A module-based approach for elimination of organic micropollutants at wastewater treatment plants

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ABSTRACT

A technical feasibility study was carried out at the wastewater treatment plant (WWTP) Hamm-West in 2018, which included preliminary planning for the improvement of the plant, using different advanced wastewater technologies. The results of the technical feasibility study show that the application of activated carbon or ozone, in combination with an additional filtration system, can not only remove organic micropollutants efficiently but can also significantly improve the quality of other standard parameters in the WWTP effluent. This technical feasibility study, along with seven other studies, is part of the module-based approach the Emschergenossenschaft and Lippeverband (EGLV) is pursuing in order to improve wastewater treatment plants with advanced treatment systems. Finally, the module-based approach can be used to pair the most suitable WWTPs with the best applicable technologies to improve the treatment process in the whole Lippe catchment area.

Key words | advanced wastewater treatment, organic micropollutants, technical feasibility study

HIGHLIGHTS

- Advanced wastewater treatment for organic micropollutants.
- Module-based approach.
- Wastewater and stormwater treatment plants.
- Technical feasibility study.
- Holistic approach.

INTRODUCTION

Organic micropollutants (OMPs) are forming an increasing threat to aquatic ecosystems and to the safety of drinking water resources (EC 2012). According to (Ternes 2001; Reemtsma et al. 2006; Kasprzyk-Hordern et al. 2009; Barbosa et al. 2016), most wastewater treatment plants (WWTPs) with conventional activated sludge processes (CAS) cannot sufficiently remove OMPs from municipal wastewater. The public concern for environmental aspects like micro-plastics and multi-drug resistant pathogens, as well as the requirements of the EU Water Framework Directive regarding nutrient concentrations in water bodies, have also raised the need to improve the treatment system.

Numerous advanced wastewater technologies have been developed as post-treatments to remove OMPs (Abeglen & Siegrist 2012), mainly making use of oxidation and adsorption processes with different technological variations, such as ozone, powdered activated carbon (PAC) and granular activated carbon (GAC). These advanced wastewater treatment technologies remove OMPs efficiently and economically (Metzger 2010; Abeglen & Siegrist 2012; Rößler & Metzger 2016) and in certain configurations, such as PAC in combination with a conventional granular-medium filter, can also further
reduce the concentrations of standard parameters (TSS, COD, TOC, N, P, etc) in WWTP effluent (Phan et al. 2018).

A technical feasibility study was carried out in 2018 at the WWTP Hamm-West (a 250,000 PE plant of the Lippeverband (LV)) with a maximal influent flowrate $Q_M$ of 6,552 m$^3$/h, to improve the WWTP's removal of OMPs. This study, along with another seven studies at WWTPs, which are representative of the capacities of WWTPs in the region (Figure 1), are components of a module-based approach for the improvement of wastewater and storm water treatment plants (SWTPs) at Emschergenossenschaft and Lippeverband (EGLV). The EGLV operates 60 WWTPs and numerous SWTPs in the densely populated and industrialised region of North Rhine-Westphalia, Germany. As such, it is essential to develop a module-based approach for the whole Lippe catchment area, which can target specific plants for improvement with flexible application of different treatment technologies to fulfil the stricter effluent standards (i.e. an elimination of 80% for micropollutant indicator substances or total phosphorus concentration in effluents lower than 0.8 mg/l). The module-based approach consists of four modules:

- Module 1: technical feasibility studies of advanced wastewater treatment stages for eight typical WWTPs, with capacities from 15,000 to 705,000 PE, in the Lippe catchment area (Figure 1).
- Module 2: monitoring of water bodies
- Module 3: environmental exposure modelling (GREAT-ER)
- Module 4: evaluation of costs and benefits for expansion scenarios of WWTPs and SWTPs

Finally, the module-based approach can be used to pair the most suitable WWTPs or SWTPs, with the best applicable technologies to improve the treatment process in the whole Lippe catchment area.

