Swedish Agency for Marine and Water Management

Treatment techniques for pharmaceuticals and micropollutants in wastewater

Description of eight projects that have received funding from the Marine and Aquatic Environment Grant for 2014-2017

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Michael Cimbritz, Ann Mattsson

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Foreword

For four years, from 2014 to 2017, the Swedish Agency for Marine and Water Management has on behalf of the government funded various projects to promote advanced treatment of wastewater by means of case funding 1:11 Actions for the Marine and Aquatic Environment. This involves supporting projects in order to develop treatment methods for municipal treatment plants to reduce emissions of pharmaceutical residues and other pollutants that are difficult to remove. 32 million Swedish kronor has been awarded to a total of eight projects over the four years. This report, provides a brief description of the most important results from these projects. For more detailed information, please see the reports and articles from the various projects.

The report has been compiled by Michael Cimbritz of Lund University and Ann Mattsson of Gryaab. Margareta Lundin Unger at the Swedish Agency for Marine and Water Management was the project coordinator for the call for projects and their coordination.

The results show that techniques are available at present that can be applied at municipal treatment plants in order to reduce micropollutants of different types, including pharmaceutical residues. This assignment has paved the way for new research and development projects in a number of instances and created a strong foundation on which to further build for the potential introduction of advanced treatment at Swedish wastewater treatment plants.

We would particularly like to thank the people at the different authorities, universities, research institutes and companies who contributed in various ways. In particular, we would like to thank Daniel Hellström and Anders Finnson (Swedish Water & Wastewater Association), Stefan Gabring (Swedish Chemicals Agency), Kia Salin (Swedish Medical Products Agency) and Linda Gårdstam and Anna-Maria Sundin (Swedish Environmental Protection Agency) for their contributions to the work, as well as the managers of the various projects, who contributed figures, illustrations, photographs and other information for the report; Emelie Ljung (RISE), Robert Sehlén (Tekniska Verken in Linköping), Ola Svahn and Erland Björklund (Kristianstad University), Christian Baresel (IVL Swedish Environmental Research Institute), Jerker Fick (Umeå University) and Berndt Björlenius (KTH Royal Institute of Technology).

Gothenburg, 12 June 2018

Björn Sjöberg
Head of Department, Swedish Agency for Marine and Water Management

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Summary

The Swedish Agency for Marine and Water Management has on behalf of the government funded eight projects with a view to evaluating treatment methods that could be applied in practice in order to reduce potentially environmentally hazardous substances that are not removed by present treatment processes at municipal wastewater treatment plants. Six different research and development projects have been completed under the management of researchers linked with RISE, Tekniska Verken in Linköping, Kristianstad University, IVL Swedish Environmental Research Institute, Umeå University, Lund University and KTH Royal Institute of Technology. Municipal water supply and sewerage organisations and companies have played key roles in the various projects. Extensive experiments have been performed, from laboratory scale to long-term, full-scale trials at wastewater treatment plants in various parts of Sweden.

In addition to these projects, a further two projects have been completed within the scope of the call for projects. An inter-calibration study was carried out at Kristianstad University with the aim of achieving enhanced analysis quality and increasing awareness of problems associated with trace analysis of pharmaceutical residues. At Lund University another project, based on literature studies and a study trip to Switzerland and Germany, was conducted to pass on information and operational experience from sewage treatment plants supplemented with advanced treatment facilities.

The results from the projects show that techniques are available at present that can be applied at Swedish municipal wastewater treatment plants with the aim of removing micropollutants of different types, including pharmaceutical residues. The technical solutions evaluated are mainly based on ozonation or filtration through activated carbon, as well as various combination solutions. In most cases, the solutions have been tested and evaluated in close cooperation with staff at wastewater treatment plants: this is a prerequisite to be able to evaluate the techniques in a credible manner and thus to be able to develop well-functioning large-scale solutions. One of the projects has also studied systems involving sorting at source.

This work has paved the way for new research and development projects where the involved parties can assist with and lead development of the wastewater treatment of the future. This concerns, for instance, ecotoxicological effects of ozonation, development and understanding of applications based on activated carbon and development of analysis techniques.

The government assignment and work on the various projects have created a strong foundation that will aid the introduction of advanced treatment at Swedish wastewater treatment plants. This report briefly describes the background to the work and the results from the projects in popular scientific form. Costs have been produced for various treatment techniques and summarised in the report, and further reading is suggested.

Introduction

For a long time, Swedish municipal wastewater treatment plants have met stringent requirements to remove organic material and phosphorus. Since the 1990s, many wastewater treatment plants have also added treatment steps to reduce nitrogen. Other substances can also be degraded by the microorganisms whose primary task is to remove organic material or nitrogen. Some substances attach to particles and can be removed with the sewage sludge, even though removal of this substance is not the specific objective. Source control is important for substances that are not degraded or removed at wastewater treatment plants and may harm the environment. This is known as upstream management and is part of the long-term cooperation between the water and wastewater utilities and the environmental authorities. Phasing out such substances has a greater general positive impact on the environment than merely improving the quality of water and sludge at wastewater treatment plants. As far as pharmaceuticals and persistent contaminants are concerned, the government has concluded that various different measures are needed in order to limit risk, both in connection with the development, manufacture and management of pharmaceuticals, as well as by supplementing wastewater treatment plants with advanced treatment methods.

Development of new treatment techniques

The Swedish Agency for Marine and Water Management has on behalf of the government funded various projects with a view to evaluating treatment methods that could be applied in practice in order to remove potentially environmentally hazardous substances that are not removed by the present treatment processes at municipal wastewater treatment plants. An initial call for projects took place in 2014 with a view to providing funding for planning, implementation, evaluation and documentation of surveys for reduction of pharmaceutical product residues and other persistent contaminants. Subsequent calls took place in 2015 and 2016. This report briefly describes the background to the work and the results from the various projects in popular scientific form.

Six different research and development projects have been conducted:

- Project 1. Pharmaceuticals in source-separated blackwater and faecal sludge – Treatment and risks – *LäK*
- Project 2. Removal of Pharmaceutical Residues Using Ozonation as Intermediate Process Step at Linköping WWTP, Sweden
- Project 3. Full-scale treatment of micropollutants – *FRAM*
- Project 4. Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances– *SystemLäk*
- Project 5. Evaluation of advanced full-scale treatment
- Project 6. Treatment of persistent contaminants in wastewater – *RESVAV*

A further two projects have been implemented besides these six major projects:

- Project 7. Knowledge synthesis on treatment to remove pharmaceutical residues and other micropollutants
- Project 8. Intercalibrated pharmaceutical analysis – a collaborative project to enhance analysis quality.

These two projects are summarised in the sections entitled *Other countries – in Europe and elsewhere* and *Analysis of micropollutants*.

What is the problem, and what happens at treatment plants?

A number of studies show that organic micropollutants, including various pharmaceuticals, are capable – or are suspected of being capable – of causing harmful effects on plant and animal life, even at very low concentrations (see, for example, Fick et al., 2010, Larsson et al., 1999). Pharmaceutical residuals appear in wastewater for a number of reasons. Most of the pharmaceutical residuals in wastewater come from pharmaceuticals that pass through the body and are excreted in the urine. Other pharmaceuticals are applied in the form of ointments and creams and may be present in water from showers and washing. A small percentage of pharmaceuticals may also end up in sewage directly from manufacturing processes, if they are present in the area, or if pharmaceuticals are discarded into the sewer systems.

As regards other organic micropollutants, some of them are substances that have been manufactured specifically to have some form of toxic effect and at the same time to be diluted and transported with water, such as herbicides (designed to kill weeds). They may also be substances in consumer products and their degradation products, with properties making them environmentally harmful. There are also substances that have been used historically or are present as contaminants in other materials (e.g. PCBs), as well as contaminants formed unintentionally (e.g. dioxins) that may end up in sewage under specific conditions. Some of these substances are now prohibited, but are still to some extent present in the community and the environment. Distribution pathways for organic micropollutants may vary widely. Some of them are present in wastewater, others are not (see Baresel et al., 2015 for a review and grouping of different organic micropollutants in wastewater). In practice, this means that both upstream management and advanced wastewater treatment are important pieces of the puzzle aiming to reduce levels of harmful substances in the environment.

Antibiotic resistance is a growing threat as treatment of various infections is being impeded or rendered impossible. Discussed risks of antibiotic resistance linked with wastewater treatment plants involve a number of different mechanisms:

- Existing antibiotic-resistant microorganisms from wastewater being spread to the environment.

- Strains of antibiotic-resistant microorganisms developing at wastewater treatment plants.
- Antibiotic resistance occurring after the wastewater treatment plant due to antibiotic residues in the treated wastewater.
- Combinations of the above.

One of the most important functions of the wastewater treatment plant is to reduce the risk of spreading waterborne infection. This is done by removing particles and by means of biological processes where bacteria that spread infection are outcompeted by bacteria that specialise in degrading organic material. This significantly decreases the number of infection-spreading bacteria in the water. If further reduction of pathogenic bacteria is needed, with or without antibiotic resistance, tried and tested methods are available for disinfecting wastewater. These include chlorination, UV light or ozonation (Keen & Montforts, 2012).

Driving forces for advanced treatment

There may be a number of driving forces for the introduction of advanced treatment. Protecting the aquatic environment is one of them. Direct or indirect reuse of wastewater as drinking water or for watering purposes is another, and the precautionary principle is a third.

For EU countries, the Water Framework Directive (2000/60/EC), with the Priority Substances Directive (2013/39/EU) and the watch list (article 8b), provide a further driving force. Some of the substances included in the watch list – diclofenac, oestradiol and ethinylestradiol – have also been recorded as river basin-specific pollutants (RBSP) according to Swedish Agency for Marine and Water Management regulations on classification and environmental quality standards for surface water (HVMFS 2015:4). With this, Sweden became one of the first countries to introduce environmental quality standards for these substances.

The precautionary principle is referred to in various contexts as an important driving force for more stringent treatment requirements at wastewater treatment plants, and hence also for the introduction of new treatment technology. This principle means that an effect does not have to be observed, but that possible or likely effects involve an obligation to take action to prevent adverse effects on the environment and health. At the same time, large-scale development of wastewater treatment plants involves environmental impact in the form of an increased need for energy and chemicals and a developed infrastructure, which should be weighed up against the benefits.

Protection of drinking water sources is another important driving force. Even if pharmaceuticals in a water body do not present any risk to human health, the very fact that certain substances have been found – albeit very low levels of these substances – may constitute a driving force for action based on a confidence perspective among water consumers (Joss et al., 2008). This is an example linked to indirect reuse of wastewater. When there is a high proportion of wastewater in the lake or river that constitutes a drinking water catchment, advanced treatment with regard to pharmaceuticals becomes increasingly relevant. This situation is prevalent in many places. In Germany it has already justified the introduction of advanced treatment at a number of wastewater treatment plants.

Six research and development projects

The various projects have been introduced in various parts of the country, focusing on different aspects. Brief descriptions of each project are presented below. Examples of results from the various projects are presented later on in this report. For further reading, please see reports and articles from each project, as summarised in Appendix I.

Project 1. Pharmaceuticals in blackwater and faecal sludge–Treatment and risks – LäK

Sewage systems with urine or blackwater sorting have considerably greater potential than conventional sewage systems to recycle – in particular – the plant nutrients nitrogen and potassium to farmland. On the other hand, a considerably greater proportion of the pharmaceuticals that in a wastewater treatment plant would risk being carried by the treated wastewater to the water body will reach farmland instead.

An important prerequisite for source-separating systems is therefore to describe which pharmaceuticals and quantities could be spread by using fertilisers from source-separated waste fractions, and what happens in the environment during plant uptake, transport and degradation in soil and groundwater. The purpose of the *LäK* project was to examine pharmaceutical residues in blackwater and faecal sludge, both before and after treatment and storage, and to calculate the pharmaceutical quantities that would be spread in agriculture compared with use of sewage sludge. This project has also calculated degradation in soil, accumulation and uptake.

The faecal sludge, which was taken from a waste plant in Norrtälje, was treated by means of digestion experiments at various temperatures. The blackwater was taken from the Telge Nät treatment plant in Hölö (Södertälje), where it is currently being treated by means of wet composting followed by urea hygienisation.

Participants and project managers

The project has been implemented in partnership by JTI (now known as RISE Agrifood and Bioscience), the Swedish University of Agricultural Sciences (SLU) and SPPD (SP Process Development). Lotta Levén and David Eveborn were the project managers. The contact person is Emelie Ljung from RISE Agrifood and Bioscience .

Project 2. Removal of Pharmaceutical Residues Using Ozonation as Intermediate Process Step at Linköping WWTP, Sweden

September 2017 saw the opening of Sweden's first permanent ozone plant for oxidation of pharmaceutical product residues at Nykvarn WWTP in Linköping. It is hoped that this will be able to reduce the pharmaceutical product residue load in the Stångån river, which runs through Östergötland and northern Småland. The purpose of this development project was to create a basis for design and operation of a full-scale plant. The plant is unique from a process engineering perspective as ozonation has been integrated with the wastewater treatment plant's nitrogen treatment in order to ensure post-treatment of the ozonated water.

