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Pharmaceuticals and other chemicals of emerging concern in leachate from a landfill and sludge storage facility

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ABSTRACT

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The occurrence of 86 chemicals of emerging concern (CECs) (including pharmaceuticals, parabens, per- and polyfluoroalkyl substances, pesticides, stimulants, an isoflavone and an industrial chemical) was investigated in this study at Hovgården, a waste facility in Uppsala (Sweden). The selected CECs were analysed using solid phase extraction, sludge extraction and ultra-high pressure liquid chromatography coupled to mass spectrometry. The main aim was to get a broader understanding regarding the occurrence of pharmaceuticals in leachate (which pharmaceuticals that were present in what quantities and where) at Hovgården, particularly from a landfill and sludge storage facility.

The occurrence of pharmaceuticals most likely originated from leachate formed in sludge from the old landfill's final cover and from the sludge storage facility. 45 out of the 56 analysed pharmaceuticals were detected in the landfill leachate, at mean concentrations ranging from 1.1 ng/L (fexofenadine and tramadol) to 4900 ng/L (erythromycin). The total mean concentration of pharmaceuticals in leachate received from the old landfill's final cover (23 000 ng/L) was 45 – 49 times greater than the active landfill. 48 pharmaceuticals were detected in leachate from the sludge storage facility, at mean concentrations ranging from 1.3 ng/L (climbazole) to 3000 ng/L (desvenlafaxine). The total mean concentration of pharmaceuticals ranged from 1500 ng/L (leachate from contaminated soil) to 16 000 ng/L (combined leachate from sludge and contaminated soil). The total mean concentration of pharmaceuticals decreased by approximately 8.1 % after the wastewater treatment plant's (WWTP's) last treatment step compared to the WWTP's influent. The WWTP's recipient had the lowest total mean concentration of pharmaceuticals (240 ng/L) of all sampling points.

It was not possible to determine whether the obtained results regarding the occurrence of pharmaceuticals were reliable, as this was the first time that this was evaluated at Hovgården. Further screenings were therefore recommended, by repeating this study, before implementing additional treatment methods. It was also recommended to include a risk characterization, to evaluate and assess whether the obtained concentrations may be harmful for the receiving environment.

In addition, the presence of CECs in waste (and consequently at waste facilities) is due to our consumption and disposal of CECs. This includes our overuse/misuse of pharmaceuticals with shown consequences for the (aquatic) environment that indirectly could harm the human health, such as increased antimicrobial resistance. The occurrence of CECs in sludge and sewage systems could thus be minimized by also increasing the public's awareness about the consequences of our actions regarding CECs.

Keywords: chemicals of emerging concern (CECs); pharmaceuticals; leachate; landfill; sludge; wastewater treatment plant (WWTP)

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REFERAT

Läkemedel och andra kemikalier i lakvatten från en deponi och slamlagringsanläggning som potentiellt är hälso- eller miljöfarliga

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I denna studie undersöktes förekomsten av 86 kemikalier som potentiellt är hälso- eller miljöfarliga (eng. *chemicals of emerging concern: CECs*) (inklusive läkemedel, parabener, per- och polyfluoralkylsubstanser (PFAS), bekämpningsmedel, stimulantia, en isoflavon och en industrikemikalie) på Hovgården, en avfallsanläggning i Uppsala (Sverige). De CECs som valdes ut analyserades med hjälp av fastfasextraktion, slamextraktion och ultra-högtrycks vätskekromatografi kopplad till masspektrometri. Huvudsyftet var att få en bredare förståelse om förekomsten av läkemedel i lakvatten (vilka läkemedel som fanns i vilka mängder och vart) på Hovgården, främst från en deponi och slamlagringsanläggning.

Förekomsten av läkemedel härrörde med största sannolikhet från lakvatten som bildats i slam från den gamla deponins sluttäckning och från slamlagringsanläggningen. 45 av de 56 analyserade läkemedlen detekterades i lakvatten från deponin, i medelkoncentrationer från 1,1 ng/L (fexofenadin och tramadol) till 4900 ng/L (erytromycin). Den totala medelkoncentrationen av läkemedel i lakvatten från den gamla deponins sluttäckning (23 000 ng/L) var 45 – 49 gånger större än den aktiva deponin. 48 läkemedel detekterades i lakvatten från slamlagringsanläggningen, i medelkoncentrationer från 1,3 ng/L (climbazole) till 3000 ng/L (desvenlafaxin). Den totala medelkoncentrationen av läkemedel varierade från 1500 ng/L (lakvatten från förorenad jord) till 16 000 ng/L (kombinerat lakvatten från slam och förorenad jord). Den totala medelkoncentrationen av läkemedel minskade med cirka 8,1 % efter avloppsreningsverkets sista behandlingssteg, jämfört med avloppsreningsverkets inflöde. Avloppsreningsverkets recipient hade den lägsta totala medelkoncentrationen av läkemedel (240 ng/L) av alla provtagningspunkter.

Det gick inte att avgöra om de erhållna resultaten gällande förekomsten av läkemedel var tillförlitliga, då detta var första gången som det utvärderades på Hovgården. Därför rekommenderades ytterligare undersökningar, genom att upprepa denna studie, innan implementering av ytterligare behandlingsmetoder. Det rekommenderades att även inkludera en riskkarakterisering för att utvärdera och bedöma om de erhållna koncentrationerna kan vara skadliga för den mottagande miljön.

Dessutom, förekomsten av CECs i avfall (och följaktligen vid avfallsanläggningar) beror på vår konsumtion samt bortskaffande av CECs. Detta inkluderar vår överanvändning/missbruk av läkemedel med påvisade konsekvenser för (akvatiska) miljön som indirekt kan skada människors hälsa, såsom ökad antimikrobiell resistens. Förekomsten av CECs i slam och avloppssystem skulle därmed kunna minimeras genom att även öka allmänhetens medvetenhet om konsekvenserna av våra handlingar gällande CECs.

Nyckelord: potentiellt hälso- eller miljöfarliga kemikalier; läkemedel; lakvatten; deponi; slam; avloppsreningsverk

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PREFACE

As the final step of the master's Program in Environmental and Water Engineering at Uppsala University and the Swedish University of Agricultural science (SLU), this master thesis was written and corresponded to 30 credits. The master thesis was conducted at SLU, in collaboration with Uppsala Vatten och Avfall AB. The supervisor was Dr Oksana Golovko, and the subject reviewer was Associate Professor Lutz Ahrens, both at the Department of Aquatic Sciences and Assessment at SLU in Uppsala.

A special thanks to Oksana for guiding and helping me throughout the project, as well as Lutz, for supporting and helping me when it was needed.

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POPULAR SCIENCE SUMMARY

Pharmaceuticals and other chemicals of emerging concern in leachate from a landfill and sludge storage facility

Victoria Runnäs

A variety of chemicals are today utilized worldwide for human or veterinary purposes, such as pharmaceuticals, per- and polyfluoroalkyl substances (PFASs), pesticides, stimulants, industrial chemicals etc. These groups of chemicals are however referred to as chemicals of emerging concern (CECs) that could have harmful effects on the health or environment. For example, pharmaceuticals have frequently been associated with causing hormone disruptions in fish, which could negatively affect their survival- and reproduction system over time. Pharmaceuticals have also been associated with causing increasing antimicrobial resistance and could therefore pose a public threat as well.

When CECs have been consumed and/or disposed of, they will be received to waste facilities as, for example, municipal solid waste or as incoming wastewater via sewage systems. The received waste will be treated to reduce contamination levels by having waste treatment. For example, wastewater treatment plants (WWTPs) and landfills, depending on the received waste type. WWTPs treats incoming wastewater (from e.g., domestic households and hospitals) by removing/degrading nutrients (such as nitrogen and phosphorus) and organic matter. The treated wastewater will then be discharged into the aquatic environment. Sludge will also be produced, which is a residue from the wastewater that has been treated. This sludge contains organic matter and nutrients, making it suitable to use as a soil improver or as a fertilizer for agricultural purposes. CECs are however challenging to remove/degrade in WWTPs since these chemicals tend to be persistent and resistant to degradation. CECs may therefore remain in unaltered forms or by only being partly removed/degraded in the treated wastewater and/or in the sludge after treatment. This means that CECs may be discharged into the aquatic environment and/or may be applied onto land. Whereas landfills are mainly utilized for decomposing municipal solid waste into non-biodegradable compounds that causes no harm on the environment or human health. Landfilling waste may however also contain CECs, due to the diverse waste types that are deposited (such as non-recyclable and hazardous waste).

If precipitation or any other moisture source encounters e.g., sludge or landfilling waste, this may generate leachate. Leachate is a liquid that contains dissolved organic matter (from the decomposed waste) that CECs may have sorbed to. This leachate may thereafter, if runoff is created, contaminate three major recipients including the surface water, groundwater and the soil zone. Which could for instance deteriorate the soil quality and harm local ecosystems.
Treating leachate in a controlled environment is therefore important to incorporate, in addition to waste treatment. For example, produced sludge from WWTPs must be treated before being applied to agricultural land. This can be performed by storing it openly in direct contact with the atmosphere at waste facility surfaces. Leachate will simultaneously be generated, which can be drained to a wastewater treatment plant (WWTP) for treatment. The purpose with this is to prevent/inhibit future decomposition from occurring after being applied to land. This reduces the risk of CECs being further transported to, for example, the three major recipients. Whereas conventional landfills are constructed with a barrier between the environment and the waste to prevent leachate from contaminating the soil and groundwater. In which covers can gradually be placed around and on top of the landfill, to prevent precipitation from passing through the waste and hence limit leachate from being generated. The amount of generated leachate can thus be prevented/reduced by having controlled leachate treatment.

Generated leachate within waste facilities can be drained to WWTPs for treatment via leachate collection systems. This is for example performed at Hovgården, a waste facility in Uppsala (Sweden). CECs may however still enter recipients depending on, for example, how efficient/properly managed the treatment methods are in removing/degrading CECs.

Furthermore, a large majority of pharmaceuticals have not been evaluated for their occurrence (such as which compounds that are present where and in what quantities), fate (where it will end up after use) or long-term toxicity in the environment (OECD 2019). They are therefore relatively new topics of interest to investigate.

Uppsala Vatten och Avfall AB owns Hovgården and were interested in gaining knowledge concerning the occurrence of pharmaceuticals at their waste facility. The aim of this master thesis was therefore to get a broader understanding regarding the occurrence of pharmaceuticals in leachate (which pharmaceuticals that are present in what quantities and where) at Hovgården. Particularly from a landfill and sludge storage facility. Other CECs were investigated as well, including parabens, PFASs, pesticides, stimulants, an isoflavone and an industrial chemical. The compounds that were selected to analyse were based on consumption in Sweden and worldwide with also including compounds that should be monitored when conducting this type of study.

The selected water sampling points corresponded to the waste facility's drainage system, which primarily collects leachate from the landfill and sludge storage facility. Water samples were also collected from the on-site WWTP as well as in the belonging recipient (surface water where the treated wastewater is discharged to). This was conducted to evaluate the occurrence of CECs before and after treatment. Solid sludge samples were also collected from the sludge storage facility, as it was of interest to investigate which compounds that remain in or that leach out from the sludge. All samples were then analysed using solid phase extraction, sludge extraction and ultra-high pressure liquid chromatography coupled to mass spectrometry, to evaluate the occurrence of CECs.

The occurrence of pharmaceuticals from the landfill and sludge storage facility most likely originated from leachate formed by sludge, since this is a common source for pharmaceuticals to be detected in. In addition, other CECs were also present in all sampling points. This study therefore showed that further monitoring and evaluation of the occurrence and dissemination of CECs should be performed since a variety were detected, including pharmaceuticals, PFASs and pesticides etc. Even more specifically in the sludge, treated wastewater and surface water, which are all potential sources for contaminating the (aquatic) environment. Recommendations were therefore suggested. The main recommendation was to conduct further screenings by repeating this study to evaluate whether the obtained results from this study were reliable, before implementing additional treatment methods. A risk characterization was also recommended to include in further screenings, to evaluate and assess how harmful the obtained concentrations may be for the receiving environment. In addition, the occurrence of CECs in sludge and sewage systems could be minimized by also increasing the public's awareness about the consequences of our actions regarding CECs.

ABBREVIATIONS AND DEFINITIONS

BOD	Biochemical oxygen demand. Measure of the amount of dissolved oxygen needed for oxygen-consuming organisms to be able to break down organic material in a certain amount of water.
CECs	Chemicals of emerging concern which are micropollutants (ranging in concentrations from a few ng/L up to several µg/L).
COD	Chemical oxygen demand. Measure of the total amount of dissolved organic carbon in water.
Detection frequency	How many compounds that were detected in a sampling point.
Dominating contributor	Compound with a mean composition ≥ 5.0 % in a sampling point.
dw	dry weight.
GS	Grab sampling.
FP / FPS	Flow-proportional / Flow-proportional sampling.
HSs	Hardened surfaces used for storing and/or treating waste, such as wood, compost and combustible waste.
LL	Landfill leachate.
Leachate	A liquid that consists of dissolved or suspended solids (from decomposed waste) and is generated from precipitation or any other moisture source that encounters the waste.
LOQ	Limit of quantification (lowest possible concentration that can be determined).
MBBR	Moving bed biofilm reactor.
Natural attenuation	Natural processes that convert pollutants to less harmful forms or immobilizes pollutants.
n.d.	no date (after referring to a source) or not detected (mean concentration ≤ LOQ).
Old landfill	No waste is deposited (no longer in use) and the landfill is completely covered.
PFASs	Per- and polyfluoroalkyl substances.
Positive / negative removal efficiency	Lower / higher CEC concentration in the WWTP effluent compared to the WWTP influent.

PP	Polypropylene.
Removal efficiency	The % of CECs that can be removed during wastewater treatment ((WWTP influent – WWTP effluent) / WWTP influent * 100).
SPE	Solid phase extraction.
Sludge cell	Synonym for "Sludge storage facility".
Sludge water	Leachate from sludge in the sludge cell.
Sewage sludge	Residue from wastewater that has been treated in a WWTP, or solid sludge samples from the sludge cell.
SP / SPs	Sampling point / Sampling points.
TI / TIS	Time-integrated / Time-integrated sampling.
UPLC-MS/MS	Ultra-high pressure liquid chromatography coupled to mass spectrometry.
WFD	Water Framework Directive.
WWTP / WWTPs	Wastewater treatment plant / Wastewater treatment plants.
WWTP effluent	Intermediate treatment step in a WWTP where treated wastewater is discharged for further treatment or where treated wastewater is discharged into a recipient after the final treatment step in a WWTP.
WWTP influent	Received wastewater that will be treated in a WWTP.
∑pharmaceuticals mean concentration	Total mean concentration of pharmaceuticals. Sum of every pharmaceutical's mean concentration in a sampling point.
∑pharmaceuticals mean concentration	Total mean composition of pharmaceuticals. Normalized mean concentration of every pharmaceutical in a sampling point.

TABLE OF CONTENTS

1 INTRODUCTION

Hundreds of thousands of tons of chemicals of emerging concern (CECs), such as pharmaceuticals, pesticides, industrial chemicals, personal care products, and per- and polyfluoroalkyl substances (PFASs), are annually consumed and dispensed worldwide. They are continuously released into the environment, in which concerns have emerged due to their potential bioaccumulative and toxic characteristics (Golovko et al. 2021). Many CECs are badly regulated or non-regulated and can therefore be harmful to humans and ecosystems (Li 2014). For instance, substances used in human medication may be non-regulated with environmental persistence and of toxic concerns. This could pose negative impacts on the (aquatic) environment due to, for example, incomplete removal in wastewater treatment plants (Golovko et al. 2021).

Uppsala Vatten och Avfall AB owns Hovgården, a waste facility with its own wastewater treatment plant (WWTP), sludge storage, landfill and other waste treatment/storage surfaces. The WWTP primarily treats generated leachate from the landfill and other waste treatment/storing surfaces.

In the 1970s and/or 1980s, some waste from the hospital in Uppsala was deposited at one of the landfills that is no longer active. Sludge has also been deposited at this landfill. In 2020, pharmaceuticals were analysed for the first time in leachate at Hovgården. Analyses of the samples revealed that the detection limit for most substances were very high and it was therefore not possible to determine whether there were significant concentrations in the water (unpublished data). Hence, there was still a knowledge gap concerning the occurrence of pharmaceuticals at Hovgården. A screening of pharmaceuticals and other CECs in water at Hovgården was therefore conducted in this study, with special attention to leachate from the landfill and the sludge storage facility.

This master thesis is a collaboration between Uppsala Vatten och Avfall AB and the Swedish University of Agricultural Sciences (SLU).

1.1 AIM AND GOALS OF THE STUDY

The aim of the master thesis was to get a broader understanding regarding the occurrence of pharmaceuticals in leachate at Hovgården, particularly from the landfill and sludge storage facility. It was also of great interest if the findings from the landfill should be considered a problem that needs measures. It was also of interest to examine which pharmaceuticals that remain in or that leach out in the sludge from the sludge storage facility. Analyses of solid sludge samples were therefore conducted as well.

1.2 LIMITATIONS

The primary subject of this master thesis was to focus on pharmaceuticals. The occurrence of other CECs (including parabens, PFASs, pesticides, stimulants, an isoflavone and an industrial chemical) in the leachate and sludge were also investigated, but less detailed discussed.

A risk characterization was not included in this study. The environmental risk of CECs to ecosystems was therefore not assessed.

2 THEORY

2.1 CHEMICALS OF EMERGING CONCERN

CECs, such as pharmaceuticals, personal care products, PFASs, pesticides and industrial chemicals are micropollutants that currently are ubiquitous in surface water and groundwater all over the world (Golovko et al. 2021). In which concerns have emerged due to their potential bioaccumulative and toxic characteristics. The concerns stems from widespread and increasing use of CECs, as well as from advances in analytical techniques that allows for the detection of chemicals at trace levels (ibid.). In which analytical methods capable of detecting CECs at trace levels have only been developed for the past 10 – 15 years, although these pollutants have been present for decades (Fernandes et al. 2021).

2.2 PHARMACEUTICALS

Pharmaceuticals are CECs that usually are polar molecules with at least one ionizable group and tend to be lipophilic or relatively soluble in water, whether natural or synthesized (Fernandes et al. 2021).

Pharmaceuticals can only be partially metabolized during therapeutic use (Frascaroli et al. 2021). Compounds may therefore be released into e.g., sewers unaltered or in the form of metabolites or conjugates after excretion, that eventually reach wastewater treatment plants (WWTPs). Furthermore, pharmaceuticals may remain active for a long time in WWTPs since most of them are designed to preserve their chemical structure throughout the therapeutic treatment (ibid.). In addition, pharmaceuticals are designed to be stable, to be able to reach and interact with target molecules. This means that they either degrade slowly or that constant use of pharmaceuticals causes a continuous discharge into the environment at rates that surpass the degradation rate. They are therefore of environmental concern, albeit at low concentrations, since they can still exhibit a pharmacological response at low doses with living systems (OECD 2019).

Pharmaceuticals are bioactive substances (Sharma et al. 2021) that have most notably been found in water bodies, soils and sediments (Puckowski et al. 2016). The concentration and impacts of pharmaceuticals in the environment are influenced by a combination of factors, such as their toxicity, degradation, persistence and mobility properties. As well as source and timing of the pollution, agriculture and veterinary practices, WWTP technology, operation and removal efficiency and the receiving environment's sensitivity and exposure history (OECD 2019).

2.2.1 Sources of pharmaceuticals

Landfills, animal-, freshwater aquaculture-, hospital-, industrial- and domestic waste are the six main pathways for pharmaceuticals to be discharged into the environment. The substances enter the environment (mainly through the surface water, groundwater and the soil zone) either via point or diffuse source pollutions (Li 2014).

Point sources are single identifiable and originates from separate locations (Li 2014), such as landfills and effluents from industries, hospitals, municipal WWTPs and septic tanks (small wastewater treatment system) (Lapworth et al. 2012; Li 2014). For example, wastewater (generated from e.g., industrial-, hospital-, aquaculture- and domestic waste) is mainly discharged to WWTPs via sewage systems and is recognized as a major point source to the soil zone and water resources (Li 2014). Of which

domestic wastewater has been acknowledged as the primary route of human pharmaceutical substances to be released into the aquatic environment (ibid.). Whereas landfills are a source of a wide range of substances due to the deposited waste, which could include expired or unwanted pharmaceuticals (Li 2014; Lu et al. 2016). Leachate generated from e.g., landfills could infiltrate through unsaturated zones into the groundwater and be discharged to the surface water (Li 2014). This can cause groundwater and surface water contamination, deteriorate soil quality and harm local ecosystems, if the generated leachate is not properly managed (Sil & Kumar 2017).

Diffuse sources occur across a wide range of geographical scales (such as in rural and urban areas) which may be difficult to identify (Li 2014). For example, leakage from waste treatment plants and systems, urban runoff from domestic waste and agricultural runoff from animal waste and manure. Sewage sludge is also a diffuse source and one of the more prominent causes of pharmaceuticals entering soils and freshwater resources. For instance, the sludge may be applied to land as a soil improver which may contain CECs that are unaltered or inadequately removed by WWTPs (ibid.). In addition, generated runoff water from sludge could for example contaminate water bodies if the sludge is used as a fertilizer in agricultural areas. The sludge can also be incinerated or disposed of at landfills, which could cause groundwater contamination (Golovko et al. 2021).

Diffuse sources generally have lower environmental loading due to a higher potential for natural attenuation in the soil and subsurface, compared to point sources (Lapworth et al. 2012; Li 2014).

2.2.2 Effects on and via the environment

The presence and occurrence of CECs in the environment can have severe effects on the (aquatic) environment and its species. Effects that are frequently associated with pharmaceuticals include increasing antibiotic resistance of microorganisms, acute or chronic toxicity and uncertainties concerning transformation products and metabolites. Pharmaceuticals could also cause other negative effects to non-target organisms, such as hormone disruptions in invertebrates and fish (Golovko et al. 2021).

The toxicity, persistence and non-biodegradability that many CECs can exhibit causes management issues, which can harm the (aquatic) environment and its species. As well as non-persistent compounds, if their continuous discharge into the environment surpasses their natural attenuation and transformation (Fernandes et al. 2021). For example, a continuous discharge of incompletely treated wastewater could have unpredictable ecological effects when mixed with other chemicals that are present in the environment. Most pharmaceuticals also have a high solubility, which causes aquatic organisms to be more receptive to their potential toxic and ecological effects (Li 2014).

The most common pharmaceuticals groups that have been identified in the soil zone, groundwater and surface water includes antibiotics, antihypertensives, antiepileptics, hormones and non-steroidal anti-inflammatory drugs (NSAID) such as analgesics (Li 2014). Some pharmaceuticals are typically used for human (e.g., analgesics, antibiotics, antidepressants, antineoplastics and hormones) or veterinary purposes (e.g., antibiotics, hormones and parasiticides). Which have been expressed as pharmaceuticals of great concern due to their unfavorable consequences on ecosystems in the environment, such as mortality and alterations to physiology, behavior and reproduction (OECD 2019).

Some ***analgesics***, such as diclofenac, could damage organs and decrease the likelihood of hatching success in fish as well as cause oxidative stress, genotoxicity and neurotoxicity in mollusks (OECD 2019). It could also cause hormone disruptions in frogs and fish (Li 2014; OECD 2019). For example, structural disruptions on fish organs could affect genes that control metabolism, which over time could negatively affect the survival- and reproduction system (Li 2014). Some ***antiepileptics*** could cause reproduction toxicity in invertebrates and development delay in fish, such as carbamazepine, which have frequently been detected in wastewater effluents (Li 2014; OECD 2019). Carbamazepine could also transform to a variety of intermediates and create biologically active chemicals via transformation reactions that occurs in soil. This may cause groundwater contamination, if transferred from the soil, in which its metabolites have been shown to cause teratogenic effects (Li 2014). Some ***anti-depressants*** could alter fish behavior, such as sertraline, making them less fearful of danger and more vulnerable to predators. Other effects involve hormone disruptions in fish as well as reproduction toxicity in invertebrates. This also concerns other psychiatric drugs such as fluoxetine and citalopram (OECD 2019). The overuse of ***antibiotics*** has been associated with causing antimicrobial resistance in humans and animals, which is the microbe's ability to withstand the effects of medication that normally would destroy or inhibit it (ibid.). The overuse/misuse of antibiotics could therefore lead to ineffective treatments which poses a public health risk, due to the formation and spread of antibiotic resistant bacteria (EFSA n.d.). Antibiotics could also reduce the growth of environmental bacteria, algae and aquatic plants (OECD 2019). For example, an induced chronic toxicity in algae could reduce its photosynthesis rate if the chloroplast functions are affected. Which increased concentrations of carbamazepine (antiepileptic) and diclofenac (analgesic) have also been reported to cause (Li 2014). Some ***antihypertensives*** such as beta blockers (e.g., propranolol) could alter the reproduction behavior in fish and reproduction toxicity in invertebrates (OECD 2019). Some ***hormones*** could affect the ability to break down pollutants (Li 2014) and have also caused feminization of fish and amphibians (Li 2014; European Commission 2019; OECD 2019). This could affect the population's ability to reproduce as a result of the endocrine's system effects (European Commission 2019). It could also increase the risk of breast or prostate cancer in humans (OECD 2019). Some ***antineoplastic*** could cause genotoxicity, mutagenicity, carcinogenicity and toxicity to fetus (ibid.).

Furthermore, humans may be exposed to pharmaceutical residues through ingestion via e.g., water, plant crops, seafood and meat (OECD 2019). Significant correlations between current pharmaceuticals and direct effects on human health has however not been established yet (European Commission 2019). A plan was put into action in 2020 by the European Commission, to address the potential effects that low doses of pharmaceuticals via the environment can have on humans (European Commission 2020b).

2.2.3 Fate and occurrence of pharmaceuticals in the environment

The fate of pharmaceuticals in the environment is generally governed by their physico-chemical properties such as its solubility in water. Also by the degree of natural attenuation (Li 2014), depending on the pharmaceutical's hydrophobicity and biodegradability and on the temperature. In which metabolization and removal via natural attenuation will lead pharmaceuticals to be present in low concentrations in the environment (WHO 2011). This includes when, for example, dispersion/dilution, sorption, biodegradation and abiotic degradation (such as photolysis, hydrolysis and volatilization) occurs (Bjerg et al. 2014; Fernandes et al. 2021). Furthermore, a lower temperature causes a decrease in biodegradation reactions (Fernandes et al. 2021).

In addition, although some pharmaceuticals can be partially removed via natural attenuation, others are resistant to degradation and environmentally persistent (Li 2014). Moreover, many pharmaceuticals are not readily biodegradable in natural waters, particularly in anaerobic conditions. Their biodegradability may therefore take several months since microorganisms are essential for biodegradation reactions to occur. The occurrence of pharmaceuticals in the soil zone and water resources is therefore ubiquitous, due to many common pharmaceuticals being resistant to degradation and/or not being completely removed via natural attenuation (ibid.).

2.3 WASTEWATER TREATMENT PLANTS

A WWTP is the physical infrastructure used to treat wastewater (Ameta 2018). The WWTP will separate incoming wastewater into treated wastewater (which will be discharged into the aquatic environment) and sewage sludge (Wennmalm 2011).

WWTPs generally has a low removal efficiency of pharmaceuticals, since WWTPs were designed to remove suspended solids, pathogens, organic matter and nutrients, excluding CECs. Pharmaceuticals can therefore pass through conventional WWTPs unaltered for example, due to their moderate to high solubility and degradation resistance during the biological and chemical processes (Fernandes et al. 2021). In which excreted and unused pharmaceuticals that for instance have been disposed into sinks and toilets may be found in the WWTP effluent (European Commission 2019). Conventional WWTPs have therefore been recognized as major/main sources of discharging CECs into aquatic environments, due to the incomplete removal (Golovko et al. 2020, 2021).

Different technical solutions have been evaluated in Sweden to improve the removal efficiency of pharmaceuticals (and other CECs) in the (aquatic) environment, such as ozonation or filtration through activated carbon as well as various combination solutions (Havs och Vattenmyndigheten 2018). However, the most advanced treatments are still not 100 % effective in removing pharmaceuticals (European Commission 2019).

2.3.1 Fate and transport of pharmaceuticals in WWTPs

The fate and transport of pharmaceuticals (and other CECs) depends on several factors, such as consumption, disposal, manufacturing volume, stability as well as the removal and transformation at WWTPs (Golovko et al. 2021).

The fate of pharmaceuticals during wastewater treatment can be controlled, depending on the removal efficiency, which is affected by different factors (Chavoshani et al. 2020). This mainly includes the CECs physico-chemical properties and the operating

conditions such as hydraulic retention time (time allowed for biodegradation and sorption), temperature and redox conditions (aerobic/anaerobic/anoxic) (ibid.). Also, the characteristics of the wastewater such as the concentration of the organic matter, ionic strength and pH. Furthermore, biotransformation and sorption are examples of factors that are more related to the CECs properties and to the potential for removal in WWTPs (ibid.).

2.3.1.1 Biotransformation (biodegradation)

Biotransformation occurs when CECs are accumulated by cell and biodegraded by bacterial enzymes, of which the CECs structure can strongly affect the resistance to biodegradation. Easily degraded substances generally include linear compounds with short-branched chains, unsaturated aliphatic compounds and compounds with electron donating functional groups. Whereas persistent CECs contain compounds with long-branched side chains, that are saturated or polycyclic compounds and compounds with sulfate, halogen or electron withdrawing functional groups (Chavoshani et al. 2020).

The removal of CECs via biodegradation can occur directly, which is carried out by a portion of the total biomass capable of degrading CECs. Also, during conversion of macropollutants. In which a co-substrate is added to serve as a growth substrate (such as ammonium or molecules that are readily biodegradable) for the biodegradation of CECs. A simultaneous degradation of the co-substrate and CECs occurs, due to the enzyme's ability to degrade many substrates (Pomiès et al. 2013; Chavoshani et al. 2020). The concentrations of CECs determine the type of biodegradation that occur. CECs are not adequate to promote growth of biomass since they are micropollutants and therefore needs energy for biotransformation to be generated. A co-substrate is therefore needed to induce the biotransformation by enzymes and will lead to persistent compounds being converted to more biotransformable intermediate products. Whereas the type and activity of the biomass (depending on the redox conditions) determines the degree of biotransformation (Alvarino et al. 2018).

2.3.1.2 Sorption

Sorption is a function of the CECs physico-chemical properties and of the sorbent agent's characteristics (Chavoshani et al. 2020) and comprises of two mechanisms; i) absorption and ii) adsorption (Sipma et al. 2010; Chavoshani et al. 2020). ***Absorption*** occurs by CECs migrating from the aqueous phase into the lipid fraction of the sludge or into the lipophilic cell membrane of the biomass, due to their hydro-phobicity. Compounds with a medium or high lipophilic nature tend to absorb onto sludge, independently of the biomass (Alvarino et al. 2018; Chavoshani et al. 2020). For example, pharmaceuticals tend to have high sorption affinity to sludge due to their lipophilic nature (Golovko et al. 2021). ***Adsorption*** occurs by CECs being retained on the solid surface due to electrostatic interactions between positively charged compounds and the negatively charged surface of biomass cells. In which ionized or dissociated CECs in the aqueous phase adsorb to the biomass (Alvarino et al. 2018; Chavoshani et al. 2020).

The octanol-water partition coefficient (K_{ow}) is frequently used to predict CECs sorption potential to solids such as sludge (Luo et al. 2014; Chavoshani et al. 2020). Compounds generally have a low sorption potential if log K_{ow} < 2.5, a moderate sorption potential if 2.5 < log K_{ow} < 4.0 and a strong sorption potential if log K_{ow} > 4.0 (Rogers 1996; Luo et al. 2014; Chavoshani et al. 2020). Compounds with strong

sorption potential (i.e., high hydrophobicity) binds more easily to sediments and suspended particulate material (Golovko et al. 2020). Hydrophobic compounds with a strong sorption potential (i.e., medium or high lipophilic behavior) have also been found to sorb to biomass (such as algae) and sludge (Alvarino et al. 2018; Gruchlik et al. 2018; Chavoshani et al. 2020). Whereas strongly or moderately hydrophilic compounds are unlikely to extensively sorb to organic matter in, for example, sedimentation ponds. These compounds will instead remain in the dissolved phase, with biodegradation and/ or photodegradation as the main mechanisms (Gruchlik et al. 2018).

The organic carbon-water sorption partioning coefficient (K_{oc}) and the sediment-water partition coefficient (K_d) can also be used to predict the fate and transport of CECs (Golovko et al. 2020). These coefficients have previously been used in studies to evaluate the adsorption capacity of chemical compounds in water and sediment (ibid.). For example, CECs with low log K_d or K_{oc} values (< 2.0) have been proposed to have high mobility in the solid phase and are more likely to exist in the aqueous phase. Whereas those with high log K_d or K_{oc} values are firmly adsorbed onto sediment or organic matter. These suggestions are however not always accurate depending on, for example, how persistent the CECs are in the water. In which carbamazepine has a moderate sorption to sediment (log K_d = 1.0 – 1.7) but can still be commonly detected in water, due to its high persistence in aquatic environments (ibid.).

2.4 SEWAGE SLUDGE

Sewage sludge ("biosolid") is a residue produced from treated wastewater by WWTPs that can contain unaltered, or only partially removed, pollutants (Li 2014).

The sludge type can be divided into three categories: i) primary ii) secondary/ activated and iii) tertiary/chemical sludge (Canziani & Spinosa 2019). The ***primary sludge*** is produced from mechanical and preliminary treatments by using gravity-settling units (ibid.). In which settleable solids i.e., the primary sludge, can be removed. This sludge generally has high content of readily biodegradable organic matter and may contain all known human pathogens due to the sorption on solid particles, as well as hydrophobic compounds (Pomiès et al. 2013; Canziani & Spinosa 2019). The ***secondary/activated sludge*** (excess growth of biomass) is produced from biological treatments (Canziani & Spinosa 2019). Which is performed since the wastewater will still be rich in biodegradable organic matter (and with high concentration levels of pollutants) (ibid.) and thus have high oxygen demand (Spinosa & Vesilind 2001). This is to be removed in this treatment step (ibid.) by generally adding biomass (containing "flocs") to assimilate the organic matter (Canziani & Spinosa 2019). The oxygen demand will be reduced and a secondary/activated sludge will simultaneously be produced. This sludge mainly consists of water with poor dewaterability since it is rich in volatile solids (ibid.), which is difficult to treat in WWTPs (Spinosa & Vesilind 2001). It also contains CECs, due to sorption to the sludge and biodegradation (Pomiès et al. 2013). The ***tertiary/chemical sludge*** is produced from final physical-chemical treatments, to improve the quality of the effluent (Canziani & Spinosa 2019). In which chemical nutrient removal or tertiary/advanced treatments are applied (such as coagulation and flocculation followed by sedimentation) to produce this sludge. This sludge's characteristics varies since it depends on the treatment option. There is a last step (such as disinfection by chlorine or ultraviolet radiation), but no sludge is produced (ibid.).

In addition, conventional WWTP typically use activated sludge process as a biological treatment to remove pharmaceuticals, whereas advanced facilities use tertiary treatment methods such as reverse osmosis, ozonation and advanced oxidation technologies (WHO 2011).

Sewage sludge contains valuable nutrients, making it suitable to use as a fertilizer or soil improver (European Commission n.d.b). About 200 000 tons of sewage sludge are annually produced in Sweden, with the majority being utilized for spread on arable land, topsoil production and to cover landfills (Statens Offentliga Utredningar 2020). 39 % of total sewage sludge was for example applied as agricultural input on farms in 2018 (Statistiska Centralbyrån 2018).

Sewage sludge that is going to be spread on agricultural land must be treated beforehand to reduce the risk of spreading unwanted pathogens, which can only be performed by certified treatment plants (Svenskt Vatten 2020a; Jordbruksverket 2021). In which specific hygiene requirements must be met if a treatment plant is supposed to be, for example, Revaq-certified. Either by storing the sludge for at least six months or by applying one of the hygiene methods specified in Naturvårdsverket's report 6580 "*Hållbar återföring av fosfor*" (Svenskt Vatten 2021).

2.5 LANDFILL

A landfill is a location for storing waste that is either on or in the ground. This does not apply to waste that is stored for less than three years before recycling or for less than one year before being disposed of at a landfill, nor to waste that is prepared for transit to another location for treatment (*Miljöbalk*, SFS 1998:808).

Municipal solid waste is mainly disposed of at "sanitary" landfills in most countries in the world (Youcai 2018a). This waste is allowed to biologically and chemically degrade into inert compounds in an enclosed environment, by using modern engineering (Hossain et al. 2011).

A conventional landfill functions as a waste containment unit with liner systems (Woodard & Curran 2006). The liner systems are constructed to form a barrier between the environment and the waste, as well as to drain leachate to treatment facilities via leachate collection systems. Which is performed to prevent soil and groundwater from being contaminated (Chabuk et al. 2018). The liner system and cover prevent rainwater, or any other moisture source, from percolating into the landfill which limits organic degradation. Thus leachate generation reduces, but can cause increased release of landfill gas and leachate if these barriers break (Mandal et al. 2017). The covering on the landfill can increase as it ages, seals can be placed around and clean groundwater and surface water will be diverted. The landfill is no longer active when it is completely covered, waste is no longer received and active measures for control and emission limitations are no longer needed (Naturvårdsverket 2008). Operating conditions can be optimized for waste degradation, such as recirculating leachate, adding certain supplemental liquids and injecting air to stimulate the microbial activity and thus accelerate the biological and chemical reactions (Mandal et al. 2017).

2.5.1 Landfill regulations in Sweden

The amount of deposited waste has fluctuated throughout time due to landfill regulations and changing societal consumption habits (Naturvårdsverket n.d.b). For instance, waste that is now categorized as non-hazardous (e.g., from households and industries) was once sent to landfills in Sweden. After the environmental protection act was enacted in 1969, landfill activities were subject to a permission requirement and hence more controlled (ibid.). The depositing of combustible waste has been prohibited since 2002, with a restriction expanded to include all organic waste since 2005 (Naturvårdsverket n.d.a). All landfill activities in Sweden must now be regulated in accordance with the Swedish Environmental Code (*Miljöbalk*, SFS 1998:808).

The designs of landfills must meet certain requirements throughout their entire life cycle in accordance with the Swedish Landfill Ordinance, to prevent and reduce the negative effects that waste landfilling may pose on the human health and the environment. Particularly in terms of polluting the global environment, soil, air, surface- and groundwater (*Förordning om deponering av avfall*, SFS 1998:808).

2.6 LEACHATE

Leachate is a liquid that consists of dissolved or suspended solids and is generated from aerobic and anaerobic microbial decomposition of waste (mainly municipal solid waste) (Sil & Kumar 2017). The leachate is formed in landfills, transfer stations, incineration- and composting plants for example, due to physical, chemical and biological changes (Youcai 2018b).

Moisture, location, transport and treatment methods all influence the quantity of leachate. The amount (from landfills) mainly depends on the precipitation-, surface runoff-, surface irrigation- and groundwater infiltration, free water in waste, leachate from covering materials and leachate produced from decomposed organic matter (Youcai 2018a). Rainwater percolation (into the waste) is the primary source of generating leachate, especially on top of a landfill or at open dumping sites (Sil & Kumar 2017). Soluble pollutants will transfer into the liquid phase from the solid phase when the rainwater infiltrates. The organic matter (in the waste) will simultaneously degrade into dissolved organic matter, such as volatile fatty acids (VFAs), by microbes and transfer into the leachate (Youcai 2018a).

In addition, factors that more specifically affect the characteristics of landfill leachate (LL) depends on the landfill age, type and moisture content of the deposited waste, waste composition, site hydrogeology, variations in seasonal weather, dilution with rain and the degree of decomposition within the landfill (Mandal et al. 2017). In which LL can be divided into three landfill ages; i) young (in operation/closed less than 5 y), which is highly biodegradable, ii) intermediate (closed for more than 5 y, but less than 10 y) and iii) mature (closed for more than 10 y), which is less biodegradable (Brennan et al. 2017; Mandal et al. 2017). The leachate's high levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) decreases as the landfill ages (ibid.). The biodegradability index (BOD/COD) also decreases when organics of greater molecular weight are released during waste degradation (Mandal et al. 2017).

Waste decomposition occurs through a series of chemical and biological processes with four phases; i) an initial transition (aerobic), ii) an acidogenic (anaerobic), iii) a methanogenic and iv) a final maturation phase (Mandal et al. 2017). Higher concentrations of VFAs of lesser molecular weight are found in the initial transition phase (ibid.). These VFAs will then degrade under anaerobic conditions in the methanogenic phase. The BOD/COD will also decrease when all readily available biodegradable compounds have been degraded. Which indicates that remaining compounds of recalcitrant or bio-refractory nature are present. For example, humic and fulvic acids in mature leachate remain, due to resisting microbial decomposition. The BOD/COD also decreases during the post-closure period as the landfill ages (ibid.). In addition, a complete degradation in landfills can take decades up to hundreds of years, before being transformed to leachate (and landfill gas) (Youcai 2018a).

