Removal of organic micropollutantns from wastewater in a Swedish context

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MAJA EKBLAD | CHEMICAL ENGINEERING | LUND UNIVERSITY

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Wastewater treatment is a cornerstone in reducing the impact of human activities on the environment. Thus, the wastewater treatment plants have been developed to target various pollutants as these substances have been identified as problematic in the receiving waters. One of the latest issues to be addressed is the release of pharmaceutical residues and other organic micropollutants.

This is a thesis on the practical aspects on removal of organic micropollutants from urban wastewater in Sweden. The main focus of the research is on ozonation and how to design a process that works well within the limitations of Swedish wastewater treatment plants. In addition, the drivers of the upgrading of said treatment plants for removal of organic micropollutants have been investigated.

MAJA EKBLAD has been working with organic micropollutants since 2014 at the Department of Chemical Engineering at Lund University. In 2016, she started her doctoral studies in Water and Environmental Engineering as an industrial PhD student employed by Sweden Water Research, working closely with Aarhus University, Gryaab, NSVA, and VA SYD.

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Removal of organic micropollutants from wastewater in a Swedish context

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Title and subtitle: Removal of organic micropollutants from wastewater in a Swedish context

Abstract:

Due to their potential environmental risk, the abatement of organic micropollutants from wastewater has gained increasing attention in recent years. This thesis evaluates the use of ozone in Swedish wastewater treatment plants (WWTPs) and the incentives for full-scale implementation of organic micropollutant removal.

This work is divided into two sections: the first segment comprises quantitative research of the removal of organic micropollutants using ozone, and the second discusses a qualitative study on the drivers of full-scale implementation of quaternary treatment in Swedish WWTPs.

Ozone oxidation of organic micropollutants was examined in Sweden, taking country-specific conditions into consideration. This technology was implemented on a pilot scale on site at a total of 14 WWTPs, and its influence on the removal rates of organic micropollutants was determined by ozone dose, hydraulic retention time (HRT), water temperature, pH, concentration of organic matter, and prior treatment level (high- or low-loaded activated sludge and post-precipitation).

The results from the first section demonstrate that ozonation is a suitable technology for Swedish conditions. In general, a slightly higher ozone dose was needed, compared with reported values, and a shorter HRT (7 min) can be applied without altering the removal efficiency. Nitrogen removal was not a prerequisite for efficient removal of organic micropollutants, and the evaluation of ozone dose was similar, whether based on dissolved organic carbon (DOC) or chemical oxygen demand (COD).

The second section indicates that two of the most prominent drivers were proactivity with regard to possible treatment requirements and the desire to protect receiving waters. There were also aspirations to increase general knowledge of organic micropollutants and the role of WWTPs in the release of these into the environment.

Key words: ozone, ozonation, organic micropollutants, quartenary treament, advanced treatment, wastewater treatment, Urban Waste Water Treatment Directive, treatment requrements, driving forces

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What you do makes a difference, and you have to decide what kind of difference you want to make.

Jane Goodall

Acknowledgements

When I was younger, I had two things on my list not to do with my life; I was not going to get a doctorate and not become a teacher. The fact that my dad defended his thesis when I was 11 and my mom was working as a teacher might have had something to do with my determination... But here I am, about to defend my own thesis and I really enjoyed the teaching I did during my years as a PhD student. I guess there is a learning opportunity here.

My time at the department of chemical engineering has been long. I started my master thesis in 2014, continued as a project engineer for 1.5 years before staring my PhD studies in March of 2016. This was also the start of my affiliation with Sweden Water Research. During this time, I have had the privilege to work with, and next to, many brilliant and kind people that has impacted both my professional and my personal life. To all of you, a warm thank you!

I want to thank my supervisors, past and present, for enabling me to follow through with this project. To Karin for acting as main supervisor during my first years. To Michael for always being on my side, helping me with the pilot plants, taking time to see both my strengths and weaknesses, and pushing me when I needed it. To Marinette for taking care of all things practical, not being afraid of voicing your concerns and always being a voice of reason. To Kai for always taking time for me and believing in my skills as an analytical chemist.

I want to thank my coworkers in the Water and Environmental Engineering Group for all collaborations, discussions, and fun times! I want to direct a special thanks to the following: To Åsa for reading this thesis, your comments helped lifting it to the next level. To Per for always having the perfect citation on top of mind and being able to find scientific relevance in any data set. To Hilde for the laughs and help with my defense. To Maria for our discussions on everything qualitative. To Salar and Hamse for introducing me to the life as a PhD student, and dill pizza. To Rubén for taking over my pilot when I was on parental leave. To Simon for being my best office mate during the pandemic. To Sofia for insightful discussions on life. To Mirjam for always playing cool music in the lab and ensuring I had things together in this process of producing a thesis. To Alexander for sharing your knowledge on your research as well as exploring the outdoors. To Sara and Ashley for taking over the torch as new PhD student and keeping the energy high. To Ellen for everything, especially our therapy sessions and over-the-phone lunch walks during the pandemic as well as not snoring in our shared hotel rooms. To Gertrud, I can’t put in words everything we have shared, but thank you for always being a good friend and colleague!

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I also want to thank Sweden Water Research for giving me this opportunity and a different context where I belong. Having two affiliations have, from time to time, been frustrating as well as rewarding. I want to direct an extra thank you to all PhD student for the insight in your reality, putting my own into perspective. I want to give a special thank you to Emma, who was by my side during my first years as a PhD student, sharing all good and bad, and who became a dear friend.

To the staff at Gryaab, NSVA and VA SYD and all other wastewater treatment plants where I have had the pleasure to work, take samples or in other ways been in contact. To the people that accepted my request for interviews and set aside time to answer all my questions, thank you. The people at Aarhus University that have helped me analyzing so many samples. To all my co-authors for your time and effort.

Finally, I want to thank my family. Especially Christian for putting up with me and for the sacrifices you have made along the way. To Otto and Irma for providing the right amount of distraction and all the cuddles, jag älskar er!

Abstract

Due to their potential environmental risk, the abatement of organic micropollutants from wastewater has gained increasing attention in recent years. This thesis evaluates the use of ozone in Swedish wastewater treatment plants (WWTPs) and the incentives for full-scale implementation of organic micropollutant removal.

This work is divided into two sections: the first segment comprises quantitative research of the removal of organic micropollutants using ozone, and the second discusses a qualitative study on the drivers of full-scale implementation of quaternary treatment in Swedish WWTPs.

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The results from the first section demonstrate that ozonation is a suitable technology for Swedish conditions. In general, a slightly higher ozone dose was needed, compared with reported values, and a shorter HRT (7 min) can be applied without altering the removal efficiency. Nitrogen removal was not a prerequisite for efficient removal of organic micropollutants, and the evaluation of ozone dose was similar, whether based on dissolved organic carbon (DOC) or chemical oxygen demand (COD).

The second section indicates that two of the most prominent drivers were proactivity with regard to possible treatment requirements and the desire to protect receiving waters. There were also aspirations to increase general knowledge of organic micropollutants and the role of WWTPs in the release of these into the environment.

Populärvetenskaplig sammanfattning

Rena bort läkemedelsrester – en god gärning för vår natur

Läkemedel och andra ämnen som vi använder i vår vardag hamnar i avloppet när vi till exempel tvättar oss eller går på toaletten. Genom våra avloppsreningsverk släpps de sedan ut i naturen där de kan ha en negativ inverkan på vår miljö. Det finns tekniker för att rena bort ämnena innan de hamnar i naturen. Jag har forskat på en av dem, så kallad ozonering. Framförallt har jag fokuserat på hur man bäst anpassar denna teknik till svenska reningsverk och hur lagstiftning och andra drivkrafter påverkar reningsverkens vilja att bygga den här typen av rening.

Med ökad kunskap kring hur de ämnen vi släpper ut med vårt avloppsvatten påverkar vår natur kommer också ett ökat ansvar att förhindra de negativa effekterna. Det handlar om läkemedel vi äter och kemikalier vi använder, ämnen som sedan hamnar i vårt avloppsvatten där avloppsreningsverken skulle kunna rena bort dem om de satsar på nya reningstekniker.

I min forskning har jag fokuserat på hur rening med ozon skulle kunna utformas för att passa på svenska reningsverk. Bland annat har jag tittat på om ozonering skulle passa oavsett var i landet ett reningsverk ligger, dvs om vattentemperatur eller typen av rening påverkar effektiviteten. Det har funnits tankar kring att ozonering inte skulle vara lämpat för reningsverk utan kväverening. I södra Sverige har man krav på att rena bort kväve, något som man inte har i norr. Det jag såg var att koncentrationen av organiskt material, och andra ämnen som kan reagera med ozonet, är det som har störst betydelse för hur mycket organiska mikroföroreningar som försvinner. Koncentrationen av organiskt material i det traditionellt renade avloppsvattnet varierar mycket mellan olika reningsverk. Variationen har dock inte speciellt mycket att göra med om reningsverket ligger i söder eller norr, därför anser jag att just kväverening inte är ett krav för att få till en effektiv ozonering.

Vissa svenska reningsverk har redan tagit steget att bygga ut sin rening med målet att minska utsläppet av organiska mikroföroreningar, dvs. läkemedel och kemikalier, till miljön. Andra har kommit en bit på vägen och utvärderat vilka tekniker de tror kan passa just deras reningsverk. En stor anledning till detta är att Naturvårdsverket har avsatt 250 miljoner kronor som de enskilda organisationerna har kunnat söka för att genomföra just denna typ av projekt. Jag har intervjuat 19 representanter från 16 VA-organisationer som fått ta del av de här pengarna. Detta har jag gjort för att ta reda på vad som driver dem att arbeta med rening från organiska mikroföroreningar.

Idag finns det inga krav på att rena bort den här typen av föroreningar i det direktiv som reglerar vad Europas reningsverk ska rena bort. En uppdatering av direktivet som ställer nya krav på organiska mikroföroreningar i avloppsvatten är just nu ute på remiss.

Min studie visar att det finns reningsverk som redan anpassat verksamheten för att kunna möta det nya direktivet. Det faktum att de förväntar sig krav på hur mycket organiska mikroföroreningar de får släppa ut ses som en viktig drivkraft i detta arbete. En annan viktig anledning till att reningsverken har valt att göra de här satsningarna är för att skydda recipienten, dvs de åar, sjöar och hav där det renade vattnet släpps ut. Möjligheten att återanvända det renade avloppsvattnet till bevattning eller dricksvatten anges också vara en faktor.

I Sverige är det framför allt två tekniker som utvärderats och används för att rena bort dessa organiska mikroföroreningar: filtrering med aktivt kol samt ozonering. I den första fungerar kolet som en tvättsvamp som suger åt sig föroreningarna. I den andra använder man gasen ozon som reagerar med föroreningarna i vattnet. När ozonet reagerar med de organiska mikroföroreningarna förändras deras form och de har, i de flesta fall, inte längre någon effekt i naturen. Ibland använder man även en kombination av de två. Varför man väljer att använda den ena eller den andra beror till exempel på hur den befintliga reningen ser ut, hur mycket ledig yta som finns tillgänglig, hur mycket mer energi som kommer gå åt och vilken känsla de som tar beslutet har för de olika teknikerna.

Det finns fortfarande mer att lära om ozon och andra reningsmetoder. Vi vet dock tillräckligt för att bygga ut våra reningsverk och rena bort läkemedelsresterna. Genom att satsa på moderna avloppsvattenreningsverk gör vi en god gärning för vår natur, nu och i framtiden.

List of Papers

Paper I

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

Paper II

Ekblad, M., Falås, P., El-taliawy, H., Nilsson, F., Bester, K., Hagman, M. & Cimbritz, M. (2019), Is dissolved COD a suitable design parameter for ozone oxidation of organic micropollutants in wastewater?, *Science of the Total Environment*, 658, 449-456

Paper III

Ekblad, M., Juárez, R., Falås, P., Bester, K., Hagman, M. & Cimbritz, M. (2021) Influence of operational conditions and wastewater composition on the removal of organic micropollutants through ozonation, *Journal of Environmental Management*, *286*, 112205

Paper IV

Ekblad, M., Hagman, M., Cimbritz, M. (YYYY), Driving forces for the upgrade of Swedish wastewater treatment plants for removal of organic micropollutants, *Manuscript*

Author's contribution to the papers

Paper I

I participated in the design and planning of parts of the experiments, conducted the sampling at one of the pilot plants and analyzed the wastewater quality parameters as well as discussing the manuscript and proofread the article.

Paper II

I participated in the design of the pilot plant and planning of the experiments, conducted most of the sampling and analyzed wastewater quality parameters. I performed the data analysis and drafted the manuscript together with Per Falås.

Paper III

I participated in the design of the pilot plant and planning of the experiments, conducted most of the sampling and analyzed wastewater quality parameters as well as the HPLC-MS-MS analysis. I performed the data analysis and drafted the manuscript.

Paper IV

I planned and executed the interview study; transcribed, coded, and performed the data analysis; and drafted the manuscript.

Related publications

The following publications are not included in the thesis. However, the work has relevance for the topic and was conducted in connection to the main research.

Ekblad, M., Cimbritz, M., Nilsson, F., Ernst, G., El-taliawy, H., Tumlin, S., Bester, K., Hagman, M., Mattsson, A., Blom, L., Stålhandske, L. & La Cour Jansen, J. (2015), Ozonering för nedbrytning av organiska mikroföroreningar – pilottester i södra Sverige, *VA-teknik Södra*, Rapport nr. 4

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

Ekblad, M., Edefell, E. & Fältström, E. (n.d.), Reducing concentrations of microplastics and organic micropollutants - upstream and downstream abatement strategies. Conference article IWA Young Water Professionals conference 2017

Kharel, S., Stapf, M., **Ekblad, M.**, Cimbritz, M., Falås, P., Nilsson, J., Sehlén, R. & Bester, K. (2020), Ozone dose dependent formation and removal of ozonation products of pharmaceuticals in pilot and full-scale municipal wastewater treatment plants, *Science of the Total Environment*, 731, 139064

Kahrel, S., Stapf, M., Miehe, U., **Ekblad, M.**, Cimbritz, M., Falås, P., Nilsson, J., Sehlén, R., Bergendahl, L. & Bester, K. (2021), Removal of pharmaceutical metabolites in wastewater ozonation including their fate in different post-treatment, *Science of the Total Environment*, 759, 143989

Nomenclature and abbreviations

ARB	Angiotensin receptor blocker
AF	Assessment factor
AS	Activated sludge
BAC	Biologically active carbon filter
BOD	Biochemical oxygen demand
CEC	Contaminant of emerging concern
COD	Chemical oxygen demand
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
EC	Effect concentration
EQS	Environmental quality standard
GAC	Granular activated carbon
HaV	Swedish Agency for Marine and Water Management (Havs- och Vattenmyndigheten)
HRT	Hydraulic retention time
LOEC	Lowest observed effect concentration
MEC	Measured effect concentration
N	Nitrogen
NDMA	*N*-Nitrosodimethylamine
NOEC	No-effect concentration
P	Phosphorous
PAC	Powdered activated carbon
PCP	Personal care product
pe	Population equivalent
PEC	Predicted effect concentration
PFAS	Per- and polyfluoroalkyl substances
PNEC	Predicted no-effect concentration
sCOD	Soluble chemical oxygen demand
SS	Suspended solids
Swedish EPA	Swedish Environmental Protection Agency (Naturvårdsverket)
TOC	Total organic carbon
WWTP	Wastewater treatment plant

Table of Contents

1 Introduction

Substances beneficial to humans have, more than once, proven to be harmful to flora and fauna when released into the environment—whether intentional, as with pesticides and biocides, or unintentional, in the case of pharmaceutical residues in treated wastewater. Whereas the precautionary principle is attractive in theory, the benefits of new substances are often assumed to be greater than the risk until proven otherwise.