Besides establishing the design criteria and highlighting operational aspects, the pilot experiments also examined diurnal variations and mass flows throughout the entire plant, potential control strategies, disruptions to subsequent biological treatment and options for reducing the problem of formation of ecotoxic by-products from ozonation. The aim was to reduce the most highly prioritised pharmaceuticals to levels that will not lead to adverse effects in the recipient.

Participants and project managers

The project has been implemented in partnership by Tekniska Verken in Linköping AB and IVL Swedish Environmental Research Institute. Robert Sehlén, Tekniska Verken, was the project manager and is the contact person.

Project 3. Full-scale treatment of micropollutants – FRAM

The aim of the FRAM project was to evaluate separation of organic micropollutants with granular activated carbon (GAC). Previous national surveys carried out on a laboratory scale indicated that GAC was, relatively expensive compared with ozonation, for example. The intention of the project was to challenge this cost scenario by carrying out filtration with GAC on a larger scale, and thereby studying whether GAC is a competitive treatment alternative in the case of more realistic modes of operation. This project was the only one that worked with GAC as a fourth treatment stage, which it would be possible to introduce separately after existing wastewater treatment plants if costs can be reduced.

A completely new technique for studying chemical binding has been developed within the project to permit evaluation of various carbon types and isolation of various pharmaceuticals. It was possible to use a new analysis laboratory at Krinova Incubator & Science Park in Kristianstad to try out appropriate carbon and then perform large-scale testing, initially at the Osby wastewater treatment plant and, later, at Kristianstad wastewater treatment plant. The project grouped various pharmaceuticals based on their chemical properties (and not medical effect). This grouping was then used to identify which parameters control the binding of pharmaceuticals to the activated carbon. Further knowledge of which chemical processes control binding can be used to develop more advanced adsorptive treatment techniques. A pilot facility was constructed for the trials, with a sand filter followed by an activated carbon filter. Besides this, a new pharmaceuticals analysis method using UPLC-MS/MS was developed which included the list of 22 substances that the Swedish Medical Products Agency proposed as environmental indicators in *"Miljöindikatorer inom ramen för nationella läkemedelssatsningen (NLS)"* [Environmental indicators within the scope of the national pharmaceutical initiative (NLS)] in 2015.

Participants and project managers

The project has been implemented in partnership by Kristianstad University (HKR), Skåne Blekinge Vattentjänst AB (SBVT), Malmberg Water AB, Krinova Incubator & Science Park and the municipality of Kristianstad. Ola Svahn and Erland Björklund, HKR, acted as project managers and are contact persons.

Project 4.Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances– SystemLäk

Within the SystemLäk project, the aim was to produce recommendations on advanced treatment technology to remove pharmaceuticals and other micropollutants from a comprehensive perspective. Experiments involving different techniques have been planned and carried out by means of extensive mapping of the knowledge base and existing gaps in knowledge. Various system solutions have been examined in terms of a number of aspects, including total environmental impact, costs and treatment efficiency. Finally, an overall assessment of the various system solutions was carried out with a view to creating recommendations for implementation at various wastewater treatment plants under different conditions, such as existing treatment process, receiving body of water, etc.

Particular emphasis has been placed throughout the project on creating a holistic approach, analysing not only pharmaceuticals but also other persistent contaminants, referred to collectively as micropollutants. A number of different techniques have also been studied, along with ongoing technical development and consequences for sludge handling, in order to create a holistic approach.

The overall assessment has described consequences in the form of costs, overall environmental impact and flexibility in respect of future challenges when introducing advanced treatment.

Participants and project managers

The project has been implemented in partnership by IVL Swedish Environmental Research Institute, Stockholm Vatten and Avfall AB, Sydvästra Stockholmsregionens VA-verksaktiebolag (SYVAB) and KTH Royal Institute of Technology (KTH).
Christian Baresel, IVL, was the project manager and is the contact person.

Project 5. Evaluation of advanced full-scale treatment

In this project, a plant for ozonation was established in Knivsta, south of Uppsala. This plant ended up becoming Sweden’s first full-scale plant as ozonation was designed to cope with the entire flow at the wastewater treatment plant, equivalent to 12,000 PE. The ozonation stage at Knivsta was constructed after the existing treatment plant but before an existing “polishing pond”, after which the treated wastewater runs out into the receiving water, the Knivstaån river. The establishment of a plant of this size is providing valuable practical experience of both implementation and operation that can be used in other projects later. Optimisation of the use of resources has been an important objective.

Reduction of 120 pharmaceuticals has been studied as part of the project. Resistance spread experiments and ecotoxicological experiments have been carried out in parallel, studying effects in the environment relating to hormone disorders, organ changes and behaviour in fish. As the entire flow from the wastewater treatment plant was processed, this project has provided a unique opportunity to describe the ecological status of the receiving water, the Knivstaån river, both before and after the introduction of advanced treatment. Figure 1 shows the plant in Knivsta.

Figure 1. Photo of the ozone plant in Knivsta. Photo: Berndt Björlenius.

Participants and project managers

The project has been implemented in partnership by Umeå University, KTH Royal Institute of Technology (KTH) in Stockholm, the University of Gothenburg and the Swedish University of Agricultural Sciences in Uppsala. Jerker Fick, Umeå University, was the project manager, and he is contact person together with Berndt Björlenius, KTH Royal Institute of Technology.

Project 6. Treatment of persistent contaminants in wastewater – RESVAV

In the RESVAV project, the aim was to develop treatment processes for reduction of pharmaceutical product residues and other persistent contaminants. In practice, this involved efforts to establish guidelines and dimensioning criteria for the operation and development of Swedish wastewater treatment plants of different types. The criteria for the introduction of advanced treatment may vary widely in different parts of the country and at different wastewater treatment plants. Some plants allow nitrogen treatment; others do not. Some wastewater treatment plants have small receiving bodies of water with low dilution, while for others the sea is the nearest receiving water.

Understanding these and other criteria provides important starting points for identification of development needs and construction of efficient, cost-effective plants. The project included implementation of pilot-scale ozonation at a number of wastewater treatment plants in southern Sweden. During the later phase of the project, the first few steps towards development of a new treatment process have been taken, combining addition of powdered activated carbon with suspended biofilm carriers.

Participants and project managers

The project has been implemented in partnership by Gryaab, Department of Sustainable Waste and Water, City of Gothenburg, North West Skåne Water and Wastewater AB (NSVA), Sweden Water Research (SWR), the Department of Chemical Engineering at Lund University, Primozone Production AB, Sweco and Aarhus University. Michael Cimbritz, Lund University, was the project manager and is the contact person.

Other countries – in Europe and elsewhere

Advanced treatment at municipal wastewater treatment plants has been introduced in a number of countries such as Germany and Switzerland. At the same time, technology for removal of organic micropollutants has been in use for a long time in respect of drinking water technology and for reuse of wastewater. Study visits under the direction of Lund University (Project 7) took place within the scope of the initiative with a view to transferring knowledge relating to the full-scale plants that were constructed.

Switzerland – the first country in the world to introduce legislation

Switzerland is the only country in the world today to have enforced legislation that results in more extensive development of the country's wastewater treatment plants. A national initiative (*Strategy Micropoll*) was formulated based on various research projects, where it was established that treatment to remove micropollutants would result in significant improvements to water quality. As such, selected wastewater treatment plants should be upgraded, initially with either ozone or activated carbon. At the same time, a political discussion began, and it was possible to establish fairly quickly that the general public was willing to pay for the upgrade. The legislation came into force in 2016, some ten years after the start of the investigations. Over the next 25 years, around 100 of the country's 700 wastewater treatment plants will be upgraded with techniques for advanced treatment. A financing fund has been set up, based primarily on an increase in water supply and sewerage tariffs. Energy consumption at sewage treatment plants is estimated to increase by 5-30%, which is equivalent to a 0.1% increase in the total energy consumption for Switzerland.

Which wastewater treatment plants are to be upgraded?

All major wastewater plants (>80,000 people) are to be upgraded. The aim of this is to reduce the overall load on the environment and is justified by the fact that all plants in Switzerland are a long way from the sea, which is why it is assumed that every initiative will make a difference for a relatively long stretch of river. This action will affect half of the population. Besides the major plants, selected treatment plants will be developed in order to protect certain drinking water sources and particularly sensitive receiving bodies of water with insufficient dilution. Small wastewater treatment plants (<8000 people) will largely be exempt from the requirements, although some smaller wastewater treatment plants will have to be upgraded. For some smaller wastewater treatment plants the water will be transferred to larger plants instead, offering more cost-effective treatment. The first plants were commissioned in 2015, with treatment by means of ozonation or addition of powdered activated carbon.

Which substances are to be removed?

The legislation means that a number of substances must be reduced by 80% on comparison between the influent and effluent water. Substances, almost only pharmaceuticals (Table 1) that are affected to a lower extent by biological treatment have been selected as indicator substances. The reduction level of the various substances that can be achieved is approximately equivalent when using either activated carbon or ozone.

Table 1. Indicator substances for checking of advanced treatment.

Substance	Type
Amisulpride	Medication, antidepressant
Carbamazepine	Medication, tranquilliser
Citalopram	Medication, antidepressant
Clarithromycin	Medication, antibiotic
Diclofenac	Medication, anti-inflammatory
Hydrochlorothiazide	Medication, antihypertensive
Metoprolol	Medication, beta blocker
Venlafaxine	Medication, antidepressant
Benzotriazole	Anticorrosion agent
Candesartan	Medication, antihypertensive
Irbesartan	Medication, antihypertensive
Mecoprop	Biocide

Other countries

Although Switzerland is the first country to introduce legislation, its neighbour Germany has more full-scale plants for removal of persistent organic contaminants, primarily in the regions of North Rhine-Westphalia and Baden-Württemberg. These plants have been constructed in order to protect drinking water sources and sensitive receiving bodies of water with low dilution. In some cases, much of the load of a persistent fraction comes from connected industries. A few wastewater treatment plants have been upgraded in France as well.

Ozone, but also activated carbon, is used relatively extensively for wastewater treatment in various places, such as Japan and the US, primarily to be able to reuse treated wastewater. This may relate to everything from flushing toilets to cleaning and watering. Although the aim was not to remove or degrade micropollutants, effective reduction of these substances can be demonstrated on many occasions. In some cases, the water is treated to produce drinking water quality and reused as drinking water. Some of the most renowned examples can be found in Windhoek, capital of Namibia, and in Singapore. The world's biggest ozone plant is being constructed in Montreal, Canada at one of the largest wastewater treatment plants in the world.

More information can be found in the report that can be downloaded from www.svensktvatten.se (in Swedish).

Analysis of micropollutants

Reliable analyses are essential to being able to evaluate and develop new technology and assess the points at which any measures in the form of advanced treatment should be deployed. The various laboratories linked with the different projects – Umeå University, Swedish University of Agricultural Sciences, IVL Swedish Environmental Research Institute, Kristianstad University and Aarhus University in Denmark – have therefore embarked upon a comparative study (intercalibration project) under the direction of Kristianstad University (Project 8). The aim of this study was to achieve enhanced analysis quality and greater awareness of problems associated with trace analysis of pharmaceuticals. This project constitutes an important first step towards enhanced quality and national coordination, allowing measurement data to be compared in various contexts. Various issues relating to analysis technology have been addressed as part of FRAM (see Appendix I) and SystemLäk (see Magnér et al., 2017) within the scope of each project.

From milligrams to nanograms

Wastewater treatment plants are currently subject to requirements in order to restrict and prevent oxygen deficiency and eutrophication. In practice, this means that it should be possible to analyse concentrations of phosphorus, nitrogen and organic material to levels in the order of a few milligrams per litre. Determining levels of various pharmaceutical residues makes completely different demands of analysis techniques and methodology, as this may involve looking for a few nanograms of a specific substance that has to be both identified and quantified. (1 nanogram is 1 million times smaller than 1 milligram.)

Ibuprofen, a painkiller and antipyretic, is a familiar substance to many people. According to FASS, the maximum daily dose for an adult is 1200 mg, and this can be taken in the form of three tablets, each of 400 mg. If these tablets were thrown into Lake Ringsjön in Skåne instead, it is possible to use current analysis techniques to determine the concentration (10 ng/l) that would theoretically occur in the lake.

Difficult and simple substances

Ten substances were selected for analysis in the intercalibration study. These substances were analysed based on standard samples and authentic samples taken from Kristianstad wastewater treatment plant and Lake Hammarsjön, which is located downstream of the wastewater treatment plant and constitutes part of the Kristianstad Vattenrike Biosphere Reserve. Figure 2 shows the preparation of samples at Kristianstad University, ready to be sent out to the four laboratories.

Figure 2. Preparation of samples at Kristianstad University for dispatch to the laboratories involved in the intercalibration project. Photo: Erland Björklund.

