Overview of advanced technologies in wastewater treatment for removal of pharmaceuticals and other micropollutants

Status in four coastal regions of the South Baltic Sea
Germany, Sweden, Poland and Lithuania

Project MORPHEUS 2017 - 2019
Deliverable 5.2

Lead Authors:
Luczkiewicz A., Fudala-Ksiazek S., Jankowska K., Szopinska M., Gdansk University of Technology, Poland

Co-authors:
Björklund, E., Svahn, O., Kristianstad University, Sweden (Lead partner)
Garnaga-Budrė G., Lithuanian EPA; Langas V., Klaipeda University, Lithuania
Tränckner J., Kaiser A., University of Rostock, Germany

Contact information:

aneta.luczkiewicz@pg.edu.pl
katarzyna.jankowska@pg.edu.pl
sylwia.fudala-ksiazek@pg.edu.pl
malgorzata.szopinska@pg.edu.pl

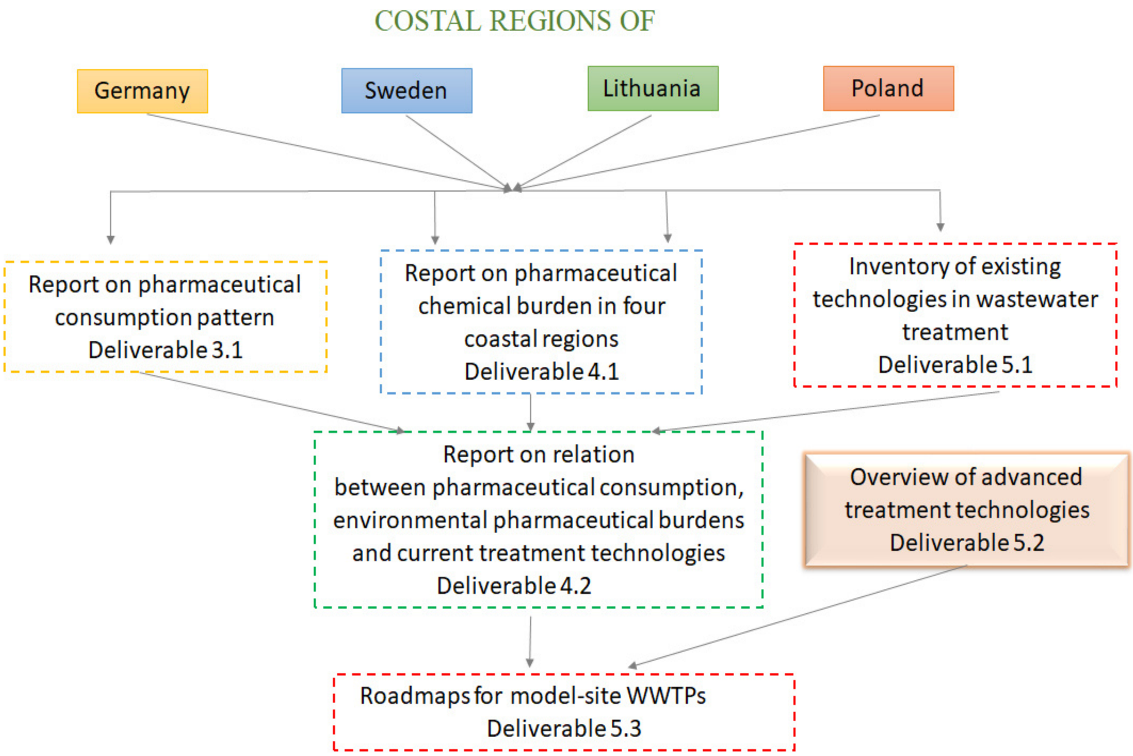
Cover photo

Left: Picture of the ozonation system at WWTP Bad Sassendorf in Lippeverband, Germany.
Right: Picture of the PAC system at WWTP Dülmen in Lippeverband, Germany.
Both pictures taken during the MORPHEUS study visit in April 2018. Photos: Franziska Stoll.

Key facts of the MORPHEUS project

MORPHEUS (Model Areas for Removal of Pharmaceutical Substances in the South Baltic) is a project financed by the European Union Interreg South Baltic Programme. The project duration is January 2017 – December 2019, with a total budget of EUR 1.6 million with a contribution from the European Regional Development Fund of EUR 1.3 million. The project has a total of 7 partners from four countries; Sweden, Germany, Poland and Lithuania: Kristianstad University (Lead Partner) – Sweden, EUCC – The Coastal Union Germany – Germany, University of Rostock – Germany, Gdansk Water Foundation – Poland, Gdansk University of Technology – Poland, Environmental Protection Agency – Lithuania and Klaipeda University – Lithuania. The project also has a total of 10 associated partners from these countries. For additional information on the project and activities please visit the MORPHEUS homepage at: www.morpheus-project.eu

The aim of this report (Deliverable 5.2, see figure below) was to extract information from available reports about advanced treatment technologies that are already installed in Sweden, Germany and Switzerland. The implementation of advanced technologies is presented in the context of strategies to reduce the release of pharmaceuticals, antibiotics, hormones and other micropollutants to the aquatic environment adopted at both EU and national levels. Besides the goals of the MPs strategies, decision-making processes, the financing programmes and important examples of full-scale implementation results are presented.



Overview of advanced technologies in wastewater treatment – visualisation of Deliverable 5.2 in the context of MORPHEUS.

Table of contents

Summary

Micropollutants (MPs) including pharmaceuticals, antibiotics and hormones can enter the aquatic environment through both diffuse and point sources, but in urbanised regions wastewater treatment plants (WWTPs) play a crucial role in their dissemination. Conventional WWTPs are effective in macropollutants removal, while MPs may go through the treatment unchanged or are removed at different rates. Most EU countries are convinced that the presence of MPs in the environment poses a serious problem, particularly in highly populated regions where surface water resources serve as a sources of potable water. Thus, an additional treatment, referred to as the “fourth” or “quaternary” step, at WWTPs, seems to be inevitable.

An essential legal EU obligation to mitigate MPs arises from the European Water Framework Directive (WFD) and the environmental quality standards (EQS) for priority substances. Currently, besides listing priority substances (45 compounds or groups of compounds), Directive 2013/39/EU has also implemented a so-called Watch List. Watch List 1, established by Commission Decision 2015/495/EU, comprises the following pharmaceuticals: Diclofenac, Amoxicillin, Ciprofloxacin, Erythromycin, Clarithromycin and Azithromycin, which should be temporary monitored in surface water to obtain high-quality data sets. Reviewing the first Watch List resulted in the second Watch List, published in 2018. The monitoring of pharmaceuticals is important not only to determine the risk posed by them to the aquatic environment, but also, in the case of antimicrobial agents, to support the European One Health Action Plan against Antimicrobial Resistance. It has already been pointed out that besides monitoring, new treatment technologies efficient in degrading or removing antimicrobials in wastewater to reduce the spread of antimicrobial resistance are of high concern. But the lack of EU recommendation on effluent standards for MPs has postponed the implementation of new technologies in the wastewater sector. Additionally, in principle the EU policy says that the polluter pays, but in terms of MPs the subject is very complex. It is not clear who the polluter is, since stakeholders are, e.g., producers of chemicals, the pharmaceutical sector, hospitals and consumer groups. Therefore, two approaches need to be developed simultaneously:

(1) substitute critical MPs production and usage (source and user measures) and (2) mitigate the dissemination of MPs by WWTPs (end-of-pipe measures). Since not all of the substances, particularly pharmaceuticals, can be replaced with harmless alternatives, end-of-pipe technologies seem to be an essential part of the solution.

From the above it can be concluded that there are no requirements to remove pharmaceuticals from wastewater within the European Union, but there is a need, posed by the European Commission and other organisations, to monitor them at a European level and to develop methods, investigate the feasibility of upgrading selected urban waste water treatment plants to more advanced treatment technologies capable of eliminating a broad range of MPs at reasonable costs.

In Europe, Switzerland was the first country that in 2016 introduced a legal basis for implementation of an additional fourth step in wastewater treatment. As a consequence, about 100 out of the total 700 WWPTs are going to be extended or upgraded until 2040. The target is hereby an 80% removal of indicator MPs (e.g. pharmaceuticals) in the upgraded WWTPs, which

together are expected to treat 50% of the total wastewater volume generated in Switzerland. Currently, two technologies have been chosen for full-scale implementation: ozone and/or activated carbon filtration, due their high removal effectiveness for a wide range of MPs, cost-effectiveness, and simplicity of operation and maintenance. It is believed that the results obtained from large-scale implementation will show positive environmental effects as well as raising the public's and politicians' acceptance of environmental problems and how they can be solved.

In Germany, the national micropollutants strategy is currently in the consulting period and in the process of defining new regulations for advanced wastewater treatment. However, there are not yet legal requirements either for the application of technologies removing MPs or for pharmaceutical thresholds. Some federal states, in particular North Rhine Westphalia and Baden-Württemberg have already equipped several WWTPs with a fourth treatment stage on a voluntary basis. Similar to Switzerland, the technologies for MP removal in Germany are mainly based on ozonation and activated carbon.

In Sweden the government has already funded several projects related to MP removal from wastewater (mainly pharmaceuticals). Currently, the knowledge and operating experience of various technical solutions are completed and available as a foundation for the full-scale introduction of advanced treatment at WWTPs. Primarily ozonation and activated carbon have been tested and suggested as realistic alternatives for upgrading Swedish WWTPs at the national scale.

In Poland and Lithuania there is neither a legal basis nor other documents related to monitoring and/or removal of pharmaceuticals from wastewater. However, both countries are introducing national regulations imposing the need to assess priority substances. In Poland, for some substances particularly harmful to the aquatic environment, the maximum permissible values of pollution indicators have been specified for industrial wastewater.

It can be concluded that, presently, various technical solutions are available and have been proven to be possible to integrate with existing treatment processes in an expedient manner. The solutions that have been evaluated are mainly based on ozonation and/or activated carbon, and various combinations thereof. A key question and concern are the costs associated with taking control of the discharges of MPs, such as pharmaceuticals, from WWTPs. On the other hand, the benefits connected with the overall improvements of water quality by removing MPs from the wastewater stream cannot be directly quantified. Nevertheless, it is expected to aid in sustainable food production, drinking water supply and rainwater use. It is also beneficial for protection of bathing waters and the welfare of aquatic ecosystems. Finally, it is believed to help reduce the spread of antimicrobial resistance. Altogether this can be of help in getting a wider social acceptance of the increased costs associated with upgrading our European WWTPs.

German Summary

Mikroschadstoffe, einschließlich pharmazeutische Spurenstoffe, wie beispielsweise Antibiotika und Hormone, können sowohl über diffuse als auch über punktuelle Quellen in aquatische Ökosysteme eingetragen werden. In Ballungsräumen spielen Kläranlagen eine entscheidende Rolle, um diesem Eintrag entgegenzuwirken. Konventionelle Kläranlagen tragen effektiv zur Entfernung von Makroverunreinigungen bei; Mikroschadstoffe werden hingegen oft nicht oder nur teilweise aus dem Abwasser entfernt. In den meisten EU-Mitgliedstaaten wird der Eintrag von Mikroschadstoffen in die Umwelt als problematisch erachtet. Dies trifft vor allem auf Regionen mit einer hohen Bevölkerungsdichte zu, in denen Oberflächengewässer für die Trinkwassergewinnung genutzt werden. Daher scheint die Einführung einer zusätzlichen „vierten" Reinigungsstufe in den Kläranlagen unabdingbar zu sein.

Eine wesentliche gesetzliche Vorgabe der EU zur Verringerung des Eintrags von Mikroschadstoffen geht aus der europäischen Wasserrahmenrichtlinie (WRRL) und den Umweltqualitätsnormen (UQN) für prioritäre Stoffe hervor. Derzeit hat die Richtlinie 2013/39/EU neben der Auflistung prioritärer Stoffe (45 Verbindungen bzw. Gruppen von Verbindungen) auch eine sogenannte Beobachtungsliste (EU-Watch-List) eingeführt. Die mit der Entscheidung 2015/495/EU der Kommission festgelegte EU-Watch-List 1 umfasst folgende Arzneimittel: Diclofenac, Amoxicillin, Ciprofloxacin, Erythromycin, Clarithromycin und Azithromycin, die in Oberflächengewässern vorübergehend überwacht werden sollen, um die bisher unzureichende Datenlage zur Belastungssituation durch zusätzliches Monitoring zu verbessern. Im Jahr 2018 wurde die überarbeitetete Version der Beobachtungsliste (EU-Watch-List 2) veröffentlicht. Das Monitoring von Arzneimittelrückständen ist nicht nur wichtig, um mögliche Risiken für die Umwelt abschätzen zu können, sondern auch im Hinblick auf die Überwachung des Eintrags antimikrobieller Wirkstoffe, um die Umsetzung des EU Aktionsplans gegen Antibiotikaresistenzen ("EU One Health Action Plan on AMR") zu unterstützen. Es wurde bereits darauf hingewiesen, dass neben der Überwachung von Arzneimittelrückständen auch neue effektive Abwasserbehandlungstechnologien von großer Bedeutung sind. Diese können den Abbau oder die Entfernung von antimikrobiellen Wirkstoffen im Abwasser ermöglichen und so zur Verringerung der Ausbreitung von Antibiotikaresistenzen beitragen. Da die EU bisher keine Grenzwertempfehlungen für Mikroschadstoffe im Abwasser gegeben hat, verzögert sich die Einführung neuer Reinigungstechnologien im Abwassersektor. Darüber hinaus wird in der EU-Politik generell das Verursacherprinzip angewendet, in Bezug auf die Mikroschadstoffe ist der Sachverhalt aber sehr komplex. Es ist nicht klar festzustellen wer der Verursacher ist, da beispielsweise Hersteller von Chemikalien, der pharmazeutische Sektor, Krankenhäuser und auch die Verbraucher beteiligt sind. Daher müssen zwei Ansätze gleichzeitig entwickelt werden:

(1) Vermeidung von Einträgen in die Umwelt bei der Produktion und Nutzung kritischer Mikroschadstoffe durch den Einsatz umweltverträglicherer Produkte (Quellen- und Verbrauchermaßnahmen) und

(2) Verringerung der Emission von Mikroschadstoffen durch Kläranlagen in die Gewässer (End-of-Pipe-Maßnahmen). Da nicht alle Substanzen, insbesondere Arzneimittel, durch harmlose Alternativen ersetzt werden können, sind End-of-Pipe-Maßnahmen ein wesentlicher Bestandteil der Problemlösung.

Bislang hat die Europäische Union keine Vorschriften zur Entfernung von Arzneimittelrückständen aus Abwässern erlassen. Jedoch besteht laut EU-Kommission sowie anderer Organisationen die Notwendigkeit, Arzneimittelrückstände auf europäischer Ebene zu überwachen, neue Methoden zu entwickeln sowie Machbarkeitsstudien zur kostensparenden Nachrüstung von ausgewählten kommunalen Kläranlagen mit fortschrittlichen Reinigungstechnologien zur Entfernung eines breiten Spektrums von Mikroschadstoffen durchzuführen.

In Europa hat die Schweiz als erstes Land im Jahr 2016 eine Rechtsgrundlage für die Umsetzung einer zusätzlichen vierten Reinigungsstufe in der Abwasserbehandlung eingeführt. Infolgedessen werden etwa 100 der insgesamt 700 Kläranlagen bis 2040 erweitert oder aufgerüstet. Ziel ist hierbei eine 80%ige Entfernung von Indikator-Mikroschadstoffen (z. B. Pharmazeutika) in den aufgerüsteten Kläranlagen, durch die zusammen 50 % des in der Schweiz anfallenden Abwasservolumens behandelt werden. Aufgrund ihres hohen Wirkungsgrades bei der Entfernung eines breiten Spektrums an Mikroschadstoffen, der Kosteneffizienz sowie der einfachen Bedienung und Wartung, wurden derzeit zwei Technologien für die vollständige Implementierung ausgewählt: Ozon- und/oder Aktivkohlefiltration. Es wird davon ausgegangen, dass diese umfassende Implementierung der Technologien positive Auswirkungen auf die Umwelt zeigen wird und das Bewusstsein sowie die Akzeptanz in der Öffentlichkeit und Politik für Umweltprobleme und potentielle Lösungsansätze gestärkt wird.

In Deutschland befindet sich die nationale Mikroschadstoffstrategie, die zur Entwicklung erster Regelungen für den Umgang mit Spurenstoffen in der Umwelt und Abwasserbehandlung beitragen soll, derzeit in der Überarbeitungsphase. Es gibt jedoch aktuell keine gesetzlichen Anforderungen für die Anwendung von Technologien, die Mikroschadstoffe entfernen oder Schwellenwerte für den Eintrag pharmazeutischer Spurenstoffe. Einige Bundesländer, insbesondere Nordrhein-Westfalen und Baden-Württemberg, haben bereits freiwillig mehrere Kläranlagen mit einer vierten Reinigungsstufe ausgestattet. Ähnlich wie in der Schweiz basieren die Technologien zur Entfernung von Mikroschadstoffen in Deutschland hauptsächlich auf Ozonierung und Aktivkohle.

In Schweden hat die Regierung bereits mehrere Projekte, die sich auf die Entfernung von Mikroschadstoffen aus Abwässern (hauptsächlich Pharmazeutika) beziehen, finanziert. Aktuell wurden bestehende Erkenntnisse und Betriebserfahrungen mit verschiedenen technischen Lösungsansätzen zusammengestellt und als Grundlage für die umfassende Einführung fortgeschrittener Reinigungstechnologien in Kläranlagen zur Verfügung gestellt. In erster Linie wurden Ozonierung und Aktivkohle getestet und als realistische Alternativen für die Aufrüstung schwedischer Kläranlagen auf nationaler Ebene vorgeschlagen.

In Polen und Litauen gibt es weder eine Rechtsgrundlage noch andere umfassende Berichte zur Überwachung und/oder Entfernung von Arzneimittelrückständen aus Abwässern. Beide Länder haben aufgrund der Notwendigkeit einer Bewertung von prioritären Stoffen Vorschriften eingeführt, dass diese untersucht werden sollen. In Polen wurden für einige Stoffe, die für die aquatische Umwelt besonders schädlich sind, zulässige Grenzwerte für Belastungsindikatoren in Industrieabwässern festgelegt.

Zusammenfassend kann festgehalten werden, dass derzeit verschiedene technische Lösungen zur Verfügung stehen, die sich sinnvoll in bestehende Abwasserbehandlungsprozesse integrieren lassen. Die bewerteten Lösungsansätze basieren dabei hauptsächlich auf Ozonierung und/oder Aktivkohlefiltration bzw. auf verschiedenen Kombinationen beider Ansätze. Eine zentrale Herausforderung ist der hohe Investitionsaufwand, der mit einer Aufrüstung und dem Betrieb einer Technologie zur Reduzierung von Mikroschadstoffen wie Pharmazeutika aus Kläranlagen verbunden ist. Andererseits lassen sich die Vorteile, die mit der allgemeinen Verbesserung der Wasserqualität durch die Entfernung von Mikroschadstoffen aus Abwässern verbunden sind, nicht direkt quantifizieren. Es wird jedoch erwartet, dass es zu einer nachhaltigen Lebensmittelproduktion, Trinkwasserversorgung und Regenwassernutzung positiv beiträgt. Gelangen weniger Arzneimittelrückstände in Kontakt mit Mikroorganismen im Gewässer, kann ebenso die Verbreitung von Antibiotikaresistenzen eingedämmt werden. Somit ist die Reduzierung von anthropogenen Belastungen vorteilhaft sowohl für den Schutz der Badegewässer als auch der aquatischen Ökosysteme. Insgesamt können all diese Aspekte dazu beitragen, die gesellschaftliche Akzeptanz für die mit der Modernisierung der europäischen Kläranlagen verbundenen Kosten zu verbessern.

Swedish Summary

Mikroföroreningar såsom läkemedel, antibiotika och hormoner kan spridas till akvatiska miljöer från både diffusa källor och punktkällor, men i urbaniserade regioner spelar avloppsreningsverk en avgörande roll för deras spridning. Konventionella avloppsreningsverk är effektiva på att avlägsna makroföroreningar, medan mikroföroreningar kan passera reningsverkens processer oförändrade eller reduceras i olika grad. De flesta EU-länder är övertygade om att förekomsten av mikroföroreningar i vattenmiljön utgör ett allvarligt problem, särskilt i (mycket) tätbefolkade regioner där ytvattenresurser fungerar som källor till dricksvatten. Således verkar ytterligare ett reningssteg, ofta benämnt det "fjärde" eller "kvartära" steget, på reningsverken vara oundvikligt.

En väsentlig laglig skyldighet inom EU för att minska mängden mikroföroreningarna kommer från EUs ramdirektiv för vatten (Vattendirektivet) och miljökvalitetsnormerna för prioriterade ämnen. För närvarande, förutom att lista prioriterade ämnen (45 föreningar eller grupper av föreningar), har direktiv 2013/39/EU också introducerat en så kallad Bevakningslista. Bevakningslista 1, upprättad genom Kommissionens beslut 2015/495/EU, omfattar följande läkemedel: Diklofenak, Amoxicillin, Ciprofloxacin, Erytromycin, Klaritromycin och Azithromycin, som temporärt ska övervakas i ytvatten för att generera högkvalitativa förekomstdata. En översyn av Bevakningslista 1 resulterade i Bevakningslista 2, som publicerades 2018. Övervakningen av läkemedel är viktigt inte bara för att bestämma riskerna de medför i vattenmiljön, utan också i fallet med antimikrobiella ämnen, att stödja ”European One Health Action Plan” mot antimikrobiell resistens.

Det har redan påpekats att förutom övervakning, är ny reningsteknik, som effektivt minskar eller ta bort antimikrobiella ämnen i avloppsvatten för att minska spridningen av antimikrobiell resistens av stor vikt. Men bristen på EU-rekommendationer för gemensamma standarder avseende mikroföroreningar i avloppsvatten har lett till att implementeringen av ny reningsteknik för avloppsvatten har skjutits upp. Dessutom säger EUs politik i princip att förorenaren ska betala, men i fråga om mikroföroreningar är detta mycket komplicerat. Det går inte att peka ut en enskild förorenare, eftersom intressenter till exempel är producenter av kemikalier, läkemedelssektorn, sjukhus och konsumentgrupper. Därför behöver två tillvägagångssätt utvecklas samtidigt: (1) byta ut produktionen och användningen av särskilt farliga mikroföroreningar (åtgärder vid källan och hos användaren) och (2) minska utsläppen av mikroföroreningar från reningsverken (åtgärder vid utsläppspunkten). Eftersom inte alla ämnen, särskilt läkemedel, kan ersättas med ofarliga alternativ, tycks åtgärder vid utsläppspunkten vara en väsentlig del av lösningen. Av ovanstående kan man dra slutsatsen att det inte finns några krav på att ta bort läkemedel från avloppsvatten inom EU, men det finns ett behov, som ställs av EU Kommissionen och andra organisationer, av att övervaka föroreningarna på europeisk nivå och att utveckla metoder samt undersöka genomförbarheten av att uppgradera utvalda reningsverk med mer avancerad reningsteknik som kan eliminera ett brett spektrum av mikroföroreningar till rimliga kostnader.

I Europa var Schweiz det första landet som 2016 införde en rättslig grund för implementering av ett fjärde steg vid rening av avloppsvatten. Som en konsekvens kommer ungefär 100 av totalt 700 reningsverk att uppgraderas till 2040. För att övervaka reningens effektivitet har ett antal mikroföroreningar valts ut som indikatorer med kravet på 80% reningsgrad. Tillsammans förväntas de uppgraderade verken behandla 50% av den totala avloppsvattenvolymen som genereras i Schweiz. För närvarande har två tekniker valts ut för fullskalig implementering: ozon och/eller filtrering med aktivt kol, på grund av deras goda förmåga att kostnadseffektivt reducera ett brett spektrum av mikroföroreningar, samtidigt som teknikerna är relativt enkla att sköta och underhålla. Det antas att de erhållna resultaten från storskalig implementering kommer att visa positiva miljöeffekter samt öka allmänhetens och politikernas acceptans av miljöproblem och hur de kan lösas.

I Tyskland är den nationella mikroföroreningsstrategin för närvarande inne i en konsultationsperiod, och genomgår en process för att definiera nya bestämmelser för avancerad avloppsvattenrening. Men det finns varken lagliga krav på att implementera teknik som tar bort mikroföroreningarna eller gränsvärden för läkemedel. Vissa federala stater, särskilt Nordrhein Westfalen och Baden-Württemberg har redan utrustat flera reningsverk med ett fjärde behandlingssteg på frivillig bas. I likhet med Schweiz är teknikerna för att reducera mikroföroreningarna i Tyskland huvudsakligen baserade på ozonering och aktivt kol.

I Sverige har Regeringen redan finansierat flera projekt relaterade till avlägsnande av mikroföroreningar från avloppsvatten (främst läkemedel). Det finns nu kunskap och driftserfarenhet av olika tekniska lösningar tillgängliga som en grund för att introducera fullskalig avancerad rening av avloppsvatten på reningsverk. Primärt ozonering och aktivt kol har testats och föreslagits som realistiska alternativ för uppgradering av svenska reningsverk i en nationell skala.

I Polen och Litauen finns det varken en rättslig grund eller andra dokument relaterade till övervakning och/eller avlägsnande av läkemedel från avloppsvatten. Men i båda länderna introduceras nationella bestämmelser som kräver en bedömning av prioriterade ämnen. I Polen har man för vissa ämnen som är särskilt skadliga för vattenmiljön, specificerat högsta tillåtna värdena för föroreningsindikatorer i industriellt avloppsvatten.

Det kan konkluderas att det för närvarande finns olika tekniska lösningar tillgängliga vilka har visat sig möjliga att på ett ändamålsenligt sätt integrera med befintliga behandlingsprocesser. De lösningar som har utvärderats baseras huvudsakligen på ozonering och/eller aktivt kol, och olika kombinationer därav. En angelägen fråga är kostnaderna för att bättre kontrollera samt reducera utsläppen av mikroföroreningar såsom läkemedel från reningsverken. Fördelarna förenade med de övergripande förbättringarna av vattenkvaliteten när man avlägsnar mikroföroreningarna från avloppsvattenströmmen kan inte kvantifieras direkt. Ändå förväntas det stödja hållbar livsmedelsproduktion, dricksvattentillförsel och regnvattenanvändning. Det förväntas också vara fördelaktigt för att skydda badvatten och akvatiska ekosystems välmående. Slutligen tros det hjälpa till att minska spridningen av antimikrobiell resistens. Sammantaget kan detta vara till hjälp för att nå en bredare social acceptans av de ökade kostnaderna för uppgradering av våra europeiska reningsverk.

Polish Summary

Mikrozanieczyszczenia (MP), w tym leki, antybiotyki i hormony, mogą przedostawać się do środowiska wodnego zarówno ze źródeł rozproszonych, jak i punktowych, jednak w rejonach zurbanizowanych kluczową rolę w ich rozprzestrzenianiu odgrywają oczyszczalnie ścieków. Konwencjonalne oczyszczalnie ścieków są skuteczne w usuwaniu makrozanieczyszczeń, podczas gdy MP mogą przejść przez procesy oczyszczania w niezmienionej formie lub być usuwane w różnym stopniu. W większości krajów panuje przekonanie, że obecność mikrozanieczyszczeń w środowisku stanowi poważny problem, szczególnie w gęsto zaludnionych regionach, w których zasoby wód powierzchniowych służą jako źródła wody pitnej. Zatem dodatkowe oczyszczanie, określane jako „czwarty" lub „czwartorzędowy" stopień na oczyszczalniach ścieków, wydaje się nieuniknione.