Figure 1 | Typical application of PAC with surface filter and ozone with downflow filter as post-treatment.
METHODS

Module 1: technical feasibility studies of WWTPs

The WWTP Hamm-West (250,000 PE) has a mechanical stage for grit removal and a biological stage with the conventional activated sludge process (Figure 3). To improve the WWTP’s capacity for OMP removal, it must be expanded by an advanced wastewater treatment stage. A technical feasibility study was carried out in 2018 to evaluate the conditions and applicable technologies for the expansion of WWTP. The study compared the application of granular activated carbon (GAC), powder activated carbon (PAC) with (Figure 1(b)) and without the return of PAC (Figure 1(a)) and ozone in combination with filtration (Figure 1(c)). From three typical application of PAC and ozone (Figure 1), eight state-of-the-art processes are developed (Table 1):

- Ozone + Downflow filtration (Variant 1.1.1, Figure 1(c))
- O3 + Granular activated carbon filter (Variant 1.3.1.1): instead of the downflow filtration, granular activated carbon filtration is built to enhance the elimination of transformation products from ozonation
- AFF + Downflow filtration (Variant 2.1.1.1, Figure 1(b)): PAC with the return of PAC and downflow filtration according to the principle Adsorption-Flocculation-Filtration (AFF)

Table 1 | Matrix to evaluate the applicability of various processes under different aspects at WWTP Hamm-West for the complete treatment ($Q_{\text{max, advance treatment}} = Q_{\text{max}} = 6,552 \text{ m}^3/\text{h}$) scenario

| Water bodies and ingredients | Ozone | PAC | GAC |
|-----------------------------|-------|-----|-----|
| OMPs removal (Ozone or Activated carbon: 2, combination of both) | 10 10,0% | 2 2.2% | 2 2.2% |
| Messaging | 5 5.0% | 2 2.2% | 3 3.3% |
| TSS | 2 2.0% | 2 2.2% | 3 3.3% |
| COD | 2 2.0% | 2 2.2% | 3 3.3% |
| Micro-plastics | 2 2.0% | 2 2.2% | 3 3.3% |
| Nanoparticles | 2 2.0% | 2 2.2% | 3 3.3% |
| Hygiene | 2 2.0% | 2 2.2% | 3 3.3% |
| Environment and resources | 25% | 44 66 46 46 63 55 76 48 |
| OMPs removal (Ozone or Activated carbon: 2, combination of both) | 10 10.0% | 2 2.2% | 2 2.2% |
| Messaging | 5 5.0% | 2 2.2% | 3 3.3% |
| TSS | 2 2.0% | 2 2.2% | 3 3.3% |
| COD | 2 2.0% | 2 2.2% | 3 3.3% |
| Micro-plastics | 2 2.0% | 2 2.2% | 3 3.3% |
| Nanoparticles | 2 2.0% | 2 2.2% | 3 3.3% |
| Hygiene | 2 2.0% | 2 2.2% | 3 3.3% |
| Risks (4 = no risk, 0 = high risk) | approx. 10% | 24 20 16 20 32 27 40 36 |
| Formation of metabolites | 4 4.0% | 4 4.0% | 2 2.2% |
| Activated carbon discharge to the environment | 2 2.0% | 2 2.2% | 3 3.3% |
| Application of polymers | 4 4.0% | 4 4.0% | 2 2.2% |
| Technology, Operation | approx. 25% | 33 27 23 32 19 23 85 22 |
| Compatible with re-investment plans (0 = not compatible) | 15 15.0% | 2 2.2% | 3 3.3% |
| Expandability / Flexibility (0 = difficult, 4 = simple) | 1 1.0% | 2 2.2% | 3 3.3% |
| Operational reliability (0 = not stable, 4 = very stable) | 1 1.0% | 2 2.2% | 3 3.3% |
| Complexity (0 = very high, 4 = very low) | 1 1.0% | 2 2.2% | 3 3.3% |
| References / Degree of testing (0 = Prototype, 4 = full-scale application) | 2 2.0% | 2 2.2% | 3 3.3% |
| Intermediate ranking (without costs) | 133 141 105 118 129 122 205 134 |
| Costs (0 = high cost, 8 = no cost) | approx. 30% | 10 10.0% | 4 4.0% | 2 2.0% |
| Operating costs | 20 20.0% | 4 4.0% | 2 2.0% |
| Overall ranking (with costs) | 100 100% | 253 241 215 208 188 202 238 224 |

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AFF + Surface filtration (Variant 2.1.2.1): instead of the downflow filtration, a surface filtration can be installed. Generally, a surface filtration has a lower demand on investment costs and places.