The degradation of leachate can be performed by several microorganisms, such as methanogenic-, nitrifying-, denitrifying-, sulfate reducing- and pathogenic bacteria. However, high concentrations of heavy metals and ammonia-nitrogen can create an imbalance of microbial nutrient elements, thus inhibiting the microbes growth and regeneration (Youcai 2018a). Also, stabilization of organics and degradation of certain higher molecular organic compounds can become challenging for microbes as the BOD/COD decreases (Mandal et al. 2017).

2.6.1 Leachate treatment

Leachate treatment can be performed, in which conventional treatments of e.g., LL can be divided into three groups; i) leachate transfer (such as combined treatment with domestic wastewater), ii) biodegradation (aerobic/anaerobic processes) and iii) chemical and physical methods (adsorption, coagulation/flocculation, sedimentation/flotation, air stripping, chemical oxidation and precipitation) (Renou et al. 2008).

It can however be challenging to treat leachate. Treatment of LL is for example influenced by its diverse composition and quantity as well as the toxicity of ammonia and heavy metals, the presence of bio-refractory compounds and the variation in pollutant concentrations across the leachate generation period (Mandal et al. 2017). Various management technologies have, and can, be applied to improve organic degradation (Mandal et al. 2017; Sil & Kumar 2017; Youcai 2018a). The most widely used combination for LL treatment is a biological process with nitrification and denitrification steps followed by membrane technology. But it may create an inhibitory effect and reduce the process efficiency of the microbial system due to certain toxic substances. Also due to the BOD/COD, since it decreases as the landfill ages (Mandal et al. 2017).

2.7 LIMIT VALUES AND WATCHLISTS IN SWEDEN

Some CECs in various environmental matrices are for example included in the EUs Water Framework Directive (WFD). Which includes a watch list of priority substances (based on risk) for action at EU level with the purpose of load reduction of those substances (European Commission n.d.a). The latest version of the watch list includes e.g., antibiotics (trimethoprim and sulfamethoxazole), antidepressants (venlafaxine and its metabolite desvenlafaxine) and azole compounds such as antifungals and pesticides (fluconazole, clotrimazole, miconazole, imazalil, metconazole, penconazole, prochloraz, tebuconazole and tetraconazole) (European Commission 2020a). The WFD is applied to all water districts in Europe with the same rules and assessments to ensure good water quality and its watchlist must be monitored by all countries within the EU (Svenskt Vatten 2020b). Each EU country can create its own lists and set limits for the most polluting substances. The Swedish Agency for Marine and Water Management determines which substances that are particularly polluting in the inland surface water, coastal water and water in transition zones in Sweden. These substances are emitted in significant amounts that pollute the aquatic environment, such as diclofenac (analgesic). Sweden's surface water is measured to ensure good water quality and is classified accordingly to the Swedish Agency for Marine and Water Management's regulations on classification and environmental quality standards regarding water (ibid.).

Pollutants identified in sewage sludge may also be included in various watchlists, such as the WFD as well as PRIO (database from the Swedish Chemical Agency). PRIO includes substances of what a Revaq-certified treatment plant in Sweden needs to monitor (Thorsén et al. 2020). For instance, salicylic acid (analgesic), pesticides (imazalil, metconazole, penconazole, prochloraz and tebuconazole) and PFASs can be found in the PRIO-database.

Furthermore, all EU countries must by January 12th 2023 adhere to a new EU drinking water directive in the national drinking water regulations that includes limit values of PFASs (Livsmedelsverket 2021).

3 MATERIAL AND METHODS

3.1 SELECTED TARGET COMPOUNDS AND CHEMICALS

56 pharmaceuticals and 30 other CECs were chosen as the target compounds to analyse in the field samples in this study. The other CECs comprised of an industrial chemical (*n = 1*), an isoflavone (*n = 1*), parabens (*n* = 3), pesticides (*n = 10*), PFASs (*n = 13*) and stimulants (*n = 2*). The selected target compounds were based on consumption in Sweden and worldwide, with also including compounds that should be monitored when conducting this type of study. See Table A1.1, A.1.2 and A2 in Appendix for details.

The native reference standards were purchased from Sigma-Aldrich (Sweden). The isotopically labeled internal standards were purchased from Wellington laboratories (Canada), Teknolab AB (Kungsbacka, Sweden), Sigma-Aldrich and Toronto Research Chemicals (Toronto, Canada). All analytical standards were of high analytical quality (> 95%). Detailed information regarding purchased substances can be found elsewhere (Golovko et al., 2020; Sörengård et al. 2019). Methanol, acetonitrile and formic acid were purchased from Sigma-Aldrich (Sweden) of high analytical quality. Milli Q Advantage Ultrapure Water purification system (Merk Millipore, Billercia, MA) produced the ultra-filtered water using a 0.22 µm Millipak Express membrane and an LC-PAK polishing unit (Merk Millipore, Billercia, MA).

3.2 SITE DESCRIPTION OF HOVGÅRDEN

Hovgården's waste facility has been in operation since 1971 and occupies an area of 57 ha northeast of Uppsala, Sweden. Waste is received from Uppsala's municipality and surrounding area, mainly from recycling centers and companies. Hovgården then handles the sorting, intermediate storage and treatment of waste on hardened surfaces (Uppsala Vatten och Avfall 2021a). The waste facility comprises of a landfill (old and active), a soil cell, a sludge cell, a storage, a sorting platform, a recycling center and a WWTP with belonging ponds. There are also separate storage areas for wood, combustible, compost and organic waste (Figure 1).

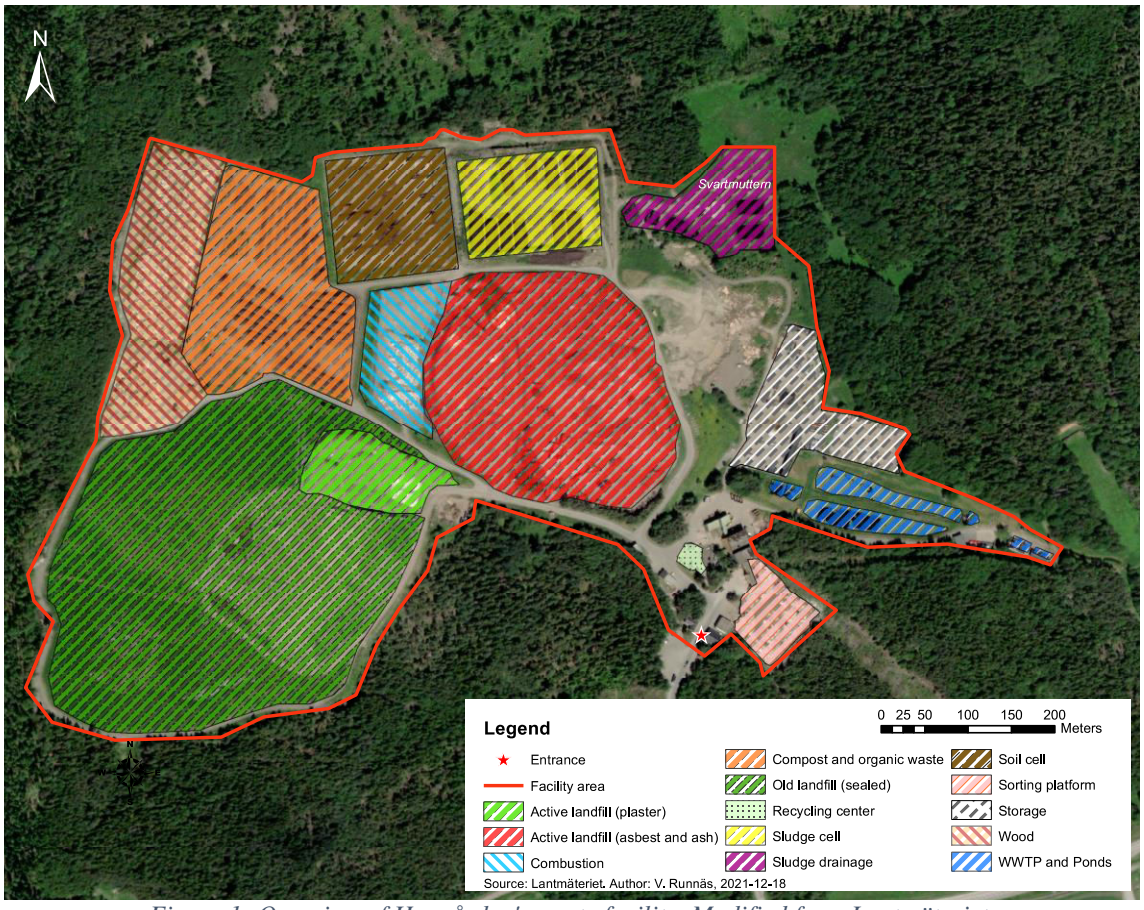

Figure 1: Overview of Hovgården's waste facility. Modified from Lantmäteriet.

A recycling center and sorting platform is situated in the southern part close to the entrance gate at Hovgården. The sorting platform is an intermediate storage that receives and sort industrial waste (Uppsala Vatten och Avfall 2021a) into different fractions such as combustible waste, wood, metal, isolation and landfill residues (S. Bjälkefur, personal communication, Dec 2, 2021). The different fractions are thereafter disposed to separate hardened surfaces. The recycling center receives bulky waste types from Uppsala's residents and small fractions of hazardous waste, which is temporarily stored in a special container, close to the sorting platform (Uppsala Vatten och Avfall 2021a).

A storage is located southeast of the facility close to the WWTP where, for example, metal, drainage pipes and diesel tanks are stored (Bonnet 2017).

The landfills (an old and an active) are located southwest/in the middle of the facility's area. The old landfill was in use from 1971 up to the turn of the year 2008/2009 and was completely covered in 2019 and is therefore not in use anymore. Industrial waste and incineration ash have mainly been deposited there as well as contaminated masses, digested sludge and sorted bottom ash used for the final coverage (Bjälkefur 2019). However, it is unknown if other waste types have been deposited here, but it is classified as a landfill for hazardous waste (Uppsala Vatten och Avfall 2021a). The active landfill functions as a conventional landfill, where waste such as asbestos, ash (originating from incinerated municipal household waste) and inert waste (such as plaster) are deposited (ibid.).

Compost and organic waste are deposited on the northwestern part of the facility. Garden, park and other residual organic waste are treated by composting and the finished soil is mostly used inside the area. Food waste from private households is only treated here in the event of operational disruptions in the biogas plant at Kungsängens gård (Uppsala Vatten och Avfall 2021a).

Wood and combustible waste are temporarily stored on separate hardened surfaces (pending transportation to incineration and energy recovery) near to the compost and organic waste area (Uppsala Vatten och Avfall 2021a). Combustible waste includes all possible waste (except from households) such as combustible fractions from the sorting platform (S. Bjälkefur, personal communication, Dec 2, 2021).

The soil and sludge cell are located on the northern part of the facility. The soil cell functions as a remedial surface for e.g., excavated petroleum-contaminated masses which are treated by composting (to reduce contamination levels). The materials are then used for leveling or other construction inside the plant. The sludge cell functions as an intermediate storage for sewage sludge, to eventually be used as fertilizer in agriculture (Uppsala Vatten och Avfall 2021a), if it is Revaq-certified. The sludge originates from treated wastewater at Kungsängsverket and the smaller WWTP Storvreta, which has been dewatered after anaerobic digestion before being transported to Hovgården (Uppsala Vatten och Avfall 2021b; c). Water from both cells is led via a sludge drainage through a sludge separator on to two sedimentation ponds and after that into two complementary ponds. A pumphouse at Svartmuttern (see Figure 1) connects this area to the remainder of Hovgården's WWTP (Uppsala Vatten och Avfall 2021a).

Untreated water at Hovgården arises partly as leachate from the landfill and partly as leachate formed by surface runoff water from all the hardened surfaces with waste treatment/storing. In which the generated leachate can be divided into two main streams. The first stream is leachate from the landfill, a sorting platform and storage areas for wood, compost and combustible waste. The LL is collected with drainage pipes at the bottom of the landfill (old and active), whereas LL generated from the top of the old landfill and leachate from the hardened surfaces is collected via pipes and surface water ditches (Uppsala Vatten och Avfall 2021a). The second stream is leachate from the soil cell and the sludge cell with a belonging sludge drainage system (Uppsala Vatten och Avfall 2018). The two streams are thereafter mixed at a pumpstation after the second stream's water has left the pumphouse at Svartmuttern (ibid.). The mixed water from both streams will then enter the "WWTP and Ponds" (Uppsala Vatten och Avfall 2021a), which is located southeast of the area (Figure 1).

3.2.1 Wastewater treatment process at Hovgården

The wastewater is treated by applying several separation methods, which mainly includes a mechanical purification step, bio-step, phosphorus precipitation and a sedimentation/polishing step (Uppsala Vatten och Avfall 2021a). The water is aerated in the mechanical purification step and an oxidation of iron, manganese and to some extent organic substances (ibid.) takes place and forms flocs. The water will thereafter pass through a lamella sedimentation after the aeration (but before the bio-step), where flocs such as oxides and other particles are separated (Uppsala Vatten och Avfall 2018). The water will then pass through a moving bed biofilm reactor (MBBR), which is the bio-step. The MBBR comprises of two basins, one of which containing plastic carriers, providing a surface for microorganisms (nitrifiers) to grow on. A degradation of organic compounds, oxygen demand and ammonium-nitrogen will be achieved via nitrification by the help of nitrifiers (Bonnet 2017; Uppsala Vatten och Avfall 2018). Denitrification it not performed at Hovgården. The water will then pass through a phosphorus precipitation step. This was put into operation in January 2020 to reduce the concentration of phosphorus in the water, by adding polyaluminium chloride and a polymer (Uppsala Vatten och Avfall 2021a). The water is thereafter directed to a sedimentation pond, where any leftover flocs and sediments can settle (Bonnet 2017). The water is then led to two large parallel polishing ponds for additional purification where bacteria and plants bind organic and inorganic substances to grow (Bonnet 2017; Uppsala Vatten och Avfall 2021a). The water is then led to an oxidation pond to increase oxygen content (to avoid reductive conditions in the receiving water course) (Bonnet 2017). The treated water is thereafter discharged via Hovgårdsbäcken to the recipient Funbosjön (Uppsala Vatten och Avfall 2021a).

3.3 SELECTION OF SAMPLING POINTS AND SAMPLE COLLECTION

Sampling points (SPs) corresponding to the drainage system at Hovgården were selected, to evaluate the CECs occurrence in water. SPs of sewage sludge were also selected, as a complementary, to evaluate which compounds that remain in or leach out from the sludge in the sludge cell. The selection of SPs was also influenced by a similar project conducted at Hovgården in 2016 (Bonnet 2017). The water and sewage sludge SPs are presented in Figure 2.

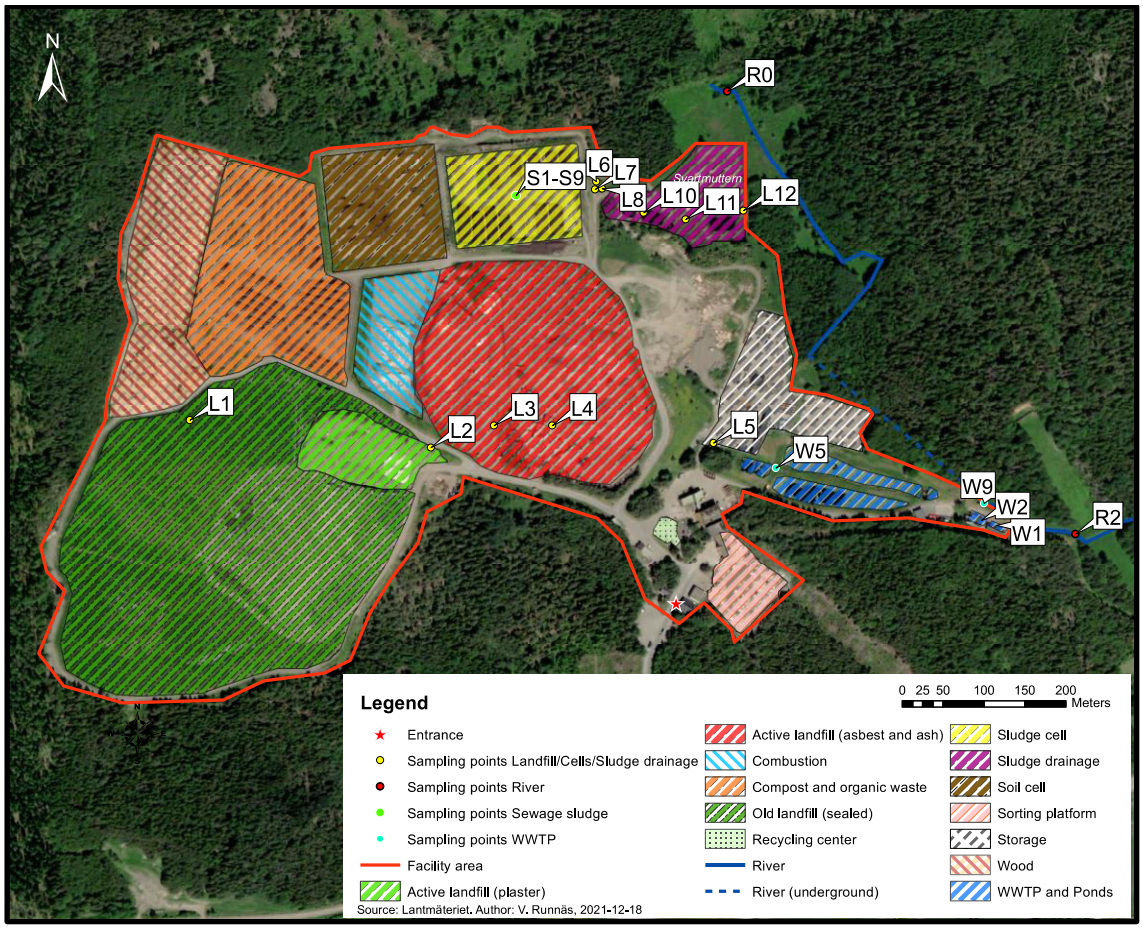

Figure 2: Overview of selected sampling points at Hovgården. L = leachate samples. W = wastewater samples from the WWTP and Ponds. S = sewage sludge samples. R = river samples from Hovgårdsbäcken. Modified from Lantmäteriet.

3.3.1 Sampling

All water samples were collected in 1 L polypropylene (PP) bottles (n_{tot} = *40*), that had been pre-rinsed with ethanol (EtOH) three times in the laboratory. Three different methods were used to collect water samples, including grab sampling (GS), time-integrated sampling (TIS) and flow-proportional sampling (FPS).

The GS method was performed for all water sampling points with a corresponding duplicate (n = *32*). L1 was excluded, due to no water accessible. An extender and a rope with a weight in the lower end (that would be attached to the 1 L PP bottle) was prepared and used if the water was not easily accessible on site.

Time-integrated (TI) autosamplers from SLU (GLS Portable Sampler, Teledyne ISCO) (n = *5*) were used for the TIS. The TI autosamplers were pre-calibrated and the container inside each autosampler was pre-rinsed with EtOH and warm tap water three times each, in the laboratory. In addition, TI autosamplers (n = *2*) (BÜHLER 1027, Hach) and a flow-proportional (FP) autosampler (n = *1*) (mjk780, MJK Automation

AB), were already on site and prepared by Uppsala Vatten. No duplicates were collected for the TIS and FPS.

Solid sludge samples were collected as grab samples in 250 mL PP bottles (n = 9), with no duplicates.

The sampling was carried out on September 22, 2021, at Hovgården's waste facility. The TIS and FPS were installed the same day as the GS was performed and collected the day after on September 23, 2021. The TI autosamplers were programmed on site to collect 50 mL water per 15 min during 24 h (9400 mL in total flow volume) to form one composite sample. The FP autosampler was programmed to collect 50 mL water for every 19 m^3 water volume that passes through per hour during 24 h to form one composite sample. All samples were then transported by car back to SLU and stored dark in a refrigerator (+4 °C), up until extraction day.

3.3.1.1 Water sampling

The water SPs corresponding to Hovgården's drainage system are presented in Figure 3. See Appendix, Figure A1, for a detailed schematic plan of the mixed stream's SPs.

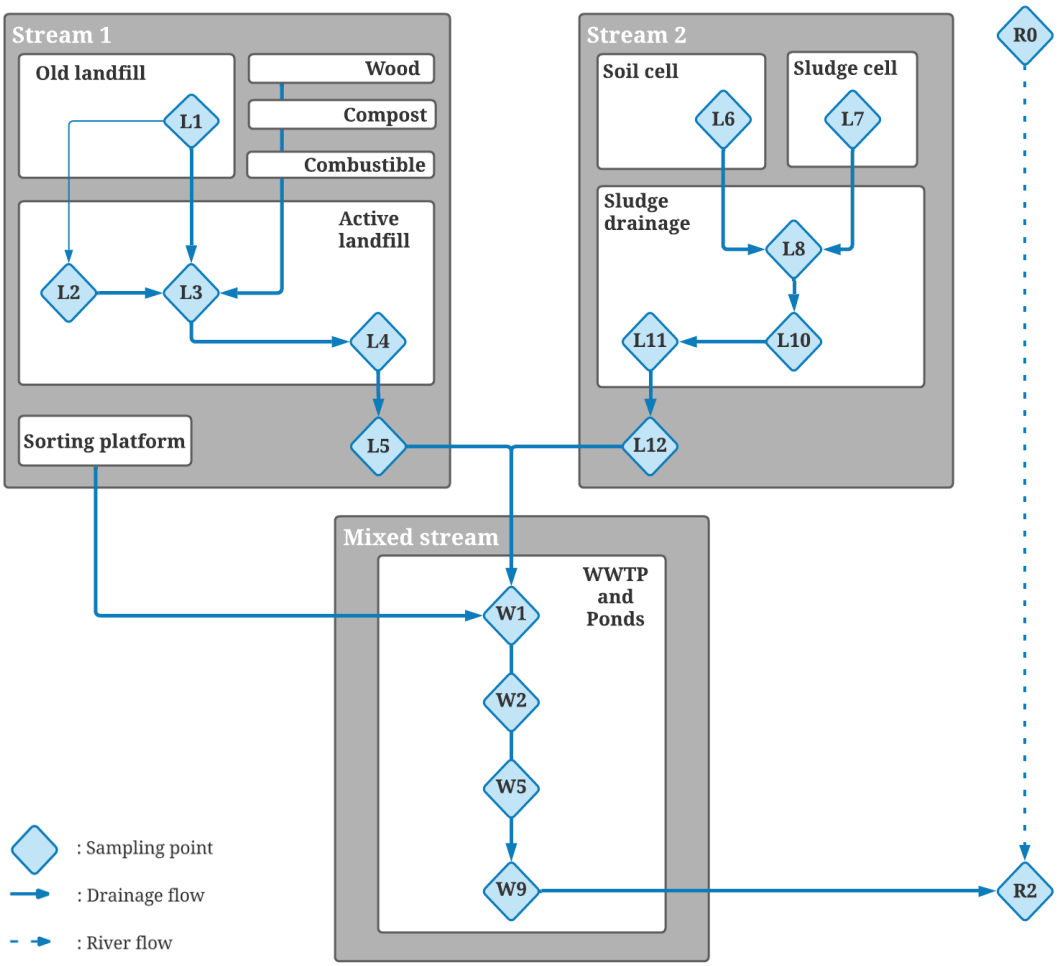

Figure 3: Schematic plan of the drainage system at Hovgården from each sampling point. Stream 1: leachate from the landfills (old and active) and the hardened surfaces (for wood, compost and combustible waste) as well as the sorting platform. Stream 2: leachate from the sludge cell and the soil cell. Mixed stream: water received from stream 1 and stream 2 that will be treated.

Stream 1 represents the drainage flow of leachate from the landfills (old and active) and the hardened surfaces (HSs) (where wood, compost, combustible waste is treated) and the sorting platform that eventually enters W1. It should be noted that formed leachate at the sorting platform is directly led to W1 (Figure 3). L1 represents LL from the old landfill. L2 represents LL that is drained from the surface (i.e., overground) of the old landfill's final cover (consisting of grass and top cover material), although it is located beneath the active landfill where plaster is deposited (Figure 2). L3 and L4 were accessed via tubes reaching down to the bottom of the drainage system of the active landfill. L3 receives leachate from asbestos, L1 and L2 (via a surface water ditch) and the HSs. Whereas L4 receives leachate from ash and L3. L5 represents the last SP where all the leachate from stream 1 (excluding the sorting platform) flows to.
No sample was collected from the sorting platform and the HSs. L1 was not sampled due to no water accessible. L2 was grab sampled using an extender connected to the bottle, whereas a weight was used for L3 – L5.

Stream 2 represents the drainage flow of leachate in the sludge cell (L7) (referred to as "sludge water") and from the soil cell (L6) with a belonging sludge drainage (L8 – L11) that eventually enters L12. L6 and L7 are separately led to L8 and were accessible via separate wells. L8 was sampled from a well inside a little house, close to L6 and L7. The combined water then flows onwards to L10 – L12. L10 was sampled after the first sedimentation pond. L11 was sampled after the second sedimentation pond. L12 represents the last SP where all the water from stream 2 flows to (Figure 3), which was collected from a pump well located in a pump house at Svartmuttern.
All SPs were grab sampled using an extender.

The mixed stream represents the drainage flow of leachate from stream 1 (L5), stream 2 (L12) and the sorting platform which represents the WWTP influent to W1 (Figure 3). W1 and W2 were sampled from separate basins in the WWTP. W1 was collected before the aeration/lamella sedimentation (the mechanical purification step). W2 was collected after. The water is then separated into the two basins where the bio-step is introduced. The water will then be led to the sedimentation pond and the polishing ponds. W5 was collected between the sedimentation pond and the polishing ponds. The water will then enter the oxidation pond (the last treatment step). W9 represents the WWTP effluent after the oxidation pond (Figure A1) and was collected in a measuring station house.
All SPs were grab sampled using an extender.

R0 and R2 represents **the river**. R0 is surface water located upstream close to Svartmuttern. It is treated as a reference point, before WWTP effluent is discharged from W9 to the recipient R2. R2 is a part of Hovgårdsbäcken located approximately 100 m downstream to the WWTP (close to W1) (Figure 3).
R0 and R2 were grab sampled without using an extender/weight.

SLU's TI autosamplers were used for the TIS of L5, L6, L8, L11 and L12, whereas Hovgården's were used for W1 and W2. Hovgården's FP autosampler was used for W9. The containers containing the composite samples were then shaken and mixed well, before the content was poured into separate 1 L PP bottles.

3.3.1.2 Sewage sludge sampling

On site in the sludge cell, there was a pile of sludge for each month from February 2021 to September 2021 and one from Storvreta's WWTP (S1 – S9 in Figure 2). 12 random points in a pile were grab sampled by using a spoon and put into one composite sample in a 250 mL plastic bottle. This was repeated for each pile.

3.4 SAMPLE PREPARATION

Procedures for preparation of water and sludge samples for instrument analysis were conducted as described in previous studies (Golovko et al. 2016; Rehrl et al. 2020).

Water samples included grab, TI and a FP sample(s) (previously described in section 3.1.1). The extraction was performed three weeks after the sampling. The samples were extracted by solid phase extraction (SPE) following the procedure described by Sörengård et al. (2019). Briefly, approximately 200 mL were filtered in an all-glass filter holder (Merck, 47 mm) using a glass microfiber filter (grade GF/F, Whatman, thickness 0.42 mm, pore size 0.7 µm). IS mixtures (20 ng per sample) were added to the aliquot (filtered sample). Oasis HLB-cartridges (6 cc, 200 mg) were conditioned with 6 mL methanol followed by 6 mL Milli-Q water with flow driven by gravity. The aliquot was subsequently loaded and the flow rate was restricted to one droplet per second under vacuum. The cartridge was then washed with 6 mL Milli-Q water, followed by 20 minutes of drying under vacuum. Compounds were thereafter eluted with 8 mL methanol by gravity into centrifuge tubes. The extracts were dried under a gentle nitrogen gas stream to 0.5 mL and then transferred to 1.5 mL glass vials (Eppendorf, Germany). The walls of the centrifuge tubes were rinsed three times with 200 µL methanol and transferred to a corresponding sample vial. The extracts in the vials were dried to exactly 0.5 mL under the same conditions as for the centrifuge tubes. The vials were vortexed and stored frozen (-16 ˚C) in darkness until instrumental analysis. The extracts were then spiked with 500 µL Milli-Q water right before injection.

Sludge samples comprised of grab samples from each sludge pile (previously described in section 3.1.1). The extraction was performed one week after the sampling. The sludge samples were prepared using an ultrasonic-based solvent approach, for which detailed information can be found elsewhere (Golovko et al. 2016). Briefly, the sludge samples were air-dried overnight in a clean fume hood. Before extraction, an IS mixture (c = 10 ng/g dw sludge) was added to 2 g dry sludge sample. 4 mL of acetonitrile and water (1/1, v/v, 0.1 % formic acid) were thereafter added to the air-dried sludge and then ultrasonicated for 15 min. The supernatant was filtered through a syringe filter (0.45 µm, regenerated cellulose, VWR, Sweden) into 10 mL glass vials. The step was repeated with a second extraction solvent mixture (acetonitrile, 2-propanol, and water (3/3/4 v/v/v with 0.1 % formic acid)). The two supernatants were combined, mixed well, and kept frozen at -20 °C until analysis, of which 1 mL of the extract was used for instrumental analysis.

3.5 INSTRUMENTAL ANALYSIS OF COMPOUNDS

The water and sludge samples were analysed by using a DIONEX UltiMate 3000 ultra-high pressure liquid chromatography (UPLC) system (Thermo Scientific, Waltham, MA, USA) coupled to a triple quadrupole mass spectrometer (MS/MS) (TSQ QUANTIVA, Thermo Scientific, Waltham, MA, USA).

An Acquity UPLC BEH-C18 column (2.1 x 50 mm, 1.7 µm particle size; Waters Corporation, Manchester, UK) was used as an analytical column for chromatographic

separation. The temperature of the column oven was set at 40 ± 2 °C. The system was equipped with a heated electrospray ion source with static spray voltage set at 3500 V positive mode and 2500 V negative mode. The temperatures of the ion transfer tube and the vaporizer were set at 325 °C and 400 °C, respectively.
The mobile phase consisted of Milli-Q water with the addition of 5 mM ammonium acetate (phase A) and acetonitrile (phase B). The same linear gradient was used in both ionization modes, with a flow rate of 0.5 mL/min. The gradient started at 2 % of phase B and increased to 99 % from 0.5 min to 10.0 min. This composition of the mobile phase was maintained for 3 minutes, until 13.0 min, after which it returned to initial conditions at 13.1 min. Such composition was maintained until the end of the analytical run, which took 15 minutes.

Xcalibur software (Thermo Fisher Scientific, San Jose, CA, USA) was thereafter used for the data acquisition and the data was evaluated using TraceFinderTM 4.1. software (Thermo Fisher).

3.6 QUALITY CONTROL

The performance of the method was assessed with regard to its linearity, blanks, limit of quantification (LOQ), relative recovery, precision, and matrix effect for CECs in the sludge- and water samples (Table A3 and A4 in Appendix).

The linearity of the calibration curve was tested in the range 0.01 ng/ml to 500 ng/ml for water samples and 0.1 ng/ml to 250 ng/ml for sludge samples. The calibration curve was measured twice, at the beginning and end of the sequence, to check instrument stability. The calibration was prepared in Milli-Q + methanol (1/1) for water samples and acetonitrile/water (1/1) for sludge samples.
LOQ was calculated as half the lowest calibration point in the calibration curve where the relative standard deviation of the average response factor was < 30% (one or two points at low concentration levels had to be removed in some cases). The peak area corresponding to this concentration was used to calculate LOQ for each individual compound in each sample.

The precision of the method was evaluated by the repeatability of the study.
Duplicate samples were prepared for the sludge- (n_{tot} = *18*) and water samples which included grab (n_{tot} = *64*), TI (n_{tot} =*16)* and FP (n_{tot} = *2)* samples.
Blanks were also included. They consisted of Milli-Q (n_{tot} = *6*) and tap water (n_{tot} = *6*) for the water samples and only Milli-Q water (n_{tot} = *1*) for the sludge samples. No target analytes were detected in the method blanks.
Fortified (FORT) samples were prepared for the water- (n_{tot} = *6*) and sludge samples (n_{tot} = *3*) to check the extraction efficiency of the method. This was performed by spiking a randomized field sample with internal and native standards before extraction.
Matrix-matched standards (MST) were used to assess the matrix effect and were prepared from a randomized field sample extract spiked with internal and native standards at concentration levels equivalent to 20 ng/L and 200 ng/L, respectively, after extraction. MST were prepared for the water- (n_{tot} = *6*) and sludge samples (n_{tot} = *3*).
The duplicates, blanks, FORT and MST samples were prepared, extracted and analysed in the same way as the water- and sludge samples. The only difference is that standards were added to MST samples after the extraction. Two blanks, one FORT and one MST sample were performed for every SPE-batch.

3.7 STATISTICAL ANALYSIS

A principal component analysis (PCA) was performed to depict (and visualize) the correlation between stream 1, stream 2, the mixed stream and the river to each other and to the 86 analysed CECs. PCA based biplots were introduced at a significance level of 5.0 %. The implemented data consisted of the ∑mean composition profile in each stream and river. Similar observations, clusters, outliers and patterns of all CECs between all streams and the river were thereafter obtained. This was also performed for all sewage sludge SPs from the sludge cell.

4 RESULTS AND DISCUSSION

The extracted water grab (n_{tot} = *61*), TI (n_{tot} = *16*) and FP (n_{tot} = *2*) as well as the sewage sludge grab (n_{tot} = *15*) samples were analysed for the 86 selected CECs (Table A1.1, A1.2 and A.2 in Appendix).

The ∑mean concentration, mean, median, detection frequency and standard deviation of all CECs, as well as the mean concentration of every CEC, in every water and sludge GS point are presented in Appendix, Table A5 – A8. The ∑mean concentration, mean, median and detection frequency of all CECs, as well as the mean concentration of every CEC, in every water TIS/FPS point is presented in Appendix, Table A9 and A10.

All water- and sludge results are described based on grab sampling approach.

4.1 PHARMACEUTICALS IN WATER AT HOVGÅRDEN

The ∑pharmaceuticals mean concentrations and ∑pharmaceuticals mean compositions in every water GS point within Hovgården's facility is presented in Appendix, Figure A2 and A3.

Six pharmaceuticals were detected in all SPs at Hovgården (carbamazepine, fexofenadine, lidocaine, losartan, metformin and metoprolol), ranging from 1.1 ng/L (fexofenadine) to 2700 ng/L (losartan). 42 pharmaceuticals were detected in at least one SP, ranging from 1.1 ng/L (tramadol) to 4900 ng/L (erythromycin). Chlorzoxazone was only detected in L11 (3.3 ng/L) (Table A5). More detailed information about the 42 pharmaceuticals that were detected at Hovgården can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.

The dominating contributors (≥ 5.0 % in mean composition) at Hovgården were fexofenadine, carbamazepine, metoprolol, erythromycin, losartan and desvenlafaxine (on average, 5.2, 6.4, 6.4, 10, 12 and 14 %, respectively) (Figure A3).

4.1.1 Pharmaceuticals in leachate from the landfill (and the HSs) in stream 1

L2 (∑pharmaceuticals mean concentration: 23 000 ng/L, mean: 420 ng/L, median: 39 ng/L) was the main contributor, covering ≈ 94 % of all ∑pharmaceuticals mean concentrations in stream 1. L4 (510 ng/L, 9.1 ng/L, 1.0 ng/L), L5 (500 ng/L, 8.9 ng/L, 0.68 ng/L) and L3 (470 ng/L, 8.4 ng/L, 1.4 ng/L) had the lowest contributions in stream 1. L3 up to L5 were ≈ 45 – 49 times lower than L2 (Table A5). The ∑pharmaceuticals mean concentrations in stream 1 is presented in Figure 4.

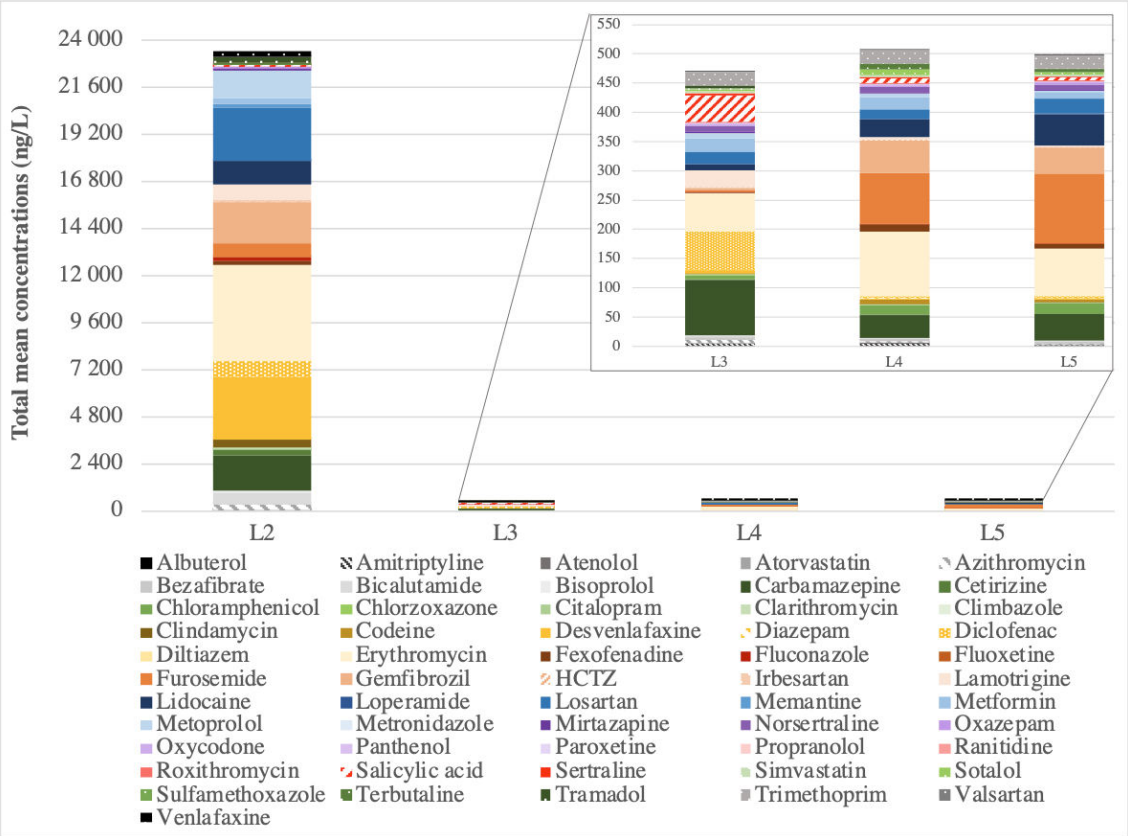

Figure 4: Total mean concentration (ng/L) of pharmaceuticals in every water grab sampling point in stream 1.

The detection frequency of pharmaceuticals was the highest in L2 (*n = 41*), followed by L3 (*n = 22*), L4 (*n = 20*) and L5 (*n = 15*). 11 pharmaceuticals were detected in all SPs, ranging from 1.1 ng/L (fexofenadine) to 4900 ng/L (erythromycin). 34 pharmaceuticals were detected in at least one SP, ranging from 1.1 ng/L (tramadol) to 3200 ng/L (desvenlafaxine). 17 of the 34 pharmaceuticals were only detected in L2, ranging from 2.9 ng/L (paroxetine) to 440 ng/L (clindamycin). Ranitidine was only detected in L3 (1.2 ng/L), as well as atenolol and sotalol in L4 (2.2 and 9.5 ng/L, respectively) (Table A5). More detailed information about which pharmaceuticals that were detected in specific SPs in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.1, and in Table A5.

The ∑pharmaceuticals mean compositions in stream 1 are presented in Figure 5. The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in all SPs included carbamazepine (7.7 – 20 %) and erythromycin (14 – 22 %) (Figure 5). Also, gemfibrozil (8.8 – 11 %) and lidocaine (5.1 – 11 %) in all SPs except in L3 and desvenlafaxine and metoprolol in L2 (14 and 6.0 %, respectively). As well as diclofenac, lamotrigine and salicylic acid in L3 (14, 5.7 and 9.6 %, respectively), furosemide

in L4 (17 %) and L5 (24 %) and losartan in L2 (12 %) and L5 (5.2 %). Erythromycin was the main contributor in L2 (21 %) and L4 (22 %), whereas it was carbamazepine in L3 (20 %) and furosemide in L5 (24 %) (ibid.). More detailed information about the mean composition profile of pharmaceuticals in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion"*, S4.1.1.