Some of the substances (carbamazepine, citalopram, diclofenac, metoprolol and sulfamethoxazole) were relatively easy to analyse, while others were more problematic (ciprofloxacin, clarithromycin, ethinylestradiol, ibuprofen and tramadol). At the same time, it is more difficult to analyse different substances in wastewater compared with analysing them in clean water, such as drinking water. Of the substances that were more difficult to analyse, clarithromycin (antibiotic) and ethinylestradiol (synthetic hormone) are particularly interesting as they are included on the EU's watch list. The levels of these substances were underestimated by several laboratories in the survey, which could of course lead to corresponding underestimation of concentrations in the environment. Ethinylestradiol has proven very problematic to analyse in various contexts, which is unfortunate as effects are seen in the aquatic environment even at very low levels.

From level to assessment

Reliable analyses are absolutely crucial in order to be able to determine whether emission of a specific substance possess risks of adverse effects in lakes or waterways. One example is the painkilling substance diclofenac, which is regulated via Swedish Agency for Marine and Water Management regulations on classification and environmental quality standards relating to surface water (HVMFS 2015). The level must not exceed an annual mean value of 100 ng/l. Several measurements are required throughout the year to be able to assess a water body, but overestimating or underestimating the level may result in the status of the surface water being underestimated or overestimated. The level of diclofenac in treated wastewater varied between 800 and 1500 ng/l in this study when the different laboratories analysed water from the same sample. Such variation may of course have consequences when assessing levels after dilution in the receiving body of water or when assessing various mass flows. In other contexts, the relative difference between two values is more interesting. This may, for example, be the case when studying reduction using different techniques and comparing influent and effluent values. Systematic errors may be of less significance in such cases, but overestimates and underestimates may also give rise to incorrect assessments in this case, thereby highlighting the importance of reliable analyses.

More information can be found in the report that can be downloaded from www.havochvatten.se (in Swedish)

Source-separating sewage systems

Source-separating sewage systems where urine and faecal matter are dealt with without diluting them with large quantities of water is in principle a good solution if the purpose is to reduce the quantity of pharmaceuticals sent to the receiving body of water. This also has the advantage of allowing more of the nitrogen and potassium to be recirculated. Necessary remodelling of existing infrastructure and fears concerning the supply of pharmaceuticals to farmland have been some of the obstacles to application.

In one of the projects funded by the Swedish Agency for Marine and Water Management, *LäK* (Project 1), JTI has examined the risks involved in the supply of pharmaceuticals to agricultural land with source-separated fractions and with sludge from wastewater treatment plants (Levén et al., 2016). In the first stage, JTI has analysed pharmaceuticals in blackwater, faecal matter and sewage sludge during various sludge treatment stages. The level of pharmaceuticals is higher in blackwater than in sewage sludge. This was compensated for in part by the fact that pharmaceuticals were reduced more effectively during aerobic treatment of blackwater and subsequent urea treatment than with mesophilic or thermophilic digestion of sewage sludge. When the differences between fertilisation strategies involving blackwater and sewage sludge are included in the calculation, the supply of pharmaceuticals to agricultural land is of the same order regardless of whether the land is fertilised using sewage sludge or blackwater.

So far, JTI has been able to base its conclusions on analyses. To be able to estimate the risks to humans of the extremely small quantities of pharmaceuticals that could conceivably be absorbed in edible parts of plants, only modelling remains as an option as these are expected to be extremely low levels, far below the detection limits. These calculations have included degradation and leaching into soil, uptake in plants and, finally, the dose to which humans may be exposed via this route. The case has been overstated in a number of regards, and since there are no risk levels for pharmaceuticals, comparisons have used one ten-thousandth of a daily dose of each pharmaceutical product. Not even then were any pharmaceuticals identified for which the uptake could even theoretically approach the risk level. JTI also turned the analysis around and calculated how many years it would take a human to consume crops containing one daily dose of any of the pharmaceuticals. This would take more than 100,000 years for all the pharmaceuticals except for two. According to these results, it would take more than 20,000 years for a human to consume enough crops to take one daily dose of the pharmaceutical in question, whether the land was fertilised with anaerobically processed sludge or aerobically processed blackwater. The researchers have a number of suggestions on how to refine the calculations and improve the data.

Overview of technical solutions

Technical solutions for removal of micropollutants at municipal wastewater treatment plants must meet a number of different requirements. It must be possible to remove a broad spectrum of problematic substances, and it must be possible to justify investment and operation costs based on the benefit perspective. One important criterion for reasonable consequences in relation to benefit is that it must be possible to integrate the technology with existing infrastructure without jeopardising requirements for treatment from oxygen-consuming substances, nitrogen and phosphorus. If the new technology affects other functions such as biogas production or sludge handling and disposal, this must also be taken into account.

Different alternatives

There are four fundamentally different methods for removing micropollutants:

- Physical
- Adsorptive
- Oxidative
- Biological

It is also possible to combine the various alternatives. Removal may relate to both degradation and isolation of various micropollutants. A large number of different substances can be removed using physical and adsorptive methods. One physical method that has proven to result in high levels of separation is membrane filtration in the form of reverse osmosis or nanofiltration. Its disadvantages are high levels of energy consumption and a concentrate that may be problematic and costly to handle. The adsorptive methods include treatment with activated carbon, by either adding powder or filtering through granules. The disadvantage of this is that the adsorbent, the activated carbon, has to be replaced or regenerated at regular intervals.

The oxidative and biological methods are based on conversion and degradation rather than isolation. There are a number of different oxidative methods, but ozonation is the most common and has proven to be effective in many studies. One disadvantage is linked to the fact that oxidation does not generally result in complete degradation. Instead, various transformation products are formed that could give rise to adverse environmental effects.

With current processes, biological treatment results in good degradation of a number of substances, while others are not degraded at all. At present, there is no biological method that gives the same broad effect as ozonation or treatment with activated carbon. However, biological treatment is an important prerequisite for any additional treatment stages for removal of pharmaceuticals, as the effect of these is improved if the water contains less organic material and fewer particles. Biological treatment may also complement certain treatment techniques such as ozonation.

To date, ozonation and treatment with activated carbon are the techniques with the most practical applications on a large scale, which is linked with their effectiveness in terms of both treatment and costs. It remains to be seen what solutions will be devised in future. Increasing numbers of studies are being implemented using combined methods that may result in removal of more substances and more resource-effective solutions.

Biological treatment

Our wastewater treatment plants are not designed to separate persistent organic substances. This does not mean that all pharmaceuticals pass unaffected through the treatment processes. Chemical treatment is applied at most treatment plants, primarily in order to reduce phosphorus, but is not an alternative to far-reaching treatment to remove pharmaceuticals. However, biological treatment, primarily in the form of active sludge and biofilm systems, has proven to have an effect on certain pharmaceuticals. With better knowledge of the mechanisms for good biological treatment, many wastewater treatment plants could make the improvements that are possible within the existing plant elements and thereby be able to achieve a certain level of reduction in output levels without excessive consumption of resources.

Activated sludge and biofilm systems

Essentially, various substances can be either adsorbed to biomass or degraded. Adsorption and degradation vary from substance to substance, depending on the properties of the substances. Pharmaceuticals are not volatile, and hence evaporation to air is considered negligible.

In Sweden, biological treatment takes place at wastewater treatment plants in the form of both activated sludge systems and biofilm systems. The most common biological method involves treatment in various types of active sludge system. In these systems, it has been demonstrated that some pharmaceuticals, such as naproxen and ketoprofen, can be degraded and that degradation is more effective in systems with nitrogen treatment. Hormones such as oestradiol and ethinylestradiol are degraded to a relatively high degree and therefore frequently demonstrate high levels of reduction (Schlüsener & Bester, 2008).

Some wastewater treatment plants use biofilm systems for biological treatment. Results from experiments involving suspended carriers have been promising, and there have been higher degradation rates for some substances such as diclofenac, which does not appear to degrade to the same extent in active sludge systems (Falås et al., 2013).

Extensive studies have been implemented to evaluate the potential of various biological treatment systems. A long retention time, high sludge age, biofilm processes and the presence of nitrifying bacteria are some of the mechanisms that have resulted in better reduction in various contexts. However, a number of micropollutants – including several pharmaceuticals – are not affected to any appreciable extent by biological treatment processes, at least not as far as current technology and treatment processes are concerned (Falås et al., 2016).

From biological treatment to advanced treatment

Thus, biological treatment results in good reduction of some substances, while others remain unaffected. There is an opportunity to optimise the degradation of certain substances if the sludge age and retention time can be extended. Biological treatment in the form of MBR (membrane bioreactor) is one variant found to result in extended degradation for certain substances. The same appears to be true if biofilm processes can be applied. The MBR technique, which also permits far-reaching particle separation, has been studied by *SystemLäk* (Allard & Wahlberg, 2017). The same project has also studied biologically active filters (BAF), which is another example of extended degradation and optimisation of biological treatment. This technique is introduced in the section relating to granular activated carbon and can be found in the list of results.

There are obstacles to exploiting the potential of biological treatment, as with all forms of subsequent advanced treatment, in the form of diluted wastewater, sewer overflow discharges before the water reaches the wastewater treatment plant and bypassing of the biological treatment in the event of high flows. As the majority of wastewater treatment plants have some form of biological treatment, it is important for this to work as effectively as possible in order to create conditions for degradation of certain substances. Effective biological treatment is also an important prerequisite for advanced treatment with activated carbon or ozone, for example. Low levels of organic material in wastewater mean less of a need for ozone and more efficient utilisation of activated carbon. Both optimised treatment processes and moderate supply flows to the treatment plant are required in order to achieve low output levels of organic material.

Ozonation

Ozone has long been used as a treatment technique in the production of drinking water for example for disinfection. When upgrading wastewater treatment plants, ozonation is one of the techniques presented as a cost-effective alternative for oxidation of persistent organic contaminants. There is also experience of ozonation for filament control at wastewater treatment plants.

Degradation or conversion?

During ozonation of wastewater, oxidation does not lead to complete degradation of the organic substances in the water. There is normally fairly little changes in the level of organic carbon (often expressed as TOC – Total Organic Carbon) for water that has undergone ozonation. This suggests transformation (conversion) rather than complete mineralisation (degradation) of substances that are detected before but not after the process. Transformation products are formed on reaction. Some are stable, while others are degradable.

At the same time, wastewater contains substances other than pharmaceuticals and other micropollutants. Some of these may form the various by-products upon ozonation. Most studies indicate that ozonation reduces the toxicity of the water, but some researchers also argue in favour of a more complex view, with various conceivable ecotoxicological effects. There are various procedures described in the literature that can be used to provide an indication of whether ozonation increases or reduces the toxicity of the water, and if so to what extent. Various toxicity tests have also been performed in connection with the ozonation experiments in Knivsta, Linköping and Stockholm (see, for example, Baresel et al. (2017 a) for further information).

Some form of biological post-treatment is often recommended in the literature in order to minimise the risk of emissions of harmful by-products or transformation products.

Process design

Only a small proportion of the organic content in wastewater consists of problematic contaminants, but the oxidation process is not selective; the entire content of organic material (and other substances that can be oxidised) will affect the need for ozone. If the water contains less organic material, less ozone – and hence less energy – is needed. This indicates that an ozone plant should ideally be placed after effective biological treatment. Figure 3 shows a typical plant design.

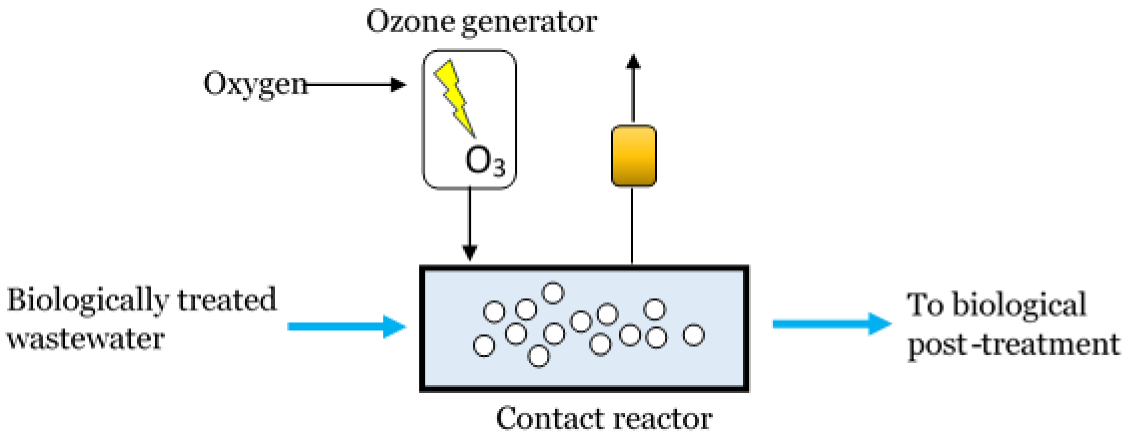

Figure 3. One of several possible process designs for ozonation of biologically treated wastewater. Illustration: Michael Cimbritz.