W prawodawstwie UE podstawowy obowiązek ograniczania mikrozanieczyszczeń wynika z europejskiej Ramowej Dyrektywy Wodnej (RDW) i środowiskowych norm jakości (EQS) dla substancji priorytetowych. Obecnie, oprócz wykazu substancji priorytetowych (45 związków lub grup związków), w Dyrektywie 2013/39/UE wdrożono również tzw. listę obserwacyjną. Lista obserwacyjna 1, ustanowiona Decyzją Komisji Europejskiej 2015/495/UE, obejmuje następujące farmaceutyki: diklofenak, amoksycylinę, cyprofloksacynę, erytromycynę, klarytromycynę i azytromycynę, które należy tymczasowo monitorować w wodach powierzchniowych w celu uzyskania wysokiej jakości zestawów danych. Przegląd pierwszej listy obserwacyjnej zaowocował powstaniem drugiej listy obserwacyjnej, opublikowanej w 2018 r. Monitorowanie farmaceutyków jest ważne nie tylko w celu określenia ryzyka, jakie stwarzają dla środowiska wodnego, ale także, w przypadku środków przeciw drobnoustrojowych, w celu wsparcia Europejskiego planu działania „Jedno *zdrowie*" na rzecz oporności na środki przeciwdrobnoustrojowe. Wskazano, że oprócz monitoringu, duże znaczenie mają również nowe technologie oczyszczania, które skutecznie ograniczają lub usuwają środki przeciwdrobnoustrojowe ze ścieków, a tym samym zapobiegają rozprzestrzenianiu się oporności na ww. środki Jednak brak wytycznych UE w sprawie norm dla dopuszczalnych zawartości mikrozanieczyszczeń w oczyszczonych ściekach spowodował opóźnienie wdrażania nowych technologii w sektorze oczyszczania ścieków. Ponadto, co do zasady polityka UE stanowi, że zanieczyszczający płaci, jednak w przypadku MP temat ten jest bardzo złożony. Nie jest jasne, kto jest zanieczyszczającym, ponieważ stronami mogą być np. producenci chemikaliów, sektor farmaceutyczny, szpitale i grupy konsumenckie. Dlatego należy opracować jednocześnie dwa podejścia:

(1) zastąpić znaczącą produkcję i stosowanie MP (działania u źródła i po stronie użytkownika) oraz (2) ograniczyć rozprzestrzenianie się MP poprzez oczyszczalnie ścieków (działania „końca rury”). Ponieważ nie wszystkie substancje, zwłaszcza farmaceutyczne, można zastąpić nieszkodliwymi zamiennikami, technologie „końca rury” wydają się być istotną częścią rozwiązania problemu.

Z powyższego można stwierdzić, że obecnie w Unii Europejskiej nie ma wymagań dotyczących usuwania środków farmaceutycznych ze ścieków, jednak,zgodnie ze stanowiskiem Komisji Europejskiej i innych organizacji, istnieje potrzeba ich monitoringu na poziomie europejskim i opracowywania metod, a także zbadania możliwości modernizacji wybranych oczyszczalni ścieków komunalnych poprzez zastosowanie bardziej zaawansowanych technologii, które będą w stanie usunąć szeroki zakres MP przy rozsądnych kosztach.

W Europie, Szwajcaria była pierwszym krajem, który w 2016 r. wprowadził podstawę prawną dla wdrożenia dodatkowego, czwartego etapu oczyszczania ścieków. W rezultacie około 100 z ogólnej 700 oczyszczalni ścieków zostanie rozbudowanych lub zmodernizowanych do 2040 r. Celem jest usunięcie 80% wskaźnikowych MP (np. farmaceutyków) w zmodernizowanych oczyszczalniach ścieków, które jak się szacuje będą oczyszczać 50% całkowitej objętości ścieków wytwarzanych w Szwajcarii. Obecnie do wdrożenia w pełnej skali zostały wybrane dwie technologie do wdrożenia w pełnej skali: filtracja ozonowa (ozonowanie) i/lub filtracja na węglu aktywnym, ze względu na ich wysoką skuteczność usuwania szerokiego zakresu MP, opłacalność oraz prostą obsługę i konserwację. Uważa się, że wyniki uzyskane na dużą skalę przyniosą pozytywne skutki dla środowiska, a także zwiększą akceptację społeczeństwa i polityków wobec problemów środowiskowych oraz sposobów ich rozwiązania.

W Niemczech krajowa strategia dotycząca mikrozanieczyszczeń jest obecnie w fazie konsultacji oraz w trakcie opracowywania nowych przepisów dotyczących zaawansowanego oczyszczania ścieków. Jednak nie istnieją jeszcze wymogi prawne dotyczące stosowania technologii usuwania MP i wartości granicznych dla farmaceutyków. Niektóre landy, w szczególności Nadrenia Północna-Westfalia i Badenia-Wirtembergia, dobrowolnie wyposażyły kilka oczyszczalni ścieków w czwarty stopień oczyszczania. Podobnie jak w Szwajcarii, technologie usuwania MP w Niemczech opierają się głównie na ozonowaniu i węglu aktywnym.

W Szwecji rząd sfinansował kilka projektów związanych z usuwaniem MP ze ścieków (głównie farmaceutyków). Obecnie wiedza i doświadczenie w zakresie stosowania różnych rozwiązań technicznych są zakończone i dostępne jako podstawa do wdrożenia zaawansowanego oczyszczania na oczyszczalniach ścieków w pełnej skali. Testowano przede wszystkim ozonowanie i filtrację na węglu aktywnym jako realistyczne alternatywy dla modernizacji szwedzkich oczyszczalni ścieków w skali krajowej.

W Polsce i na Litwie nie ma ani podstawy prawnej ani innych dokumentów związanych z monitorowaniem i/lub usuwaniem farmaceutyków ze ścieków. Oba kraje wprowadzają jednak krajowe przepisy nakładające potrzebę oceny substancji priorytetowych. W Polsce dla niektórych substancji szczególnie szkodliwych dla środowiska wodnego określono maksymalne dopuszczalne wartości wskaźników zanieczyszczeń w ściekach przemysłowych.

Podsumowując, można stwierdzić, że obecnie dostępne są różne sprawdzone rozwiązania techniczne, które mogą być w odpowiedni sposób zintegrowane z istniejącymi procesami oczyszczania. Rozwiązania, które zostały poddane ocenie, oparte są głównie na ozonowaniu i/lub węglu aktywnym oraz różnych ich kombinacjach. Kluczowym pytaniem i obawą są koszty związane z przejęciem kontroli nad zrzutami MP, takich jak farmaceutyki, z oczyszczalni ścieków. Z drugiej strony nie można bezpośrednio oszacować korzyści związanych z ogólną poprawą jakości wody dzięki usunięciu MP ze ścieków. Niemniej jednak można się spodziewać, iż poprawa ta pomoże w zrównoważonej produkcji żywności, zaopatrzeniu w wodę pitną i wykorzystaniu wody deszczowej. Działania będą miały również korzystny wpływ dla ochrony wód w kąpieliskach i dobrostanu ekosystemów wodnych. Wreszcie, uważa się, że pomogą one zmniejszyć rozprzestrzenianie się oporności na środki przeciwdrobnoustrojowe. Ogółem mogą one pomóc w uzyskaniu szerszej akceptacji społecznej dla wyższych kosztów związanych z modernizacją europejskich oczyszczalni ścieków.

Lithuanian Summary

Mikro teršalai (MT), įskaitant farmacines medžiagas, antibiotikus ir hormonus, gali patekti į vandens aplinką iš pasklidosios ir sutelktosios taršos šaltinių, bet urbanizuotuose regionuose lemiamos įtakos jų pasklidimui turi nuotekų valymo įrenginiai (NVĮ). Įprastiniai NVĮ efektyviai šalina MT, tačiau, nepaisant to, MT, valymo proceso metu, gali išlikti nepakitę arba gali būti pašalinti skirtingi jų kiekiai. Daugumoje ES šalių laikomasi nuomonės, kad aplinkoje esantys MT yra rimta problema, ypač tankiai apgyvendintuose regionuose, kur paviršinio vandens telkiniai naudojami kaip geriamojo vandens šaltiniai. Vadinasi, papildomas valymas, vadinamas "ketvirta" arba "ketvirtine" pakopa, nuotekų valymo įrenginiuose, atrodo, yra neišvengiamas.

Svarbus teisinis ES įsipareigojimas – sumažinti MT lygį, kyla iš Europos Bendrosios vandens politikos direktyvos (WFD) ir aplinkos kokybės standartų (AKS), taikomų prioritetinėms medžiagoms. Dabartiniu metu, be prioritetinių medžiagų sąrašo (45 junginiai arba junginių grupės), Direktyvoje 2013/39/ES taip pat įtvirtintas vadinamasis stebėsenos sąrašas. Į pirmąjį stebėsenos sąrašą, kuris buvo patvirtintas Komisijos sprendimu 2015/495/ES, įtraukti šie medikamentai: diklofenakas, amoksicilinas, *ciprofloksacinas*, eritromicinas, klaritromicinas ir azitromicinas, kurių stebėsena laikinai turi būti vykdoma paviršinio vandens telkiniuose, siekiant gauti aukštos kokybės duomenų rinkinius. Peržiūrėjus pirmąjį stebėsenos sąrašą, sudarytas antrasis stebėsenos sąrašas, kuris buvo paskelbtas 2018 m. Farmacinių medžiagų stebėsena svarbi ne tik siekiant nustatyti jų keliamą riziką vandens aplinkai, bet taip pat, antimikrobinių medžiagų atveju, siekiant prisidėti prie Bendros sveikatos koncepcija grindžiamo Europos kovos su atsparumu antimikrobinėms medžiagoms veiksmų plano įgyvendinimo. Kaip jau buvo pažymėta, naujos valymo technologijos, kurios veiksmingai suskaido arba pašalina nuotekose esančias antimikrobines medžiagas, yra labai svarbios siekiant sumažinti antimikrobinio atsparumo plitimą. Tačiau, kadangi nėra ES rekomendacijų dėl nuotekose esančių MT normų, vėlinamas naujų technologijų įdiegimas nuotekų tvarkymo sektoriuje. Be to, iš principo, ES politikos nuostata yra tokia, kad moka teršėjas, tačiau, kalbant apie MT, problema yra labai sudėtinga. Neaišku kas yra teršėjas, kadangi su tuo susiję subjektai yra, pavyzdžiui, cheminių medžiagų gamintojai, farmacijos sektorius, ligoninės ar vartotojų grupės. Todėl, tuo pat metu turi būti plėtojamos dvi strategijos:

(1) pakeisti pavojų keliančią MT gamybą ir vartojimą (šaltinio ir vartotojo priemonės) ir (2) sumažinti MT pasklidimą per nuotekų valymo įrenginius (*„end-of-pipe“* - gamybos ciklo pabaigoje

įdiegtas priemones). Kadangi ne visas medžiagas, ypač farmacines medžiagas, galima pakeisti alternatyviomis nekenksmingomis medžiagomis, manoma, kad pagrindinę sprendimo dalį galėtų sudaryti valymo technologijos gamybos ciklo pabaigoje.

Atsižvelgiant į tai, kad buvo pasakyta pirmiau, galima daryti išvadą, kad Europos Sąjungoje nenumatyti jokie reikalavimai iš nuotekų pašalinti farmacines medžiagas, tačiau egzistuoja poreikis, kurį suformulavo Europos Komisija ir kitos organizacijos, vykdyti šių medžiagų stebėseną Europos lygiu ir kurti metodus, ir atlikti atrinktų miesto nuotekų valymo įrenginių modernizavimo, pereinant prie pažangesnių valymo technologijų, kurios užtikrintų plataus MT spektro pašalinimą pagrįstomis sąnaudomis, galimybių tyrimą.

Europoje, Šveicarija buvo pirmoji šalis, kuri 2016 m. nustatė teisinį pagrindą papildomos ketvirtos nuotekų valymo pakopos įdiegimui. Atsižvelgiant į tai, apie 100 nuotekų valymo įrenginių, kurių iš viso yra 700, turi būti išplėsti arba modernizuoti iki 2040 m. Užduotis – modernizuotuose nuotekų valymo įrenginiuose, kuriuose, numatoma, bendrai bus išvaloma 50% viso Šveicarijoje susidarančių nuotekų kiekio, pasiekti 80% MT (pvz., farmacinių medžiagų) pašalinimo rodiklį. Šiuo metu pasirinktos dvi technologijos, kurios yra diegiamos nuotekų valyklose susidarančių visų nuotekų išvalymui: ozonavimas ir/arba filtravimas naudojant aktyvuotą anglį. Šios technologijos buvo pasirinktos dėl jų efektyvumo šalinant platų MT spektrą, rentabilumo, paprasto eksploatavimo ir techninės priežiūros. Tikimasi, kad, plačiai diegiamos, šios technologijos turės teigiamą poveikį aplinkai ir taip pat prisidės prie visuomenės bei politikų geresnio aplinkos problemų ir jų sprendimo būdų supratimo.

Vokietijoje, šiuo metu vyksta konsultacijos dėl nacionalinės mikro teršalų strategijos ir naujų taisyklių, reglamentuojančių pažangesnį nuotekų valymą, rengimo procesas. Tačiau, teisiniai reikalavimai MT šalinimo technologijų taikymui arba farmacinių medžiagų ribinėms vertėms dar nenustatyti. Kai kuriose Vokietijos federalinėse žemėse, ypač Šiaurės Reino-Vestfalijos ir Badeno-Viurtenbergo žemėse, keliose nuotekų valyklose savanoriškai įdiegta ketvirta valymo pakopa. Panašiai kaip Šveicarijoje, Vokietijoje MT šalinimo technologijos daugiausia yra pagrįstos ozonavimu ir aktyvuotos anglies naudojimu.

Švedijoje vyriausybė jau skyrė finansavimą keletui projektų, susijusių su MT šalinimu iš nuotekų (daugiausia farmacinių medžiagų). Šiuo metu, sukauptos žinios ir darbo patirtis, susijusi su įvairiais techniniais sprendimais, gali būti naudojamos kaip pagrindas plataus masto pažangių valymo technologijų įdiegimui nuotekų valymo įrenginiuose. Pirmiausia, buvo išbandyti ozonavimo ir aktyvuotos anglies metodai, ir manoma, kad šie metodai galėtų būti realios Švedijos nuotekų valymo įrenginių modernizavimo alternatyvos nacionaliniu mastu.

Lenkijoje ir Lietuvoje nėra nei teisinio pagrindo nei kitų dokumentų, susijusių su nuotekose esančių farmacinių medžiagų stebėsena ir/arba jų šalinimu. Tačiau, abiejose šalyse galioja nacionaliniai teisės aktai, įtvirtinantys prioritetinių medžiagų vertinimo reikalingumą. Lenkijoje, kai kurioms vandens aplinkai ypač kenksmingoms medžiagoms nustatytos pramoninių nuotekų taršos rodiklių didžiausios leistinos vertės.

Galima daryti išvadą, kad šiuo metu egzistuoja įvairūs techniniai sprendimai, kurie gali būti racionaliai integruoti į esamus valymo procesus. Sprendimai, kurie buvo vertinami, daugiausia pagrįsti ozonavimu ir/arba aktyvuotos anglies naudojimu bei įvairiais šių technologijų deriniais. Pagrindinis klausimas ir susirūpinimą keliantis dalykas yra nuotekose MT šalinimo/valymo, pavyzdžiui, farmacinių medžiagų, išlaidos. Iš kitos pusės, neįmanoma tiesiogiai kiekybiškai įvertinti naudą, susijusią su bendru vandens kokybės pagerėjimu iš nuotekų srauto pašalinus MT. Nepaisant to, tikimasi, kad tai gali prisidėti prie tvarios maisto gamybos, geriamojo vandens tiekimo ir lietaus vandens panaudojimo. Tai taip pat būtų naudinga užtikrinant maudyklų vandens apsaugą ir vandens ekosistemų gerovę. Pagaliau, tikimasi, kad tai padėtų sumažinti atsparumo antimikrobinėms medžiagoms plitimą. Bendrai, tai gali paskatinti platesnį visuomenės pritarimą padidėjusioms išlaidoms, susijusioms su Europos nuotekų valymo įrenginių modernizavimu.

1 Introduction

Surface and underground water bodies have a variety of functions. They are important ecosystems with great natural diversity, leisure areas, hydroelectric reservoirs and water reserves for drinking/irrigation purpose. To supply all current needs and to maintain them for future generations, water reserves have to be sustainably managed. Such aims were defined in the Water Framework Directive EU[1] by achieving good ecological and chemical status of water bodies. It is estimated that over 30,000 different substances are in daily use in industrial, commercial and domestic applications (e.g. human and veterinary pharmaceuticals, plant protection products, biocides, personal care products, household chemicals and detergents).[2] These compounds may enter our surrounding water bodies through both point and non-point pathways. In this respect waste water treatment plants (WWTPs) are particular local point sources of harmful chemicals. Such anthropogenic compounds are an important stress factor, since their presence in the aquatic environment – even at very low concentrations (below µg/L) – may cause considerable toxicological concerns.[3] The fact that they often occur at low concentrations has caused them to be referred to as "micropollutants" (MPs). The primary focus of this report is the release of pharmaceuticals, antibiotics and hormones to our environment. To reduce their impact on the environment, a complex and coordinated strategy is required, including mitigation strategies on both the source and user side, as well as at end-of-pipe locations, as summarised in Figure 1.

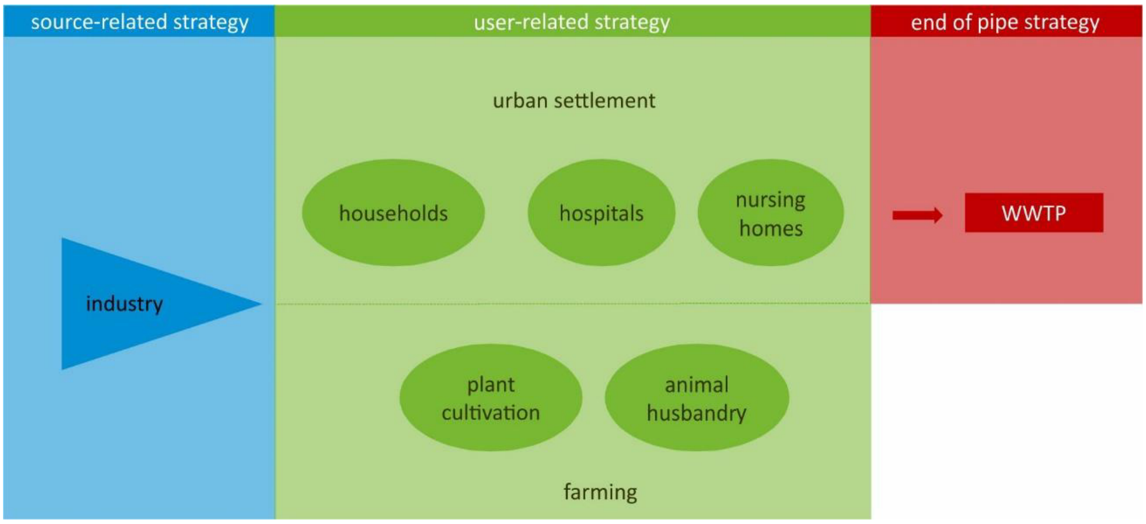


Figure 1. Strategies required to reduce the release of pharmaceuticals, antibiotics, hormones and other MPs to the aquatic environment

[1] Water Framework Directive 2000/60/EC
[2] OECD (2017), OECD Environmental Performance Reviews: Switzerland 2017, OECD Publishing, Paris,
[3] Schwarzenbach et al. (2006) The challenge of micropollutants in aquatic systems. Science 313(5790):1072-7.

A recent suggested proposal for a source- and user-related approach to reducing the release and impact of pharmaceuticals is presented below:

Pharmaceuticals:[4,5]

- to provide all environmentally relevant data for active substances,
- to collect data of active substances sales and usage,
- to do more research on environmentally friendly ingredients that can replace human and veterinary drugs that are persistent, bioaccumulative, and/or toxic to the environment,
- to inform physicians, pharmacists and patients about the environmental aspects of pharmaceuticals and about the disposal of unused drugs.

Separate strategies have also been formulated for personal care products, cleaning agents and biocides (for disinfection, material protection, pest control), as well as plant protection products,[6] but will not be further addressed here.

All actions mentioned above require both time and large resources in order to create measurable reductions in MPs at the production and usage stages. Additionally, not all inputs of pharmaceuticals can be prevented by these strategies, since some pharmaceuticals used in medical applications are absolutely essential in our healthcare systems and cannot easily be replaced by environmentally friendly alternatives. One group of special concern is antibiotics. According to the WHO, antibiotic resistance is one of the biggest threats to global health and development today.[7] The WHO states that there are some new antibiotics in development, but none of them are expected to be effective against the most dangerous forms of antibiotic-resistant bacteria. It is therefore not realistic to put additional constraints, such as environmental factors, on the development of effective antibiotics.

Since most pharmaceuticals end up in the municipal wastewater system, it has been suggested that the main emphasis should be placed on wastewater treatment processes (end-of-pipe technologies), which today are a major protective barrier in the water pollution control against organic material, nitrogen and phosphorous.[8] However, the effective removal of a broad spectrum of pharmaceuticals can today also be achieved with the help of an advanced fourth (quaternary) stage of waste water treatment, as discussed below.

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[4] The Strategic Approach to Pharmaceuticals in the Environment currently being developed by the European Commission (EC) according to the Article 8c of Directive 2008/105/EC (amended by Directive 2013/39/EU), for details see: Road map Strategic approach to pharmaceuticals in the environment (Ref. Ares(2017)2210630 – 28/04/2017)

[5] EurEau Position Paper. EurEau's Contribution to the European Commission Strategic Approach on Pharmaceuticals in the Environment, 26 May 2014, www.eureau.org

[6] Kümmerer et al. (2015) Long-Term Strategies for Tackling Micropollutants. In: Fatta-Kassinos D., Dionysiou D., Kümmerer K. (eds) Advanced Treatment Technologies for Urban Wastewater Reuse. The Handbook of Environmental Chemistry, vol 45. Springer, Cham

[7] https://www.who.int/news-room/fact-sheets/detail/antibiotic-resistance [Accessed February 2019]

[8] Hillenbrand et al. (2017) Recommendations from the Multi-Stakeholder Dialogue on the Trace Substance Strategy of the German Federal Government to policy-makers on options to reduce trace substance inputs to the aquatic environment. Bundesministerium für Umwelt, Naturschutz, Bau und Reaktorsicherheit und Umweltbundesamt. Berlin, 33 pp. http://publica.fraunhofer.de/documents/N-466986.html

2 Current wastewater treatment systems and MP removal, including pharmaceuticals

Historical and ongoing research programmes in the EU and elsewhere that investigated a wide range of different possible MP discharge sources recognised that WWTPs are the key pathway and important point sources for the introduction and dissemination of MPs such as pharmaceuticals into the environment.[9] Of special concern are receivers with a high fraction of treated wastewater, such as small streams, lakes and coastal waters. In such water bodies different MPs may be discharged simultaneously and a combined toxic effect of all MPs could be achieved. Furthermore, besides parent compounds, transformation products also have to be considered in order to understand the overall effect on biota in aquatic environments.

In EU law the strategy to prevent and reduce the inputs of MPs to the environment is guided by “the precautionary principle” and “the polluter pays principle”.[10] Consequently, a multi-stakeholder dialogue is needed for a holistic understanding and approach to (I) proposing indicator substances for monitoring of MPs at national and EU-wide level (II) developing practical and viable technological solutions for MP removal and (III) efficient financial programmes for investments in new infrastructure (Figure 2).

Figure 2. Multiple stakeholders involved in the MP-removal dialogue (adapted from[11])

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[9] Eggen et al. (2014) Reducing the discharge of micropollutants in the aquatic environment: the benefits of upgrading wastewater treatment plants. Environ Sci Technol. 15;48(14):7683-9.

[10] Directive 2004/35/EC of the European Parliament and of the Council of 21 April 2004 on environmental liability with regard to the prevention and remedying of environmental damage (ELD) establishes a framework based on the polluter pays principle to prevent and remedy environmental damage. The polluter pays principle is set out in the Treaty on the Functioning of the European Union (Article 191(2) TFEU).

[11] Hillenbrand et al. (2017) Recommendations from the Multi-Stakeholder Dialogue on the Trace Substance Strategy of the German Federal Government to policy-makers on options to reduce trace substance inputs to the aquatic environment, https://www.bmu.de/en/download/recommendations-from-the-muliti-stakeholder-dialogue-on-the-trace-substance-strategy-of-the-german-f

Conventional WWTPs based on activated sludge have historically been successfully developed and applied to control the dissemination of organic matters and nutrients such as nitrogen and phosphorus. Today it is clear, however, that MPs, including a number of pharmaceuticals, antibiotics and hormones, are removed with rather limited effectiveness by such technologies. Some MPs with low sorption coefficient, high water solubility and/or persistence to biodegradation may act as inert contaminants in the wastewater treatment process, passing unaltered through the WWTPs. There are several factors that determine the effectiveness of MP removal from wastewater. One of the most important is the properties of the MPs, as shown in Figure 3.

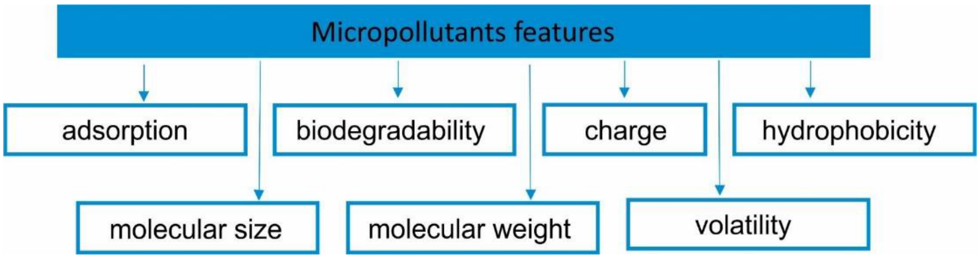

Figure 3. MP properties that are important in the processes of their removal from wastewater

It should be noted that pharmaceuticals, antibiotics and hormones are a very diverse and inhomogeneous group of MPs and that they cover a broad variety of differing properties. This may include, for example, size, charge and hydrophobicity. Apart from the properties of the MPs, other factors, such as plant configuration and operating conditions at the WWTP, are also important. This includes, for example, hydraulic retention time (HRT), wastewater pH and temperature. However, it is assumed that substances with similar physicochemical characteristic behave rather consistently towards treatment.

Before looking in more detail at both conventional and advanced treatment technologies' ability to remove pharmaceuticals and antibiotics from wastewater, a short overview of the legal and recommended bases for monitoring and removal of these MPs is warranted.

3 Legal and recommended basis for monitoring and removal of pharmaceuticals

The Water Framework Directive[12] (WFD) introduced a strategy for water protection, while Decision 2455/2001/EC[13] established a list of 33 priority substances, including 13 identified as “priority hazardous substances” (for details, see Annex A at the end of the report). This first list was replaced by Annex II of the Directive on Environmental Quality Standards (Directive 2008/105/EC)[14] (EQSD), which limits the concentrations of 33 priority substances (Annex II) and 8 other pollutants (Annex III) in surface waters (for details, see Annex B of this report). The Commission subsequently reviewed this list. Additionally, in 2013, Directive 2013/39/EU[15] amended both WFD and the EQSD (see Annex A) and established a Watch List mechanism. The Watch List is a list of potential water pollutants that should be temporarily monitored in surface waters to obtain a high-quality Union-wide dataset that would allow the risk these pollutants pose to the aquatic environment to be determined. The first Watch List, was published in 2015,[16] and included ten substances or groups of substances (see Table 1). Reviewing the first Watch List resulted in the second Watch List[17]. The Commission decided to remove five substances or groups of substances (diclofenac, the herbicides oxadiazon and triallate, the sunscreen ingredient 2-ethylhexyl-4-methoxycinnamate and the industrial compound 2,6-di-tert-butyl-4-methylphenol) and three new substances were included the pesticide metaflumizone and the two antibiotics amoxicillin and ciprofloxacin. The inclusion of the antibiotics on the second Watch List is consistent with the European One Health Action Plan against Antimicrobial Resistance (AMR),[18] which, among others, supports the use of the Watch List to improve knowledge and to evaluate the risks to human and animal health posed by the presence of antimicrobials in the environment. The updated second Watch List was published in 2018 (Table 1).