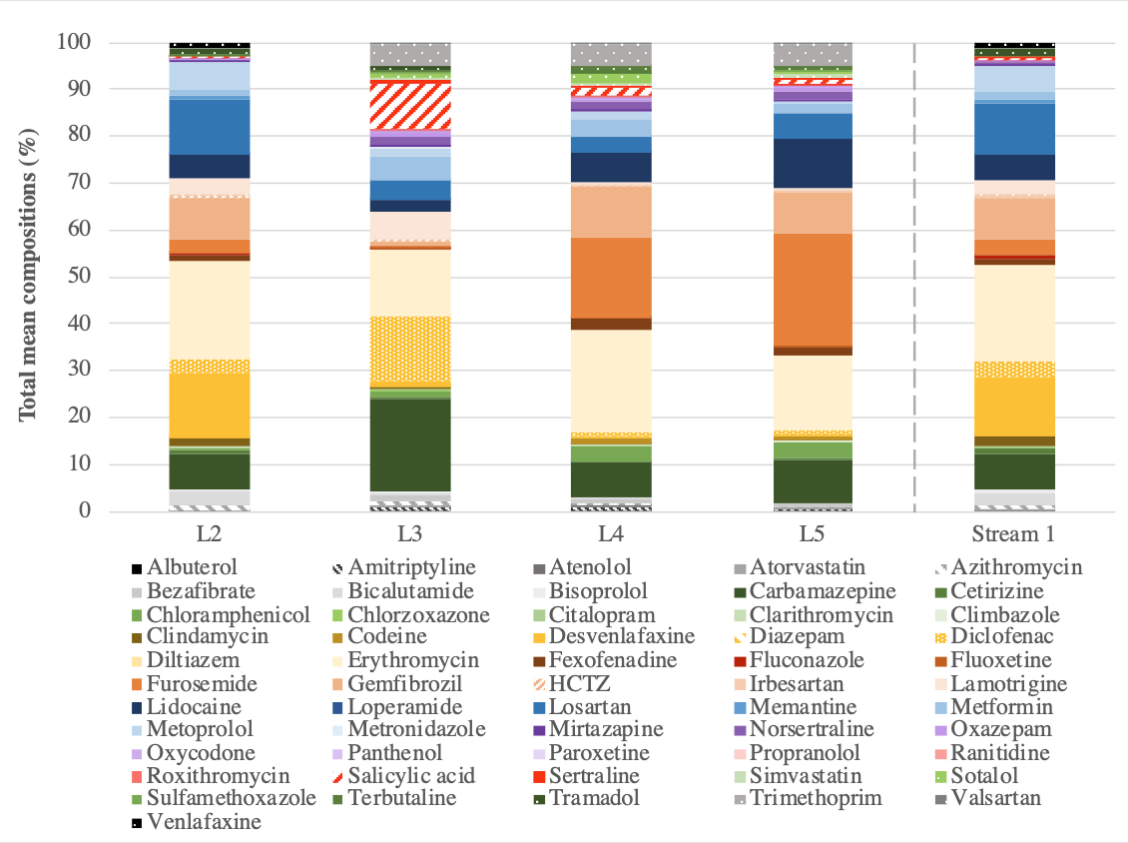

Figure 5: Total mean composition profile (%) in every water grab sampling point and in stream 1, based on normalized mean concentrations of the pharmaceuticals.

The decrease in the ∑pharmaceuticals mean concentration from L2 to L5 (Figure 4) and detection frequency (by 2.7 times lower from L2 to L5) as well as discrepancies in concentrations and causes for the occurrence are most likely a combination of several factors. This may include **i)** exposure history with pharmaceuticals that are persistent and resistant to degradation, **ii)** the CECs physico-chemical properties, **iii)** natural attenuation, **iv)** poor representation, **v)** uncertainties (e.g., concentrations close to the detection limit) and **vi)** analytical errors.

Firstly, no previous research was found about pharmaceuticals being commonly present in ash and asbestos. This could explain why the ∑pharmaceuticals mean concentration in L3 – L5 were lower than L2 (Figure 4). It also indicates that the occurrence of pharmaceuticals in L3 – L5 is influenced by leachate that is transported from L2. However, no sample was collected from L1 or the hardened surfaces (where wood, compost and combustible waste are treated). Which means that iv) and v) are contributing factors as well. It can therefore not be ruled out if the pharmaceutical occurrence in L3 – L5 is also influenced by leachate from other surfaces. Which could for example explain why codeine and sotalol were detected in L4 but not in L2 nor L3 (Table A5). On the other hand, this could also be due to i) and/or vi). In addition, i) could for example explain L2's high mean concentrations of the 10 pharmaceuticals that deviated the most in stream 1 (bicalutamide, carbamazepine, desvenlafaxine, diclo-

fenac, erythromycin, gemfibrozil, lamotrigine, lidocaine, losartan and metoprolol) (Table A5). Carbamazepine has for instance been recognized to have these properties (Li 2014; Golovko et al. 2020; Golovko et al. 2021; Chavoshani et al. 2020) as well as erythromycin (Minski et al. 2021), metoprolol and lamotrigine (Golovko et al. 2021). Also, i) could explain why erythromycin was detected in a higher mean concentration in L4 compared to L3 (1.7 times higher) (Table A5). As well as why furosemide and gemfibrozil were detected in all SPs, except in L3 (ibid.). Also, why some pharmaceuticals were only detected in one SP, such as the 17 pharmaceuticals that were only detected in L2 (S4.1.1 in Appendix). On the other hand, this could also be due to ii) and iii) since the fate and transport of pharmaceuticals are generally governed by natural attenuation (such as sorption, biodegradation and abiotic degradation and dispersion/ dilution, depending on the compound's hydrophobicity and biodegradability) and by their physico-chemical properties (such as water solubility) in the environment (WHO 2011; Bjerg et al. 2014; Li 2014; Fernandes et al. 2021). This may be the main cause for the decrease in the ∑pharmaceuticals mean concentration and detection frequency from L2 to L5. Furthermore, fexofenadine had a mean concentration (1.1 ng/L) close to its LOQ-value (1.0 ng/L) in L3 (Table A3 and A5). Tramadol also had a mean concentration (1.1 ng/L) close to its LOQ-value (1.0 ng/L) in L4 (ibid.). Which means that there are uncertainties (i.e., v)) regarding pharmaceuticals that were detected in a concentration close to their LOQ-value. Moreover, vi) may explain pharmaceuticals that were only detected in one SP in low mean concentrations, such as paroxetine in L2, ranitidine in L3 and atenolol in L4 (2.9, 1.2 and 2.2 ng/L, respectively) (Table A5).
Anyhow, the ∑pharmaceuticals mean concentration in L2 is most likely caused by precipitation infiltrating into the old landfill's final cover, where leachate is transported from a drainage pipe above the sealing layer to the surface water ditch where L2 was sampled. L2's high detection frequency is most likely due to the sludge in the layer, since pharmaceuticals have frequently been mentioned to be present in sludge (Li 2014; Klatte et al. 2017; Patel et al. 2019; Ekane et al. 2021; Golovko et al. 2021). This is also the more likely cause for the pharmaceutical occurrence in L3 – L5 as well, since no previous research was found about pharmaceuticals being commonly present in ash and asbestos. Which is also due to observing much lower ∑pharmaceuticals mean concentrations and detection frequencies in L3 – L5 compared to L2 (Table A5). It should however be noted that it cannot be ruled out if L1 and/or other surfaces may contribute as previously mentioned (p. 24).

Moreover, a study conducted by Yu et al. (2020) compared 50 research articles/reports from 1993 – 2018 about pharmaceuticals that were analysed in LL. There it was concluded that compounds such as carbamazepine (antiepileptic), diclofenac (analgesic), gemfibrozil (antilipemic), metoprolol (antihypertensive) and erythromycin (antibiotic) were frequently detected (Yu et al. 2020). Which is of interest since these were dominating/main contributors in stream 1 (pp. 23, 24). Which could indicate that these are common pharmaceuticals in LL. Moreover, some of these pharmaceutical groups have often been detected in the surface water, groundwater and the soil zone and have been associated with causing harmful effects on the (aquatic) environment (pp. 3, 4). This therefore shows that it is important to have further monitoring and evaluations of the occurrence and dissemination of pharmaceuticals in LL.

Furthermore, the ∑pharmaceuticals mean concentrations were 45 – 49 times lower in L3 – L5 compared to L2 (p. 23). This could indicate that a high fraction of pharmaceuticals is removed via natural attenuation from and/or that they are persistent in L2.

Which may indicate that no measures are required to improve the treatment of generated LL. There is however a possibility that leachate from L2 may be transported to L3 – L5 in even higher concentrations in the future, which eventually could reach the WWTP. This means that measures may be required, depending on how efficient the WWTP is in removing/degrading pharmaceuticals before the treated wastewater is discharged into the aquatic environment. However, the reliability of the obtained measurements from this study can be questioned since this was the first time an evaluation of the pharmaceutical occurrence at Hovgården was performed. The results from this study would therefore be more reliable if further screenings are performed. For example by repeating this study to compare the results. Which would be beneficial before considering measures such as implementing additional treatment methods. In addition, a risk characterization was not included in this study. This means that it could not be determined if the concentrations of the analysed pharmaceuticals may be harmful for the receiving environment. If a risk characterization would've been included, this could've decreased the uncertainty concerning if measures are necessary.

4.1.2 Pharmaceuticals in leachate from the sludge (and the soil) cell in stream 2

L8 (∑pharmaceuticals mean concentration: 16 000 ng/L, mean: 290 ng/L, median: 54 ng/L) was the main contributor in stream 2, followed by L7 (13 000 ng/L, 230 ng/L, 34 ng/L). The contribution was lower in L10 (5900 ng/L, 110 ng/L, 14 ng/L), L11 (5900 ng/L, 100 ng/L, 16 ng/L) and L12 (4200 ng/L, 74 ng/L, 16 ng/L), which were 2.7 – 3.8 times lower than L8. L6 (1500 ng/L, 28 ng/L, 6.6 ng/L) had the lowest contribution in stream 2, which was 11 times lower than L8 (Table A5). The ∑pharmaceuticals mean concentrations in stream 2 are presented in Figure 6.

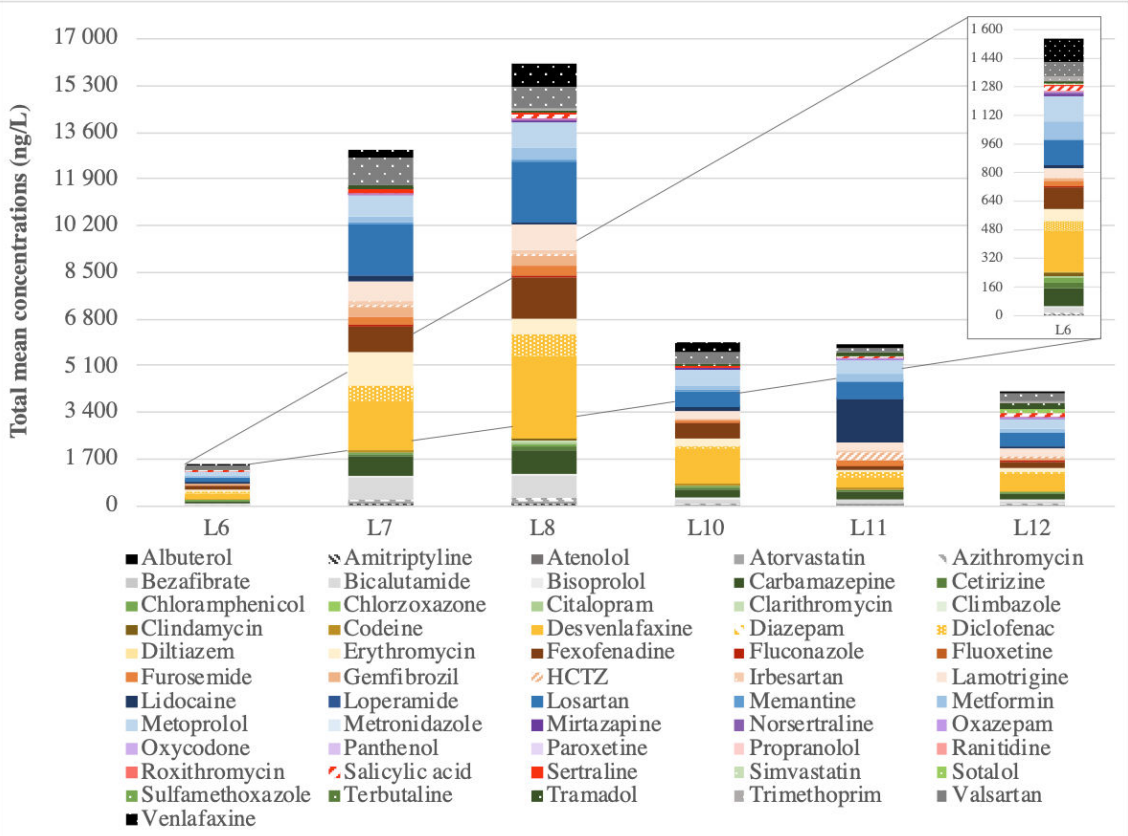

Figure 6: Total mean concentration (ng/L) of pharmaceuticals in every water grab sampling point in stream 2.

The detection frequency of pharmaceuticals was the highest in L11 (*n = 43*), followed by L8 (*n = 40*), L7 (*n = 39*), L10 and L12 (*n = 38*, respectively) and L6 (*n = 30*). 29 pharmaceuticals were detected in all SPs, ranging from 1.6 ng/L (azithromycin) to 3000 ng/L (desvenlafaxine). 19 pharmaceuticals were detected in at least one SP, ranging from 1.3 ng/L (climbazole) to 320 ng/L (HCTZ). Loperamide was only detected in L7 (3.2 ng/L), as well as paroxetine in L10 (1.5 ng/L), chlorzoxazone and ranitidine in L11 (3.3 and 1.7 ng/L, respectively) (Table A5). More detailed information about which pharmaceuticals that were detected in specific SPs in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.2, and in Table A5.

The ∑pharmaceuticals mean compositions in stream 2 are presented in Figure 7. The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in all SPs included carbamazepine (5.0 – 6.5 %), desvenlafaxine (7.0 – 22 %), losartan (9.0 – 15 %) and metoprolol (5.7 – 11 %) (Figure 7). Fexofenadine (5.0 – 9.3 %) and valsartan (5.1 – 7.7 %) were dominating contributors in all SPs except in L11. Also, metformin in L6 (6.5 %), erythromycin in L7 (9.3 %), diclofenac in L8 (5.1 %), tramadol in L12 (6.5 %) and HCTZ and lidocaine in L11 (5.5 and 27 %, respectively). As well as bicalutamide in L7 (5.8 %) and L8 (5.2 %), lamotrigine in L7, L8, L11 and L12 (5.7, 6.0, 5.0 and 6.7 %, respectively) and venlafaxine in L6, L8 and L10 (8.4, 5.2 and 5.4 %, respectively) (ibid.). Losartan (15 %) was the main contributor in L7, whereas it was lidocaine (27 %) in L11 and desvenlafaxine in the remaining SPs (15 – 22 %) (ibid.). More detailed information about the mean composition profile of pharmaceuticals in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.2.

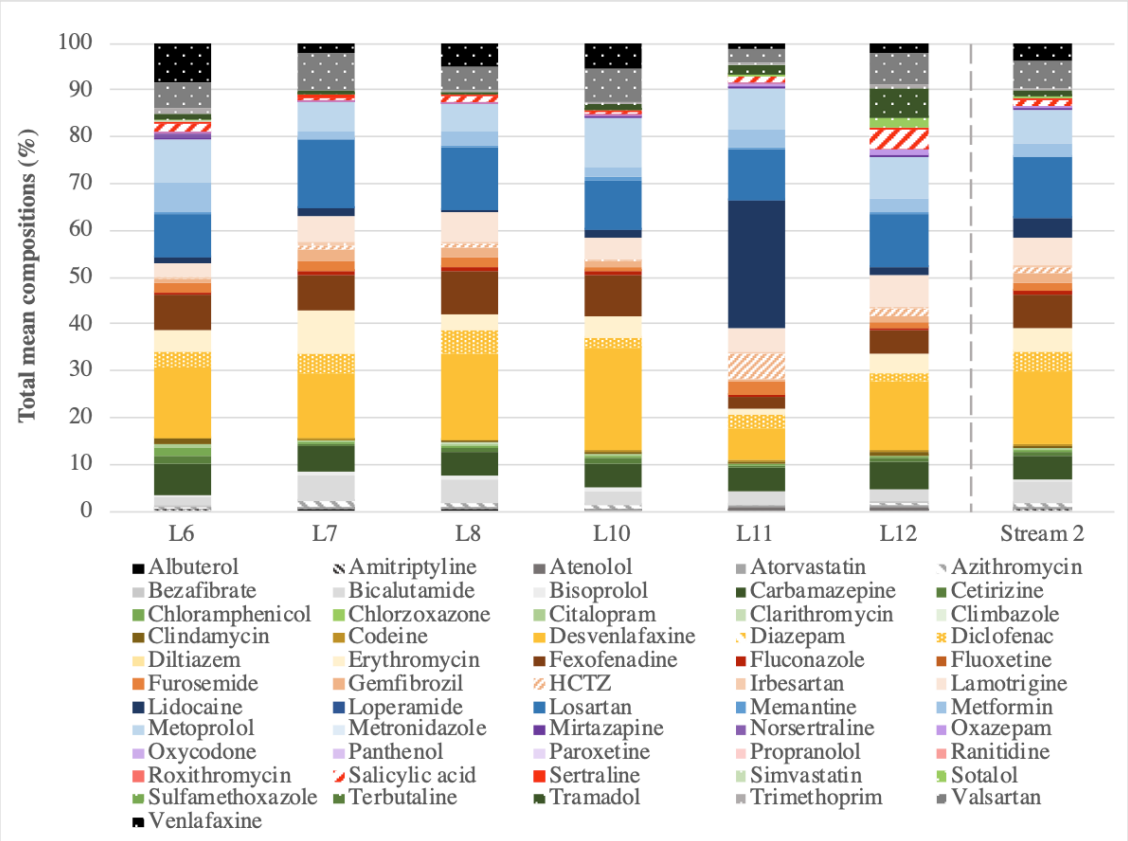

Figure 7: Total mean composition profile (%) in every water grab sampling point and in stream 2, based on normalized mean concentrations of the pharmaceuticals.

The occurrence of pharmaceuticals in L6 is most likely caused by sludge water, since sludge was reported to be stored at L6 in the middle of 2021 (S. Bjälkefur, personal communication, March 6, 2022). There is a possibility that the excavated masses, which is treated in the soil cell at L6, may have contained pharmaceuticals. For example, if sludge has been spread on agricultural land with possibly containing pharmaceuticals that may have dispersed to areas where excavation has been performed. This possibility was however not found in previous research. It is therefore more reasonable that the pharmaceutical occurrence at L6 is caused by sludge that has previously been stored in the soil cell. It is also reasonable that L7 had a higher ∑pharmaceuticals mean concentration (Figure 6) and detection frequency (1.3 times higher) (Table A5) than L6, since the sludge is mainly stored in the sludge cell (where L7 is the first receiving SP of sludge water). However, only one of four samples were extracted from L7. This could question the reliability of L7's ∑pharmaceuticals mean concentration and its detection frequency compared to L6 (and the other SPs). But it is still more reasonable that L7 had a higher ∑pharmaceuticals mean concentration and detection frequency than L6, since pharmaceuticals have frequently been mentioned to be present in sludge (Li 2014; Klatte et al. 2017; Patel et al. 2019; Ekane et al. 2021; Golovko et al. 2021). Which was also confirmed in the sludge samples (Table A6). This could also explain why the ∑pharmaceuticals mean concentrations in L8 – L12 were higher than L6 (Figure 6), as well as the detection frequencies (on average, 1.3 times higher) (Table A5).

Discrepancies in detection frequencies, ∑mean concentration and mean concentrations of pharmaceuticals in stream 2 could be a cause of: **i)** exposure history with pharmaceuticals that are persistent and resistant to degradation, **ii)** the CECs physico-chemical properties (such as water solubility), **iii)** natural attenuation, **iv)** poor representation, **v)** uncertainties (e.g., concentrations close to the detection limit) and **vi)** analytical errors.
Firstly, the ∑pharmaceuticals mean concentration increased from L6 to L8 in the GS and TIS points (Figure A22). Which indicates that it is of regular pattern (that it increases from L6 to L8) and hence that iv) and vi) are minor factors for this observation. Furthermore, ii) may have contributed to L8 having a higher ∑pharmaceuticals mean concentration than both L6 and L7 (Figure 6). For example, if pharmaceuticals have high mobility from these SPs. This may also be a result of i). For instance, carbamazepine's, lamotrigine's and metoprolol's mean concentrations were higher in L8 (on average, 910 ng/L) compared to L6 and L7 (on average, 96 and 740 ng/L, respectively) (Table A5) and have been recognized to have these characteristics (Golovko et al. 2021) and indicates that i) may have contributed to L8 having a higher ∑pharmaceuticals mean concentration than L6 and L7. Desvenlafaxine and fexofenadine were also detected in a higher mean concentration in L8 (3000 and 1500 ng/L, respectively) compared to all other GS points in stream 2 (on average, 870 and 390 ng/L, respectively) (Table A5). This was also confirmed in the TIS point of L8 (i.e., L8TI) (1900 and 1100 ng/L, respectively) compared to all other TIS points in stream 2 (on average, 370 and 120 ng/L, respectively) (Table A9). Which indicates that desvenlafaxine and fexofenadine may also be prone to have these characteristics. Moreover, it was noted during the SPE-filtration that the grab sample of L8 had a high fraction of dissolved/suspended solids compared to L6 and L7. This may indicate that iv) is a contributing factor. Which could for example explain why citalopram, lamotrigine, losartan, salicylic acid and venlafaxine were detected in higher mean concentrations in L8 compared to L6 and L7 (on average, 850, 71 and 600 ng/L, respectively) (Table A5), although they are more expected to adsorb to suspended solids and sediments in water

(PubChem n.d.a, n.d.d, n.d.b, n.d.e, n.d.f). Which questions the reliability of the measurements of L8 and may also contradict L8 as the main contributor in stream 2.
The ∑pharmaceuticals mean concentrations then decreased from L8 to L12 (Figure 6). This is most likely due to ii) with hydrophobic pharmaceuticals that have adsorbed to suspended solid surfaces and settled in the sedimentation ponds (before and after L10). Whereas pharmaceuticals that were detected in L10 – L12 are most likely moderately or strongly hydrophilic and have therefore remained in the dissolved phase, with bio-degradation and/or photodegradation as main mechanisms (Gruchlik et al. 2018). Hence, iii) is also a main factor here. This could for instance explain why the mean concentration of desvenlafaxine, diclofenac, fexofenadine, losartan and valsartan decreased from L8 to L10 (on average, 1700 and 600 ng/L, respectively) (Table A5). However, the detection frequency in L10 was 1.1 times lower than L11 (ibid.). This indicates that i) may have contributed and/or that the grab sample of L11 had iv). It was for instance noted during the SPE-filtration that L11's grab sample had a high fraction of dissolved/suspended solids. Which could explain why lidocaine, which is more expected to adsorb to solids when released into water (PubChem n.d.c), had a higher mean concentration in L11 compared to L10 (1600 and 90 ng/L, respectively) (Table A5). In addition, i) and/ or iv) of L11 could also explain why HCTZ was detected in a higher mean concentration in L11 compared to L10 (320 and 10 ng/L, respectively) (Table A5). Moreover, desvenlafaxine's and fexofenadine's mean concentration was higher in L12 (620 and 210 ng/L, respectively) compared to L11 (410 and 150 ng/L, respectively) (Table A5). Which was also confirmed in the TIS point of L12 (i.e., L12TI) (630 and 200 ng/L, respectively) and L11 (i.e., L11TI) (410 and 150 ng/L, respectively) (Table A9). This means that the mean concentrations of these compounds did not change when comparing the GS to its corresponding TIS point of L11 and L12. Which could indicate that desvenlafaxine and fexofenadine are persistent and hence that i) have contributed to this difference. This could also explain if compounds were detected in higher mean concentrations in, for example, L12 compared to L11.

It cannot be concluded which SP that is the main contributor to L8 since the leachate from L6 and L7 are mixed at L8. However, the pharmaceutical occurrence in stream 2 is more likely to originate from pharmaceuticals leaching out from the sludge than from excavated masses (treated in the soil cell), due to sludge frequently being mentioned as a source of pharmaceuticals. It is also based on not finding previous research about excavated masses containing pharmaceuticals and with sludge previously being stored in the soil cell. Also, L7 is the originating source for the pharmaceutical occurrence in stream 2, if sludge is the primary source, since L7 is the first SP that receives sludge water from the cell where sludge is mainly stored. This could however suggest that the ∑pharmaceuticals mean concentration should be higher in L7 than L8, although it was the opposite in this study. But this also depends on, for example, if pharmaceuticals are persistent and resistant to degradation in L8 and/or if there is a high mobility from L7 to L8. It may also be a result of poor representation in L8's grab samples since a higher fraction of dissolved/suspended solids in a water sample could lead to pharmaceuticals being detected in higher concentrations, as they tend to also sorb onto solids (pp. 6, 7).
Since multiple factors could have affected the results and that a TI sample of L7 was not conducted, it cannot be confirmed with high certainty if it is of regular pattern that L8's ∑pharmaceuticals mean concentration is higher than L7. Which means that there is an uncertainty concerning which SP that is the main contributor in stream 2. The reliability of the obtained measurements from this study could be strengthened if further screenings are performed. For example, by repeating this study to compare the results.

Also, the ∑pharmaceuticals mean concentrations and detection frequencies were high in stream 2. It is therefore important to have further monitoring and evaluations since the detected pharmaceuticals may reach the WWTP in even higher concentrations in the future. Which, consequently, could be discharged into the aquatic environment in higher concentrations, depending on how efficient the WWTP is in removing/degrading pharmaceuticals. Further monitoring and evaluations would improve the knowledge about the pharmaceutical occurrence in the sludge water and if additional treatment methods are necessary. Furthermore, it could not be determined if the concentrations of the analysed pharmaceuticals may be harmful for the receiving environment since a risk characterization was not included in this study. The uncertainty concerning if measures are necessary could've also decreased if a risk characterization had been included.

4.1.3 Pharmaceuticals in combined water in the mixed stream and in the river

W1 (∑pharmaceuticals mean concentration: 740 ng/L, mean: 13 ng/L, median: 1.1 ng/L) was the main contributor in the mixed stream, followed by W9 (680 ng/L, 12 ng/L, 1.4 ng/L) and W2 (590 ng/L, 11 ng/L, 2.4 ng/L). W5 (490 ng/L, 8.7 ng/L, 1.3 ng/L) had the lowest contribution in the mixed stream, which was 1.5 times lower than W1. R0 (470 ng/L, 8.4 ng/L, 1.3 ng/L) was the main contributor in the river, followed by R2 (240 ng/L, 4.2 ng/L, 0.94 ng/L), which was 2.0 times lower than R0. R2 also had the lowest contribution compared to all the SPs from the mixed stream, where it was 3.1 times lower than W1 (Table A5). The ∑pharmaceuticals mean concentrations in the mixed stream and in the river are presented in Figure 8.

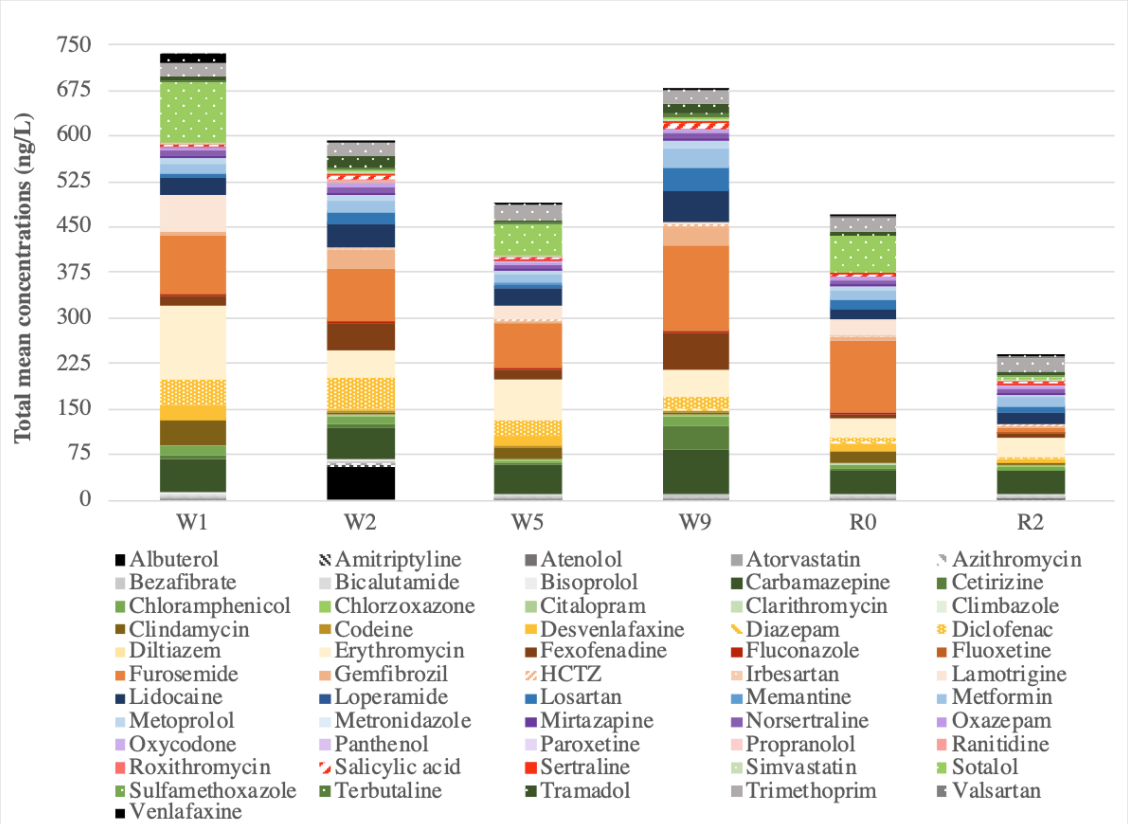

Figure 8: Total mean concentration (ng/L) of pharmaceuticals in every water grab sampling point in the mixed stream and in the river.

The detection frequency of pharmaceuticals was the highest in W2 (*n = 31*), followed by W5 (*n = 20*), W1 and R0 (*n = 19*), W9 (*n = 18*) and R2 (*n = 12*) (Table A5). Nine pharmaceuticals were detected in all SPs in both the mixed stream and in the river,

ranging from 1.3 ng/L (clindamycin) and 140 ng/L (furosemide). 29 pharmaceuticals were detected in at least one SP, ranging from 1.2 ng/L (azithromycin) to 120 ng/L (erythromycin). 11 of the 29 pharmaceuticals were only detected in W2, ranging from 1.3 ng/L (bisoprolol and citalopram) to 55 ng/L (albuterol). HCTZ and memantine were only detected in W5 (2.7 and 1.6 ng/L, respectively) (Table A5). More detailed information about which pharmaceuticals that were detected in specific SPs in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.3, and in Table A5.

The ∑pharmaceuticals mean compositions in the mixed stream and in the river are presented in Figure 9. The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in all SPs in the mixed stream and in the river included carbamazepine (8.3 – 16 %) and erythromycin (6.4 – 16 %) (Figure 9). Furosemide was a dominating contributor in all SPs except in R2 (13 – 26 %) as well as lidocaine except in W1 and R0 (5.3 – 8.0 %). Also, clindamycin in W1 (5.6 %), albuterol in W2 (9.3 %), cetirizine and losartan in W9 (5.9 and 5.5 %, respectively) and metformin and trimethoprim in R2 (6.8 and 9.7 %, respectively). As well as fexofenadine and gemfibrozil in W2 (7.8 and 5.6 %, respectively) and W9 (9.3 and 5.0 %, respectively) and lamotrigine in W1 and R0 (8.1 and 6.0 %, respectively). Also, diclofenac in W1, W2 and W5 (5.7, 8.5 and 5.3 %, respectively) and sotalol in W1, W5 and R0 (14, 11 and 13 %, respectively) (ibid.). Furosemide was the main contributor in all SPs (15 – 26 %) except in W1 and R2 where it was erythromycin (16 %) and carbamazepine (16 %), respectively (ibid.). More detailed information about the mean composition profile of pharmaceuticals in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.1.2.

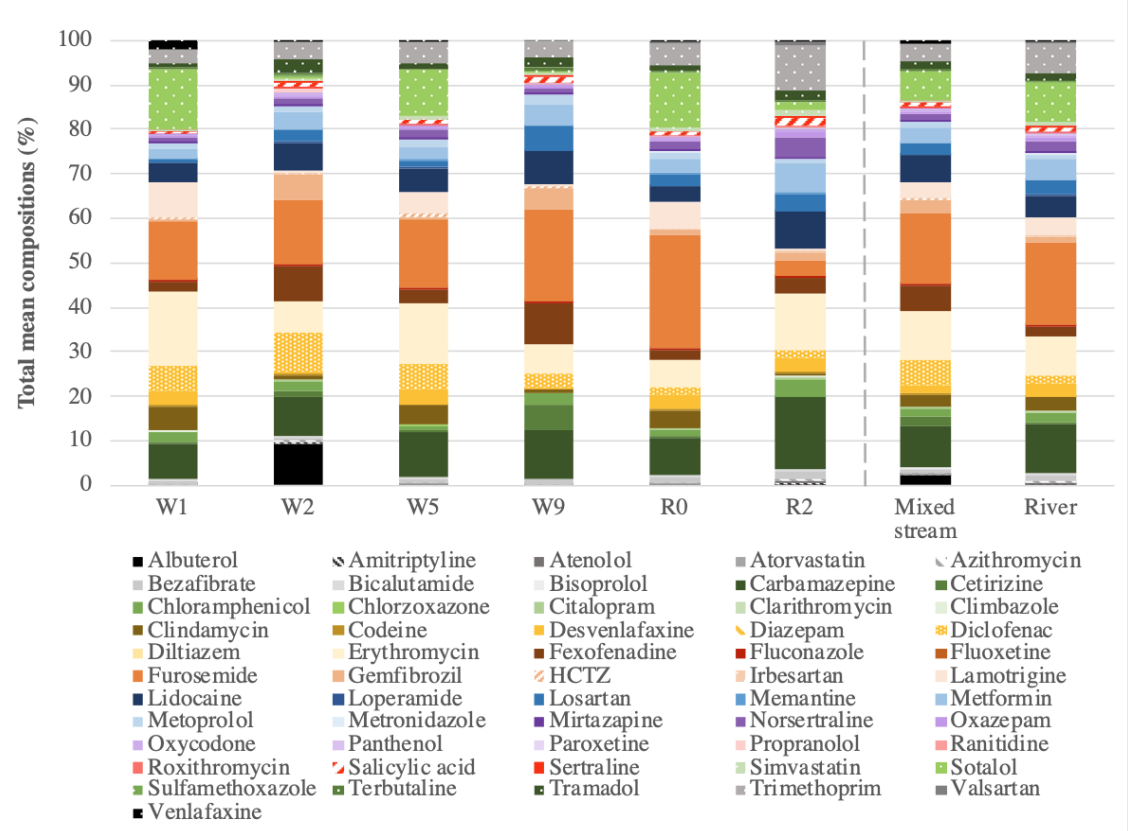

Figure 9: Total mean composition profile (%) in every water grab sampling point, in the mixed stream and in the river, based on normalized mean concentrations of the pharmaceuticals.

The pharmaceutical occurrence in the mixed stream is more likely influenced by stream 1 (L5) and stream 2 (L12) than by the sorting platform which has a direct drainage to W1 (Figure 3). This is heavily based on the discoveries in this study about pharmaceuticals being present in those streams (Figure 4 and 6). However, it cannot be ruled out whether the sorting platform contributes to the pharmaceutical occurrence in the incoming water at W1, since no sample was collected from this site. Furthermore, it cannot be concluded if L5 or L12 is the main contributor to W1 since the water from L5 and L12 are mixed before entering W1.

The ∑pharmaceuticals mean concentration decreased from W1 to W2 and from W2 to W5 (Figure 8), showing a positive removal efficiency of 20 % and 17 %, respectively (Table A5). Which means that there was a lower ∑pharmaceuticals mean concentrations in the WWTP effluent compared to its influent. This was also confirmed in the TIS points of W1 up to W5 (Figure A22), where it showed a positive removal efficiency of 20 % and 11 %, respectively (Table A9). This could be due to microbial degradation or sorption to solids, which have been mentioned to cause decrease of CEC concentrations in WWTP influents (Golovko et al. 2021). In other words, the aeration and lamella sedimentation (after W1), the MBBR-process and the sedimentation pond (where CECs have time to settle down and to be accumulated by plants) (after W2 but before passing through W5) could have influenced the degradation and sorption of pharmaceuticals.
The ∑pharmaceuticals mean concentration then increased from W5 to W9 (Figure 8), showing a negative removal efficiency of 39 % (Table A5). Which means that there was a higher ∑pharmaceuticals mean concentration in the WWTP effluent compared to its influent. This was confirmed in the TIS/FPS points of W5 and W9 as well (Figure A22), where it showed a negative removal efficiency of 125 % (Table A9). The cause for a higher concentration of CECs in the WWTP effluent compared to its influent could be a cause of: **i)** precursors degrading to target CECs, **ii)** compounds sorbed to the solid phase migrate to the aqueous phase, **iii)** influent and effluent samples being collected at the same time but with different portions of the wastewater due to different hydraulic retention times, **iv)** uncertainties (e.g., concentrations close to the detection limit) and **v)** analytical errors (Golovko et al. 2021). For instance, more water-soluble CECs may migrate over time to the aqueous phase, under well-mixed conditions (Golovko et al. 2021). In which the water from W5 is led to an oxidation pond before entering W9 to increase oxygen content. This may have stimulated microbial degradation and improved mixing conditions and hence may have caused i) and ii). The increase in detection frequency and/or if different compounds were detected between the GS points could also be a result of i) and ii). This could explain why W2 had the highest detection frequency and why it increased (by 39 %) from W1 to W2 (Table A5). Azithromycin, bisoprolol, citalopram and irbesartan were for example detected in W2, whereas they were not detected (n.d.) in W1. On the other hand, this may be a result of iv) and v) since these compounds mean concentration (1.2 – 1.4 ng/L) were close to their corresponding LOQ-value (0.50 ng/L – 0.55 ng/L) in W2 (Table A3 and A5). Another example is clindamycin, which was detected in R2 in a mean concentration (1.3 ng/L) close to its LOQ-value (1.0 ng/L) (ibid.).
The same pattern was obtained with the ∑pharmaceuticals mean concentration decreasing from W1 to W5 and with then increasing from W5 to W9, independently of which sampling method that was used (Figure A22). This may indicate that iii), v) and iv) are minor factors. It should however be noted that the ∑pharmaceuticals mean concentration in the GS point of W1 to W9 showed a positive removal efficiency of 8.1 %, whereas the TIS/FPS point of W1 to W9 (i.e., W1TI to W9FP) showed a negative

removal efficiency of 60 % (Table A5 and A9). This questions the performance of the WWTP's general removal efficiency and the reliability of the obtained measurements.

The ∑pharmaceuticals mean concentration decreased from W9 to the WWTP recipient i.e., R2 (Figure 8), showing a positive removal efficiency of 65 %. The detection frequency also decreased from W9 to R2, by 33 % (Table A5). CECs concentrations are typically lower in surface water than in the WWTP effluent due to natural attenuation (such as dilution, sorption, biodegradation and photodegradation) (Li 2014; Golovko et al. 2021). Similar findings have been discovered in previous research as well (Sörengård et al. 2019; Golovko et al. 2021). In addition, R0 may be a contributor to the pharmaceutical occurrence in R2 although WWTP effluents has been mentioned to be point/main sources to CECs occurrence in aquatic environments (Lapworth et al. 2012; Li 2014; Sörengård et al., 2019; Golovko et al. 2021). For instance, a higher ∑pharmaceuticals mean concentration and detection frequency was observed in R0 compared to R2 (2.0 and 1.6 times higher) (Table A5). However, the surface water in R0 may be more susceptible for natural attenuation, since R0 is located further away from R2 than W9. Which means that R0 may have a lower environmental load on R2 compared to W9. This could for example explain why the mean concentration of lidocaine and salicylic acid were higher in R2 (by 1.2 times, respectively), compared to R0 (Table A5). However, it cannot be concluded whether W9 is the main contributor to R2. Which is based on not being able to compare with previous results regarding the occurrence of pharmaceuticals at Hovgården, since this was the first time that this was evaluated. For example, R2's pharmaceutical occurrence may be a result of other factors such as exposure history with persistent pharmaceuticals that are resistant to degradation.

Since it was of interest to evaluate the pharmaceutical occurrence in the LL and in the leachate from the sludge storage facility, it would therefore be of interest to evaluate their declination to the last receiving SP, i.e., R2. However, it would give a wrong impression to present how much the ∑pharmaceuticals mean concentration decreased from the final SPs in stream 1 (L5) and stream 2 (L12) to R2, since the water from these SPs are mixed at W1. It is however apparent that it decreased from the streams to R2, since the average sum of all ∑pharmaceuticals mean concentration in the LL (6100 ng/L) and sludge water (7800 ng/L, including L6) were higher than R2 (240 ng/L). It is also apparent that the number of detected pharmaceuticals decreased. The detection frequency in the LL and sludge water (including L6) was on average at 25 and 38, respectively, whereas it was at 12 in R2 (21 % of all analysed pharmaceuticals) (Table A5).