One notable example is DDT, which was considered a miracle substance that could be used to solve almost any problem until Rachel Carson published her famous book, *Silent Spring* (Carson, 1962), highlighting the enormous cost to the environment. The result was a global ban on DDT, with few exceptions. Although more precautions are now taken before introducing new substances to the market, it is not unusual for researchers to find that their negative effects in nature are caused by said manmade substances. A recent example is the use of neonicotinoids as pest control in agriculture and their effects on the bee population (Muth and Leonard, 2019), for which a partial ban was placed in 2018 by the European Union (BeeLife, 2018).

Banning or restricting the use of a substance can be efficient in reducing the adverse effects of pesticides, for example. In other cases, it might be immoral to forbid specific compounds. Few, if any, legislators would agree to outlaw a certain pharmaceutical if there were no substitutions that were available that targeted the same disease or symptom. Thus, other methods to reduce such risks to the environment are needed. This is where the wastewater treatment plants (WWTPs) comes into the picture.

The constituents of a wastewater reflect the needs and habits of the population that produces it. Anything that is flushed or poured down the drain will reach the same endpoint: the WWTP—i.e., what we consume will be found in our wastewater. A tremendous opportunity for researchers who study drug use, pharmaceutical consumption, and viral infections (e.g., SARS-CoV-2 and seasonal flu) as well as introducing measures to reduce environmental pollution.

Viewing WWTPs as a route by which pollution is discharged has driven the development of the treatment. These facilities, originally built to collect and transport waste out of cities to reduce odors and improve sanitation, are now closely monitored and controlled environments that are built to remove many compounds—

such as organic carbon, phosphorous (P), and nitrogen (N)—to protect the aquatic environment and our drinking water.

Although new facilities continue to be designed and built, many Swedish WWTPs were established in the mid-1900s and have since been fitted and retrofitted with new and increasingly more advanced treatment steps to meet evolving treatment requirements (Persson, 1998), often resulting in complex designs and illogical flowcharts. These treatment requirements are dictated by directives on several levels. The Swedish requirements are ultimately regulated by the European Union and Urban Waste Water Treatment Directive (91/271/EEC). However, this directive was imposed before the issue of organic micropollutants in wastewater was widely discussed. Thus, there are no specific treatment requirements for these substances in the directive, yet.

This lack of treatment requirements concerning organic micropollutants is being deliberated in ongoing discussions on multiple levels, from the European Union, Swedish government, departments, county boards, and wastewater utilities to individual WWTPs. A current proposal advocates for updating the Urban Waste Water Treatment Directive (2022/0345/COD), suggesting ambitious goals for the removal of organic micropollutants.

Similarly, the Swedish government has not set specific treatment requirements, awaiting the updated EU directive. Nevertheless, measures have been taken toward a large-scale removal of organic micropollutants. Grants for evaluating and implementing technologies that target organic micropollutants have been awarded to Swedish wastewater utilities by the Swedish Environmental Protection Agency (Swedish EPA) and Swedish Agency for Marine and Water Management (HaV).

The current consensus is that organic micropollutants, such as pharmaceuticals and antibiotics, in wastewater poses a risk to the environment, access to clean drinking water and, somewhat ironically, human health. However, there are many outstanding issues with regard to their removal, including the substances that should be removed, the extent to which they should be eliminated, how they should be eliminated, what substances that should be focused on, and not to forget, who should bear the costs?

Although these concerns are unresolved, several utilities in Sweden have implemented treatments that target these organic micropollutants. I have examined the drivers behind this movement, how the utilities motivate the additional costs of wastewater treatment to decision makers and consumers, and whether the result satisfies the proposed additions to the European Urban Waste Water Treatment Directive.

1.1 Aim

The aim of the work in this thesis was to evaluate the possible treatments for the removal of organic micropollutants at Swedish WWTPs, potential issues with such activity, and the driving forces behind their implementation, focusing on pharmaceuticals and ozonation. My objectives were to:

- Evaluate the compatibility of ozonation at Swedish WWTPs with various treatment configurations.
- Determine the influence of wastewater characteristics (i.e., temperature, pH, concentrations of nitrogen and phosphorous species, and organic matter) and operational parameters (e.g., hydraulic retention time, ozone dose, and dispersion method) on removal efficiencies and suggest design parameters that are suited for Swedish conditions.
- Examine whether the required ozone dose can be calculated based on COD measurements and dose-response curves for organic micropollutants.
- Identify the drivers of the implementation of treatment technologies that target organic micropollutants in Sweden today and in the future.

1.2 Outline of the thesis

This thesis is based on four papers. Papers I, II, and III introduce and discuss the ozonation pilot plant trials, the results of which are presented in Chapter 4. Paper IV was an interview study that identified the drivers of national implementation of the removal of organic micropollutants; the findings of this study are presented in Chapter 5.

2 Contaminants of emerging concern and organic micropollutants

In recent years, *contaminants of emerging concern* (CECs) have attracted interest, becoming a go to term in academia and with decision-makers. But what does this term mean, and is there a standard definition of what contaminants are considered an *emerging concern*?

This term is highly inclusive and often refers to chemical compounds that are found in groundwater, surface water, wastewater, drinking water, and food, usually at low concentrations. This definition includes pharmaceuticals, personal care products (PCPs), pesticides, and biocides—all of which have an array of adverse effects on the environment and human health. The main issues with these substances are that their toxicological effects are largely unknown and that due to their low concentrations, it is difficult to analyze them, if it is at all possible (Rosenfeld and Feng, 2011). Microplastics can be included in the term *CECs* (Lambert and Wagner, 2018), widening its definition from chemical compounds to all contaminants with potential adverse effects in the environment.

Such a broad and inclusive term is useful but only if it has a clear definition. Because this term includes contaminants that are not part of the scope of my research, I use the phrase *organic micropollutants*.

2.1 Organic micropollutants in the environment

Organic micropollutants include a variety of organic substances. An organic substance contains one or more carbon atoms, in combination with hydrogen, oxygen, or nitrogen. *Micro-* refers to the concentrations at which they are found (µg/L) in both natural waters and wastewater. This term is somewhat misleading because today's analytical techniques are more sensitive than when it was coined, detecting substances down to a few nanograms per liter (ng/L) or even lower. *-pollutants* indicates that these substances pose a risk to the environment at these low concentrations—a definition that pharmaceuticals, synthetic hormones, antibiotics, PCPs, biocides, and pesticides meet. This thesis examines an urban setting, thus excluding the diffuse sources of pharmaceuticals and biocides from animal production and farmland.

Organic micropollutants have several common characteristics. Because they are developed to target specific biological processes, they must be recalcitrant to biological degradation. A pharmaceutical that is easily degraded never reaches its site of activity and is thus not an efficacious substance. In addition, most pharmaceuticals are highly water-soluble, targeting organisms whose main constituent is water (Larsson and Lööf, 2015). These properties are favorable, as long as the compounds act on the target organism, whether it is a human, insect, or algae. Once they are excreted or otherwise released into wastewater or natural waters, these qualities become problematic (Wittmer et al., 2010).

2.1.1 Pathways

The routes by which organic micropollutants reach the environment and our natural water bodies differ, depending on their intended use. Figure 2.1 shows an overview of such pathways of pharmaceuticals, PCPs, and biocides in an urban setting.

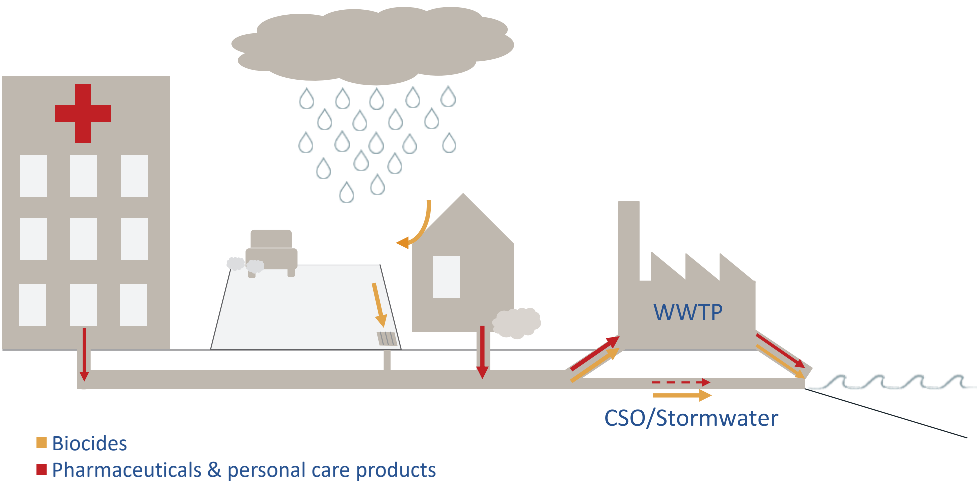

Figure 2.1
Sources and pathways of organic micropollutants in an urban setting.

In Sweden, the main pathway for pharmaceuticals (Figure 2.2) is through consumption and excretion in the home (Naturvårdsverket, 2017). Parts of the active substance are degraded in the body, and some are excreted unaltered. Regardless, they end up in in wastewater and are transported to the WWTP. An estimated 5% of all pharmaceuticals that are prescribed or bought are never consumed—of which roughly 75% is returned to a pharmacy for destruction (Castensson and Ekdahl, 2010); most of the remaining 25% is believed to be discarded in household garbage, and a small fraction is most likely flushed down the drain (Larsson and Lööf, 2015). Hospitals only contribute a minor fraction of all consumed pharmaceuticals, given that most patients are prescribed medicine that is later consumed at home (Björklund et al., 2020).

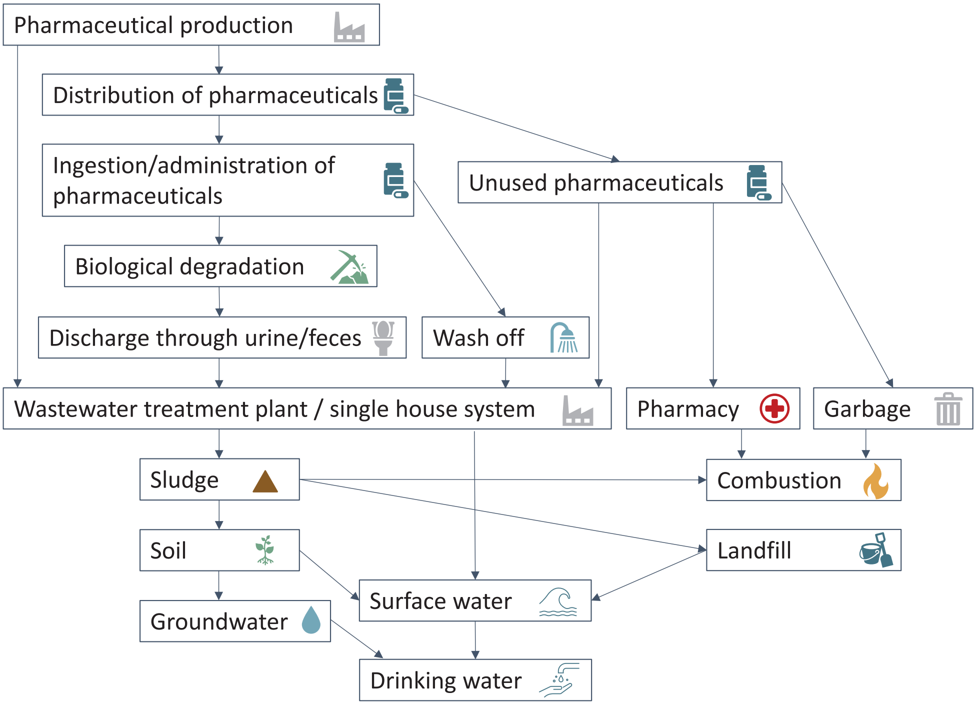

Figure 2.2
Possible pathways of pharmaceuticals intended for human use.

Once it enters wastewater, a substance is subject to three fates: separation to sludge, biological degradation, or release into the environment.

Most wastewater treatment of today is based on physical separation, biological degradation, and chemical precipitation. Micropollutants with high water solubility are less likely to adsorb to sludge and be removed through separation, as is the case for most pharmaceuticals (Gao et al., 2012, Ternes et al., 2004). If micropollutants end up in sludge, they can reach the soil through spreading of sludge on farmland (Radjenović et al., 2009, Jones-Lepp and Stevens, 2007, Golet et al., 2003) and in turn enter surface water or groundwater.

Certain substances are susceptible to biological degradation and will be removed to a significant extent in the activated sludge treatment in WWTPs. One such substance is ibuprofen, up to 99% of which is degraded, compared with only 20% of diclofenac, a substance that has similar purposes (Joss et al., 2005).

Although the scope of this thesis does not include veterinary medicines from animal production, these substances have distinct pathways in the urban setting. Pets, such as cats and dogs, are treated with a plethora of pharmaceuticals, antibiotics, and hormones. Most dogs, and many cats, do their business on sidewalks, lamp posts, and lawns; thus, any residual substance risks entering the stormwater system. In 2021, 750 kg of antibiotics, 580 kg of anti-inflammatory substances, 70 kg of hormones (including contraceptives and steroids, such as hydrocortisone), and 25 kg of antiparasitic agents were sold in Sweden to treat domestic dogs and cats. The yearly sale antibiotics for canine and feline consumption corresponds to 8% of the total amount sold for veterinary use (Jordbruksverket, 2022). In comparison, 53.3 tons of antibiotics was sold for human consumption, versus 9.3 tons for veterinary use, during the same timeframe (Swedres-Svarm, 2021). Thus, the contribution of these substances, used to treat cats and dogs, might not be insignificant, especially in separated systems without stormwater treatment.

The pathways for biocides (Figure 2.3) differ slightly from those of pharmaceuticals. Biocides have diverse applications and are found in household items; in paints and roofing materials as surface protection; gardens; and public areas (Wittmer et al., 2010). Thus, once they reach surface waters, their origin can be difficult to determine—more so if a system combines sources of water (both wastewater and stormwater in the same pipes). However, biocides found in stormwater originate primarily from paints, roofing materials and weed control (Wittmer et al., 2010).

All substances that are not removed in existing WWTPs are released to receiving waters. Whether they pose a risk to the aquatic environment depends on two overarching factors: concentration and specific toxicity.

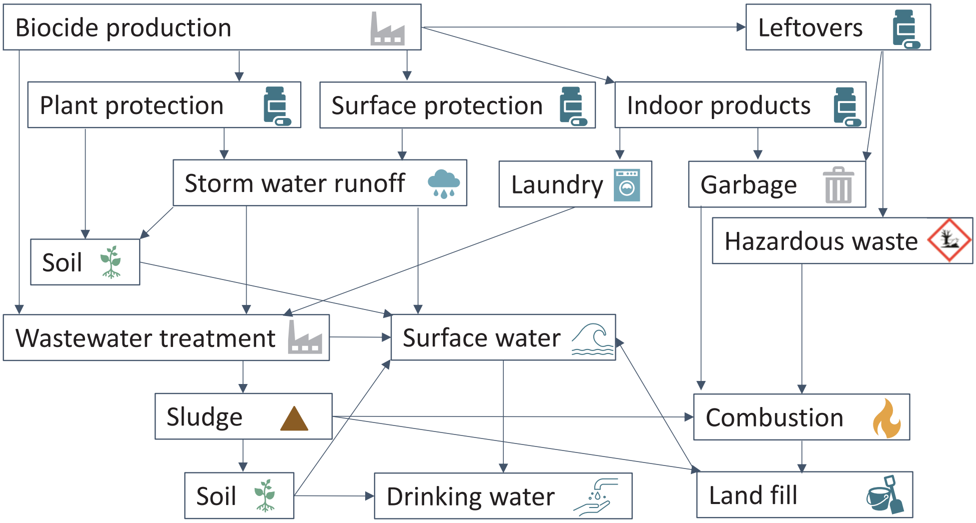

Figure 2.3
Possible pathways of biocides in an urban setting.

2.1.2 Effects on the environment

Pharmaceuticals are commonly designed to interact with proteins on the cellular level. These proteins are often not specific to humans and can be expressed in other organisms, especially other vertebrates, including fish.