From the above it can be concluded that the removal of pharmaceuticals, antibiotics or hormones from wastewater is today not required within the European Union. However, there is a need expressed by the European Commission and other organisations to monitor them at a European level, as described in the EU Watch List 1 and Watch List 2. Within the European One Health Action Plan against Antimicrobial Resistance, it was also stated that multidisciplinary efforts are needed to develop (I) new tools for monitoring antimicrobials and microorganisms resistant against antimicrobials in the environment, and (II) new technologies that enable efficient and rapid

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[12] EU Water Framework Directive 2000/60/EC, http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32000L0060

[13] Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy, and amending Directive 2000/60/EC

[14] Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council. http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=celex%3A32008L0105

[15] Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2013:226:0001:0017:EN:PDF

[16] Commission Implementing Decision (EU) 2015/495 of 20 March 2015 establishing a watch list of substances for Union-wide monitoring in the field of water policy pursuant to Directive 2008/105/EC of the European Parliament and of the Council (notified under document C(2015) 1756)

[17] Commission Implementing Decision (EU) 2018/840 of 5 June 2018 establishing a watch list of substances for Union-wide monitoring in the field of water policy pursuant to Directive 2008/105/EC of the European Parliament and of the Council and repealing Commission Implementing Decision (EU) 2015/495

[18] https://ec.europa.eu/health/amr/action_eu_en

degradation of antimicrobials in wastewater and the environment to reduce the spread of antimicrobial resistance. In response to the EU legal basis and recommendations, some national level actions have been undertaken related to identifying which compounds should be monitored. Within the MORPHEUS project the national regulations and/or recommendations for the four participating countries Sweden, Germany, Poland and Lithuania are discussed below, as summarised in (Annex C).

Table 1. Micropollutants, including pharmaceuticals, antibiotics and hormones included in EU Watch List 1 and Watch List 2.

Compounds	Watch List 1 2015	Watch List 2 2018
Pharmaceuticals		
Diclofenac	X	-
Ciprofloxacin	-	X
Amoxicillin	-	X
Macrolide antibiotics (Erythromycin, Clarithromycin, Azithromycin)	X	X
Synthetic and natural hormones		
Estrone (E1)	X	X
17-Beta-estradiol (E2),	X	X
17-Alpha-ethinylestradiol (EE2)	X	X
Sunscreen ingredients		
2-Ethylhexyl 4-methoxycinnamate	X	-
Pesticides		
Methiocarb	X	X
Herbicides		
Tri-allate	X	-
Oxadiazon	X	-
Insecticides		
Neonicotinoids (Imidacloprid, Thiacloprid, Thiamethoxam, Clothianidin, Acetamiprid)	X	X
Metaflumizone	-	X
Industrial compounds		
2,6-ditert-butyl-4-methylphenol	X	-

3.1 SWEDEN

According to the Swedish EPA there are more than 1,000 active pharmaceutical ingredients (APIs) in use on the Swedish market today.[19] The ability to analyse and identify these in complex environmental water samples depends on the availability of advanced technologies based on

[19] https://www.naturvardsverket.se/Sa-mar-miljon/Manniska/Miljogifter/Organiska-miljogifter/Lakemedel/

liquid-chromatography couples to tandem mass spectrometry (LC-MS/MS).[20,21] Additionally, specific methods directed towards the analysis of a selected number of compounds must be developed. Over the past three decades, more and more pharmaceuticals, antibiotics and hormones have been added to these methods, which today may cover more than 100 compounds. As a result, our knowledge about the presence of MPs in wastewater, surface water, ground water and our surrounding seas has increased dramatically.

The backside of including an ever-increasing number of compounds to the methods is an increased complexity which may hamper both the quality and the interpretation and of the data. Additionally, this complexity leads to increased costs of analysis. Thirdly the comparability between different might also be poor when the applied methods differ.

From a Swedish perspective the Swedish Medical Products Agency identified the problems associated with analysis of pharmaceuticals, antibiotics and hormones in water and stated that there was a need for coordinated national analyses (compare with chapter 8. Swedish strategy). In 2015 they issued a report in Swedish named *"Miljöindikatorer inom ramen för nationella läkemedelsstrategin (NLS) 2015"* [Environmental indicators in the scope of the national pharmaceuticals strategy (NLS)].[22] In the report they stated: *"The working group considered the indicator "measure levels of pharmaceutical substances in environment" to be of the very highest priority. This is because, besides it being of major importance to monitor the development of drug remnants in the environment over time to evaluate the effect of implemented measures, the working group felt that there is considerable potential to optimise the use of the public resources through a better coordination of measurements in the environment. Many measurements have been taken historically by different public actors without any coordination."* Furthermore, the Swedish Medical Products Agency writes: *"The working group's continued work came to focus on preparing proposals on substances that should be monitored in the environment, i.e. measurement of the occurrence of pharmaceutical substances in water, sludge, inlet and outlet water of treatment plants, biota, etc."* At the end of the report they suggested monitoring of 22 "indicator pharmaceuticals" as listed (for details, see Table 7). The analysis of these compounds is not required by law, but it should be mentioned that some of the proposed substances are included in the European Commission's Watch List of substances for Union-wide monitoring in the field of water policy (EU) 2015/4954.

In 2013, the Swedish Agency for Marine and Water Management published statutes containing regulations on classification and environmental quality standards for surface water.[23] These included assessment grounds for specific pollutants in inland surface water as well as in coastal water. This list of compounds included the pharmaceutical diclofenac. In order for a surface water to be considered to be of good environmental status the maximum concentration of diclofenac

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

[21] Hermes et al. (2018) Quantification of more than 150 micropollutants including transformation products in aqueous samples by liquid chromatography-tandem mass spectrometry using scheduled multiple reaction monitoring. Journal of Chromatography A. Vol. 1531, 64-73.

[22] Report from the Office of the Centre for Better Use of Pharmaceuticals, Swedish Medical Products Agency 07/09/2015 - Miljöindikatorer inom ramen för nationella läkemedelsstrategin (NLS) [Environmental indicators in the scope of the national pharmaceuticals strategy (NLS)]; 7 pages.

[23] Havs- och vattenmyndighetens föreskrifter om klassificering och miljökvalitetsnormer avseende ytvatten. HVMFS 2013:19, Came into force 2013-09-01.

was set to 100 ng/L, expressed as a yearly average. The corresponding concentration for coastal water was ten times lower, with a maximum of 10 ng/L. These values may be further revised, as diclofenac has received much attention from different stakeholder groups lately, due to its high consumption as an OTC drug, and for its harmful effects on the environment.[24]

3.2 GERMANY

Since many years, the German environmental sector is aware of micropollutants which is directly shown in the established national regulations, for example the Surface Water Ordinance (OGewVO, for the details see Annex C). This Ordinance implements the European Requirements concerning EQS and regulates the emissions of so-called priority substances. However, this list of substances does not yet include pharmaceuticals even though suggestions for EQS have been made already by the Federal Environment Agency (UBA) (compare chapter 7. German strategy). Another research funding program concerning risk management of new pollutants and pathogens in the water cycle (RiSKWa) developed a guideline for a list of indicator substances with the purpose to identify the sources of the pollution, indicate anthropogenic changes in water quality and control/monitor natural and technical treatment processes[25]. These chemical indicator substances included several pharmaceuticals but with the mentioned functional purpose – they do not represent the degree of pollution or water quality standards itself, human- and ecotoxicological criteria were not included. Until now, these indicators are not adopted to legal regulations.

There have also been activities at the source of pharmaceutical products within admission of new substances. Since 1998, it is obligatory to perform an environmental risk assessment (ERA) within the admission procedure of human and veterinary pharmaceuticals in Germany according to the German Medical Products Act[26]. The European Medicines Agency published a guidelide for the ERA of human pharmaceuticals wherein the potential ecotoxicological risk has to be evaluated[27].

On a political level, a micropollutants strategy is currently under discussion, and some federal states, in particular North Rhine Westphalia and Baden-Württemberg, have already upgraded some WWTPs with a fourth waste treatment stage for MPs removal (see chapter German strategy below). These regional activities are mainly the reason of the pressure to act: The river basin Rhine-Ruhr is driven by a high population density. Besides resulting larger consumption loads of pharmaceuticals, this also leads to a high wastewater ratio in flow of natural water bodies. Hence, the issue on micropollutants including pharmaceuticals raised first in the affected regions of Germany.

Nevertheless, both regional and national research is ongoing and a common national strategy is on the way to harmonize the concept and measures to reduce the discharge of pharmaceuticals into the aquatic systems.

[24] Ringbom et al. (2017) Tonvis med diklofenak i våra vatten – regeländring behövsLäkartidningen; 114:EWL6
[25] Jekel & Dott (2013): RiSKWa-Leitfaden: Polare organische Spurenstoffe als Indikatoren im anthropogen beeinflussten Wasserkreislauf. Ergebnisse des Querschnittthemas Indikatorsubstanzen
[26] Ebert et al. (2010): Umweltrisikobewertung von Humanarzneimitteln. Pharmazeutische Industrie, 72, 1517-20
[27] EMA (2006). Guideline on the environmental risk assessment of medicinal products for human use

3.3 POLAND

In Poland there is neither a legal basis nor other documents related to the analysis or removal of pharmaceuticals from wastewater. It should be mentioned, however, that the legal basis for the national regulations imposing the need to assess the priority substances is the Water Act of 20 July 2017.[28] In 2016 the Ministry of Environment (Journal of Laws No. 2016, item. 681)[29] published a list of 45 priority substances in accordance with Directive 2013/39/EU. Currently the Polish Inspection for Environmental Protection is monitoring priority substances for which environmental quality standards have been specified in flora and fauna, priority substances that tend to accumulate in sediments, and substances particularly harmful to the aquatic environment included in the Watch List. Additionally, pursuant to Art. 45 Section 1 Point 1 of this binding Water Act, the Ministry of the Environment is required to issue the Ordinance on the conditions to be met when introducing wastewater into water or soil and on substances particularly harmful to the aquatic environment (amending the current legal basis: Journal of Laws. No. 2014, item 1800[30]).

Up to now, no changes have been made in the existing legislation. Currently, for some substances particularly harmful to the aquatic environment, the maximum permissible values of pollution indicators were specified for industrial wastewater (for details, see Annex D). Poland, however, has indicated in the National Environmental Monitoring Programme for the years 2016–2020, the need to continue existing tasks (and to implement new ones) connected with EU requirements for the environmental monitoring system, especially the implementation of the Directive 2013/39/EU of the European Parliament and of the Council regarding priority substances in the field of water policy. In the aquatic environment the monitoring of harmful substances (including priority substances) should be carried out annually on water bodies at representative points. If the results obtained in the first full annual monitoring cycle show that the concentration of this substance does not exceed the permissible limit values, the frequency of monitoring can be reduced to a minimum of 4 measurements per year (minimum every 3 months). Data collected during the monitoring are submitted to the Ekoinfonet IT system[31] belonging to the Inspection of Environmental Protection and are available on request in written or electronic form.

3.4 LITHUANIA

In Lithuania long- and mid-term environmental approaches and water management policies are determined by two strategic documents adopted in recent years.

In 2015, Lithuanian Seimas (Parliament) approved the National Environmental Protection Strategy. The Strategy define the priority areas of the environmental protection policy, long-term objectives up to 2030 and a vision for the Lithuanian environment, including water resources management up to 2050. For the reduction of dangerous chemicals in water bodies, the Strategy

[28] http://prawo.sejm.gov.pl/isap.nsf/DocDetails.xsp?id=WDU20170001566
[29] http://prawo.sejm.gov.pl/isap.nsf/DocDetails.xsp?id=WDU20160000681
[30] http://prawo.sejm.gov.pl/isap.nsf/DocDetails.xsp?id=WDU20140001800
[31] http://ekoinfonet.gios.gov.pl/

emphasises the importance of applying innovative technologies and well-balanced use of plant protection substances.

To prevent harmful effects of wastewater discharges these key implementing directions have been identified by the Strategy: *to raise public awareness about the aquatic environmental impact of wastewater; ensure that enterprises control priority hazardous substances that may be released into wastewater and that all generated wastewater is collected and managed in conformity with the established requirements; ensure the development and modernisation of wastewater management infrastructure through the efficient use of EU financial instruments.* More attention must also be paid to strengthened control of economic facilities, as well as implementation of an integrated pollution prevention and control system.

Specific indicators and measures are set in the medium-term Water Sector Development Programme for 2017–2023 approved by the Lithuanian Government and the Programme implementation Action Plan endorsed by Ministries of Environment and Agriculture in 2017.

A wide range of planned measures are designed to improve the status of surface and groundwater bodies, to achieve and maintain good environmental status of the Baltic Sea, and to reduce flood risks and their consequences throughout the country, etc. The Action Plan will primarily contribute to the prevention of pollution and the reduction of diffuse agricultural pollution. Impact mitigation measures of river dams and hydromorphological changes on water bodies and pollution reduction from point sources are also provided in the Plan. For point sources, the following main measures are foreseen: to increase effectiveness of wastewater treatment in 12 WWTP's; to ensure that all generated wastewater is collected and treated in conformity with the established requirements; to ensure the development and modernisation of wastewater infrastructure; and to enhance the accessibility of water supply and wastewater treatment services, especially in small towns and settlement of 200–2,000 inhabitants, with priority for EU investments in agglomerations with more than 2 000 inhabitants, etc. Additional requirements imposed to remove nitrogen and phosphorus in wastewater from single houses and small agglomerations.

For reducing emissions of harmful substances, the planned measures mainly focused on the strengthening of environmental permits, control and enforcement, monitoring and research.

However, no measures are integrated into these strategic planning documents to implement advanced treatment for the removal of micropollutants, including pharmaceuticals, in wastewater. Nevertheless, pilot investments in technological solutions for removing pharmaceuticals and other micropollutants are planned to be introduced in Kretinga town WWTP (for details, see chapter 10. Lithuanian strategy).

4 Advanced technologies for pharmaceutical removal

Up to now, several processes such as adsorption to granular and powdered activated carbon, membrane systems (sometimes combined with biological degradation) and advanced oxidation processes (such as O_3, UV/H_2O_2 or O_3/H_2O_2) have been developed for the removal of MPs from wastewater. Various lab-scale, pilot-scale and full-scale studies have been conducted in several countries in the past decade-and-a-half in order to investigate the application of advanced treatment in WWTPs. Processes and methods used to remove MPs including pharmaceuticals and antibiotics are listed in Table 2, while their effectiveness according to recent literature reviews is shown in Table 3. It should also be noted, though, that Table 3 is not a comprehensive list of pharmaceuticals, antibiotics and hormones, but a selection thereof. Still, it gives an idea of the similarities and differences between technologies.

The high variability in MP removal reported in the literature (Table 3) can be explained mainly by the different conditions of experiments. These include differences in treatment scale ranging from small lab-scale systems to large full-scale treatment facilities, and the origin of wastewater, which varied from synthetic wastewater to real wastewater obtained from working WWTPs. Other differences included variations in the tested WWTPs' overall performance, and differing seasons when the tests were conducted. Additionally, most studies focus only on the wastewater treatment line, while the sewage sludge management line is usually neglected. It is nonetheless obvious that some MPs tend to attach to sludge particles and are recirculated in the system with recirculated sludge or are removed with excess sludge. The fate of MPs in both recirculated and excess sludge is largely unknown. It should be noted, however, that the wastewater rejected from excess sludge processes (e.g. dewatering) is usually redirected to the wastewater line after mechanical treatment. For this reason, when calculating the MP balance in a WWTP system, the MP load removed/introduced from the sewage sludge processes should be included.

High differences in existing wastewater treatment systems (each WWTP is unique) mean that there is a large number of possibilities for combining them with advanced technologies. Thus, it is not possible to devise a single general solution for removal of all MPs such as pharmaceuticals at all WWTPs. For this reason, if removal of MPs is planned as a complementary treatment by WWTPs, this decision should be preceded by:

- MP monitoring in treated wastewater and receivers to prioritise the MPs of concern,
- defining the requirements for MP removal rate,
- performance of comparable, pilot-scale and on-site tests (I) to confirm the effectiveness of MP removal by specific technology in existing WWTPs, (II) to estimate the costs of this technology, and (III) to check whether this technology affects the existing wastewater treatment process as well as sewage sludge management,
- knowledge transfer, by e.g. study visits at other WWTPs, where the same technology has already been implemented.

All the above will be discussed in detail in Deliverable 5.3 of the MORPHEUS project.

Table 2. Processes and methods used for MP removal

Process	Method	Advantages	Disadvantages	Notes
Physical	- reverse osmosis - nanofiltration - microfiltration	- effective for a large number of different MPs - MP removal effectiveness quite/very stable	- by-product (concentrate) is problematic and costly to handle - high energy consumption	- alone, they cannot be regarded as treatment methods – they just separate permeate from concentrate (concentrated pollutants)
Biological	- membrane bioreactor (MBR) - moving bed biofilm reactor (MBBR) - other biofilm processes	- MPs are removed from wastewater via biodegradation and adsorption to activated sludge (removed from the system as excess sludge) - MP removal effectiveness quite stable	- MPs removal is a substrate- and microbial-community-dependent process - conversion and degradation of MPs is not well controlled (unknown intermediates)	- biological degradation is often combined with filtration (e.g. micro/nanofiltration), to ensure a more stable MP removal rate - formation of biofilms ensures higher diversity of bacterial consortia, longer retention time, and high sludge age, which is suspected to better reduce MPs - qualified staff needed
Adsorptive	- granular activated carbon (GAC) - powdered activated carbon (PAC)	- effective to a large number of different MPs - MP removal effectiveness quite stable	- regular replacement/ regeneration of GAC - in PAC technology the excess sludge produced has to be dewatered and incinerated - high energy requirement for regeneration of activated carbon - in presence of DOC/TOC competitive adsorption may occur	- activated carbon production and regeneration contribute to environmental footprint - effective biological step (low DOC/TOC concentration in effluent) is an important prerequisite for MP removal by activated carbon or ozone
Oxidative	- ozonation - UV/H_2O_2 - O_3/H_2O_2	- easily changed ozone dosage - MP removal effectiveness quite stable	- incomplete MPs degradation - high energy consumption	- ozone dosing depends on requirements and DOC/TOC concentration - ozonation requires the post-treatment step to destroy ozone residues and limit dissemination of any harmful transformation products. - qualified staff needed

Table 3. Example of efficiencies of selected MP removal by different wastewater treatment technologies reported in the literature[32,33]

	CAS	MBR	MBBR	O_3 + sand filtration	GAC	PAC + sand filtration	CW
(%)							
Antimicrobials							
Clarithromycin	37	0-99	47-61	>80	>80	>80	11-98
Sulfamethoxazole	35-84	0-90	(-28)-28	>80	30-60	60-80	0-75
Ciprofloxacine	63-90	15-94	2-96	30-60	30-60	60-80	N.A.
Hormones							
Estradiol (E2)	91-96	39-100	95-100	>80	>80	>80	0-100
Analgesics and Anti-Inflammatories							
Diclofenac	<0-81	<0-87	25-100	>80	60-80	60-80	0-75
Ibuprofen	-	-	-	30-60	60-80	>80	-
Beta Blockes							
Metoprolol	-	-	-	30-60	>80	>80	-
Contrast Agents							
Iopamidol	-	-	-	30-60	30-60	30-60	-
Diatrizoic acid	-	-	-	<30	<30	<30	-
Other							
Mecoprop	-	-	-	30-60	60-80	30-60	-
Benzotriazole	30-91	15-74	43-76	60-80	>80	>80	8-100

CAS – conventional activated sludge, MBR – membrane bioreactors, MBBR – moving bed biofilm reactor, GAC – granular activated carbon; PAC – powdered activated carbon, CW– constructed wetlands

[32] Krzeminski et al.. (2019) Performance of secondary wastewater treatment methods for the removal of contaminants of emerging concern implicated in crop uptake and antibiotic resistance spread: A review. Sci Total Environ. 648:1052-1081

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

Up to now, a number of studies have concluded that primarily two technologies are capable of eliminating a broad range of micropollutants at reasonable costs: ozonation and activated carbon treatment,[34,35] and thus these will be the main focus of this report.

4.1 Ozonation

Ozonation is in general an effective technology to reduce MPs in WWTPs. One of the benefits of using ozonation in aqueous solutions is that the hydroxyl ($OH^{\bullet}$) radicals, which are generated through the self-decomposition of ozone in water, react non-selectively with pharmaceuticals and other MPs. Ozonation additionally reduces some ecotoxic effects, especially estrogenic activity. The disinfectant properties of ozone are also considered important advantages of this method in some cases.[36]

There are also, however, some disadvantages to this method. For instance, in the production of potable water, the usage of ozone is limited if the concentration of natural bromide (Br^-) is significant, due to the formation of the carcinogenic bromate (BrO_3^-) in treated water. US EPA[37] and EU[38] quality standards limit the concentrations of BrO_3^- in drinking water to 10 µg/L. In wastewater technology, it is suggested that several MPs are not completely mineralised under the ozone dosages applied today, which are about 0.6–1.0 g O_3 per g DOC (dissolved organic carbon) and hydraulic retention times of about 20–30 minutes. Consequently, during ozonation, the MPs are transformed into other compounds, which may not be completely removed from the effluent. This formation of intermediates, which can be more toxic than the parent compounds, is a critical, extensively studied topic of ozonation.[39] Up to now, however, significant production of toxic by-products in full-scale WWTPs has not been noted. Nonetheless, in Germany and Switzerland it is advised to implement ozonation with a post-biological or sand-filtration step, to remove any biodegradable transformation by-products. The effectiveness of sand filtration in removing reactive compounds is, however, not fully recognised.

From a technological point of view, it can be noted that ozone is unstable, and thus cannot be stored on site, but must be produced directly prior to its application. The ozone is generated from pure oxygen or air through electrical discharge. After ozone has been generated it is mixed by injectors or diffusers with the effluent water of the WWTPs in a contact basin. It has been noted that energy consumption is slightly higher for the injectors and no increased removal of MPs was found compared to the use of diffusers.[40] Therefore, the latter are regarded as the better solution. The ozone–wastewater contact basin is air-tight, as the remaining ozone in gaseous form has to

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[34] Swedish Environmental Protection Agency Report 6766 (2018) Advanced wastewater treatment for separation and removal of pharmaceutical residues and other hazardous substances - Needs, technologies and impacts

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

[36] Zimmermann et al.. (2011) Kinetic assessment and modeling of an ozonation step for full-scale municipal wastewater treatment: micropollutant oxidation, by-product formation and disinfection. Water Res. 45(2):605-17.

[37] EPA, 2010. Revised State Implementation Guidance for the Consumer Confidence Report (CCR) Rule (EPA 816-R-09-010).

[38] EU, 1998. Official Journal of the European Community L 330: Directive 98/83/EG, Official Journal of the European Community L 330: Directive 98/83/EG

[39] as in [36] Zimmermann

[40] as in [35] Mulder

be treated. The effluent of the contact basin is then passed through a sand filter to remove any biodegradable metabolites.

It should be noted that during ozone generation, oxidation of nitrogen can also take place, which in the presence of moisture may form nitric acid. For this reason, to avoid corrosion of the ozonator, the air or oxygen must be moisture-free, which is achieved by cooling or drying the gas. Additionally, it is important to note that a concentration of ozone in air greater than 1 ppm is considered unsafe for prolonged human exposure. In legislation developed by the EU concerning air quality standards, the maximum allowable concentration of ozone in air should not exceed 120 μg/m^3 for continuous human exposure for 8 hours or 180 μg/m^3 for one-hour exposure.[41] WHO standards set a value of 100 μg/m^3 exposure per 8 hours.[42] Consequently, ozone detectors and warning systems should be present in buildings and other places where ozone is produced and used. A diagram of a typical ozonation system at a WWTP is shown in Figure 4, while Germany and Switzerland's suggested general design criteria for MP removal are given in Table 4.

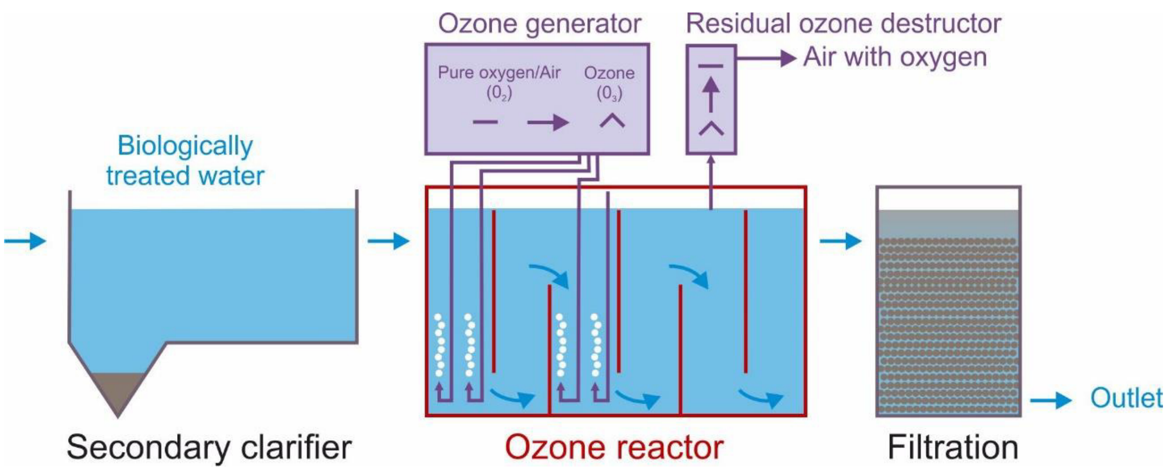

Figure 4. Example of ozonation system at WWTP (modified from [43])

[41] EU Air Quality Directive (2008/50/EC)
[42] WHO, 2006, Air quality guidelines: Global update 2005.
[43] Abegglen C. & Siegrist H. (2012): Mikroverunreinigungen aus kommunalem Abwasser. Verfahren zur weitergehenden Elimination auf Kläranlagen. Bundesamt fur Umwelt, Bern, Umwelt-Wissen Nr.1214: 210 S.

Table 4. General design criteria for removal of MPs from biologically treated wastewater by ozonation unit in Germany and Switzerland[44]

Subject	Unit	Value
Ozonation		
Dosage	$g\ O_3$ / g DOC	0.6–0.9
	$mg\ O_3/l^{a)}$	4–14
Hydraulic Retention Time Contact Tank	minutes	15–30 (reactor 10–25 min; removing remaining ozone 5 min)
Power consumption	kWh/kg O_3 × h	10
	W/treated m^3	45
Sand filtration after ozonation [b)]		
Upflow velocity	m/h	12
Backwash water	% of incoming flow	5–10
Power consumption	W/treated m^3	15

[a)] based on dissolved organic carbon (DOC) content in WWTP effluent of 7–15 mg/L

[b)] similar criteria for sand filtration after PAC

4.2 Activated Carbon

Activated carbon is commercially available in granular (GAC) and powdered (PAC) form, and is widely used as an adsorbent in many industrial processes due to its microporous, homogeneous structure. GAC typically has a particle size diameter ranging between 1.2 and 1.6 mm, while PAC has a particle size diameter smaller than 0.2 mm, typically in the range of 5–50 µm. The surface area of activated carbon is very large, normally ranging from 500 to 1,400 m^2/g[45]. Activated carbon properties depend on the surface area, pore volume and distribution of pore size, and the material used for production (Figure 5). Currently, activated carbons can be produced from a variety of materials of high carbon content that are activated at high temperatures (>700°C). Common raw

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

[45] Cecen F. & Aktaş O. (2011) Removal of NOM, Nutrients, and Micropollutants in BAC Filtration, in Activated Carbon for Water and Wastewater Treatment: Integration of Adsorption and Biological Treatment, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.

materials are coals (anthracite, bituminous and lignite), coconut shells, wood, peat and petroleum residues.[46]

The effectiveness of activated carbon in organic matter removal, including MPs, is generally connected to the physical properties of the compounds. Commonly, hydrophilic compounds are less adsorbed than hydrophobic substances. However, the charge of the MPs is also of great importance, where negatively charged pharmaceuticals bind less hard than those positively charged. Neutral pharmaceuticals bind more strongly than negative ones, but less so than positive pharmaceuticals. Additionally, the removal rate of hydrophilic MPs is greatly influenced by the presence of other organic matter, especially hydrophobic contaminants, due to the competitive adsorption (hydrophobic compounds are usually more easily and strongly adsorbed to activated carbon). A schematic overview of the binding and occurrence of both small and large organic molecules within the interior of an activated carbon particle is shown in Figure 5.