Although conventional WWTPs are not designed to have a high removal efficiency of CECs, it was observed that this WWTP may have a positive removal efficiency to some extent. The declination of ∑pharmaceuticals mean concentrations from W1 to W5, with then increasing from W5 to W9, showed the same pattern independently of which sampling method that was used (Figure A22). This indicates that the WWTP's treatment steps has both positive (W1 to W5) and negative removal efficiencies (from W5 to W9). However, the general removal efficiency was positive in the GS point of W1 to W9, but negative in the TIS/FPS point of W1 to W9 (pp. 32, 33). Which is of importance to evaluate since this affects the reliability in the results from the GS method and consequently whether measures regarding additional treatment methods are necessary.
Furthermore, R2 had an occurrence of pharmaceuticals such as carbamazepine (antiepileptic), metformin (antidiabetic), tramadol (analgesic), furosemide and metoprolol (antihypertensives) (on average, 14 ng/L) (Table A5). These substances have previously been detected in surface water and have been mentioned as commonly found

pharmaceutical groups in this matrix (Li 2014; Golovko et al. 2021). Metformin has also been mentioned to be one of the most frequently detected pharmaceuticals in surface water worldwide. Also for being persistent and resistant to degradation in the environment, as well as carbamazepine (ibid.). This may be of concern, since some of these groups (analgesics, antiepileptics and antihypertensives) have been proven to cause adverse effects on the (aquatic) environment (pp. 3, 4). Moreover, desvenlafaxine (metabolite of antidepressant) was detected in R2, which is included in the WFD and is thus of environmental concern (European Commission 2020a). It is therefore important to conduct further screenings, to evaluate if the obtained measurements from this study are reliable. Which in this case mainly concerns the occurrence of pharmaceuticals in R2 and if the obtained concentrations from R2 may be of environmental concern.

4.2 PHARMACEUTICALS IN SEWAGE SLUDGE

The sewage sludge pile representing June 21 (∑pharmaceuticals mean concentration: 3600 ng/g dw, mean: 65 ng/g dw, median: 4.5 ng/g dw) was the main contributor. The contribution was lower in Aug 21 (2600 ng/g dw, 47 ng/g dw, 5.3 ng/g dw), July 21 (2200 ng/g dw, 39 ng/g dw, 4.4 ng/g dw), May 21 (2000 ng/g dw, 36 ng/g dw, 4.1 ng/g dw), Storvreta (1800 ng/g dw, 33 ng/g dw, 2.4 ng/g dw) and Sep 21 (1800 ng/g dw, 32 ng/g dw, 2.9 ng/g dw). April 21 (1500 ng/g dw, 27 ng/g dw, 2.7 ng/g dw), Feb 21 (1400 ng/g dw, 26 ng/g dw, 2.5 ng/g dw) and March 21 (1400 ng/g dw, 25 ng/g dw, 3 ng/g dw) had the lowest contributions of all sludge piles in the sludge cell. Of which Feb 21 up to April 21 were 2.4 – 2.6 times lower than June 21 (Table A6). The ∑pharmaceuticals mean concentration in every sewage sludge SP from the sludge cell is presented in Figure 10.

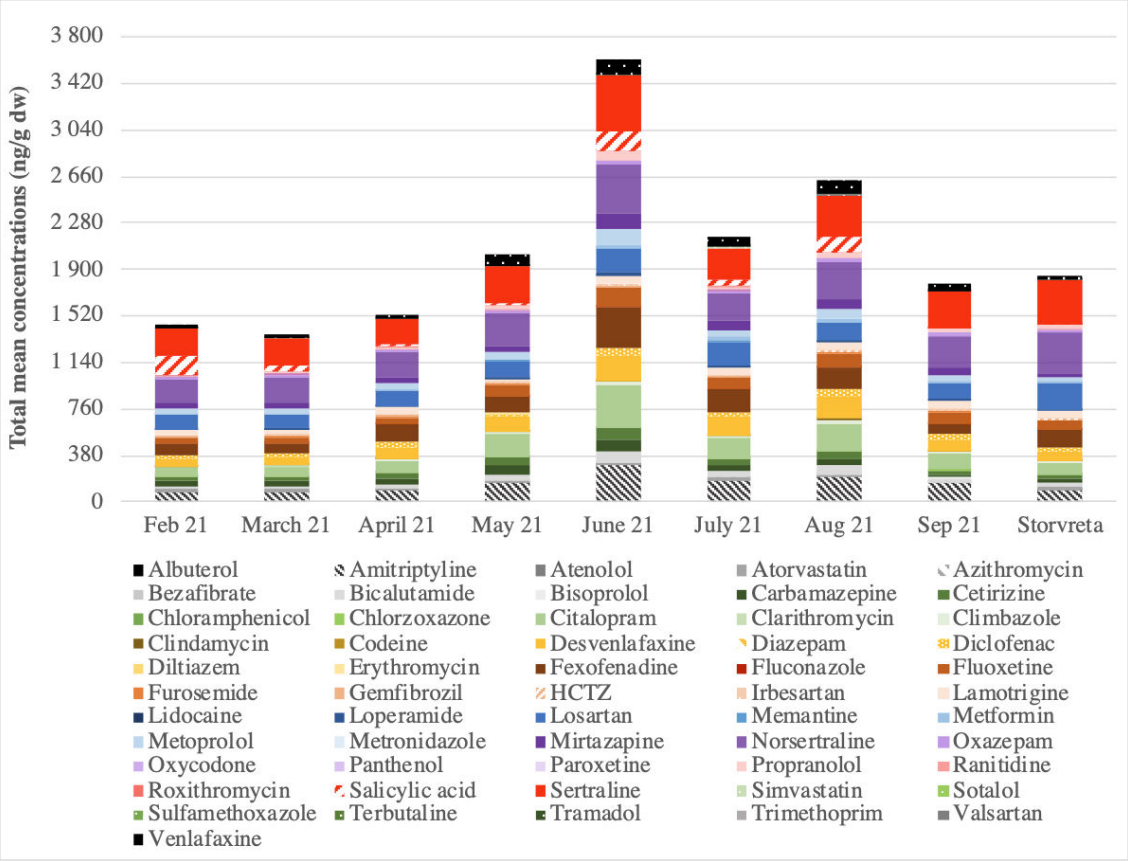

Figure 10: Total mean concentration (ng/g dw) of pharmaceuticals in every sewage sludge grab sampling point in the sludge cell.

The detection frequency of pharmaceuticals was the highest in the sewage sludge piles representing June 21 up to Sep 21 (*n = 32*, respectively), followed by May 21 (*n = 31*), March 21 and April 21 (*n = 30*, respectively), Feb 21 (*n = 29*) and Storvreta (*n = 26*). 24 pharmaceuticals were detected in all SPs, ranging from 1.3 ng/g dw (memantine) to 460 ng/g dw (sertraline). 11 pharmaceuticals were detected in at least one SP, ranging from 1.3 ng/g dw (bisoprolol and clindamycin) to 150 ng/g dw (salicylic acid) (Table A6). More detailed information about which pharmaceuticals that were detected in the specific sewage sludge SPs can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.2, and in Table A6.

Nine of the 48 detected pharmaceuticals (19 %) in L7 (stream 2) were n.d. in the sludge piles in the sludge cell (albuterol, chloramphenicol, clarithromycin, codeine, diazepam, fluconazole, lidocaine, terbutaline and tramadol). Four of the 35 pharmaceuticals that were detected in at least one sludge pile (11 %) were n.d. in L7 (climbazole, norsertraline, oxazepam and panthenol) (Table A5 and A6). The pharmaceuticals that were detected in the sludge piles but n.d. in L7 remain in the sludge instead of leaching out and vice versa.

The ∑pharmaceuticals mean compositions in every sewage sludge SP from the sludge cell is presented in Figure 11. The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in all SPs included amitriptyline (5.1 – 8.4 %), citalopram (5.2 – 9.7 %), losartan (5.3 – 11 %), norsertraline (10 – 18 %) and sertraline (12 – 20 %) (Figure 11). Fexofenadine was a dominating contributor in all SPs except in Sep 21 (5.4 – 9.8 %). Also, fluoxetine in Sep 21 (5.0 %), salicylic acid in Feb 21 and Aug 21 (10 and 5.3 %, respectively) and desvenlafaxine in April 21 up to Sep 21 (5.5 – 7.0 %). Sertraline (12 – 20 %) and norsertraline (10 – 18 %) were the main contributors in all SPs (ibid.). More detailed information about the mean composition profile of pharmaceuticals in the sewage sludge can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.2.

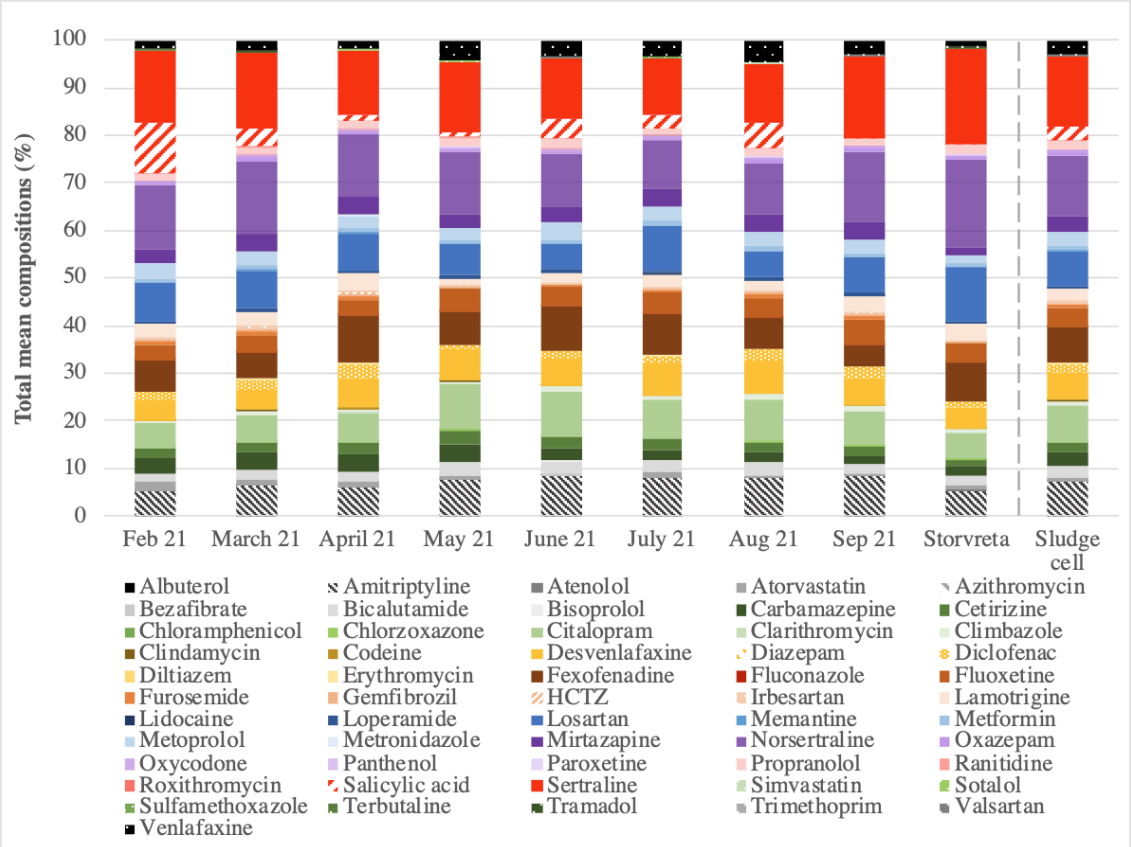

Figure 11: Total mean composition profile (%) in every sewage sludge grab sampling point and in the sludge cell, based on normalized mean concentrations of the pharmaceuticals.

The lowest ∑pharmaceuticals mean concentrations were observed during the spring i.e., in the sludge pile from March 21 to May 21 (on average, 1600 ng/g dw) (Table A6). This could be a result of natural attenuation (such as biodegradation) and the pharmaceuticals physico-chemical properties (such as water solubility). However, the highest ∑pharmaceuticals mean concentrations were observed during the summer i.e., from June 21 to Aug 21 (on average, 2800 ng/g dw), followed by the fall i.e., Sep 21 (1800 ng/g dw) (ibid.). The decrease in ∑pharmaceuticals mean concentrations from summer to fall indicates that not only natural attenuation influences. It could for instance be a result of higher consumption and disposal of pharmaceuticals during the summer and/or that pharmaceuticals are more stable (e.g., resistant to degradation) in the sludge piles from the summer months. Moreover, the pharmaceutical occurrence in the sludge is most likely due to our consumption and disposal since it originates from treated wastewater at Kungsängsverket and Storvreta.

The ∑pharmaceuticals mean concentrations were generally higher in stream 2 (including L6) compared to all sludge piles (on average, 7800 ng/L and 2000 ng/g dw, respectively). As well as the detection frequency (on average, 1.3 times higher in stream 2) (Table A5 and A6). This could be a result of biodegradation reactions, which are less active in the water zone compared to the soil zone as described by Li (2014). This may also be caused by the pharmaceuticals lipo- or hydrophilic nature as well. Their hydrophilic nature could explain why the ∑pharmaceuticals mean concentrations were generally higher in stream 2 compared to the sludge piles. It could also be due to exposure history with pharmaceuticals that are persistent and resistant to degradation. For example, venlafaxine has been mentioned to have high affinity to sludge (Golovko et al. 2021), although higher mean concentrations were found in stream 2 than in the

sludge piles (on average, 290 ng/L and 63 ng/g dw, respectively) (Table A5 and A6). Compounds such as desvenlafaxine, diclofenac, erythromycin, fexofenadine, losartan and valsartan were also detected in L7 in higher mean concentrations (on average, 1200 ng/L) compared to the sludge piles (on average, 60 – 130 ng/g dw) (ibid.). This could be a result of the compounds being water soluble. It could also be due to compounds being persistent and resistant to degradation, such as desvenlafaxine and fexofenadine, which are compounds that were previously suspected of having these characteristics in this study (pp. 28, 29). Whereas the pharmaceuticals lipophilic nature could explain why some substances were detected in the sludge pile(s), but not in the first SP that receives sludge water (L7) (p. 35). As well as why e.g., fluoxetine, mirtazapine, amitriptyline, citalopram and sertraline had higher mean concentrations in the sludge piles compared to stream 2 (including L6) (on average, 150 ng/g dw and 21 ng/L, respectively) (Table A5 and A6). In which these psychoactive compounds have previously been reported to have a lipophilic nature and thus high sorption affinity to sludge (Golovko et al. 2021).

Only one sample was extracted for Feb 21, April 21 and Storvreta, instead of two, which could question the reliability of the obtained measurements. However, substances that were frequently detected (> 90%) in a study conducted by Golovko et al. (2021), combined with all sludge piles having similar dominating contributors in this study (p. 35) says otherwise. For example, compounds detected in all sludge piles (with some being dominating/main contributors) were included in that study (such as amitriptyline, bicalutamide, carbamazepine, cetirizine, citalopram, diclofenac, fexofenadine, fluoxetine, lamotrigine, losartan, metformin, metoprolol, mirtazapine, norsertraline, propranolol, sertraline and venlafaxine) (Figure 11 and Golovko et al. 2021).

Furthermore, a metabolic transformation of pharmaceuticals can occur within human and animal bodies, of which its metabolites consequently reach WWTPs after excretion (Han & Lee 2017). This can also occur via biotransformation during wastewater treatment, in which pharmaceutical metabolites can be detected in higher concentration than its parent compounds (Golovko et al. 2020). Which indicates that the metabolite has stronger persistence (ibid.). This could for example explain the lower mean concentration of venlafaxine compared to its metabolite desvenlafaxine in all sludge piles (on average, 63 and 120 ng/g dw, respectively) and in stream 2 (on average, 290 and 360 ng/L, respectively) (Table A5 and A6). Due to the uncertainties regarding effects that transformation products could cause (Golovko et al. 2021) combined with studies showing that some metabolites could pose a greater toxicity than the parent compound in the aquatic environment (Han & Lee 2017; Golovko et al. 2021), this shows the importance of monitoring metabolites as well. For example, norsertraline (antidepressant metabolite of sertraline) showed higher sorption to all sludge piles (on average, 260 ng/g dw) compared to stream 2 (n.d. in all SPs i.e., < 10 ng/L) (Table A5 and A6). If norsertraline is spread on agricultural land, it could perhaps harm the (aquatic) environment if e.g., runoff occurs and/or if it infiltrates into the groundwater.

In addition, salicylic acid was detected in seven out of the nine sludge piles, ranging from 15 ng/g dw (April 21) to 150 ng/g dw (Feb 21) (Table A6). This pharmaceutical is included in PRIO's database and is therefore a compound that Revaq-certified treatment plants need to monitor (p. 11). Which shows that it is important to monitor and evaluate the occurrence of pharmaceuticals in sludge as well.

The results (Table A5 and A6) showed that some pharmaceuticals leach out rather than remaining absorbed to the sludge, since 19 % of the detected pharmaceuticals in L7 were n.d. in all sludge piles. However, some compounds exhibited more sorption to the sludge, of which 11 % of the detected pharmaceuticals in sludge were n.d. in L7 (p. 35). The sludge piles were only compared to L7 regarding substances that remain in or leach out from the sludge since this is the first SP that receives sludge water. For example, L8 – L12 may be influenced by L6, which would've increased the uncertainty regarding which pharmaceuticals that remain in or leach out from the sludge in the sludge cell.
Furthermore, stream 2 generally had higher detection frequencies and ∑pharmaceuticals mean concentrations compared to all sludge piles as previously mentioned (pp. 36, 37). This indicates that sludge is a prominent pathway for pharmaceuticals to be transported via leachate and to potentially be released into the (aquatic) environment. However, the difference in magnitude between stream 2 and the sludge piles should be taken on lightly since other factors may have contributed. For example, earlier exposure with persistent pharmaceuticals that are resistant to degradation in the sludge water. As well as other waste types being stored in the soil cell such as excavated masses (p. 14). However, this waste is not believed to contribute to the occurrence of pharmaceuticals as previously described (p. 28). Furthermore, there is an uncertainty about how representative the grab sampling is and thus how reliable the measurements are, since this was the first time that the pharmaceutical occurrence was evaluated at Hovgården (with no previous results to compare with). Further screenings would provide more information about which pharmaceuticals that leach out or that remain absorbed to the sludge. Thus, which pharmaceuticals that are persistent in the matrix and which ones that should be evaluated in terms of environmental concern. It would also decrease uncertainties regarding the magnitude of the pharmaceutical occurrence and its causes.

4.3 PCA OF PHARMACEUTICALS IN THE STREAMS, RIVER AND SLUDGE

The PCA biplot of the pharmaceuticals (i.e., observation points) related to all streams and the river (i.e., variables) is presented in Figure 12.
All variables had large positive loadings on PC1 and were positively correlated to each other. Stream 2 and stream 1 were more correlated to each other, as well as the mixed stream and the river. Stream 1 showed a higher correlation than stream 2 to the mixed stream and the river. Stream 2 and the river had the lowest correlation (Figure 12).
The clustered observation points located close to the left of the origin ($n = 36$) were highly correlated to each other, with a variance close to the average (ibid.). These pharmaceuticals had little or no influence on PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones on the right. Whereas chloramphenicol, norsertraline (closer to the mixed stream and the river), bicalutamide, valsartan and venlafaxine (closer to stream 2 and stream 1) were more related to the variables they were closer to (ibid.).
The observation points located to the right of the origin ($n = 15$) had more influence on PC1 and PC2, and thus on the correlations between the variables. The ones of most significance were carbamazepine, desvenlafaxine, erythromycin, furosemide and losartan (ibid.). Desvenlafaxine and losartan had the highest mean concentrations in stream 2 (on average, 1200 and 1000 ng/L, respectively) and in stream 1 (on average, 800 and 690 ng/L, respectively) (Table A5). Carbamazepine, erythromycin and furosemide also had the highest mean concentrations in stream 1 (on average, 490, 1300 and 220 ng/L, respectively) and in stream 2 (on average, 420, 390 and 150 ng/L, respectively) (ibid.). Whereas desvenlafaxine, losartan, carbamazepine, erythromycin and furosemide were much lower in the mixed stream (on average, 11, 17, 58, 68 and

99 ng/L, respectively) and in the river (on average, 10, 11, 39, 30 and 64 ng/L, respectively) (ibid.). Which indicates that carbamazepine, desvenlafaxine, erythromycin, furosemide and losartan are persistent in stream 1 and stream 2. Also, the mean compositions of these compounds varied between the streams and the river. For example, desvenlafaxine's and losartan's mean composition were lower in the mixed stream and the river compared to stream 1 and stream 2 (Figure 5, 7 and 9). In which desvenlafaxine was the main contributor in stream 2 and losartan was a dominating contributor in stream 1 (S4.1.1 and S4.1.2 in Appendix). Whereas furosemide's mean composition was lower in stream 2 and stream 1 compared to the mixed stream and the river (Figure 5, 7 and 9), where it was the main contributor (S4.1.3 in Appendix). This indicates that furosemide had a higher presence than desvenlafaxine and losartan in the river and in the mixed stream (and the opposite for stream 1 and stream 2). Moreover, carbamazepine and erythromycin were dominating contributors in all variables, with the lowest mean composition in stream 2 (5.4 and 5.0 %, respectively) (S4.1.1 – S.4.1.3 in Appendix). Carbamazepine's mean composition was the highest in the river, followed by the mixed stream and stream 1 (11, 9.2 and 8.0 %, respectively). Whereas erythromycin's was the highest in stream 1 (and the main contributor), followed by the mixed stream and the river (21, 11 and 8.5 %, respectively) (ibid.). This means that carbamazepine had a higher presence than erythromycin in the river and stream 2, whereas it was the opposite in the mixed stream and stream 1.

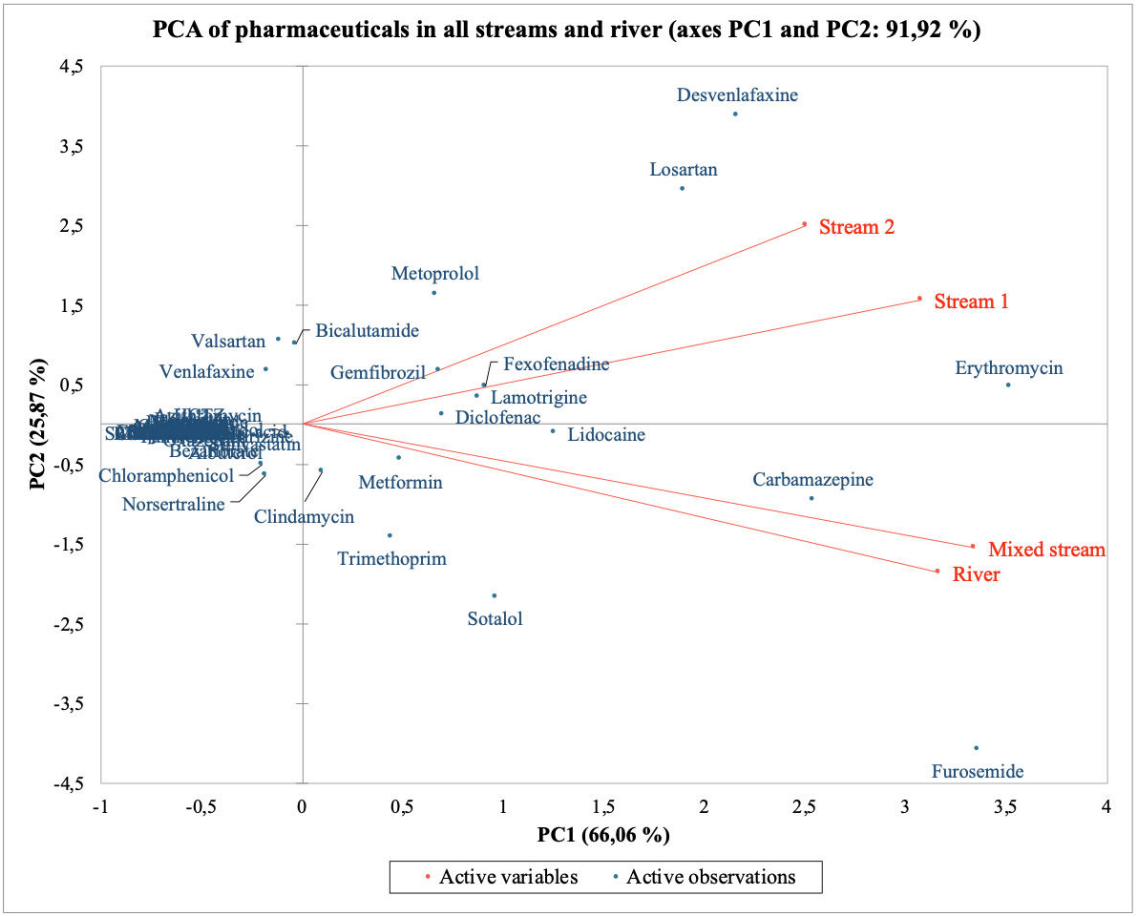

Figure 12: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

The PCA biplot of the pharmaceuticals (i.e., observation points) related to all sewage sludge SPs (i.e., variables) is presented in Figure 13.
All variables had large positive loadings on PC1 and were positively correlated to each other. The most significant observation was that Storvreta had the lowest correlation to June 21 and Aug 21 and the highest correlation to March 21 (Figure 13). Which indicates that similar pharmaceuticals were consumed in the variables that were the most correlated. A PCA for the sludge from solely Kungsängsverket was performed as well, with all vectors still being positively correlated (Figure A26 in Appendix). Which indicates that sludge from Kungsängsverket and Storvreta are similar, although months may now have different correlations, such as Feb 21 and March 21 that would now have the highest correlation (Figure 13 and A26). The similarities in the sludge from Kungsängsverket and Storvreta indicates that the pharmaceutical occurrence in sludge water may be influenced by the sludge from both treatment plants.
The clustered observation points located to the left of, or near, the origin ($n = 40$) were highly correlated to each other, with a variance close to the average (Figure 13). These compounds had little or no influence on PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones on the right (ibid.).
The observation points located to the right of the origin ($n = 16$) had more influence on PC1 and PC2 and therefore on the correlations between the variables as well. The ones of most significance were amitriptyline, citalopram, desvenlafaxine, fexofenadine, losartan, norsertraline and sertraline (ibid.). Which were dominating contributors in all variables (except fexofenadine in Sep 21 and desvenlafaxine in Feb 21 and March 21) (p. 35). Storvreta had the highest mean concentration of losartan (followed by July 21 and June 21) (Table A6). Whereas June 21 had the highest mean concentration of amitriptyline, citalopram, desvenlafaxine, fexofenadine, norsertraline and sertraline (followed by Aug 21 of citalopram and desvenlafaxine, and Storvreta of norsertraline and sertraline) (ibid.). This may indicate that there is a higher consumption of these pharmaceuticals during these months and in the sludge from Storvreta compared to the ones with lower mean concentrations. Furthermore, all variables were the most related to norsertraline and sertraline since they were the main contributors (p. 35). Storvreta, March 21, Feb 21 and July 21 were thereafter more related to losartan as well as Sep 21 to amitriptyline, April 21 to fexofenadine and May 21, June 21 and Aug 21 to citalopram (Figure 11). Which may indicate a higher consumption of these pharmaceuticals during these months and in the sludge from Storvreta.

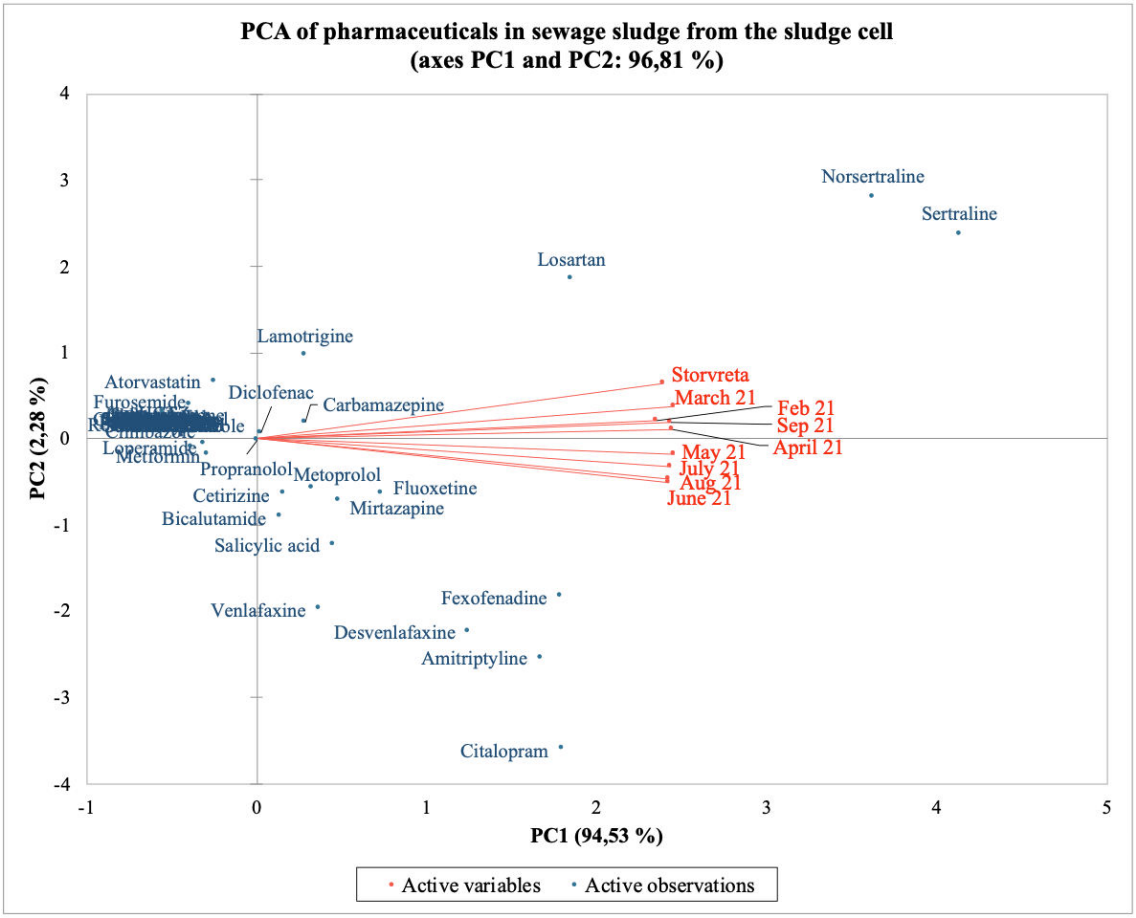

Figure 13: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

In addition, the PCA biplot of only the sludge from Kungsängsverket showed different placements of some observation points when comparing Figure 13 to A26. For example salicylic acid, which had a high mean composition in Feb 21, compared to the other observations on the right side (excluding norsertraline and sertraline) (Figure 11, 13 and Figure A26). Feb 21 and June 21 (and Aug 21) also had the highest mean concentration of salicylic acid compared to all SPs, whereas it was n.d. in Sep 21 and Storvreta (Table A6). This means that Storvreta affected the correlations between the variables and the observations points which should be taken into consideration when observing Figure 13.

4.4 OTHER CECs IN WATER

The other CECs will be analysed based on which group the compound belongs to; PFASs or remaining CECs. The ∑PFASs and ∑remaining CECs mean concentrations and mean compositions in every water GS point within Hovgården's facility are presented in Appendix, Figure A4.1 – A4.2 and A5.1 – A5.2.

Nine of the other CECs were detected in all SPs (PFBS, PFHxA, PFOA, PFOS_linear and PFPeA, caffeine, DEET, nicotine and tolytriazole), ranging from 11 ng/L (PFOS_linear) to 590 ng/L (PFOA) of PFASs and from 2.3 ng/L (caffeine) to 5300 ng/L (caffeine) of the remaining CECs. 19 of the other CECs were detected in at least one SP. The mean concentrations ranged from 1.7 ng/L (PFTeDA) to 290 ng/L (PFHpA) of

PFASs and from 1.7 ng/L (clotrimazole) to 3300 ng/L (BAM) of the remaining CECs. FOSA was only detected in W2 (3.7 ng/L) (Table A7). More detailed information about the 19 of the other CECs that were detected at Hovgården can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.

The dominating contributors (≥ 5.0 % in mean composition) of PFASs at Hovgården included PFHxS, PFOS_linear, PFBS, PFHpA, PFPeA and PFHxA (6.6, 8.0, 11, 13, 16 and 23 %, respectively) (Figure A5.1). Whereas it included tolytriazole, nicotine, DEET, BAM and caffeine (10, 12, 14, 20 and 39 %, respectively) of the remaining CECs (Figure A5.2).

4.4.1 Other CECs in leachate from the landfill (and the HSs) in stream 1

L4 (∑PFASs mean concentration: 2200 ng/L, mean: 170 ng/L, median: 160 ng/L) was the main contributor of PFASs, covering ≈ 37 % of all ∑PFASs mean concentrations in stream 1. The contribution was lower in L5 (2000 ng/L, 160 ng/L, 150 ng/L) and L3 (1100 ng/L, 82 ng/L, 59 ng/L). L2 (710 ng/L, 54 ng/L, 11 ng/L) had the lowest contribution in stream 1, which was 3.1 times lower than L4 (Table A7). The ∑PFASs mean concentrations in stream 1 are presented in Figure A6.

The detection frequency of PFASs was the highest in L3 and L5 (*n = 12*, respectively), followed by L4 (*n = 9*) and L2 (*n = 7*) (Table A7). Seven PFASs were detected in all SPs and five PFASs were detected in at least one SP (Table A7). More detailed information about which PFASs that were detected in specific SPs in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.1, and in Table A7.

The ∑PFASs mean compositions in stream 1 are presented in Figure A7. The dominating contributors (≥ 5.0 % in mean composition) of PFASs in all SPs included PFBS, PFHpA, PFHxA, PFOA and PFPeA (Figure A7). Also, PFHxS and PFOS_linear in all SPs except in L2. PFOA was the main contributor in all SPs (8.8 – 27 %), except in L2 where it was PFHxA (37 %) (ibid.). More detailed information about the mean composition profile of PFASs in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.1.

L4 (∑remaining CECs mean concentration: 1800 ng/L, mean: 100 ng/L, median: 2.1 ng/L) was the main contributor of the remaining CECs, covering ≈ 37 % of all ∑remaining CECs mean concentrations in stream 1. The contribution was lower in L2 (1500 ng/L, 86 ng/L, 3.7 ng/L) and L5 (1400 ng/L, 84 ng/L, 2.1 ng/L). L3 (150 ng/L, 8.9 ng/L, 2.1 ng/L) had the lowest contribution in stream 1, which was 12 times lower than L4 (Table A7). The ∑remaining CECs mean concentrations in stream 1 are presented in Figure A8.

The detection frequency of the remaining CECs was the highest in L2 (*n = 9*), followed by L3, L4 and L5 (*n = 6*, respectively) (Table A7). Four of the remaining CECs were detected in all SPs and eight of the remaining CECs were detected in at least one SP. Clotrimazole, ethylparaben, imazalil, prochloraz and tebuconazole were only detected in L2, as well as propylparaben in L3 (Table A7). More detailed information about which of the remaining CECs that were detected in specific SPs in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.1, and in Table A7.

The ∑remaining CECs mean compositions in stream 1 are presented in Figure A9. The dominating contributor (≥ 5.0 % in mean composition) of the remaining CECs in all SPs included DEET (Figure A9). Also, BAM in all SPs except in L2, ethylparaben and methylparaben in L3 and tolytriazole in L2 and L3. DEET was the main contributor in all SPs (ranging between 27 – 85 %). Tolytriazole was also a main contributor in L2 (42 %) (ibid.). More detailed information about the mean composition profile of the remaining CECs in stream 1 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.1.

4.4.2 Other CECs in leachate from the sludge (and the soil) cell in stream 2

L12 (∑PFASs mean concentration: 580 ng/L, mean: 44 ng/L, median: 40 ng/L) was the main contributor of PFASs, covering ≈ 29 % of all ∑PFASs mean concentrations in stream 2. The contribution was lower in L11 (400 ng/L, 31 ng/L, 20 ng/L), L10 (370 ng/L, 29 ng/L, 23 ng/L), L7 (260 ng/L, 20 ng/L, 10 ng/L) and L8 (200 ng/L, 16 ng/L, 3.6 ng/L). L6 (150 ng/L, 11 ng/L, 3.2 ng/L) had the lowest contribution in stream 2, which was 3.9 times lower than L12 (Table A7). The ∑PFASs mean concentrations in stream 2 are presented in Figure A10.

The detection frequency of PFASs was the highest in L10 (*n = 12*), followed by L11 and L12 (*n = 11*, respectively), L8 (*n = 10*), L7 (*n = 9*) and L6 (*n = 7*) (Table A7). Six PFASs were detected in all SPs and six PFASs were detected in at least one SP (Table A7). More detailed information about which PFASs that were detected in specific SPs in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.2, and in Table A7.

The ∑PFASs mean compositions in stream 2 are presented in Figure A11. The dominating contributors (≥ 5.0 % in mean composition) of PFASs in all SPs included PFBS, PFHxA, PFOS_linear and PFPeA (Figure A11). Also, PFOA in all SPs except in L10, PFDoDA and PFUnDA in L10, PFHxS in L8, L11 and L12 and PFHpA in L6, L10, L11 and L12. PFHxA was the main contributor in all SPs (17 – 49 %), except in L12 where it was PFOA (24 %). PFOA was also a main contributor in L11 (23 %) (ibid.). More detailed information about the mean composition profile of PFASs in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.2.

L11 (∑remaining CECs mean concentration: 8700 ng/L, mean: 510 ng/L, median: 3.7 ng/L) was the main contributor of the remaining CECs, covering ≈ 35 % of all ∑remaining CECs mean concentrations in stream 2. The contribution was lower in L8 (6300 ng/L, 370 ng/L, 6.6 ng/L), L12 (4200 ng/L, 250 ng/L, 3.9 ng/L), L7 (3800 ng/L, 220 ng/L, 3.7 ng/L) and L10 (1600 ng/L, 93 ng/L, 3.7 ng/L), which were ≈ 1.4 – 5.4 times lower than L11. L6 (370 ng/L, 22 ng/L, 2.1 ng/L) had the lowest contribution in stream 2, which was ≈ 24 times lower than L11 (Figure A12 and Table A7). The ∑remaining CECs mean concentrations in stream 2 are presented in Figure A12.

The detection frequency of the remaining CECs was the highest in L12 and L8 (*n = 9*), L11 and L7 (*n = 8*) and lowest in L10 and L6 (*n = 7*, respectively) (Table A7). Five of the remaining CECs were detected in all SPs and six of the remaining CECs were detected in at least one SP (Table A7). More detailed information about which of the remaining CECs that were detected in specific SPs in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.2, and in Table A7.

The ∑remaining CECs mean compositions in stream 2 are presented in Figure A13. The dominating contributors (≥ 5.0 % in mean composition) of the remaining CECs in all SPs included caffeine and tolytriazole (Figure A13). Also, BAM in all SPs except in L11, nicotine in all SPs except in L6, DEET in L6 and daidzein in L7 and L8. BAM was the main contributor in L6, L7, L8 and L10 (40 – 69 %) whereas it was caffeine in L11 (61 %) and L12 (43 %) (ibid.). More detailed information about the mean composition profile of the remaining CECs in stream 2 can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.2.

4.4.3 Other CECs in combined water in the mixed stream and in the river

W2 (∑PFASs mean concentration: 2100 ng/L, mean: 160 ng/L, median: 170 ng/L) was the main contributor of PFASs in the mixed stream, followed by W9 (2000 ng/L, 150 ng/L, 170 ng/L) and W5 (1400 ng/L, 110 ng/L, 29 ng/L). W1 (1300 ng/L, 99 ng/L, 41 ng/L) had the lowest contribution in the mixed stream, 1.6 times lower than W2. R0 (870 ng/L, 67 ng/L, 18 ng/L) was the main contributor in the river, followed by R2 (840 ng/L, 65 ng/L, 38 ng/L). R2 also had the lowest contribution compared to all the SPs from the mixed stream, which was 2.5 times lower than W2 (Table A7). The ∑PFASs mean concentrations in the mixed stream and in the river are presented in Figure A14.

The ∑PFAS concentrations increased from W1 to W9 (Figure A14), showing a negative removal efficiency of 54 % in general. This was not confirmed in the corresponding TIS/FPS points (Figure A23.1), where it instead showed a positive removal efficiency of 18 % from W1TI to W9FP. Furthermore, the ∑PFASs mean concentration decreased from W9 to R2 (Figure A14), showing a positive removal efficiency of 58 %.