AFF + Ultrafiltration (Variant 2.1.3.1): instead of the downflow filtration, an ultrafiltration (membrane) can be installed. A membrane system has extra high quality of effluent; however, it has a high demand on investment and operation costs.

PAC + Surface filtration (Variant 2.4.3, Figure 1(a)): PAC applied without the return of PAC. This configuration has a low demand on investment costs and places.

PAC + MBR (Variant 2.4.1): the aeration tanks are converted to membrane bioreactors and PAC can be dosed to MBR directly.

Granular activated carbon filter (Variant 3.3.1.2): a granular activated carbon filter is built after the secondary clarifier.

As the OMPs concentrations in WWTP influent vary from dry weather to wet weather, the expansion of the WWTP has been considered for two scenarios. First, the expansion with the maximal dry-weather WWTP has been considered for two scenarios. First, the expansion with the maximal dry-weather to wet weather, the expansion of the WWTP. Second, the expansion with the maximal influent flowrate of 3,276 m³/h and partial flow treatment. Second, the expansion with the maximal influent flowrate of 6,552 m³/h and full flow treatment.

For the application of ozone, lab-scale experiments were carried out in 2018 to investigate the estrogenic activity (A-YES Assay) and the formation of bromate and nitrosamines. 24-hour composite effluent samples were collected then analysed for bromide, bromate, nitrosamines and estrogenic activity. Afterwards, the samples were treated with different ozone to dissolved organic carbon (DOC) ratios, from 0.3 to 1.6 mg O₃/mg DOC (Table 2). Finally, the products after ozoning were measured again for bromide, bromate, nitrosamines.

To evaluate the eight state-of-the-art processes for advanced wastewater treatment at the WWTP Hamm-West, a rating matrix was developed. Each element of the matrix was rated on a scale from one to four (except costs from 0 to 8) and consists of the following criteria:

- Water bodies and ingredients (0 = no improvement, 4 = significant improvement)
- Environment and resources (0 = high consumption, 4 = no consumption)
- Risks (4 = no risk, 0 = high risk)
- Technology, Operation
- Costs (0 = high cost, 8 = no cost)

According to the rating matrix, eight state-of-the-art processes for advanced wastewater treatment were evaluated, considering everything from water quality and effluent parameters to overall costs.

Instead of carrying out feasibility studies and examining 30 WWTPs individually, comprehensive monitoring and technical feasibility studies at representative selected WWTPs aim at a holistic, integrated water assessment in the Lippe catchment area. In cooperation with the authorities, WWTP Hamm-West and seven other representative WWTPs (Figure 2) were selected for the technical feasibility studies, based on a prioritization of criteria (size class, the wastewater ratio in the receiving water body and surface water quality). The seven feasibility studies for WWTPs Reken, Hünxe, Herten-Westerholt, Dortmund-Deusen, Dortmund-Scharnhorst, Soest, and Bönne have been in progress since 2019.

| Effluent of WWTP Hamm-West | Input O₃ mg/L | DOC mg/L | Specific ozone dose mgO₃/mgDOC | Bromate μg/L | N-Nitrosodimethylamin μg/L |
|---------------------------|--------------|----------|-------------------------------|--------------|----------------------------|
| Sample A                  | 2            | 8.0      | 0.3                           | 3.7          | 0.0090                     |
| Sample A                  | 5            | 8.0      | 0.6                           | 9.2          | 0.010                      |
| Sample A                  | 10           | 8.0      | 1.3                           | 50           | <0.005                     |
| Sample B                  | 2            | 6.3      | 0.3                           | 5.0          | 0.011                      |
| Sample B                  | 5            | 6.3      | 0.8                           | 13           | 0.012                      |
| Sample B                  | 10           | 6.3      | 1.6                           | 52           | <0.005                     |

Module 2: monitoring of water bodies

In order to get an overview of the treatment efficiency of the conventional WWTPs and the water quality with regard to organic micropollutants in the Lippe catchment area, monitoring was carried out. The monitoring program includes the WWTP influent and effluent of the eight selected WWTPs and surface water samples.

The monitoring program ran six times over a period of 12 months. 24-hour mixed samples were taken in the WWTP influent and effluent. The surface waters (scoop samples) were examined above and below the outflow of the WWTP into the receiving rivers. Additionally, surface water was measured at eight gauging sites along the Lippe river and its tributaries. The surface water samples were taken on the same day as the other samples (water phase only).
The WWTP data was used to calculate the current treatment efficiency and the per capita excretion. The river flow rates served to determine the loads of the key parameters (s.b.) and to calibrate the model in Module 3.