Synthetic hormones, such as ethinylestradiol, used in contraceptives, affect the fertility of fish at very low concentrations (approximately 1 ng/L) (Parrott and Blunt, 2005). The predicted no-effect concentration (PNEC; for explanation and calculation, see Section 2.1.3) of ethinylestradiol is even lower (0.035 ng/L). These concentrations have been recorded in treated wastewater and surface waters (Klaic and Jirsa, 2022). In addition, other substances with similar hormonal effects are found in treated wastewater. Natural hormones, such as natural estrogens, and industrial chemicals, including nonylphenol and bisphenol A, can contribute to the total hormonal effect of the water (Gonsioroski et al., 2020)—sometimes called a "cocktail effect," which can be difficult to predict and quantify.

Diclofenac, an anti-inflammatory substance that is used as a veterinary drug and by humans, has received more attention than most pharmaceuticals as it may cause adverse effects in the environment. In Sweden, it is more commonly known under the trade name Voltaren. Diclofenac caused the mass death of vultures in India and Pakistan in the early 2000s due to fatal liver failure, as a result of their consumption of deceased livestock in which it had accumulated (Oaks et al., 2004). Diclofenac has also been shown to cause changes in liver, kidney, and gill tissue in trout at concentrations as low as 1 µg/L (Triebskorn et al., 2004).

There is an increased risk of antibiotic-resistant bacteria due to historical overuse, especially of broad-spectrum antibiotics. Although the concentrations that are released by WWTPs have not been shown to be sufficient to select for resistance (Larsson and Flach, 2022), laboratory studies have reported that already resistant bacteria can be favored at very low concentrations of antibiotics (Gullberg et al., 2011).

2.1.3 Environmental risk assessment

The risk of releasing treated wastewater can be determined using the predicted no-effect concentration (PNEC), together with a measured environmental concentration (MEC) or predicted environmental concentration (PEC).

A PNEC value for freshwater is derived through toxicological tests of three trophic levels—usually algae, daphnia, and fish. The most frequently used toxicological endpoints are mortality, growth, and reproduction. Determining the no-effect concentration (NOEC, the highest concentration at which no effects can be measured) for each organism and using the most sensitive organism as a baseline, the PNEC can be calculated using an assessment factor (AF; usually 10 for freshwater) as follows: PNEC=NOEC/AF. The PNEC for seawater can be similarly derived, or if no data are available, it can be calculated as the PNEC for freshwater divided by a factor of 10. A risk factor can be calculated as MEC/PNEC or PEC/PNEC. If MEC/PNEC is larger than 1, there is a risk of negative effects (ChemSafetyPRO, 2016). This procedure must be repeated for each substance that we want to evaluate.

2.2 Abatement strategies

Now that we have ascertained that organic micropollutants exist in natural water bodies and that they might have negative effects on the biota, we are interested in the measures that can be taken to reduce the amounts of pollutants that reach the environment. Considering the routes by which organic micropollutants enter the environment, as described in Section 2.1, we can identify several strategic nodes at which some form of abatement would be beneficial (Figure 2.4). I have divided such abatements strategies into *upstream*, based on whether they are introduced before the inlet to a WWTP, and *downstream,* if they are introduced after it.

Upstream abatement strategies include preventive interventions and technical innovations. The term *preventive* describes such interventions as legislation, substitution, and behavioral changes. Legislation includes bans on specific compounds and rules on prescription or distribution. Substitution is merely the exchange of one substance for another. Behavioral changes are more difficult to quantify. One such example, nudging, involves changing someone's behavior on their own terms, without bans or regulations—by providing information that allows a consumer to make "better" decisions, whether it is in their own interest or for the greater good (Tahler and Sunstein, 2021). Examples of technical innovations include filters that are installed in drainpipes and manholes to target biocides and small-scale treatment at the household level or at point sources.

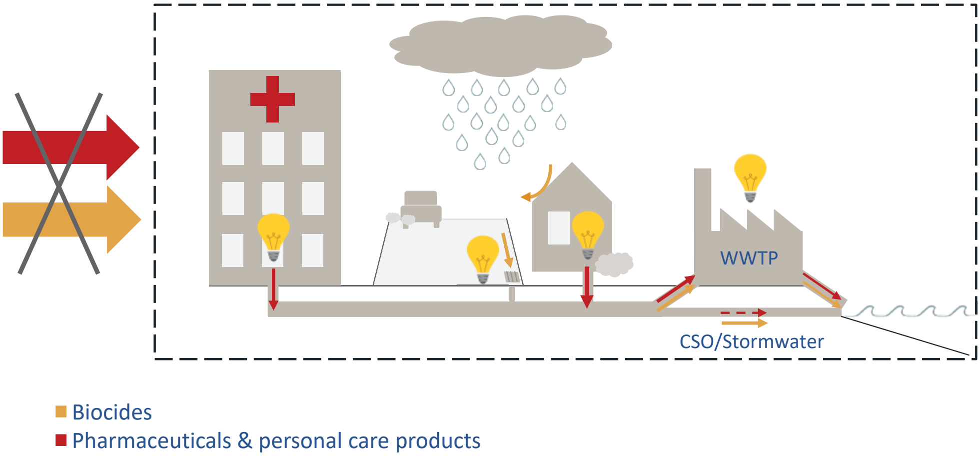

Figure 2.4
Abatement strategies. From left to right: Prevention, local interventions at point sources, local interventions treating storm water, local interventions in households, and interventions at WWTPs.

2.2.1 Advanced or quaternary treatment?

Abatement is most efficient when a combination of upstream and downstream abatement strategies is implemented (Kümmerer et al., 2018, Kümmerer et al., 2019). Preventive interventions and local abatement strategies can be effective when targeting specific micropollutants, but a downstream strategy can be more potent when trying to target a broader range of micropollutants. This strategy entails the use of technology that transforms or removes the contaminants in wastewater. Such treatment is sometimes referred to as advanced treatment or quaternary treatment. Neither term describes the phenomena ideally, because all implementations are not advanced, and depending on the placement of the technology, it might not constitute a fourth treatment step. I will use the term *quaternary treatment* when referring to these downstream abatement strategies, which is the term that is used by the European Union.

2.3 Legislation and strategies

A variety of approaches have been proposed in the development of legislation and strategies that address organic micropollutants on various decision levels (municipal, regional, national, and international). In the following sections, I discuss such attempts by the European Union, Sweden, and Switzerland. Switzerland is notable, being the first country to adopt legislation that is related to the removal of organic micropollutants.

2.3.1 European legislation over time

2.3.1.1 The beginning (1970-2000)

In the 1970s and 1980s, the first European acts of legislation were established as a step toward protecting drinking water sources and natural waters. Quality standards for drinking water sources, bathing water, and groundwater were adopted, and emissions were controlled in the Dangerous Substance Directive (67/548/EEC). In the 1990s, several directives that addressed water-related issues were adopted. For example, the Urban Waste Water Treatment Directive (91/271/EEC) was enacted in 1991 to ensure biological treatment of wastewater. Due to the sheer number of directives that targeted water, there was a desire to generate a comprehensive directive with an integrated river basin management strategy. In an attempt to combine and coordinate the objectives of the various directives, a new act was proposed in 1995 (European Commission website, nd).

2.3.1.2 The early 2000's

The European Water Framework Directive, the result of this work, was adopted in 2000 (2000/60/EC). As part of this directive, a list of 33 priority substances—primarily heavy metals, biocides, and pesticides—was established alongside environmental quality standards (EQSs) for them. This list was revised in 2013, increasing to 45 substances (2013/39/EU). No pharmaceuticals, antibiotics, or PCPs are on these lists.

In 2008, the original water framework directive was amended (2008/105/EC) to state that a watchlist of potentially harmful substances was to be established and continuously updated. The first list was adopted in March of 2015 (2015/495/EU) and comprised 10 substances that were to be monitored by the member states. This list has been updated regularly (Table 2.1) (2018/840/EU and 2020/1161/EU), and at the time of writing, the last update was published in August of 2022 (2022/1307/EU). As a result, the number of substances on the watchlist has increased, and the types of substances has changed over the years.

2.3.1.3 The present

The priority substance list was revised most recently in 2022, adding 25 substances, including per- and polyfluoroalkyl substances (PFAS), certain pesticides, bisphenol A, pharmaceutical compounds that are found in painkillers and anticonvulsants, and antibiotics (European Commission, 2022). In addition to other updates and changes, a new list of 73 priority substances and subsequent EQSs is awaiting approval (European Commission website, 2022), and a new proposal for an updated Urban Waste Water Treatment Directive was published in 2022 (2022/0345/COD). These revisions are expected to reduce the pollution from urban sources, improve the quality of European surface waters, and positively impact the goals in the Marine Strategy Framework Directive (2008/56/EC) and Bathing Water Directive (2006/7/EC).

2.3.1.4 The future

The proposal for a new Urban Waste Water Directive calls for more stringent treatment requirements of nitrogen and phosphorus and a higher degree of connected households. It also includes two new articles that are directed toward micropollutants—one on treatment and one on producer responsibility. The first, Article 8, would affect the implementation of treatment steps to remove micropollutants at all large WWTPs (> 100,000 population equivalents or pe) before 2035 and, by 2040, at WWTPs above 10,000 pe where the concentration or accumulation of micropollutants poses a risk to human health or the environment. The substances that are to be analyzed and removed are presented in the annexes to the proposal, and this list reflects the substances in the Swiss legislation. (2022/0345/COD). Implementation at all European WWTPs > 100,000 pe is estimated to reduce the cumulative toxic discharge by 40% (Pistocchi et al., 2022a) and by 75% at the point of discharge (Pistocchi et al., 2022b).

The second, Article 9 calls for producers and importers of these substances to help defray the increased costs of wastewater treatment. The portion to be paid would be determined by the amount and toxicity of a substance (2022/0345/COD). Hopefully, this measure would incentivize reduced consumption and the development of less harmful substances.

In addition to formulating directives and lists of priority substances, the European Union also grants funding for projects. Usually, these grants are handled by different programs. The *Joint Baltic Sea Research and Development Programme,* or BONUS, awarded grants to 48 projects in five different calls, allocating 100 million euros to research on approaches that promote a cleaner Baltic Sea (BONUS EEIG, 2021). One project that was funded through this program was *BONUS CleanWater*, a collaboration between Swedish, Danish, and German partners that examined sources of organic micropollutants and microplastics and developed technologies for their degradation and separation; I was one of many PhD students who worked on this project.

2.3.2 Waiting for legislation or leaping into action

Sometimes, legislation is needed to get the ball rolling but the legislative process is often slow, creating an urgency to act. Other actors, apart from legislators, have been working parallel to the European Union, both in Sweden and elsewhere. Key events, projects, and decisions are described in the next several subsections, generally in chronological order.

2.3.2.1 The beginning – local initiatives

In the early 2000s, the Stockholm County Council adopted a new pharmaceutical management strategy to protect human health, which entailed informing authorities and producers of pharmaceuticals on its strategy, educating doctors and patients on the negative environmental effects of pharmaceuticals, and collaborating with water authorities for monitoring purposes (Wennmalm and Gunnarsson, 2005). To ease matters for health care providers and patients and promote the development of pharmaceuticals with less risk, Wennmalm and Gunnarsson (2005) suggested classifying pharmaceuticals based on persistence, ecotoxicity, and potential for bioaccumulation.

In 2005, Stockholm Water Company received funds from the City of Stockholm to examine tools and methods for preventing pharmaceutical discharge; map concentrations of pharmaceutical residues in effluent water from WWTPs, in recipients, and in drinking water; study removal rates in existing WWTPs; and evaluate advanced treatment technologies (Wahlberg et al., 2010).

2.3.2.2 The government investigates

The water framework directive was integrated into Swedish legislation in 2004. Since then, Swedish water management has been regulated primarily through the Swedish environmental code (Ds 2000:61), water management regulations (SFS 2004:660), and instructions to county boards (Regulation 2002:864) (Sveriges Geologiska Undersökning, 2020).

In 2005, the Swedish EPA was assigned by the government to assess the capacity of WWTPs to remove pharmaceutical residues and other harmful substances. In their final report, they highlighted a knowledge gap in on the topic of abatement of organic micropollutants and suggested evaluating complementary technical interventions at WWTPs, such as ozonation, a combination of UV and hydrogen peroxide, membrane filtration, and filtration through activated carbon. They also noted the importance of upstream abatement, mainly in the form of regulation and substitution (Naturvårdsverket, 2008).

Table 2.1
Compounds and groups of compounds in the four versions of the EU watchlist. H: hormone; ph: pharmaceutical; PCP: found in personal care products; a: antibiotic; p: pesticide; h: herbicide; i: insecticide; ad: antidepressant; f: fungicide. *Substance included in the analytical scheme.

2015	2018	2020	2022
17-Alpha-ethinylestradiol (EE2) (H)	17-Alpha-ethinylestradiol (EE2) (H)		
17-Beta-estradiol (E2), Estrone (E1) (H)	17-Beta-estradiol (E2), Estrone (E1) (H)		
Diclofenac* (ph)			
2,6-Ditert-butyl-4-methylphenol (PCP)			
2-Ethylhexyl 4-methoxycinnamate (PCP)			
Macrolide antibiotics* (a)	Macrolide antibiotics* (a)		
Methiocarb (p)	Methiocarb (p)		
Neonicotinoids (i)	Neonicotinoids (i)		
Oxadiazon (h)			
Tri-allate (p)			
	Metaflumizone (p)	Metaflumizone (p)	
	Amoxicillin (a)	Amoxicillin (a)	
	Ciprofloxacin* (a)	Ciprofloxacin* (a)	
		Sulfamethoxazole* (a)	Sulfamethoxazole (a)
		Trimethoprim* (a)	Trimethoprim (a)
		Venlafaxine* and O-des-methylvenlafaxine (ad)	Venlafaxine* and O-des-methylvenlafaxine (ad)
		Azole compounds (f) Fluconazole, Metconazole, Miconazole, Tebuconazole*, Tetraconazole, and others	*Azole compounds* (f) Fluconazole, Metconazole, Miconazole, Tebuconazole*, Tetraconazole, and others
		Dimoxystrobin (f)	Dimoxystrobin (f)
		Famoxadone (f)	Famoxadone (f)
			Diflufenican (h)
			Fipronil (i)
			Clindamycin* (a)
			Ofloxacin (a)
			Metformin (ph) and Guanylurea (ph)
			3 sunscreen agents (PCP)

2.3.2.3 The government invests

Between 2008 and 2015, researchers in the MistraPharma project, funded by the Swedish Foundation for Strategic Environmental Research, attempted to identify pharmaceuticals that pose a risk to the environment by analyzing water samples, performing toxicity tests, determining the risk of antibiotic resistance, and proposing risk management strategies and suitable wastewater treatment technologies address these issues (MistaPharma, 2016).

In 2013, the Swedish Agency for Marine and Water Management (HaV) adopted a regulation on EQSs and classification of surface water (HVMFS 2013:19), which included a list of river basin-specific pollutants with subsequent EQSs. This list was based in part on the list of priority substances in the water framework directive but also encompassed pharmaceuticals, antibiotics, and hormones, making Sweden one of the first countries with EQSs for such compounds (Cimbritz and Mattsson, 2018). An updated version of the regulation was ratified in 2019 (HVMFS 2019:25).

In 2014, HaV allocated 32 million SEK to eight projects that aimed to develop technologies for removal of organic micropollutants from wastewater (Havs- och Vattenmyndigheten, 2018). At this time, it was assumed that ozone would be the best match for Swedish WWTPs, as also evidenced by the focus of the eight projects, six of which concentrated on testing treatment technologies. Out of these six only one project—FRAM, led by Kristianstad University—focused solely on granular activated carbon (GAC), several examined powdered activated carbon (PAC), and two resulted in full-scale ozonation plants. The remaining two projects aimed to intercalibrate the analysis techniques in the other projects and acquire knowledge from full-scale treatment facilities abroad (Cimbritz and Mattsson, 2018). That ozone was the predominant strategy can be explained by findings from early studies of the high costs of GAC compared with PAC and ozonation (Abegglen and Siegrist, 2012).