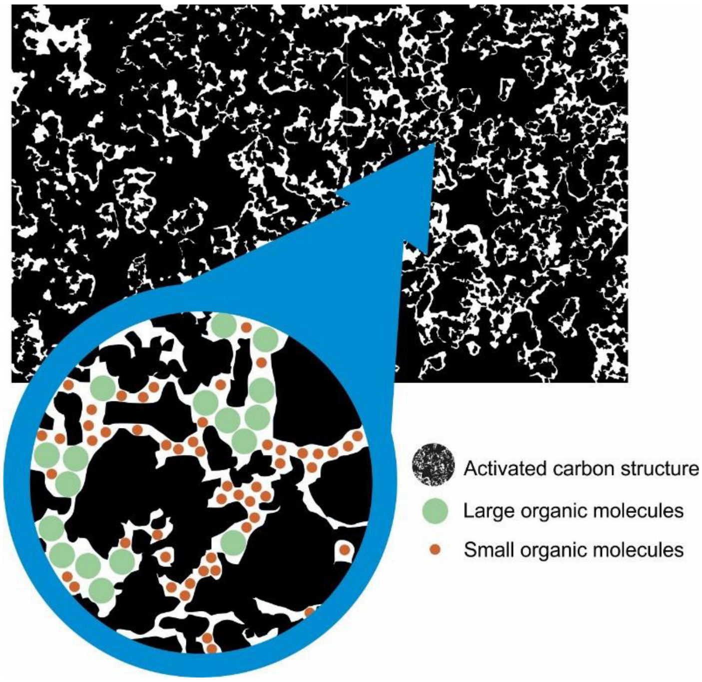

Figure 5. Schematic illustration of the adsorption of small and large organic molecules onto an activated carbon particle.

4.2.1 GAC technology

The advantages of MP removal by GAC technology include its simple application, operation and maintenance. GAC treatment in WWTPs is often applied as a single filtration step by a fixed bed filter as exemplified in Figure 6.

[46] https://www.desotec.com/en/carbonology/carbonology-academy/raw-materials-activated-carbon

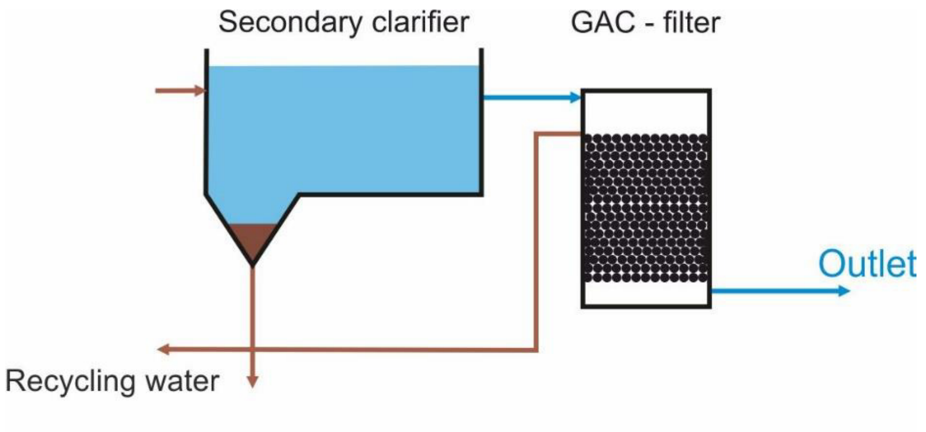

Figure 6. Example of a GAC system at a WWTP (modified from [47])

The incoming water flows downward under the force of gravity through the GAC medium, which is usually placed in a cylindrical tank. It should be noted that the presence of organic matter may reduce the effectiveness of MP removal due to competitive adsorption. Additionally, the blocking of GAC filter pores may become faster, when high amount of organic material is present. For this reason, if the settling tank at a full-scale WWTP does not function well, and the incoming water to the GAC filter contains suspended solids at higher than 10 mg/L, the GAC-filter should be bypassed.[48] To avoid clogging of the GAC filters, they are therefore sometimes preceded by sand filters. Parameters important in MP removal are presented in Figure 7.

Periodically, to remove organic matter and prevent blockage, the GAC filters may have to be flushed backwards with clean water, and this “backwashed” water is then directed back to the WWTP. If backwashing with water does not solve this problem, the GAC filter can be flushed with pressurised air. Additionally, the GAC filters have to be replaced once the effectiveness of targeted compound removal begins to drop. This means that all adsorption sites on the activated carbon are filled with contaminants. Reduced performance of the GAC filters is a signal to refill the filter with new or reactivated GAC. The advantage of GAC technology over PAC technology is connected with the possibility of GAC thermal regeneration. During this process, all adsorbates are volatilised and/or degraded (mineralised) and adsorption capacity is completely restored. However, during regeneration, about 10% of the GAC mass is lost. Some general design criteria suggested in Germany and Switzerland for MP removal by GAC are given in Table 5.

[47] Abegglen C. & Siegrist H. (2012): Mikroverunreinigungen aus kommunalem Abwasser. Verfahren zur weitergehenden Elimination auf Kläranlagen. Bundesamt fur Umwelt, Bern, Umwelt-Wissen Nr.1214: 210 S.
[48] Mulder et al. (2015) Costs of Removal of Micropollutants from Effluents of Municipal Wastewater Treatment Plants - General Cost Estimates for the Netherlands based on Implemented Full Scale Post Treatments of Effluents of Wastewater Treatment Plants in Germany and Switzerland. STOWA and Waterboard the Dommel, The Netherlands

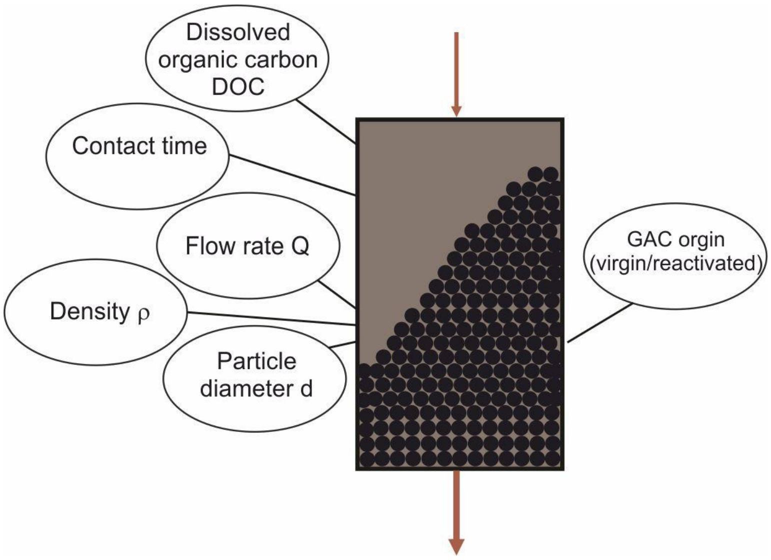

Figure 7. Parameters important in MP removal by a GAC treatment step (modified from [49])

Table 5. General design criteria for removal of MPs from biologically treated wastewater by GAC units in Germany and Switzerland[50]

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

[49] Abegglen C. & Siegrist H. (2012): Mikroverunreinigungen aus kommunalem Abwasser. Verfahren zur weitergehenden Elimination auf Kläranlagen. Bundesamt fur Umwelt, Bern, Umwelt-Wissen Nr.1214: 210 S.
[50] as in [48] Mulder M.

4.2.2 PAC technology

In PAC technology, the effluent of WWTPs is treated in a separate system, consisting of a contact tank, a settling tank and a filter. The PAC system is usually located after the existing biological stage (Figure 8a). To the contact tank, together with PAC, flocculants and coagulants are dosed (e.g. Al/Fe solutions). Due to the small size of PAC, its particles remain in the effluent, and a post-treatment is thus needed, mainly as a filtration step: sand, membrane, activated carbon filtration (see Figure 8).

The sludge from the PAC system is usually partly recycled to the contact tank but, optionally, can also be recycled to the biological step, e.g. to the aeration zone. Alternatively, the PAC can be dosed directly into the aeration tank of the activated sludge step (Figure 8, bottom panel) or to the inlet of existing sand filters. Direct dosing to biological treatment may significantly reduce the investment costs but this solution is still under investigation, since it is still not clear how either final or direct PAC dosing influences the effectiveness of MP removal or the effectiveness of the existing treatment system.

The dosages of PAC normally applied to WWTP effluents vary from 10 to 20 mg PAC/L. It should be noted that PAC dosage increase the amount of routinely dosed polymers and precipitation solutions by approximately 10% to 20%. Several PAC storages and feeding systems into wastewater are currently commercially available. The installations usually consist of a storage module, gravimetric feeding devices and a dissolving/mixing unit, which provides the optimal dose, depending on the volume and quantity of wastewater. Importantly, high feeding accuracy lowers operating costs. Importantly, too, the storage and dosage PAC systems should be designed in accordance to the special regulations if sparks can occur, since PAC can be explosive in the form of dust. Additional PAC can react with oxygen, releasing heat.

Other disadvantages of PAC treatment for MP removal is the clogging of the carbon slurry transport systems. This is mainly the result of undersized piping systems, short and sharp radius bends, insufficient velocity, and lack of cleaning. Additionally, abrasion of pipes transporting the slurry is a common problem. Those problems can be solved by increasing the size of the piping, using glass- or rubber-lined-steel or coated-cast-iron pipes.

Additionally, MP removal requires a certain PAC dosage. Spent PAC is continuously removed from the system, and usually dewatered, dried and finally incinerated, which limits the further dissemination of the pollutants into another environment. Thus, from a carbon-use perspective, PAC is less economical than GAC. Some general design criteria suggested in Germany and Switzerland for MP removal by PAC are given in Table 6.

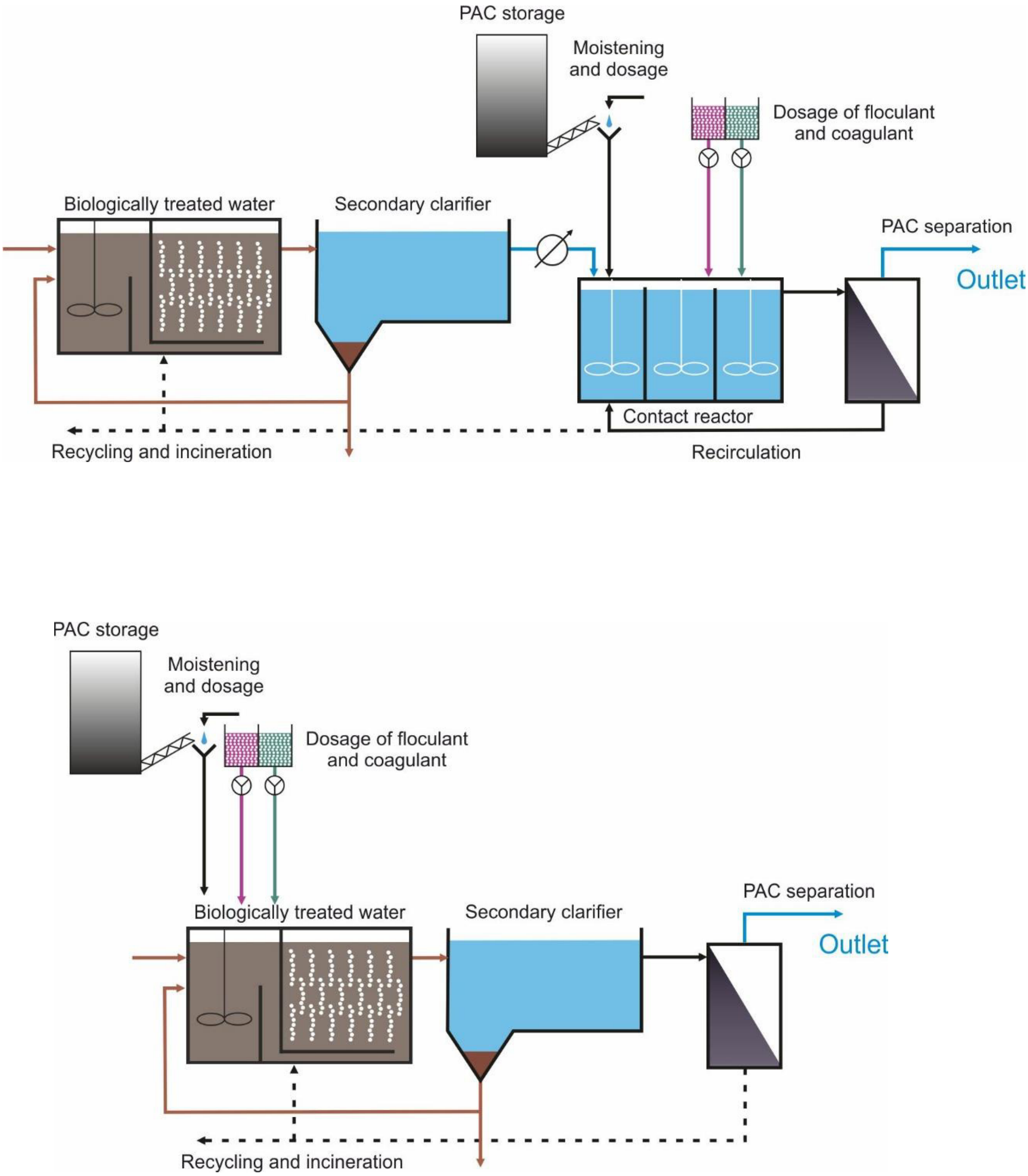

Figure 8. Example of two PAC systems at WWTPs (modified from [51]). Top panel: PAC system located after the existing biological stage. Bottom panel: PAC system dosing directly into the aeration tank of activated sludge stage

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

Table 6. General design criteria for removal of MPs from biologically treated wastewater by PAC unit in Germany and Switzerland[52]

Subject	Unit	Value
PAC		
Dosage	g PAC / g DOC	0.7–1.4
	mg PAC /L[a)]	10–20
Dosage coagulant	mg/L	4–6
Dosage polymer	mg 100% active /L	0.2–0.3
Hydraulic Retention Time Contact Tank	minutes	30–40
Surface load settler	m/h	2.0
Recycle factor PAC	-	0.5–1.0
Power consumption	W/treated m^3	45
Sand filtration after PAC [b)]		

[a)] based on dissolved organic carbon (DOC) content in WWTP effluent of 7–15 mg/L

[b)] similar criteria as for sand filtration after ozonation (see Table 4)

4.3 Process automation

The process parameters and effectiveness of MP removal should be easily controlled by process automation. In the case of GAC filters, the important issue is the accuracy of velocity through the filter bed volume. Another important issue is the clogging of the GAC filter, which can be measured through pressure changes. Pressure increasing beyond a threshold value should automatically start the back-flushing of the filter bed with air or water. To estimate the lifetime of a GAC filter and the need to replace the activated carbon, break-through curves need to be established by periodical MPs measurement in the quaternary treated effluent.

For both ozone and PAC technology, accurate dosage is an important issue in MP removal. Currently, ozone and PAC can be easily over- or under-estimated because dosages are adjusted to the flow of incoming biologically treated wastewater, while the effectiveness of those

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

technologies is strongly linked to the DOC concentration. At full-scale installations direct online measurement of DOC concentration is rather difficult and not accurate enough. For this reason, at several applications, indirect measurement was applied based on the loss of UV light absorption at 254 nm (UVA254).[53] Besides the dosage, the contact time is also important in MP removal; for this reason, further research is needed into ozone and PAC dosage.

4.4 Cost estimation

In estimating the costs of MP removal from wastewater, several parameters need to be taken into account, as shown in Figure 9.

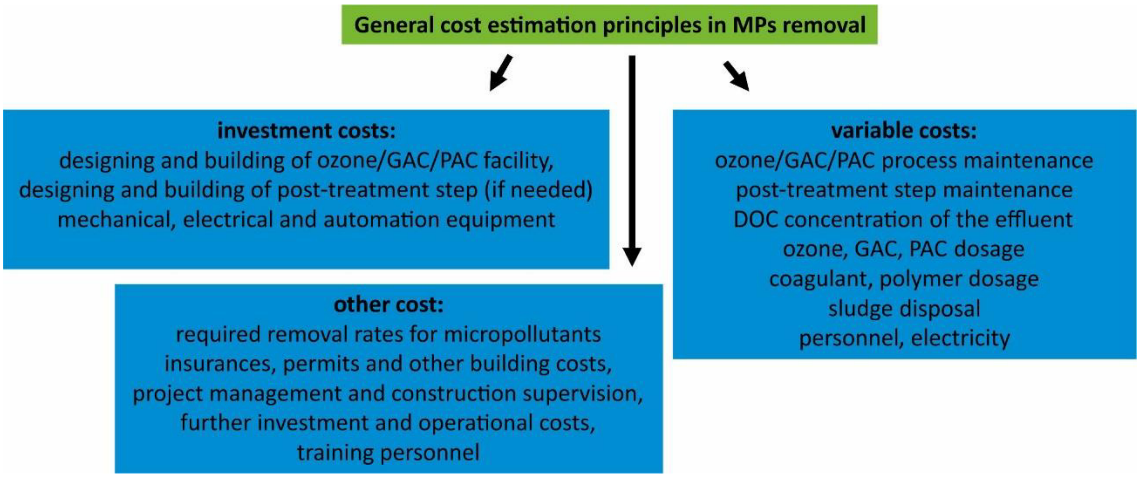

Figure 9. Parameters to consider in calculating the cost of MP removal from wastewater[54]

In general, it can be concluded that the overall investments costs for ozonation and PAC treatment are similar, while for GAC treatment they are usually significantly lower, due to the simplicity of this technology's installation.[55] In energy consumption, ozone technology usually requires double the energy of PAC treatment and up to 12 times more than GAC treatment. However, the overall maintenance costs of ozonation are the lowest, since PAC treatment requires continuous dosage of PAC, coagulants and polymers, as well as sludge treatment (usually dewatering and thermal processing), while for GAC treatment the maintenance costs are connected with the periodical need of activated carbon exchange/regeneration. When comparing the viable costs of GAC and PAC treatment, it was assumed that GAC technology is about 5 times more expensive per m^3 of treated wastewater than PAC technology,[56] including both GAC replacement (usually after 6

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[53] Bahr et al. (2007). UVA as control parameter for the effective ozonation of organic pollutants in secondary effluent. Water Sci Technol. 55 (12), 267-274.
[54] Mulder et al. (2015) Costs of Removal of Micropollutants from Effluents of Municipal Wastewater Treatment Plants - General Cost Estimates for the Netherlands based on Implemented Full Scale Post Treatments of Effluents of Wastewater Treatment Plants in Germany and Switzerland. STOWA and Waterboard the Dommel, The Netherlands
[55] as in [54] Mulder M.
[56] as in [54] Mulder M.

months) and the sludge processing with a post-treatment step in PAC treatment. This is also connected with the higher amount of GAC needed to achieve the same removal rates as PAC, which is more effective per g of coal (see point 4.2). However, a very recent 4-year Swedish study in the FRAM project (Full-scale treatment of micropollutants)[57] performed at Kristianstad University (LP) shows that the cost of GAC filtration can be reduced dramatically by an appropriate protection of the GAC filter from unwanted organic material using a common sand filter. This is discussed below at some length in the section "Swedish strategy" (see chapter 8).

There are also trials to reduce the costs of this technology by using biochar instead of GAC. For example, the SystemLäk project (Systems for the purification of pharmaceutical residues and other emerging substances)[58] conducted adsorption tests using different types of biochar, and some could reduce pharmaceutical residues from particular treated wastewater with a capacity comparable to that of commercially available activated carbon. Thus, the obtained results are very promising, and we expect the costs of this technology to drop in the future.

In cost calculations one has also to be aware that when costs are presented in the literature, the MP removal is sometimes separated from post-treatment, so costs connected with the design, building and maintenance of post-treatment installations may not be included in the overall costs needed to build and maintain MP removal technology. It was concluded, that depending on the technique, the costs of post-treatment steps for MP removal may vary from 0.16 to 0.33 EUR/m^3 in treated effluent.[59] Between countries, differences in, for example, electricity and labour expenditure may also influence the final cost calculations for advanced technologies.

[57] Björklund E. & Svahn O., 2019, FRAM - Fullskalig Rening Av Mikroföroreningar 2014–2018. Filtrering av avloppsvatten genom sand och granulerat aktivt kol (GAK) för avskiljning av läkemedel och antibiotika som alternativ till ozonering. Report Kristianstad University, Sweden, in preparation.

[58] www.hammarbysjostadsverk.se.

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

5 Other options in wastewater treatment for MP removal

5.1 Relevance of entry pathways from healthcare facilities

It is estimated that in some regions about 20% of the active pharmaceutical ingredients in municipal wastewater may originate from hospitals, nursery houses and other healthcare facilities, and the remaining share from households.[60] For this reason, decentralised treatment of wastewater from healthcare facilities to eliminate certain micropollutants used in human medicine has been considered. A well-known example from Denmark is the direct treatment of raw hospital wastewater from Herlev Hospital, which has a total of 700 beds and produces 150,000 m^3 wastewater yearly as presented in a report from 2016.[61] From May 2014 the hospital wastewater has been cleaned by a combination of MBR-Ozone-GAC-UV in series, and after two years of operation the evaluation revealed a 99.9 % removal of a large number of pharmaceuticals and antibiotics. Additionally, the water was no longer toxic to aquatic organisms and no traces of bacteria occurred, including antibiotic-resistant bacteria that cause hard-to-treat diseases. Pictures from the treatment facility re shown in Figure 10.

Figure 10. Pictures of the direct treatment of raw waste water from Herlev Hospital Denmark using a combination of advanced treatment technologies reducing the occurrence of pharmaceuticals with more than 99%. Photo: Erland Björklund 2018.

The incentive to use such advanced treatment technologies was reduced operating costs for the treatment plant in relation to fee for discharge to the public sewer. The former costed 1.45 EUR/m^3, while the latter 3.41 EUR/m^3, meaning a saving of 1.96 EUR/m^3.

[60] Ahting et al. (2018) Recommendations for reducing micropollutants in waters. Ed. Helmecke M. (II 2.1) German Environment Agency Section II 2.1 General Aspects of Water and Soil. https://www.umweltbundesamt.de/sites/default/files/medien/1410/publikationen/180709_uba_pos_mikroverunreinigung_en_bf.pdf

[61] Full scale advanced wastewater treatment at Herlev Hospital - Treatment performance and evaluation. DHI Report May 2016.

A few comparative studies and research projects such as noPILLS[62] and Sauber+ [63] indicated that in most cases there is no greater entry of medicinal product residues, toxic substances, antibiotic-resistant bacteria or resistance determinants from healthcare facilities. For this reason, separate wastewater treatment is reasonable only in isolated cases. It cannot be, however, generalised, and each relevant entry pathway should be separately analysed, especially for separate collection/disposal of radiocontrast agents.

[62] http://www.no-pills.eu/conference/BS_NoPills_Final%20 Report_summary_EN.pdf
[63] https://www.sauberplus.de/

6 Swiss strategy

6.1 Current situation: Switzerland

In Europe, Switzerland is regarded as a pioneering country in enforcing legal obligations for the monitoring and removal of MPs, including several pharmaceuticals. In 2011 the Swiss parliament approved the proposal of the Federal Office for the Environment to reduce MP loadings by 80% at selected WWTP outlets. This was the result of several studies that confirmed the contribution of municipal WWTPs to the total water pollution. After public consultations in the years 2012–2014, the revised Water Protection Ordinance came into force in January 2016, and requires that Swiss municipalities implement technical measures to remove MPs in selected WWTPs.[64] In Switzerland it is going to be achieved mainly by upgrading existing WWTPs. However, wastewater pre-treatment at some sources, such as hospitals, nursing home, etc. was also regarded as reasonable if they represented a high proportion of the total pharmaceutical load in a catchment (see section 5.1).

The decision to start reducing MP dissemination via WWTPs was preceded by a wide range of pilot- and full-scale monitoring programmes focusing on MP removal technologies' levels of effectiveness, their ecotoxicological impact and their energy demand. According to the obtained results it was decided that both WWTPs and technical innovations should follow the below criteria:[65]

Selection of WWTPs for upgrading has to be based on:

- the anticipated MP load of WWTPs serving >80,000 persons, upstream responsibility,
- dilution capacity of wastewater receiver if wastewater consists of >10% of dry-season stream flow,
- protection of sensitive areas and water bodies feeding drinking water reservoirs.

Selection of technical innovations has to be based on:

- effectiveness on as broad a range of micropollutants as possible,
- flexibility and accessibility for implementation in existing infrastructure without disturbing existing processes,
- acceptable cost/benefit ratio.

This upgrading procedure will be complemented by closing down small WWTPs, and diverting their wastewater to larger plants.

Up to now, several technologies have been tested in terms of MP removal in Switzerland. The technical feasibility and costs support combining existing biological treatment steps with ozone or

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64 Gewässerschutzgesetz GSchG; https://www.admin.ch/opc/de/classified-compilation/19910022/index.html
65 Logar et al. (2014) Cost-benefit analysis of the Swiss national policy on reducing micropollutants in treated wastewater, Environmental Science and Technology, 48(21), 12500-12508

powdered activated carbon (PAC). Thus, the Swiss strategy in MP removal from wastewater has focused on ozone and PAC, since:

- they can eliminate a broad range of micropollutants,
- they are both economically feasible and manageable for WWTP personnel.

MP removal effectiveness should be controlled from 8 to 24 times per year (depending on WWTP size), using 24-h or 48-h composite samples and the presence of indicator substances according Table 7. The indicators Amisulpride, Carbamazepine, Citalopram, Clarithromycin, Diclofenac, Hydrochlorothiazid, Metoprolol, Venlafaxine, Benzotriazole, Candesartan, Irbesartan, Mecoprop were chosen according to the following criteria:

- They are present in sufficiently high concentrations in influent of most Swiss WWTPs with small load variation.
- Their chemical properties are similar to other MPs typically present in wastewater.
- Their removal by conventional Swiss WWTPs (biology) is little or non-existent.
- They can be assessed simply during a single run with LC/MS/MS.

The scope of the treatment is to obtain 80% removal (primary clarified wastewater vs. final effluent) measured with minimum 6 out of 12 indicator compounds: four from category 1 (“compounds very easily eliminated by advanced treatment”) and two from category 2 (“compounds easily eliminated by advanced treatment”) (indicate, whether advanced treatment is operated correctly). For details, see Table 7.

It should be noted that the Swiss strategy for MPs removal has also been criticised for a number of reasons: (I) not considering all ecological aspects, (II) imposing a financial burden on society, and (III) providing ambiguous environmental benefits.[66] Up to now, large-scale implementation has not proven its environmental effects, since it requires long-term evidence. Notwithstanding, all the actions taken so far have brought increased awareness of environmental issues among the public and politicians.

[66] Johnson A.C. & Sumpter J.P. (2015) Improving the quality of wastewater to tackle trace organic contaminants: Think before you act! Environ. Sci. Technol. 49, 3999– 4000

Table 7. Indicator substances for checking the performance of advanced treatment of MPs in Switzerland[67]

Type	Substance	Category
Substances of medical origin		
Antibiotics	Clarithromycin	1
Antidepressants	Amisulpride	1
	Citalopram	1
	Venlafaxine	1
Antihypertensives	Irbesartan	2
	Hydrochlorothiazide	1
Anti-inflammatories	Diclofenac	1
Beta blockers	Metoprolol	1
Tranquillisers	Carbamazepine	1
	Candesartan	2
Other substances		
Anticorrosion agents	Benzotriazole	2
Biocides	Mecoprop	2

Category 1 and 2: compounds eliminated by advanced treatment technologies very easily and easily, respectively.

6.2 Preliminary cost and energy consumption analysis: Switzerland[68]

Preliminary cost/benefit and energy consumption analysis indicated that:

- the energy consumption will increase by 10 to 30% per WWTP. By 5–10 % for larger WWTPs and 15–30 % for smaller. Nationally, total energy consumption will increase by 0.1%.
- the costs will increase from 5 to 35% per WWTP: the smaller the plant, the higher the costs. It is estimated to increase the national annual costs of wastewater treatment by 12%, and the annual costs of wastewater disposal by 6%.

In Switzerland, 100 WWTPs larger than 10,000 PE are being consider for upgrade for MP removal, with investment costs estimated at ca. 1,200 million CHF. The financial contribution to the project follows the “polluter-pays principle”, and is divided in two streams:

[67] Eggen et al. (2014) Reducing the discharge of micropollutants in the aquatic environment: the benefits of upgrading wastewater treatment plants. Environ Sci Technol. 48:7683-9
[68] http://www.water2020.eu/sites/default/files/keynote_adriano_joss_eawag_switzerland.pdf

1. 75% of the investment paid by the national budget:
   - municipalities pay 9 CHF/person/year into a fund
   - municipalities with upgraded WWTPs are exempted
   - only direct costs for upgrading for MP removal are covered (nutrient removal not covered)
   - financing starts in 2016 and ends in 2040
2. 25% of investment + operation costs covered by municipalities.