The detection frequency of PFASs was the highest in W2 (*n = 11*), followed by W1 (*n = 10*), W5, W9 and R0 (*n = 9*, respectively) and R2 (*n = 8*) (Table A7). Eight PFASs were detected in all SPs and three PFASs were detected in at least one SP. FOSA was only detected in W2 (Table A7). More detailed information about which PFASs that were detected in specific SPs in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.3, and in Table A7.

The ∑PFASs mean compositions in the mixed stream and in the river are presented in Figure A15. The dominating contributors (≥ 5.0 % in mean composition) of PFASs in all SPs included PFBS, PFHpA, PFHxA, PFOA and PFPeA (Figure A15). Also, PFHxS in all SPs except in R2 and PFOS_linear in W2, W9 and R0. PFHxA was the main contributor in all SPs (ranging between 27 – 29 %), except in W2 and W9 of which it was PFOA (28 and 24 %, respectively) (ibid.). More detailed information about the mean composition profile of PFASs in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.3.

W9 (∑remaining CECs concentration: 6900 ng/L, mean: 410 ng/L, median: 3.7 ng/L), was the main contributor of the remaining CECs, followed by W2 (3300 ng/L, 190 ng/L, 3.8 ng/L). W1 (270 ng/L, 16 ng/L, 2.9 ng/L) and W5 (260 ng/L, 15 ng/L, 3.5 ng/L) had the lowest contribution in the mixed stream, which was 26 and 27 times lower than W9, respectively. R2 (380 ng/L, 22 ng/L, 2.1 ng/L) was the main contributor in the river, followed by R0 (300 ng/L, 18 ng/L, 2.1 ng/L), which were 18 and 23 times

lower than W9, respectively (Table A7). The ∑remaining CECs mean concentrations in the mixed stream and in the river are presented in Figure A16.

The ∑remaining CECs concentrations increased from W1 to W9 (Figure A16), showing a general negative removal efficiency of 2500 %. This was not confirmed in the corresponding TIS/FPS points (Figure A23.2), where it instead showed a positive removal efficiency of 80 % in general. Furthermore, the ∑remaining CECs mean concentration decreased from W9 to R2 (Figure A16), showing a positive removal efficiency of 94 % (Table A7).

The detection frequency of the remaining CECs was the highest in W2 ($n = 8$), followed by W9 ($n = 7$), W1, W5 and R0 ($n = 6$, respectively) and R2 ($n = 5$). Four of the remaining CECs were detected in all SPs and six of the remaining CECs were detected in at least one SP. Ethylparaben and penconazole were only detected in W2, as well as propylparaben in W9. Furthermore, daidzein (detected in W9 and R0), propylparaben and tebuconazole (detected in W9) were n.d. in R2. Also, BAM was detected in the river, but not in W9 (Table A7). More detailed information about which of the remaining CECs that were detected in specific SPs in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.3, and in Table A7.

The ∑remaining CECs mean compositions in the mixed stream and in the river are presented in Figure A17. The dominating contributors (≥ 5.0 % in mean composition) of the remaining CECs in all SPs included caffeine and DEET (Figure A17). Also, BAM in R0 and R2, nicotine in W9, R0 and R2 and tolytriazole in W1, W5 and R0. Caffeine was the main contributor in W2 (70 %) and W9 (76 %), whereas it was DEET in W1 (48 %) and W5 (≈ 50 %) and nicotine in R0 (36 %) and R2 (37 %) (ibid.). More detailed information about the mean composition profile of the remaining CECs in the mixed stream and in the river can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.4.3.

R2 is the only SP that will be further analysed due to the limitations of this study and represents the occurrence of PFASs and of the remaining CECs in surface water. 62 % of the analysed PFASs (PFBS, PFHpA, PFHxA, PFHxS, PFNA, PFOA, PFOS_linear, PFPeA) and 24 % of the remaining CECs (BAM, caffeine, DEET, nicotine and tolytriazole) were detected in R2. The mean concentrations ranged from 25 ng/L (PFHxS) to 240 ng/L (PFHxA) of PFASs and from 17 ng/L (tolytriazole) to 140 ng/L (nicotine) of the remaining CECs that were detected in R2 (Table A7). Furthermore, R0 had a lower ∑remaining CECs mean concentration than R2 (Figure A16), which could indicate that the WWTP effluent may be the main contributor to the occurrence of the remaining CECs in the surface water. Of which caffeine has been suggested to serve as a suitable indicator and can be a strong indicator for wastewater contamination (Golovko et al. 2021), which was detected in R2 (at 28 ng/L). This may also indicate that the WWTP effluent is the main contributor for the occurrence of PFASs and pharmaceuticals in R2 as well.

Since PFASs are widely known for being persistent, it does not come to any surprise that they were detected in R2. However, although this surface water is not intended for drinking water use, it could perhaps cause a dispersion of PFASs to areas where this is conducted. This causes concerns since some PFASs exceeded 90 ng/L (PFBS, PFHpA,

PFHxA and PFPeA) (Table A7), which is the recommended limit value for PFASs concentrations in drinking water in Sweden (Livsmedelsverket 2021).

Furthermore, nicotine is a widely detected CEC in the environment and is among the most widely used stimulants (Golovko et al. 2021), which would explain it being the main contributor in R2 (and in R0) (p. 45).

With knowing that CECs tend to have bioaccumulative and toxic characteristics and with observing the above-mentioned findings (pp. 44, 45), this suggests that further monitoring and evaluation of other CECs at Hovgården should be conducted as well.

4.5 OTHER CECs IN SEWAGE SLUDGE

The sewage sludge pile representing June 21 (∑PFASs mean concentration: 30 ng/g dw, mean: 2.3 ng/g dw, median: 0.50 ng/g dw) was the main contributor of PFASs, covering ≈ 24 % of all ∑PFASs mean concentrations. Storvreta (7.3 ng/g dw, 0.56 ng/g dw, 0.50 ng/g dw) had the lowest contribution, which was 4.1 times lower than June 21 (Table A8). The ∑PFASs mean concentrations in every sewage sludge SP from the sludge cell is presented in Figure A18.

The detection frequency of PFASs was very low in the sludge cell. It was the highest in May 21, June 21 and Aug 21 (*n = 3*, respectively). No PFASs were detected in Storvreta. PFOS_linear was detected in all SPs (excluding Storvreta) and four PFASs were detected in at least one SP (Table A8). More detailed information about which PFASs that were detected in the specific sewage sludge SPs can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.5, and in Table A8.

The ∑PFASs mean compositions in every sewage sludge SP from the sludge cell is presented in Figure A19. The dominating contributor (≥ 5.0 % in mean composition) of PFASs in all SPs included PFOS_linear (Figure A19). Also, PFUnDA in all SPs except in June 21. PFOS_linear was the main contributor in all SPs (25 – 37 %), except in Storvreta where it was PFUnDA (12 %). PFHxA was also a main contributor in June 21 (36 %) (ibid.). More detailed information about the mean composition profile of PFASs in the sewage sludge can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.5.

June 21 (∑remaining CECs mean concentration: 750 ng/g dw, mean: 44 ng/g dw, median: 6.5 ng/g dw) was the main contributor of the remaining CECs, covering ≈ 17 % of all ∑remaining CECs mean concentrations. Storvreta (260 ng/g dw, 15 ng/g dw, 2.6 ng/g dw) had the lowest contribution, which was 2.9 times lower than June 21 (Table A8). The ∑remaining CECs mean concentrations in every sewage sludge SP from the sludge cell is presented in Figure A20.

The detection frequency of the remaining CECs was the highest in June 21 (*n = 11*), followed by July 21, Aug 21 and Sep 21 (*n = 10*, respectively), May 21 (*n = 9*), Feb 21, March 21 and Storvreta (*n = 8*, respectively) and April 21 (*n = 6*) (Table A8). Six of the remaining CECs were detected in all SPs and six of the remaining CECs were detected in at least one SP. BAM was only detected in March 21, as well as ethylparaben in June 21 (Table A8). More detailed information about which of the remaining CECs that were detected in the specific sewage sludge SPs can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.5, and in Table A8.

The ∑remaining CECs mean compositions in every sewage sludge SP from the sludge cell is presented in Figure A21. The dominating contributors (≥ 5.0 % in mean composition) of the remaining CECs in all SPs included clotrimazole, imazalil and miconazole (Figure A21). Also, nicotine in Sep 21, tolytriazole in Storvreta and tebuconazole in Feb 21, June 21, Aug 21, Sep 21 and Storvreta. Imazalil was the main contributor in all SPs (47 – 70 %) (ibid.). More detailed information about the mean composition profile of the remaining CECs in the sewage sludge can be found in Appendix, section "*Supplementary information: Results and discussion",* S4.5.

The presence of PFASs and the remaining CECs could be a result of being disposed of into sewage systems and being absorbed to the sludge during treatment at Kungsängsverket and Storvreta. It is therefore important to have further monitoring and evaluations of other CECs in sludge at Hovgården as well since this sludge is used to eventually be spread onto, for example, agricultural land.

4.6 PCA OF OTHER CECs IN THE STREAMS, RIVER AND SLUDGE

The PCA biplot of the PFASs (i.e., observation points) related to all streams and the river (i.e., variables) is presented in Figure A27.
All variables had large positive loadings on PC1 and were positively correlated to each other. The river and stream 2 were more correlated to each other, as well as the mixed stream and stream 1. Stream 2 showed a higher correlation than the river to the mixed stream and stream 1. The river and stream 1 had the lowest correlation (Figure A27).
The observation points that are located close to each other to the left of the origin (PFDoDA, PFNA, PFUnDA, PFDA, FOSA and PFTeDA) had similar variances. These PFASs had little or no influence on PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones on the right (ibid.).
The observation points located to the right of the origin (PFBS, PFHpA, PFHxA, PFOA, PFOS_linear and PFPeA) had more influence on PC1 and PC2, and thus on the correlations between the variables. The ones of most significance were PFOA and PFHxA (ibid.). PFOA and PFHxA had higher mean concentrations in the mixed stream (on average, 370 and 380 ng/L, respectively), followed by stream 1 (on average, 350 and 300 ng/L, respectively), the river (81 and 240 ng/L on average, respectively) and stream 2 (on average, 50 and 89 ng/L, respectively) (Table A7). These compounds were also dominating contributors in all variables (S4.4.1 – S4.4.3 in Appendix). PFHxA had a higher mean composition than PFOA in the river and stream 2 (Figure A11 and A15). Whereas PFOA had a higher mean composition than PFHxA in stream 1 and the mixed stream (Figure A7 and A15). Which indicates that PFHxA had a higher presence than PFOA in the river (+19 %) and stream 2 (+12 %) (Figure A11 and A15), whereas the presence of PFOA and PFHxA were quite equivalent in stream 1 (± 3.0 %) and in the mixed stream (± 0.0 %) (Figure A7 and A15). Moreover, the river had a higher mean composition i.e., higher presence, of PFPeA (and PFHpA and PFBS) than of PFOA (Figure A15).

The PCA biplot of the PFASs (i.e., observation points) related to all sewage sludge SPs (i.e., variables) is presented in Figure A28.
All variables had large positive loadings on PC1, except Storvreta which had large positive loadings on PC2. All variables with sludge from Kungsängsverket were positively correlated to each other, of which May 21 and June 21 had the lowest correlation to each other. Storvreta showed no correlation to Feb 21 – May 21 and a

negative correlation to June 21 – Sep 21 (Figure A28). This indicates that similar PFASs were present in the variables that were the most correlated.
The observation points located to the left of the origin on the negative PC2-axis ($n = 8$) had little or no influence PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones on the right and on the positive PC2-axis (ibid.). PFNA was more related to Feb 21 – May 21 (and Storvreta). Whereas PFPeA and PFUnDA had large positive associations to Storvreta (ibid.). It should however be noted that they were n.d. in Storvreta (Table A8). The ones of most significance were PFOS_linear and PFHxA (Figure A28), which had the highest mean concentrations in June 21 (Table A8). PFOS_linear had a high mean composition i.e., high presence in all variables (excluding Storvreta) as well as PFHxA in June 21, Aug 21, July 21 and June 21 (Figure A19).

The PCA biplot of the remaining CECs (i.e., observation points) related to all streams and the river (i.e., variables) is presented in Figure A29.
All variables had large positive loadings on PC1, except stream 1 which had larger positive loadings on PC2. All variables were positively correlated. Stream 1 and the river were more correlated to each other, as well as the mixed stream and stream 2. The river showed a higher correlation than stream 1 to stream 2 and the mixed stream. Stream 1 and the mixed stream had the lowest correlation to each other (Figure A29).
The clustered observation points located to the left of the origin ($n = 11$) were highly correlated to each other, with a variance close to the average. These CECs had little or no influence on PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones to the right of the origin (ibid.). It should however be noted that daidzein was only a dominating contributor in stream 2 (S4.4.1 – S4.4.3 in Appendix) and is therefore more related to this stream than to the other variables.
The observation points located to the right of the origin (BAM, caffeine, DEET, nicotine and tolytriazole) had more influence on PC1 and PC2, and thus on the correlations between the variables as well. The ones of most significance were BAM, caffeine, DEET and nicotine (Figure A29). The mean composition of BAM was the highest in stream 2 (and the river) followed by stream 1, whereas it was the lowest in the mixed stream (Figure A9, A13 and A17). Caffeine's was the highest in the mixed stream (and stream 2) whereas nicotine was the highest in the river (and stream 2). The lowest mean composition of BAM and caffeine were identified in stream 1. DEET's was the highest in stream 1, followed by the river and mixed stream with being the lowest in stream 2 (ibid.). Which indicates that DEET had a higher presence than BAM, nicotine and caffeine in stream 1. Whereas stream 2 had a higher presence of BAM than DEET as well as caffeine in the mixed stream and nicotine in the river when comparing to DEET (ibid.). Furthermore, DEET's mean concentration was higher in stream 1 and in the mixed stream (on average, 820 and 460 ng/L, respectively) compared to stream 2 and the river (71 ng/L on average, respectively) (Table A7). This indicates that DEET has a higher persistence in stream 1 and in the mixed stream. Whereas BAM's and nicotine's were higher in stream 2 (on average, 1200 and 700 ng/L, respectively) compared to stream 1 (on average, 120 and 9.0 ng/L, respectively), the mixed stream (on average, 48 and 160 ng/L, respectively) and the river (on average, 70 and 125 ng/L, respectively) (ibid.). As well as caffeine's in the mixed stream followed by stream 2, with being much lower in the river and stream 1 (on average, 1900, 1300, 29 and 13 ng/L, respectively) (ibid.).

The PCA biplot of the remaining CECs (i.e., observation points) related to all sewage sludge SPs (i.e., variables) is presented in Figure A30.
All variables had large positive loadings on PC1 and were positively correlated to each other, of which Storvreta had the largest positive loading on PC2 (Figure A30).
The observation points located close to the left of the origin (*n = 13*) had little or no influence on PC1 and PC2, and thus on the correlations between the variables as well, compared to the ones on the right of the origin (ibid.). It should however be noted that tolytriazole was only a dominating contributor in Storvreta (p. 47). The observation points of most significance included imazalil, miconazole and tebuconazole (Figure A30). They were dominating contributors in all variables (except for tebuconazole in March 21, July 21 and May 21) (p. 47). Of which all variables were the most related to imazalil since it was the main contributor in all variables (ibid.). This means that all variables had the highest presence of this compound, of which it was the lowest in Storvreta's sludge pile (Figure A21). All variables were then more related to miconazole, except for Aug 21 which was more related to clotrimazole, as well as Storvreta which was more related to tebuconazole. All variables then related to clotrimazole, except for Aug 21 where it was miconazole (ibid.).

4.7 DIFFERENCE BETWEEN DIFFERENT SAMPLING METHODS

Three different sampling methods were performed for the water SPs to evaluate the reliability of the measurements obtained from the GS points, by comparing them to their corresponding TIS or FPS point. It should be noted that "every SP" in this section will further on refer to L5, L6, L8, L11, L12, W1, W2, W5 and W9.

The ∑target compounds mean concentrations and mean composition profiles in every SP with regards to the GS and corresponding TIS/FPS point is presented in Appendix, Figure A22 – A25.2. It was noticeable that the ∑target compounds mean concentration of every GS point that was compared to its corresponding TIS/FPS point varied (Figure A22, A23.1 and A23.2).

The ∑pharmaceuticals mean concentrations, in every SP (excluding W9), increased within the range of 2.4 % (L12) up to 39 % (W1 and W2) (mean: 21 %). Whereas W9's decreased (5.9 %), when the GS was compared to the TIS/FPS method (Figure A22). The detection frequency, in every SP (excluding W9), increased within the range of 0.0 % (L8) up to 55 % (W2) (mean: 24 %). Whereas W9's decreased (5.6 %), when the GS was compared to the TIS/FPS method (Table A5 and A9).

The ∑PFASs mean concentrations, in every SP (excluding L6 and W1), increased within the range of 0.0 % (L5 and W5) up to 33 % (W2) (mean: 13 %). Whereas L6's and W1's decreased (40 and 31 %, respectively), when the GS was compared to the TIS/FPS method (Figure A23.1).
The detection frequency, in every SP (excluding L6), increased within the range of 0.0 % (L12, W1, W5 and W9) up to 40 % (L8) (mean: 15 %). Whereas L6's decreased (22 %), when the GS was compared to the TIS/FPS method (Table A7 and A10).

The ∑remaining CECs mean concentrations, in every SP (excluding L6, W1 and W5), increased within the range of 10 % (L11) up to 97 % (W9) (mean: 48 %). Whereas L6's, W1's and W5's ∑remaining CECs mean concentration decreased (11, 340 and 65 %, respectively), when the GS was compared to the TIS/FPS method (Figure A23.2).
The detection frequency, in every SP (excluding L5), increased within the range of 0.0 % (W5) up to 43 % (W9) (mean: 22 %). Whereas L5's decreased (17 %), when the GS was compared to the TIS/FPS method (Table A7 and A10).

All sampling methods showed an occurrence of CECs, with generally having the highest ∑mean concentrations and detection frequencies in the SPs from the GS method (Table A5, A7, A9 and A10). The ∑mean composition profiles (Figure A24 and A25.1 – A25.2) therefore varied as well. Differences in detection frequencies and/or ∑mean concentrations between the GS and TIS/FPS method indicates that more/less compounds were detected as well as in higher/lower concentrations. It should be noted that SPs with equivalent detection frequencies does not necessarily indicate that the same compounds were detected. For instance, simvastatin was n.d. in the GS of L8 but detected in L8TI, whereas memantine was detected in L8 but not in L8TI (Table A5 and A9). Which causes an uncertainty in the reliability of the CECs concentrations and detection of them.

The influence of adopting a GS, TIS and FPS method to evaluate the occurrence of micropollutants in wastewaters has previously been studied by Verlicchi & Ghirardini (2019). In which three key compounds with distinct properties (representative for a

group of compounds) were considered. Conclusively, FPS provided the most reliable measurements of concentrations and removal efficiencies, followed by TIS. Whereas GS showed the highest uncertainties regarding evaluated mean concentrations of micro-pollutants in the water environment, which agreed with previous research as well (ibid.). The difference in mean concentrations (and thus in ∑mean concentrations) and detection frequencies between the three sampling methods in this study should therefore be taken into consideration with relying more on the measurements from the FPS and TIS, followed by the GS method. Of which the GS and corresponding TIS point that are the most similar may be more reliable when evaluating the occurrence of CECs. However, the GS points are still representative for the wastewaters condition at the time of the collection. The only difference is that it cannot represent the average characteristics of the wastewater during the composition period, which TI and FP samples can (US EPA 2017). L2, L3, L4, L7, R0 and R2 will therefore only portray the CECs occurrence at one specific occasion with, unfortunately, higher uncertainties. This concerns for example when determining if the occurrence of CECs is a result of exposure history with CECs that are persistent and resistant to degradation and/or if it is due to the transported leachate/treated water from a previous SP.

Furthermore, the removal efficiency of pharmaceuticals in the GS points from W1 to W9 was in general positive, whereas it was negative from W1TI to W9FP (pp. 32, 33). In addition, the removal efficiency of PFASs and of the remaining CECs in the GS points from W1 to W9 were in general negative, whereas they were positive from W1TI to W9FP (pp. 44, 45). This would suggest that one should rely more on the measurements from W1TI to W9FP, based on previous studies (Verlicchi & Ghirardini 2019). However, further screenings should first be conducted to evaluate whether the removal efficiency is negative or positive before implementing treatment methods, since this was the first time that the occurrence of pharmaceuticals was evaluated. The occurrence of other CECs has previously been investigated at Hovgården (e.g., Bonnet 2017), but this was not further evaluated in this study since it was not the primary scope. In other words, no comparison was made between the present study and previous studies concerning the occurrence of other CECs at Hovgården and thus it was not determined whether the obtained measurements from this study are reliable.

5 CONCLUSIONS

- A total of 76 CECs (48 pharmaceuticals and 28 other CECs) were detected in the water samples, in mean concentrations ranging from 1.1 ng/L to 4900 ng/L (erythromycin) of pharmaceuticals, from 1.8 ng/L to 590 ng/L (PFOA) of PFASs and from 1.7 ng/L to 5300 ng/L (caffeine) of the remaining CECs. The most frequently detected CECs in the LL, sludge water, WWTP influent and effluent and surface water included six pharmaceuticals (carbamazepine, fexofenadine, lidocaine, losartan, metformin and metoprolol), five PFASs (PFBS, PFHxA, PFOA, PFOS_linear and PFPeA), a pesticide (DEET), an industrial chemical (tolytriazole) and two stimulants (caffeine and nicotine).
- A total of 46 CECs (35 pharmaceuticals and 11 other CECs) were detected in the sludge samples, in mean concentrations ranging from 1.3 ng/g dw to 460 ng/g dw (sertraline) of pharmaceuticals, from 1.5 ng/g dw to 11 ng/g dw (PFOS_linear and PFHxA) of PFASs and from 10 ng/g dw to 480 ng/g dw (imazalil) of the remaining CECs. The most frequently detected CECs in the sludge included 24 pharmaceuticals (amitriptyline, atorvastatin, bicalutamide, carbamazepine, cetirizine, citalopram, climbazole, desvenlafaxine, diclofenac, fexofenadine, fluoxetine, irbesartan, lamotrigine, loperamide, losartan, memantine, metoprolol, mirtazapine, norsertraline, oxazepam, paroxetine, propranolol, sertraline and venlafaxine), a PFAS (PFOS_linear), four pesticides (clotrimazole, imazalil, miconazole and tebuconazole), a stimulant (caffeine) and an industrial chemical (tolytriazole).

- The highest ∑CECs mean concentrations of all water grab SPs (including LL, sludge water, WWTP influent and effluent and surface water) were found in:
  - L2 (LL from the top of the old landfill's final cover) at 23 000 ng/L of all the ∑pharmaceuticals mean concentrations, followed by L8 (leachate from the soil- and the sludge cell) at 16 000 ng/L and L7 (leachate from the sludge cell) at 13 000 ng/L.
  - L4 (leachate from L2, the active landfill and possibly leachate from the hardened surfaces including wood, compost and combustible waste) at 2200 ng/L of all the ∑PFASs mean concentrations.
  - L11 (leachate from the soil- and the sludge cell) at 8700 ng/L of all the ∑remaining CECs mean concentrations.

- The pharmaceutical occurrence in the LL and sludge water most likely originated from leachate formed in sludge in the old landfill's final cover and in sewage sludge. The average sum of ∑pharmaceuticals mean concentrations in the GS points were higher in the sludge water (7800 ng/L) than in the LL (6100 ng/L).

- No conclusions can be drawn whether measures are required at the landfill. Further screenings and a risk characterization could provide more information concerning the reliability of the measurements obtained from this study and if the obtained concentrations may be considered as harmful for the receiving environment.

- Albuterol, chloramphenicol, clarithromycin, codeine, diazepam, fluconazole, lidocaine, terbutaline and tramadol were only detected in the sludge water whereas climbazole, norsertraline, oxazepam and panthenol were only detected in the sludge in the sludge cell, when L7 (the first SP that receives sludge water) was compared to the sludge. The ∑pharmaceuticals mean concentrations and detection frequencies were generally higher in the sludge water in stream 2 compared to the solid sludge in the sludge cell.

- The ∑pharmaceuticals mean concentration in the GS points decreased in general in the WWTP. The treatment steps showed both positive and negative removal efficiencies. It decreased by 20 % after aeration and sedimentation and by 17 % after the MBBR process and sedimentation ponds. It increased by 39 % after passing through the polishing- and oxidation pond(s). The removal efficiency was in general positive from the WWTP influent (W1) to the effluent (W9). The ∑pharmaceuticals mean concentration decreased by 65 % from W9 to the WWTP recipient (R2). R2's ∑pharmaceutical mean concentration (240 ng/L) and detection frequency (*n = 12*) was lower than the upstream reference point of the river (R0), indicating that not only the WWTP effluent may affect the recipient's water quality.
- The WWTP's GS points generally showed a negative removal efficiency of PFASs and of the remaining CECs. The ∑PFASs and ∑remining CECs mean concentration of R2 (840 and 380 ng/L, respectively) was lower than W9. Caffeine was detected, indicating that the WWTP's effluent may be a main cause for CECs to occur in R2.
- The TIS/FPS points removal efficiencies from W1 to W9 showed different results. Further evaluations are therefore necessary to evaluate the reliability of the measurements, before implementing additional treatment methods.

CECs were identified in all sample matrices including LL, sludge water, sludge, WWTP influent and effluent as well as in the surface water. It is therefore important to continue with monitoring the occurrence and dissemination of CECs from these treatment/storing areas, to evaluate whether it may be of concern for the receiving environment.

6 FUTURE RECOMMENDATIONS

It is recommended to conduct further screenings by repeating this study, before implementing treatment methods. This is heavily based on this study being the first to evaluate the occurrence of pharmaceuticals at Hovgården and it can therefore not be said that the results from this study are reliable. It could for example not be concluded if the WWTP effluent was the primary cause for the pharmaceutical occurrence in the recipient R2. This uncertainty will decrease if further screenings are conducted.

It is recommended to continue with collecting samples from the SPs that were chosen in this study, to first evaluate if the obtained results are of regular pattern and thus reliable, since CECs were detected in all SPs. Continuous monitoring of L1 is important to include although no water was accessible during this field sampling since leachate may be generated in the future. It is also recommended to integrate GS and TIS/FPS methods (depending on offered supply), since different CECs were detected (in higher/lower concentrations as well). A TI/FP sample of L2, L3, L4, L7, R0 and R2 could be of value to include, to limit uncertainties concerning detection frequencies, concentrations and the cause of the pharmaceutical occurrence in the various sample matrices (such as in the old and active landfill as well as in the recipient). It would mainly be recommended to include a TI/FP sample of R2, since this SP represents the presence of CECs in the aquatic environment. Multiple grab samples could be collected manually if a TIS/FPS is not practicable. Samples of other hardened surfaces could also be included (such as from the sorting platform), to limit uncertainties concerning the originating source.

A risk characterization is recommended to include in further screenings. This would increase knowledge about which CECs that should be especially reviewed, such as at the landfill. Which could be beneficial while deciding on the most suitable treatment method for the most harmful pollutants. Although the guideline values of the Swedish Food Agency, WFD and PRIO could be used, they do not include all CECs that were analysed and detected in this study. Another approach could therefore be to integrate acute toxicity data with measure of predicted environmental concentration, which is a standard tool for risk estimation according to Kuzmanovic et al. (2013). This would also be suitable to include for sludge to evaluate and assess if the CECs concentrations are of concern, before being distributed on agricultural land for instance. Calculations of, for example, log K_d, log K_{ow} and log K_{oc} could also be sufficient to include, as it could provide information concerning which CECs that are more persistent and resistant to degradation. These coefficients have previously been used in research to interpret/ determine/predict sorption behavior of CECs (pp. 6, 7) and could limit uncertainties regarding causes for the fate and transport of pharmaceuticals for instance. This could, in the long run, help to select the most effective treatment approach for improving the removal efficiency.

Other initiatives could also contribute to the minimization of CECs, such as carrying out upstream work. The presence of CECs in waste (and consequently at waste facilities) are due to our consumption and disposal of CECs. This includes our overuse/misuse of pharmaceuticals with shown consequences for the (aquatic) environment that indirectly could harm the human health, such as increased antimicrobial resistance. The occurrence of CECs in sludge and sewage systems could thus be minimized by also increasing the public's awareness about the consequences of our actions regarding CECs.

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APPENDIX

FIGURES

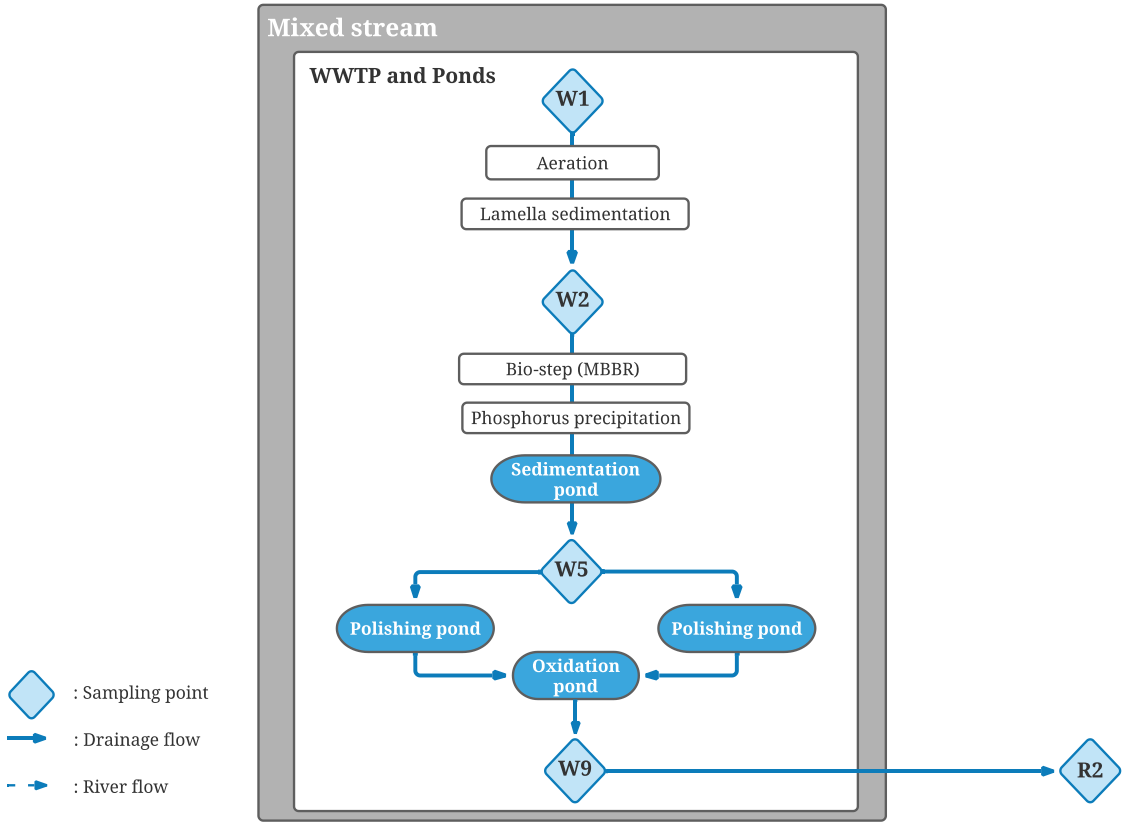

Figure A1: The treatment steps (detailed) in the wastewater treatment plant (WWTP) and Ponds, which represents the mixed stream.

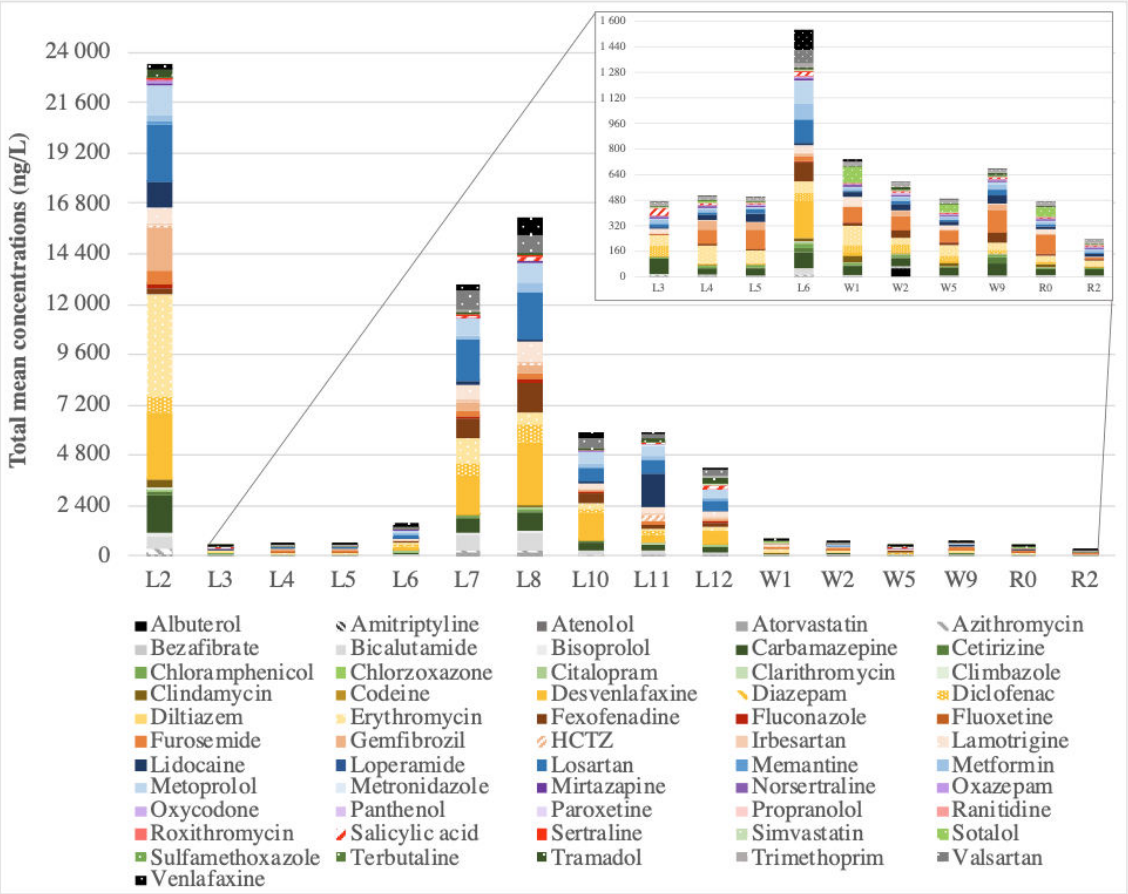

Figure A2: Total mean concentration (ng/L) of pharmaceuticals in every water grab sampling point at Hovgården.

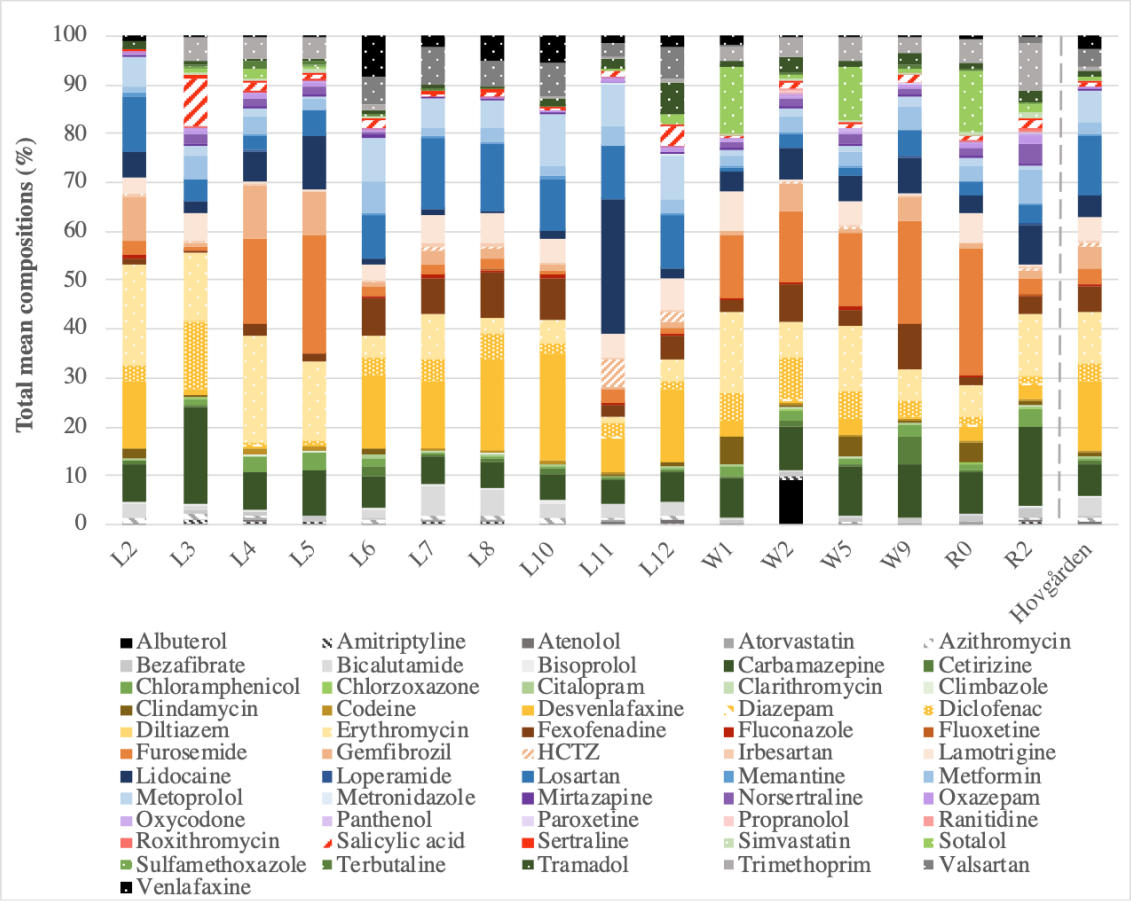

Figure A3: Total mean composition profile (%) in every water grab sampling point and at Hovgården, based on normalized mean concentrations of the pharmaceuticals.

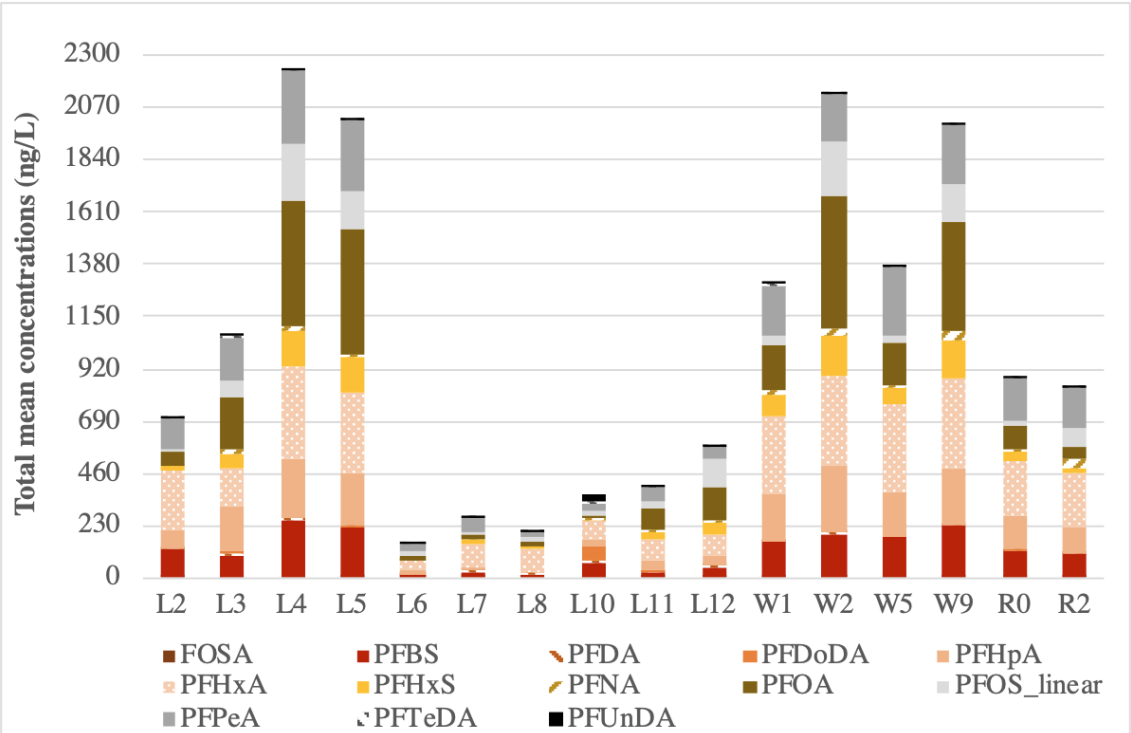

Figure A4.1: Total mean concentration (ng/L) of the per- and polyfluoroalkyl substances (PFASs) in every water grab sampling point at Hovgården.