With regard to the two projects that yielded full-scale treatment with ozonation, the first Swedish ozonation plant for treating wastewater was built at Knivsta WWTP as a full-scale pilot plant, treating its entire flow after the conventional treatment (Björlenius, 2018). The first permanent full-scale ozonation treatment step was up and running in Linköping in 2017 (Sehlén and Nilsson, 2020), the design of which was based on pilot-scale trials from 2014-2015 (Sehlén et al., 2015, Baresel et al., 2016).

2.3.2.4 The government investigates further

In 2015, the Swedish EPA received a new assignment from the government to "investigate the prerequisites for the use of advanced treatment in order to remove pharmaceutical residues from wastewater in order to protect the aquatic environment." The results of this study were presented in 2017 (Naturvårdsverket, 2017). The key message in this report was that the EPA recognizes the need for quaternary treatment for removal of pharmaceutical residues in wastewater, or advanced wastewater treatment, as they chose to call it. They went on to say that the number of WWTPs that were to be upgraded could not be determined based on available knowledge but outlined the following criteria for prioritizing facilities:

- The amount of organic micropollutants released to receiving waters
- The dilution ratio and water recharge rate of receiving waters
- Multiple WWTPs discharging to the same receiving water
- The sensitivity of the receiving water—i.e., its ecological sensitivity
- Fluctuations in water recharge rate and effluent volumes from the WWTP throughout the year

They stated that technologies are available and evaluated them based on performance and cost (including energy and environmental costs). In addition, they presented a blueprint, the first step of which was to identify the WWTPs with the greatest need for these technologies and determine the governance and controls that were needed for implementation to ensure socioeconomic efficiency (Naturvårdsverket, 2017). In parallel, the Swedish Environmental Research Institute (IVL), who conducted the study, published a handbook on technologies for the removal of organic micropollutants (Baresel et al., 2017).

2.3.2.5 The present

As a direct result of the 2017 report by the Swedish EPA, the Swedish government, through the Ministry of Environment, introduced a regulation (2018:495) on grants for the removal of pharmaceutical residues from wastewater. As of 2018, the Swedish EPA has been tasked with distributing annual grants to municipalities, municipal corporations, and municipal associations that want to either evaluate the need for treatment or invest in full-scale treatment for the removal of organic micropollutants. In total, 51 wastewater utilities and municipalities have received grants for 71 projects through January of 2023: 11 WWTPs that are to be upgraded and 60 for which the need was to be evaluated or technologies tested (Björlenius, 2023).

To collect and manage the findings and experience of these projects, the Swedish Water and Wastewater Association (Svenskt Vatten) formed a client group (Beställargrupp för minskade utsläpp av läkemedelsrester, mikroplaster och andra föroreningar via avloppsreningsverk), an initiative that was also funded by the EPA. The client group has become a platform for applicants to share results, information about contractors and technologies, and discuss courses of action (Svenskt Vatten, 2022a). To improve the comparability between projects, the Swedish EPA published a list of recommended substances for inclusion in the analysis of wastewater (Naturvårdsverket, nd).

2.3.2.6 The future

The proposal for a new and updated Urban Waste Water Treatment Directive will affect the future governance of Swedish WWTPs. If this proposal is accepted without any changes, approximately 20 WWTPs will have to incorporate quaternary treatment that targets organic micropollutants, based solely on size (>100,000 pe). In addition, roughly 100 WWTPs (10,000–100,000 pe) will need to perform risk assessments to establish the need for quaternary treatment (Svenskt Vatten, 2021).

2.3.3 Switzerland – A precursor

Switzerland has been at the forefront of wastewater treatment that targets organic micropollutants. They began addressing this issue in the early 2000s, culminating in a water protection act that dictated which WWTPs required an upgrade in 2016 (Bourgin et al., 2018). This pioneering has been attributed to several factors:

- The desire to protect drinking water sources. As a landlocked country, Switzerland relies on rivers as its main source of drinking water, in which major cities lie along these rivers and treated wastewater is released back into them, becoming drinking water for downstream cities, through direct use or groundwater recharge. An additional treatment step constitutes an extra barrier that protects these drinking water sources and thus human health. Further, it helps protect the aquatic environment (Abegglen and Siegrist, 2012).
- Switzerland has the expertise and economic capacity to research and develop treatment technologies and implement them on a large scale. Its inhabitants have also shown great willingness to pay for this investment (Logar et al., 2014).
- Switzerland is not a member of the European Union; thus, it does not have to await European legislation or incorporate its directives into local laws, although the changes are compatible with EU law (FOEN, 2015a).

The water protection act states that approximately 130 WWTPs are to be upgraded with quaternary treatment. These plants were selected based on a list of parameters, including the size of the WWTP, dilution ratios, ecological sensitivity, and size of the receiving water (Bourgin et al., 2018). No specific technologies are mentioned, but at least 80% of a set number of organic micropollutants are to be removed across the entire WWTP (Eggen et al., 2014, FOEN, 2015b). A list of 12 possible pollutants to evaluate have been specified (Table 2.2) and selected as representatives of larger groups of substances, based on their properties, such as biodegradability, water solubility, and toxicity (Bourgin et al., 2018). This upgrade is funded primarily through taxation of all inhabitants who are connected to a WWTP and government subsidies to the affected WWTPs (FOEN, 2015a).

Germany has also been implementing treatment for removal of organic micropollutants on a larger scale—on a voluntary basis—promoted and financed in part by federal programs. Most of these implementations can be found in the southwest—in the regions of Nordrhein-Westfalen and Baden Württemberg. WWTPs first implemented treatments that target organic micropollutants in the 1970s to abate industrial pollution. In the early 2000s, the scope shifted to focus on applications that covered a wider range of pollutants. PAC-based treatments have been the most prevalent, followed by ozone and then GAC filters (Krahnstöver et al., 2022).

Why do we in Sweden and other countries devote time and effort to research and development when Switzerland and Germany have already made such advances? There are several fundamental differences between countries, such as the quality of wastewater that enters the treatment plants [e.g., dissolved organic carbon (DOC) content], sensitivity of receiving waters, reuse of sludge, and level of treatment (i.e., with or without nitrogen removal and high-loaded vs low-loaded activated sludge treatment). These differences are significant enough for each country to conduct its own research to optimize treatments. A group of Dutch scientists have compared German and Swiss key figures with Dutch equivalents, concluding that they have no universal validity and that the specific conditions in each country must be established before any nation-specific estimations are made (Mulder et al., 2021).

Table 2.2
List of indicator substances for evaluating the efficiency of upgraded WWTPs. This list was established by the Swiss Federal Office for the Environment (Bourgin et al., 2018). *Included in the analytical scheme.

Compund	Classification	Reactivity with ozone
Amisulpride	Pharmaceutical – Antipsychotics	high (tertiary amine, $k_{O3,pH7} = 1.5 \cdot 10^5$ $M^{-1}s^{-1}$)
Carbamazpine*	Pharmaceutical – Antiepileptic	high (olefin, $k_{O3,pH7} = 3.0 \cdot 10^5$ $M^{-1}s^{-1}$)
Citalopram*	Pharmaceutical – Antidepressant	high (tertiary amine)
Clarithromycin*	Pharmaceutical – Antibacterial	high (tertiary amine $k_{O3,pH7} = 4 \cdot 10^4$ $M^{-1}s^{-1}$)
Diclofenac*	Pharmaceutical – Anti-inflammatory	high (aniline, $k_{O3,pH7} = 6.8 \cdot 10^5$ $M^{-1}s^{-1}$)
Hydrochlorothiazide	Pharmaceutical – Diuretic	high (aniline, $k_{O3,pH7} = 8.4 \cdot 10^4$ $M^{-1}s^{-1}$)
Metoprolol*	Pharmaceutical – Beta-blocker	high (tertiary amine, $k_{O3,pH7} = 2.0 \cdot 10^3$ $M^{-1}s^{-1}$)
Venlafaxine*	Pharmaceutical – Antidepressant	high (tertiary amine)
Benzotriazole*	Industrial – Corrosion inhibitor	intermediate (deactivated aromatic, $k_{O3,pH7} = 140$ $M^{-1}s^{-1}$)
Methylbenzotriazole	Industrial – Corrosion inhibitor	intermediate (deactivated aromatic, $k_{O3,pH7} = 460$ $M^{-1}s^{-1}$)
Candesartan*	Pharmaceutical – Antihypertensive	intermediate (deactivated aromatic, $k_{O3,pH7} = 563$ $M^{-1}s^{-1}$)
Irbesartan*	Pharmaceutical – Antihypertensive	Low (deactivated aromatic, $k_{O3,pH7} = 23$ $M^{-1}s^{-1}$)

2.4 Ozone for abatement of organic micropollutants

The following two sections briefly introduce the two predominant treatment technologies that are used for the abatement of organic micropollutants at WWTPs: activated carbon and ozone. My research focuses on ozone, which is reflected in the description of the technologies.

Ozone is a highly reactive gas that consists of three oxygen molecules. The reactivity of the ozone molecule is selective, reacting only with electron-rich moieties, such as secondary and tertiary amines, activated aromatic rings, and olefins (von Sonntag and von Gunten, 2012). Thus, the degradation of organic micropollutants through ozonation is substance-specific (Nöthe et al., 2009, von Sonntag and von Gunten, 2012). For example, carbamazepine and diclofenac have higher reactivity with ozone than oxazepam and iodinated contrast media (compare k-values in Table 2.2) (Lee et al., 2014). When bubbled in wastewater, ozone gas decays and can produce hydroxyl radicals (OH radicals). These radicals are non-selective oxidants and highly reactive; thus, two types of reactions can take place: a more selective reaction with ozone radicals and reactions with OH radicals (Figure 2.5) (von Sonntag and von Gunten, 2012).

Other parameters also affect the efficiency with which ozone removes organic micropollutants, such as the properties of the wastewater with regard to the concentrations of nitrite and organic carbon, pH, and water temperature (Zucker et al., 2015, Hansen et al., 2016, Antoniou et al., 2013, Ekblad et al., 2019). Nitrite will consume ozone at a ratio of 3.43 mg O_3:1 mg NO_2-N—an important property when calculating ozone consumption at a specific WWTP.

2.4.1 Design of an ozonation treatment step

Ozone treatment is relatively easy to implement. Ozone production requires pure oxygen gas, which can be produced on site or delivered by truck. The ozone is generated by subjecting oxygen gas to an electrical field, an energy-intense procedure. Ozone gas can be mixed with water using several approaches, the most common of which are diffusors, static mixers, and Venturi injectors, sometimes in combination. A contact tank with a volume that ensures hydraulic retention times (HRTs) that are sufficiently long to deplete all ozone gas and a biologic post polishing step, such as a sand filter, to handle transformation and byproducts is needed. The dose can be based on several parameters (e.g., flow, organic matter, and UV absorbance), some of which can be measured online. Thus, the process can be monitored and adjusted in real time.

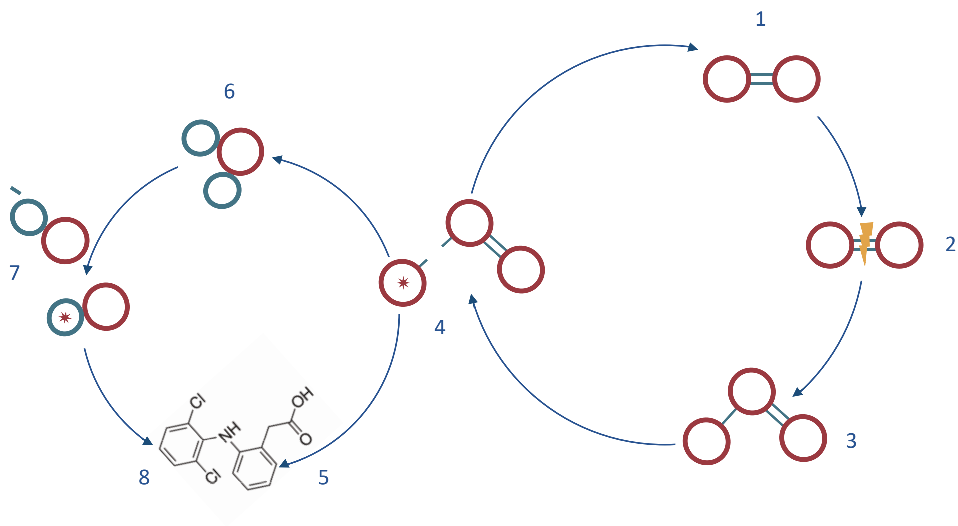

Figure 2.5
The production and degradation of ozone in water. 1) An oxygen molecule (O_2); 2) The oxygen molecule is split when subjected to high energy, such as electrical plasma; 3) Ozone (O_3) is formed when a single oxygen atom connects with an oxygen molecule (O_2); 4) The single oxygen atom will detach and either 5) oxidize an organic compound with an electron rich moitiy (in this case diclofenac) or 6) react with a water molecule; 7) One hydroxyl radical (•OH) and one hydroxide ion (OH^-) are formed; 8) The hydroxyl radical reacts with organics in the water.

2.4.2 Risks with ozonation

As a highly corrosive gas, ozone is associated with health risks. Long-term exposure to low concentrations and short-term exposure to high concentrations have negative health effects. The exposure limits are 0.1 ppm for long-term exposure (8 hours) and 0.3 ppm for short-term exposure, becoming life-threatening at 5 ppm. However, ozone has a distinct odor that is detectable at low concentrations (0.02-0.05 ppm) (Ozone Solutions, 2021), allowing one to avoid exposure. The use of personal ozone alarms or a sniffer that detects elevated levels of ozone is a simple method of preventing exposure to dangerous levels.

2.4.2.1 Byproducts of concern

During the ozonation of wastewater, toxic byproducts can form. At high concentrations of bromide, the formation of bromate is possible (Hollender et al., 2009, von Gunten, 2018, von Gunten and Hoigne, 1994); bromate is carcinogenic and thus a risk to human health (WHO, 2005). Subsequently, drinking water limits of 10 µg/L (EU, 1998, USEPA, 2006) and environmental limits of 50 µg/L (Oekotoxzentrum, 2015) have been established for bromide. Certain coastal Swedish WWTPs have high concentrations of bromide in the wastewater

(>0.7 mg/L), even in the absence of anthropogenic sources (e.g., landfills, incineration plants, and industry), which have been attributed to saltwater intrusion, thus correlating bromide concentrations and sea levels (Falås et al., 2022).

Ozonation can still be implemented at WWTPs with elevated bromate concentrations, given several approaches to preventing bromide from entering the environment. One method is to maintain a low ozone dose, because bromate formation is dose-dependent (Falås et al., 2022, Soltermann et al., 2017). To attain sufficient removal of organic micropollutants at low ozone doses, multiple dosing points can be used. Another technique is to transform the bromate back to bromide with denitrifying bacteria. This approach has been examined on a lab scale by Falås et al. (2022) and shows promise, although further testing is needed before its full-scale implementation.

Another concerning group of byproducts are *N*-nitrosamines, especially *N*-nitrosodimethylamine (NDMA), given their potent carcinogenicity (Krauss and Hollender, 2008). These substances are byproducts of disinfection through chlorination (Schreiber and Mitch, 2006) and ozonation (Najm and Trussell, 2001). The precursors occur naturally in both drinking water and wastewater, but elevated concentrations can stem from industrial discharge and the use of polymers at WWTPs. Thus, wastewater must be tested for NDMA and bromide to assess the risk for each WWTP (VSA, 2017).

2.4.2.2 Transformation products of concern

In addition to byproducts, transformation products that are more toxic than their parent compounds can also be generated, as discussed in a report from 2022 (Formas, 2022). This report includes a systematic review of 35 studies on their effects on various organisms, such as primary producers (bacteria and algae), primary consumers (zoo plankton, damselfly larvae, and mussels), and secondary consumers (species of fish), caused by conventionally treated wastewater and quaternary wastewater. The group concluded that there are negative effects on all trophic levels when they are exposed to conventionally treated wastewater, including decreased algal growth, increased mortality in daphnia, and hormonal changes and oxidative stress in fish.