A list with 127 parameters was selected (see appendix), including 56 general chemical parameters and metals, 12 perfluorinated tensides, 51 pesticides, one anticorrosive and seven drugs and metabolites. The parameters were chosen from the results of the WFD-monitoring and under consideration of the guidelines of the Competence Center for Micropolllutants NRW (ARGE Kompetenzzentrum Mikroschadstoffe.NRW 2016).

The further modeling focused on the seven key parameters: carbamazepine, clarithromycin, diclofenac, metoprolol, sulfamethoxazole, terbutryn and 1H-benzotriazole. Again, the guidelines of the Competence Center for Micropolllutants NRW (ARGE Kompetenzzentrum Mikroschadstoffe. NRW 2016) were taken into account for establishing this list.

Module 3: environmental exposure modelling

In a next step, the model GREAT-ER 4.1 was applied. GREAT-ER stands for Geography-Referenced Regional Exposure Assessment Tool for European Rivers. The GREAT-ER model is a geo-referenced exposure model for water catchment areas, which was originally developed for the simulation of typical household and industrial chemicals. It calculates the chemical concentrations and water quality of the seven key substances along the surface waters. The model estimates concentration distributions at different points in the water, assuming steady state and constant emissions. GREAT-ER is used as an ArcGIS® add-in. The model of the Lippe catchment was created by the University of Osnabrück (IUSF) on the basis of the existing NRW model (Klasmeier et al. 2011). The checking and correction of the input data was carried out by the Lippeverband.

The model divides the waters into segments with a section length up to 2,000 m. The load $L$ (mg/day) of the substance in the water phase is calculated at the beginning $L_0$ and at the end $L_{end}$ of each water section. The concentration ($\mu$g/liter) in the water sections is determined based on this load and the corresponding mean discharge value. In a given segment, loss processes are modelled assuming a kinetic (pseudo) first order with the rate constant $k$ (1/h). The model takes into account the loss processes of hydrolysis, biodegradation, photolysis and sedimentation. In the case where only little information about the behaviour of the substances in the water body is available, the loss process can be described with a single degradations rate. So
for the poorly degradable, for example, diclofenac, the processes of photolysis and sedimentation are mainly responsible for the reduction of concentration. For diclofenac, the model uses literature values for the total degradation rate, which is 0.002 l/h. The load is reduced according to the following equation where $HRT$ is the hydraulic retention time spent in a segment. $HRT$ was not investigated, it was taken from the existing NRW model. The residence time is taken to be the quotient of the segment length $l$ (m) and the flow velocity $u$ (m/sec) of a river.

$$L_{\text{end}} = L_0 \times \exp(-k \times HRT) \quad HRT = \frac{l}{u}$$

GREAT-ER requires a geo-referenced, fully connected, topologically correct water network, and the geo-referenced connection of point dischargers. The simulation takes substance-specific data into account; for example, per capita consumption values, excretion rates and degradation rates in wastewater treatment plants. The consideration is made for medium mean flow (mean discharge at a measuring point in one normal year) and low mean flow (arithmetic mean of the lowest outflows of similar periods). To calibrate the model hydraulically, data from a flood forecasting model was used. The calibration of the key parameter concentration could be carried out with the monitoring data.

**Module 4: evaluation of costs and benefits for expansion scenarios of WWTPs and SWTPs**

The costs from module 1 are evaluated with the benefits, which are results from module 2 and 3. This evaluation can identify the most suitable WWTPs and SWTPs for expansion.

**RESULTS AND DISCUSSION**

**Module 1: technical feasibility studies of WWTP**

At the technical feasibility study of WWTP Hamm-West, the rating matrix for the complete treatment scenario ($Q_{\text{max, advance treatment}} = Q_M = 6,552$ m$^3$/h) shows that three technological variants ($O_3 +$ granular activated carbon filter, powdered activated carbon $+$ hybrid-membrane bioreactor and granular activated carbon filter) would have the most advantages to the other five technological variants, if the costs are not considered. However, the variants $O_3 +$ down filter and PAC $+$ surface filter are ranked better because of their moderate investment and operational costs, in comparison with the membrane and GAC variants at the final ranking (Table 1). For all the variants, it is expected to achieve an 80% elimination of micropollutant indicator substances.
Preliminary planning for the WWTP improvement with the four highest ranked variants was also included in the technical feasibility study.