6.3 Full-scale case studies: Switzerland

In Switzerland, large-scale pilot experiments were carried out at three WWTPs (ozonation and PAC treatment). The questions to be answered with the full-scale studies were the following:

- What is the elimination capacity of the process?
- How does the wastewater quality change in terms of other parameters such as ecotoxicology, pathogens, foaming, odour, etc.?
- Are there any undesired side effects or products, e.g. waste, toxic substances or interference with biological treatment?
- Is the technology technically and operationally applicable in terms of infrastructure, process engineering, control, safety, etc.?
- How much energy is required?
- What are the costs for construction and operation?

Swiss Example 1: WWTP Neugut in Dübendorf full-scale ozonation system[69]

At the WWTP Neugut in Dübendorf, a full-scale ozonation system has been in effect since April 2014, as shown in Figure 11.

[69] Schachtler M. & Hubaux N. Ozonation planning/implementation HOLINGER AG, Liestal, Ingenieurbüro Gujer AG, Rümlang, ARANeugut, Otto-Jaag-Strasse15 CH-8600 Dübendorf, www.neugut.ch

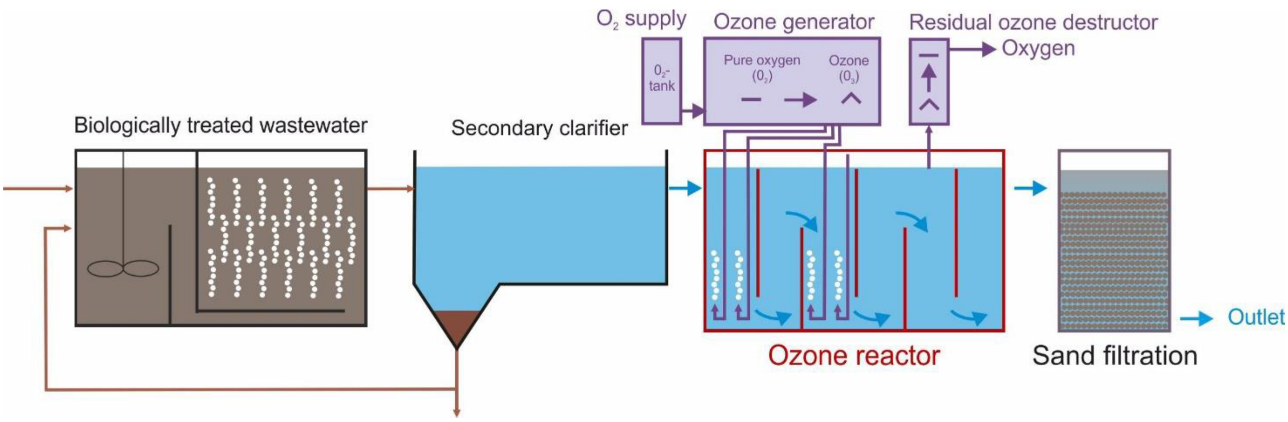

Figure 11. WWTP Neugut in Dübendorf with a full-scale ozonation system (modified from [65])

Some basic characteristics of the WWTP are listed below:

- ***WWTP characteristic***: plant size: 155,000 PE (105,000 inhabitants and 55,000 industry); flow range: Q = 13,000–57,000 m³/d ($Q_{min\text{-}max}$ = 70–660 L/s)
- ***ozone unit inflow wastewater characteristic***: COD = 16 mg/L; DOC = 5.3 mg/L; N_{NH4} = 0.08 mg/l; N_{N02} = 0.03 mg/L; pH = 7.4 mg/L; Q = 70–660 L/s
- ***Ozonation unit characteristic***: pure oxygen tank 80 m^3; ozone generators: 2 × 5.5 kg O_3/h; ozone reactor: V = 530 m^3 (divided in two ozonation chambers with ceramic diffusers) water depth 6.0 m; mean residence time 37 min (min. residence time 13 min.)
- ***Ozone dosage***: 0.33–0.50 g O_3/g DOC and 1.6–2.7 g O_3/m^3
- ***Ozonation unit energy requirements***: pure oxygen 28 g/m^3; electricity: 0.024 kWh/m^3; entire plant: 0.42 kWh/m^3
- ***Costs of ozonation***: Gross investment (excl. deduction of federal subsidy): 3.27 million CHF; amortisation, maintenance: 0.025 CHF/m^3; operating costs: 0.014 CHF/m^3; total costs per inhabitant: 6 CHF/year; ozonation operating costs per year: 110,000 CHF/year including 40% pure oxygen; 20% electricity; 20% indicator compound analysis; 20% personnel and overheads
- ***Removal efficiency of MPs***: average elimination of 12 indicator substances from wastewater and varied between 80% and 86%, as shown in details in Table 8

Table 8. Removal of selected MP indicators by ozonation achieved by WWTP Neugut across the entire plant, including sand filtration. The ozone dose was 2.2 mg O3/L or 0.42 g O3/g DOC.

Substance	**Tradename[1)]**	**Medication**	**Removal rate**
		Pharmaceuticals	
Carbamazepine	Tegretol	tranquilliser (epilepsy and neuropathic pain)	>95%
Diclofenac	Voltaren	pain and inflammatory diseases (gout)	>95%
Metoprolol	Lopressor	beta blocker	>85%
Irbesartan	Aprovel, Karvea, and Avapro	antihypertensive	63–65%
Hydrochlorothiazide	large number of brand names		80–85%
Amisulpride	large number of brand names	antidepressant	85–95%
Citalopram			85–95%
Venlafaxine			75–85%
Clarithromycin	Biaxin, Klacid	antibiotic	>90%
		Other MPs	
Benzotriazole	-	Anticorrosion agent	79–82%
Methylobenzotriazole			80–83%

[1)] examples of trade names; trade names may differ between countries

Currently, the ecotoxicological measurements are being carried out on effluent after ozonation, and different types of ozonated-effluent post-treatments are also being tested, such as sand filter, fluidised sand bed filter and granular activated carbon filter. This is very important because more WWTPs are planned to be upgraded. The planning and construction of other WWTPs will be performed based on the experience gained at Neugut.

Swiss Example 2: WWTP ARA Thunersee full-scale PAC system[70]

WWTP Thunersee treats municipal wastewater from 38 communities in the area around the city of Thun. The WWTP went into operation in 1972, and since then the plant has continuously been extended. The plant has an impressively high treatment performance level, low operating costs, and extraordinarily good energy values. Since June 2018, a PAC system has been operated for removal of MPs, as shown in Figure 12.

[70] Pulveraktivkohledosierung (PAK) ARA Thunersee Plattform Verfahrenstechnik Mikroverunreinigungen, www.micropoll.ch

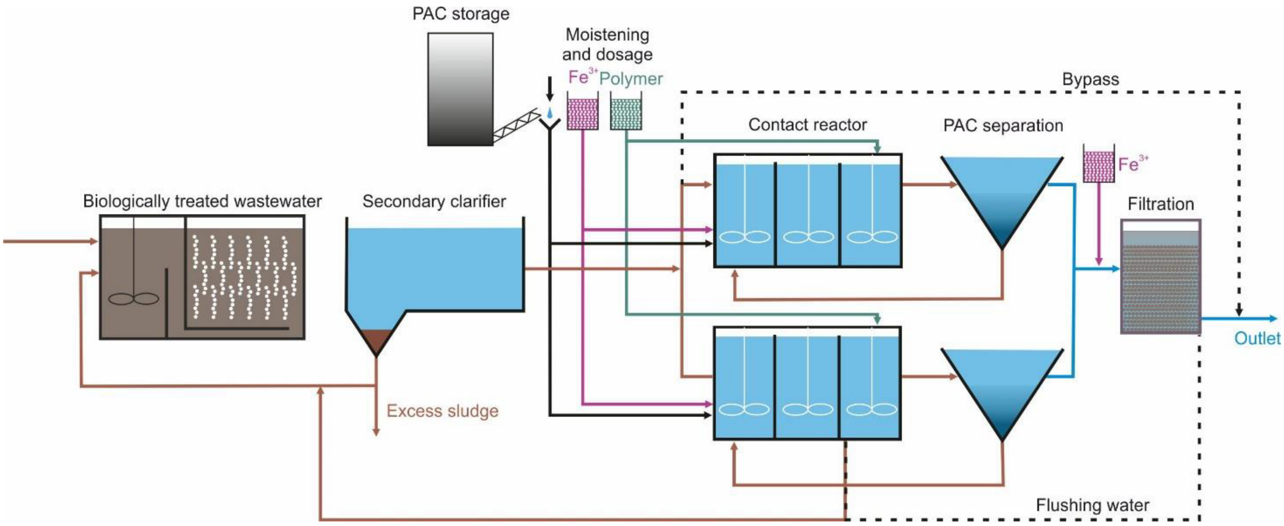

Figure 12. WWTP Thunersee with a full-scale PAC system (modified from [71])

Some basic characteristics of the WWTP are listed below:

- ***WWTP characteristic***: plant size: 150,000 PE (124,000 inhabitants and 26,000 industry); flow range: Q = 500 L/s – 1,350 L/s, in dry and rainy weather, respectively; WWTP treats over 13 million m^3 annually
- ***treatment process***: mechanical step contains: coarse and fine screen, sand trap, and primary clarifier; biological stage is based on activated sludge system (nitrification, denitrification, bio-P) combined with secondary clarifier; chemical precipitation (addition of ferric chloride in return sludge) to ensure effective phosphate removal
- ***PAC unit characteristic***: 2 PAC silos (80 m^3), 2 PAC dosing stations, 2 PAC contact basins (1,100 m^3 each, contact time: 46 mins), 4 sedimentation basins (1,944 m^3 each, residence time 2.7 h) and 8 filtration cells (42.2 m^2 each, sand and anthracite, max. filter speed 9.8 m/h), the PAC excess sludge is channelled into the return biological part of wastewater treatment sludge

[71] Pulveraktivkohledosierung (PAK) ARA Thunersee Plattform Verfahrenstechnik Mikroverunreinigungen, www.micropoll.ch

7 German strategy

7.1 Current situation: Germany

The micropollutants strategy in Germany is in progress and will mainly be developed at both the national and the regional level by involved actors such as the Federal Environmental Agency (abbrev. “UBA”) and the German Working Group on water issues of the Federal States and the Federal Government, represented by the Federal Environment Ministry (abbrev. LAWA)[72]. In order to evaluate this work appropriately, the legal basis in Germany regarding the topic of micropollutants, i.e. pharmaceuticals, needs to be considered.

At the national level, the Federal Environmental Agency investigated and also published comprehensive reports in recent years on measures to reduce the discharge of micropollutants into waters (see UBA 2014 and UBA 2016)[73] [74]. Considering legal requirements at the EU-level, the WFD requires Environmental Quality Standards (EQS) for polluting substances[75]. Hence, Germany’s Federal Environmental Agency (UBA) also initiated a project to update the environmental quality standards (EQS) for 10 (non-pharmaceutical) pollutants specific to river basins according to the Surface Water Ordinance (OGewV), and to compile suggestions for environmental quality standards, including EQS proposals for pharmaceuticals. Within this project EQS proposals were derived for, *inter alia*, Bezafibrate, Carbamazepine, Erythromycin, Metoprolol, Roxithromycin and Sulfamethoxazole (UBA 2015)[76]. Here, already existing EQS concentration levels (µg/L) for different matrices are collected and updated accordingly to recent research activities.

Additionally, another policy instrument, this one based on financial incentive systems, is the Wastewater Levy Act[77,78]. It provides the possibility to charge the total loads discharged via wastewater into the environment. If this financial charge system were strengthened, investment costs for, e.g., advanced treatment technologies could be re-financed indirectly by this instrument. This may appear as a suitable and interesting option in German wastewater regulation in the future and could represent an economic instrument to be implemented into the micropollutant strategy.

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[72] https://www.bmu.de/fileadmin/Daten_BMU/Download_PDF/Binnengewaesser/spurenstoffstrategie_policy_paper_bf.pdf

[73] Hillenbrand et al. (2014) Measures to reduce micropollutant emissions to water. Summary. https://www.umweltbundesamt.de/sites/default/files/medien/378/publikationen/texte_87_2014_mikroschadstoffe_summary.pdf

[74] Hillenbrand et al. (2016) Measures to reduce micropollutant emissions to water – Phase 2. de/sites/default/files/medien/377/publikationen/ mikroschadstoffen_in_die_gewasser-phase_2.pdf

[75] Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy

[76] Wenzel et al. (2015) Revision der Umweltqualitätsnormen der Bundes-Oberflächengewässerverordnung nach Ende der Übergangsfrist für Richtlinie 2006/11/EG und Fortschreibung der europäischen Umweltqualitätsziele für prioritäre Stoffe. https://www.bmu.de/fileadmin/Daten_BMU/Pools/Forschungsdatenbank/fkz_3712_28_232_umweltqualitaetsnormen_bf.pdf

[77] Gesetz über Abgaben für das Einleiten von Abwasser in Gewässer (Abwasserabgabengesetz - AbwAG), https://www.gesetze-im-internet.de/abwag/AbwAG.pdf

[78] Gawel et al. (2017) Arzneimittelabgabe – Inpflichtnahme des Arzneimittelsektors für Maßnahmen zur Reduktion von Mikroschadstoffen in Gewässern https://www.umweltbundesamt.de/sites/default/files/medien/1410/publikationen/2017-12-19_texte_15-2017_arzneimittelabgabe.pdf

When focusing on the substance itself, despite numerous suggestions for EQS, no legal thresholds for pharmaceuticals have been determined to date. However, several monitoring data on micropollutants not regulated by law are reported for several surface waters within different Federal States. Even though the data has been produced with different research aims in deviating monitoring campaigns, the pharmaceutical burden has been determined, and emphasised the need for a national strategy (LAWA report 2016). Additionally, regional studies, especially at the Federal State Level, established specialised competence centres for micropollutants to bring together and transfer the knowledge gained by smaller-scale research results. Distinguishing itself from others is the Environmental Agency in North Rhine Westphalia, which already has developed a list of trace substances including the pharmaceuticals Benzotriazole, Carbamazepine, Diclofenac, Metoprolol, Clarithromycin and Sulfamethoxazole in addition to other priority substances[79]. Further advanced and comprehensive studies are also provided by the International Commission for Rhine Protection (ICRP) which evaluated the pharmaceutical burden at total-catchment scale[80]. In their report, the basic research for a common strategy on reduction and prevention of diverse micropollutants discharged into the Rhine and its backwaters was developed by improving the knowledge on emissions, ecotoxicological effects in nature and efficient treatment technologies. Due to the immense number of different chemicals used within the Rhine catchment, the main important substance groups were selected and evaluated regarding their occurrence in the surface waters. The report shows their actual burden in 2017 compared to 2011. Additionally, the status of planned and implemented reduction measures in the different related states are described and serves as basis for further discussion on a future action plan to be developed[81]. In order to transfer the regional experience, a so-called policy paper "Multi-Stakeholder Dialogue on the Trace Substance Strategy of the German Federal Government to policy-makers on options to reduce trace substance inputs to the aquatic environment"[82] was initiated by Actors of the Federal States and water boards, guided by the German water protection policy.

The main goal of the German micropollutants strategy is to harmonise both regional and national approaches and data of finalised studies. In November 2016 the multi-stakeholder dialogue was launched as part of the preparations for a federal-government strategy to mitigate MPs in the aquatic environment. Here, details are discussed in an ongoing process to define the overall objectives and regulations, i.e. for wastewater treatment. The outcomes of this dialogue are presented in Table 9[83].

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[79] ARGE Kompetenzzentrum Mikroschadstoffe.NRW in 2016. https://www.masterplan-wasser.nrw.de/fileadmin/user_upload/Broschueren_PDFs_und_Titel_JPGs/Machbarkeitsstudie_11_2016.pdf

[80] Mikroverunreinigungen im Rheineinzugsgebiet Bilanz 2017. IKSR-CIPR-ICBR 2017, https://www.iksr.org/fileadmin/user_upload/DKDM/Dokumente/Fachberichte/DE/rp_De_0246.pdf

[81] Mikroverunreinigungen im Rheineinzugsgebiet Bilanz 2017. IKSR-CIPR-ICBR 2017, https://www.iksr.org/fileadmin/user_upload/DKDM/Dokumente/Fachberichte/DE/rp_De_0246.pdf

[82] Hillenbrand et al. (2017) Recommendations from the Multi-Stakeholder Dialogue on the Trace Substance Strategy of the German Federal Government to policy-makers on options to reduce trace substance inputs to the aquatic environment. Bundesministerium für Umwelt, Naturschutz, Bau und Reaktorsicherheit und Umweltbundesamt. Berlin, 33 pp. http://publica.fraunhofer.de/documents/N-466986.html

[83] Hillenbrand et al. (2017) Recommendations from the Multi-Stakeholder Dialogue on the Trace Substance Strategy of the German Federal Government to policy-makers on options to reduce trace substance inputs to the aquatic environment. Bundesministerium für Umwelt, Naturschutz, Bau und Reaktorsicherheit und Umweltbundesamt. Berlin, 33 pp.

Table 9. Overview of the topics and recommendations from the multi-stakeholder dialogue on the strategy to reduce trace-substance inputs to the aquatic environment

Source-related	Use-related	End-of-pipe
Recommendation 1:		
1.1 Determination of relevant trace substances		
Recommendation 2:	**Recommendation 3:**	**Recommendation 4:**
2.1 Communicating the findings of environmental risk assessment and closing knowledge gaps	3.1 Joint information campaign on the relevance of trace substances to the aquatic environment	4.1 Oriented framework for additional effluent treatment in sewage plants
2.2 Producers' recommendations on behaviour to mitigate trace-substance inputs to waters	3.2 Taking up the issue of the relevance of trace substances to the aquatic environment in initial/advanced training and advisory programmes	4.2 Research and development, if appropriate, of measures specific to precipitation-water discharge and combined storm-water–sewage discharge
2.3 Recommendations for reducing where appropriate effluent discharges from production and processing	3.3 target-group-focused labelling schemes	4.3 Exchange of information and R&D on the upgrading of municipal sewage in fracture
2.4 Reducing trace substances in important productions	3.4 Development or refinement of specific measures on the user side	4.4 Proper disposal of residues and wastes
Recommendation 5:		
5.1 Costs of implementing the Trace Substances strategy		

Among them Recommendations 4.1 to 4.4 address end-of-pipe measures, which involve technical or organisational measures necessary to prevent or reduce dissemination of relevant trace substances. It was suggested that WWTPs are important steps in reducing MPs, but due to the limited effectiveness of the existing conventional systems, the relevant substances should be prevented or reduced at source/user level, before entering the wastewater (see Figure 1). For proper disposal of the residues containing relevant MPs, user information should be intensified (Recommendation 4.4). In cases of the receiver's sensitivity, high pollution load etc., the fourth step of treatment in WWTPs is recommended and justified (Recommendation 4.1). However, a need for a uniform selection procedure of WWTPs intended for upgrade was strongly suggested to be carried at a national level. Currently, the following main criteria are suggested for use:

- ecological sensitivity of receiving water body,

- conservation of water resources for potable and leisure uses (bathing waters) – upstream responsibility,
- efficiency and cost-effectiveness criteria (such as the size and state of wastewater management facilities),
- pollution charge of receiving waters.

In 2018 the German Environment Agency (UBA) issued recommendations for reducing MPs in waters.[84] It is currently assumed that an effective implementation of source-related strategies (manufacturers' and users' responsibility) can be obtained only in the long term (>10 years). For this reason, effective removal of a broad spectrum of MPs should be achieved with the help of an advanced fourth (quaternary) treatment stage.

In Germany, 16 full-scale treatment plants in North Rhine-Westphalia and in Baden-Württemberg have currently been upgraded with a fourth treatment stage, 6 installations are currently under construction, and such treatment is planned for another 11 WWTPs. There are plans also for WWTPs in other federal states (e. g. Berlin, Bavaria and Hesse). It is supposed that the experience gained with these plants can be compared with the results of various lab-scale research projects. This is a fundamental approach for understanding both treatment effectiveness and costs. The large-scale and full-scale implementations of MP removal at German WWTPs in North Rhine-Westphalia and in Baden-Württemberg are listed in Table 10.

[84] Ahting et al. (2018) Recommendations for reducing micropollutants in waters. Ed. Helmecke M. (II 2.1) German Environment Agency Section II 2.1 General Aspects of Water and Soil. www.umweltbundesamt.de

Table 10. Large-scale and full-scale upgraded for removal of micropollutants from municipal wastewater in Germany (as of January 2019): implemented (I), under construction (UC) and under design (UD)[85, 86]. WWTPs shaded in grey (Bad Sassendorf using ozone and Dülmen using PAC) are described in more detail below. These were also included as study visits in the MORPHEUS project

	I	UC	UD	Plant size [PE]	Technology	Treated amount of wastewater
WWTP in North Rhine-Westphalia						
Aachen-Soers			x	480,000	Ozone	-
Bad Sassendorf	x			13,000	Ozone	Qmax = 300 m^3/h
Bad Oeynhausen	x			78,500	GAC	Qmax = 370 m^3/h
Barntrup	x			12,000	PAC	Qmax = 300 m^3/h
Detmold	x			135,000	Ozone	Qmax = 300 m^3/h
Duisburg Verlinden	x			30,000	Ozone	Qmax= 400 m^3/h
Dülmen		x		55,000	PAC	-
Espelkamp			x	33,000	Ozone	-
Gutersloh	x			150,600	GAC	Qmax = 840 m^3/h
Harsewinkel			x	570,000		Qmax = 300 m^3/h
Neuss Ost			x	280,000		
Obere Lutter	x			380,000	GAC	Qmax = 960 m^3/h
Rietberg		x		46,500	GAC	Qmax = 360 m^3/h
Rheda			x	94,000	Ozone	Qmax = 1,000 m^3/h
Schwerte	x			50,000	Ozon/ PAC	Qmax = 1,100 m^3/h
Warburg		x		70,000	Ozone	
WWTP in Baden-Württemberg						
Albstadt	x			125,000	PAC	Qmax = 3,500 m^3/h
Busnau		x		9,680	GAC	Qmax = 70 m^3/h
Emmingen-Liptingen	x			7,500	GAC	Qmax = 70 m^3/h
Freiburg			x	600,000		
Hechingen	x			57,200	PAC	Qmax = 1,440 m^3/h
Karlsruhe			x	700,000	PAC	
Kressbron	x			24,000	PAC	Qmax = 900 m^3/h
Lahr	x			100,000	PAC	Qmax = 1,260 m^3/h

[85] https://koms-bw.de/en/
[86] https://www.masterplan-wasser.nrw.de/das-kompetenzzentrum/

	I	UC	UD	Plant size [PE]	Technology	Treated amount of wastewater
WWTP in Baden-Württemberg						
Laichingen		x		35,000	PAC	Qmax = 540 m^3/h
Lautingen		x		36,000	PAC	Qmax = 800 m^3/h
Mannheim	x			725,000	PAC	Qmax = 1,100 m^3/h planned Qmax = 5,400 m^3/h
Ohringen			x	46,000	PAC	
Ravensburg	x			184,000	PAC	Qmax = 4,000 m^3/h
Sindelfingen	x			250,000	PAC	Qmax = 4,000 m^3/h
Stockacher Aach	x			43,000	PAC	Qmax = 900 m^3/h
Stuttgart Muhlhausen			x	1,200,000		
Ulm (Steinhaule)	x	x		440,000	PAC	Qmax = 5,000 m^3/h planned Qmax = 9400 m^3/h
Wendlingen			x	170,000	PAC	-
Westerheim			x	5,500	GAC	-

Various advanced processes are available for MP removal. But in Germany, similarly to the Swiss strategy, so far only two methods are regarded as technically feasible on a larger scale: I) oxidation with ozone, and II) adsorption onto activated carbon (PAC or GAC), or a combination of these two methods. They are feasible for plant operators, and it is assumed that, appropriately equipped and managed, WWTPs may obtain a 80% reduction in many MPs. The elimination rate is, however substance-specific and depends on the treatment technology. It is also expected that besides the MPs, ozonation and activated carbon give opportunity to enhance the removal of other organic compounds and/or to improve the hygienic quality of the WWTP's effluent. The disadvantage of these methods (except for GAC) is connected with the need for a post-treatment stage (see point 4.1).

7.2 Preliminary cost and energy consumption analysis: Germany

Financing is a key element of a Trace Substance Strategy implementation, and according to Recommendation 5 must be elaborated by the Federal Government. The final costs will depend on the final level/goal of protection that is going to be achieved. The costs are suggested to be shared between producers, distributors, water resource management institutions and citizens (as consumers). It is, however, not sure which of available instruments will be used for this purpose (taxes, existing wastewater levies, special funds, etc.).

It is assumed that, in Germany, advanced wastewater treatment may cause about 5–30% higher energy consumption over normal operation.[87] The costs depend on the size of the WWTP, the raw wastewater quality and the treatment method used. Many WWTPs have, however, considerable potential for energy savings or production. Nonetheless, it was assumed by the German Environment Agency (UBA) that additional costs for the expansion of all 230 large WWTPS in Germany (PE>1,000,000, German size category GK 5), which treat 50% of total wastewater in Germany, will cost in total 10.4 to 10.9 billion Euro over a period of 25 years. According to the above estimation, from 415 to 435 million Euro *per annum* will be spent for the elimination of MPs, including post-treatment.[88] The UBA calculates that the additional average costs for the expansion of a large WWTP will be around 16 euros per person per year.

Other costs connected with the manufacturers' responsibility, such as environmental risk assessments, large-scale informational and awareness-raising campaigns, increased research for eco-label substitutes, data provision and industrial wastewater treatment, as well as users' responsibility (substituting certain MPs with eco-label products) have not been quantified so far. Nonetheless, it is suspected that the federal government ensures that manufacturers and marketers of medicinal products and other MPs will be adequately involved in the financial responsibility of removing MPs from the aquatic environment, according to the "polluter pays" concept.

7.3 Full-scale case studies: Germany

As part of the MORPHEUS project, two German WWTPs, in Bad Sassendorf and Dülmen, with full-scale advanced treatment of pharmaceuticals in operation were visited, as indicated in Table 10 – one applying ozonation (Bad Sassendorf) and one utilising PAC (Dülmen), which are described in some detail below. The "Kompetenzzentrum Mikroschadstoffe.NRW" provides profiles on the wastewater treatment plants in North Rhine-Westphalia on their website.

German Example 1: Full scale ozonation system: WWTP Bad Sassendorf in Lippeverband[89]

WWTP Bad Sassendorf in Lippeverband is a one-stage, conventional mechanical–biological wastewater treatment plant, as shown in Figure 13.