Ten of the articles included in the 2022 report have reported the consequences of ozonation, indicating that the effects on algae persist but that the lifespan and vitality of daphnia increase and that hormonal changes and endpoints of exposure to toxicity decrease in fish. Four studies determined the effects of wastewater after treatment with activated carbon, in which fish experienced fewer hormonal changes and lower exposure to toxicity, although the small number of studies makes it difficult to draw any conclusions.

The conclusions of the report indicate that wastewater treatment with ozonation and activated carbon has fewer negative effects on the biota than conventionally treated wastewater. Another study on the effects of hormone-disrupting substances and genotoxicity before and after quaternary treatment (with ozone and activated carbon) at six Swedish WWTPs showed that toxicity declines significantly after quaternary treatment (Holm and Önnby, 2022).

2.5 Activated carbon for abatement of organic micropollutants

Activated carbon is a highly porous medium that can be produced from nonrenewable resources, such as coal and lignite, or renewable resources, including coconut husks, wood, and sludge. Depending on the material and method of activation, the end product can vary with regard to properties and quality (Heidarinejad et al., 2020). In this method, organic micropollutants are separated through adsorption to the carbon particles.

At a WWTP, activated carbon can be dosed continuously—as PAC—or used as filter media—as GAC. Continuous dosing can be performed in an existing treatment step—for example, in an activated sludge treatment basin. The carbon, with the adsorbed organic micropollutants, is recycled and removed with the sludge. This method is a viable solution in places where sludge is incinerated. In Sweden, sludge is traditionally used as fertilizer on farmland, which is why PAC has never been a serious option here.

For now, GAC is better suited for Swedish conditions, because most treatment plants use polishing sand filters. The sand can be substituted with GAC to reduce the organic micropollutant load or be used as a pre-polishing step to improve the lifespan of the GAC. In discussions of the cost efficiency of such a filter, it all comes down to bed volume as the chief determinant—i.e., how much water can be filtered before the granules become saturated with organics. Ensuring low DOC concentrations will improve the lifespan of the GAC, because they will compete for available sites for adsorption (Corwin and Summers, 2011).

Because bacteria will grow everywhere, biofilm will develop in the GAC filter (Gibert et al., 2013). Thus, the GAC filter becomes a biologically active carbon filter, or a BAC filter for short. Researchers have suggested that certain compounds can be degraded by bacteria that grow on the filter media, prolonging the life of the filter (Edefell et al., 2022, Reungoat et al., 2010, Altmann et al., 2016). When the breakthrough of organic micropollutants is too great (a point that is subject to discussion and can vary, based on treatment requirements and how they are formulated), the GAC can be regenerated, and the filter media be reused (San Miguel et al., 2002). At the time of writing, there are no regeneration plants in Sweden, and all filter media must be shipped abroad to be regenerated.

The possibility to regenerate the GAC is a key factor in lowering the carbon footprint of this technology. Ozone tends to have the smallest carbon footprint among technologies. Whereas PAC has a footprint that is six times that of ozone, the carbon footprint of GAC is only twice that of ozone due to the possibility of regeneration (Meier and Remy, 2020). In addition, the carbon footprint of a GAC filter depends highly on the material that is used (Baresel et al., 2017, Pistocchi et al., 2022b).

2.5.1 Combining GAC and ozone

Combinations of ozonation and GAC, primarily with ozonation as a pretreatment step before adsorption to activated carbon, are not uncommon, aiming to reduce the ozone dose and increase the lifespan of the filter media. Usually, the ozone is then used as pretreatment for the GAC filter (Boehler et al., 2020, Guillossou et al., 2020, Reungoat et al., 2010, Schollée et al., 2021). The adsorption of parent compounds increases after ozonation, due to the change in chemical structure of the organic carbon, resulting in less competition for adsorption sites (Świetlik et al., 2004, Treguer et al., 2010, Betsholtz et al., 2022). However, the transformation products that form during ozonation are less likely to be adsorbed to the activated carbon, and a negative adsorption trend was observed with increasing ozone doses (Betsholtz et al., 2022). This finding suggests that ozonation with lower ozone doses, to prolong the life of a GAC filter, is beneficial, whereas using the GAC filter as a polishing step after a higher ozone dose is applied might not be advisable.

3 Methods

This chapter introduces the methods that I have used in my research. The main focus has been on pilot-scale experiments, performed on site at WWTPs; the analysis of organic micropollutants in the wastewater; and their subsequent removal through ozonation (Papers I, II, and III)—as part of the overarching effort to optimize the treatment in the Swedish context. In addition, I have conducted a qualitative study on the drivers of the upgrade of Swedish WWTPs for removal of organic micropollutants. The data in this study were collected through semi-structured interviews with representatives from utilities, governmental agencies, and county boards and consultants (Paper IV).

3.1 Working in pilot scale

In my research, I have had the opportunity to work with four different pilot plants at 14 WWTPs in the south of Sweden. I have been working hands on with three of the pilot plants at four WWTPs (Figure 3.1-3.4). The specific designs and placements of all pilot plants are described in Papers I, II, and III. This approach of working on a larger scale (water flows between 1.5 and 4.2 m^3/h) on site had many benefits—and a few drawbacks. I have been able to study the effects of natural fluctuations in temperature, water flow, and wastewater quality. In addition, my work has facilitated the exchange of knowledge on quaternary treatment between the participants and organizations in the projects.

Working in a laboratory setting enables control of various parameters, such as temperature, pH, contact time, water quality, and initial concentrations of organic micropollutants through spiking of samples. Whereas such a practice might be beneficial for studying the oxidation rates of specific compounds or the influence of a single parameter, it might not translate to the more chaotic real-life scenario at a WWTP. The pilot plants in which I worked did not allow influent wastewater to be spiked with organic micropollutants, providing a realistic profile of what substances are present. Nevertheless, this inability can complicate the analysis, particularly when comparing results between samples from the same WWTP at various time points or from different WWTPs. It can also cause issues when mapping the removal efficiency using multiple ozone doses, because the initial concentrations are too low, preventing dose-response curves for these substances from being calculated.

Figure 3.1
Photos of the pilot plant at Ryaverket in Gothenburg. Top left: the exterior and placement at the WWTP. Bottom left: parts of the interior, including ozone gas bubbling through a contact column. Right: a sampling point, where a sampling flask is being filled with ozonated water.

Nearly all processes take longer on a pilot scale compared with the lab scale. Changing a parameter, such as ozone dose or HRT, means waiting 15 to 60 minutes before sampling (generally three times the HRT), limiting the number of variables that can be tested in a day. Thus, it might take several days, or until all wastewater quality parameters have been analyzed, to realize that things went wrong, requiring a new sampling campaign to be scheduled.

Working with ozone in general requires specific materials, because it is highly corrosive, any leakage of which poses a health risk. Thus, all pipes, contact tanks, and other equipment must meet specific standards, as do the sampling and storage of water for the analysis of organic micropollutants.

In the first pilot-scale trials (Papers I and II), I used pilot plants that were designed by other people and had to work within the limitations of the existing design. In the CleanWater project, funded by BONUS EU, I had the opportunity participate in the design of the pilot plant using my experiences from earlier pilot plants (Paper III). I found it valuable to work on a pilot scale—for the technical experience, the experience of working at a WWTP, and my interaction with the personnel at these sites. If nothing else, I became good at finding creative solutions to most problems.

Figure 3.2
The ozonation pilot plant, placed at 10 WWTPs, built and run by Filip Nilsson. Bottom left: ozone production unit, including the oxygen purification and cooling system; a stainless steel contact tank; and an equalization tank. Right: the drum filter used before ozonation.

Figure 3.3
The pilot plant used at the Klagshamn and Sjölunda WWTPs in Malmö. The larger picture shows the container, inside which an ozone production unit can be seen. The inset shows the contact tanks, an offgas ozone destructor, and the ozone injection point.

Figure 3.4
Photos from the pilot plant at the Lundåkra WWTP in Landskrona. The top panel shows, from right to left, the oxygen purification unit, ozone production unit, control panel, ozone injection points (static mixer or Venturi injector), and reactor tanks (photo by Ellen Edefell). The bottom left shows the container being lifted into place at the WWTP (photo by Ellen Edefell). The bottom right shows the control panel with a schematic flowchart with the components and measured data.

3.2 Analysis of organic micropollutants

The selection of organic micropollutants to analyze is critical. We can only say something about the substances that we measure (unless we look at general toxicity of the treated wastewater). Thus, the substances that we analyze should pose a documented or potential risk or represent a larger group of substances. In total, 52 organic micropollutants were screened for, eight of which were not detected in any water sample (Table 3.1). For most of the remaining compounds, their detection varied between sampling sessions and locations. For more details on specific compounds, see Papers I, II, and III.

Comparing this list of analyzed substances with the EU watchlist (Table 2.1), the Swiss list of indicator substances (Table 2.2), the Swedish list of river basin-specific pollutants, and the recommended substances from the Swedish EPA (Naturvårdsverket, nd), we note an overlap of 22 substances, indicating that the analyzed substances are relevant from comparative and toxicological perspectives. As part of the initial analytical package, we examined four steroid hormones: estrone, 17β-estradiol, estriol, and 17α-etinylestradiol. At the time, the detection limits were too high, and only one substance was detected in four of 14 samples (Ekblad et al., 2015).

Table 3.1
Analyzed organic micropollutants and their classification (Papers I, II, and III). [1]Compounds on the EU Watchlist. [2]Compounds on the Swiss list of indicator substances. [3]Compounds on the Swedish list of river basin-specific pollutants. [4]Compounds on the list of recommended substances from the Swedish EPA. *Not detected in any water sample.

Compound		Classification	Compound		Classification
Atenolol	[4]	Beta-blocker	Iopromide	*	Contrast media
Atrazine		Herbicide	Irbesartan	[2]	Antihypertensive
Azithromycin	[1]	Macrolide antibiotic	Isoproturon		Herbicide
Benzoisothiazolinone	*	Biocide, PCP	Losartan	[4]	Antihypertensive
Benzotriazole	[2]	Anticorrosive	Mecoprop	[3]	Herbicide
Candesartan	[2]*	Antihypertensive	Methylisothiazolinone	*	Biocide, PCP
Carbamazepine	[2,4]	Antiepileptic	Metoprolol	[2,4]	Beta-blocker
Carbendazim		Fungicide	Mycophenolic acid		Immunosuppressant
Ciprofloxacin	[1,3,4]	Antibiotic	Octylisothiazolinone		Biocide
Citalopram	[2,4]	Antidepressant	Olmesartan	*	Antihypertensive
Clarithromycin	[1,2,4]	Macrolide antibiotic	Oxazepam	[4]	Antianxiety
Clindamycin	[1]	Antibiotic	Phenazone		Anti-inflammatory
Cybutryne		Biocide, antifouling	Propiconazole		Fungicide
Diatrizoic acid		Contrast media	Propranolol		Beta-blocker
Dichlorobezamide		Herbicide	Roxithromycin	*	Antibiotic
Dichloroisothiazolinone		Biocide	Sotalol		Beta-blocker
Diclofenac	[1,2,3,4]	Anti-inflammatory	Sulfadiazine		Antibiotic
Diuron		Herbicide	Sulfamethizole		Antibiotic
Eprosartan		Antihypertensive	Sulfamethoxazole	[1,4]	Antibiotic
Erythromycin	[1,4]	Macrolide antibiotic	Tebuconazole		Fungicide
Gabapentin		Antiepileptic	Terbutryn		Herbicide
Ibuprofen	[4]	Anti-inflammatory	Tramadol	[4]	Opioid
Iodocarb	*	Fungicide	Triclosan	[3]*	Antibacterial
Iohexol		Contrast media	Trimethoprim	[1,4]	Antibiotic
Iomeprol		Contrast media	Valsartan		Antihypertensive
Iopamidol		Contrast media	Venlafaxine	[1,4]	Antidepressant

All organic micropollutants were quantified by high-performance liquid chromatography with tandem mass spectrometry (HPLC/MS-MS). The samples that were drawn in connection with Papers I and II were analyzed and interpreted by PhD students at Aarhus University. I prepared the samples (e.g., adding internal standards) for the analysis in Paper II. For Paper III, I oversaw the entire process, from taking water samples at the pilot plant, preparing the samples for analysis, and adding the procedures in the data analysis program to integrating and interpreting the output, giving me a better overview and understanding of this process.

3.3 Analysis of wastewater quality

In addition to organic micropollutants, the analysis of wastewater quality parameters is vital—especially nitrogen species (total nitrogen concentration, nitrite, nitrate, and ammonium), phosphorous species (total phosphorous concentration and phosphate), organic matter (total organic carbon, dissolved organic carbon, chemical oxygen demand, soluble chemical oxygen demand, and suspended solids), adsorption of UV light at 254 nm, water temperature, and pH. Water temperature and pH are preferably measured at the time of sampling. The other parameters must be analyzed within 24 hours of sampling to prevent degradation or other changes. Thus, one day of sampling at the pilot plant is always followed by at least one day in the lab.

The analytical procedures are standardized, and most parameters were analyzed spectrophotometrically in Hach-Lange cuvettes (Hach, Düsseldorf, Germany). Suspended solids were analyzed per ISO 11923, and UV adsorption was measured on a spectrophotometer.

3.4 Qualitative methods and semi-structured interviews

In my last study (Paper IV), I focused on an ongoing project that is evaluating the possibility of upgrading Swedish WWTPs to improve their removal of organic micropollutants. I wanted to determine the driving forces and motivations for individual WWTPs that were to go through these evaluations and to implement additional treatment steps, given that there are no laws that regulate the release of organic micropollutants, as of this writing. I also wanted to understand how those in these organizations view this issue and how they perceive the process. To this end, I had to abandon the safety and well-known world of quantitative methods and instead venture into the more obscure lands of qualitative research.

I decided to conduct an interview study, using semi-structured interviews. This method allows a set of questions to be used as the basis for the interview and ad hoc follow-up questions to be posed to gain a deeper understanding of the answers yet maintain a relaxed conversation with the interviewee.

I used the criterion-based sampling method to select interviewees, in which one reviews and studies all cases that meet the predetermined criteria (Patton, 1990). My criteria were as follows: The interviewee must:

- work at a water and wastewater utility that had completed or begun upgrading at least one 1 of its WWTPs for the removal of organic micropollutants or
- work at one of the following: Swedish EPA, the Swedish Agency for Marine and Water Management, a county administrative board, or the Swedish Water and Wastewater Association, and
- have taken an active role in work on this issue, regardless of the connection.

Based on these criteria, I chose 19 representatives from 16 water and wastewater organizations, one representative from the Swedish Water and Wastewater Association, one from a county administrative board in southern Sweden, two consultants, and one representative from the Swedish EPA. I conducted the interviews online, recording the conversations for transcription. All transcription and coding of the transcripts were performed in NVivo, a qualitative data analysis program that facilitates organization and analysis of qualitative data, such as interviews.

I used the iterative phronetic approach, synthesized developed by S.J. Tracy (2019). This approach allows a researcher without much experience with data analysis in quantitative research to obtain end results, providing a framework for engaging in qualitative data analysis. Initially, it prompts for the formulation of a specific issue, dilemma, or concern as a starting point for the research. In my work, this issue was the lack of regulation regarding organic micropollutants and the drivers of the development of solutions in Sweden. During the coding, the method suggests starting with descriptive codes as a first step. These codes are generally describing the “who, what, when, where, and how.” An example of codes that are used in the first step is presented in Table 3.2. In the second step, the coding should describe the data and interpret their meaning, answering the “how, why, or because.”

Table 3.2
An excerpt from the coding tree used in the first step of coding the interview transcripts. Three mother codes and the subordinate daughter codes used to categorize the answers of the interviewees are presented.