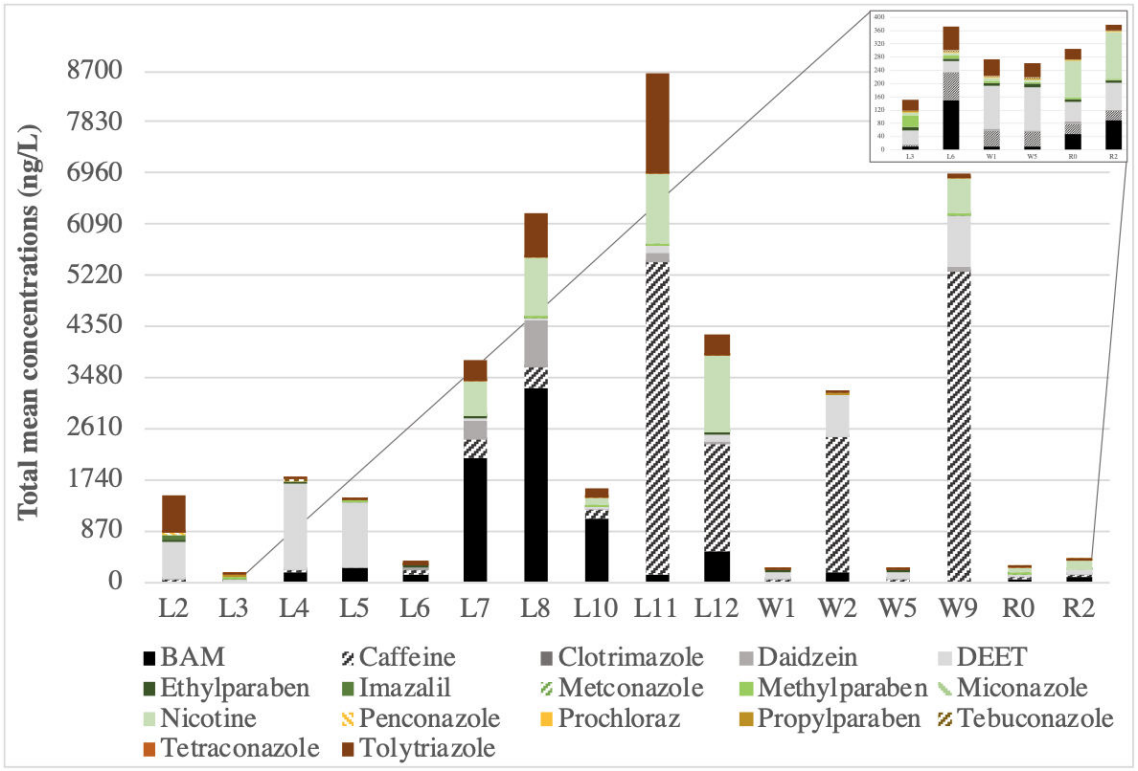

Figure A4.2: Total mean concentration (ng/L) of the remaining chemicals of emerging concern (CECs) in every water grab sampling point at Hovgården.

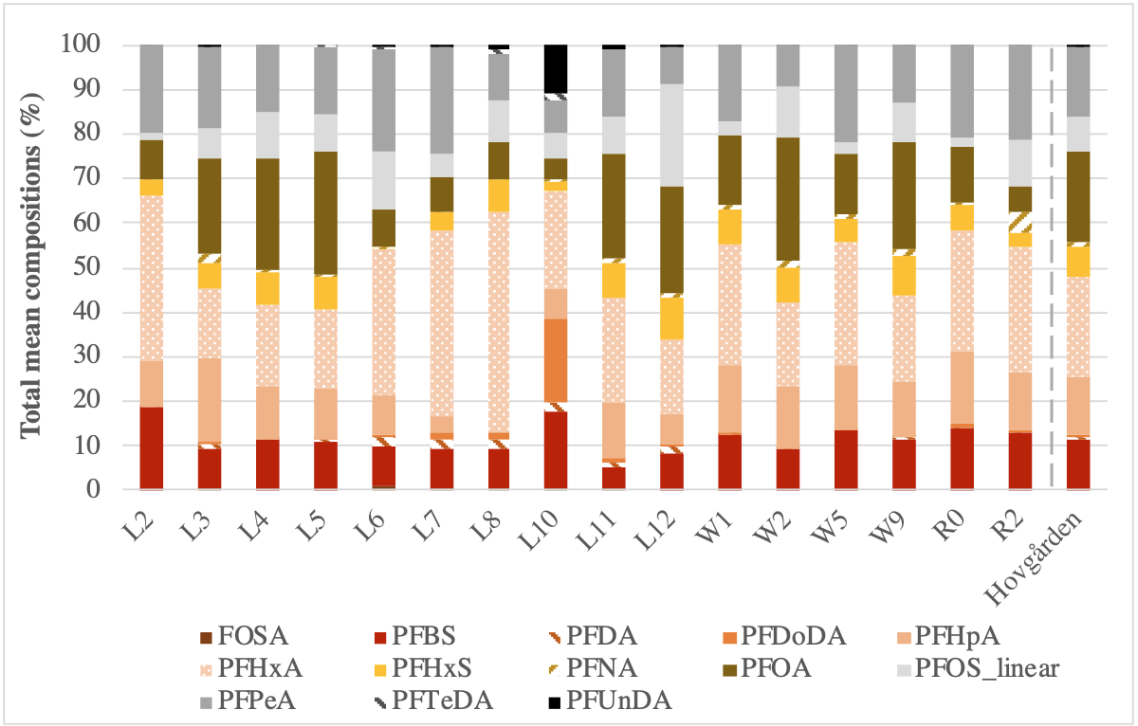

Figure A5.1: Total mean composition profile (%) in every water grab sampling point and at Hovgården, based on normalized mean concentrations of per- and polyfluoroalkyl substances (PFASs).

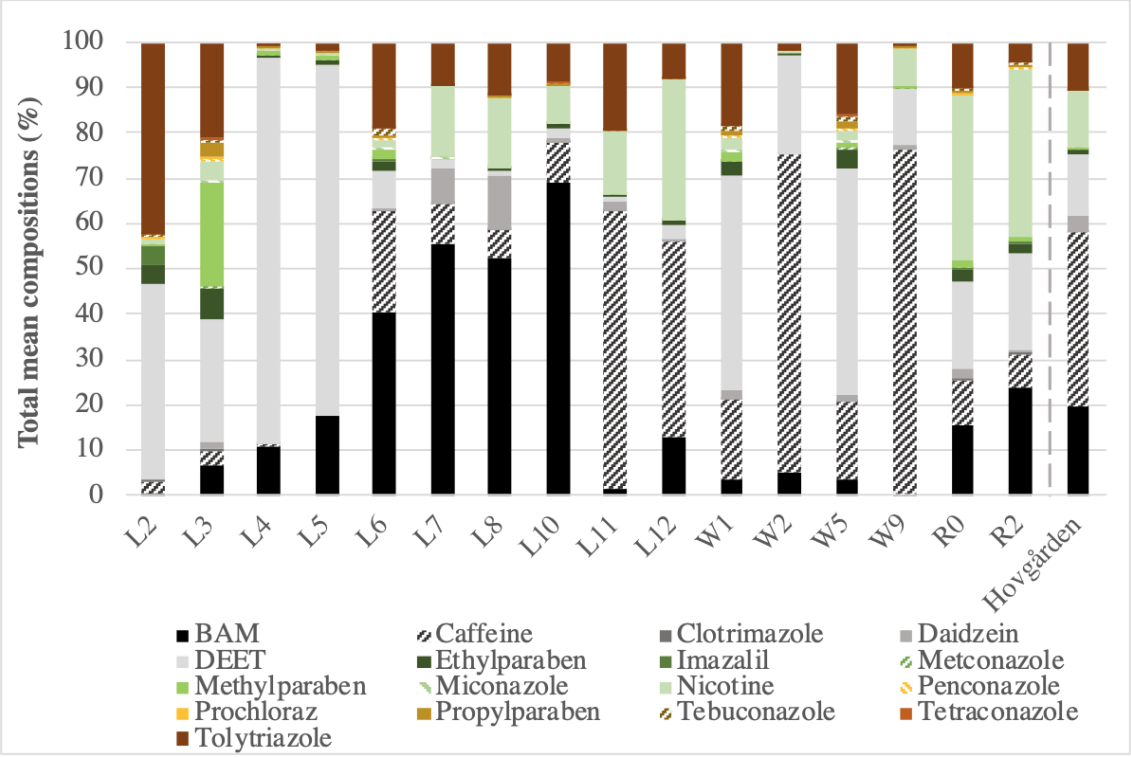

Figure A5.2: Total mean composition profile (%) in every water grab sampling point and at Hovgården, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

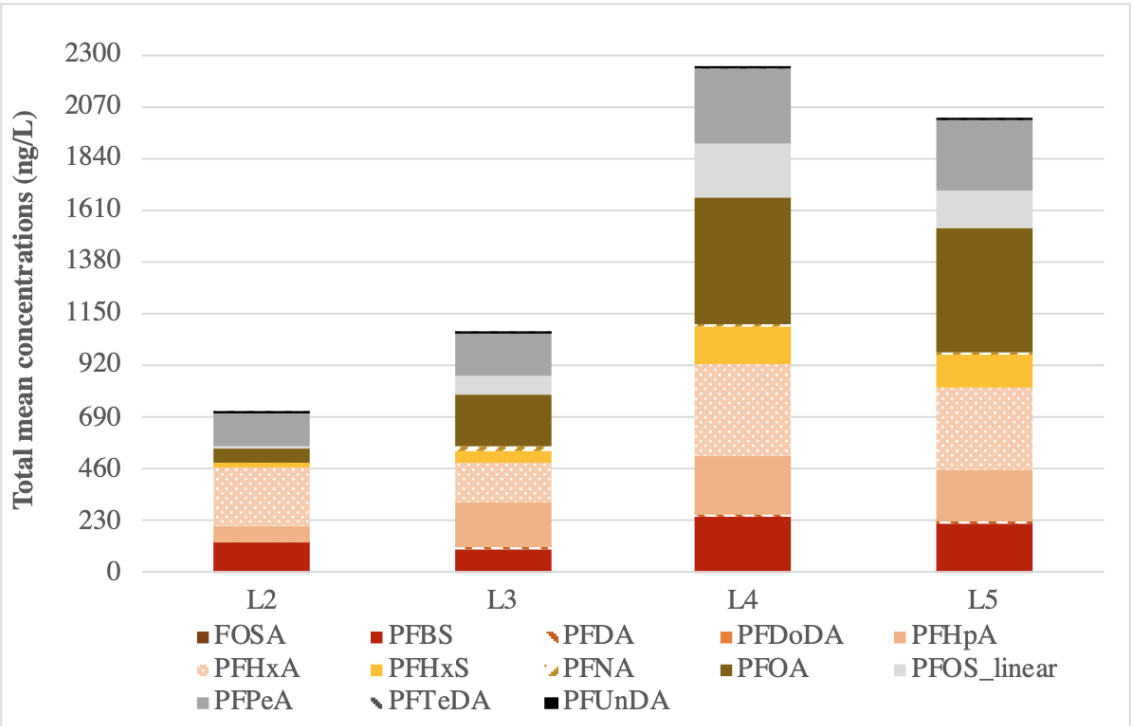

Figure A6: Total mean concentration (ng/L) of per- and polyfluoroalkyl substances (PFASs) in every water grab sampling point in stream 1.

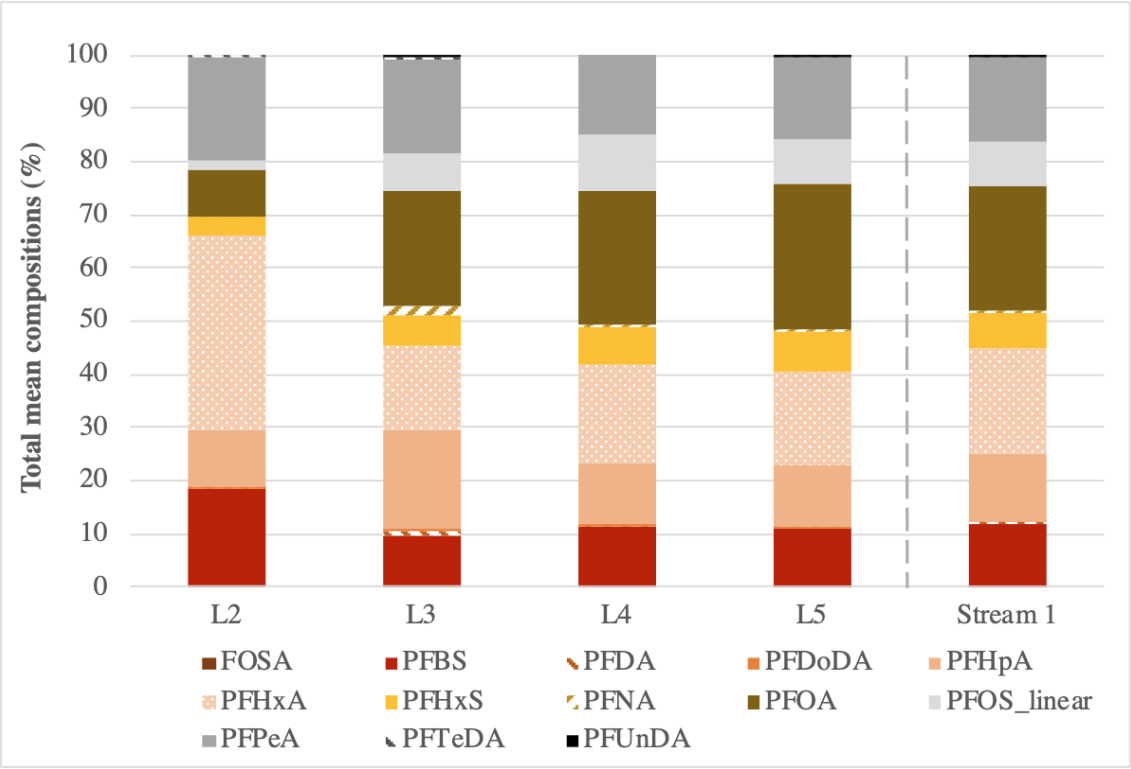

Figure A7: Total mean composition profile (%) in every water grab sampling point and in stream 1, based on normalized mean concentrations of per- and polyfluoroalkyl substances (PFASs).

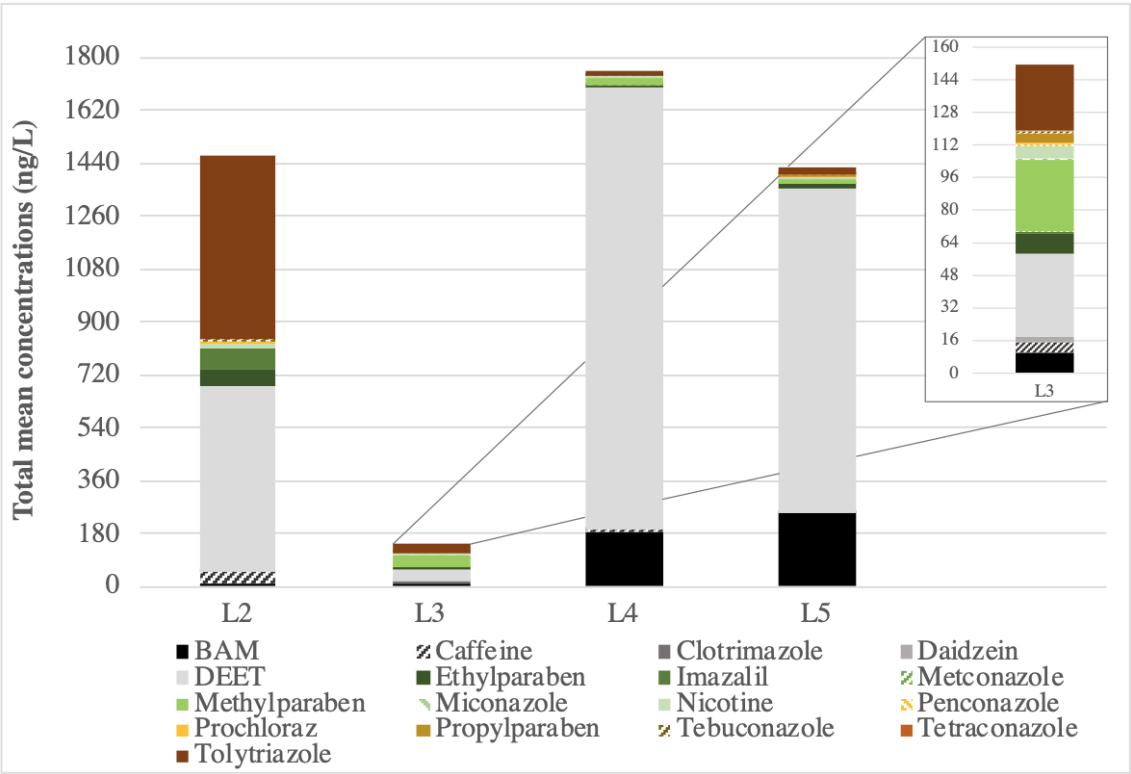

Figure A8: Total mean concentration (ng/L) of the remaining chemicals of emerging concern (CECs) in every water grab sampling point in stream 1.

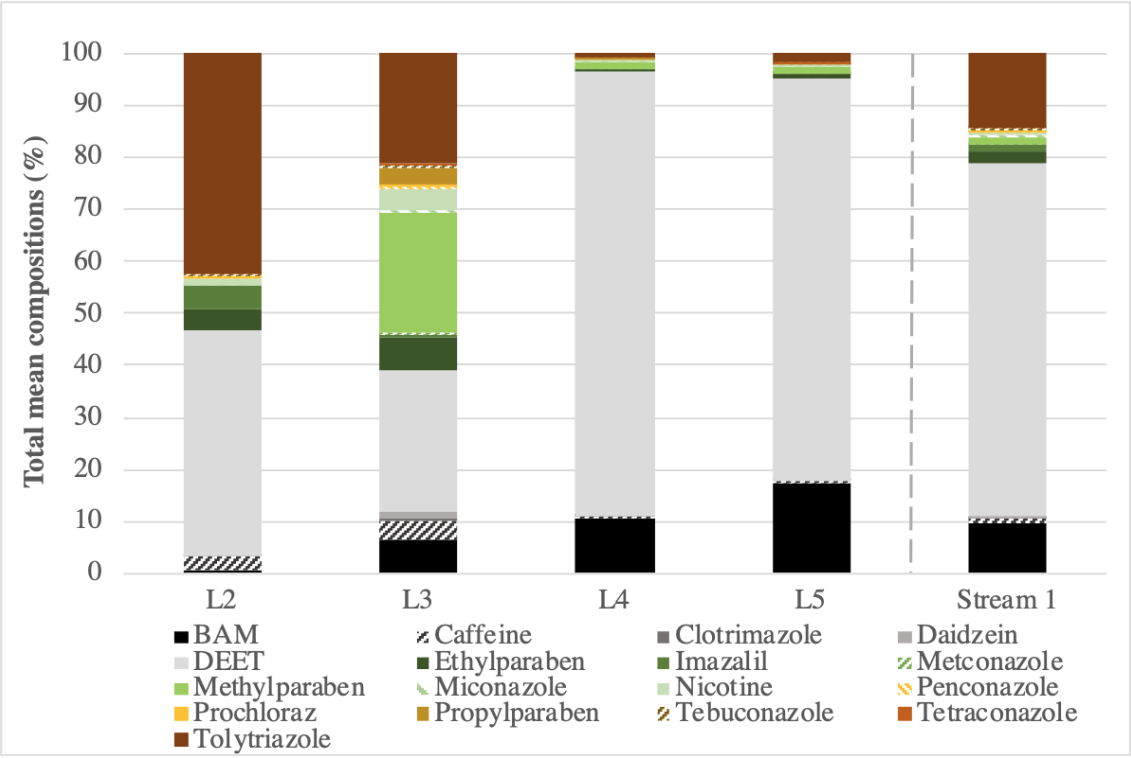

Figure A9: Total mean composition profile (%) in every water grab sampling point and in stream 1, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

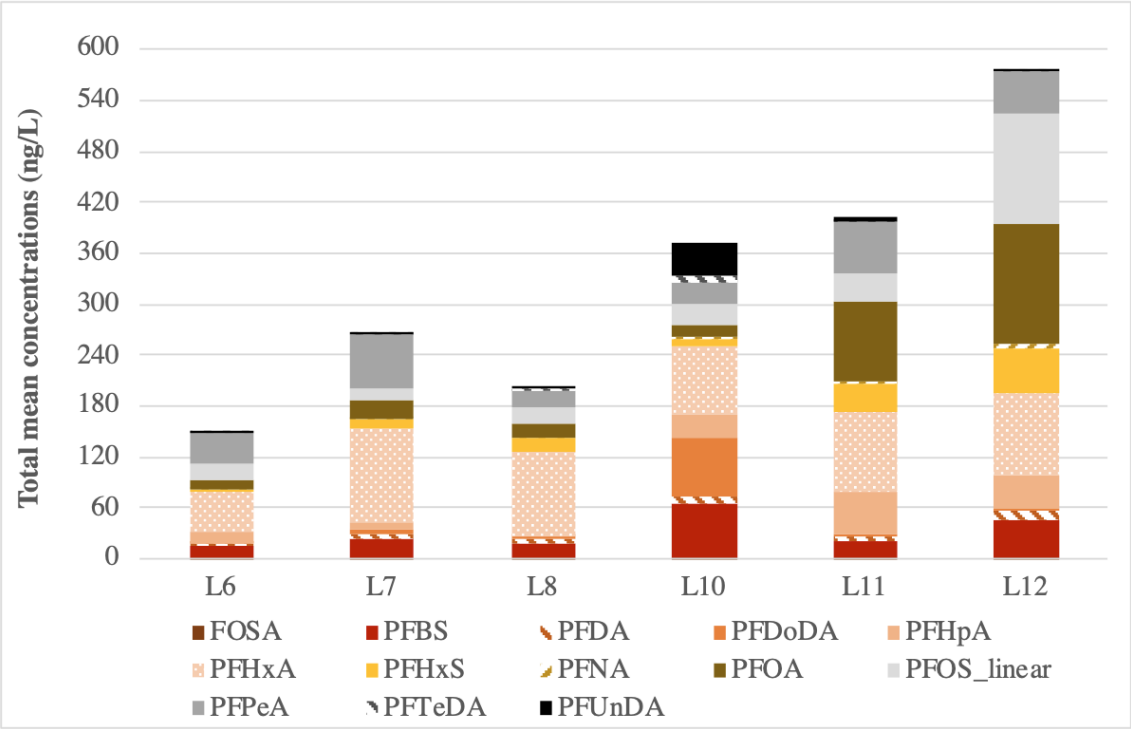

Figure A10: Total mean concentration (ng/L) of per- and polyfluoroalkyl substances (PFASs) in every water grab sampling point in stream 2.

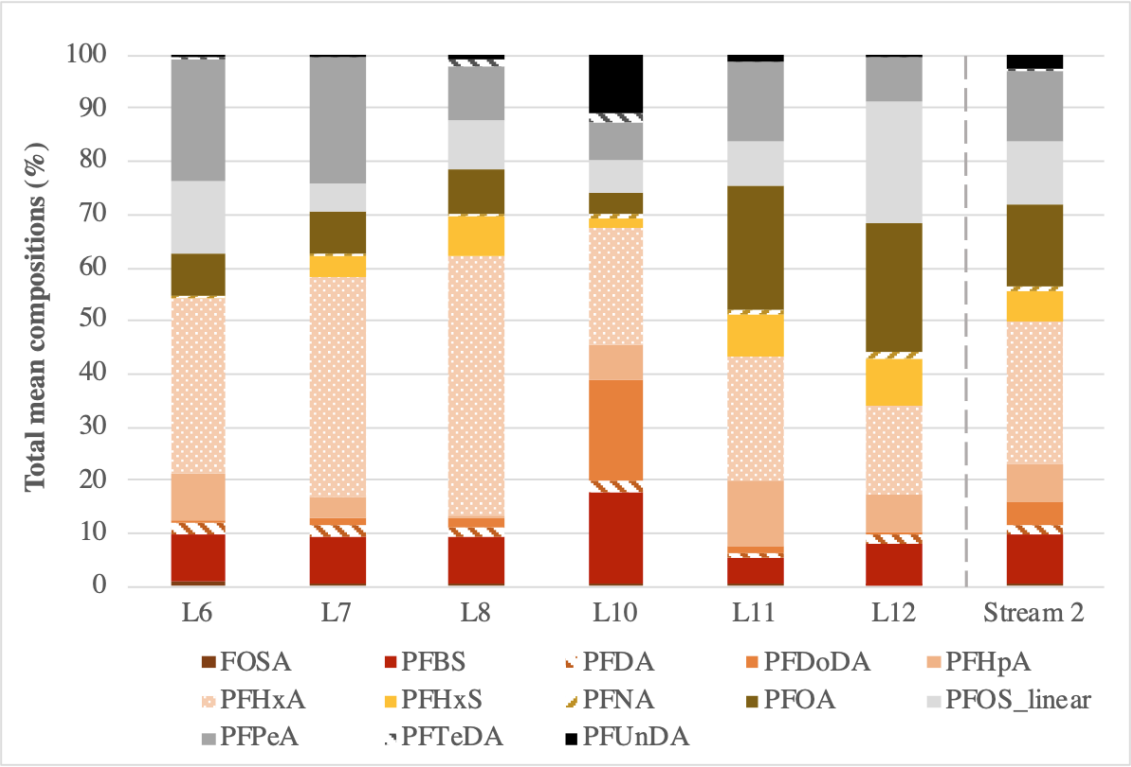

Figure A11: Total mean composition profile (%) in every water grab sampling point and in stream 2, based on normalized mean concentrations of per- and polyfluoroalkyl substances (PFASs).

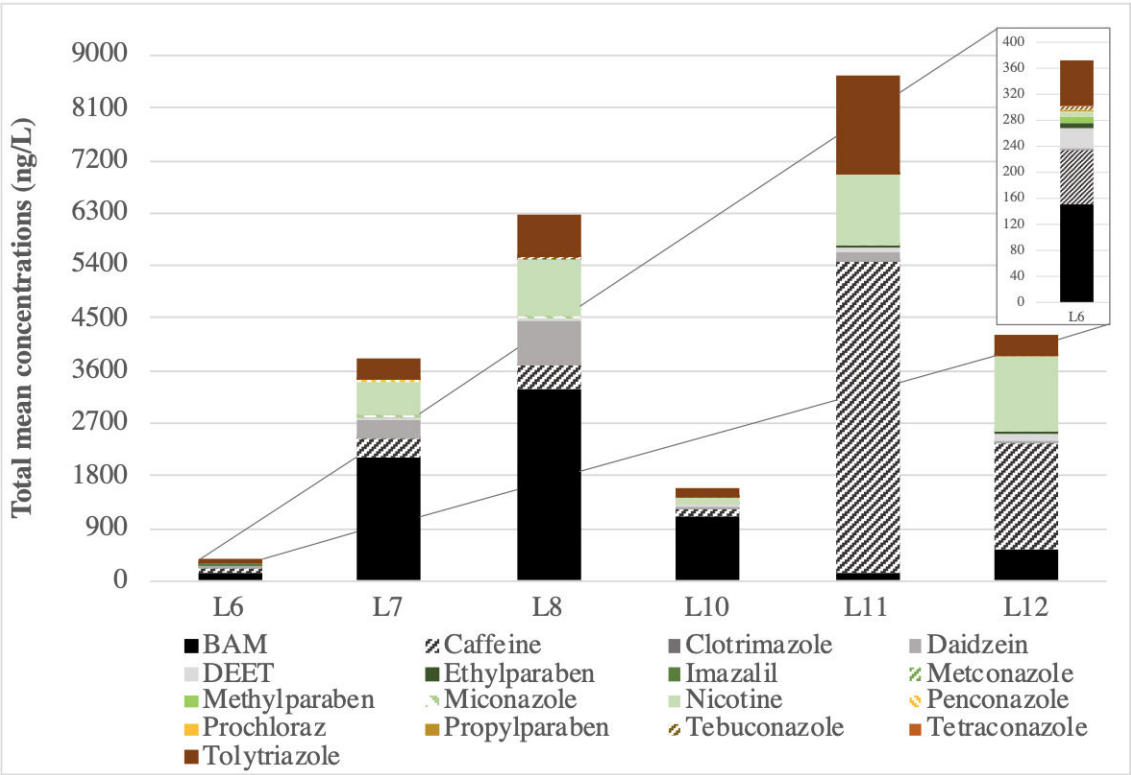

Figure A12: Total mean concentration (ng/L) of the remaining chemicals of emerging concern (CECs) in every water grab sampling point in stream 2.

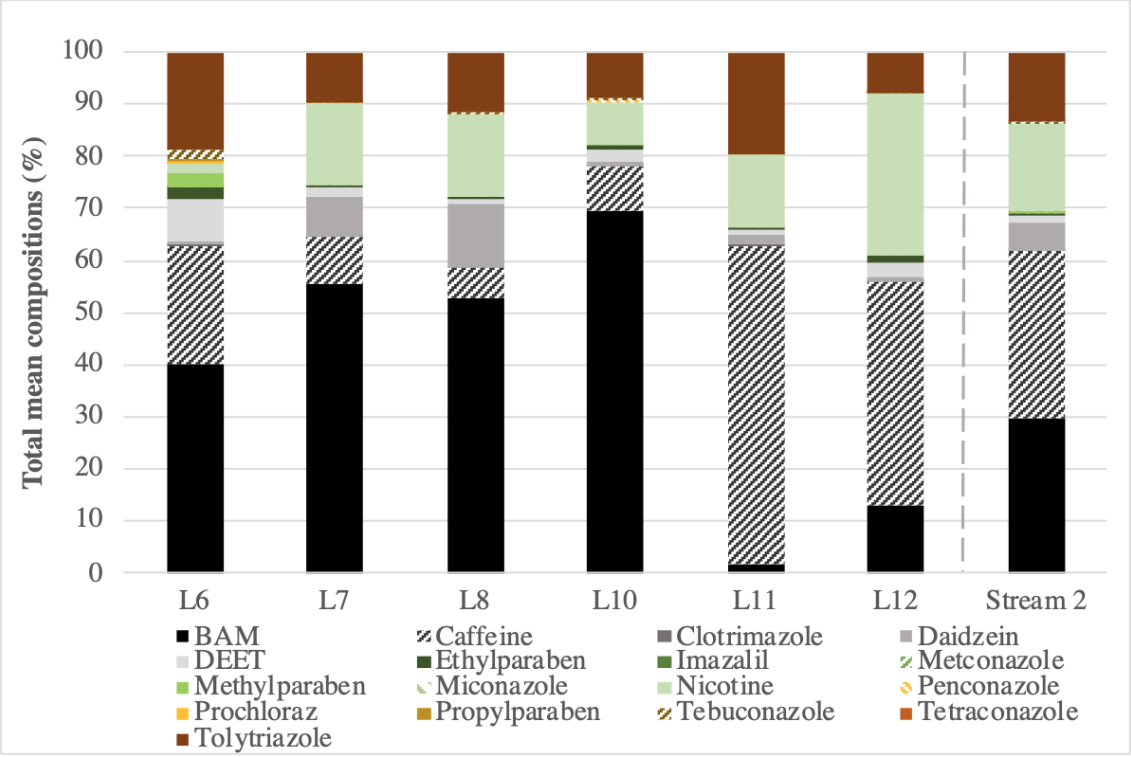

Figure A13: Total mean composition profile (%) in every water grab sampling point and in stream 2, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

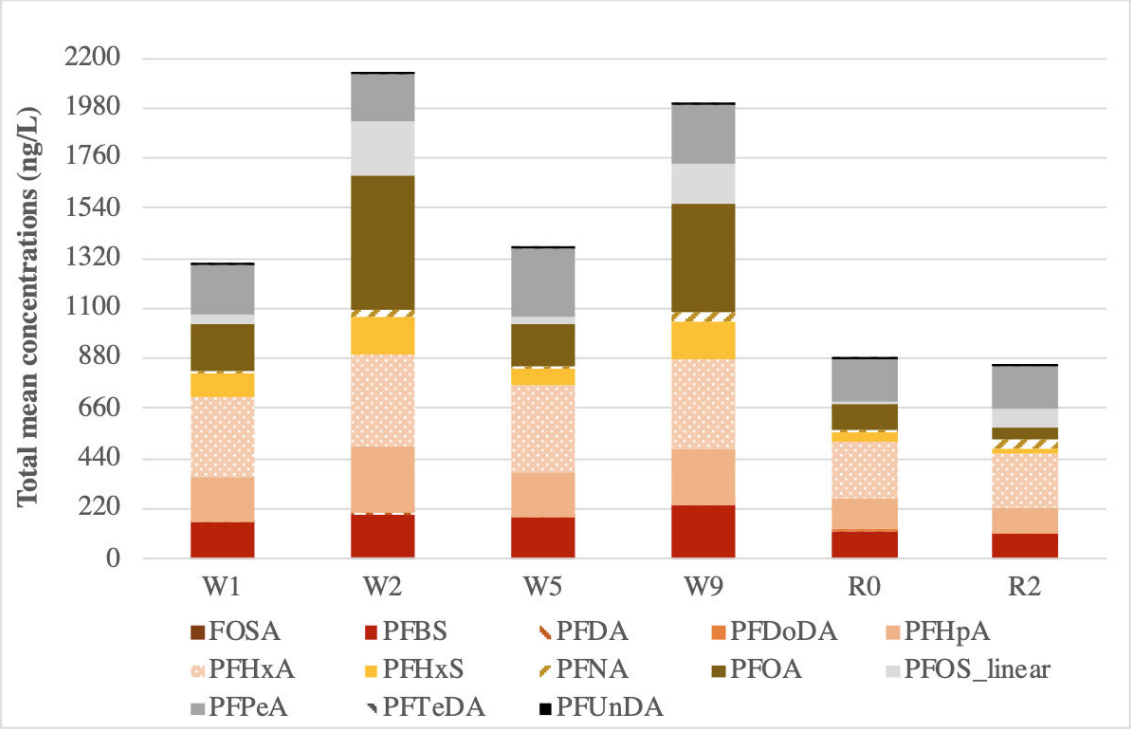

Figure A14: Total mean concentration (ng/L) of per- and polyfluoroalkyl substances (PFASs) in every water grab sampling point in the mixed stream and in the river.

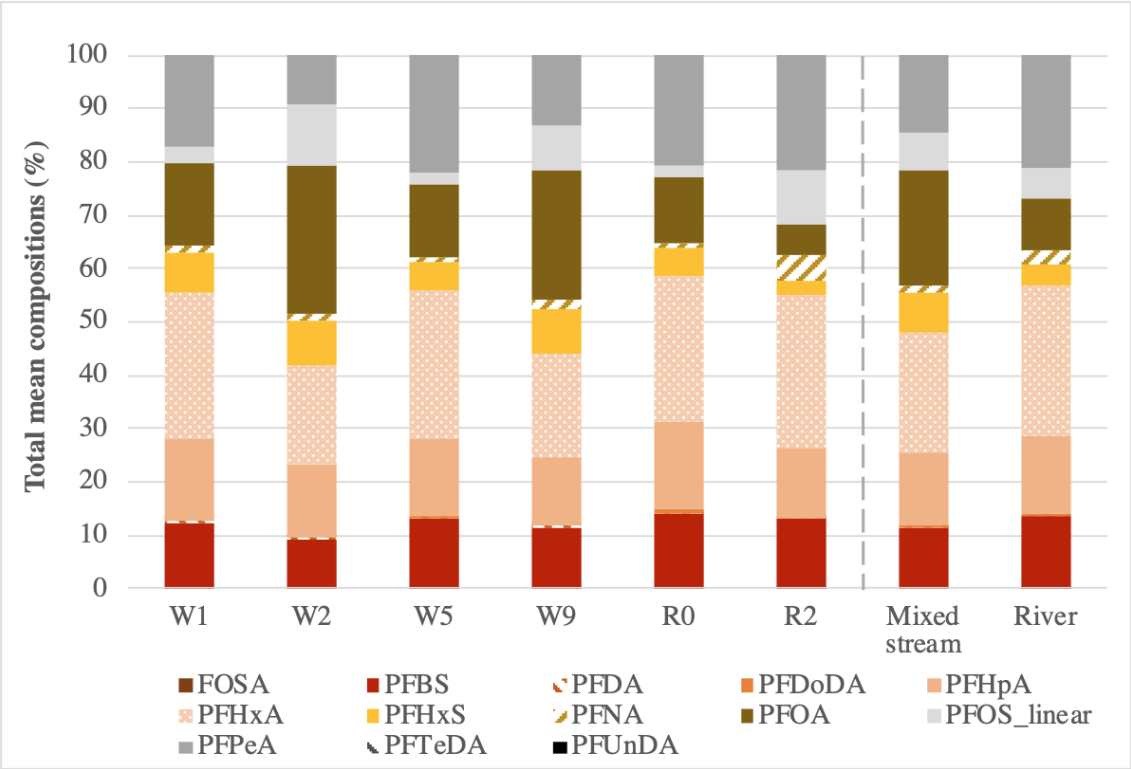

Figure A15: Total mean composition profile (%) in every water grab sampling point, in the mixed stream and in the river, based on normalized mean concentrations of per- and polyfluoroalkyl substances (PFASs).

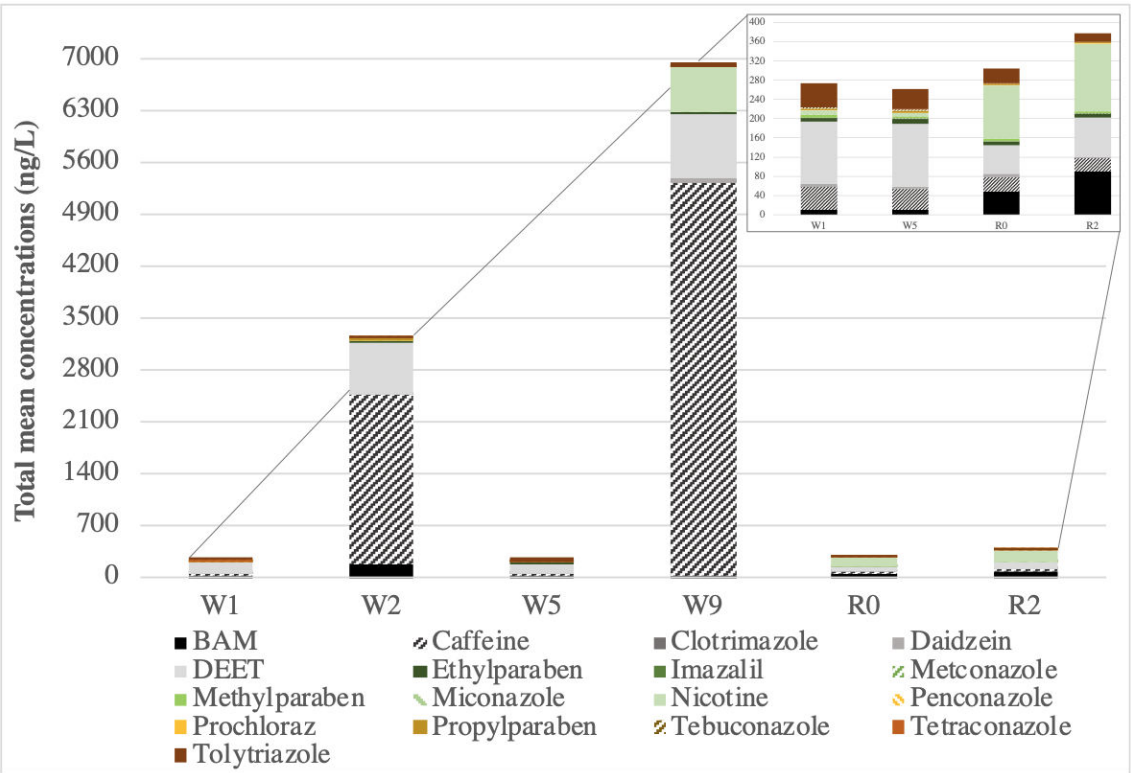

Figure A16: Total mean concentration (ng/L) of the remaining chemicals of emerging concern (CECs) in every water grab sampling point in the mixed stream and in the river.