The ozonation stage in Knivsta (Project 5) was designed and laid out in a similar manner to the system shown in Figure 3, but after final chemical precipitation (after biological treatment) so as to be loaded with as little suspended and organic material as possible. Contact filters were placed after the ozonation stage to destroy ozone residues and provide an opportunity for establishment of a biofilm.

Ozone gas (O_3) is both explosive and unstable. Therefore, it is not easy to store or transport it. In practice, this means that ozone must be generated in situ. Oxygen (O_2), which is used to produce ozone, can either be produced from air adjacent to the ozone plant or be supplied in liquid form and stored in an oxygen tank. The retention time in the contact reactor is in the order of 10-25 minutes, which means that relatively little space is required in relation to many other elements at a wastewater treatment plant. During the subsequent polishing stage, a biofilm is expected to result in further degradation of the by-products and transformation products from ozonation. Post-treatment can also be arranged in other biological processes. At Nykvarnsverket in Linköping, post-treatment takes place in a biological treatment stage for nitrogen removal with suspended biofilm carriers (Baresel et al., 2016).

Ozonation is a flexible technique where the dosage can be changed easily depending on the treatment needs. However, optimised dosage requires advanced monitoring and control. Such technical processes are being developed and are a prerequisite for energy streamlining. Ozone generation is a relatively energy-intensive process, so it is important to optimise ozone dosing in relation to the desired effect.

Energy usage

To estimate energy usage, it is important to assess total energy usage for ozonation of wastewater. If liquid oxygen is produced outside the plant, this needs to be included in the calculation when comparing alternatives. Besides ozone production, cooling of the ozone generator and pumping require a significant amount of energy. This means that energy consumption may vary depending on the design of the plant and the structure of the existing infrastructure. As dosing is in proportion to the amount of organic material, upstream treatment will affect energy usage. During the full-scale treatment in Knivsta, energy consumption amounted to just under 0.1 kWh/m^3 of wastewater. Baresel et al. (2017 b) report the increase in electricity consumption at the wastewater treatment plant to be 0.1-0.2 kWh/m^3.

Key indicators

Key indicators for dimensioning have been produced based on the experiments performed within the various projects:

- Retention time in contact reactor: 10-25 minutes
- Specific ozone dosage: 0.4-0.9 g O_3/g DOC

There are always uncertain elements linked with key indicators. For example, there may be nitrite or other inorganic substances that consume further ozone. It may also be added that the actual retention times vary widely at different full-scale plants. Good reduction results have been achieved in both *SystemLäk* and *RESVAV* with comparatively low contact times.

Powdered activated carbon

Activated carbon permits a high level of separation of a broad spectrum of organic substances, including many pharmaceuticals. Separation takes place by means of adsorption to the carbon structure and is based on isolation rather than degradation. Therefore, a high specific area is important for effective separation. Wastewater can be treated by adding powdered activated carbon (PAC) or filtered through granular activated carbon (GAC) as a final stage. The latter alternative is described in detail in the next section.

Process design

PAC addition requires that powdered carbon can be stored at the wastewater treatment plant. The powder is mixed with water prior to addition in order to be able to control the addition and reduce dust. Stringent demands are made of steel quality for critical equipment elements, as handling and pumping of powdered activated carbon results in high levels of wear. Storage of PAC also requires EX classification due to the risk of explosion. Handling of PAC can be facilitated by using inert gas. Figure 4 shows how PAC can be added at a wastewater treatment plant.

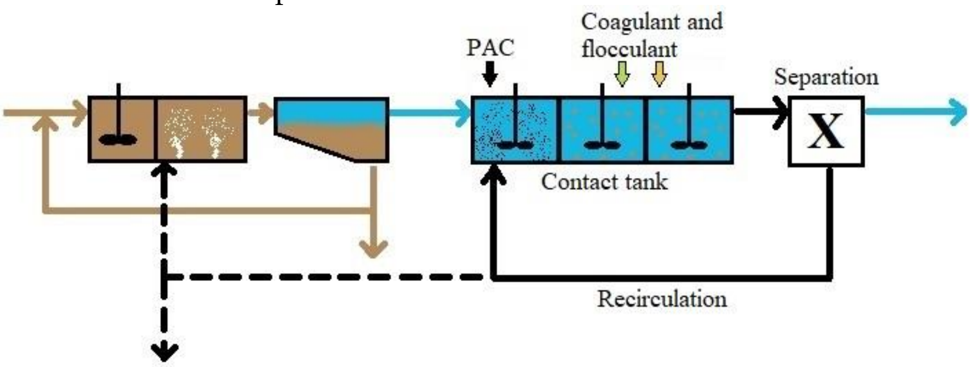

Figure 4. General solution for how PAC can be added to a contact reactor in a supplementary stage, where X is equivalent to a separation process for removal of PAC. (Högstrand & Ignell, 2018).

After addition, a reaction time in the order of 30 minutes is required before the carbon can be separated with adsorbed substances. The PAC sludge can be separated by means of precipitation and flocculation and sedimentation followed by sand filtration. Other methods can be used for separating carbon, such as membrane filtration. Production of activated carbon is an energy-intensive process, so efficient utilisation of capacity for adsorption of various substances is important. In practice, this means that the carbon is recirculated in the process in order to minimise the need to add more carbon. At the plants constructed in Switzerland and Germany, consumed carbon is returned to the biological treatment facility before then leaving the plant with the surplus sludge, which is often incinerated. The activated carbon should not be mixed with the sludge if the sludge is to be recycled to farmland.

There are other process designs, such as direct addition of PAC to the active sludge process. No separate reaction tanks are required for such a configuration. *SystemLäk* has studied the combination of membrane bioreactor (MBR) + PAC, which permits both far-reaching degradation and adsorption. *RESVAV* has successfully tested the option of adding PAC directly to a biofilm process (MBBR) on bench scale. As most full-scale plants integrate powdered activated carbon with biological treatment, experiences of the handling of sludge mostly comprising activated carbon are limited.

Energy usage

Most process solutions that involve adding powdered activated carbon involve a marginal increase in electricity consumption at the wastewater treatment plant itself. That said, it is important to remember that producing activated carbon is an energy-intensive process. Energy usage increases enormously if this is included. With energy consumption equivalent to 30 kWh/kg of PAC (Abegglen & Siegrist, 2012) and a dosage of 15 g/m^3, power equivalent to 0.45 kWh/m^3 is needed, that is to say 65 kWh/PE/year (if every person generates 400 l of wastewater per day).

Key indicators

PAC can be integrated in the treatment processes in several different ways. Required dosage depend on the desired separation and other factors, primarily the extent to which the activated carbon is recirculated in the process. A list compiled by Baresel et al. (2017) reports key indicators corresponding to:

- Dosage of 10-20 mg PAC/l
- Retention times equivalent to approx. 30 minutes

If the dosage is integrated with the biological treatment, no extra contact reactor is required as the retention time in the active sludge pond is sufficient for adsorption of pharmaceuticals to the activated carbon. The activated carbon is separated together with the sludge.

Granular activated carbon

One alternative to adding powdered activated carbon is filtration through a bed of granular activated carbon (GAC). Just as powdered activated carbon, low levels of organic material are desirable so that the carbon can be utilised as efficiently as possible. In other words, avoiding use of the adsorption capacity of the carbon for organic material that could have been separated in previous treatment stages. It is also important to ensure that the particle content is low so that the macrostructure of the filter is not blocked, thereby preventing efficient utilisation of the microstructure of the carbon. Other terms are also used in connection with carbon filtration. BAC, biologically activated carbon, is used in order to emphasise the fact that a biofilm is developed in the carbon filter that may assist with separation of various substances by means of degradation. The text below sometimes refers to the acronyms BAC (biologically activated carbon) or BAF (biologically active filtration).

Process design

Figure 5 shows a process design that has been evaluated with a great deal of success in the *FRAM* project. In this design, an existing sand filter is used after biological treatment as pre-filtration for the GAC filter.

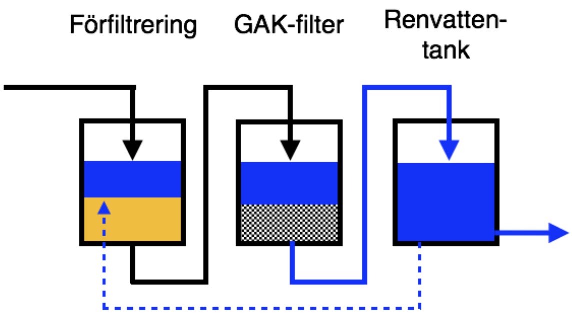

Figure 5. General sketch of pre-filtration with subsequent filtration through granular activated carbon in the FRAM project (Svahn & Björklund, 2018).

Operation of an activated carbon filter is essentially similar to operation of a sand filter for polishing wastewater. However, the density of the carbon is lower than the density of the sand, which affects backflushing. The degree of separation decreases over time, and after a certain time – frequently expressed as the number of filtered bed volumes – what is known as a breakthrough is eventually reached, which means that separation is impaired for one or more substances. The limit for what is regarded as a breakthrough varies and is ultimately dependent on the final objective of the treatment. In the *FRAM* project, breakthrough of small, negatively charged molecules such as sulfamethoxazole took place first, which was entirely in line with the studies previously performed on a laboratory scale as part of the project (Svahn & Björklund, 2015). Upon breakthrough, the carbon has to be replaced with new carbon or regenerated. During regeneration, the carbon is heated and the substances that were adsorbed are mineralised. Approximately ten per cent new activated carbon has to be added after regeneration in order to compensate for losses.

Energy usage

Process solutions involving filtration through a GAC filter result in a relatively small increase in energy consumption at the wastewater treatment plant, in a similar way to adding powdered activated carbon. Energy use is linked primarily with the flushing of the filter. However, manufacturing and regenerating activated carbon requires energy, precisely as is the case when manufacturing PAC.

Key indicators

GAC filters are designed according to approximately the same criteria as sand filters. Flushing of the filter is adapted after upstream processes and any pre-treatment and is generally performed considerably less frequently than for a sand filter. Key indicators for dimensioning have been produced based on the experiments performed within the various projects:

- Contact time in the filter: >10 minutes
- Filtration rate: 6-10 m/h
- Regeneration: >20,000 bed volumes

The number of bed volumes (often referred to as *EBCT, Empty Bed Contact Time*) varies from study to study, but this figure amounted to more than 20,000 bed volumes in the experiments performed within the *SystemLäk* and *FRAM* projects. Long intervals between carbon replacements are more economical and resource-efficient.

Results from the projects

There are currently no specific requirements to relate to as regards discharge of pharmaceuticals from wastewater treatment plants so how various techniques should be evaluated is not obvious. Levels of various substances in the receiving water are crucial in order to avoid effects on plant and animal life in lakes and waterways. However, reduction of various substances is evaluated in many studies of different techniques. Looking at reduction sometimes makes it easier to compare treatment techniques, but at the same time, there is no guarantee that high reduction will result in a sufficiently low level in the receiving water, or that the same reduction will be needed to protect all receiving bodies of water. It may also be more difficult to achieve a certain percentage reduction if the water is diluted from the outset, which is why a requirement for a certain level of reduction may be more difficult to meet than a level requirement for a treatment plant receiving a lot of infiltration and inflow water.

It may be worth noting that the legislation introduced in Switzerland is based on percentage reduction of a fairly small number of substances. However, the idea is to remove substances – even those not covered by the requirements – to a corresponding level, and to ensure that the target average percentage reduction is sufficiently high to have the desired effect in lakes and waterways. Simplification is necessary, as there are many different substances that occur in different concentrations and may give rise to different effects. It is not practicably possible or economically reasonable to monitor the reduction or level of each individual substance frequently. Assessing how any requirements are to be defined is a challenge, for a number of reasons. The need to reduce the supply of pharmaceuticals to a receiving water varies, depending on dilution and other local factors. Analysis of pharmaceuticals is relatively costly and probably requires some form of standardisation. Some of the substances to be removed also vary in incidence and over time (see the section entitled “Specific loads”), and this presents a challenge.

It is important to look at and discuss results against this background. Preferred levels or reduction levels are not fixed, at least not from a regulatory perspective. Therefore, different techniques have not necessarily been evaluated against a single target in the various projects. That said, the potential of the techniques themselves has been examined and evaluated. *Examples* of results from the various projects are described below, focusing on reduction of various micropollutants in accordance with the purpose of the call for projects. The *SystemLäk* project has tested a number of different techniques and combinations of techniques. An overall assessment of this project is provided in the final section, entitled *Choice of technique*.

Specific loads

When designing wastewater treatment plants, specific loads of nitrogen, phosphorus and organic material provide an important starting point. It may be interesting to study corresponding loads for various pharmaceuticals. A study of this kind has been performed as part of the RESVAV project, where the influx of various substances was studied over time for a number of wastewater treatment plants. Table 2 shows a list of various substances.

Table 2. Removal and loading of various pharmaceuticals at a number of Swedish wastewater treatment plants (Paxéus et al., 2016 and Paxéus, 2018).