Mother code	1st degree daughter code	2nd degree daughter code
Driving forces	Environmental protection	
	Feeling of duty	
	Human health	
	Leading edge	
	Monetary aid	
	Personal interest	Board member Consultant or company Within organization Political interest
	Recipient protection	
	Treatment requirements	Planning for future Received requirements
Treatment technology	Choice of technology	
	Factors for choice	Byproducts Cost Energy Knowledge Proposals Safety Space Target substances
	Personal feelings	
Knowledge	Consultants	Sufficient Insufficient
	Governmental agencies	Sufficient Insufficient
	Universities	Sufficient Insufficient
	Wastewater utility	Sufficient Insufficient

4 Design parameters for ozonation

When I started working with abatement of organic micropollutants the discourse within the research community focused on how to adapt available methods to existing WWTPs and determine the optimal technologies for the Swedish framework. In one of the first meetings that I attended, several placements for ozone injection and dosing of PAC were proposed, based on the specific criteria for Swedish wastewater treatment and the expertise and experiences of two authorities from Germany and Switzerland: Norbert Jardin and Christian Abegglen (Ekblad et al., 2015). One of these suggestions is depicted in Figure 4.1.

Notably, GAC was discarded as a viable technology at this point due to results that suggested that the costs would be too high compared with PAC and ozonation. Further, the Swedish ambition to spread sludge on farmland rendered the use of PAC less compelling, because new contact basins and separate sludge handling would be needed to maintain the separation of carbon from the sludge at a traditional WWTP that used activated sludge. These factors weighed heavily in establishing the direction of my research.

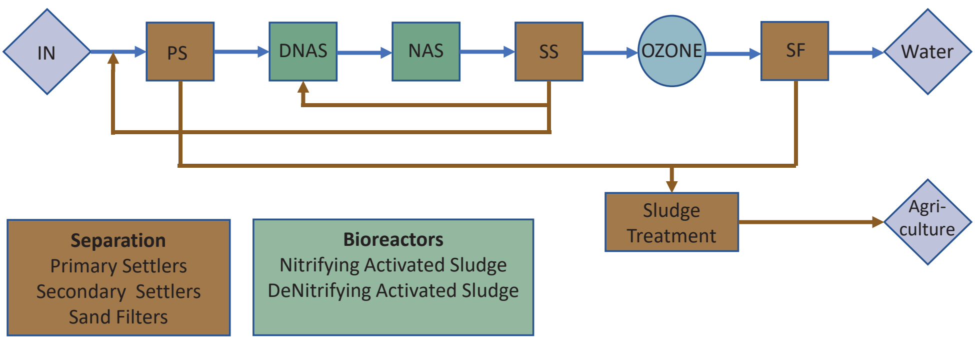

Figure 4.1
Schematic of a typical Swedish WWTP, with nitrification and denitrification and the addition of an ozonation treatment step, as discussed in 2015 (Ekblad et al., 2015).

4.1 The initial evaluation of ozone

The first step in my research was to test whether ozonating effluent water from various WWTPs, using the same equipment, would yield the same results and whether the knowledge that was gained from Germany and Switzerland was transferable to Swedish plants. The tests resulted in a wide variation in the removal rate of organic micropollutants, depending on the wastewater (Figure 4.2; Paper I), confirming that the Swiss and German values are not suitable for direct use, as claimed by Mulder et al. (2021), and that it is not possible to transfer settings between Swedish WWTPs either.

However, sufficient removal rates could be obtained at most WWTPs. At this point, we evaluated the outcome based on 90% removal of all easily and moderately easily degradable substances and proposed that a specific ozone dose of 0.7 mg O_3/mg TOC would be adequate at most WWTPs. If the more relevant 80% removal rate (based on the proposed treatment requirements from the European Union) is applied, up to 50% more ozone is required at some WWTPs compared with others, whereas this level was unachievable at other facilities (Figure 4.2). This pattern is compelling with regard to energy consumption and treatment costs.

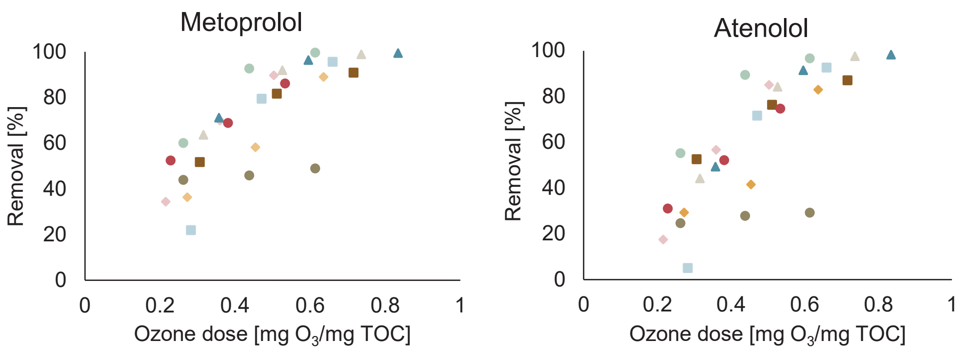

Figure 4.2
Removal of two pharmaceutical compounds, metoprolol and atenolol, at nine WWTPs. The scattering of datapoints highlight the range of removal efficiencies at similar ozone doses.

4.2 Effects on wastewater quality parameters

Based on the initial trials, we could see that the composition of wastewater affects the efficiency of ozonation. It is also important to identify how ozonation affects the composition of wastewater. All Swedish utilities operate their facilities to reach specific treatment requirements, which are usually based on organic matter, phosphorous, and, in some cases, nitrogen. Any changes to the treatment can alter the water quality, thus jeopardizing compliance with these requirements. When a new treatment step is introduced, such as ozonation, we must understand how such changes impact these parameters.

Wastewater quality parameters are used to evaluate the function of a WWTP and compare waters from various points in an individual WWTP or from several WWTPs. Because the removal of nitrogen, phosphorous, and organic matter is the main purpose of a WWTP, such parameters as nitrogen and phosphorous species (tot-N, NO_3-N, NO_2-N, NH_4-N, tot-P and PO_4-P), total organic carbon (TOC), dissolved organic carbon (DOC), and chemical oxygen demand (COD) are important to monitor. Other closely monitored parameters include suspended solids (SS) and pH.

The sampling at 11 WWTPs in Paper I allowed us to evaluate the impact of ozonation on wastewater quality parameters (Table 4.1). Considering the proxies for organic matter, TOC and COD, we observed a decrease in both parameters after ozonation. TOC concentrations declined by 6%, and COD fell by 17% on average.

With regard to nitrogen species, total nitrogen was stable, with 85% of the nitrite (NO_2^-) being oxidized to nitrate (NO_3^-). Ideally, the difference in nitrite should be visible in the nitrate that is produced. The increase in nitrate was on average slightly higher than the decrease in nitrite but can be explained by uncertainties in the analysis at these low concentrations. That nitrite reacts with ozone is significant, because an abundance in nitrite will affect the amount of ozone that is needed to attain a certain removal rate.

Where it could be detected, no significant differences in ammonium (NH_4^+) were seen, implying that ozone does not react with ammonium at these pH values. Total phosphorous (Tot-P) and phosphate (PO_4^{3-}) were stable—variations in them lay within the error of the method (Hach Lange, 2013). The concentrations of suspended solids decreased by an average of 60% after ozonation, and pH was unaffected by ozonation. In conclusion, there were no negative effects of ozonation on wastewater quality parameters.

Table 4.1
Wastewater quality parameters at 11 WWTPs before (in) and after (out) ozonation with 5 g O_3/m^3 (approximately 0.5 mg O_3/mg TOC). All concentrations are presented as mg/L. na: no measurement *Values below the level of quantification.

	TOC		COD		Tot-N		NO_3^--N		NO_2^--N		NH_4^+-N		Tot-P		PO_4^{3-}-P		pH		SS	
	in	out	in	out	in	out	in	out	in	out	in	out	in	out	in	out	in	out	in	out
Karlshamn	13.9	13.5	33.6	17.3	3.3	3.2	2.7	2.6	0*	0*	na	na	0.09	0.09	0.03	0.02	6.89	6.92	4.4	na
Ystad	10.6	9.8	30	26.7	3.0	2.4	0.63	0.47	0.16	0.01	na	na	0.17	.08	0.20	0.05	7.23	7.19	na	na
Åstorp	11.4	8.1	35.5	22.8	10.5	11.0	0.13	2.41	0.18	0	2.5	2.6	0.21	0.14	0.26	0.04	6.78	6.68	na	na
Torekov	8.7	8.6	26.6	24.8	3.5	3.5	2.7	2.8	0.1	0*	0*	0*	0.09	0.08	0.04*	0.02*	7.33	7.35	1.5	0.7
Malmö	10.8	10.6	30.9	32.0	4.3	4.3	0.8	1.2	0.6	0.01	2.3	2.3	0.11	0.11	0.05*	0.06	8.15	8.06	3.3	0.3
Lund	9.0	8.0	20.8	20.5	8.9	8.7	7.7	8.4	0.25	0*	0.1*	0.1*	0.09	0.06	0.05*	0.03*	7.08	6.81	1.7	0
Ellinge	13.1	12.5	30.3	25.0	6.3	6.5	0.64	0.58	0.05	0*	4.7	4.7	0.08	0.08	0.01*	0.01*	6.79	6.91	1.9	1.6
Kävlinge	8.2	7.7	25.5	19.8	4.9	4.9	3.8	3.7	0.09	0.0*	0.4*	0.4*	0.85	0.12	0.94	0.32	7	7.03	12.7	1.6
Svedala	12.3	12.5	34.4	32.3	5	5.0	3.3	3.2	0.14	0.17	0.5*	0.5*	0.11	0.09	0.18	0.10	7.43	7.51	8.7	2.0
Halmstad	11.4	10.8	28.5	24.2	6	5.6	2.1	2.0	0.1	0.08	2.8	3.0	0.28	0.18	na	na	6.86	7.02	9.2	7.5
Göteborg	13.2	13.2	31.1	26.3	6.1	6.0	0.59	0.75	0.09	0*	4.3	3.8	0.21	0.18	0.10	0.10	7.58	na	3.5	2.4

4.3 Impact of operational conditions

In the CleanWater project, I used my experiences with previous pilot plants to participate in the design of a brand-new pilot plant. This endeavor resulted in a pilot plant in which the HRTs could be set between 7 and 20 minutes, the dispersion method could be switched between a static mixer and a Veturi injector, and a facility that could be operated with two different waters (before and after post-precipitation). The main goal of this study was to examine the impact of operational conditions and wastewater properties on the removal of organic micropollutants (Paper III), as the basis for the design of this pilot plant.

4.3.1 pH

In theory, pH is important for the oxidation of organic micropollutants with ozone. pH affects the depletion rate of ozone molecules and the production of hydroxyl radicals—at higher pHs, ozone has a shorter life and more hydroxyl radicals are formed (Lee and von Gunten, 2010). pH also influences the speciation of protonatable molecules; thus, specific reactive sites can be deactivated through protonation, changing their reactivity with ozone (von Sonntag and von Gunten, 2012). However, in working with wastewater, I saw no evidence that pH (in the range in the analyzed wastewater) had any effect on the removal efficiency of organic micropollutants (Papers I and II).

4.3.2 Water temperature

The temperature of wastewater can vary in temperate regions, such as Sweden, where temperatures can differ by 10°C to 20°C (Davis, 2010). Because the solubility of ozone is temperature-dependent, with higher solubility at lower temperatures, I examined the possibility that water temperature impacts the removal efficiency of organic micropollutants and that a lower ozone dose during winter could affect sufficient removal of organic micropollutants. The pilot plant at Lundåkra WWTP in Landskrona was operated over seven-months, allowing me to test this hypothesis on wastewater with naturally fluctuating temperatures (12.7-21.4°C). Although there are theoretical implications of temperature dependence, the results did not demonstrate any significant differences between seasons (Figure 4.3, Paper III).

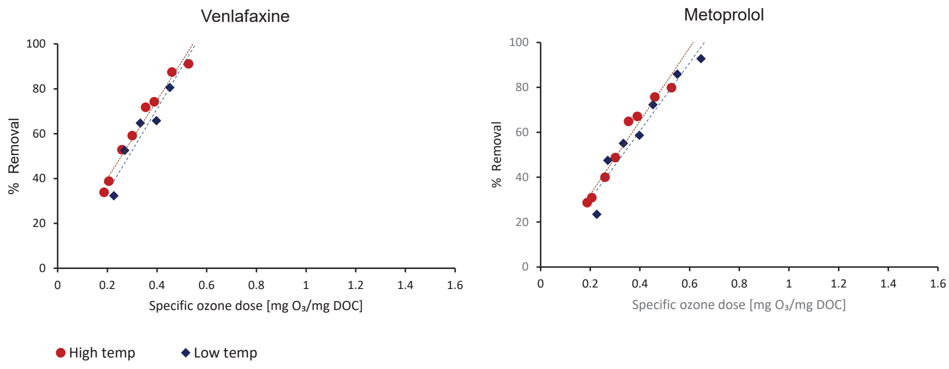

Figure 4.3
Dose-response curves for two organic micropollutants, showing removal in warm and cold weather. Mean water temperatures ranged between 13°C in cold weather and 20°C in warm weather (Paper III).

4.3.3 Ozone dispersion methods

There are several means of introducing ozone gas into wastewater. Gas can be dispersed in a side stream that is then mixed with the main wastewater stream. Alternatively, it can be dispersed directly into the main water stream or reaction tank. When gas is added directly to a reaction or contact tank, diffusors can be placed at the bottom. This type of diffusor is well established in wastewater treatment, as they are commonly used to aerate wastewater. However, these diffusors are susceptible to fouling and require regular cleaning (USEPA, 1999), which can be problematic in ozonation, given that the contact tanks should be sealed. In addition to being susceptible to fouling through biological growth, wastewater can be corrosive, underscoring the importance of the selection of materials used in these set-ups (USEPA, 1999).

Wastewater can contain debris, such as cotton swabs and other foreign objects, that increase the risk of clogging. At the Rya WWTP in Gothenburg (Paper I), we encountered issues with water snails clogging our pumps. These incidents highlight some of the more technical aspects to consider when choosing how to introduce ozone into wastewater.

When ozone is dosed directly into the main water stream through pressurizing, a static mixer can be used to disperse large bubbles into smaller ones to increase the transfer of gas, the dimensions of which can be tailored to the water flow. In addition, the mixer contains no moving parts, rendering it suitable for use in wastewater. The drawback is that bubble size is difficult to control, and the bubbles that are produced might be suboptimal in size.

To this end, the Venturi injector increases water flow, producing a slight vacuum in the gas pipe, sucking the gas into the water and creating small bubbles. A rapid increase in pipe dimension ensures adequate mixing of the water and gas. This design makes the apparatus sensitive to blockages and clogging, necessitating an additional pump to ensure sufficient water flow through the injector.

In testing the efficiency of a static mixer versus a Venturi injector (Paper III), my hypothesis was that a Venturi injector would improve removal rates, because it generates smaller bubbles. The size of the bubbles influences the gas transfer rate (Gao et al., 2019), and a higher transfer rate correlates with high removal rates. Although these tests demonstrated slightly greater removal of organic micropollutants using the Venturi injector at the lowest ozone dose (0.2 mg O_3/mg DOC) (Figure 4.4), there was no difference between methods at doses at which sufficient removal was attained. Thus, other parameters, such as water quality, design of the entire treatment step, and energy consumption, are more important when selecting the optimal method for ozone dispersion.

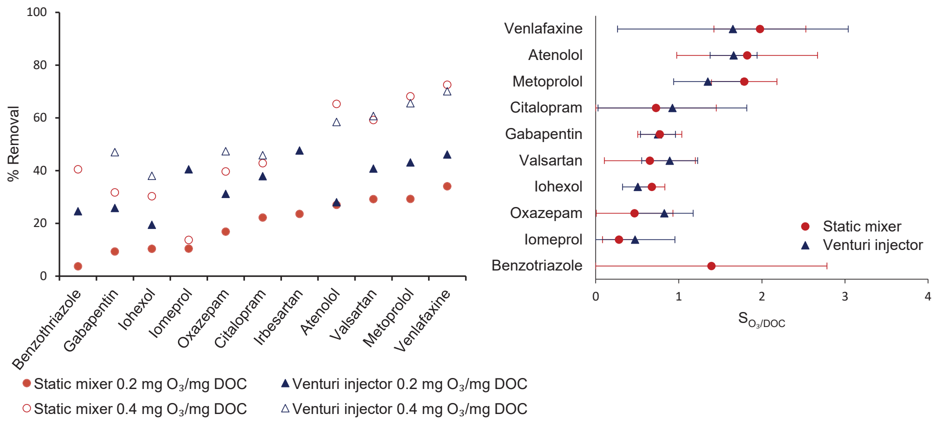

Figure 4.4
Left: percent removal of 11 organic micropollutants at two ozone doses (0.2 and 0.4 mg O_3/mg DOC) using a static mixer or a Venturi injector to disperse the ozone. Right: calculated removal constants ($S_{O_3/DOC}$) and their 95% confidence intervals (Paper III).