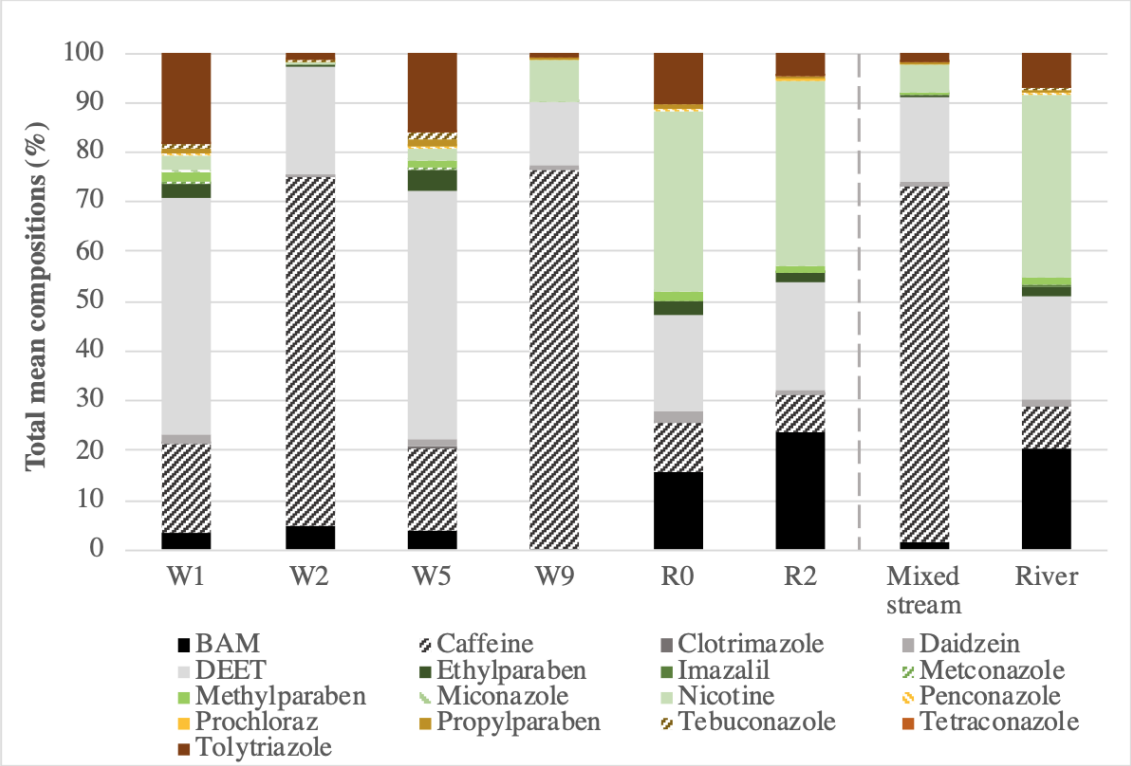

Figure A17: Total mean composition profile (%) in every water grab sampling point, in the mixed stream and in the river, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

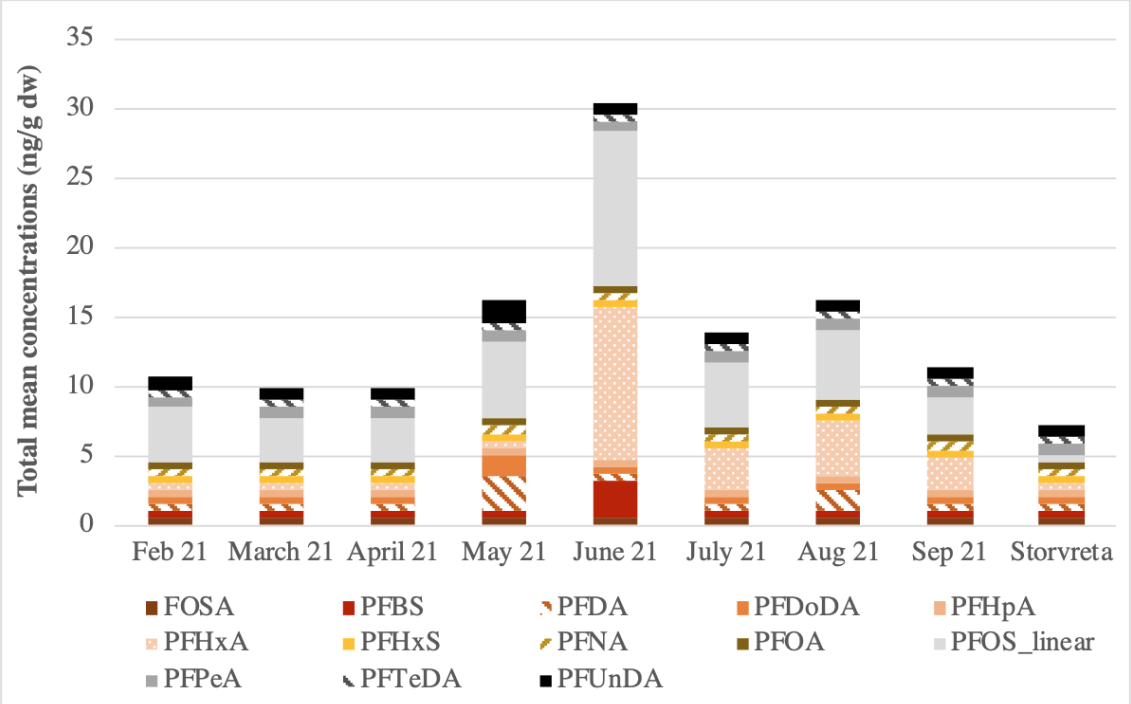

Figure A18: Total mean concentration (ng/g dw) of per- and polyfluoroalkyl substances (PFASs) in every sewage sludge grab sampling point in the sludge cell.

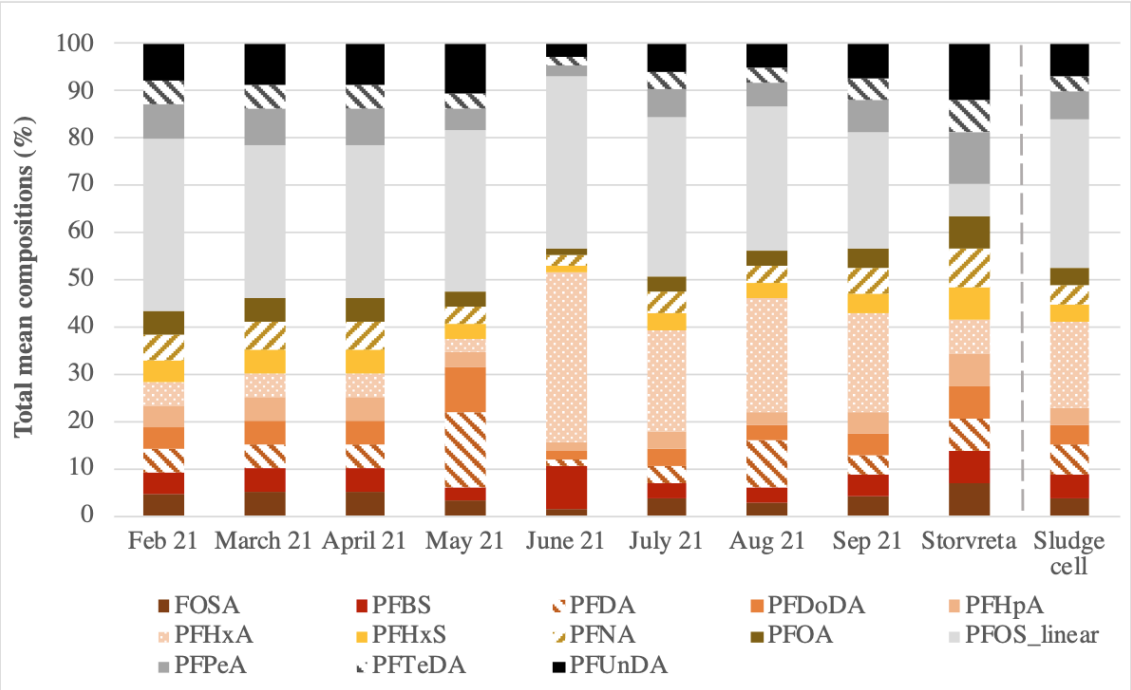

Figure A19: Total mean composition profile (%) in every sewage sludge grab sampling point and in the sludge cell, based on normalized mean concentrations of all per- and polyfluoroalkyl substances (PFASs).

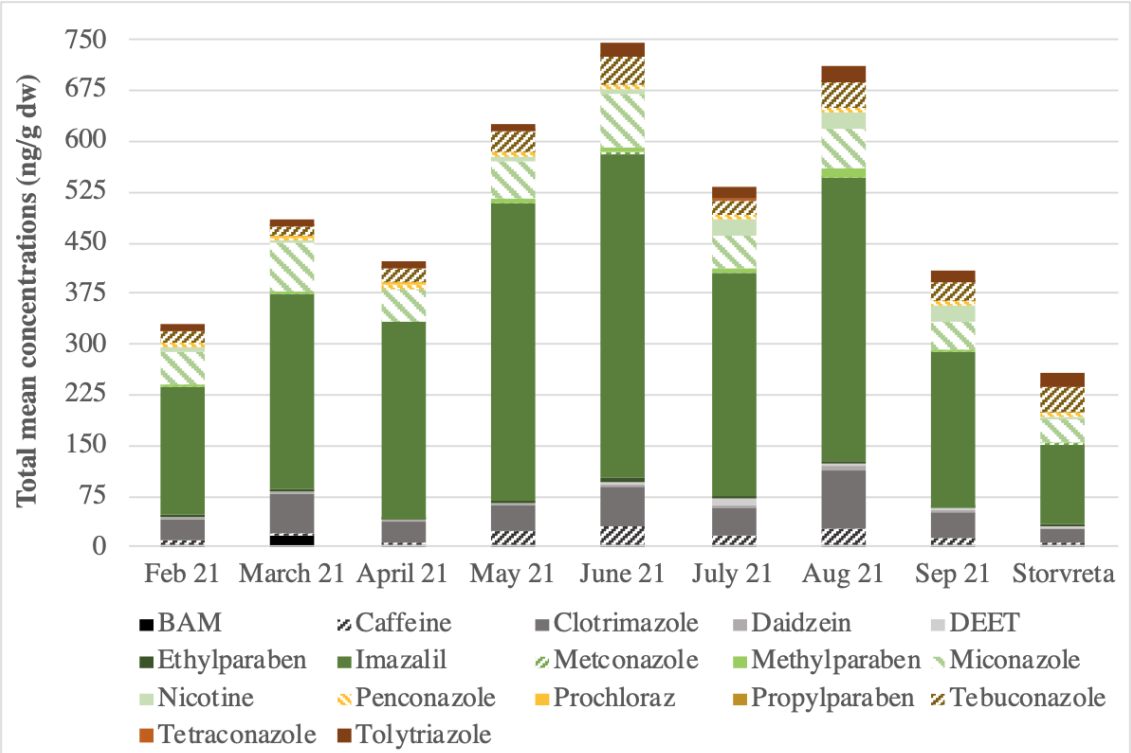

Figure A20: Total mean concentration (ng/g dw) of the remaining chemicals of emerging concern (CECs) in every sewage sludge grab sampling point in the sludge cell.

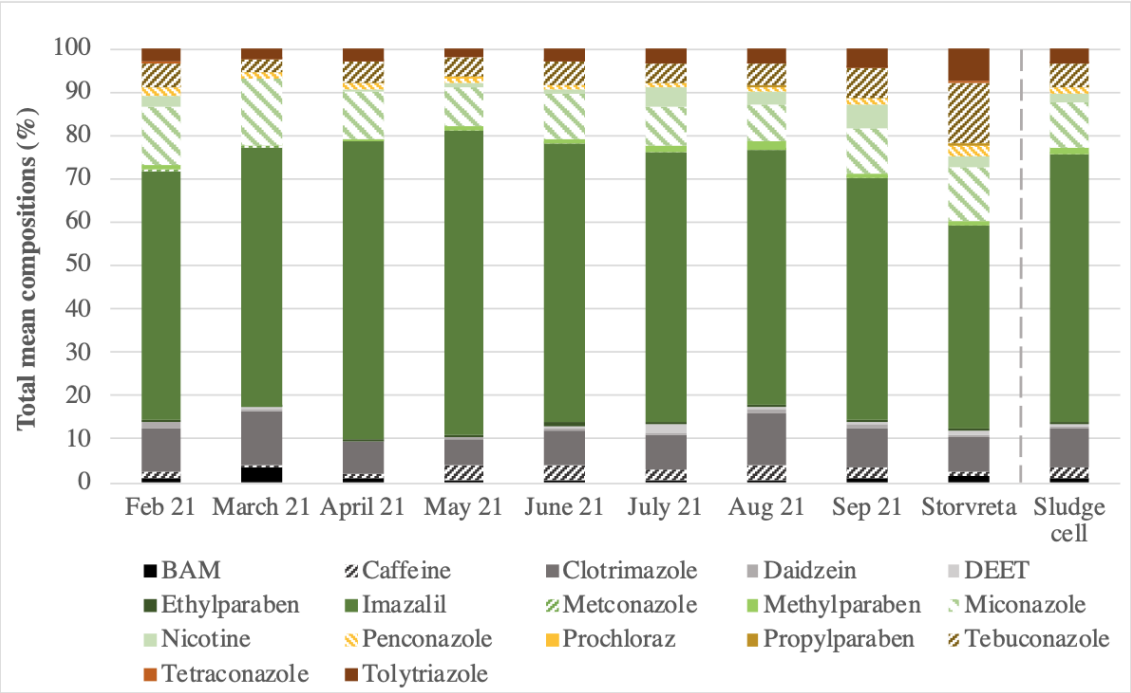

Figure A21: Total mean composition profile (%) in every sewage sludge grab sampling point and in the sludge cell, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

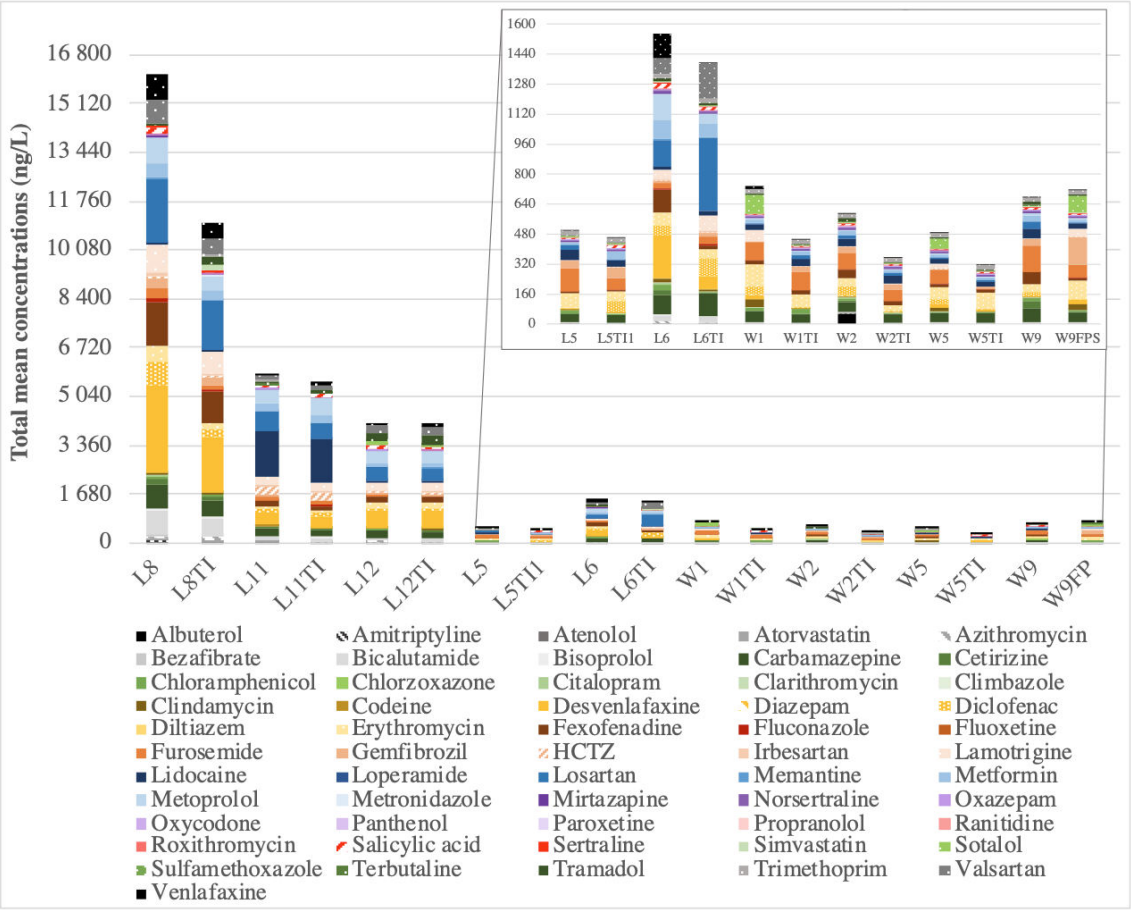

Figure A22: Total mean concentration (ng/L) of pharmaceuticals in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling (GS) point at Hovgården.

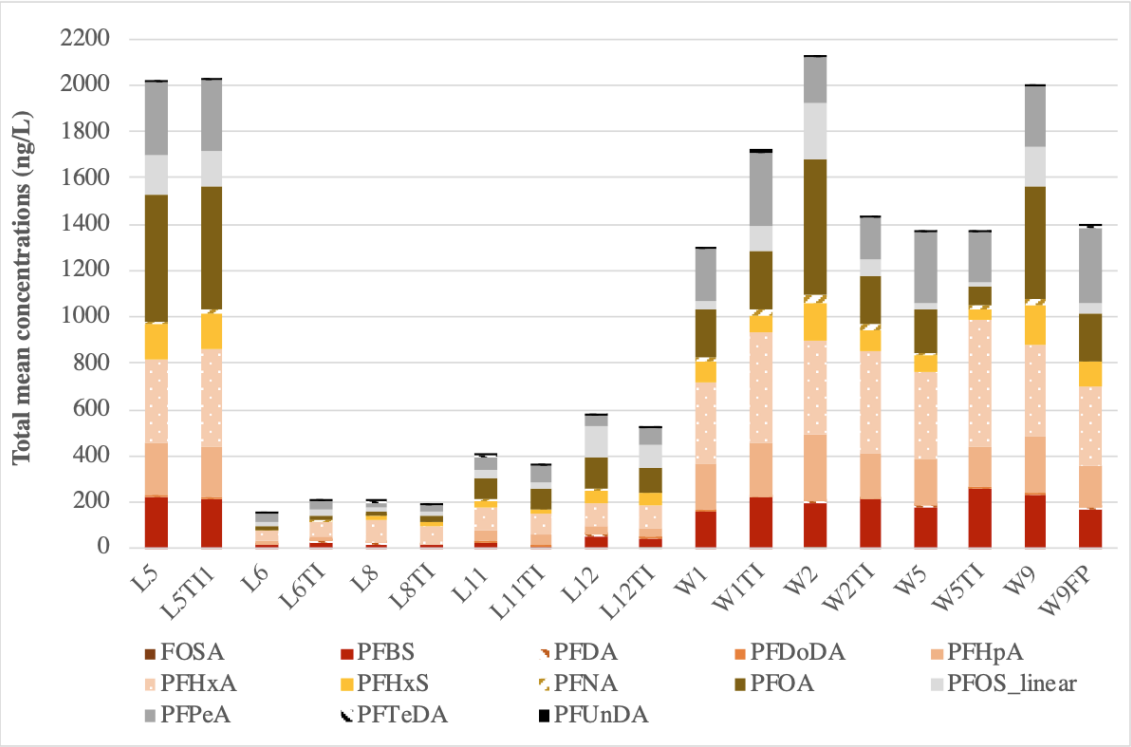

Figure A23.1: Total mean concentration (ng/L) of per- and polyfluoroalkyl substances (PFASs) in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling point at Hovgården.

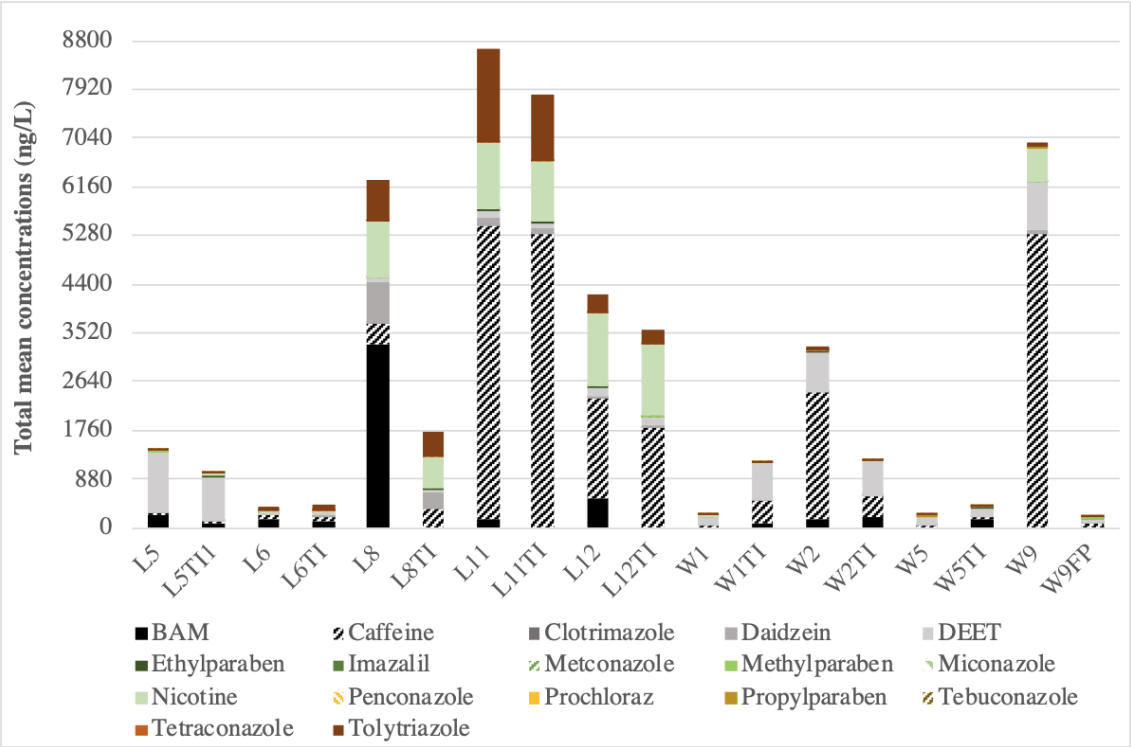

Figure A23.2: Total mean concentration (ng/L) of the remaining chemicals of emerging concern (CECs) in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling point at Hovgården.

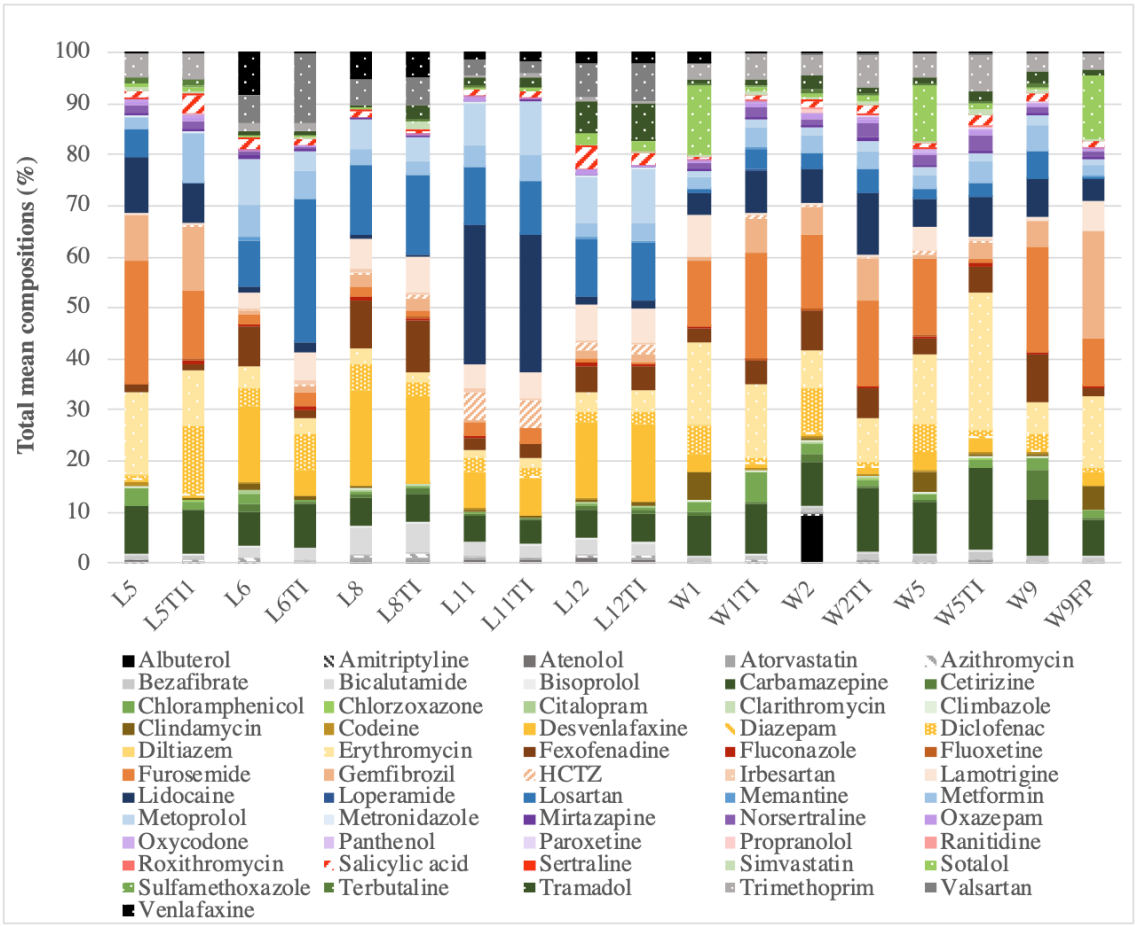

Figure A24: Total mean composition profile (%) in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling point at Hovgården, based on normalized mean concentrations of the pharmaceuticals.

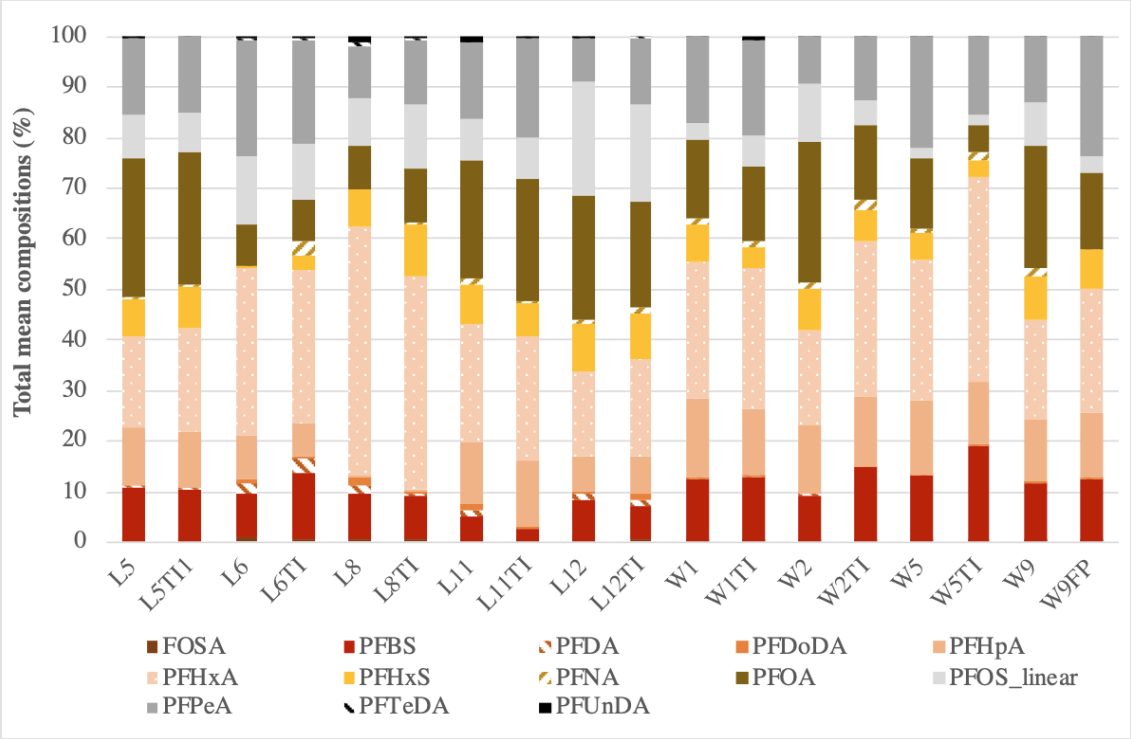

Figure A25.1: Total mean composition profile (%) in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling point at Hovgården, based on normalized mean concentrations of per- and polyfluoroalkyl substances (PFASs).

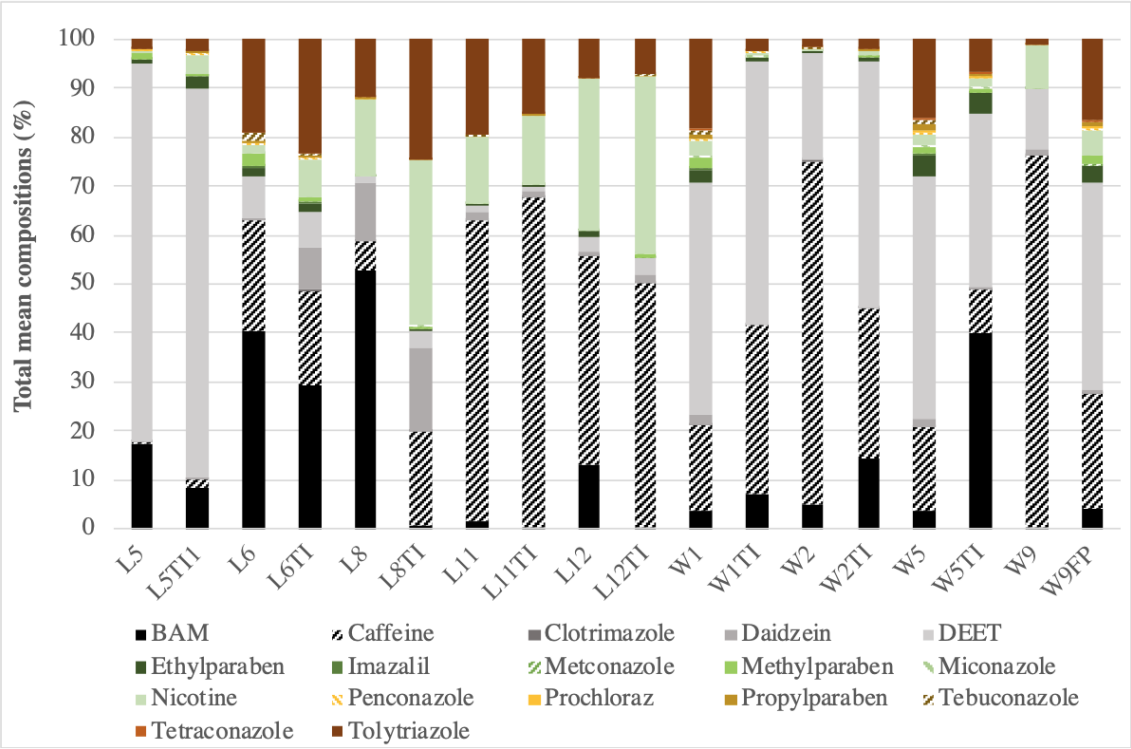

Figure A25.2: Total mean composition profile (%) in the time-integrated (TI) / flow-proportional (FP) sampling point(s) compared to their corresponding grab sampling point at Hovgården, based on normalized mean concentrations of the remaining chemicals of emerging concern (CECs).