Pharmaceutical product	Removal in WWTPs, %	To WWTPs mg/person, day	Loading trend
Atenolol	15-60	0.5	Declining
Metoprolol	Approx. 20	0.3	Major spread Declining
Propranolol	0-20	0.025	Declining
Sotalol		0.09	
Trimethoprim	0-50	0.02	Declining
Sulfamethoxazole	30-75	0.02-0.4	Declining
Citalopram	0-20	0.08	Declining
Tramadol		0.1-0.4	
Carbamazepine	0-20	0.1	Slightly declining
Ibuprofen	80-100	2.0	Stable
Diclofenac	0-25	0.15-0.4	

Various estimates – of different mass flows, for example – can be carried out based on this type of data. If there is knowledge of various treatment processes and recipient conditions, specific loads for different pharmaceuticals can be used in different contexts in which needs and the design of advanced treatment are discussed. One step in this direction has been taken with the LUSKA project (Pharmaceutical emissions from wastewater treatment plants 2017 in southern Sweden), where samples have been taken upstream of, downstream of and in effluent water from various wastewater treatment plants (Svahn & Björklund, 2017). The percentage reduction has been studied for a number of different pharmaceuticals and confirms the findings, with relatively low reduction for many substances.

Ozonation

Ozonation has been demonstrated and evaluated in a number of the projects and proven to give good results for a broad spectrum of organic micropollutants, not least pharmaceutical residues. A few examples are presented below.

Project 2. Removal of Pharmaceutical Residues using ozonation as Intermediate Process Step at Linköping WWTP, Sweden

Experiments carried out in Linköping tested ozonation with a view to creating a basis for design of the full-scale plant, which was then designed at Nykvarnsverket after the end of the project. However, the purpose of the experiments was not just to evaluate the effect of ozonation on different pharmaceuticals, but also to evaluate how ozonation could be integrated in the existing treatment process. Ozonation has been combined with biological treatment at the plant and placed between the active sludge process and the plant's biofilm process for nitrogen removal. It was possible to note in the experiments that ozonation did not disrupt the subsequent biofilm process and that biological post-treatment could be completed without designing an additional treatment stage. Figure 6 shows the pilot facility.

Figure 6. Pilot facility for ozonation at Nykvarnsverket wastewater treatment plant in Linköping.

One important element of the experiments was to show that different pharmaceuticals actually could be reduced, and at what dosage. Figure 7 shows how the remaining amounts of certain pharmaceuticals decline as the dosages increase.

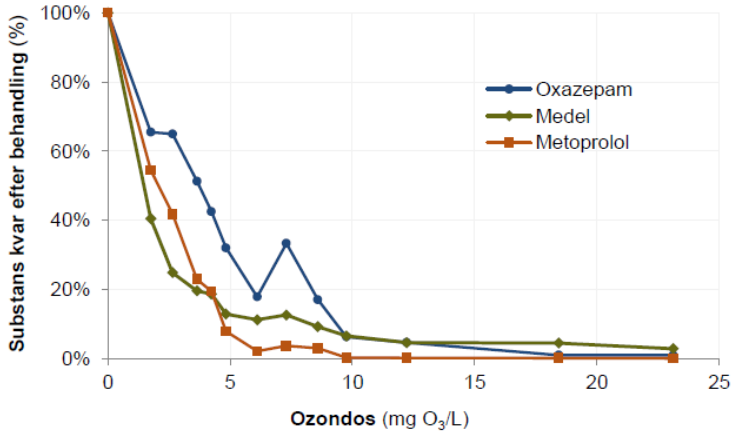

Figure 7. Remaining amount of pharmaceuticals as a function of ozone dosage. “Mean” constitutes a mean for the pharmaceuticals analysed (Sehlén et al., 2015).

Oxazepam is one of the substances requiring a relatively high ozone dose for reduction, while metoprolol is part of a group that is relatively simple to oxidise with ozone. However, at levels in the order of 5 mg O_3/l it was possible to remove almost 90% for most of the pharmaceuticals analysed. Based on this type of data, it was possible to estimate levels of various substances in the wastewater treatment plant’s receiving water (the Stångån river) and evaluate these against the concentrations that can be expected to give rise to a risk of adverse environmental effects.

Another important element of the experiments in Linköping involved various ecotoxicological experiments. None of these (oestrogen activity, green and red algae, Nitocra and Ames tests) indicated any adverse influence at the tested ozone dosages (up to 18 mg O_3/l). One parameter that is frequently referred to in connection with ozonation is the formation of the carcinogenic compound bromate, but high levels of this could not be detected at reasonable ozone dosages (<10 mg O_3/l). Overall, the various experiments helped to provide an overall view in which ozonation was considered an appropriate solution for Nykvarnsverket in Linköping.

Project 5. Evaluation of advanced full-scale treatment

A full-scale ozonation plant was established at the Knivsta wastewater treatment plant, south of Uppsala. This treatment plant is designed for 12,000 PE and for a design flow of 300 m^3/h. The size of the ozone plant and the fact that it was constructed for ozonation of the entire wastewater flow make it particularly interesting from both a national and an international perspective. The fact that the wastewater treatment plant’s entire flow was processed with ozone created a unique opportunity to study the receiving body of water, the

Knivstaån River, *before*, *during* and *after* ozonation was operational. This was done by taking samples at several different points upstream and downstream of the wastewater treatment plant, the last 8 km downstream.

Pharmaceuticals were analysed in both water and biota. An extensive inventory of invertebrates, a microbial evaluation and an exposure experiment with fish were carried out in addition to this. Figure 8 shows how the mass flow for the pharmaceuticals analysed in water changed with and without ozonation.

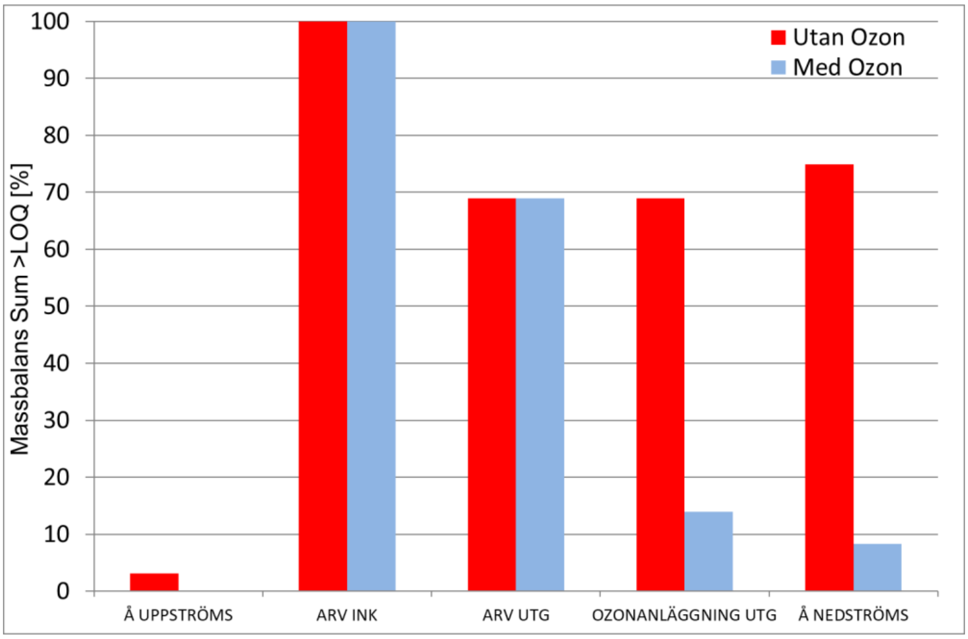

Figure 8. The diagram shows how the total amount of studied pharmaceuticals changes through the wastewater treatment plant and in the receiving body of water, with and without ozonation. (LOQ stands for Limit of Quantification, and so the mass balance relates to the total amount of the substances that could be quantified at various points.)

Reduction of pharmaceutical residues at the existing treatment plant was equivalent to approx. 30%, and with ozonation, the total reduction was in excess of 90%.

Reduction of diclofenac, a focus substance in the water management, was in excess of 99%. A high level of reduction could be established for the vast majority of substances. However, a number of substances proved to be more difficult to remove such as irbesartan (antihypertensive), tramadol (painkiller) and, in particular, fluconazole (antifungal). The effect on fish of pharmaceutical treatment with ozone was studied over a period of three weeks by placing zebra fish in aquaria at the ozonation plant in Knivsta. The experiments showed through an increase in ovulation that the zebra fish were affected by ozonation, but there was also a slight increase in induction of vitellogenin in males, which indicates that oestrogen-like substances may have formed in connection with ozonation (Pohl et al., 2018). Previous Swedish studies with rainbow trout have indicated reduced induction of vitellogenin after ozonation. The various outcomes indicate that more exposure studies should be carried out and any necessary adjustments to the treatment process should be made, in parallel with implementation of ozonation on a broad front for treatment to remove pharmaceutical residues and other micropollutants.

Ozonation also proved to lead to significant reductions of pharmaceutical residues in biota in the Knivstaån River, where various mayflies and other species were studied. In connection with these experiments, an extensive inventory of invertebrates has also been compiled which includes no fewer than 140,000 invertebrates. Significant changes in the number of individuals have been identified for mayflies and caddis flies, leeches and molluscs. The number of mayflies has increased, while the number of leeches and molluscs has decreased since ozonation began. Ozonation also reduced the presence of viruses (Wang et al., 2018).

Project 6. Treatment of persistent contaminants in wastewater (RESVAV)

Ozonation was tested in a number of different pilot experiments within the scope of the *RESVAV* project. A study was performed in order to compare ozonation at an actived sludge plant under high load and low load, given the fact that there are actived sludge systems in Sweden both with and without nitrogen removal

In another study, a pilot facility was relocated for short-term experiments at ten different wastewater treatment plants in southern Sweden. The purpose of this study was to compare different wastewater treatment plants and study how the ozone requirement varies in order to achieve the same level or reduction of a given substance with a view to creating dimensioning data for practical applications. Table 3 shows a selection of treatment results from this study.

Table 3. Reduction of different pharmaceuticals as a function of ozone dosage.
Red: <50%, yellow: 50-79%, light blue: 80-89%, dark blue: >90%. For a full list including additional substances, please see Ekblad et al. (2015).

Substance	Group	5 g O_3/m^3	5 g O_3/m^3	7 g O_3/m^3	10 g O_3/m^3
Diclofenac	*Anti-inflammatory*	Dark blue	Dark blue	Dark blue	Dark blue
Ibuprofen	*Anti-inflammatory*	Red	Yellow	Yellow	Yellow
Atenolol	*Beta blocker*	Red	Yellow	Light blue	Dark blue
Metoprolol	*Beta blocker*	Yellow	Yellow	Light blue	Dark blue
Ciprofloxacin	*Antibiotic*	Red	Red	Yellow	Dark blue
Clarithromycin	*Antibiotic*	Red	Red	Red	Red
Sulfamethoxazole	*Antibiotic*	Light blue	Light blue	Dark blue	Dark blue
Carbamazepine	*Tranquilliser*	Dark blue	Dark blue	Dark blue	Dark blue
Venlafaxine	*Antidepressant*	Yellow	Yellow	Light blue	Dark blue
Tramadol	*Painkiller*	Yellow	Yellow	Light blue	Light blue
Iohexol	*Contrast agent*	Red	Red	Red	Yellow
Carbendazim	*Biocide*	Yellow	Yellow	Dark blue	Dark blue
Diuron	*Biocide*	Yellow	Yellow	Yellow	Light blue

The table shows that certain substances, such as diclofenac and carbamazepine, can be reduced even at fairly low ozone dosages. Higher dosages are required for other substances, while certain substances do not seem to be affected to any appreciable extent. This is applicable to various types of contrast medium, for example, but even the antibiotic clarithromycin, from the EU watch list, undergoes poor oxidation even at high ozone dosages. It does not seem to be possible to achieve high reduction for some substances, even at high dosages. One such example is ibuprofen. However, it should be noted that it is often possible to demonstrate very high reduction for ibuprofen even in an activated sludge system with nitrogen removal. The remaining level, which can be reduced in subsequent ozone treatment, is very low in this case. The example of ibuprofen shows that it is important to study levels as well and understand the big picture, i.e. which processes are upstream and how these affect different substances. Reduction across the entire wastewater treatment plant may be very high, almost 100%, even if reduction in the advanced treatment – ozonation in this case – is significantly lower.

Activated carbon

Activated carbon, in both pulverised and granular form, has been evaluated in a number of the projects. Dosages of powdered activated carbon have been tested successfully within the scope of *SystemLäk* (Project 4) and *RESVAV* (Project 6), involving an MBR (membrane bioreactor) in *SystemLäk* and an MBBR (Moving Bed Biofilm Reactor) in *RESVAV*. Figure 9 shows a picture of the experiments where PAC was added to a process with suspended biofilm carriers.

Figure 9. Carrier material from pilot experiments at the Sjölunda wastewater treatment plant, carried out as part of the RESVAV project. The carrier on the left comes from a reactor to which powdered activated carbon has been added, while the carrier on the right comes from a reference reactor with no dosage.