[87] UBA (2015): Organische Mikroverunreinigungen in Gewässern – Vierte Reinigungsstufe für weniger Einträge. https://www.umweltbundesamt. de/publikationen/organische-mikroverunreinigungen-in-gewaesser

[88] Hillenbrand et al. (2016) Measures to reduce micropollutant emissions to water – Phase 2. de/sites/default/files/medien/377/publikationen/ mikroschadstoffen_in_die_gewasser-phase_2.pdf

[89] ARGE TP6 2011, Elimination von Arzneimittelrückständen in kommunalen Kläranlagen; Schlussbericht Phase 1 (http://www.lanuv.nrw.de/wasser/abwasser/forschung/pdf/Arzneimittelr_Abschlussbericht.pdf)

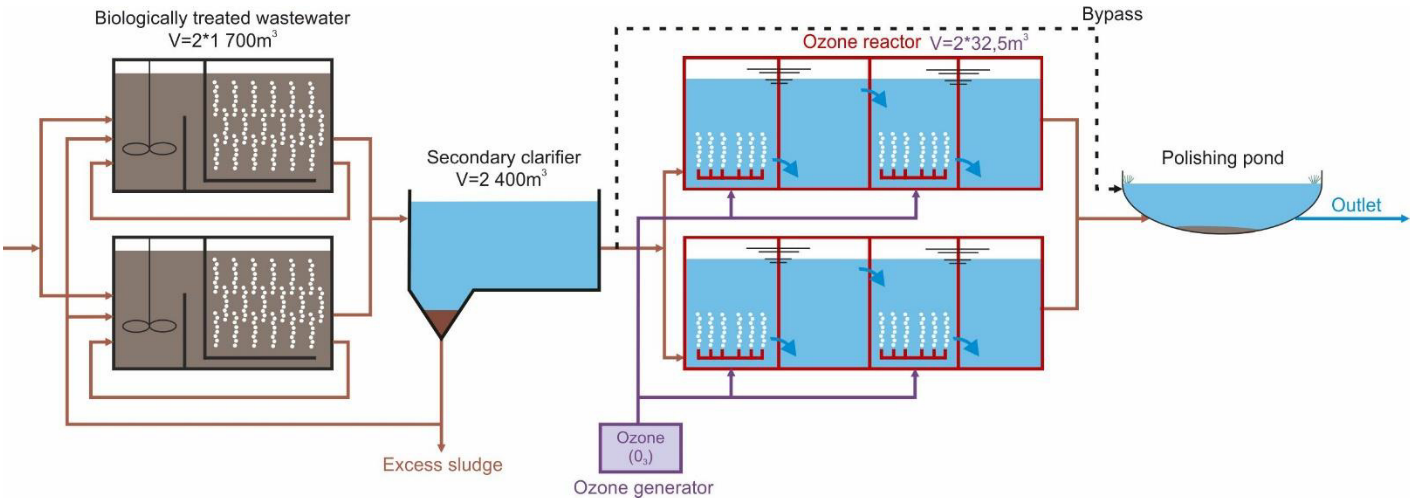

Figure 13. Scheme of the ozonation system at WWTP Bad Sassendorf in Lippeverband (adapted from[90])

The ozonation was installed in 2009 and the unit is implemented after the secondary clarification, and followed by post-treatment in a polishing pond, discharging to Rosenaue river. Some basic characteristics of the WWTP are listed below:

- ***WWTP characteristic***: plant size: 13,000 PE; flow range: Q = 300–650 m³/h; annual amount of treated wastewater: 1.7 million m³ per year;
- ***raw wastewater characteristic***: COD <60 mg/L; N_{total} <18 mg/L; P_{total} <2 mg/L;
- ***ozonation unit characteristic***: ozone system, manufacturer: Xylem (Wedeco); ozone generator, type: SMO 500 with ceramic diffusers; reaction volume: 2 × 32.5 m³ = 65 m³; ozone dosage: 5–15 mg O_3/L; Control alternatively via: Q SAK254;
- ***post treatment***: polishing pond;
- ***costs***: the investment costs for the construction of the ozonation plant amounted to about 1.0 million EURO net. The operating costs depend on the ozone consumption and are currently being determined;
- ***removal efficiency of MPs*** is substance- and dose-dependent, and varies as shown for indicator MPs in Table 11.

[90] ARGE TP6 2011, Elimination von Arzneimittelrückständen in kommunalen Kläranlagen; Schlussbericht Phase 1 (http://www.lanuv.nrw.de/wasser/abwasser/forschung/pdf/Arzneimittelr_Abschlussbericht.pdf)

Table 11. WWTP Bad Sassendorf: removal of selected indicator MPs by ozonation using an ozone dose of 2 mg O_3/L.

Substance	Medication	Removal rate
Pharmaceuticals		
Carbamazepine	Epilepsy and neuropathic pain	86–87%
Diclofenac	Pain and inflammatory diseases (gout)	91–92%
Metoprolol	Beta blocker	26–34%
Sulfamethoxazole	Sulfonamide antibiotic[1)]	77–79%
other MPs		
Bisphenol A	Xenoestrogen, Oestrogen-mimicking, hormone-like properties	-15–53%

[1)] used in combination with trimethoprim

Pictures of the ozonation system at WWTP Bad Sassendorf in Lippeverband, taken during the MORPHEUS study visit in April 2018, are shown in Figure 14.

Figure 14. Pictures of the ozonation system at WWTP Bad Sassendorf in Lippeverband, taken during the MORPHEUS study visit in April 2018. The removal of selected MPs' indicators is shown in Table 11. Photo: Erland Björklund.

German Example 2: Full-scale PAC adsorption system: WWTP Dülmen in Lippeverband[91]

WWTP Dülmen in Lippeverband is a single-stage, conventional mechanical–biological wastewater treatment plant as shown in Figure 15.

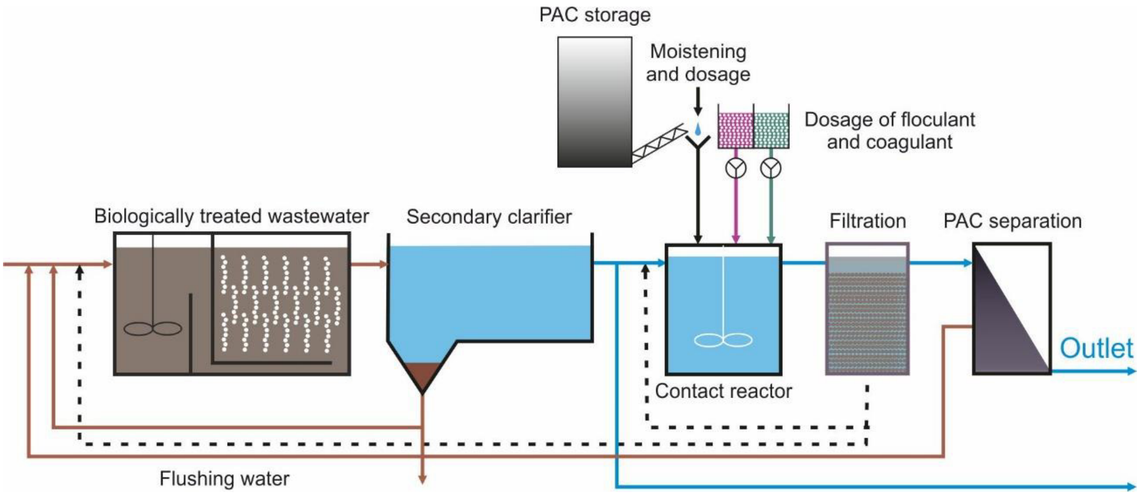

Figure 15. Scheme of the PAC system at WWTP Dülmen (adapted from [92])

The PAC unit consists of a contact basin (Vges. = 270 m^3), two converted filter cells, a newly built sedimentation basin (A = 360 m^2, V = 1440 m^3) and the remaining filter system (three filter cells). Before discharging into the receiving river Tiberbach, the clarification outflow is treated in the adsorption step for removal of MPs. Some basic characteristics of the WWTP are listed below:

- ***WWTP characteristic***: plant size: 55,000 PE; flow range: Q = 450–720 m³/h; annual amount of treated wastewater: 3 million m³ per year.
- ***raw wastewater characteristic***: COD <60 mg/L; N_{total} <18 mg/l; P_{total} <1 mg/L
- ***applied PAC technology***: PAC storage and dosage system; PAC contact basin (V = 270 m^3), residence time: 22–150 min; PAC dosage from 10 to 20 mg /L; Sedimentation basin: V = 1470 m^3 (area = 370 m^2); three residual filter cells; filter type: two-layer spatial filtration (area per filter cell 28 m²); Filtration speed: 7.5–13 m/h; treated wastewater flow: Q = 30–200 dm³/s
- ***costs***: the investment costs for the construction and adaptation of existing WWTP to PAC system was 4.0 million Euro.
- ***removal effectiveness of MPs*** are given in Table 12.

[91] ARGE TP6 2011, Elimination von Arzneimittelrückständen in kommunalen Kläranlagen; Schlussbericht Phase 1 (http://www.lanuv.nrw.de/wasser/abwasser/forschung/pdf/Arzneimittelr_Abschlussbericht.pdf)
[92] ARGE TP6 2011, Elimination von Arzneimittelrückständen in kommunalen Kläranlagen; Schlussbericht Phase 1 (http://www.lanuv.nrw.de/wasser/abwasser/forschung/pdf/Arzneimittelr_Abschlussbericht.pdf)

Table 12. WWTP Dülmen: removal of selected indicator MPs by PAC (PAC dose 10 mg /l); n=10[93]

Substance	Concentration inflow WWTP [µg/L]	Elimination rate before dosing PAC [%]	Elimination rate after dosing PAC (outflow WWTP) [%]
1H-Bezotriazol	16.9	52±16	93±4 (95±3)
Carbamazepine	0.34	-23±20	88±6 (90±5)
Diclofenac	2.92	-2±19	81±10 (82±9)
Metoprolol	1.32	40±12	97±2 (98±1)
Clarithromycin	0.43	24±22	91±6 (92±4)
Sulfamethoxazole	0.49	45±27	72±11 (74±12)

Pictures of the PAC system at WWTP Dülmen in Lippeverband, taken during the MORPHEUS study visit in April 2018, are shown in Figure 16.

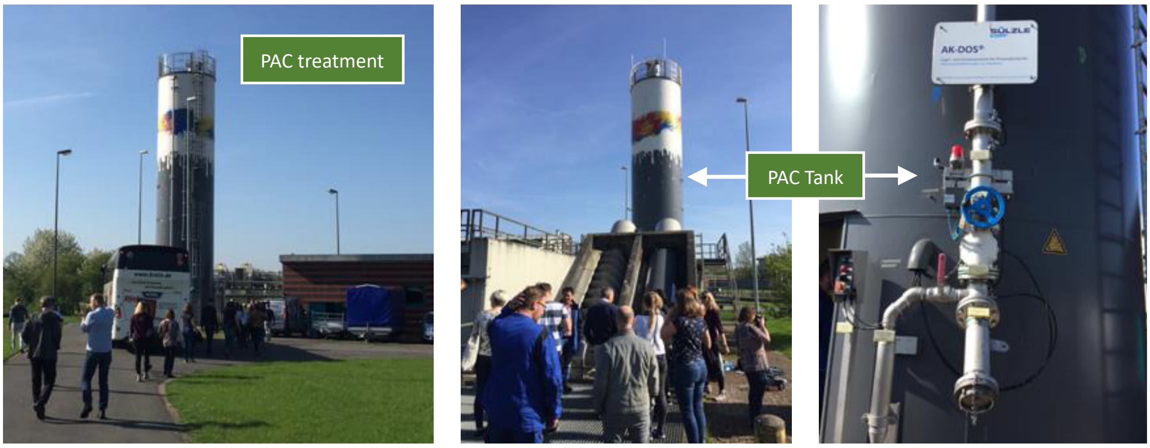

Figure 16. Pictures of the PAC system at WWTP Dülmen in Lippeverband, taken during the MORPHEUS study visit in April 2018. The removal of selected indicator MPs is shown in Table 12.
Photo: Erland Björklund

7.4 Overview comparison of ozonation and PAC in Germany

Based on the relatively large number of studies performed primarily on ozonation and PAC there are several documents available that make it possible to compare the two technologies from a more general point of view. An overview of the experiments made in Germany is shown in Table 13 by comparing system costs and design criteria.

93 EGLV, Presentation Sven Lyko in November 2018 at “Verfahrenstechnische Möglichkeiten für die Umsetzung einer 4. Reinigungsstufe und großtechnische Betriebserfahrungen” in Prenzlau, Germany

Table 13. Overview of comparison of system costs and design criteria for ozonation and PAC during implementation in German WWTPs[94,95,96,97]

Criteria	Ozonation	PAC
Primary Energy demand	0.09–0.37 kWh/m³	0.05–0.08 kWh/m³
Primary Energy demand (production and transport)	0.03–0.09 kWh/m³	0.36–0.72 kWh/m³
CO_2 emission	60–130 g CO_2/m³	150–240 g CO_2/m³
Yearly costs	0.02–0.14 €/m³	0.04–0.20 €/m³
Operation	High degree of automatisation	Low degree of automation
Space requirement	Low	High for contact and sedimentation tanks, low for dosing into filters or activated sludge reactor
Advantages	Slight disinfection	Loaded carbon coal (incinerator)
Disadvantages	Transformation products, biological follow-up treatment required)	Expanding the sludge amount, potentially PAC-drifting into receiving water, increasing CO_2 emissions

94 https://www.umweltbundesamt.de/sites/default/files/medien/377/publikationen/mikroschadstoffen_in_die_gewasser-phase_2.pdf
95 https://www.umweltbundesamt.de/sites/default/files/medien/378/publikationen/texte_85_2015_massnahmen_zur_verminderung_des_eintrages_von_mikroschadstoffen_anhang.pdf
96 http://www.lawa.de/documents/Uml242016_20160126_LAWA_Bericht_Mikroschadstoffe_in_Gewaessern_final_207.pdf
97 https://www.umweltbundesamt.de/sites/default/files/medien/378/publikationen/texte_47_2015_revision_der_umweltqualitaetsnormen_der_bundes-oberflaechengewaesserverordnung_2.pdf

8 Swedish strategy

8.1 Current situation: Sweden

In Sweden, a number of research and development projects dealing with pharmaceuticals in the environment have been carried out since 2005. Not all of these will be covered, but Figure 17 gives an overview of four major national projects run during 2005–2019 that together will have spent roughly 22 million Euro by the end of 2020.

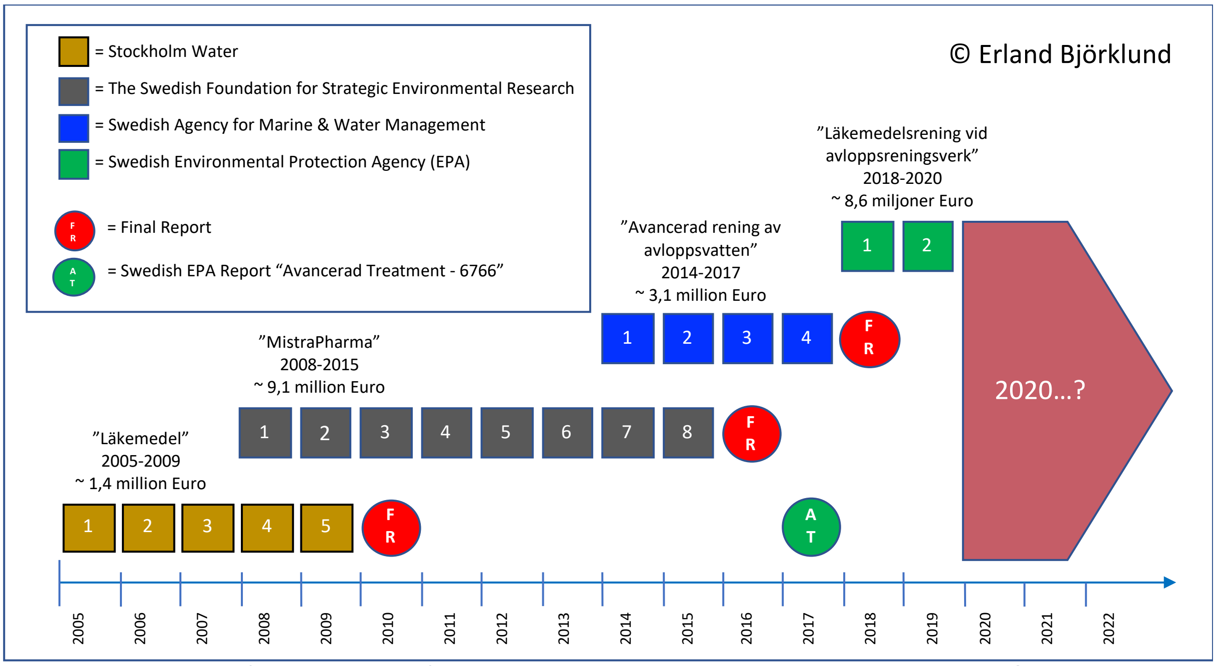

Figure 17. Examples of major research funding related to pharmaceuticals in the environment, Sweden, 2005–2018

From 2005 to 2009, a local but large Swedish project was run by Stockholm Water called "Pharmaceuticals - occurrence in the aquatic environment, preventive measures and possible treatment methods" (Figure 17). The final report was published in 2010.[98] In the project various complementary methods were tested based on either biological, oxidizing or separating principles and it was found that several of these methods worked well for removing pharmaceuticals from wastewater. However, ozone or activated carbon were the most promising technologies from a holistic perspective. The main results were published separately in a technological magazine[99] where it was stated that "*Activated carbon is the best, ozone second best.*" This was based on the fact that purification was most comprehensive with new activated carbon, which adsorbed 46 substances to 98%. In the study, however, it was observed that the cleaning effect dropped slightly as the carbon became saturated.

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[98] Wahlberg et al. (2010) Läkemedelsrester I Stockholms vattenmiljö -Förekomst, förebyggande åtgärder och rening av avloppsvatten, Stockholm Vatten 2010, 140 pages.
[99] Karlsson-Ottosson (2009) Aktivt kol bäst mot medicinrester, NyTeknik 2009-12-10. https://www.nyteknik.se/industri/aktivt-kol-bast-mot-medicinrester-6407880

Additionally, the study ranked the various techniques with a scoring system based on how the toxicity of wastewater decreased compared to having only one sand filter as the last step in the purification and then came to the following ranking:

- Active carbon: 10 points
- Ozone (5 mg/L): 9 points
- Ozone (15 mg/L): 8 points
- MBBR Biofilm system: 3 points
- MBR Membrane Bioreactor: 1 point

In the report by Stockholm Water the following statement was made: *"Since some of the supplementary purification methods can give rise to oxidation products that risk being harmful to the aquatic environment, ecotoxicological tests were carried out on the wastewater. Fish, algae, crustaceans and bacteria were exposed to wastewater purified by the various techniques. The collective researchers' overall assessment showed that activated carbon was the best treatment method followed by ozonation. But the ozone dose should not exceed 5g/m^3. At higher doses, more adverse effects occur in the aquatic organisms studied."*

It is noteworthy that already in 2009, from a Swedish perspective, it was stated that activated carbon had both physicochemical and ecotoxicological benefits. This is probably due to the different mechanisms by which the two technologies reduce the presence of pharmaceutical residues in water. Activated carbon adsorbs the substances so that they can no longer be detected by the analytical chemical technique (LC-MS/MS) since they are physically separated from the water phase. Ozone, on the other hand, converts the substances into new "unknown" chemical compounds with "unknown" effects on organisms. Admittedly, the pharmaceuticals can no longer be detected with LC-MS/MS after treatment with ozone (which is chemically very reactive), but this is not the same as there being no active molecules left in the water after this oxidative process. Despite several advantages of activated carbon and an uncertainty associated with ozonation from an ecotoxicological perspective, ozone became the technology that was ultimately recommended in the report by Stockholm Water 2010. This was based on cost calculations for both technologies according to the following formulation: "*Ozonation and activated carbon show the most promising results. The cost of the activated carbon filters is about six times higher than for ozonation. In terms of resource use, low-dose ozonation is the main alternative for reducing drug residues. Activated carbon requires more resources in the form of new coal.*" The cost aspects are further discussed in section 8.2 below, but it should be stressed that costs associated with activated carbon have decreased substantially in the past decade as the technology has been further developed, making it a competitive alternative to ozone.

Between 2008 and 2015, the Swedish Foundation for Strategic Environmental Research funded a project called MistraPharma with a total of 9.1 million Euros (Figure 17).[100] MistraPharma was at the time one of the world's largest research programmes in the field and for eight years several Swedish research groups worked on the following topics:

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[100] https://www.mistrapharma.se

- identifying human drugs that can pose a problem to aquatic ecosystems,
- addressing the risk of developing antibiotic resistance in the environment,
- proposing risk management strategies in the form of improved regulatory testing requirements,
- suggesting recommendations for improved waste water treatment technology.

The projects resulted in a large number of scientific articles on a great variety of topics, and a final report published in 2016.[101] The key outcome of the research was a policy brief with ten recommendations for improving environmental risk assessment,[102] but part of the research was devoted to wastewater treatment technologies. According to the final report various pilot plants for ozonation and activated carbon (GAC and PAC) were constructed.

The key finding for ozone was that with an appropriate ozone dose of 5–7 g O_3/m^3, ozonation reached 85–95% removal efficiency, with lower biomarker responses than today's effluent. However, sand filter treatment after ozonation did not improve the removal of pharmaceuticals.

Key findings for activated carbon was that PAC and GAC systems showed the highest removal of pharmaceuticals, 95–98%. The dose of activated carbon was in the range of 15–70 g prod./m^3. In PAC systems, the activated carbon consumption was typically one half to one third that of GAC systems. This was explained by diffusion limitations and less area displayed in the GAC filters. An important result was also that without a final sand filter at the treatment plant prior to the GAC filter, the uptime for GAC was limited. Consequently, to increase the uptime for the GAC filters a pre-treatment, in the form of sand filters, was built ahead of the pilot plant.

According to the report, several attempts were also made to evaluate the ecotoxicity. Biomarker response in rainbow trout exposures showed that both GAC filtration and ozonation reduced biomarker responses as compared to the positive control using effluent from the treatment plant. Additionally, ozonation of the water did not result in any increased oxidative stress response in the fish. Biotests have also been performed on *Daphnia magna* but data analysis is not ready at the time of reporting. Growth inhibition tests performed on algae showed mixed results, and the reader is referred to the final report for details.

As a logical continuation of the above two projects, the Swedish Agency for Marine and Water Management, on behalf of the Swedish government, between 2014 and 2018 funded eight projects with a total of 3.1 million Euro (Figure 17). The aims were to recognise the presence of MPs in the environment, and to develop and test advanced treatment technologies that effectively could limit the discharge of MPs into the aquatic environment. An English version of the final report was published in 2018.[103] Six of the projects were directly working with research and development on advanced wastewater treatment:

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[101] Identification and Reduction of Environmental Risks Caused by Human Pharmaceuticals MistraPharma Research 2008–2015 Final Report 2016.
[102] https://www.mistrapharma.se/outcomes/policy-brief-27166372
[103] Cimbritz M & Mattsson A. (2018) 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. Swedish Agency for Marine and Water Management report 2018:7

1. Pharmaceuticals in source-separated blackwater and faecal sludge – Treatment and risks – LäK
2. Removal of pharmaceutical residues using ozonation as an intermediate process step at Linköping WWTP, Sweden
3. Full-scale treatment of micropollutants – FRAM
4. Sustainable treatment systems for the removal of pharmaceutical residues and other emerging substances – SystemLäk
5. Evaluation of advanced full-scale treatment
6. Treatment of persistent contaminants in wastewater – RESVAV

In these projects ozonation and/or activated carbon were the chief technologies evaluated, but with a strong tendency towards ozonation. Two of these projects are described in section 8.3 below as Swedish examples: Project #2 using ozonation and Project # 3 using filtration through GAC (section 8.3).

Similarly as in Switzerland and Germany, the main reasons to perform the projects were the precautionary principle in protecting the aquatic environment and the desire to be able to reuse wastewater for irrigation and potable purposes. Conducted at different scales, the projects proved that MPs present in wastewater are easily-, slowly- or non-biodegraded by current wastewater treatment systems used in Sweden, which are mainly based on activated sludge technology supported by chemical precipitation. It was also concluded that different measures are needed to limit the fate of pharmaceuticals and other persistent MPs in the environment, including the previously mentioned end-of-pipe strategy by supplementing WWTPs with advanced treatment methods, as well as by promoting environmentally friendly chemical substances and safe MP disposal. Most importantly though, was the conclusion that techniques are now available that can be implemented at municipal wastewater treatment plants in order to remove MPs, including pharmaceuticals, from wastewater. The question of where advanced treatment should be implemented had no clear answer, but it was concluded that it will depend on various factors. It was stated that both knowledge and operating experience of various technical solutions are available to tackle a number of different scenarios. Additionally, the techniques were evaluated in close cooperation with staff at WWTPs all over Sweden, which is a prerequisite to be able to evaluate the techniques in a credible manner.

At about the same time as the advanced treatment projects were running, in December 2015, the Swedish Government commissioned the Swedish EPA to investigate whether it was possible to introduce advanced treatment of wastewater to hinder the discharge of pharmaceuticals to aquatic environments. The report should include potential advantages and disadvantages of various techniques and other possible consequences of their use. The results were presented in 2017 in a final report "Advanced treatment 6766" (Figure 17).[104] It was estimated that in Sweden about 90% of the discharges originate from WWTPs of capacity greater than 2,000 PE. A conventional WWTP consists of mechanical, chemical and biological stages. In Sweden, biological treatment takes place at wastewater treatment plants in the form of both activated

[104] Swedish Environmental Protection Agency Report 6766 (2018) Advanced wastewater treatment for separation and removal of pharmaceutical residues and other hazardous substances - Needs, technologies and impacts

sludge systems and biofilm systems. The most common biological method involves treatment in various types of active sludge systems. In these systems, it has been demonstrated that some pharmaceuticals are more effectively degraded in systems with nitrogen removal. It should be noted however that WWTPs in the north of Sweden do not use biological treatment, due to the lack of requirements for nitrogen removal (governed by regulations NFS 2016:6[105]). Some WWTPs that use biofilm at the biological treatment stage and/or can extend sludge age and retention time (e.g. membrane bioreactor, MBR, moving bed biofilm reactor, MBBR, and biologically active filters, BAF) reported promising results connected with a higher removal rate for some MPs compared with conventional activated sludge systems.[106]

In the EPA report it was also established that pharmaceutical emissions can be prevented by equipping Swedish WWTPs plants with more advanced technology, such as carbon filters or ozone treatment. It was concluded that the next step should be an investigation of where the technology primarily should be introduced. However, existing Swedish data on the environmental occurrence of pharmaceuticals was not sufficient to specify this. The Swedish EPA concluded that before implementing additional treatment steps for pharmaceutical residues and other unwanted substances, the following local conditions should be considered:

- The amount of pharmaceutical substances and other persistent pollutants released into receiving waters
- The water recharge rate of the receiving waters, where the receiving waters with low initial dilution and low water renewal are more likely to reach the threshold values as stated in the specific pollutant criteria and impact levels
- The presence of several treatment plants that discharge to the same receiving water body
- The receiving water body's sensitivity, such as ecological sensitivity
- Fluctuations in water recharge rate over the year in the receiving waters, and variations in effluent volumes from the wastewater treatment plant

The journey of identifying where action should be taken is now even more important since, during the fall of 2018, via the Swedish EPA, the Swedish Government moved forward and launched another 8.6 million Euros to finance pre-studies and infrastructure installations for "Treatment of pharmaceutical residues" at selected Swedish WWTPs during 2019–2020 (Figure 17). Notably, the funding was now NOT intended for research projects but should be applied for by Swedish municipalities wanting to try out new technologies as a direct pre-cursor to upgrading their WWTPs with advanced technologies. The distribution of this recent funding in Sweden is shown in Table 14. The funding is divided into two parts, either funding for smaller local pre-studies or funding in direct investments in new infrastructure. In short, funding in Sweden has now slowly turned from research-based to application-based projects.

[105] Regulation NFS 2016:6, Swedish Environmental Protection Agency
[106] Allard A.-S. & Wahlberg C. (2017) Förekomst och reduktion av fokusämnen i fyra reningsverk. Delrapport SystemLäk projekt. IVL Swedish Environmental Research Institute, Report B2279.

Table 14. Funding for infrastructure investment pre-studies in Sweden from the Swedish Government distributed via the Swedish EPA in November 2018. The total amount distributed was 84,956,108 SEK. Funding directed to projects in Region Skåne, South Baltic area are shown in grey

Pre-studies	**Amount**	**Infrastructure**	**Amount**
Borlänge Energi	1,035,000	Kristianstad Kommun	11,070,000
Boråsenergi	549,900	Lidköping Kommun	13,494,972
Falu Energi & Vatten AB	3,009,600	NSVA AB (H+)	6,033,600
NSVA AB Öresundsverket	1,000,000	Simrishamns Kommun	19,124,100
NSVA AB Lundåkraverket	1,000,000	Tierps Miljö & Energi AB	10,440,000
Syvab	2,097,000	Östra Göinge Kommun	9,450,000
VA Syd	1,607,400		
Vivab	2,214,936		
Växjö Kommun	2,559,600		
Örebro Kommun	270,000		
Total to pre-studies	15,343,436	**Total infrastructure**	69,612,672
Total to Region Skåne	3,607,400 (23.5%)	**Total to Region Skåne**	45,677,700 (65.6%)
Overall investments: 84,956,108			
Overall to Region Skåne: 49,285,100 (58.0%)			

In the MORPHEUS project Sweden is represented by Region Skåne, which is also a Swedish region of interest from a South Baltic perspective. Despite Skåne only representing 13.1% of the Swedish population and 2.5% of the Swedish area, it can be seen from Table 14 that a majority of the national funding has landed in Skåne – a total of 49 million SEK, corresponding to 58% of the entire budget. The distribution of this funding is visualised in Figure 18.

