondly, testing will also increase the already high water service costs. Providing financial assistance to the water company should be considered for further testing and operating pilot advanced treatment equipment.

The Klaipėda WWTP should be given priority based on the following criteria:

- Klaipėda WWTP discharges the largest volumes of treated wastewater and releases the highest amounts of pharmaceuticals to surface water bodies
- The introduction of pilot-scale and/or on-site tests or even full-scale advanced treatment will have least effect on water services prices compared to other coastal WWTPs
- The water company has the greatest potential for recruiting qualified specialists for the maintenance and operation of advanced wastewater treatment systems.

Considering the long-term reduction targets, upgrading the Klaipėda WWTP with advanced treatment technologies, such as ozonation or activated carbon, would significantly reduce the direct pollution load of pharmaceuticals and other micropollutants to the Lithuanian coastal waters. To achieve these goals in the future, multi-stakeholder dialogue and agreement is needed. Further steps could currently be initiated, e.g. the filling of monitoring gaps and a search for viable technological solutions by pilot-scale and/or on-site tests as well as available funds/financing schemes.

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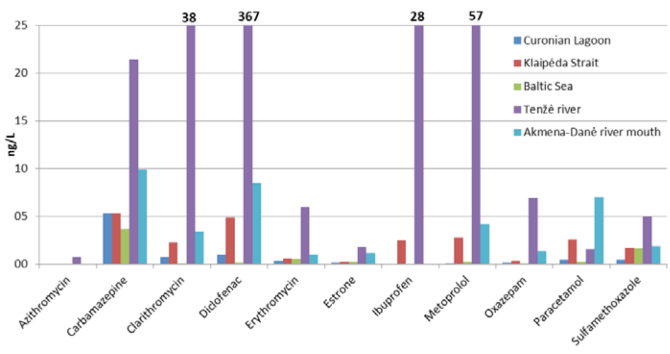

Figure 1. Average concentrations (summer, winter, downstream, background monitoring stations in the Curonian Lagoon, Klaipėda Strait and the Baltic Sea) of pharmaceuticals in Lithuanian waterbodies. Except for the Baltic Sea, values measured near the outlets of WWTPs are not included.

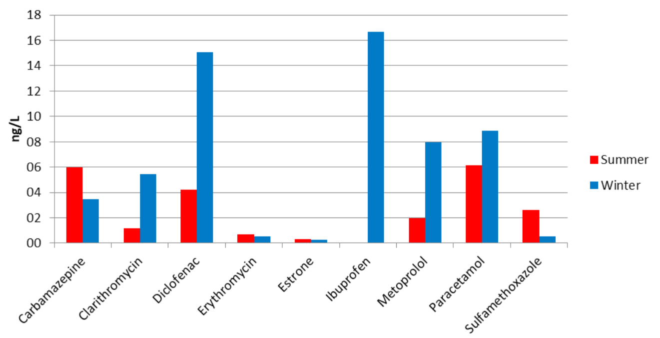

Figure 2. Average concentrations (near the outlet and downstream) of pharmaceuticals in Klaipėda Strait in summer 2017 and winter 2018.

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Advanced technologies for the removal of pharmaceutical substances in wastewater

Most EU countries are convinced that the presence of micropollutants (MPs) such as pharmaceuticals in the environment pose a serious problem for aquatic life, but also for humans, due to the presence of MPs in surface drinking water resources and to the development of antimicrobial resistance by the release of antibiotics. However, there is currently a lack of EU recommendation on WWTP effluent standards for many MPs including pharmaceuticals. In addition, the 'polluter pays' principle is very complex in terms of MPs since it is unclear who the polluter is, e.g. chemical producers, the pharmaceutical sector, hospitals or the consumer groups. Therefore, two approaches need to be developed simultaneously: (1) the limitation of critical MPs and usage (source and user measures) and (2) the mitigation of the dissemination of MPs by WWTPs (end-of-pipe measures). Since pharmaceuticals are rather essential in our healthcare systems and cannot be fully replaced with harmless alternatives, end-of-pipe technologies seem to be an essential part of the solution.
Currently there is a need, posed by the European Commission and other organizations, to monitor MPs and to develop methods and investigate the feasibility of upgrading selected urban WWTPs with advanced treatment to eliminate a broad range of MPs at reasonable costs.