However, adding powdered activated carbon assumes a change in sludge handling, which is not the case with filtration through granular activated carbon, which was tested and evaluated within the scope of both *SystemLäk* and *FRAM* (Project 3).

Project 3. Full-scale treatment of micropollutants – FRAM

The primary objective of the *FRAM* project was to evaluate the ability of granular activated carbon to remove different micropollutants on a full-scale level. While the filtration plant was being constructed, the commercial carbon types that most effectively adsorbed micropollutants were tested based on new knowledge of chemical interaction and not on a “trial and error” basis.

A number of commercial activated carbons are currently available on the market. Manufacturing procedures and starting materials vary. Besides the condensed carbon structure that constitutes the basic prerequisite for adsorption, activated carbon also contains functional groups that include nitrogen, oxygen and hydrogen. These will affect the binding of both humic substances and micropollutants. In chemical terms, pharmaceuticals are a very heterogeneous group of substances, and a new analysis instrument was developed in order to get an idea of how pharmaceuticals of different types bind to different organic matrix types and activated carbon. The results showed that the interactions of the pharmaceuticals with fixed matrices is very strongly linked with the physical-chemical properties of the molecules. Diclofenac is one example of a molecule with acidic properties. It can donate a proton and form a negative ion and is thus repelled by the negatively charged organic fraction in the treatment plant's organic sludge phase. The new analysis instrument could be used to evaluate the binding capability of nine commercial carbon varieties in a short period of time. The carbons showed major differences in ability to bind pharmaceuticals, and the two most suitable carbon types – 400 kg of each – were purchased for testing at the pilot facility. This new chemical tool, as well as the starting point of grouping pharmaceuticals according to chemical properties and not medical effect for water treatment, is new and provides valuable knowledge on which parameters control the binding of pharmaceuticals, which in turn will lead to more controlled and evidence-based development of a flexible and future-proof treatment method.

A new analysis method based on the Swedish Medical Products Agency's 22 environmental indicators and the EU watch list has been developed as part of the project. Organic trace analysis of micropollutants is costly but necessary to be able to assess whether the treatment technique works. Over the years, researchers at various laboratories have developed a very large number of methods for analysis of micropollutants, and what is known as a "multi-method" may accommodate more than 100 pharmaceuticals. This results in more analysis data on the one hand, but also in greater complexity, which may result in greater measurement uncertainty and increased costs in respect of analysis work. Not measuring the same substances in the different methods may also impede the comparability of different analyses.

This project produced a limited but carefully selected list of approx. 50 substances to carry on working with. The selected substances represent a broad spectrum of physical-chemical properties such as anions, cations and zwitterions, and varying hydrophobicity, thereby ensuring that as many existing and future micropollutants in the "chemical cocktail" as possible are covered.

After testing of activated carbon and designing the plant, the plant was initially sited at the wastewater treatment facility in Osby near to Lake Osbysjön, and subsequently at Kristianstad WWTP at Lake Hammarsjön. Both treatment plants are indirectly linked, via their lakes, to the Helge å River, which empties into Hanöbukten. Figure 10 shows the plant.

Figure 10. Picture of the plant with filtration of treated wastewater at the Osby treatment plant (Skåne), first through sand and then through granular activated carbon (GAC). Designer Måns Hansson, who at the time was chief engineer at Malmberg Water AB in Yngsjö, is pictured on the left, while the man on the right is Ola Svahn, a researcher at Kristianstad University. Photo: Erland Björklund.

The sand filter should act as protection for the activated carbon phase, with the hope of extending the service life of the carbon.

Initial chemical analysis (on 29 May 2015) indicated a very high separation level for all substances examined according to Table 4, which clarifies the generally good adsorption capability of activated carbon for a broad spectrum of substances. Treated wastewater is rich in dissolved organic carbon (DOC), which will reach the carbon filter and, together with the micropollutants, will help to saturate the carbon filter.

Table 4. Level of reduction of different substances over the lifetime of the project.

Pharmaceutical product	Date		
	29/05/2015	**22/11/2017**	**27/01/2018**
Atenolol	100	93	96
Ciprofloxacin	100	76	68
Citalopram	100	99	98
Clarithromycin	99	95	91
Diclofenac	99	88	86
Erithromycin	100	89	86
Estrone	97	100	97
Fluconazole	100	61	53
Furosemide	86	89	91
Imidacloprid	100	97	68
Carbamazepine	100	84	82
Losartan	100	84	90
Metoprolol	100	96	96
Naproxen	100	92	93
Oxazepam	99	84	79
Propranolol	100	100	100
Sertraline	96	94	98
Sulfamethoxazole	100	26	8
Tramadol	100	99	89
Trimethoprim	99	98	98
Venlafaxine	100	85	82
Zolpidem	99	100	93

Very high levels of separation were achieved for the studied substances during the experiments. An example showing diclofenac is presented in Figure 11 below. Only traces of diclofenac could be detected over the two operating periods in Osby and up to the end of June in Kristianstad (21 June 2017), with a treatment level of >99%. A decision was made in mid-June to increase the flow, at which time an elevated concentration of diclofenac was measured after the carbon filter. To determine whether the breakthrough was due to filter saturation or an insufficient retention time in the filter, the flow was reduced again, resulting in reduced diclofenac levels. This indicated that the filter was not yet saturated.

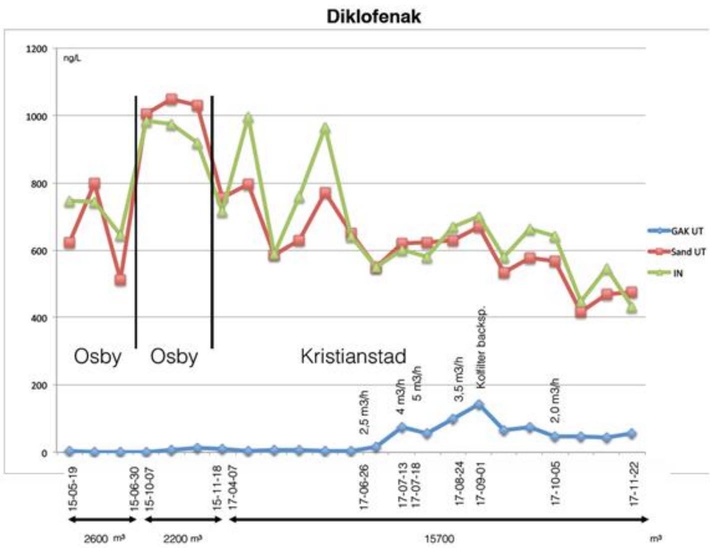

Figure 11. Overview of analysed concentrations of diclofenac. The green line (IN) shows the concentrations in the water that had already been treated at the Osby treatment plant and Kristianstads Centrala WWTP and that was passed into the sand filter. The red line (Sand OUT) shows the concentrations after the wastewater had passed the sand filter. The blue line (GAC OUT) shows effluent concentrations after the granular activated carbon.

The filter was evaluated regularly throughout the autumn of 2017, and 20,500 m3 of water had been treated by the end of November (22 November 2017). The antibiotics sulfamethoxazole and fluconazole clearly indicated breakthrough (Table 5), but the reduction was still good to very good for other substances. The pilot facility therefore continued to be operated until the end of January 2018. 23,000 m3 (equivalent to 23,000 bed volumes) had passed by this time, which is the volume used to place a new price tag on the cost of large-scale filtration with granular activated carbon (GAC).

Project 4. Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances – SystemLäk

A number of different techniques and combinations of techniques have been evaluated within the *SystemLäk* project (for more information, see the section entitled *Choice of technique*), including filtration through activated carbon. Figure 12 shows the reduction of a few selected pharmaceuticals as a function of filtration time from experiments carried out at Hammarby Sjöstadsverket in Stockholm.

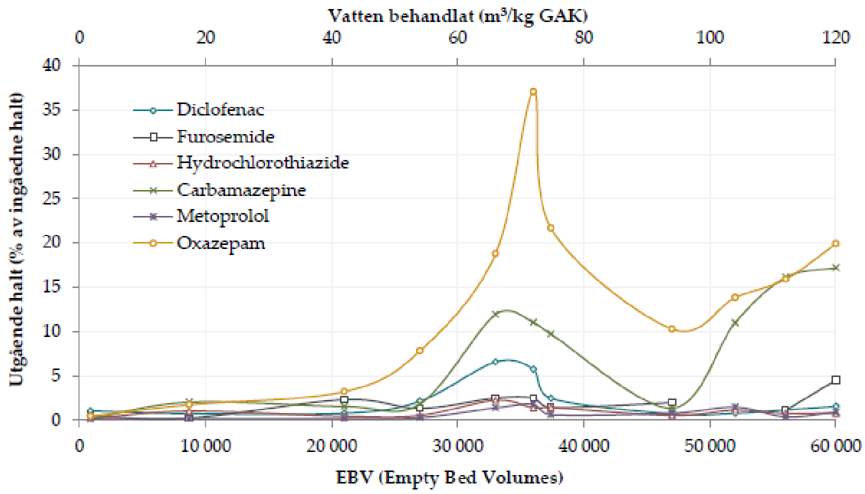

Figure 12. Reduction level of different substances (in %) after filtration through granular activated carbon. The reduction level is specified as a function of the number of bed volumes, i.e. the amount of water filtered. (Baresel et al. 2017 a)

The figure shows that reduction of the various pharmaceuticals can be achieved directly after start up and maintained for at least 25,000 bed volumes, which is equivalent to more than six months of operation at a reasonable filtration rate (approximately 5 m/h).

Influent and effluent levels of different substances were estimated and compared based on the long-term experiments carried out (two years). The difference was then compared with the volumes found in the carbon: see Table 5.

Table 5. Material balance for substances that could usually be quantified after BAF(GAC).

Pharmaceutical product	Total separated mg/kg GAC	Analysed in carbon mg/kg GAC	Found %
Citalopram	29.2	1.09	3.7
Diclofenac	67.9	0.13	0.2
Furosemide	49.2	0.57	1.2
Hydrochlorothiazide	143.4	3.97	2.8
Ibuprofen	8.1	0.01	0.1
Carbamazepine	41.2	13.1	31.8
Metoprolol	82.5	3.15	3.8
Oxazepam	54.3	7.03	12.9
Propranolol	6.7	0.87	12.0

Most of the various substances have not been found. This may be explained by the fact that they have been degraded, entirely or partly. If a GAC filter permits both adsorption and biological degradation, the capacity in a filter system can be increased, as well as bringing about a very good treatment effect, which means reduced costs. Alternative operating methods (connection of filters in series) have also been proposed based on results arrived at as part of the *SystemLäk* project, and these would permit reduction of costs and environmental impact.

Choice of technique

None of the treatment techniques provides complete treatment to remove all micropollutants, and all techniques have their advantages and disadvantages. However, activated carbon and ozonation both provide high levels of production for the vast majority of pharmaceuticals and many other organic micropollutants.

Project 4. Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances – SystemLäk

As part of the *SystemLäk* project, one of the objectives was to perform a comprehensive assessment of various system solutions and submit recommendations for implementation at different types of wastewater treatment plants. A number of aspects that are sometimes overlooked in the debate on pharmaceutical residues have been emphasised in particular within the project; for example, the fact that a number of both organic and non-organic contaminants are included in the term “micropollutants”. In other words, this does not just relate to pharmaceuticals. Removing micropollutants from wastewater may in many cases mean that several micropollutants end up in the sludge, which may require alternative sludge handling. Ongoing technical development should also be brought up in discussions on any future treatment requirements.

Which technical solution should be selected in the long run is a complex issue and requires consideration of many different aspects, as well as an understanding of the conditions prevailing at the wastewater treatment plant in question. One important result from this work is the discovery that effective treatment to remove pharmaceutical residues and other micropollutants is achieved by combining different techniques. Ozonation, for example, requires a polishing stage in order to eliminate any toxicity, but this post-treatment can also be achieved by means of integration in an existing process. Biofilters with GAC (referred to below as BAF(GAC)), with a combination of adsorption and biological degradation, are considered to provide high capacity and good reduction and are proposed as a solution after a membrane bioreactor. For wastewater treatment plants with far-reaching nitrogen, phosphorus and BOD treatment and low particulate levels in outgoing water, a combination of ozonation and BAF(GAC) is highlighted as a good alternative. Table 6 shows the treatment effect for various substances based on different treatment techniques and combinations of techniques.

From the project, the importance of effective primary treatment is also emphasised, with a view to reduce phosphorus, nitrogen and oxygen-consuming substances, which in practice means that this should be assured before introducing advanced treatment.

Table 6. Reductions for various substances, using different techniques (Baresel et al., 2017). UF – Ultrafiltration, GAC – Granular activated carbon, PAC – Powdered activated carbon, BAF – Biological aerated filter, O_3 – Ozonation, BAF(GAC) relates to biological filtration using GAC as the filter material.