4.3.4 Hydraulic retention time and hydraulic loading

What constitutes a sufficient HRT depends on the parameter that is evaluated. A few seconds to several minutes is normally adequate time for dissolved ozone to react with the organic micropollutants in water (von Sonntag and von Gunten, 2012). Because there might be residual ozone in the gas phase, increasing the retention time will allow further gas transfer and ensure that all ozone has reacted—an important objective with regard to health and safety aspect.

Recommended HRTs vary between 15 and 70 minutes (Bourgin et al., 2018, Lee et al., 2012, Margot et al., 2013, Schollée et al., 2018, Östman et al., 2019). The pilot plants at which I have worked were run with HRTs of between 5 and 20 minutes (Papers I, II, and III). In Paper III, I performed a methodical test of the impact of HRTs of between 7 and 20 minutes, but the results gave no indication that removal efficiency differs with contact time. Ultimately, the availability of space and funds and safety aspect will be the determinants for the applied HRT—larger volumes for treatment will require larger contact reactors and greater investment.

The sewer systems in many Swedish cities are old, which means that their pipes could be cracked or sometimes made of pervious materials, such as clay, which will lead to groundwater infiltration. Old systems are often combined systems, in which stormwater and household wastewater enter the same pipes. The infiltration of groundwater and the collection of stormwater can cause large variations in water flow to WWTPs, complicating the design of ozonation treatments, because the system must be flexible with regard to water flow, which affects HRTs and water quality.

One of the pilot plants was operated at the Rya WWTP in Gothenburg (Paper I), in which its large sewer system resulted in significant variations in flow, depending on weather conditions (between 2.5 and 14 m^3/s). However, the removal rate of organic micropollutants was not significantly affected by the change in hydraulic loading at an ozone dose of 0.5 mg O_3/mg TOC when an HRT of 10 minutes was maintained. Preserving a specific HRT will be challenging at full scale, requiring compromises—one could choose to treat all water at shorter HRTs or maintain the HRT by bypassing part of the flow. Many WWTPs are constructed to bypass sections of the biological treatment during periods of high hydraulic loading.

4.4 Organic compounds in the water

Investigating all the previously presented parameters show that some have a potential to impact the removal rate of organic micropollutants but in the setting of a WWTP have no or a minor actual impact. As there are differences in removal rates between the WWTPs there must be something causing it. This something is called ozone scavengers and part of it can be explained by nitrite being oxidized to nitrate. The majority is usually accredited to the dissolved organic matter (DOM). These organic compounds contain the type of electron rich moieties that react with ozone (von Sonntag and von Gunten, 2012). DOM in wastewater is usually found in mg/L, compared to the organic micropollutants that are found in concentrations in the range of ng to µg/L (Nöthe et al., 2009). In order to keep the ozone dose low and the removal rate of organic micropollutants high, ozone is usually dosed after a well-functioning biological treatment with nitrogen removal (El-taliawy et al., 2017, Hollender et al., 2009, Schaar et al., 2010).

To illustrate the impact of organics in wastewater, I compared the removal rates of 11 organic micropollutants before and after post-precipitation with aluminum (Paper III). The goal of the post-precipitation was to remove phosphate. However, the concentrations of suspended solids (from 7.5 ± 6.9 to 1.8 ± 0.9 mg SS/L) and COD (from 38.1 ± 9.8 to 27.5 ± 1.8 mg/L) were also lower; DOC concentrations remained constant. There was a slight improvement in the removal of organic micropollutants after ozonation of post-precipitated water (13 ± 5%) (Figure 4.5). Although the calculated confidence intervals suggest uncertainties, this trend indicates a positive correlation with small changes in suspended solids and COD.

A more in-depth examination of the impact of suspended solids on the removal efficiency of organic micropollutants was conducted by Juárez et al. (2021), who studied the interaction between ozone and suspended solids and the removal of organic micropollutants at various concentrations of suspended solids. Their results showed that the concentration of suspended solids decreased and dissolved fraction increased with rising ozone doses. This finding suggests that ozone breaks the particles. However, this effect predominated at solids concentrations >25 mg SS/L and high ozone doses, indicating that interference by suspended solids is negligible at the concentrations that normally exist in effluent wastewater.

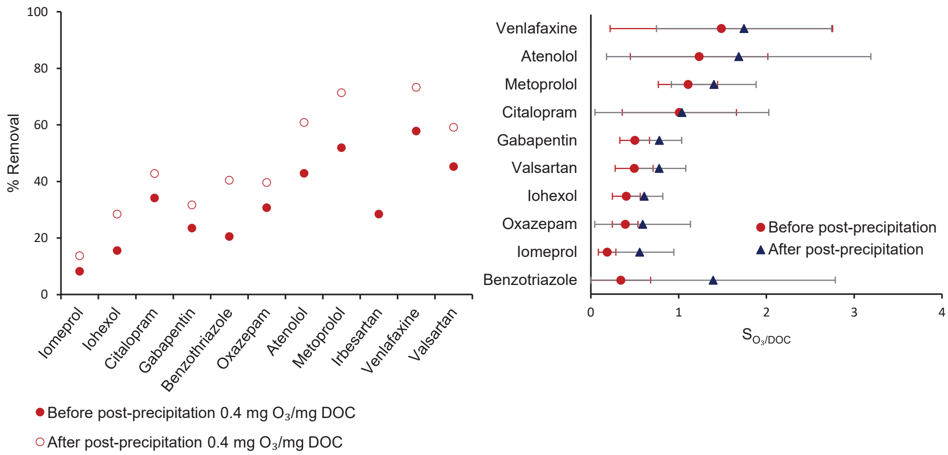

Figure 4.5
Left: Comparison of removal of organic micropollutants at Lundåkra WWTP before and after post-precipitation at 0.4 mg O_3/mg DOC. Right: Calculated $S_{O_3/DOC}$ values for organic micropollutants. The error bars represent 95% confidence intervals (Paper III).

The organic fraction of wastewater is normally represented by DOC. When removal rates of organic micropollutants are presented, DOC is commonly used to normalize ozone doses (with compensation for nitrite) (Altmann et al., 2014, Reungoat et al., 2010, Singh et al., 2015), allowing removal efficiencies to be compared between different wastewaters (Lee et al., 2013). However, normalization by DOC merely accounts for ozone scavenging by the DOC, because the measurements of DOC provide information on the amount of carbon in organic compounds. Other parameters, such as the oxidation state of the organic matter, and the presence of inorganic species that may be oxidized by ozone are unaccounted for (von Sonntag and von Gunten, 2012). COD measurements convey information on the number of electrons that is required for complete oxidation of organic compounds to carbon dioxide, water, and other inorganic products. For example, ozone-mediated oxidation of a phenol to a benzoquinone or catechol (Ramseier and Gunten, 2009) would alter COD but not DOC.

In Paper II, I examined normalization with DOC and soluble COD (sCOD), using the removal rates of organic micropollutants from two WWTPs in Malmö—Sjölunda (a low-loaded AS) and Klagsham (a high-loaded AS)—and compared the suitability of the parameters (Figure 4.6). I noted an offset between the dose-response curves that were normalized with DOC that disappeared with COD. This offset can be explained in part by the presence of nitrite in the wastewater at Klagshamn (between 10% and 20%, depending on the ozone dose). The remaining offset could be attributed to other inorganic constituents that could have acted as ozone scavengers. When Juárez et al. (2021) tested this theory on effluent water from six WWTPs, this difference could not be seen. However, they found that COD, DOC, and $SUVA_{254}$ ratios correlated well, suggesting that COD is not a superior option to DOC but that these values can be used (more or less) interchangeably.

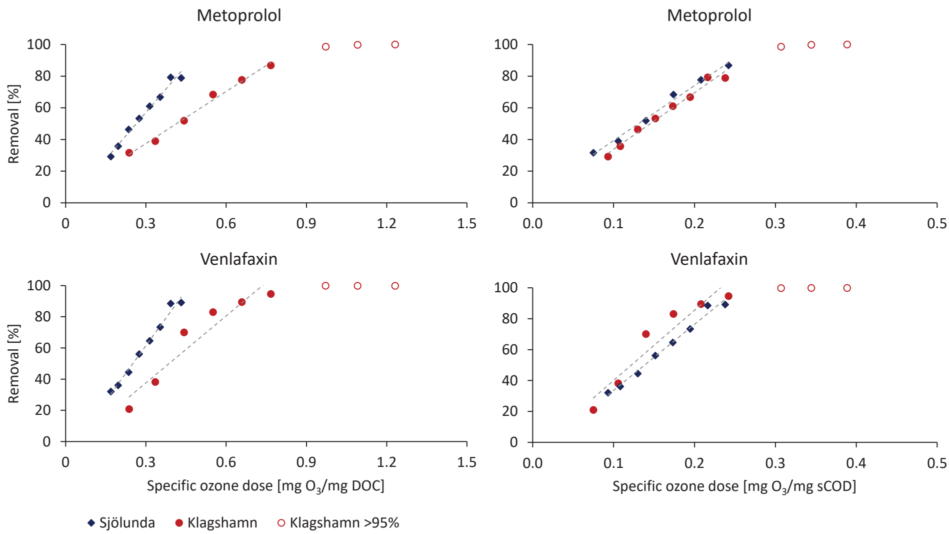

Figure 4.6
Dose-response curves for metoprolol and venlafaxin after normalization of ozone dose with DOC (left) and sCOD (right). The wastewater originated from two WWTPs: one with a high-loaded activated sludge process (Sjölunda) and one with a low-loaded activated sludge process (Klagshamn) (Paper II).

It can be assumed that DOC, with nitrite, is sufficient for evaluating ozone consumption for most domestic wastewaters unless there are industrial sources of inorganic ozone scavengers, which could be accounted for by using COD measurements. In addition, DOC is not always measured at Swedish WWTPs. COD, however, is a regulatory parameter, for which data are readily available for calculations and comparisons between WWTPs (Figure 4.7 A & B). This property is also why we used COD as the basis of our prediction model for determining the ozone dose that was required for 80% removal of organic micropollutants (Figure 4.7 C) (Paper II).

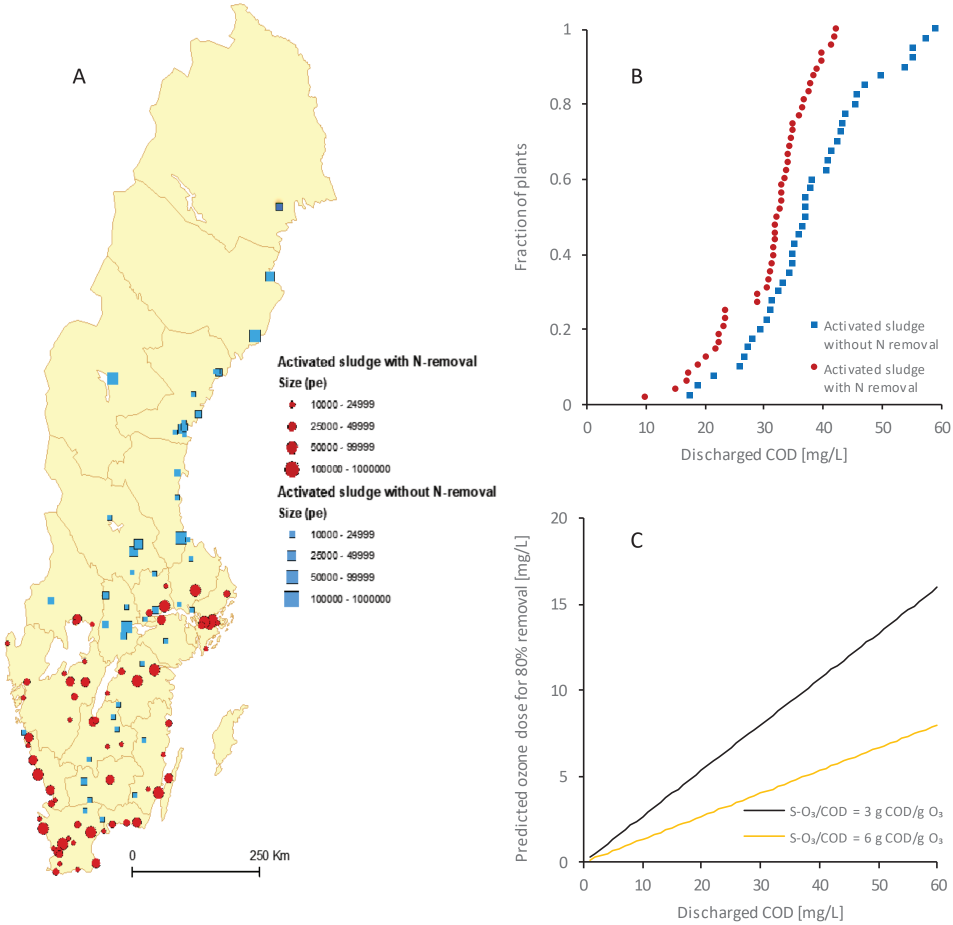

Figure 4.7
A. Geographic distribution of 88 Swedish WWTPs with activated sludge treatment, with and without requirements for nitrogen removal. Red dots indicate WWTPs with nitrogen removal, and blue squares denote WWTPs without nitrogen removal. The size of the marker indicates the size of each WWTP, in pe. B. Annual average COD concentration in wastewater released from Swedish WWTPs—48 with nitrogen removal and 40 without it. C. Estimated ozone demand for 80% removal of organic micropollutants at two reactivity constants ($S_{O_3/COD}$). A high $S_{O_3/COD}$ indicates greater reactivity with ozone; thus, a lower ozone dose is required to attain 80% removal (Paper II).

In developing the prediction model, we calculated the reactivity constants for all organic micropollutants detected in wastewater from Klagshamn and Sjölunda (Figure 4.8). These substances fall into three main categories: high reactivity with ozone ($S_{O_3/COD}$ larger than 6), medium reactivity ($S_{O_3/COD}$ between 3 and 6), and low reactivity ($S_{O_3/COD}$ below 3). Using this reactivity constant, we generated a function to calculate the predicted removal:

$$R = 100 \times S_{O_3/a} \times \frac{O_3}{A} \quad (1)$$

where *R* is the predicted removal (%), *A* is the concentration of DOC or sCOD (mg/L), O_3 is the applied ozone dose (mg/L), and $S_{O_3/a}$ is the removal constant, expressed as the slope of the dose-response function for a specific organic micropollutant (*a* denotes DOC or sCOD). If, instead, the ozone dose that is required to achieve a specific level of removal of a substance with a known reactivity constant is desired, the function can be rewritten as:

$$O_3 = \frac{R \times A}{100 \times S_{O_3/a}} \quad (2)$$

These values are visualized in Figure 4.7 C for two hypothetical organic micropollutants with removal constants that correspond to substances with high and medium reactivity with ozone. We can also see the impact of COD concentration on ozone dose, which is particularly notable when comparing WWTPs with and without requirements on nitrogen removal.