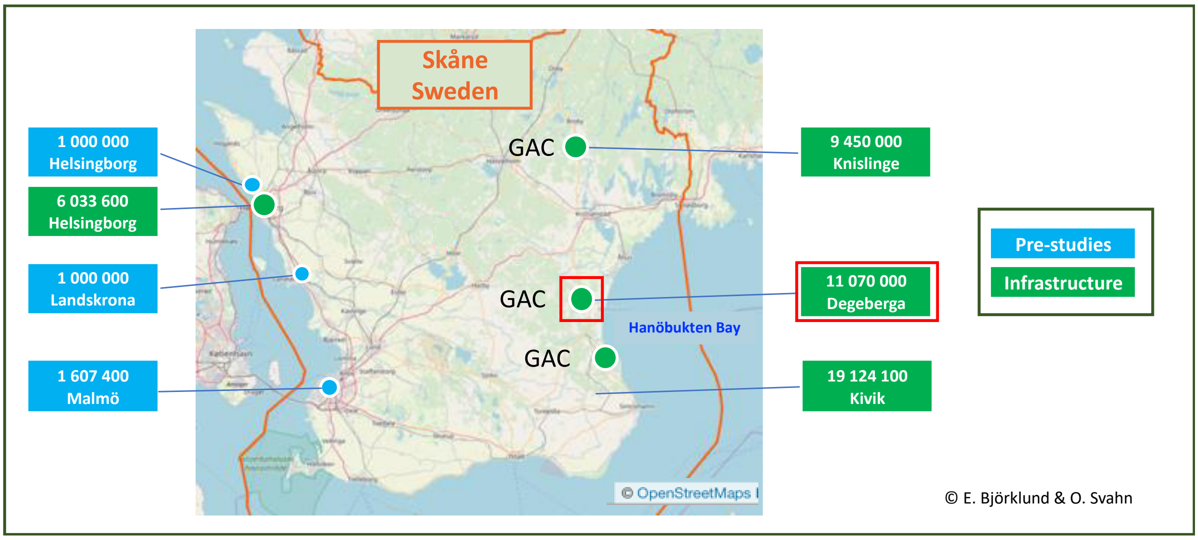

Figure 18. Major investments (in SEK) in advanced MP treatment technologies in Skåne, 2018–2020, funded via the Swedish EPA 2018. Highlighted in red: Degeberga WWTP (part of the MORPHEUS project). Knislinge, Degeberga and Kivik WWTP will all introduce filtration through GAC, as indicated.

It is worth noting that most of the large investments in infrastructure are located in eastern Skåne (Knislinge WWTP, Degeberga WWTP, Kivik WWTP), which is directly or indirectly connected to Hanöbukten Bay in the Baltic Sea (Figure 18). This bay has for decades suffered from severe unsolved environmental problems such as wounded and declining fish,[107,108] and it is therefore a great step that some of the treatment plants will decrease their MP burden in this region. At the time of writing this report it was not known which technologies should be implement at all sites. However, after contact with personnel at the three WWTPs in eastern Skåne it became clear that they all intend to introduce large-scale filtration through granulated activated carbon (GAC). Of these, Degeberga WWTP has been monitored for its release of pharmaceuticals into Segeholmsån river and Hanöbukten Bay during 2017 and 2018 as part of the MORPHEUS project (see Deliverable 4.1). These MORPHEUS occurrence data were also part of the application for funding submitted by Kristianstad Municipality during the fall of 2018 to the Swedish EPA, which was successfully approved.

As already mentioned in Section 3.1 above, in order to harmonise the Swedish investigations on occurrence of pharmaceuticals and MPs, the Swedish Medical Products Agency in 2015 proposed a total of 22 pharmaceuticals as indicators to be monitored in water [see ref. earlier]. At about the same time, in 2014, the County Administrative Board of Scania issued a supervisory guide on drug residues in wastewater [TVL-Info 2014:12, see earlier reference] where they state: "*The County Administrative Board of Skåne also considers that sampling of pharmaceutical substances shall take place with regard to outlet wastewater from treatment plants dimensioned for more than 200 PE and upstream and downstream of the treatment plant. This applies to both municipal treatment plants and private treatment plants in industrial parks, conference facilities,*

[107] Hanöbukten – Regeringsuppdrag. Havs- och Vattenmyndighetens Rapport 2013-10-29; 107 pages
[108] Miljön i Hanöbukten 2015-2017 -finns det ett samband mellan tillståndet för fisken, dess hälsa och belastningen av miljöfarliga ämnen? Havs- och vattenmyndighetens rapport 2018:10, 81 pages

treatment centres and the like." These points are illustrated together with a fourth sampling point at the wastewater treatment plant's inlet water in Figure 19.

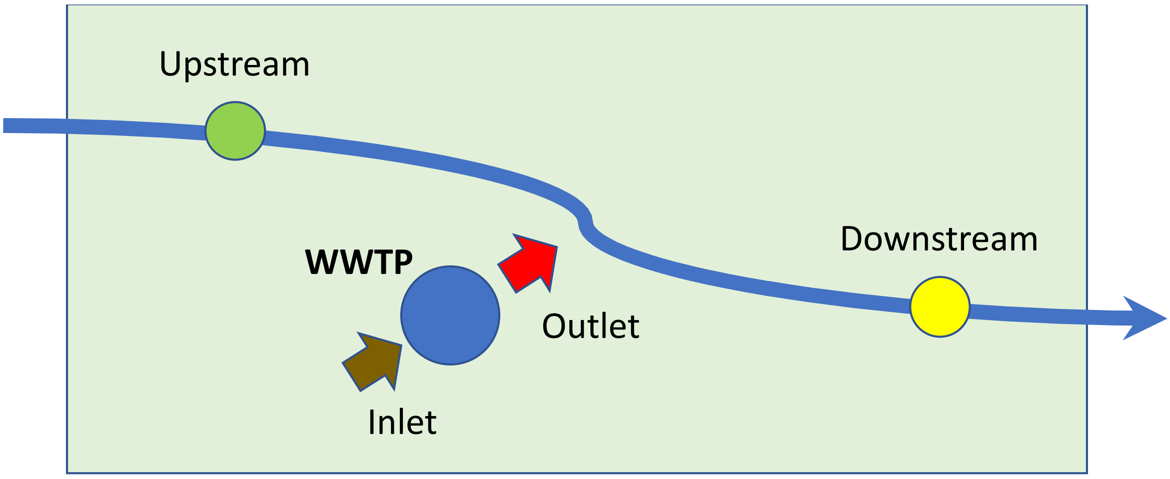

Figure 19. Three sampling points suggested by the County Administrative Board of Skåne 2014, plus an additional point at the inlet of the WWTP

Up to now very few (if any) studies in Sweden and Skåne have been conducted with such a high geographical resolution as that expressed by the County Administrative board of Scania applying a harmonised analysis protocol suggested by the Swedish Medical Products Agency. However, in two recent regional projects called LUSKA 2017[109] and KARSK 2018–2019 (ongoing), funded by Region Skåne and Kristianstad University, data are now becoming available to aid in the understanding of where to upgrade WWTPs in Region Skåne with advanced technologies. Likewise, from a South Baltic perspective, data are now also available via the MORPHEUS project, covering a total of 15 WWTPs and their recipients in Sweden (3 WWTPs), Germany (4 WWTPs), Poland (4 WWTPs) and Lithuania (4 WWTPs). These data are presented in Deliverable 4.1 of the MORPHEUS project.

8.2 Preliminary cost and energy consumption analysis: Sweden

8.2.1 Stockholm Vatten Report – 2010.

Several attempts have been made to estimate costs and energy consumption in Sweden. These are discussed here chronologically. In 2010 Stockholm Vatten made preliminary cost estimates of activated carbon and ozone. They stated that ozonation is relatively cheap, with a production cost of ca SEK 10,000/tonne at very large-scale on-site production. For a dosage of 10 g/m3, this would mean about 0.10 SEK/m3 of wastewater, but the price increases in smaller plants. Stockholm Vatten also estimated an electricity consumption of about 0.15 kWh/m3, which

[109] Svahn O. & Björklund E. (2018) LUSKA Läkemedelsutsläpp från skånska avloppsreningsverk 2017. Rapport Högskolan Kristianstad, 58 pages.

resulted in a total cost of roughly 0.6 SEK m3 for ozonation in sewage treatment plants for more than 100,000 people. For activated carbon, Stockholm Vatten wrote that the cost depends on the carbon consumption, and stated that the operating cost of activated carbon was six times greater to obtain at least as good results as ozonation. As will be seen later in this text, this price tag is today much too high as a consequence of technological development. Even so, Stockholm Vatten concluded that with a coal price of 20 SEK/kg and a dose of 120 g/m^3 to give a good treatment result, this would mean 2.4 SEK/m^3 treated wastewater. Additionally, there are costs of carbon filter basins and for disposal of the coal. The total cost of activated carbon filtration at WWTPs for more than 100,000 people was therefore estimated at 2.9 SEK/m^3, but refers to the pricing level of 2009.

Stockholm Vatten also made a rough estimate of the overall operating, investment and capital costs for Swedish conditions and showed that the added average costs for reducing pharmaceutical residues ranged between 0.6 and 19 SEK/m^3, depending on the choice of technology and WWTP size. Except for the reverse osmosis method, which is the most expensive technology, the cost would fall to between 0.6 and 12 SEK/m^3. Stockholm Vatten also estimated some energy use at the WWTPs and stated that with ozonation it would increase by 50%, and with UV/H_2O_2 by 200%. This would correspond to the amount of electricity needed to heat up between 6,500 and 26,000 villas with direct-acting electricity (20,000 kWh/villa, year).

Extrapolating the cost to all WWTPs in Sweden, the total cost was estimated to SEK 1.2–5.7 billion/year or SEK 150–750 per person per year. This was compared to the operation of all Sweden's WWTPs (water and sewage), which cost 14.3 billion in 2003. Thereby, at best, the reduction of pharmaceutical residues would cause a 10% increase on the current cost, but at worst would increase it by 40%.

8.2.2 Swedish EPA Report 6766 – 2017

The report published by the Swedish EPA in 2017 estimated both the energy consumption and the costs of introducing advanced treatment. A summary of energy and costs for a number of technologies is shown in Table 15.

One of the first things to be noted is that when comparing the two previously recommended technologies – filtration through GAC and ozonation – the price difference is no longer as dramatic as previously indicated by Stockholm Vatten. Additionally, the energy consumption during operation is also much lower for a GAC filter than for an ozonation unit. Table 15 also shows that the implementation of any type of advanced treatment will result in an increase in energy consumption. For ozonation and UF the increased energy usage is mainly related to operation, while for PAC and GAC it is mainly the production of activated carbon that demands energy.

Supplementary PAC and GAC systems were estimated to result in increased energy consumption of about 2–10% (1–6 kWh/PE/yr), at large WWTPs of >100,000 PE, while ozonation would increase energy consumption by about 20–60% (10–36 kWh/PE/yr), and finally a UF step would require up to 100% more energy (60 kWh/PE/yr). This last energy amount is equal to 3,000 villas with direct effect electricity for one year (assuming that 20,000 kWh/yr is needed). In general, larger WWTPs were also stated to be more energy efficient than smaller ones.

Table 15. Estimated costs and energy consumption for advanced treatment technologies in Sweden as published by the Swedish EPA in 2017.[110] These values were based on original data by Baresel and co-workers

Treatment technology								
Number of PE	**UF**	**GAC**	**PAC**	**BAF**	**O_3**	**PAK-UF**	**O_3-BAF (GAK)**	**UF-BAF (GAK)**
Total cost [SEK/m³]								
2,000	3.5–4.5	1.0–1.2	1.1	1.0–1.20	0.55–0.90	5.3	1.5	4.5–5.7
10,000	1.0–1.5	0.80–1.0	0.70	0.70–1.0	0.25–0.55	2.1	1.1	1.7–2.5
20,000	0.70–1.1	0.70–1.0	0.60	0.50–0.80	0.23–0.35	1.6	0.75	1.2–1.9
100,000	0.50–0.75	0.50–0.70	0.57	0.35–0.60	0.19–0.20	1.3	0.50	0.80–1.4
500,000	**0.40–0.65**	0.30–0.60	0.55	0.20–0.50	0.14–0.15	1.2	0.40	0.60–1.2
Energy consumption during operation (kWh/m³)								
	0.1–0.5	<0.01	0.01–0.05	<0.01	0.1–0.3	0.1–0.55	0.1–0.3	0.1–0.5

UF = ultrafiltration, GAC = granulated activated carbon, PAC = pulverised activated carbon, BAF = biological active filter, O_3 = ozonation, PAC-UF = combination PAC and UF, O_3-BAF(GAC) = combination O_3 and BAF with GAC as filter material, UF-BAF(GAC) = combination UF and BAF with GAC as filter material

The Swedish EPA also attempted to extrapolate the total costs to all wastewater treatment plants in Sweden greater than 2,000 PE, as shown in Table 16.

[110] Baresel et al. (2017). Tekniska lösningar för avancerad rening av avloppsreningsvatten. IVL Svenska Miljöinstitutet, report No. C 235.

Table 16. Extrapolation of total treatment costs for introducing advanced treatment technologies in Sweden, as published by the Swedish EPA in 2017.

431 WWTPs with a total of 8,049,753 connected persons (PE)
WWTPs 2,000–10,000 PE (246 WWTPs)
150 m^3/PE/yr*678,682 PE = 101.8 million m^3/yr*0.55–5.7 SEK/m^3 Total: = 56–580 million SEK/yr
WWTPs 10,000–20,000 PE (71 WWTPs)
150 m^3/PE/yr*602,021 PE = 90.3 million m^3/yr*0.25–2.5 SEK/m^3 Total: = 23–226 million SEK/yr
WWTPs 20,000–100,000 PE (95 WWTPs)
150 m^3/PE/yr*2,542,267 PE = 381 million m^3/yr*0.19–1.4 SEK/m^3 Total: 73–534 million SEK/yr
WWTPs 100,000–500,000 PE (19 WWTPs)
150 m^3/PE/yr*4,226,783 PE = 634 million m^3/yr*0.14–1.2 SEK/m^3 Total: 89–761 million SEK/yr
ALL 431 WWTPs
Total: 241 million–2.1 billion SEK/year

As seen from Table 16 the total estimated cost has a very wide range, between approximately 241 million and 2.1 billion SEK/year. This was calculated to correspond to approximately 55 and 480 SEK/household per year. These estimates are a bit lower than those previously reported by Stockholm Vatten 2010 above. The Swedish EPA also compared the estimated total cost to the total cost of operating all Swedish water companies, including water and sewage, which amounted to a total of 17 billion SEK in 2012 (according to experts at The Swedish Water & Wastewater Association). A historical Swedish perspective also shows that 40 years ago (1979), the Swedish government invested around SEK 1.5 billion SEK for the expansion of Swedish WWTPs,[111] which corresponds to approximately SEK 5.5 billion in 2016.

[111] Naturvårdsverket (2014). Rening av avloppsvatten i Sverige 2014. ISBN 978-91- 620-8728-9.

8.3 Full-scale case studies: Sweden

Swedish Example 1: Full scale ozonation system: Nykvarnsverket WWTP in Linköping

In 2017, the first large-scale WWTP for MP removal by ozonation was completed at Nykvarnsverket WWTP in Linköping. This implementation was preceded by a pilot study, conducted to properly evaluate the dosage and effectiveness of ozonation for different MPs, with a final report published in 2015.[112] The completed pilot project showed that ozone treatment before the de-nitrification step is a viable alternative for reducing pharmaceutical residues in wastewater. Experiments showed that the optimal ozone dose to remove all investigated pharmaceuticals to such an extent that no harmful effects were observed in the recipient Stångån river, was around 0.5–0.8 mg O_3/mg DOC. In the conclusions of the report it was stated that the reduction of pharmaceuticals at Nykvarn WWTP, when applying ozonation, was higher than in previous studies at other investigated WWTPs. When adding the ozonation step before the de-nitrification step, the total flow of pharmaceutical residues could be reduced by just over 90% on average. However, the authors pointed out that the total mass flow is a very blunt instrument in evaluating the purification effect. The reduction differs very much between various pharmaceuticals and depends largely on which compounds are analysed and how these are reported. It was also noteworthy that no adverse effects were demonstrated on red algae, green algae or Nitocra in the applied ozonation dose range. Additionally, there results from conducted Ames tests indicate no increased formation of mutagenic by-products and hence no genotoxicity. Based on the results from the pilot study, a full-scale ozonation treatment unit was built.

An overview of the Nykvarnsverket WWTP in Linköping is shown in Figure 20.

The final ozonation treatment plant is today described by the operators on their webpage as follows:[113]

With the help of ozone, over 90% of the incoming pharmaceutical residues are reduced in the wastewater. The ozone is mixed into the wastewater in a reactor that holds 600 m³. The ozone reactor is designed so that all ozone can react before the water proceeds to the next purification step. In the building containing the ozonation unit, there are double safety systems that quickly shut down the ozone production if a leak is detected. In this way, a safe and good working environment and external environment is ensured around the ozone reactor. After the ozone treatment, the water is treated in a biological process that captures degradation products from the ozone treatment. In addition to removing pharmaceuticals, the combination of ozone purification and the subsequent biological purification can improve the overall purification of organic carbon and nitrogen.

[112] Sehlén et al. (2015) Pilotanläggning för ozonoxidation av läkemedelsrester i avloppsvatten, NR B 2218 Februari 2015 RAPPORT, 60 pages.

[113] https://www.tekniskaverken.se/om-oss/innovation/innovativa-projekt/rening-av-lakemedelsrester/

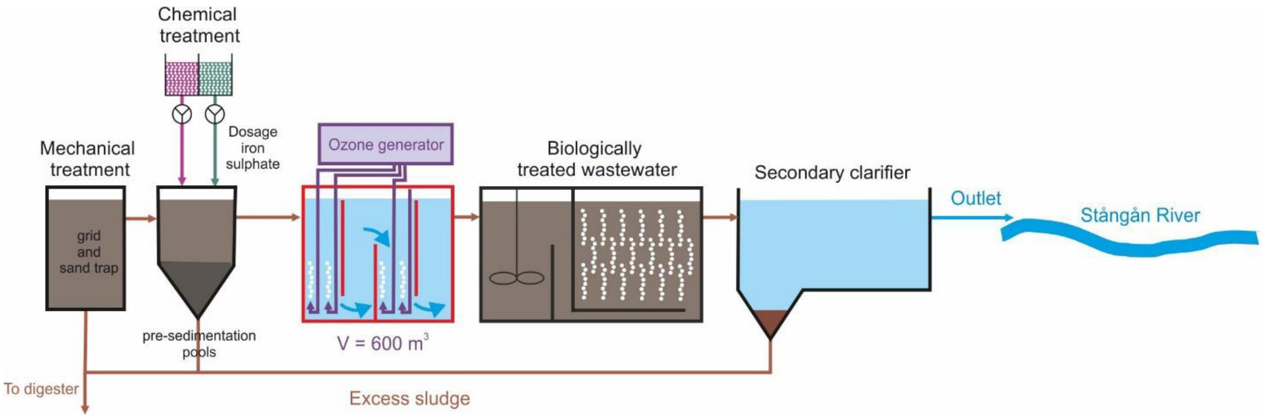

Figure 20. Scheme of the ozonation system at Nykvarnsverket WWTP in Linköping (adapted from [114])

Some basic characteristics of the WWTP before it was rebuilt could be identified in an environmental report from 2016,[115] which was the most recent report we could identify from the operator.

- ***WWTP characteristic***: plant size: 143,200 PE; flow range: Q = 1,154–3,233 m³/h; amount of treated wastewater: 13.9 million m³/yr.
- ***raw incoming wastewater characteristic***: BOD_7 431 mg/L; N_{total} 58 mg/L; P_{total} 7.3 mg/L
- ***applied technology***: Ozonation system using 0.5–0.8 mg O_3/mg DOC, and a reactor that holds 600 m³
- ***costs***: the actual cost could not be identified, but according to articles in technical magazines it has been estimated to around 25 million SEK.[116]
- ***removal efficiency of MPs***: 90% for around 40 pharmaceuticals, but with compound-dependent differences.

Pictures of the ozonation system at Nykvarnsverket WWTP in Linköping are shown in Figure 21.

[114] https://www.tekniskaverken.se/om-oss/anlaggningar/avloppsreningsverk2/
[115] Miljörapport 2016, Nykvarnsverket, Linköping, Tekniska verken i Linköping AB (publ), 62 pages.
[116] https://www.nyteknik.se/innovation/linkoping-bygger-sveriges-forsta-lakemedelsrening-6806943

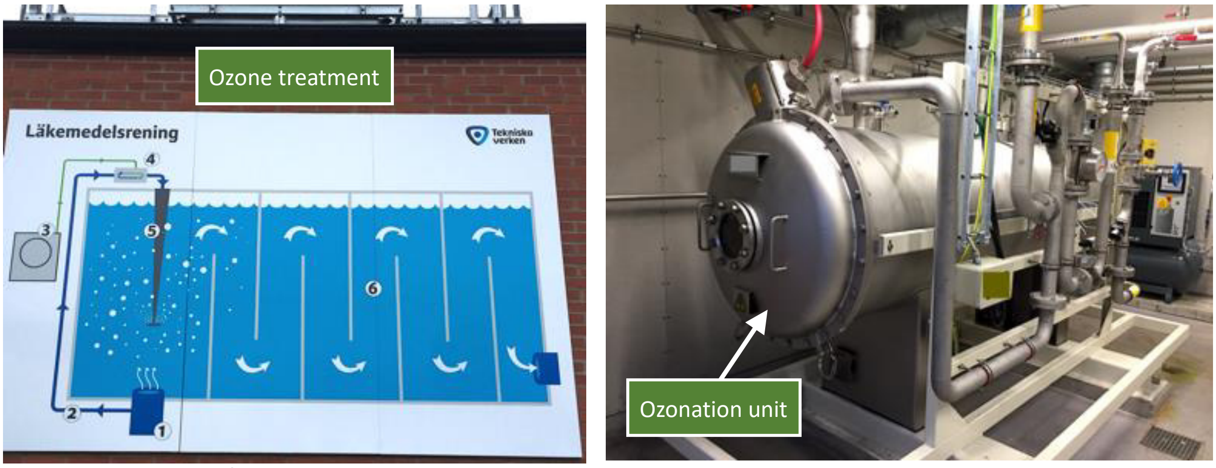

Figure 21. Pictures of the ozonation system at Nykvarnsverket WWTP in January 2018. Photo: Erland Björklund 2018

Swedish Example 2: GAC filtering system: Kristianstads WWTP in Skåne

The GAC filtering system was placed at the outlet of Kristianstad WWTP as a fourth-stage treatment step, treating a fraction of the outgoing water as shown in Figure 22.

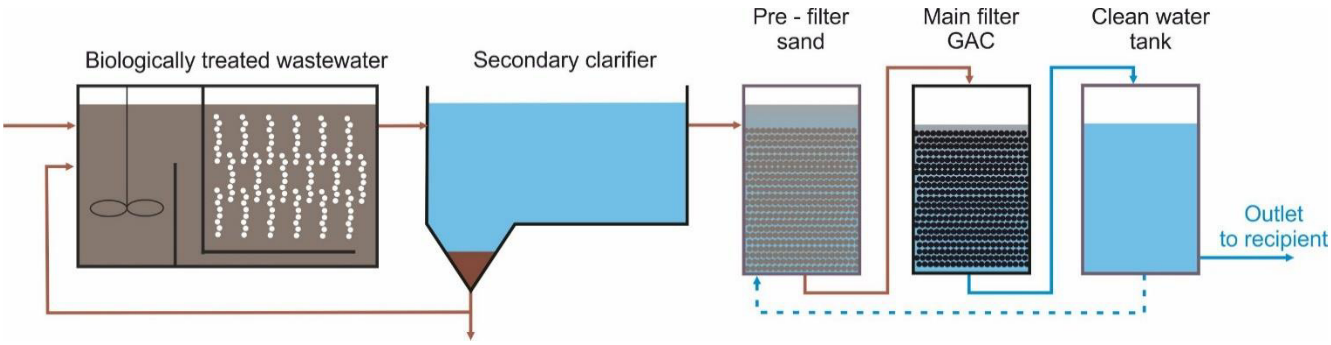

Figure 22. Scheme of the GAC filtering system at Kristianstad WWTP Sweden, adapted from final report from the Swedish Agency of Marine and Water Management 2018

The GAC unit consisted of pre-filter filled with 1 m^3 of sand to remove part of the organic material and thereby “protect” the GAC filter from high loads of such materials coming from the outlet water of the WWTP. This increased the uptime of the GAC filter. After filtration through sand, the water is pumped to the main GAC filter containing 1 m^3 of GAC (325 kg) to remove pharmaceuticals and other MPs. The flow through the system was 2 m^3/h, but the system can easily be scaled up to more or less any size. Presently, a larger version of the filter system is under construction at Degeberga WWTP (financed by the Swedish EPA, see above) which will have a flow of around 20 m^3/h. The GAC filter at Kristianstad WWTP has been in operation for more than 12 months as described in the final report by the Swedish Agency for Marine and Water Management 2018. This corresponds to more than 23,000 BV (or 23,000 m^3, since the filter volume is 1 m^3). The sand filter was back-flushed around 3–4 times a week while the GAC filter did not need any back-flushing during the entire year. Some basic characteristics of the treated outlet water at

Kristianstad WWTP according to an environmental report from 2017, which is water being treated by the GAC filter, is shown below:

- ***WWTP characteristic***: plant size: 118,000 PE; flow range: Q = 750–1,604 m^3/h; amount of treated wastewater: 8.4 million m^3/yr
- ***conventionally treated wastewater characteristics***: BOD_7 1.7 mg/L; N_{total} 7 mg/L; P_{total} 0.095 mg/L
- ***applied GAC technology***: pre-filtration through 1 m^3 sand followed by filtration through 1 m^3 of GAC. Flow: 2 m^3/h
- ***costs***: the cost for this add-on fourth stage filter was roughly 1 million SEK. Price will be reduced if applied at larger scale
- ***removal efficiency of MPs*** is in most cases more than 90% after >20,000 bed volumes (BV)

The treatment ability of the GAC filter after 20,500 BV and 23,000 BV for some selected compounds are shown in Table 17.

Depending on what compounds need to be treated and to what level they should be removed, removal may be also continued to be sufficiently effective after more than 1 year of operation. Primarily negatively charged pharmaceuticals such as sulfamethoxazole tend to break through the filter first while a majority are still being removed to more than 80% after 23,000 BV.

Pictures of the GAC filter at Kristianstad WWTP in Skåne, taken during the MORPHEUS study visit in May 2017, are shown in Figure 23.

Figure 23. Pictures of the GAC filter at Kristianstad WWTP taken during the MORPHEUS study visit in May 2017. The removal of selected indicator MPs is shown in Table 17. Photo: Erland Björklund.

Table 17. GAC filter at Kristianstad WWTP Skåne: removal of selected indicator MPs after 20,500 bed volumes (BV) and 23,000 BV.

Substance	Removal (%) 20,500 BV	Removal (%) 23,000 BV
Atenolol	93	96
Ciprofloxacin	76	68
Citalopram	99	98
Clarithromycin	95	91
Diclofenac	88	86
Erithromycin	89	86
Estrone	100	97
Fluconazole	61	53
Furosemide	89	91
Imidacloprid	97	68
Carbamazepine	84	82
Losartan	84	90
Metoprolol	96	96
Naproxen	92	93
Oxazepam	84	79
Propranolol	100	100
Sertraline	94	98
Sulfamethoxazole	26	8
Tramadol	99	89
Trimethoprim	98	98
Venlafaxine	85	82
Zolpidem	100	93

9 Polish strategy

Currently in Poland, no pilot or large-scale systems aiming to limit the discharge of pharmaceuticals and MPs into the aquatic environment are being implemented at municipal WWTPs. However, in 2015 the National Environmental Monitoring Programme for the years 2016–2020 was established, and adapts the current European strategic documents in water monitoring, in particular the need to monitor priority substances in the field of water policy as provided by Directive 2013/39/EU (for details, see Section 3 and Annex C). There is also an interest in this topic, as expressed by the participation of Polish institutions in European projects aiming to test new, cost-effective technological solutions for the removal of pharmaceuticals and other micropollutants by upgrading existing wastewater treatment systems (e.g.: MORPHEUS and LESS IS MORE). The questionnaires sent by Gdansk Water Foundation as part of the MORPHEUS project indicated that in Poland the problem with pharmaceuticals in the aquatic environment is known at a very basic level. Not many conferences/workshops are devoted to this topic, and if any does, usually only the presence of pharmaceuticals in the aquatic environment, their impact on living organisms and the potential consequences for humans are discussed. The technologies effective for pharmaceutical removal from wastewater are very rarely discussed.