	Treatment technique/combination				
Prioritised micropollutants and effects	**O_3 [1]**	**BAF(GAC)**	**PAC-UF**	**O_3-BAF(GAC)**	**UF-BAF(GAC)**
Azithromycin *(antibiotic)*					
Ciprofloxacin *(antibiotic)*					
Clarithromycin *(antibiotic)*		#	#	#	#
Diclofenac *(anti-inflammatory)*					
E2 (17β-oestradiol) *(hormone)*				#	
EE2 (17α-ethinylestradiol) *(synthetic hormone)*				#	
Erythromycin *(antibiotic)*		#	#	#	#
Ibuprofen *(anti-inflammatory and painkiller)*					
Carbamazepine *(antidepressant)*					
Levonorgestrel *(synthetic hormone)*		#	#	#	#
Metoprolol *(beta blocker, antihypertensive)*					
Oxazepam *(anxiolytic and tranquilliser)*					
Propranolol *(beta blocker, antihypertensive)*					
Sertraline *(antidepressant)*					
Sulfamethoxazole *(antibiotic)*					
Trimethroprim *(antibiotic)*			#		
Risk of infection *(bacteria, pathogens)*					
Antibiotic resistance (ARB)					
Oestrogenic effects (YES) *(effect of hormones)*			#		#
Bisphenol A *(plastic chemical, endocrine disruptor)*					
Cybutryne/Irgarol *(herbicide)*		#	#	#	#
Dioxins and PCBs *(e.g. in coolants)*		#	#	#	#
Endotoxins *(toxic bioaerosols)*		#	#	#	#
Phthalates (e.g. DEHP) *(plasticisers in plastic products)*					
Flame retardants (e.g. HBCD)					
Chloroalkanes (C10 to C13) *(lubricants)*		#	#	#	#
Linear alkylsulphonates (LAS) (C10 to C13)		#	#	#	#
Nonylphenol *(e.g. additive in cleaning products)*					
Octylphenol *(e.g. additive in cleaning products)*					
PFAS (inc. PFOS) *(tenside)*					
Sucralose *(sweetener)*					
Terbutryn *(herbicide)*		#	#	#	#
Tributyltin (TBT) *(biocide)*		#	#	#	#
Trichlorobenzene *(solvent and insecticide)*		#	#	#	#
Triclosan *(antiseptic)*					
Heavy metals[2] (lower priority)		#	#	#	#
Microplastics 1 µm - 5 mm (lower priority)					
Phosphorus					
Nitrogen					
Organic material COD/BOD	*				

Costs and resource consumption

Costs and environmental impact when introducing advanced treatment are important pieces of the puzzle which will allow decisions to be made on advanced treatment. Costs have been estimated within a number of the projects. Life cycle assessments have also been performed within the scope of the *SystemLäk* project.

Resource usage and environmental impact

To be able to describe resource usage and emissions to the environment, IVL has performed a life cycle assessment within the *SystemLäk* project according to ISO14044:2206 (with modelling in Gabi 7.2 software). Production of electricity and chemicals needed for treatment at the wastewater treatment plant is taken into account with this methodology in the evaluation of various techniques. The analysis has been performed based on various environmental impact indicators: climate impact, acidification potential, eutrophication potential and depletion of non-renewable energy and material resources. Toxic effects are not included, preferably due to restrictions and uncertainties in assessment of toxicity potential.

The most important conclusions from the analysis are that:

- the larger the plant, the lesser the environmental impact per m^3 wastewater treated.
- combination solutions result in greater environmental impact than individual techniques, but at the same time they provide more benefit to the environment as more micropollutants can be removed.
- manufacturing activated carbon has a relatively major environmental impact, but development towards biochar could potentially reduce this environmental impact significantly.
- ozonation has a low environmental impact in relation to other solutions, but in this case, recommended post-treatment methods such as ecotoxological effects of degradation products have not been included in the assessment.

Although ozonation requires more energy at the wastewater treatment plant than filtration through activated carbon, the environmental impact is deemed to be lower depending on the CO_2 emissions occurring during the manufacture of activated carbon. Although the activated carbon is regenerated, there is relatively high environmental impact as approx. 10% new carbon needs to be added for every regeneration. However, the risk of the occurrence of toxic transformation products during ozonation should be considered when comparing ozonation and treatment with activated carbon. This risk can be reduced if there is a subsequent biological stage.

Costs

Different data is available concerning the cost of introducing pharmaceutical treatment at wastewater treatment plants. The differences may be due to factors such as which treatment level is aimed for, and for which substances, the size of the flow peaks for which the plant is designed, and which pre-treatment or other circumstances are required. Different costs have been reported even within the project funded by the Swedish Agency for Marine and Water Management.

The *RESVAV* project's costs are estimated costs for a typical design plant. This includes capital costs and operating costs, with the exception of payroll and maintenance costs. VAT is not included.

The *SystemLäk* project has defined conditions for every technique for which cost estimates were compiled. This includes process-specific key indicators such as contact time, dosages, type of activated carbon, etc. Based on this dimensioning data, a number of Swedish and foreign suppliers have then submitted specific tenders for installation and operation of a number of typical treatment plants of varying size. Cost estimates from suppliers that failed to use the defined dimensioning data have been omitted from the report.

The actual costs have been specified for the plants in Linköping, Knivsta and Kristianstad. However, note that the costs for the Kristianstad plant are operating costs only. Please see each project for more details. The costs, together with the criteria for the cost estimates, have been specified in Table 7 (ozonation) and Table 8 (GAC).

Table 7. Some key indicators for treatment plants with ozone treatment. Relates to conditions for a plant for 100,000 population equivalents (PE) unless otherwise specified.

	SystemLäk	RESVAV	Tekniska Verken, Linköping	Knivsta
Cost, SEK/m^3	0.19 - 0.20	0.3 - 0.4	0.25 -0.3***	0.7 ** (0.9*)
Cost, SEK/PE/year	28 - 30	41 - 54	17 - 20	74
Average flow to WWTP, l/PE/d	410	370	185	290
Max. flow to ozone plant, l/PE/d	410	410	306	1120
Retention time in contact tank average/max/ flow, minutes	>10	33/30	20/12	30/11
Contact tankvolume, l/PE	-	8.5	2.6	8.3
Ozone dosage, g O_3/m^3	5	7	4 – 8	7 (3 - 9)
Ozone consumption, g O_3/PE/year	-	850	447	636
Electricity consumption for plant, kWh/PE/year	15 / 45	17	10	8.1
Subsequent biological stage included in cost	None	None	None	Contact filter
Adaptations to existing WWTP included in cost	None	None	Clearance prior to construction	Pumping
Production of O_2 included in specified electricity consumption	Yes – the higher figure	Yes	No	No
Production of ozone from O_2	Yes	Yes	Yes	Yes

* For the actual load, which was 12,000 PE. ** Scaled up to 100,000 PE. *** 235,000 PE.

The treatment level for the actual ozone treatment stage is stated as being from 80% to more than 90% for the various pharmaceutical product packages assessed or analysed in the studies. The treatment level over the entire treatment plant may be higher when the treatment in other treatment stages is included or lower because not all water passes through the treatment stage for pharmaceutical treatment at high flows. The higher cost per cubic metre at the plant in Knivsta is partly explained by the fact that ozone treatment is dimensioned for a very high flow so as to be able to process all water throughout the entire year. It is also explained that the costs for Knivsta include a subsequent contact filter and pumping from the existing plant. Differences in flow variations and other criteria mean that different contact pond sizes per person are expected in the various projects. The specified ozone consumption for all projects assumes that the wastewater is biologically treated initially in a nitrifying plant (to < 10 mg DOC/l) in order to reduce the amount of dissolved organic carbon consuming ozone. The *SystemLäk* project points out in this regard that any other ozone-consuming substances, such as iron or nitrite, must be taken into account.

Table 8. Some key indicators for treatment plants using granular activated carbon. Relates to conditions for 100,000 population equivalents (PE) unless otherwise specified.

	SystemLäk	RESVAV	Knivsta	Kristianstad
Cost, SEK/m³	0.35 – 0.60	0.8 – 1.1	1.1*	(0.17**)
Cost, SEK/PE/year	52 – 90	108 - 148	-	(12)**
Average flow, l/PE/d	410	370	-	194
Max. flow through filter, l/PE/d	410	410	400	486
Retention time in filter (EBCT) at average/max., minutes	x/15	23/20	17/12	30/12
Surface load on filter at average/max. flow, m/h	3.5/4.8	2.6/2.8	3.4/4.7	2/5
Volume in filter, l/PE		6	2 - 4	4.0
Time between backwashes, hours	48 – 2000	48	35	***
Proportion of processed flow for backwashing, %	-	5	6	***
Proportion of carbon regenerated, %/year	-	100	106	100
Electricity consumption for plant, kWh/PE/year	<1.5	3		0
Included for GAC, regeneration	Yes	Yes		Yes
... incineration			Yes	

* Pilot facility for 8 PE, scaled up to 100,000 PE. ** Operating cost for pilot facility for 250 PE (not capital costs).*** The filter is not backwashed.

For treatment with granular activated carbon, the conditions vary, but the projects estimate the reduction level in the actual filter to be 90–95%, and it is assumed that the water is biologically treated effectively before the filter. The assumed retention time in the filter, as well as the interval between backwashes, varies slightly between the projects. This, together with different assumptions in respect of flow distribution, means that the filter volume needed per PE, and hence the cost, varies. It is assumed that the filter washwater is recirculated to the treatment plant for the plants that wash their filters, but no costs for this have been included. It is calculated that all carbon will be regenerated in approximately one year and that 10% new carbon has to be added during regeneration. Electricity consumption for GAC is low, particularly if there is no need to pump water to or from the filter, as in the case of Kristianstad.

Cost estimates compiled in Germany, the Netherlands and Switzerland indicate slightly higher costs per m^3. However, it is important to remember that the prerequisites are different (see Cimbritz et al., 2016 for more information).

Conclusions

The work from the various projects shows that techniques are available that can be applied at municipal wastewater treatment plants in order to remove micropollutants of different types, including pharmaceutical residues. Where measures should be implemented depend on various factors, but through the work completed both knowledge and operating experience of various technical solutions are now available. The solutions that have been evaluated are mainly based on ozonation or activated carbon and various combination solutions, where it has proven to be very important to integrate the new treatment techniques with existing treatment processes in an expedient manner. The solutions have been tested and evaluated in close cooperation with staff at wastewater treatment plants all over Sweden. This is a prerequisite to be able to evaluate the techniques in a credible manner.

The different projects have not only tested new technology but have also been able to describe how various technical solutions can be adapted to different Swedish sewage treatment plants. At the same time, studies have been completed which pave the way for new research and development projects where the parties working as part of the current call for projects can assist with and lead development of the wastewater treatment of the future. This relates to factors such as ecotoxicological effects of ozonation, development and understanding of applications using activated carbon and development of analysis techniques.

With the introduction of new technology for treatment to remove substances that occur in very low concentrations in a difficult matrix (wastewater), we face a number of challenges in terms of analysis technology. Based on the analysis work carried out by the various projects and with the intercalibration study completed, important steps have been taken towards greater understanding of the problems surrounding the analysis of micropollutants. It is very important for this work to continue and for more steps to be taken toward experience exchange and standardisation.

Various solutions have been demonstrated in both pilot scale and full scale, by virtue of the work completed. The project in Linköping demonstrated how to make the transition from pilot scale to full scale, and the project in Knivsta showed how a full-scale ozonation solution can be applied and what effects this will have on the recipient. The project in Lund evaluated ozonation in pilot experiments at various different types of wastewater treatment plant and the first steps towards an integrated process with PAC and MBBR were taken. IVL has tested a number of different techniques and used life cycle assessment to show how these can be applied and combined in order to create resource-effective system solutions. The project in Kristianstad demonstrated the potential for application of activated carbon filters in parallel with development of analysis technology in order to enhance understanding of what happens in a carbon filter and how activated carbon should be selected. JTI (now known as RISE) has shown that not all solutions necessarily need to be based on new technology, and that system changes may be justified. The government initiative and work on the various projects have created a very strong foundation on which to further build for the potential introduction of advanced treatment at Swedish wastewater treatment plants.

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Pohl, J, Björlenius, B, Brodin, T, Carlsson, G, Fick, J, Larsson, D, Norrgren, L & Örn, S. (2018). Effects of Ozonated Sewage Effluent on Reproduction and Behavioral Endpoints in Zebrafish (Danio rerio), *Aquatic Toxicology,* January *2018.*

Schlüsener, M., Bester, K. (2008). Behavior of Steroid Hormones and Conjugates During Wastewater Treatment – A Comparison of Three Sewage Treatment Plants, *Clean Soil Air Water,* 2008, 36 (1), s. 25 – 33.

Sehlén, R., Malmborg, J., Baresel, C., Ek, M., Magnér, J., Allard, A-S., Yang, J. (2015). *Pilotanläggning för ozonoxidation av läkemedelsrester i avloppsvatten.* IVL Swedish Environmental Research Institute, Report B2218.