For the variants with ozone, the preconditions for the ozone process were verified by running the experiments outlined in Wunderlin (2017). The estrogenic analysis shows that the estrogen concentrations in the effluent lie under the determination limit of 0.01 ngEQ/l. Neither bromate nor nitrosamines were detected in the effluent of the WWTP Hamm-West. The limits of detection for bromate and nitrosamines are 1 μg/l and 0.005 μg/l respectively. For bromide, an average concentration of 190 μg/L Br⁻ was measured in collected effluent samples.

It can be seen from Table 2 that bromate can already form at input O₃ concentrations as low as 2 mg/l O₃ or 0.3 mg O₃/mg DOC, while typical applied specific ozone doses range between 0.5 and 0.7 mg O₃/mg DOC. The environmental quality standard recommends limiting bromate concentrations to 50 μg/L (Wunderlin 2017). At an O₃ dosing up to 0.6–0.8 mg O₃/mg DOC, the formed bromate concentrations are under the environmental quality standard of 50 μg/L. However, the bromate concentrations would be higher than the recommended quality standard for O₃ dosing above 0.8 mg O₃/mg DOC. For the nitrosamines, NDMA could only be detected from samples where the O₃ dosing was above 0.8 mg O₃/mg DOC. In these cases, the NDMA concentrations could exceed the drinking water limit of 10 ng/l. However, NDMA can be degraded in post-treatment, for example using a downflow filter or GAC-filter (Böhler et al. 2017). To sum up, the effluent from the conventional activated sludge stage of WWTP Hamm-West should be suitable for application of ozone with a post-treatment.

Figure 3 shows the flow diagram of variants with ozone. In these variants, the wastewater is treated by a downflow filter after ozoning. At this stage, transformation products from the ozoning process (e.g. NDMA) can be biodegraded by microorganisms in the filter medium. The transformation products can also be adsorbed by GAC, if the post-treatment is done with a GAC filter. For the variants with ozone, necessary plant equipment comprises the following major units:

- an ozone supply system consisting of four ozone generators, with a total maximum capacity of 80 kg O₃/h (O₃-dose 2–10 mg/l),
- a reaction tank (V = 2,185 m³) for a minimal hydraulic retention time of 20 min,
- a downflow filter with 16 filter chambers and total filter area of 440 m². The maximal filtration rate is 15 m/h.

The filter medium is anthracite. For the variants with GAC-filter, the anthracite will be replaced by GAC.

For the activated carbon variant where PAC is recirculated between a sedimentation tank and the contact tank (Adsorption-Floculation-Filtration (AFF) process or Ulmer process in Germany, Figure 2(b)), the available space for the sedimentation tanks is a limiting factor. However, PAC can also be dosed directly before the filter (surface filter or membrane filter) and then returned to the CAS stage by backwashing. The used PAC is then removed from the system through the excess sludge removal process.

The technical feasibility study shows that both the application of ozone with post-treatment and PAC without recirculation are feasible for the advanced wastewater treatment stage at WWTP Hamm-West, in both partial flow and full flow treatment scenarios. The advanced wastewater treatment stage can reduce numerous OMPs from the wastewater and improve the effluent quality. The overall costs of ozone application with downflow filter and PAC with surface filter are similar. Specific costs for the solutions involving the downflow filter and PAC with a surface filter are about 0.1 €/m³ treated wastewater for partial flow treatment and 0.12 €/m³ treated wastewater for full flow treatment. The application of PAC with a membrane filter would result in the best effluent quality based on OMPs, total suspended solids and total phosphorus, but also have a double overall cost in comparison with two other applications.

Module 2: monitoring of water bodies

The results of the water monitoring confirmed those of the official monitoring (Water Framework Directive). Area-wide exceedances could be demonstrated for the metals (copper, zinc) and pharmaceuticals (diclofenac, ibuprofen) as well as sporadic exceedances for perfluorooctansulfonacid and crop protection products. Polycyclic aromatic hydrocarbons were not measured and mercury was unremarkable in the water phase.