Strategies for micropollutants removal from wastewater

Switzerland as an up-stream country, in 2016 (first in Europe) introduced a legal basis for the implementation of advanced treatment technologies in wastewater treatment. About 100 out of the total 700 WWPTs in Switzerland are going to be extended or upgraded by 2040 with the aim to eliminate 80% of indicator MPs (e.g. pharmaceuticals) and to treat 50% of the total wastewater volume generated there.
In Germany, there are as yet, no legal requirements for the application of technologies to remove MPs. The national micropollutant strategy is currently in the consulting phase. However, some federal states, in particular North Rhine Westphalia and Baden-Württemberg, have already equipped several WWTPs with advanced treatment on a voluntary basis.
The Swedish government has already funded several projects related to MPs (mainly pharmaceuticals) removal from wastewater. Currently, the knowledge and operating experiences of various technical solutions are complete and available as a foundation for a full-scale introduction of advanced treatment technologies.
In Poland and Lithuania there is neither a legal basis nor other documents related to monitoring and/or the removal of pharmaceuticals from wastewater. However, both countries are introducing national regulations imposing the need to assess priority substances.

Available technologies

Various technical solutions are available and have proved to be effective in MP removal with possible integration with existing treatment processes in an expedient manner. The solutions that have been evaluated are mainly based on activated carbon and/or advanced oxidation processes like ozonation (Figure 1 and 2), and various combinations thereof. Both possibilities and limitations of the techniques are widely discussed in Deliverable 5.2 of the MORPHEUS project (for details see *www.morpheus-project.eu/downloads*). Nonetheless, at a particular WWTP, the decision-making process should be supported by a wide range of monitoring programmes assessing the MP baseline in wastewater and receiving water body. Additionally, the MP strategy should have a broad societal and political acceptance. In the framework of the MORPHEUS project, roadmaps for the upgrade of selected WWTPs in Sweden, Germany, Poland and Lithuania have been developed and can be found at: *www.morpheus-project.eu*.

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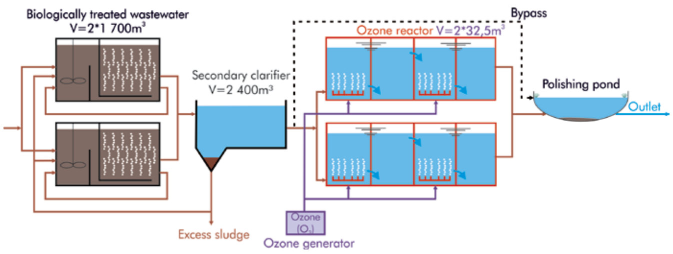

Figure 1. Scheme of the ozonation system at WWTP Bad Sassendorf in Lippeverband, Germany

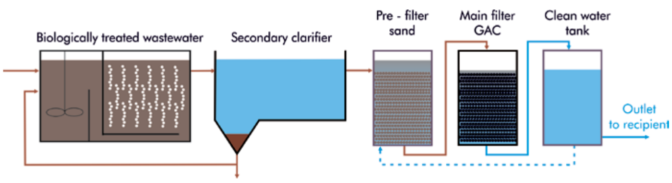

Figure 2. Scheme of the granulated activated carbon (GAC) filtering system at Kristianstad WWTP, Sweden

Clear waters from pharmaceuticals (CWPharma)

CWPharma will give tools and recommendations to policy makers, authorities and municipalities on the best ways to reduce emissions of active pharmaceutical ingredients (APIs) in the Baltic Sea Region. Screening of a wide range of APIs will be performed in six river basin districts to get a more complete picture of sources, emissions and environmental concentrations of APIs. Based on the data from case studies and literature, the overall emissions of APIs and their impact on the environment in the BSR will be assessed. Different emission reduction measures will be evaluated by CWPharma. They include advanced municipal wastewater treatment, improved take-back schemes and disposal for unused medicines, dissemination of environmental data on pharmaceutical products, and environmental permitting of pharmaceutical plants. The best existing practices of the partner countries will be shared to promote the sustainable management of APIs in the Baltic Sea Region.

CWPharma is funded by the EU Interreg Baltic Sea Region Programme until September 2020.

www.cwpharma.fi

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Finnish Environment Institute (SYKE)

Less is More – Energy-efficient technologies for removal of pharmaceuticals and other contaminants of emerging concern

Micropollutants, or contaminants of emerging concern (CEC), referring to a number of various substances such as pharmaceuticals, biocides and endocrine disruptors, may have adverse environmental effects, especially in cases with sensitive recipients. Besides impacts on aquatic life, release of micropollutants is a threat to drinking water resources in areas where the access to fresh water for drinking water production is limited. The reduction of micropollutants reaching the wastewater treatment plants (WWTPs) from industries, households and hospitals is poor for a majority of the contaminants. This makes the WWTPs point sources for many of these substances. Therefore upgrading of WWTPs will be needed. Advanced treatment for removal of micropollutants will also enable reuse of water for various purposes. The overall idea of the project is to test new, cost-effective technological solutions, for removal of pharmaceuticals and other CECs as well as antibiotic resistant bacteria, suitable for small and medium WWTPs and to disseminate information on new technologies to the end users.