Svahn, O. & Björklund, E. (2015). Describing sorption of pharmaceuticals to lake and river sediments, and sewage sludge from UNESCO Biosphere Reserve Kristianstads Vattenrike by chromatographic asymmetry factors and recovery measurements, *Journal of Chromatography A*, 1415 (2015) 73–82.

Wang, H., Sikora, P., Rutgersson, C., Lindh, M., Brodin, T., Björlenius, B., Larsson, D.G.J., Norder, H. (2018). Differential removal of human pathogenic viruses from sewage by conventional and ozone treatments. *International Journal of Hygiene and Environmental Health, 221, 479-488.*

Appendix I – Further reading

A large number of different publications are available for anyone who is interested in continuing to read up on the subject in greater depth. A number of these are available online for example via IVL Swedish Environmental Research Institute or the websites of the various universities. Work is also in progress on a variety of publications at the time of writing. This is why titles are provided for planned scientific articles in some cases. It may be a good idea to get in touch with the relevant project manager in order to find out which articles have actually been published or are expected to be published in the near future.

The various projects have attracted a great deal of interest all over Sweden and have been referred to in a variety of ways in the press and on radio and TV. Please contact the relevant project manager if you are interested in finding out what has been written *about* the various projects. Below is a list of the publications produced by the various projects.

Pharmaceuticals in source-separated blackwater and faecal sludge – Treatment and risks (LäK)

Levén, L., Eveborn, D., Ljung, E., Gros Calvo, M., Dalahmeh, S., Jönsson, H., Ahrens, L., Wiberg, K., Lundin, G. (2016) *Läkemedel i källsorterat klosettvatten och latrin*. RKA 54. JTI – Institutet för jordbruks- och miljöteknik.

Planned publication

Gros M. et al. Occurrence and fate of pharmaceuticals in source separated sanitation systems: fecal sludge anaerobic digestion and blackwater liquid composting followed by ammonia treatment.

Removal of Pharmaceutical Residues Using Ozonation as Intermediate Process Step at Linköping WWTP, Sweden

Baresel, C., Ek, M., Malmborg, J., & Sehlen, R. (2016) Removal of pharmaceutical residues using ozonation as intermediate process step at Linkoping WWTP, Sweden, *Water Science and Technology*, 73, 8, pp. 2017-2024.

Sehlén, R., Malmborg, J., Baresel, C., Ek, M., Magnér, J., Allard, A-S., Yang, J. (2015), *Pilotanläggning för ozonoxidation av läkemedelsrester i avloppsvatten*. Report no. B 2218, February 2015. IVL Swedish Environmental Research Institute.

Full-scale treatment of micropollutants – FRAM

FRAM has appeared frequently in various media, such as Sveriges Radio P4, SVT, TV4, Kristianstadsbladet and a number of different journals. Please contact the project manager for a full list.

Björklund, E. & Svahn, O. (2017) Interkalibrerad läkemedelsanalys 2017 – ett samarbetsprojekt för ökad analyskvalité, Kristianstad University Report (2017); 62 pages.

Björklund, E. & Svahn, O. (2017) LUSKA Pharmaceutical emissions from Scanian wastewater 2017. Ett utvecklings- och samverkansprojekt på Högskolan Kristianstad i samarbete med Region Skåne och 6 skånska reningsverksaktörer, Kristianstad University Report (2017); 58 pages.

Björklund, E., Svahn, O., Bak, S., Bekoe, S., Hansen, M. (2016) Pharmaceutical Residues Affecting the UNESCO Biosphere Reserve Kristianstads Vattenrike Wetlands: Sources and Sinks. Archives of Environmental Contamination and Toxicology, 71 (2016) 423–436.

Svahn, O., Björklund, E., Ross, E. (2017) Analysis of Macrolides and Three Other Antibiotic Classes at Low ppt Levels in WWTP Effluent and Surface Waters by LC-MS/MS., Technology Brief, May (2017), ©2017 Waters Corporation.

Svahn, O. & Björklund, E. (2016) Increased electrospray ionization intensities and expanded chromatographic possibilities for emerging contaminants using mobile phases of different pH, Journal of Chromatography B, 1033 (2016) 1–10.

Svahn, O. (2016) Tillämpad miljöanalytisk kemi för monitorering och åtgärder av antibiotic- och läkemedelsrester i Vattenriket. Center for Environmental Studies and Climate Research, Lund University. ISBN 978-91-7623-774-8

Svahn, O. & Björklund, E. (2015) Describing sorption of pharmaceuticals to lake and river sediments, and sewage sludge from UNESCO Biosphere Reserve Kristianstads Vattenrike by chromatographic asymmetry factors and recovery measurements, Journal of Chromatography A, 1415 (2015) 73–82.

Slam i kolonnen ger resultat. Kemivärlden Biotech med Kemisk Tidskrift, No. 7 November 2015; 30-31.

Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances– SystemLäk

Allard, A.-S., Wahlberg, C. (2017) *Förekomst och reduktion av fokusämnen i fyra reningsverk*. Delrapport SystemLäk projekt. IVL Swedish Environmental Research Institute, Report B2279.

Baresel, C., Ek, M., Ejhed, H., Allard, A.-S., Magnér, J., Dahlgren, L., Westling, K., Wahlberg, C., Fortkamp, U., Søhr, S. (2017) *Handbok för rening av mikroföroreningar vid avloppsreningsverk – Planering och installation av reningstekniker för läkemedelsrester och andra mikroföroreningar*. Slutrapport SystemLäk projekt. IVL Swedish Environmental Research Institute, Report B2288.

Baresel, C., Ek, M., Harding, M., Magnér, J., Allard, A.-S., Karlsson, J. (2017) *Kompletterande tester för en resurseffektiv avancerad rening av avloppsvatten*. Delrapport SystemLäk projekt. IVL Swedish Environmental Research Institute, Report B2287.

Baresel, C., Cousins, A.P., Hörsing, M., Ek, M., Ejhed, H., Allard, A.S., Magnér, J., Westling, K., Wahlberg, C., Fortkamp, U., Söhr, S. (2015) *Läkemedelsrester och andra skadliga ämnen i avloppsreningsverk – koncentrationer, kvantifiering, beteende och reningsalternativ* – Swedish summary of Report B2226. IVL Swedish Environmental Research Institute, Report B2226-P, Stockholm.

Conference contributions

Baresel, C., Ek, M., Harding, M., Magnér, J. (2017) *Long-time experiment with biological active filter (BAF) for removal of pharmaceutical residues*. NORDIWA – Nordic Wastewater Conference, 10-12 October, Aarhus, Denmark.

Magnér, J., Örtlund, L., Fång, J., Baresel, C. (2017) *Fate of pharmaceuticals in sewage and sludge*. NORDIWA – Nordic Wastewater Conference, 10-12 October, Aarhus, Denmark.

Baresel, C., Ejhed, H., Westling, K., Fortkamp, U., Hörsing, M., Magnér, J., Allard, A.S., Wahlberg, C., Söhr, S. (2015) *Removal of Pharmaceutical Residues and Other Priority Contaminants in the Effluent of Sewage Treatment Plants*. WWTP-0101, 12th IWA LWWTPL, September 2015, Prague.

Theses

Alcala Borao, R. (2015) *Oxidation of pharmaceuticals by chlorine dioxide in wastewater effluent*. Thesis LWR – EX – 2015:16, KTH Royal Institute of Technology & IVL Swedish Environmental Research Institute, Stockholm.

Mparmpagianni, S. (2016) *Powdered activated carbon (PAC) addition in membrane bioreactor (MBR) for increased removal of organic pollutants in municipal wastewater*. Dept. of Environmental Science and Analytical Chemistry, Division of Analytical and Toxicological Chemistry, Stockholm University IVL Swedish Environmental Research Institute.

Other information

Brochure – Rening av mikroföroreningar vid ARV

Leaflet – Removal of micropollutants in WWTPs

Presentation – How-to Guide – Removal of micropollutants in WWTPs

Presentation – Vägledning – Rening av mikroföroreningar vid ARV

Evaluation of advanced full-scale treatment

Pohl, J, Björlenius, B, Brodin, T, Carlsson, G, Fick, J, Larsson, D, Norrgren, L, & Örn, S. (2018), Effects of Ozonated Sewage Effluent on Reproduction and Behavioral Endpoints in Zebrafish (Danio rerio), *Aquatic Toxicology,* January *2018.*

Wang, H., Sikora, P., Rutgersson, C., Lindh, M., Brodin, T., Björlenius, B., Larsson, D.G.J., Norder, H. (2018). Differential removal of human pathogenic viruses from sewage by conventional and ozone treatments. *International Journal of Hygiene and Environmental Health, 221, pp. 479-488.*

Planned publications

Another three publications are in the pipeline: one focusing on the process at the treatment plant during ozonation, one focusing on the ecological effects in recipients, and one looking at the microbial effect and occurrence of resistance genes. A summary report is also planned for the autumn.

Treatment of persistent contaminants in wastewater – RESVAV

Cimbritz, M., Tumlin, S., Hagman, M., Dimitrova, I., Hey, G., Mases, M., Åstrand, N., Jansen, J. la Cour (2016) *Rening från läkemedelsrester och andra mikroföroreningar – En kunskapssammanställning*. Svenskt Vatten Utveckling, Report 2016-04.

Ekblad, M., Cimbritz, M., Nilsson, F., Ernst, G., El-taliawy, H., Tumlin, S., Bester, K., Hagman, M., Mattsson, A., Blom, L., Stålhandske, L., Jansen, J. la Cour (2015) *Ozonering för nedbrytning av organiska mikroföroreningar – Pilottester i södra Sverige*. VA-teknik Södra, Report No. 04.

El-taliawy, H, Ekblad, M, Nilsson, F, Hagman, M, Paxeus, N, Jönsson, K, Cimbritz, M, la Cour Jansen, J, & Bester, K. (2017) Ozonation efficiency in removing organic micro pollutants from wastewater with respect to hydraulic loading rates and different wastewaters, *Chemical Engineering Journal*, 325, 310-321.

El-taliawy, H, Casas, M, & Bester, K. (2018) Removal of ozonation products of pharmaceuticals in laboratory Moving Bed Biofilm Reactors (MBBRs), *Journal of Hazardous Materials*, 347, 288-298.

Mases, M., Wärff, C., Öhrström, E. (2017) Kostnadsbedömning för införande av tekniker för avskiljning av svårnedbrytbara ämnen. Report, Assignment Number 1234211000, SWECO, Malmö.

Nilsson, F., Ekblad, M., Jansen, J. la Cour., Jönsson, K. (2017) Removal of pharmaceuticals with ozone at 10 Swedish wastewater treatment plants, Water, Practice & Technology, 12(4), 871-881.

Paxéus, N (2018) *Läkemedelsbelastningen till svenska avloppsreningsverk – en sammanställning*. VA-teknik Södra, Report No. 09.

Paxeus, N, Bester, K, & El-Taliawy, H. (2016) Temporal variations and trends in loads of commonly used pharmaceuticals to large wastewater treatment plants in Sweden, a case study (Ryaverket), *Water Science And Technology*, 73, 12, 3049-3056

Conference contributions

Ekblad, M., Cimbritz, M., Mattsson, A., Bester, K., El-taliawy, H., Jansen, J. la Cour (2015) *Considerations for removal of organic micropollutants at Swedish wastewater treatment plants.* Poster vid NORDIWA 2015-Nordic Wastewater Conference, 4-6 November 2015, Bergen, Norway.

Theses

Isgaard, P., Thörnqvist, E (2016) *Integration of powdered activated carbon in tertiary disc filtration of wastewater.* Department of Chemical Engineering, Lund University, Thesis, October 2016.

Högstrand, S., Ignell, M (2018) *Möjligheten att kombinera pulveriserat aktivt kol (PAK) och MBBR för avskiljning av organiska mikroföroreningar*. Department of Chemical Engineering, Lund University, Thesis, February 2018.

Planned publications

Ekblad, M., Falås, P., El-taliawy, H., Nilsson, F., Bester, K., Hagman, M., Cimbritz, M. Is dissolved COD a suitable design parameter for ozone oxidation of organic micropollutants in wastewater?

Cimbritz, M., Edefell, E., Thörnqvist, E., Ekblad, M., El-taliway, H., Ekenberg, M., Bester, K., Hagman, M., Falås, P. Combining PAC-adsorption and nitrification in an MBBR.

Treatment techniques for pharmaceuticals and micropollutants in wastewater

Description of eight projects that have received funding from the Marine and Aquatic Environment Grant for 2014-2017

The Swedish Agency for Marine and Water Management has been working on behalf of the government between 2014 and 2017 to fund various projects for the development of treatment techniques with a view to reducing emissions of pharmaceutical residues and other micropollutants from municipal wastewater treatment plants. This report briefly describes the background to the work and the results from the various projects in popular scientific form. Costs have been produced for various treatment techniques and summarised in the report, which also offers further reading for anyone interested. The results show that techniques are available at present that can be applied at municipal treatment plants to reduce micropollutants of different types, including pharmaceutical residues. The research in the various projects have created a strong foundation on which further building can take place for the potential introduction of advanced treatment at Swedish wastewater treatment plants.

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