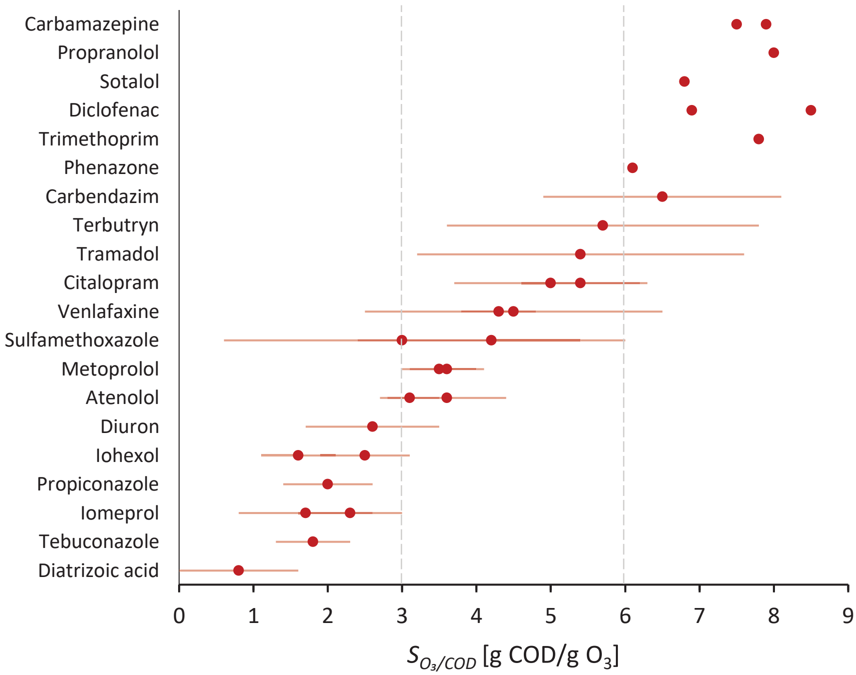

Figure 4.8
Calculated removal constants ($S_{O_3/COD}$) from two biologically treated wastewaters.

If the new proposal for an updated Urban Waste Water Treatment Directive (2022/0345/COD) is accepted without changes, the regional differences might become less pronounced, because the proposal calls for more stringent nitrogen removal at WWTPs above 10,000 pe. Until this change is completed, the average Swedish WWTP without nitrogen removal will need to apply an ozone dose that is approximately 20% higher to obtain the same removal rates as that with nitrogen removal (Paper II). Whether this is feasible must be evaluated on a case-by-case basis, but ozone can be applied at WWTPs without nitrogen removal.

4.5 Meeting new requirements

The proposal for a new Urban Waste Water Treatment Directive includes treatment requirements with regard to 12 substances, at least six of which should be measured, and stipulates that an average removal rate of 80% must be reached. Table 4.2 shows the ozone doses that are required to attain an 80% removal rate of ten of the 12 substances. Six doses are based on data from several WWTPs (Papers I, II, and III). As a result, an ozone dose of 0.5 mg O_3/mg DOC should be sufficient to attain an average removal rate of 80% at most Swedish WWTPs. Nevertheless, case-specific evaluation of wastewater quality and removal efficiencies should be performed before full-scale implementation.

Table 4.2
Organic micropollutants in the proposal for a new Urban Waste Water Treatment Directive and the corresponding ozone dose needed to attain 80% removal of the substance, based on data from Papers I, II, and III. nd: not detected at concentrations above LOQ; na: not analyzed.

Substance	Required ozone dose for 80% removal
Amisulprid	<0.3 mg O_3/mg DOC *
Carbamazepine	<0.3 mg O_3/mg DOC
Citalopram	0.6 mg O_3/mg DOC
Clarithromycin	<0,3 mg O_3/mg DOC *
Diclofenac	<0.3 mg O_3/mg DOC
Hydrochlorothiazide	0.4 mg O_3/mg DOC
Metoprolol	0.7 mg O_3/mg DOC
Venlafaxine	0.6 mg O_3/mg DOC
Benzotriazole	0.5 mg O_3/mg DOC *
Candesartan	nd
Irbesartan	0.5 mg O_3/mg DOC *
4-Metylbensotriazole and 6-Metylbensotriazole	na

*Values from eight sampling sessions at a Swedish WWTP (Hoyer et al., 2022)

5 Upgrading Swedish WWTPs

The results of my interview study suggest that the issue of organic micropollutants in wastewater has been part of the collective consciousness of wastewater utilities for at least several years and that awareness of it has risen with increased reporting via research and the media.

5.1 Does size matter?

There is a general trend in which larger organizations have assumed a more active role in gathering knowledge on the removal of organic micropollutants, participating in or leading research projects or gathering knowledge from other parts of the world through business intelligence. Cases in which smaller organizations lead such efforts can generally be described as being driven by personal interest from one or a few enthusiasts. This difference can be explained by the disparity in economic resources and personnel.

Such size-based differences are not prevalent when comparing utilities that have applied for grants (Svenskt Vatten, nd). However, there is a strong correlation between the size of a WWTP and the type of project for which they have received grants. Of 10 treatment plants that have been awarded investment grants, eight have treatment capacities below 10,000 pe; the remaining two have reported capacities of 15,000 and 61,000 pe, respectively (Svenskt Vatten, nd). This pattern can be explained in part by economic practicalities: the entire budget for the grants that were posted by the Swedish EPA was 250 million SEK over a 5-year period (2018-2022) (Björlenius, 2023), and the estimated investment for one of the largest WWTPs in Sweden (ca 1,000,000 pe) amounted to 400-500 million SEK, as reported by one of the interviewees. Another interviewee responded with an estimated investment of 300 million SEK for a WWTP with a treatment capacity of 900,000 pe and 70-80 million SEK for a plant with a 200,000-pe capacity. The largest sum that was granted for investing in new treatment technologies in this program was 19 million SEK, covering 90% of the total investment cost for upgrading the Kivik WWTP with GAK filtration (Svenskt Vatten, nd).

Another reason that was given for choosing an evaluation project over an investment project was the lack of treatment requirements. The utilities desired to remain updated on the issue by evaluating the need for treatment and, in some cases, performing pilot-scale testing of a specific technology but chose to await a decision from authorities before investing.

5.2 Driving forces

The possibility of receiving grants was mentioned as an igniting spark in most of the interviews—i.e., the organization would not have proceeded with the evaluations or investments without financial incentive. However, when analyzing the interviews, other important drivers were identified.

5.2.1 Protection of receiving waters

The desire to protect recipient waters is an important factor when deciding to evaluate or reduce the impact of treated wastewater. The interviewees who cited this driver also commented that the recipient is small, sensitive, or exposed in other ways. That multiple WWTPs release treated water in or close to protected areas, such as marine nature reserves and Natura 2000 areas, was presented as incitement for evaluation or implementation of quaternary treatment. Three interviewees mentioned that the respective recipient is critical for fish (salmon and trout) and, in one case, the endangered freshwater pearl mussel (flodpärlmussla). In addition, the impact on bath water quality was mentioned as a positive side effect in three cases: two coastal WWTPs and one that released water to a lake. The protection of drinking water sources does not appear to be a primary driver but is considered when discussing improving the status of the recipient.

5.2.2 Water reuse

The possibility of water reuse was broached by two persons who represented two different utilities in Skåne. One cited the potential for water reuse as one of its main drivers for implementing the removal of organic micropollutants. That these utilities discuss this motivation is notable, because parts of Skåne have experienced water scarcity since 2018, with peak shortages during summers (Svenskt Vatten, 2022b). The topic of water reuse has also been actualized with the construction of desalination and reuse plants on Gotland and Öland (Takman et al., 2023). No other representatives of utilities mentioned the possibility of reuse at all, perhaps because they reside in areas where this issue does not exist or is not prioritized.

There is a conspicuous discrepancy between utilities and representatives from the Swedish EPA, Swedish Water and Wastewater Association and the two consultants. It seems that this discourse is more prevalent in the governmental organizations, because all of the representatives discussed the possibilities and opportunities with regard to wastewater reuse. The reason behind this discrepancy in the discourse is hard to determine, but I speculate that utilities are more focused on solving more immediate issues, whereas the other parties are attempting to predict a more distant future and find ways to encourage implementation on a larger scale. Perhaps water reuse is currently a buzz word among certain groups that may reach utilities in due time, especially with the proposed Urban Waste Water Treatment Directive highlighting the issue.

5.2.3 Justification of increased costs

Motivating the initial investment and an increase in operational costs associated with technologies for removal of organic micropollutants is an issue mentioned by the utility representatives. The balance between benefit, measured primarily as the removal rate or improved water quality in the recipient, and cost—monetary, energy, and environmental (i.e., carbon footprint)—is acknowledged as a determinant of implementation. Many also opine that the knowledge of the actual gain is insufficient to justify investment, choosing to evaluate the need for quaternary treatment and assess one or more technologies to fill this knowledge gap.

My impression is that there is a general issue regarding the transfer of knowledge from universities, research institutes, and governmental agencies to utilities and between utilities. An initiative with a client group that was organized by the Swedish Water and Wastewater Association has been instrumental in creating a platform for sharing knowledge and information. When I performed the interviews, this group was new, and the pandemic prevented in-person meetings and workshops. This shortcoming was echoed in the impressions and opinions of the interviewees when evaluating the value of the group.

5.2.4 Anticipating treatment requirements

The most prevalent driving force, mentioned by all interviewees regardless of affiliation, was the imminent risk of new treatment requirements for organic micropollutants. Without this risk, there would have been much less interest in applying for grants. These interviews were conducted before the proposal for a new Urban Waste Water Treatment Directive was released. Regardless, all interviewees recognized the future arrival of treatment requirements, although with disparate visions of their content and how they will affect a specific WWTP.

Most subjects wished for requirements that were based on the needs of the recipient, wherein some hoped that the size of the WWTP would not be the only prerequisite or a parameter at all. Nonetheless, interviewees hoped for clear requirements, regardless of the organization that issued them or their exact wording. Because most utilities were in the process of applying for new permits or planning an expansion or new development in the near future, they recognized the need to evaluate their ability to meet such demands on treatment efficiency. Utilities that had invested in new treatment technologies were not worried that their treatments would be insufficient to meet any requirements.

5.3 Attitudes on full-scale implementation

With regard to technology choices, of the 19 evaluation projects in this study, 13 included evaluations of at least two technologies. The predominant technologies were ozone, GAC, or their combination. In one case, PAC was included in the evaluation. One notable finding was the number of projects that included the combination of ozone and GAC, which was chosen as the setup in three evaluation projects and two of six investment projects. This selection is sometimes motivated by the prolonged life of the GAC filter, as a precaution against byproducts and transformation products from ozonation, or the desire to abate a broader spectrum of organic micropollutants compared with the individual technologies.

In general, discussing the various technologies invoke strong feelings. Sometimes, but not always, these feelings are grounded in facts and research. In certain cases, this objectivity has influenced the choice of technical setup. In cases in which the evaluation led to a decision on a suitable technology for a WWTP, five determined ozone to be the most appropriate, whereas four selected GAC. With regard to investment projects, one decided to implement an ozonation step, and three chose GAC. When discussing the possibility for GAC, that there is no regeneration plant in Sweden was mentioned as a drawback, but no one considered it a reason for avoiding it. One of the most important aspects in deciding for or against a technology, according to the interviewees, was its environmental impact. In comparing energy consumption, the carbon footprint and removal efficiency were considered to be important, especially because many organizations strive for carbon and energy neutrality.

There is hesitancy surrounding the implementation of large-scale removal of organic micropollutants. As discussed, there are several reasons for this indecision, including the large investment that is required, increased energy consumption, lack of regulations, a perceived lack of knowledge on the topic, skepticism of the technologies, and trust in dilution. Another explanation of this reluctance is the anticipation of new unknown issues that will need a solution in the future. There is hope that by delaying the incorporation of removal treatments, such variables will be revealed and a technology that can handle multiple types of pollution can be developed and implemented.

One such issue is PFAS, which have gained significant attention in recent years. They are not susceptible to ozone in general, but it is possible to remove them using GAC. Thus, breakthrough will occur earlier than with other organic micropollutants, shortening the lifespan of the filter and increasing the frequency of reactivation (Kaiser et al., 2021). That these persistent substances are found worldwide should compel stakeholder to focus not only on the concentrations but the total amount of substances in the water that is released from our WWTPs.

I hope that the knowledge that has been gained in these projects is managed such that it can be transferred to other interested parties and that there will be adequate follow-up on the outcomes of the evaluation and investment projects, regarding technical parameters and removal efficiencies over time. These aspirations were also expressed by the interviewees—particularly the importance of evaluating the results and the perceived lack of plans for this transfer of knowledge.

6 Conclusions

The focus of my research has been to evaluate the possibility of using ozonation for the removal of organic micropollutants and examine what drives progress toward implementation of quaternary treatment at Swedish WWTPs. Based on my results, I conclude that ozonation is a technology that is well suited for Swedish conditions.

In my research, I have found that:

- The concentration of organic matter is the parameter that most influences the removal efficiency. Level of treatment, wastewater temperature, pH, and dispersion method are less influential under these conditions.
- Dose-response curves for organic micropollutants are generally linear in the removal range up to 95%, generating key figures that can be used to estimate ozone demand.
- Applying an ozone dose of 0.5 mg O_3/mg DOC or a corresponding dose that is based on COD is sufficient to meet the proposed treatment requirements.
- An HRT of 5-10 minutes is sufficient for the removal of organic micropollutants and ensures ozone depletion.

In addition, I recommend that utilities that are interested in implementing full-scale ozonation for the removal of organic micropollutants do the following:

- Evaluate concentrations of ozone scavengers—DOC and nitrogen or COD. Removal of organic micropollutants is possible at higher concentrations but will be more costly in terms of energy input. If possible, add or improve the treatment to reduce the concentrations of ozone scavengers.
- Consider the flow for which the design should account and the water quality that is treated, because these parameters will have large impact on the size of contact tanks and ozone production units.
- Run lab-scale tests with ozone to determine the suitability of specific wastewater. Pilot plant trials have much to offer but are time-consuming and costly, and we now have a deeper understanding of the technology and the parameters that impact removal efficiency and ozone consumption. Unless experience with operating a larger ozonation plant is desired by the personnel at a WWTP, lab-scale trials should thus be sufficient.

The number of full-scale implementations that target organic micropollutants in Sweden is increasing, and even more wastewater utilities have evaluated the need for them at their WWTPs. Ozone remains one of the two most common technologies to be considered or implemented. However, there are certain reservations due to the risk of transformation and byproducts and the poor impact on PFAS concentrations. Thus, GAC filters and combinations of ozone and GAC are favored.

With the Swedish EPA investing 250 million SEK in evaluating the need for treatment and building treatment steps at Swedish WWTPs over a five-year period and the Swedish Water and Wastewater Association coordinating a client group, Sweden is at an advantageous starting point when and if the proposal for a new Urban Waste Water Treatment Directive is accepted. To ensure this advantage, I hope that funds will be allocated toward evaluation and follow-up of the projects to allow for further transfer of knowledge between organizations. This is especially important regarding the investment projects.

If we can manage the knowledge that we have gained, through academic research and during the evaluation and implementation of full-scale removal, and ensure that it reaches the correct stakeholders, I believe that we will form a strong base on which treatments for the abatement of organic micropollutants can be developed.

7 Future work

Reducing the amount of ozone that is needed to obtain certain removal rates of organic micropollutants remains an important target to save energy, striving toward climate neutrality. Determining the impact of various types of organics in wastewater would be desirable because uncertainties surrounding this issue persist, the evaluation of which the use of fractionation of organic matter could be a tool.

As the number of full-scale implementations increases, the importance of online control of ozone dosing rises. Identifying the ideal parameter to use as a proxy for ozone dose is essential, necessitating a deep understanding of how various parameters correlate with ozone consumption.

Post-treatment of ozonated wastewater is an important means of abating the release of degradation products and byproducts. Finding efficient and well-functioning alternatives increasing our understanding of these processes is an interesting research field.

Further work with bromide and the removal of bromate that forms during ozonation will be a critical topic, because Swedish WWTP have reported high concentrations in wastewater.

With the expansion of WWTPs resulting in cleaner wastewater, it would be advisable to examine alternatives to releasing treated water into the environment. Particularly given the growing desire for circulating nutrients and increasing water scarcity, the reuse of treated wastewater is gaining interest, for which the disinfecting properties of ozone and the removal of organic micropollutants are compelling.

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