The project is funded by the EU Interreg South Baltic Programme until September 2020.

www.gfw.pl/projekty/less-is-more

Michael Cimbritz
Lund University

BONUS CLEANWATER – Eco-technological solutions to remove micro-pollutants and micro-plastic from contaminated water

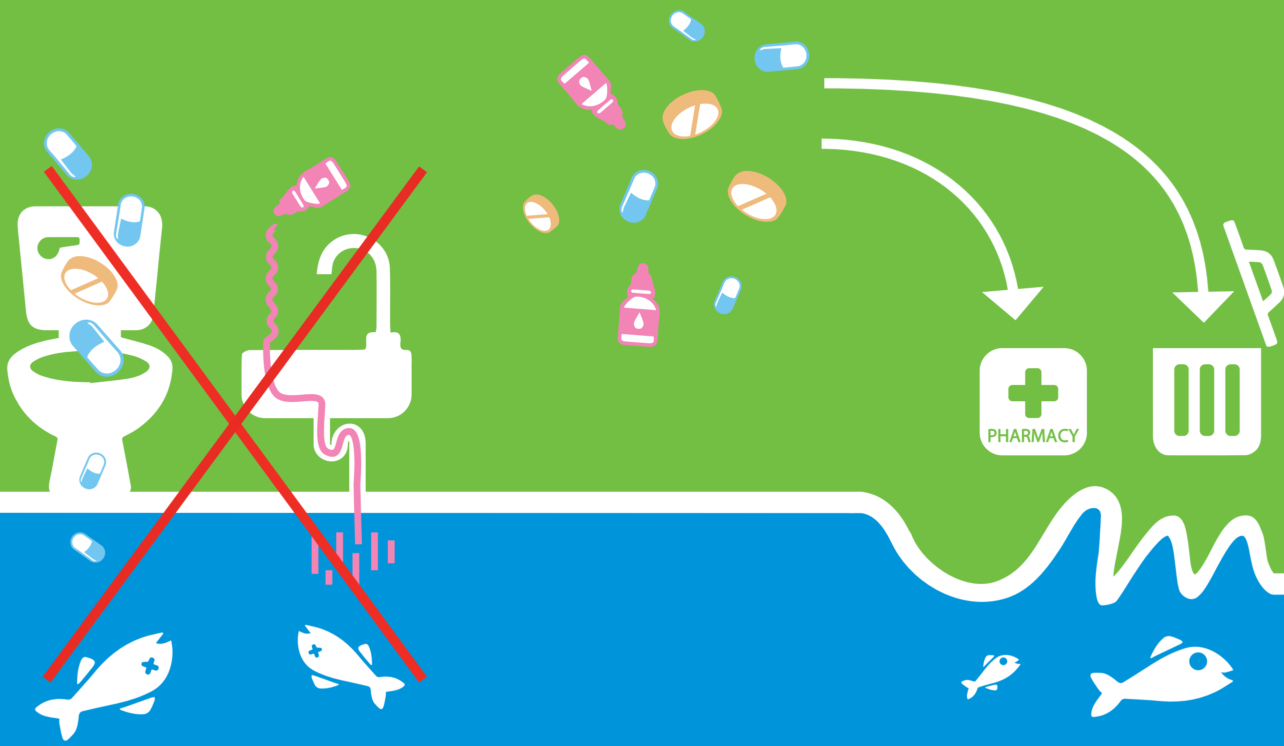
BONUS CLEANWATER is a research project focusing on reducing the input of micropollutants and microplastic from wastewater into the Baltic Sea by exploring, developing and comparing innovative eco-technological approaches. BONUS CLEANWATER will test removal technologies for selected hydrophilic (pharmaceuticals, etc.), lipophilic compounds (fragrances, flame retardants) as well as microplastics with different profiles in respect of matureness, energy demand and infrastructure profile. Innovative approaches for chemical oxidation, biofilm reactors, membrane bioreactors and biofilters will be tested and critical process parameters will be optimized. Adaptations of the respective stormwater treatment technologies will be performed. Innovative methods for testing for these compounds including metabolites and particles will be used and further developed.

The BONUS CLEANWATER project has received funding from BONUS, funded jointly by the EU and Innovation Fund Denmark, Sweden's innovation agency VINNOVA and the German Ministry for Education and Science (BMBF). The project's duration is until March 2020.

www.bonuscleanwater.com

Kai Bester
Aarhus University

DON'T FLUSH! DISPOSE OF MEDICINES CORRECTLY



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MORPHEUS
Model Areas for Removal of Pharmaceutical Substances in the South Baltic

Interreg South Baltic

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