10 Lithuanian strategy

Long- and mid-term environmental approaches in water management policies have been determined by two strategic documents adopted in recent years: the **National Environmental Protection Strategy** and the **Water Sector Development Programme for 2017–2023** (for details, see Section 3 and Annex C).

To improve the status of surface and groundwater bodies and to achieve and maintain good environmental status of the Baltic Sea, plans mainly involve reducing agricultural pollution (diffuse sources) and increasing treatment efficiency in 12 WWTPs (point source). However, no measures are integrated in the mentioned strategic planning documents to implement advanced treatment for the removal of micropollutants, including pharmaceuticals, in wastewater. Nevertheless, pilot investments in technological solutions for removing pharmaceuticals and other micropollutants are planned to be introduced in Kretinga town WWTP (Figure 24). This innovative approach is partly supported by the EU Interreg South Baltic programme project “LESS is MORE”.

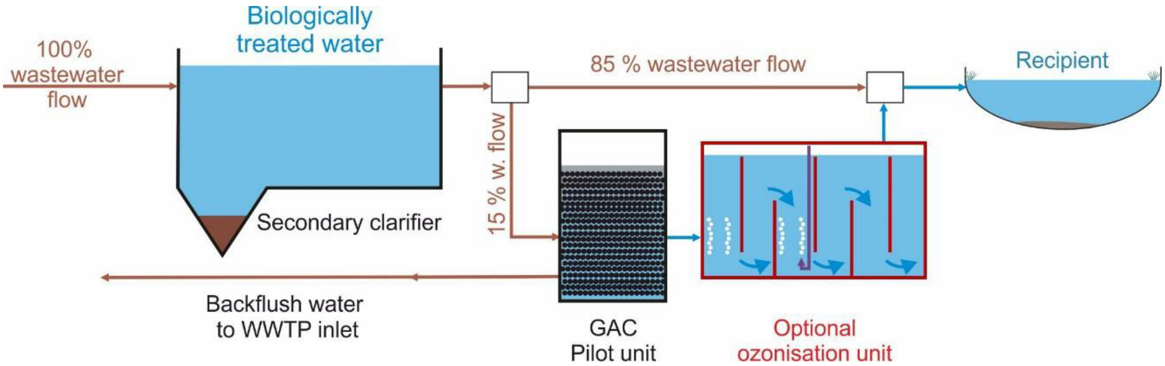

Figure. 24 Concept for pilot tests in Kretinga town WWTP (http://www.gfwlm.nazwa.pl/projekty/less-is-more/)

Substances from the Watch List are included in the Lithuanian State Environmental Monitoring Programme for 2018-2023. Monitoring of these substances is planned at four monitoring stations once per year annually. Some data of pharmaceuticals are provided for several WWTPs and surface water bodies in Western Lithuania by the current MORPHEUS project and in four rivers by the EEA Financial Mechanism supported project.

Environmental authorities will consider various options for further monitoring of pharmaceuticals and other micropollutants, as well as those included in Watch List 2 by EU Commission Implementing Decision 2018/840.

11 Conclusion: lessons learnt

WWTPs in Europe and in the South Baltic area have been recognised as relevant point sources of pharmaceuticals and other MPs. Thus, it is expected that in order to protect drinking water supplies and aquatic organisms' welfare, and to reduce antimicrobial resistance dissemination, MP control strategies should most likely be initiated by authorities and come from the government level. In Europe, such efforts have been undertaken by several countries, mainly Switzerland, but also, e.g., Germany and Sweden. From their experiences it can be concluded that:

- the goals of the MP strategy have to be clearly defined and reflect both legal obligations and practical implementation measures,
- the decision-making process should be supported by a wide range of monitoring programmes assessing the current baseline of MPs' fate in WWTPs and in the environment,
- the monitoring should combine lab tests with pilot plants and large-scale *in situ* implementation to assess the effectiveness of MP removal via different methods,
- the effectiveness of a fourth-step implementation in existing WWTPs should be evaluated according to the following criteria: (i) a sound scientific and technical basis, (ii) technical feasibility, (iii) manageability, (iv) time adaptability, and (vii) financial feasibility,
- the financing programme and cost estimation for the MPs strategy should be prepared with due diligence and be subject to public consultation,
- the MPs strategy should need broad societal and political acceptance,
- the consulting period should be carried over a long time perspective, engaging all relevant stakeholders and raising awareness and understanding of the problem among the public, scientists, practitioners and politicians,
- a wide range of actions should be undertaken, such as meetings, popular and scientific publications, advertising, and other avenues.

Currently there are several advanced treatment methods capable of eliminating a broad range of MPs from wastewater. Among them, ozonation and activated carbon are regarded as the most technically and financially feasible. It is suspected that their large-scale implementation will provide the answers to such questions as:

- What is the process's MP elimination capacity?
- How does the process influence the existing wastewater treatment system?
- Are there any undesired side effects or by-products?
- Is the technology technically and operationally applicable (including process engineering, control, and safety)?
- How much energy is required?
- What are the costs of construction and operation?

It is anticipated that even large-scale implementation of advanced treatment of wastewater will in most cases not show an immediate environmental effect, as it often requires long-term monitoring of the receiving aquatic ecosystems to observe changes. However, analysis of the WWTP outlet and surface water in terms of MP concentrations will still allow it to be ensured that critical levels of certain harmful MPs are not exceeded. Likewise, it allows for the absolute reduction in kilograms per year of analysed MPs to be calculated, which may improve the public and political perception of environmental issues. As a result, in addition to the control of MP dissemination by WWTPs (end-of-pipe measures), substituting the production and use of critical MPs may also attain wider social acceptance (source and user measures).

Annex A

List of priority substances in the field of EU water policy

No	CAS number	EU number	Compound	Directive 2000/60/EC[1]	Directive 2008/105/EC[2]	Directive 2013/39/EU[3]
1	15972-60-8	240-110-8	Alachlor			
2	120-12-7	204-371-1	Anthracene	X[4]	X	X
3	1912-24-9	217-617-8	Atrazine	X[4]		
4	71-43-2	200-753-7	Benzene			
5	n.a.	n.a.	Brominated diphenylether	X[4]	X[4]	X[4]
6	7440-43-9	231-152-8	Cadmium and its compounds	X[4]	X	X
7	85535-84-8	287-476-5	Chloroalkanes, C10-13	X[4]	X[4]	X
8	470-90-6	207-432-0	Chlorfenvinphos			
9	2921-88-2	220-864-4	Chlorpyrifos	X[4]		
10	107-06-2	203-458-1	1,2-dichloroethane			
11	75-09-2	200-838-9	Dichloromethane			
12	117-81-7	204-211-0	Phthalic acid 2-ethylhexylester (DEHP)	X[4]		X
13	330-54-1	206-354-4	Diuron	X[4]		
14	115-29-7	204-079-4	Endosulfan	X[4]	X	X
15	206-44-0	205-912-4	Fluoranthene	[4]	[4]	
16	118-74-1	204-273-9	Hexachlorobenzene	X	X	X
17	87-68-3	201-765-5	Hexachlorobutadiene	X	X	X
18	608-73-1	210-158-9	Hexachlorocyclohexane	X[4]	X	X
19	34123-59-6	251-835-4	Isoproturon	X[4]		
20	7439-92-1	231-100-4	Lead and its compounds	X[4]		
21	7439-97-6	231-106-7	Mercury and its compounds	X	X	X
22	91-20-3	202-049-5	Naphthalene	X[4]		
23	7440-02-0	231-111-4	Nickel and its compounds			
24	25154-52-3	246-672-0	Nonylphenols	X[4]	X[4]	X[4]
25	1806-26-4	217-302-5	Octylphenols	X[4]	[4]	[4]
26	608-93-5	210-172-5	Pentachlorobenzene	X	X	X
27	87-86-5	201-778-6	Pentachlorophenol	X		

No	CAS number	EU number	Compound	Directive 2000/60/EC[1]	Directive 2008/105/EC[2]	Directive 2013/39/EU[3]
28	n.a.	n.a.	Polyaromatic hydrocarbons	X[4]	X[4]	X[4]
29	122-34-9	204-535-2	Simazine	X[4]		
30	688-73-3	211-704-4	Tributyltin compounds	X[4]	X[4]	[4]
31	12002-48-1	234-413-4	Trichlorobenzenes	X[4]		
32	67-66-3	200-663-8	Trichloromethane (chloroform)			
33	1582-09-8	216-428-8	Trifluralin	X[4]		X
34	115-32-2	204-082-0	Dicofol	n.incl.	n.incl.[5]	X
35	1763-23-1	217-179-8	Perfluorooctane sulfonic acid and its derivatives (PFOS)	n.incl.	n.incl.[5]	X
36	124495-18-7	n.a.	Quinoxyfen	n.incl.	n.incl.[5]	X
37	n.a.	n.a.	Dioxins and dioxin-like compounds	n.incl.	n.incl.[5]	X[4]
38	74070-46-5	277-704-1	Aclonifen	n.incl.	n.incl.	
39	42576-02-3	255-894-7	Bifenox	n.incl.	n.incl.	
40	28159-98-0	248-872-3	Cybutryne	n.incl.	n.incl.	
41	52315-07-8	257-842-9	Cypermethrin	n.incl.	n.incl.	[4]
42	62-73-7	200-547-7	Dichlorvos	n.incl.	n.incl.	
43	n.a.	n.a.	Hexabromocyclododecanes (HBCDD)	n.incl.	n.incl.	X[4]
44	76-44-8/ 1024-57-3	200-962-3/ 213-831-0	Heptachlor and heptachlor epoxide	n.incl.	n.incl.	X
45	886-50-0	212-950-5	Terbutryn	n.incl.	n.incl.	

n.a. - not applicable, n.incl. – not included, [1)] according to Annex X of the Decision no 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 and amending Directive 2000/60/EC; [2)] according to Annex II of the Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council; [3)] according to Annex I of the Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy; [4)] for details see relevant directive; [5)] substances included in Annex III of this Directive as substances subject to review for possible identification as priority substances or priority hazardous substances (for details see Annex B in this report)

Annex B

Substances subject to review for possible identification as priority substances or priority hazardous substances in the field of the EU water policy according to Annex III of the Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC and amending Directive 2000/60/EC of the European Parliament and of the Council

No.	CAS number	EU number	Compound
1	1066-51-9	-	AMPA
2	25057-89-0	246-585-8	Bentazon
3	80-05-7	-	Bisphenol-A
4	115-32-2	204-082-0	Dicofol[1]
5	60-00-4	200-449-4	EDTA
6	57-12-5		Free cyanide
7	1071-83-6	213-997-4	Glyphosate
8	7085-19-0	230-386-8	Mecoprop (MCPP)
9	81-15-2	201-329-4	Musk xylene
10	1763-23-1	-	Perfluorooctane sulphonic acid (PFOS) [1]
11	124495-18-7	-	Quinoxyfen (5,7-dichloro-4-(p-fluorophenoxy)quinoline) [1] Dioxins[1] PCB

1) substances included in the list of priority substances in the field of water policy according to Annex I of the Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy

Annex C

Legal bases for handling micropollutants

EU Regulations	
Water Framework Directive (WFD) 2000/60/EC Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC	Objective to achieve/maintain good chemical and ecological status of waters; prohibition of deterioration; measures for reducing relevant pollutants/pollutant groups (Annex VIII of the WFD); demand to phase out priority hazardous substances
Groundwater Directive (GWD) 2006/118/EC	Specifications for good groundwater chemical status; reversal of significant and sustained upward trends in concentrations of pollutants; environmental quality standards (EQS) for pesticides and parameters for threshold values Measures for achieving/maintaining good water status and for preventing or limiting the input of pollutants
Directive 2008/105/EC on Environmental Quality Standards, amended by 2013/39/EU	Environmental quality standards (EQS) for so-called priority and priority hazardous substances (Annex X of the WFD), defining “good chemical status” with respect to surface waters. Presently, EQSs are defined for 45 substances; 12 of which have only been included in the assessment of chemical status since 2018. The list is revised every 6 years. A “watch list” is being created to facilitate the future prioritisation process.
Marine Strategy Framework Directive (MSFD) 2008/56/EU	Objective to achieve/maintain good status of the marine environment; prohibition of deterioration; measures for reducing relevant pollutants/pollutant groups
REACH Regulation (EC) 1907/2006 (Registration, Evaluation, Authorisation & Restriction of Chemicals)	Registration, evaluation, authorisation and restriction of chemicals; official evaluation of dossiers and substances ensures sufficient information is known about the substances. Official instruments for risk management exist in the form of identification of substances of very high concern (SVHC) (from an environmental perspective these would be PBT, vPvB substances and endocrine disruptors (ED)), possible authorisation requirements and restrictions.
CLP Regulation (EC) 1272/2008 (Classification, Labelling & Packaging)	Classification and labelling inventory (approximately 114,000 substances classified as hazardous)
Plant Protection Product Regulation (EC) No. 1107/2009	Authorisation, placing on the market, use and control of plant protection products. List of active substances approved in the EU.
Directive 2009/128/EC on the sustainable use of pesticides	Commitment to a sustainable, permanent, environmentally friendly use of pesticides; creation of National Action Plans for the Member States
Regulation (EU) 528/2012 on biocidal products	Authorisation of biocidal products based on an environmental risk assessment of active biocidal substances and biocidal products. List of active substances approved in the EU.

Directive 2001/83/EC (amended by 2004/27/EC) on the Community code relating to medicinal products for human use	The authorisation of human medicinal products requires testing for potential impacts on the environment. If a risk to the environment is identified, denial of authorisation is not possible; authorisation can be subjected to conditions for the protection of the environment.
Directive 2001/82/EC (amended by 2004/28/EC and 2009/9/EC) on the Community code relating to veterinary medicinal products	The authorisation of veterinary medicines requires testing for all possible impacts on the environment. If a risk to the environment is identified, authorisation can be denied or be subjected to conditions for the protection of the environment.
Regulation (EC) No. 726/2004 on Community authorisation procedures and establishing a European Medicines Agency	Additional legal requirements for the authorisation of new human and veterinary medicinal products
Regulation (EC) No. 648/2004 on detergents	Regulates complete aerobic biodegradation of surfactants and derogations for placing surfactants on the market
Directive 2010/75/EU on industrial emissions	Sets out the requirements for the construction, operation and cessation of operations of industrial installations. Industrial operations may require an EU-wide permit and must be operated according to the best available techniques.
Commission Implementing Decision (EU) 2015/495 of 20 March 2015 establishing a watch list of substances for Union-wide monitoring in the field of water policy	amendment in 2018
German Regulations	
Federal Water Act (Wasserhaushaltsgesetz, WHG)	Federal objectives for managing waters (§§ 27, 44 and 47 WHG) and pollution control specifications (§§ 32, 45 and 48 WHG): Specifications for achieving objectives and avoiding any deterioration of the chemical status of waters and detrimental changes to the water composition Allows discharge of wastewater into water bodies only if the amount and harmfulness of the wastewater can be kept as low as possible by applying the best available techniques (§ 57 WHG); Permission can also be denied if the management objectives cannot be achieved with the best available techniques (§ 12 WHG). Lays down safety requirements for facilities that handle substances hazardous to water (§ 62 in conjunction with the Ordinance on Facilities for Handling Substances Hazardous to Water (AwSV))
Surface Water Ordinance (Oberflächengewässerverordnung, OGewV)	Implementation of the EQS Regulation in national law; specification of river-basin-specific pollutants
Groundwater Ordinance (Grundwasserverordnung, GrwV)	Implementation of the GWD in German law; specification of groundwater threshold values (including plant protection products and active biocidal substances)
Plant Protection Act (Pflanzenschutzgesetz, PflSchG)	Authorisation and use of plant protection products
Chemicals Act (Chemikaliengesetz, ChemG) – Section IIA	Authorisation procedure for biocidal products. Testing and evaluating all impacts on human health and the environment

(implementing Regulation (EU) 528/2012)	
Medicinal Products Act (Arzneimittelgesetz, AMG) of 1976, last amended 10 Dec. 2015	Authorisation and trade with medicinal products for human beings and animals
Detergents and Cleaning Products Act (Wasch- und Reinigungsmittelgesetz, WRMG)	Regulates the manufacture, labelling and sale of washing detergents and cleaning products in Germany; also regulates the primary biodegradability of surfactants and cosmetic products
Wastewater Ordinance (Abwasserverordnung AbwV)	Regulation on requirements for the discharge of wastewater into waters
Wastewater Levy Act (Abwasserabgabengesetz AbwAG)	Act on charges for the discharge of wastewater into waters
Polish Regulations	
Water Act (Journal of Laws No. 2017 item 1566)	According to article 3 law the surface water quality tests (including priority substances) belong to the competence of the Voivodeship Inspector for Environmental Protection.
Regulation of the Ministry of Environment List of priority substances (Journal of Laws No. 2016, item. 681)	List of 45 priority substances in accordance with the Directive 2013/39/EU, amending the previous regulation of the Ministry of Environment from 2011 (Journal of Laws No. 2011, item. 1528)
Regulation of the Ministry of Environment (Journal of Laws No. 2016, item. 85)	Criteria and method for assessment of the status of underground water bodies
Regulation of the Ministry of Environment (Journal of Laws No. 2016, item 1187)	Criteria and method for assessment of the status of surface water bodies and environmental quality standards for priority substances
The National Environmental Monitoring Programme for 2016-2023 approved by Resolution of the LT Government	Adapts the current European strategic documents in the water policy, e.g.: - Decision No 1386/2013/EU of the European Parliament and of the Council of 20 November 2013 on a General Union Environment Action Programme to 2020 (7th EAP); - EU Biodiversity strategy to 2020: Our life insurance, our natural capital (COM/2011/0244); - Europe 2020, A European strategy for smart, sustainable and inclusive growth - Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 amending Directives 2000/60/EC and 2008/105/EC as regards priority substances in the field of water policy
Regulation of the Ministry of Environment (Journal of Laws No. 2014, item 1800)	Conditions for wastewater discharge into the water or soil, and on substances particularly harmful to the aquatic environment For some substances particularly harmful to the aquatic environment (no pharmaceuticals), the maximum permissible values of pollution indicators were specified, but only for industrial wastewater.
Act of 8 March 2013 on Plant Protection Products [pesticides] (Journal of Laws 2013, item 455).	Provisions for the use of pesticides in protecting plants and crops

Lithuanian Regulations	
Law on Water	Requires that all water bodies, i.e. rivers, lakes, groundwater, transitional waters and coastal waters, achieve good ecological status, and that heavily modified water bodies and artificial water bodies achieve good ecological potential. The Law also states that wastewater must be collected and treated using the best available technologies and discharged of with minimal environment impact. The law prohibits to discharge wastewater directly into underground water bodies.
Law on Environmental Impact Assessment of the Proposed Economic Activity	Establishes pre-decision-making preventive measures designed to reduce the impacts of a proposed project or development on environmental components, including surface water bodies and groundwater. Impact on the environment is reduced by selecting the most suitable territory, best available technologies, construction solutions and conditions of operation.
Law on Plant Protection	The State Plant Service under the Ministry of Agriculture is authorised to register plant protection products, issue marketing licenses and implement control of their import, sale, storage, packaging-labelling and use, including phytosanitary control, including for genetically modified plants and plant products that are not intended for human food and animal feed. To date, more than 400 plant protection products have been registered in Lithuania.
HELCOM Baltic Sea Action Plan and Recommendations.	Implementation of HELCOM Baltic Sea Action Plan and relevant HELCOM Recommendations i.e.: - identification of sources of the selected hazardous substances or substance groups; - a ban or restrictions on the use of identified relevant hazardous substances or substance groups; - substitution of the selected hazardous substances or substance groups with less hazardous substances, etc.
The National Environmental Monitoring Programme for 2018-2023 approved by Resolution of the LT Government	The aim of the Programme is to provide reliable information on the state of the natural environment (all components) and changes caused by anthropogenic impact. The objectives of the programme in water status monitoring also include the hazardous substances listed in EU and international agreements, such as: - Water Framework Directive (2000/60/EB), - Directive on environmental quality standards in the field of water policy (2008/105/EB), - Marine Strategy Framework Directive (2008/56/EC), - Quality of water intended for human consumption directive (98/83/EB), - Groundwater directive (2006/118/EB), - Minamata Convention on Mercury, - Stockholm Convention on Persistent Organic Pollutants and EU Regulation on persistent organic pollutants (850/2004),

	- Helsinki Convention and other international LT agreements, national strategies and programmes in water sector.
"Wastewater Management Regulation" approved by the order of the Minister of Environment	The main legal instrument that establishes basic environmental requirements for the collection, treatment and discharge of wastewater, specifies maximum allowable concentrations for hazardous and priority hazardous substances, and some other pollutants in wastewater discharges into the surface water bodies and into a sewer system. Sets EQS for surface water bodies and biota. Identifies/lists hazardous substances and other pollutants to be controlled in industrial wastewater by type of industrial activity
Order of the Minister of the Environment On the approval of the Rules on the Issuing, Renewal and Revocation of the Integrated Pollution Prevention Permits (IPPC permit).	Operators of industrial installations operating activities covered by Annex I of the IPPC rules are required to obtain an environmental permit. The IPPC permits first of all require implementation of all available pollution prevention measures and introduction of the Best Available Techniques (BAT). Apart from these general requirements, the permits specify pollution limit values as well as requiring, if necessary, the development of programmes for reducing water pollution with dangerous substances.
Regulatory document LAND 20-2001 "Requirements for the Use of Sewage Sludge for Fertilisation" approved by order of the Minister of the Environment	Regulates/establishes the classification and permission system for the use of sewage sludge in agriculture, forestry, plantation, growing of energetic plants, recultivation of damaged areas, etc. Requirements apply to sludge from municipal and similar industrial (e.g. food industry) WWTPs. Establishes limit values for hazardous (heavy metals) and other substances.
Programme for the Reduction of Water Pollution by Hazardous Substances approved by Minister of Environment	The aim of the 2015–2021 programme is to reduce pollution of surface water bodies with List II hazardous substances (Directive 2006/11/EB). Programme lists economic activities – potential sources of hazardous substances entering the water environment. Identifies main assessment and environment control measures with set deadlines. Developed additional reduction measures shall be included in updated river basin district management plans and programmes of measures.

Annex D

MPs indicators specified for industrial wastewater in Poland (List I and List II)

List I Substances especially harmful for aquatic environment, which should be eliminated

The substances particularly harmful to the aquatic environment that should be eliminated include:

1. organohalogen compounds or substances that can form such compounds in an aqueous environment
2. organophosphorus compounds
3. organotin compounds
4. substances that have carcinogenic, mutagenic or teratogenic properties in the aquatic environment
5. mercury and mercury compounds
6. cadmium and cadmium compounds
7. solid mineral oils and petroleum hydrocarbons
8. persistent synthetic substances that may float, be suspended or drown and which may influence the usage of surface water

The maximum permissible values (depending on the type of industrial wastewater) were defined for 19 indicators of substances listed above. The indicators, which are related to the MPs are given in Table AD1.

Table AD1. The highest acceptable values of selected indicators, which are related to MPs, and should be eliminated from industrial wastewater

	Indicator	**The highest acceptable value** **depends on the type of industrial wastewater**
1	Hexachlorocyclohexane (HCH)	0 mg HCH/L
2	Tetrachloromethane (CCl_4)	3 mg CCl_4/L
3	Pentachlorophenol (PCP) and its salts	2 mg PCP/L
4	Dieldrin, aldrin, endrin, isodrin	0 mg /L
5	Dichlorodiphenyltrichloroethane (DDT)	0 mg DDT/L
6	Polychlorinated biphenyl (PCB)	0 mg PCB/L
7	Polychlorinated terphenyls (PCT)	0 mg PCT/L
8	Hexachlorobenzene (HCB)	from 0 HCB mg /L to 3 mg HCB /L
9	Hexachlorobutadiene (HCBD)	3 HCBD mg /L
10	Chloroform (trichloromethane) $CHCl_3$	2 mg $CHCl_3$/L

11	1,2-dichloroethane (EDC)	from 0.2 mg EDC/L to 5 mg EDC/L
12	Trichloroethylene (TRI)	from 0.1 mg TRI/L to 0.5 mg TRI/L
13	Tetrachloroethylene (PER)	from 0.5 mg PER/L to 1 .25 mg PER/L
14	Trichlorobenzene (TCB)	from 0.05 mg TCB/L to 1.0 mg TCB/L

List II substances especially harmful for aquatic environment, which should be limited

The substances particularly harmful to the aquatic environment that should be limited include:

1. substances belonging to families and groups of substances from List I, but not included in table AD1
2. certain substances or categories of substances belonging to the families and groups of substances listed below that have a deleterious effect on the aquatic environment:

a) non-metals and metals and their compounds:

b) biocides and their derivatives not included in List I,

(c) substances which have a deleterious effect on the taste or smell of products intended for consumption by people coming from the aquatic environment, and compounds that can give rise to such substances in the water, which would make these waters unfit for human consumption,

(d) toxic or persistent organic compounds of silicon and substances which may give rise to such compounds in water, with the exception of those that are biologically harmless or which are rapidly converted into harmless substances in water,

e) inorganic phosphorus compounds and unbound phosphorus,

f) unstable mineral oils and petroleum hydrocarbons,

g) fluorides,

h) cyanides,

i) substances that negatively affect the oxygen balance in water, in particular ammonia and nitrite.

The maximum permissible values (depending on the type of industrial wastewater) were defined for indicators of substances listed above. The indicators, which are related to the MPs are given in Table AD2.

Table AD2. The highest acceptable values of selected indicators, which are related to MPs, and should be limited in industrial wastewater

	Indicator	The highest acceptable value depends on the type of industrial wastewater
1	Free cyanide (CN)	0.1 mg CN /L
2	Thiocyanate (CNS)	10 mg CNS/L
3	Formaldehyde	2 mg /L
4	Acrylonitrile	20 mg /L
5	Volatile phenols (phenolic index)	0.1 mg /L
6	Insecticides from the group of chlorinated hydrocarbons	0.5 µg /L
7	Organophosphorus and carbamate insecticides	10 µg /L
8	Caprolactam	10 mg /L
9	Anionic surfactant group	5 mg /L
10	Non-ionic surfactants	10 mg /L
11	BTX (benzene, toluene, xylene)	0.1 mg /L
12	Adsorbable Organic Halides (AOX)	from 0.5 mg Cl/L to 5 mg Cl/L

Annex E

Assessment matrix for selected measures important in reducing micropollutants (including pharmaceuticals) in water[117]

Criteria	Expected effectiveness	Substance-specific/ broad spectrum	Costs	Effectiveness horizon	Feasibility
Assessment matrix of selected measures for wastewater					
Fourth treatment stage	high	broad spectrum effect	moderate	medium to long term (>10 years)	immediately feasible
Advanced centralised treatment of rainwater	moderate	broad spectrum effect	moderate	medium to long term (>10 years)	immediately feasible
Advanced decentralised treatment of rainwater	moderate	broad spectrum effect	moderate	medium to long term (>10 years)	immediately feasible
Advanced centralised treatment of combined sewer discharges	high	broad spectrum effect	moderate	medium to long term (>10 years)	immediately feasible
Separate collection/disposal of radiocontrast agents	high	substance-specific effect	low	short to medium term (up to 10 years)	not yet immediately feasible
Assessment matrix of selected measures for detergents					
Creating an information system for the ingredients of detergents	high	substance-specific effect	low/ moderate	medium to long term (>10 years)	immediately feasible
Information campaigns for sustainable handling of detergents	high	broad spectrum effect	low	medium to long term (>10 years)	immediately feasible
Information campaigns on the correct dosing of detergents	high	broad spectrum effect	low	medium to long term (>10 years)	immediately feasible
Development of the criteria for eco-labels for detergents	moderate	substance-specific effect	low	short to medium term (up to 10 years)	immediately feasible

[117] M. Ahting, F. Brauer, A. Duffek, I. Ebert, A. Eckhardt, E. Hassold, M. Helmecke, I. Kirst, B. Krause, P. Lepom, S. Leuthold, C. Mathan, V. Mohaupt, J. F. Moltmann, A. Müller, I. Nöh, C. Pickl, U. Pirntke, K. Pohl, J. Rechenberg, M. Suhr, C. Thierbach, L. Tietjen, P. Von der Ohe, C. Winde, 2018. Recommendations for reducing micropollutants in waters. Eds Helmecke, M. (II 2.1). German Environment Agency Section II 2.1 General Aspects of Water and Soil. www.umweltbundesamt.de

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