Table 2 shows the treatment efficiency of the seven key parameters calculated from the conventional WWTPs (CAS) influent and effluent. With carbamazepine, a negative efficiency is obtained because the breakdown products are included. Götz et al. (2015) also confirms the effect of negative efficiency (Table 3). Here, various pharmaceuticals were evaluated with regard to their degradability. The results are comparable with these tests in the order of magnitude.
The model GREAT-ER has already been applied in several catchment areas in Germany. It was used in general for the development of strategies to reduce OMPs and for the cost-benefit assessment of introducing an advanced wastewater technology (Kolisch et al. 2016; Knerr et al. 2018; Matthies et al. 2003).

GREAT-ER enables a comprehensive visualisation of predicted chemical concentrations in the Lippe catchment area. The aim was to optimise the expansion of individual WWTP with regard to water quality. So different scenarios had to be defined to create a management basis. In the development of these scenarios, various environmental objectives were discussed and corresponding criteria included:

1. A load reduction is achieved through a selection via the wastewater treatment plant sizes. The focus here is on large wastewater treatment plants (>100,000 PE). The load out of the whole catchment should be reduced and can be compared to the measures across regions.

2. Regarding the percentage of wastewater from wastewater treatment plants in the water, the focus is usually on the smaller rivers. It is about improving the water quality on the longest possible waterways. Figure 4 shows the rivers with a wastewater fraction over 1/3 of the medium mean flow. Especially the upper courses are affected and the main stream Lippe because of the high population density.

3. A third criteria could be to bring the pollutant concentrations in the water below the limit values or to reduce them by 80% in general. Thus, compliance with the target concentrations at the monitoring points takes into account the legal environmental quality standards.

The statements of the model refer to mean discharges. The substances were also selected with regard to their main entry path via the wastewater treatment plants. Concerning discharges from storm water treatment plants, they cannot be considered as separate point sources with this model. So instead they must be included proportionally via the discharge rate of the catchment areas. The calculations outside the model assume that only an amount of 3–4% of the key substances loads (with the exception of terbutryn) are led past the wastewater treatment plants and enter the water body through the storm water treatment plants without biological treatment. Knerr et al. (2016) showed similar results in their model calculation. About

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**Table 3** | Measured medium treatment efficiency in eight selected WWTP

| Substances (key parameters) | Efficiency CAS |
|----------------------------|---------------|
| Lippe, Germany | Switzerland |
| Pharmaceuticals | | |
| Carbamazepine (anti-epileptic) | – 14% | – 22% |
| Clarithromycin (antibiotic) | 10% | 35% |
| Diclofenac (analgetic) | 29% | 29% |
| Metoprolol (betablocker) | 31% | 24% |
| Sulfamethoxazole (antibiotic) | 37% | 65% |
| Pesticide | | |
| Terbutryn | 13% | not measured |
| Anticorrosive | | |
| 1-H benzotriazole | 58% | 31% |

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**Figure 4** | WWTPs in the river ‘Lippe’ catchment area with the sewage fraction in the receiving rivers.
7% of the annual load of diclofenac could arrive in the surface water due to combined sewer overflows.

The modelling could represent that with the improvement of the wastewater treatment plants, the concentration and load of the key parameters in the water body can be significantly reduced. The model also showed that it is difficult to reduce the concentrations of all key parameters below the quality objectives. In this densely populated area, the wastewater fractions in the water bodies are simply too high.

With this module-based approach, we have a differentiated basis for further management planning.

CONCLUSIONS

The technical feasibility study of WWTP Hamm-West and other technical feasibility studies in module 1 show that the applications of ozone and PAC should be suitable for the eight studied WWTPs. For the application of ozone, an investigation of effluent CAS for transformation products, especially bromate formation, is necessary. In spite of high costs, the application of PAC with membrane filter is an interesting solution due to its high performance in particle separation.

The stormwater treatment plants are mainly under discussion with regard to the input of sediment-bound substances (metals and phosphorus). Here, measurements and possibly further modelling must help to determine the main points of discharge.

The monitoring showed that benzotriazole was detected at every water monitoring station site and the pharmaceuticals and the biocide terbutryn at almost all water monitoring sites.

The results of these modelling scenarios will be a basis for the further decision-making process. The discussions will include cost-benefit aspects as well before it will lead to a plan of measures that considerably reduces OMPs.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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