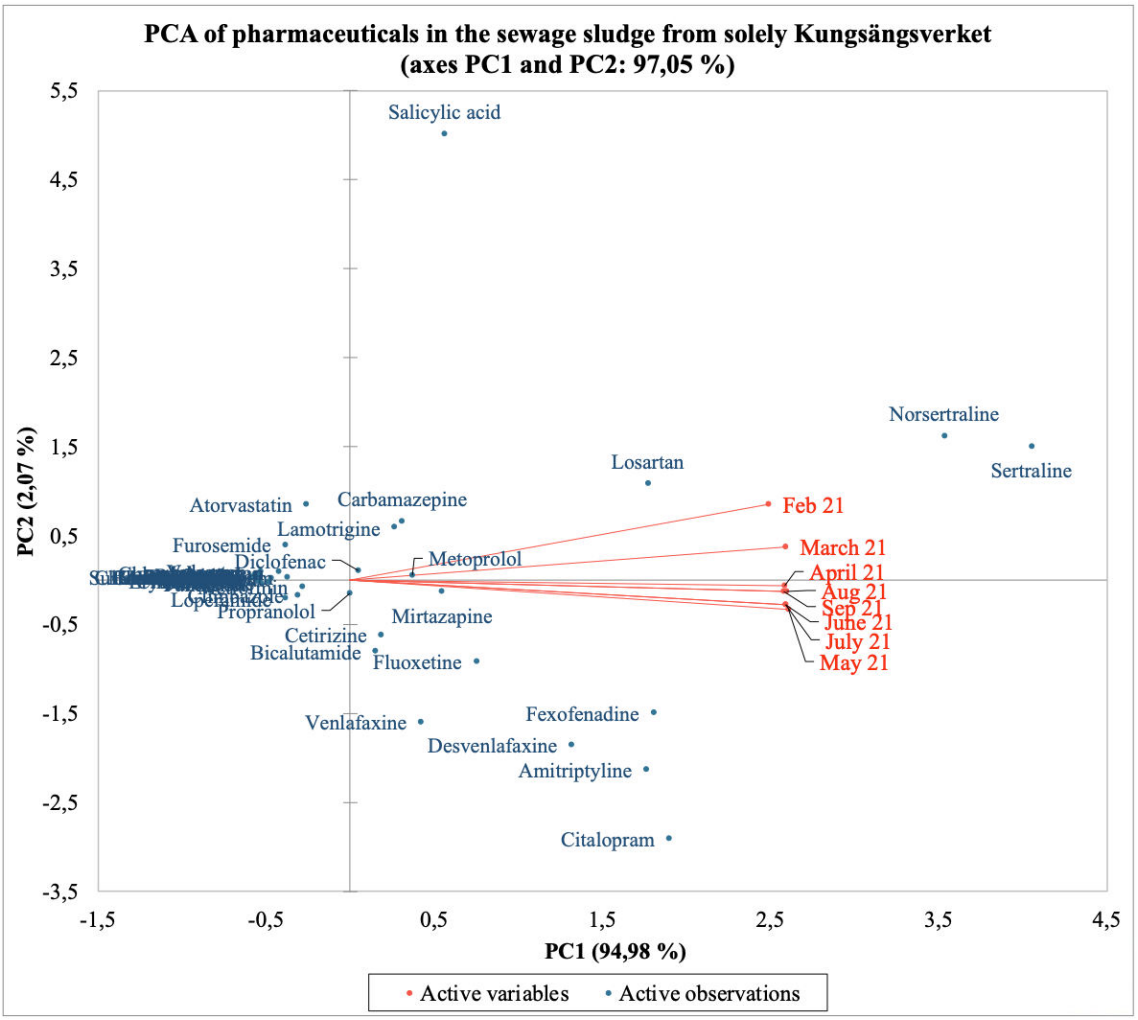

Figure A26: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

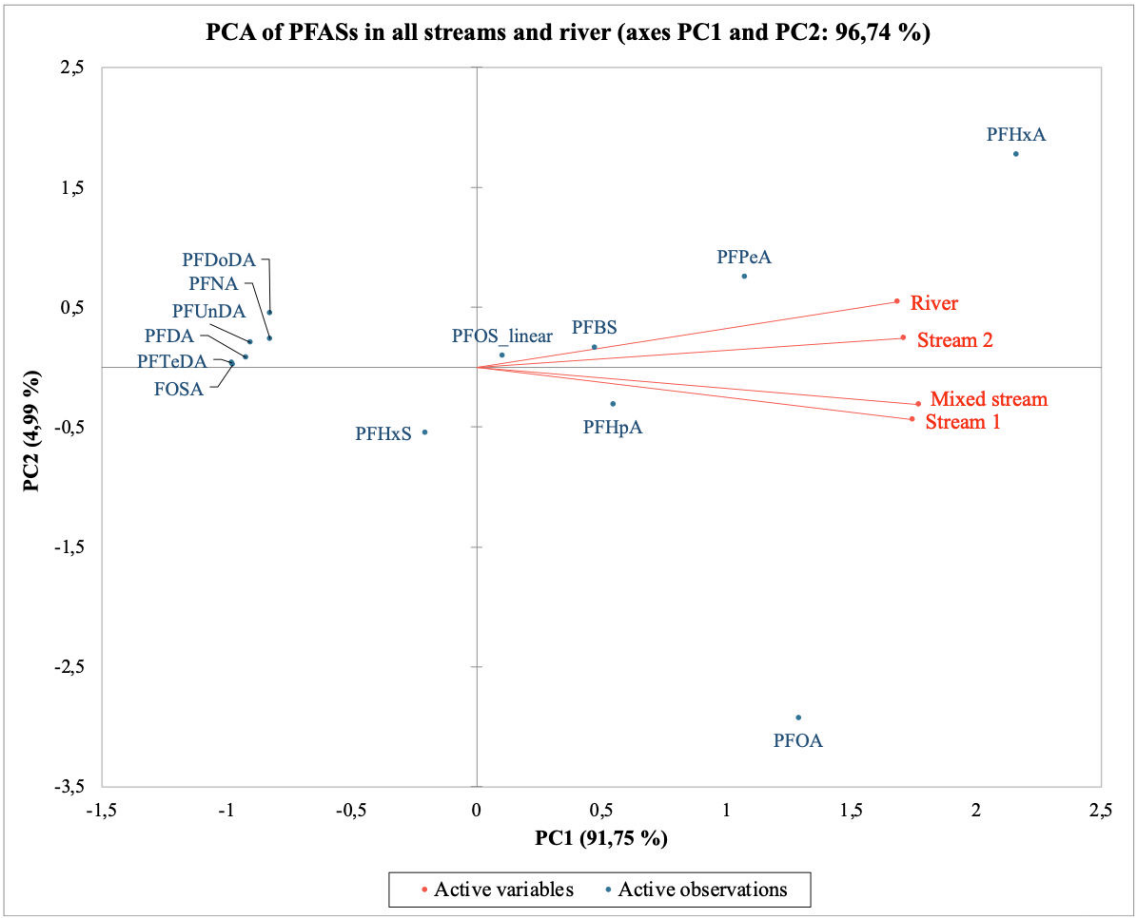

Figure A27: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

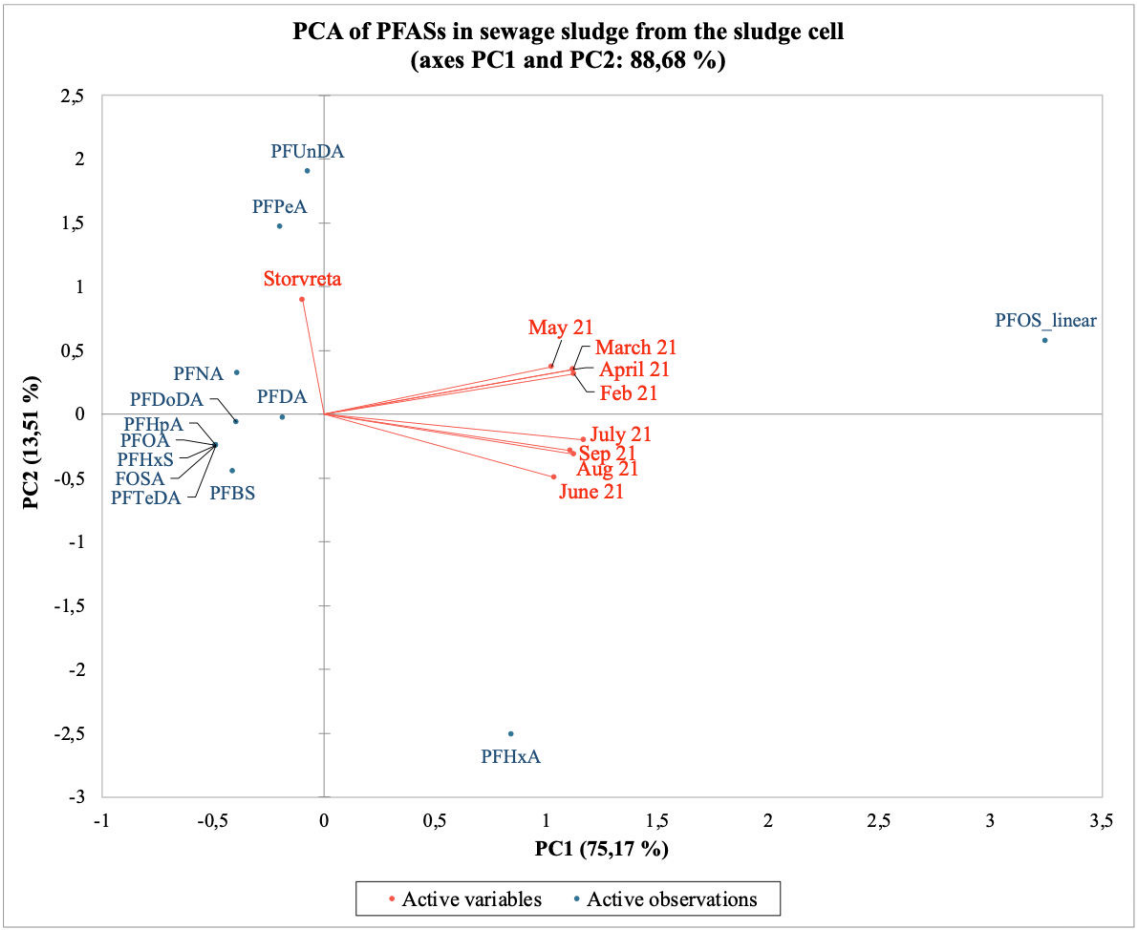

Figure A28: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

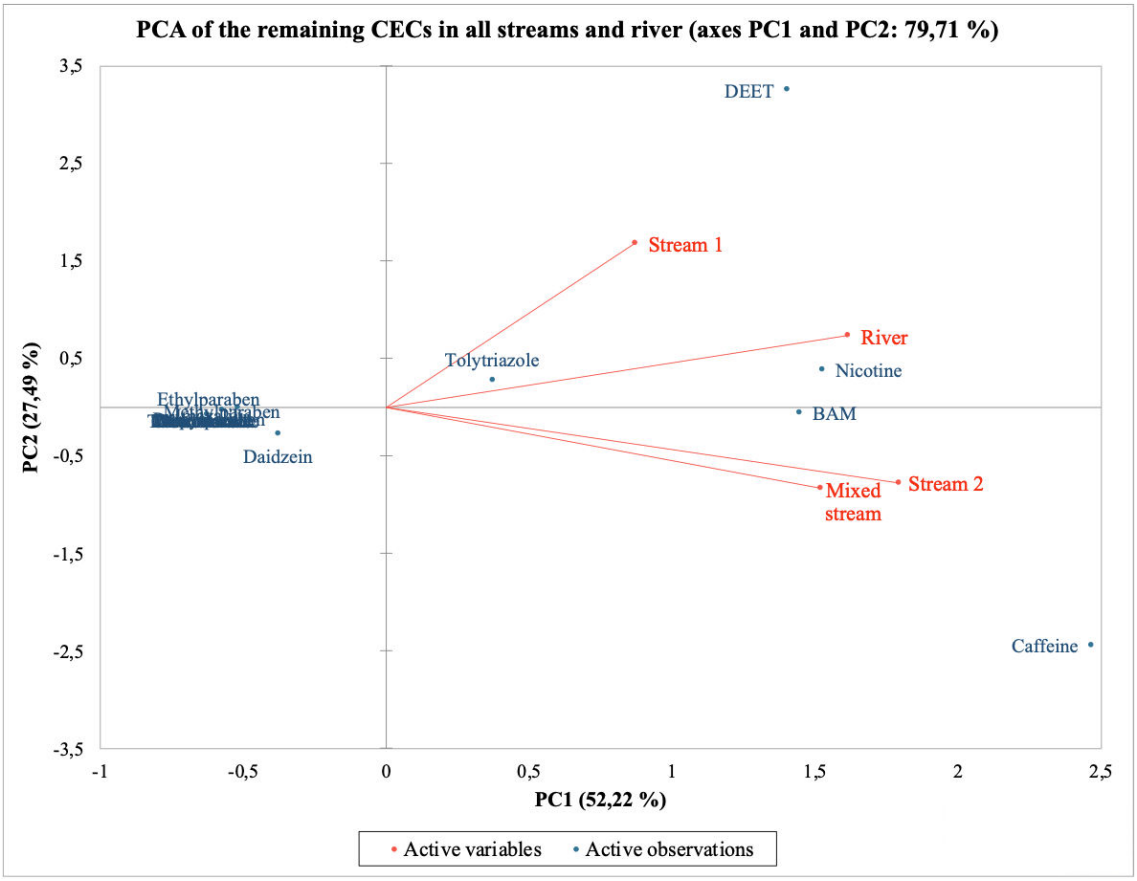

Figure A29: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

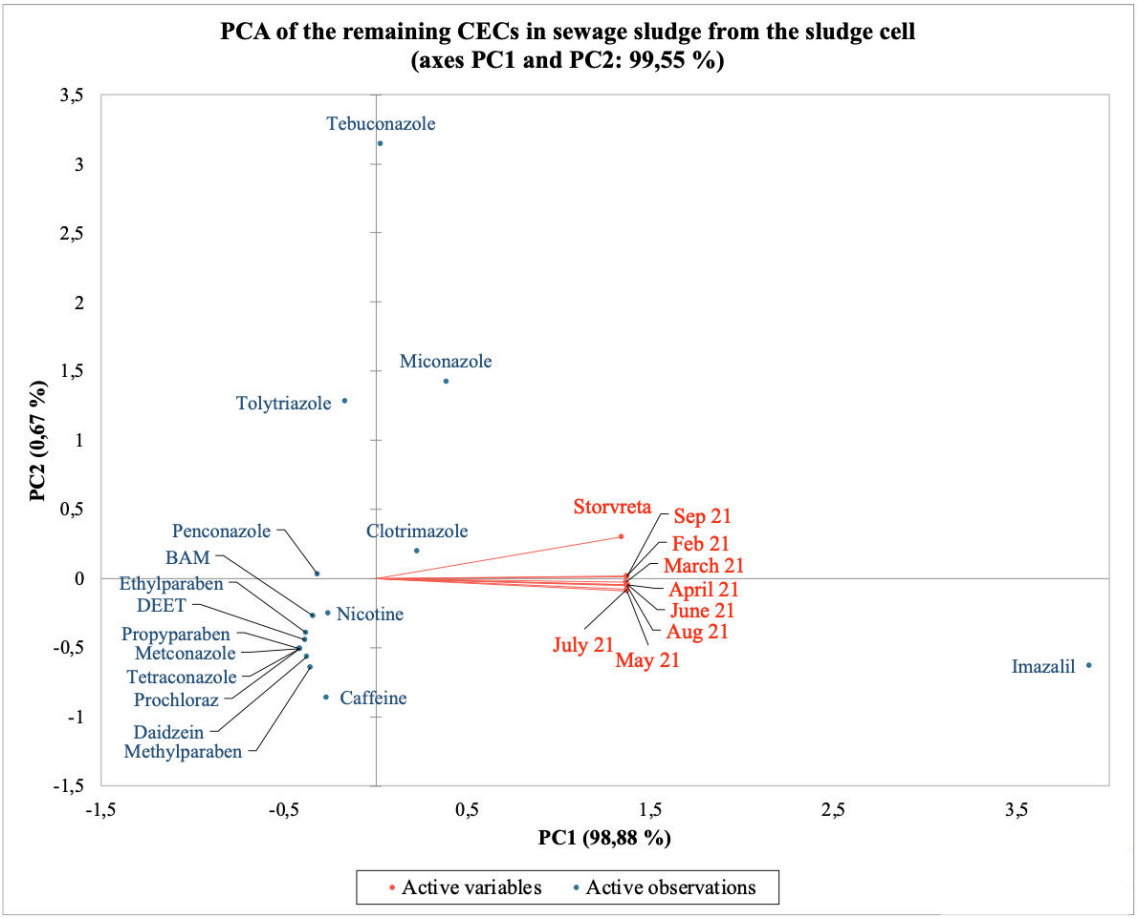

Figure A30: Biplot of observed data and dependent variables in terms of principal components PC1 (variance in mean composition) and PC2 (how frequent the difference is between the variables). PC1 and PC2 together explain the total variation (maximal amount of variance) in the observed data. The vectors (red lines) represent the correlation between the variables, and their loading (weight) on PC1 and/or PC2. A <, > or ≈ 90 ° angle between the vectors implies a positive, negative or no correlation.

TABLES

Table A1.1: Analysed pharmaceuticals. Sources: Chemspider and Pubchem.

Compound	Group / type	Molecular formula	CAS number
Memantine	Alzheimer	$C_{21}H_{21}N$	41100-52-1
Codeine	Analgesic	$C_{18}H_{21}NO_3$	76-57-3
Diclofenac	Analgesic, antipyretic	$C_{14}H_{11}Cl_2NO_2$	15307-86-5
Oxycodone	Analgesic	$C_{18}H_{21}NO_4$	76-42-6
Salicylic acid	Analgesic	$C_7H_6O_3$	69-72-7
Tramadol	Analgesic	$C_{16}H_{25}NO_2$	27203-92-5
Lidocaine	Anesthetic	$C_{14}H_{22}N_2O$	137-58-6
Azithromycin	Antibiotic	$C_{38}H_{72}N_2O_{12}$	83905-01-5
Chloramphenicol	Antibiotic	$C_{11}H_{12}Cl_2N_2O_5$	56-75-7
Clarithromycin	Antibiotic	$C_{38}H_{69}NO_{13}$	81103-11-9
Clindamycin	Antibiotic	$C_{18}H_{33}ClN_2O_5S$	21462-39-5
Erythromycin	Antibiotic	$C_{37}H_{67}NO_{13}$	114-07-8
Metronidazole	Antibiotic	$C_6H_9N_3O_3$	443-48-1
Roxithromycin	Antibiotic	$C_{41}H_{76}N_2O_{15}$	80214-83-1
Sulfamethoxazole	Antibiotic	$C_{10}H_{11}N_3O_3S$	723-46-6
Trimethoprim	Antibiotic	$C_{14}H_{18}N_4O_3$	738-70-5
Amitriptyline	Antidepressant	$C_{20}H_{23}N$	50-48-6
Citalopram	Antidepressant	$C_{20}H_{21}FN_2O$	59729-33-8
Desvenlafaxine	Antidepressent	$C_{16}H_{25}NO_2$	93413-62-8
Fluoxetine	Antidepressant	$C_{17}H_{18}F_3NO$	54910-89-3
Mirtazapine	Antidepressant	$C_{17}H_{19}N_3$	85650-52-8
Norsertraline	Antidepressant	$C_{16}H_{15}Cl_2N$	87857-41-8
Paroxetine	Antidepressant	$C_{19}H_{20}FNO_3$	61869-08-7
Sertraline	Antidepressant	$C_{17}H_{17}Cl_2N$	79617-96-2
Venlafaxine	Antidepressant	$C_{17}H_{27}NO_2$	93413-69-5
Metformin	Antidiabetic	$C_4H_{11}N_5$	657-24-9
Loperamide	Antidiarrhoeal	$C_{29}H_{33}ClN_2O_2$	53179-11-6
Carbamazepine	Antiepileptic	$C_{15}H_{12}N_2O$	298-46-4
Lamotrigine	Antiepileptic	$C_9H_7Cl_2N_5$	84057-84-1

Table A1.2: Analysed pharmaceuticals. Sources: Chemspider and Pubchem.

Compound	Group / type	Molecular formula	CAS number
Climbazole	Antifungal	$C_{15}H_{17}ClN_2O_2$	38083-17-9
Fluconazole	Antifungal	$C_{13}H_{12}F_2N_6O$	86386-73-4
Cetirizine	Antihistamine	$C_{21}H_{25}ClN_2O_3$	83881-51-0
Fexofenadine	Antihistamine	$C_{32}H_{39}NO_4$	153439-40-8
Atenolol	Antihypertensive, antiarrhytmic	$C_{14}H_{22}N_2O_3$	29122-68-7
Bisoprolol	Antihypertensive, antiarrhytmic	$C_{18}H_{31}NO_4$	6722-44-9
Diltiazem	Antihypertensive	$C_{22}H_{26}N_2O_4S$	42399-41-7
Furosemide	Antihypertensive	$C_{12}H_{11}ClN_2O_5S$	54-31-9
HCTZ (hydrochlorothiazide)	Antihypertensive	$C_7H_8ClN_3O_4S_2$	58-93-5
Irbesartan	Antihypertensive	$C_{25}H_{28}N_6O$	138402-11-6
Losartan	Antihypertensive	$C_{22}H_{23}ClN_6O$	114798-26-4
Metoprolol	Antihypertensive, antiarrhytmic	$C_{15}H_{25}NO_3$	51384-51-1
Propranolol	Antihypertensive, antitremorous	$C_{16}H_{21}NO_2$	525-66-6
Sotalol	Antihypertensive, antiarrhytmic	$C_{12}H_{20}N_2O_3S$	3930-20-9
Valsartan	Antihypertensive	$C_{24}H_{29}N_5O_3$	137862-53-4
Atorvastatin	Antilipemic	$C_{33}H_{35}FN_2O_5$	134523-00-5
Bezafibrate	Antilipemic	$C_{19}H_{20}ClNO_4$	41859-67-0
Gemfibrozil	Antilipemic	$C_{15}H_{22}O_3$	25812-30-0
Simvastatin	Antilipemic	$C_{25}H_{38}O_5$	79902-63-9
Bicalutamide	Antineoplastic agent	$C_{18}H_{14}F4N_2O_4S$	90357-06-5
Ranitidine	Antisecretory	$C_{13}H_{22}N_4O_3S$	66357-35-5
Albuterol (salbutamol)	Bronchal dilator (beta-adrenergic agonist)	$C_{13}H_{21}NO_3$	18559-94-9
Terbutaline	Bronchal dilator (beta-adrenergic agonist)	$C_{12}H_{19}NO_3$	23031-25-6
Panthenol	Moisturizer	$C_9H_{19}NO_4$	16485-10-2
Chlorzoxazone	Muscle relaxant	$C_7H_4ClNO_2$	95-25-0
Diazepam	Sedative	$C_{16}H_{13}ClN_2O$	439-14-5
Oxazepam	Sedative	$C_{15}H_{11}ClN_2O_2$	604-75-1

Table A2: Other analysed chemicals of emerging concern (CECs). PFAS = per- and polyfluoroalkyl substance. Sources: Chemspider and Pubchem.

Compound	Group / type	Molecular formula	CAS number
Tolytriazole	Industrial chemical	$C_7H_7N_3$	29878-31-7
Daidzein	Isoflavone	$C_{15}H_{10}O_4$	486-66-8
Methylparaben	Paraben	$C_8H_8O_3$	99-76-3
Ethylparaben	Paraben	$C_9H_{10}O_3$	120-47-8
Propylparaben	Paraben	$C_{10}H_{12}O_3$	94-13-3
BAM (dichlorobenzamide)	Pesticide	$C_7H_5Cl_2NO$	2008-58-4
Clotrimazole	Pesticide	$C_{22}H_{17}ClN_2$	23593-75-1
DEET (N,N-diethyl-m-toluamide)	Pesticide	$C_{12}H_{17}NO$	134-62-3
Imazalil	Pesticide	$C_{14}H_{14}Cl_2N_2O$	35554-44-0
Metconazole	Pesticide	$C_{17}H_{22}ClN_3O$	125116-23-6
Miconazole	Pesticide	$C_{18}H_{14}Cl_4N_2O$	22916-47-8
Penconazole	Pesticide	$C_{13}H_{15}Cl_2N_3$	66246-88-6
Prochloraz	Pesticide	$C_{15}H_{16}Cl_3N_3O_2$	67747-09-5
Tebuconazole	Pesticide	$C_{16}H_{22}ClN_3O$	107534-96-3
Tetraconazole	Pesticide	$C_{13}H_{11}Cl_2F_4N_3O$	112281-77-3
FOSA (perfluorooctane sulfonamide)	PFAS	$C_8H_2F_{17}NO_2S$	754-91-6
PFBS (perfluorobutanesulfonic acid)	PFAS	$C_4HF_9O_3S$	375-73-5
PFDA (perfluorodecanoic acid)	PFAS	$C_{10}HF_{19}O_2$	335-76-2
PFDoDA (perfluorododecanoic acid)	PFAS	$C_{12}HF_{23}O_2$	307-55-1
PFHpA (perfluoroheptanoic acid)	PFAS	$C_7HF_{13}O_2$	375-85-9
PFHxA (perfluorohexanoic acid)	PFAS	$C_6HF_{11}O_2$	307-24-4
PFHxS (perfluorohexanesulfonic acid)	PFAS	$C_6HF_{13}O_3S$	355-46-4
PFNA (perfluorononanoic acid)	PFAS	$C_9HF_{17}O_2$	375-95-1
PFOA (perfluorooctanoic acid)	PFAS	$C_8HF_{15}O_2$	335-67-1
PFOS_linear (perfluorooctanesulfonic acid)	PFAS	$C_8HF_{17}O_3S$	1763-23-1
PFPeA (perfluoropentanoic acid)	PFAS	$C_5HF_9O_2$	2706-90-3
PFTeDA (perfluorotetradecanoic acid)	PFAS	$C_{14}HF_{27}O_2$	376-06-7
PFUnDA (perfluoroundecanoic acid)	PFAS	$C_{11}HF_{21}O_2$	2058-94-8
Caffeine	Stimulant	$C_8H_{10}N_4O_2$	1958-08-02
Nicotine	Stimulant	$C_{10}H_{14}N_2$	1954-11-05

Table A3: Average relative recovery (ARR), matrix effect, limit of quantification (LOQ) and LOQ/2 of the analysed pharmaceuticals.

Pharmaceutical	Sludge samples				Water samples			
	ARR (%)	Matrix effect (%)	LOQ (ng/g dw)	LOQ/2 (ng/g dw)	ARR (%)	Matrix effect (%)	LOQ (ng/L)	LOQ/2 (ng/L)
Albuterol	140	-33	1.7	0.85	120	-130	1	0.5
Amitriptyline	150	-10	1	0.5	160	-110	1	0.5
Atenolol	130	-83	1	0.5	100	-220	1	0.5
Atorvastatin	67	-140	1	0.5	54	-130	1	0.5
Azithromycin	72	-2.5	1	0.5	180	-79	1.1	0.55
Bezafibrate	130	-200	1	0.5	110	-180	8.9	4.5
Bicalutamide	100	-10	1	0.5	160	-23	1	0.5
Bisoprolol	120	-69	1	0.5	110	-120	1	0.5
Carbamazepine	130	-110	1	0.5	110	-120	1	0.5
Cetirizine	120	-99	1	0.5	110	-170	1	0.5
Chloramphenicol	140	-48	1	0.5	100	68	9	4.5
Chlorzoxazone	86	-7.1	4.3	2.2	130	-170	1.1	0.55
Citalopram	110	-33	1	0.5	99	-150	1	0.5
Clarithromycin	120	-67	1	0.5	120	-110	1	0.5
Climbazole	99	-89	1	0.5	120	-64	1	0.5
Clindamycin	99	-20	1	0.5	130	-21	1	0.5
Codeine	120	-110	1	0.5	86	67	2.3	1.2
Desvenlafaxine	140	-86	1	0.5	170	-270	3.5	1.8
Diazepam	110	-97	1	0.5	120	-120	1	0.5
Diclofenac	44	-36	4.9	2.5	160	-51	6.7	3.4
Diltiazem	130	-100	1	0.5	120	-160	1	0.5
Erythromycin	270	-69	2.8	1.4	240	-410	59	30
Fexofenadine	110	-2.9	1	0.5	140	-100	1	0.5
Fluconazole	140	-270	1	0.5	110	58	1	0.5
Fluoxetine	120	-34	1	0.5	120	100	1	0.5
Furosemide	140	-140	5.6	2.8	140	-240	5.7	2.9
Gemfibrozil	140	-13	4.6	2.3	190	-32	7.9	4
HCTZ	160	-61	2.4	1.2	140	-170	1.9	0.95
Irbesartan	130	-98	0.96	0.48	120	-110	1.1	0.55
Lamotrigine	110	-140	1	0.5	120	-480	2.5	1.3
Lidocaine	140	-43	1	0.5	89	76	1	0.5
Loperamide	90	-64	1	0.5	83	-56	1	0.5
Losartan	130	1	1	0.5	130	-71	1.4	0.7
Memantine	89	-70	1	0.5	120	-150	1	0.5
Metformin	90	-100	24	12	16	-170	2.4	1.2
Metoprolol	150	-100	3.8	1.9	120	-160	1	0.5
Metronidazole	130	-13	1	0.5	180	-340	1	0.5
Mirtazapine	82	-61	1	0.5	100	-81	1	0.5
Norsertraline	130	-83	20	10	97	-87	20	10
Oxazepam	120	-89	1	0.5	110	-48	6.1	3.1
Oxycodone	77	-73	1	0.5	110	-240	1	0.5
Panthenol	78	-460	3.6	1.8	8.4	-170	2.5	1.3
Paroxetine	95	-120	1	0.5	67	-88	1	0.5
Propranolol	94	-73	1	0.5	110	-90	1	0.5
Ranitidine	59	-160	2.7	1.4	95	-320	1.1	0.55
Roxithromycin	210	-53	1	0.5	170	-110	1.1	0.55
Salicylic acid	94	9	2	1	14	1.4	2.9	1.5
Sertraline	110	25	1	0.5	110	-110	1.5	0.75
Simvastatin	110	-41	2.1	1.1	55	-43	5.8	2.9
Sotalol	160	11	2.6	1.3	110	-140	7.3	3.7
Sulfamethoxazole	210	-100	1	0.5	210	-150	1.6	0.8
Terbutaline	170	9.7	2.2	1.1	170	-86	1	0.5
Tramadol	120	-150	1	0.5	97	69	1	0.5
Trimethoprim	210	-8.5	1	0.5	230	-500	45	23
Valsartan	140	-170	1	0.5	100	71	1.9	0.95
Venlafaxine	120	-54	1	0.5	130	-140	1	0.5

Table A4: Average relative recovery (ARR), matrix effect, limit of quantification (LOQ) and LOQ/2 of the other analysed chemicals of emerging concern (CECs).

Other CECs	Sludge samples				Water samples			
	ARR (%)	Matrix effect (%)	LOQ (ng/g dw)	LOQ/2 (ng/g dw)	ARR (%)	Matrix effect (%)	LOQ (ng/L)	LOQ/2 (ng/L)
BAM	150	-96	7.3	3.7	160	-110	20	10
Caffeine	130	-110	1.8	0.9	93	41	2	1
Clotrimazole	60	-43	1	0.5	60	-63	1	0.5
Daidzein	120	-260	1.8	0.9	150	-550	4.1	2.1
DEET	130	-86	1	0.5	120	-39	1.1	0.55
Ethylparaben	160	-87	4.4	2.2	110	-190	15	7.5
Imazalil	60	49	1.4	0.7	60	0.079	1.1	0.53
Metconazole	60	-74	1	0.5	60	-62	1	0.5
Methylparaben	150	-120	3	1.5	130	-310	7.4	3.7
Miconazole	60	-45	1	0.5	0.72	-48	1	0.5
Nicotine	88	-330	1	0.5	120	28	1	0.5
Penconazole	60	2	13	6.5	60	-0.24	2.2	1.1
Prochloraz	60	-97	1	0.5	60	-38	1	0.5
Propylparaben	150	-110	1	0.5	110	-120	4.2	2.1
Tebuconazole	60	-73	2.8	1.4	7.3	-57	1.4	0.7
Tetraconazole	60	-99	1	0.5	60	-57	1	0.5
Tolytriazole	110	1.9	1.6	0.8	150	-170	8.2	4.1
FOSA	90	-60	1	0.5	85	-79	2.6	1.3
PFBS	78	-47	1	0.5	90	-33	1	0.5
PFDA	82	-86	1	0.5	71	-110	1	0.5
PFDoDA	100	-80	1	0.5	75	-57	1.7	0.85
PFHpA	92	-150	1	0.5	79	-180	1	0.5
PFHxA	76	-59	1	0.5	68	-59	4.2	2.1
PFHxS	38	-23	1	0.5	100	-44	1	0.5
PFNA	64	-14	1.2	0.6	69	-50	1	0.5
PFOA	62	-42	1	0.5	73	-66	2.6	1.3
PFOS_linear	96	-140	1	0.5	76	-110	1	0.5
PFPeA	64	-76	1.6	0.8	54	-230	6.1	3.1
PFTeDA	58	-29	1	0.5	17	-83	1	0.5
PFUnDA	94	-70	1.7	0.85	80	-85	1.4	0.7

Table A5: ∑mean concentrations, mean concentrations, means, medians and standard deviations (STDs) (bolded ones are the 10 most significant in each area) (ng/L) of pharmaceuticals in all water grab sampling points within Hovgården's facility with belonging detection frequencies. If the concentration of the pharmaceutical was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A pharmaceutical was not detected if its mean concentration was ≤ LOQ (red box).

Pharmaceutical	**L2**	**L3**	**L4**	**L5**	**STD**	**L6**	**L7**	**L8**	**L10**	**L11**	**L12**	**STD**	**W1**	**W2**	**W5**	**W9**	**R0**	**R2**	**STD**
Albuterol	5.1	0.5	0.5	0.5	2.3	0.5	40	46	4.6	3.7	0.5	21	0.5	55	0.5	0.5	0.5	0.5	**22**
Amitriptyline	41	3.6	3.4	1.9	19	4	21	27	7.1	2.8	4.9	10	0.5	2.6	0.5	0.5	0.5	0.95	0.84
Atenolol	0.5	0.5	2.2	0.5	0.85	0.5	0.5	0.5	0.5	38	38	19	0.5	0.5	0.5	0.5	0.5	0.5	0
Atorvastatin	6.5	0.5	0.5	0.5	3	1	72	79	5.4	27	5	35	0.5	0.5	0.5	0.5	0.5	0.5	0
Azithromycin	270	5.6	2.5	0.55	130	9.9	120	120	59	1.6	23	54	0.55	1.2	0.55	0.55	1.7	0.96	0.47
Bezafibrate	4.5	4.5	4.5	4.5	0	4.5	4.5	13	4.5	8.1	9.7	0	4.5	4.5	4.5	6.6	4.5	4.5	0
Bicalutamide	630	2.2	0.5	0.5	**310**	27	750	830	180	160	110	**350**	4.3	1.4	2.3	0.5	2.2	0.5	1.4
Bisoprolol	110	1.7	0.5	0.5	55	6.6	67	79	29	13	11	31	0.5	1.3	0.5	0.5	0.5	0.5	0.33
Carbamazepine	1800	94	39	46	**870**	100	720	840	310	290	240	**290**	57	51	49	73	39	38	13
Cetirizine	230	1.8	0.5	0.5	110	28	85	150	85	19	26	51	4.6	8.4	2.2	40	2.4	0.5	**15**
Chloramphenicol	24	6.6	16	17	7.1	27	33	71	22	19	20	20	15	11	4.5	16	6.1	8.8	4.7
Chlorzoxazone	0.55	0.55	0.55	0.55	0	0.55	0.55	0.55	0.55	3.3	0.55	0	0.55	1.5	0.55	0.55	0.55	0.55	0
Citalopram	63	0.5	0.5	0.5	31	11	26	76	12	0.5	5.1	28	0.5	1.3	0.5	0.5	0.5	0.5	0.33
Clarithromycin	22	0.5	0.5	0.5	11	0.93	11	12	3.3	2.1	0.93	5.1	0.5	1	0.5	0.5	0.5	0.5	0.2
Climbazole	14	0.5	0.5	0.5	6.8	0.5	0.5	6.7	4.2	1.3	0.5	2.6	0.5	0.5	0.5	0.5	0.5	0.5	0
Clindamycin	440	0.5	0.5	0.5	220	20	8.9	61	12	18	29	19	41	3.5	20	3.9	19	1.3	**15**
Codeine	1.2	1.2	7.3	4.9	3	2.3	35	24	34	23	13	13	1.2	2.6	1.2	1.9	1.2	1.2	0.59
Desvenlafaxine	3200	4.9	1.8	1.8	**1600**	230	1800	3000	1300	410	620	**1000**	24	2.6	17	1.8	14	6.8	8.8
Diazepam	5.3	0.5	0.5	0.5	2.4	0.5	4.4	2.1	0.5	4.6	0.5	2	0.5	1.6	0.5	0.5	0.5	0.5	0.45
Diclofenac	740	65	3.4	3.4	**360**	54	570	820	120	170	70	**320**	42	50	26	21	7.7	3.4	**18**
Diltiazem	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0
Erythromycin	4900	66	110	80	**2400**	67	1200	530	280	75	170	**430**	120	43	66	43	30	30	**34**
Fexofenadine	280	1.1	12	8	140	120	950	1500	520	150	210	**550**	18	46	16	63	8.7	8.3	**23**
Fluconazole	130	0.5	0.5	0.5	65	6.6	99	110	44	14	23	45	2.9	1.9	2.2	1.8	1.7	0.5	0.78
Fluoxetine	5.8	0.5	0.5	0.5	2.7	0.5	9.2	11	0.5	0.5	0.5	5	0.5	0.5	0.5	0.5	0.5	0.5	0
Furosemide	670	2.9	87	120	300	31	280	330	40	170	45	130	96	86	73	140	120	7.9	**46**
Gemfibrozil	2100	4	56	44	**1000**	13	350	350	69	22	55	160	4	33	4	34	5.4	4	**15**
HCTZ	19	0.95	0.95	0.95	9	0.95	90	84	10	320	74	120	0.95	0.95	2.7	0.95	0.95	0.95	0.71
Irbesartan	140	1.3	0.55	0.55	70	7	100	120	22	30	17	48	0.55	1.4	0.55	0.55	0.55	0.55	0.35
Lamotrigine	780	27	3.6	1.8	**380**	48	740	960	290	290	280	**340**	60	2.1	24	4.5	28	1.3	**23**
Lidocaine	1200	11	31	54	**580**	17	180	76	90	1600	70	**620**	30	38	26	50	16	19	13
Loperamide	6	0.5	0.5	0.5	2.8	0.5	3.2	0.98	0.5	0.5	0.5	1.1	0.5	1	0.5	0.5	0.5	0.5	0.2
Losartan	2700	20	16	26	**1300**	140	1900	2200	620	650	470	**840**	6	17	8	37	13	8.9	11
Memantine	210	0.5	0.5	0.5	100	6.5	34	47	42	3.7	15	19	0.5	0.5	1.6	0.5	0.5	0.93	0.45
Metformin	290	23	20	11	140	100	230	480	130	240	120	140	16	21	13	32	16	16	6.9
Metoprolol	1400	8.9	6.9	1.6	**700**	140	770	920	620	500	370	280	9.2	8.5	7.1	14	6	2	4
Metronidazole	10	0.5	0.5	0.5	4.8	0.5	0.5	0.5	0.5	9.9	7.7	4.3	0.5	0.5	0.5	0.5	0.5	0.5	0
Mirtazapine	81	1.4	0.5	0.5	40	8.9	22	41	11	3.2	3.2	15	0.5	0.5	0.5	0.5	0.5	0.5	0
Norsertraline	10	10	10	10	0	10	10	10	10	10	10	0	10	10	10	10	10	10	0
Oxazepam	62	3.1	3.1	3.1	29	3.1	3.1	3.1	15	44	40	19	3.1	3.1	3.1	3.1	3.1	3.1	0
Oxycodone	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0
Panthenol	1.3	1.3	1.3	1.3	0	1.3	1.3	1.3	1.3	1.3	1.3	0	1.3	1.3	1.3	1.3	1.3	1.3	0
Paroxetine	2.9	0.5	0.5	0.5	1.2	0.5	0.5	0.5	1.5	0.5	0.5	0.41	0.5	1.8	0.5	1.6	0.5	0.5	0.62
Propranolol	58	0.5	0.5	0.5	29	4.9	22	41	8.7	4.9	5.2	15	0.5	4.8	0.5	0.5	0.5	0.5	1.8
Ranitidine	0.55	1.2	0.55	0.55	0.33	0.55	0.55	0.55	0.55	1.7	0.55	0.47	0.55	2	0.55	0.55	0.55	0.55	0.59
Roxithromycin	0.55	0.55	0.55	0.55	0	0.55	0.55	0.55	0.55	0.55	0.55	0	0.55	0.55	0.55	0.55	0.55	0.55	0
Salicylic acid	37	45	8.1	5.5	20	26	49	180	19	73	180	74	2.9	5.1	3.9	10	3.6	4.3	2.6
Sertraline	74	3.6	2.6	1.6	36	5.8	110	76	27	2.3	6.8	45	0.75	3.4	0.75	1.5	0.75	0.75	1.1
Simvastatin	2.9	2.9	2.9	2.9	0	2.9	2.9	2.9	2.9	2.9	2.9	0	2.9	2.9	2.9	2.9	2.9	2.9	0
Sotalol	3.7	3.7	9.5	3.7	2.9	3.7	3.7	3.7	3.7	19	89	34	100	3.7	52	3.7	59	3.7	**40**
Sulfamethoxazole	0.8	0.8	0.8	0.8	0	0.8	0.8	0.8	0.8	0.8	0.8	0	0.8	0.8	0.8	0.8	0.8	0.8	0
Terbutaline	13	1.9	8.1	5	4.7	0.5	21	6.9	0.5	11	0.5	8.2	0.5	3.1	0.5	5	0.5	0.5	1.9
Tramadol	340	4	1.1	0.5	170	14	98	78	80	110	270	86	6.8	18	5.9	16	6.1	5.8	5.6
Trimethoprim	23	23	23	23	0	23	23	23	23	23	23	0	23	23	23	23	23	23	0
Valsartan	0.95	0.95	0.95	0.95	0	84	1000	820	420	170	280	**370**	0.95	2	0.95	0.95	0.95	2.7	0
Venlafaxine	260	0.5	0.5	0.5	130	130	280	830	320	84	90	280	15	0.5	0.5	0.5	2.2	0.5	5.8
∑mean concentration	23000	470	510	500		1500	13000	16000	5900	5900	4200		740	590	490	680	470	240	
Mean	420	8.4	9.1	8.9		28	230	290	110	100	74		13	11	8.7	12	8.4	4.2	
Median	39	1.4	1	0.68		6.6	34	54	14	16	16		1.1	2.4	1.3	1.4	1.3	0.94	
Detection frequency	41	22	20	15		30	39	40	39	43	38		19	31	20	18	19	12	

Table A6: ∑mean concentrations, mean concentrations, means, medians and standard deviations (STDs) (bolded ones are the 10 most significant in each area) (ng/g dw) of pharmaceuticals in the solid sewage samples from the sludge cell with belonging detection frequencies. If the concentration of the pharmaceutical was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A pharmaceutical was not detected if its mean concentration was ≤ LOQ (red box).

Pharmaceutical	Feb 21	March 21	April 21	May 21	June 21	July 21	Aug 21	Sep 21	Storvreta	STD
Albuterol	0.85	0.85	0.85	0.85	0.85	0.85	0.85	0.85	0.85	0
Amitriptyline	74	85	89	150	300	170	210	150	99	**73**
Atenolol	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Atorvastatin	27	20	21	17	10	28	14	6.5	21	7.2
Azithromycin	1.4	0.5	0.5	2.2	3.3	0.5	0.5	1.4	0.5	1
Bezafibrate	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Bicalutamide	23	24	27	54	100	54	73	36	33	26
Bisoprolol	1.3	2.3	2.4	0.5	4.3	2.7	2.7	2.5	2.5	1
Carbamazepine	48	48	55	77	97	39	47	26	36	22
Cetirizine	26	28	38	62	82	57	62	37	27	20
Chloramphenicol	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Chlorzoxazone	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2	0
Citalopram	75	81	95	190	350	170	230	130	100	**90**
Clarithromycin	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Climbazole	8.5	11	9.7	14	31	20	30	18	13	8.4
Clindamycin	0.5	2.3	2.1	3.5	2.8	1.5	3.7	1.3	0.5	1.2
Codeine	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Desvenlafaxine	64	55	95	130	210	150	180	98	81	**53**
Diazepam	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Diclofenac	21	31	47	15	62	28	64	43	22	18
Diltiazem	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Erythromycin	1.4	1.4	1.4	3.2	1.4	4.7	2.6	3	1.4	1.2
Fexofenadine	95	74	150	140	340	190	170	82	150	**80**
Fluconazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Fluoxetine	47	50	51	95	150	94	110	89	77	**33**
Furosemide	11	13	16	4.6	7.9	7.1	15	14	2.8	4.8
Gemfibrozil	3.9	6.3	5.7	7.9	5.5	6.6	12	7.8	2.3	2.7
HCTZ	1.2	1.2	1.2	1.2	1.2	1.2	1.2	3.4	3.3	0.95
Irbesartan	6.3	8.7	9.8	5.9	15	8.2	14	9.3	3.8	3.6
Lamotrigine	41	42	55	26	74	60	52	58	66	15
Lidocaine	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Loperamide	5.7	7.1	6.7	16	23	13	21	13	4.5	6.8
Losartan	120	110	120	130	190	200	140	130	210	**39**
Memantine	1.3	2.6	2.4	3	4.7	4	3	2.4	4	1
Metformin	12	12	12	12	32	30	32	12	12	9.7
Metoprolol	43	41	43	52	130	57	75	51	34	29
Metronidazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Mirtazapine	45	50	56	62	120	78	91	67	29	27
Norsertraline	190	210	200	260	400	220	290	260	340	**70**
Oxazepam	7.9	11	11	10	20	14	18	13	9.5	4
Oxycodone	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Panthenol	1.8	1.8	1.8	7.3	3.3	3.5	6.8	1.8	1.8	2.2
Paroxetine	4.1	3.5	3.3	7.9	12	5.8	8.6	6	8.7	2.9
Propranolol	24	23	29	37	81	34	44	29	39	18
Ranitidine	1.4	1.4	1.4	1.4	1.4	1.4	1.4	1.4	1.4	0
Roxithromycin	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Salicylic acid	150	50	15	19	150	57	140	1	1	**64**
Sertraline	220	220	210	300	460	260	330	300	370	**82**
Simvastatin	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	0
Sotalol	1.3	1.3	1.3	1.3	1.3	1.3	1.3	1.3	1.3	0
Sulfamethoxazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Terbutaline	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	0
Tramadol	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Trimethoprim	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Valsartan	2.8	3.4	2.9	2.3	2.8	3.5	3.3	2.7	0.5	0.9
Venlafaxine	25	29	25	84	130	73	120	57	28	**41**
∑mean concentration	1400	1400	1500	2000	3600	2200	2600	1800	1800	
Mean	26	25	27	36	65	39	47	32	33	
Median	2.5	3	2.7	4.1	4.5	4.4	5.3	2.9	2.4	
Detection frequency	29	30	30	31	32	32	32	32	26	

Table A7: ∑mean concentrations, mean concentrations, means, medians and standard deviations (STDs) (bolded ones are the 10 most significant in each area) (ng/L) of other chemicals of emerging concern (CECs) in the water grab sampling points within Hovgården's facility with belonging detection frequencies. If the concentration of the CEC was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A CEC was not detected if its mean concentration was ≤ LOQ (red box).

PFASs	**L2**	**L3**	**L4**	**L5**	**STD**	**L6**	**L7**	**L8**	**L10**	**L11**	**L12**	**STD**	**W1**	**W2**	**W5**	**W9**	**R0**	**R2**	**STD**
FOSA	1.3	2.6	2.5	1.3	0.72	1.3	1.3	1.3	1.3	1.3	1.3	0	1.3	3.7	1.3	1.3	1.3	1.3	0.98
PFBS	130	100	250	220	71	13	23	18	65	20	46	**20**	160	190	180	230	120	110	45
PFDA	0.5	8.4	7.6	4.3	3.6	3.2	6.4	3.6	7.8	4.3	9.1	2.4	1.9	8.3	0.5	4.1	0.5	0.5	3.1
PFDoDA	0.85	5.4	0.85	3.1	2.2	0.85	3.5	3.2	70	4.6	2.2	**27**	1.8	2.1	2	1.7	10	0.85	3.4
PFHpA	75	200	260	230	**81**	13	10	0.5	25	49	40	19	200	290	200	250	140	110	**67**
PFHxA	260	170	410	360	**110**	49	110	100	82	94	96	**21**	350	400	380	390	240	240	**74**
PFHxS	25	59	160	150	67	0.5	11	15	6.5	32	53	19	96	170	69	170	48	25	**62**
PFNA	0.5	21	14	12	8.5	0.5	0.5	0.5	3.1	4.4	6.8	2.6	18	31	12	35	5.8	38	13
PFOA	62	230	560	550	**250**	12	21	17	16	94	140	**54**	200	590	190	480	110	51	**220**
PFOS_linear	11	74	240	170	**100**	20	14	19	23	33	130	**45**	41	240	29	170	18	85	**89**
PFPeA	140	190	330	310	**92**	34	63	21	26	61	49	18	220	200	300	260	180	180	48
PFTeDA	0.5	2.6	0.5	1.7	1	0.5	0.5	2.1	6.8	0.5	0.5	2.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFUnDA	0.7	5	0.7	3.3	2.1	0.7	0.7	2	40	4.3	1.9	16	0.7	0.7	0.7	0.7	0.7	0.7	0
∑mean concentration	710	1100	2200	2000		150	260	200	370	400	580		1300	2100	1400	2000	870	840	
Mean	54	82	170	160		11	20	16	29	31	44		99	160	110	150	67	65	
Median	11	59	160	150		3.2	10	3.6	23	20	40		41	170	29	170	18	38	
Detection frequency	7	12	9	12		7	9	10	12	11	11		10	11	9	9	9	8	
Remaining CECs	**L2**	**L3**	**L4**	**L5**	**STD**	**L6**	**L7**	**L8**	**L10**	**L11**	**L12**	**STD**	**W1**	**W2**	**W5**	**W9**	**R0**	**R2**	**STD**
BAM	10	10	190	250	**120**	150	2100	3300	1100	150	550	**1300**	10	160	10	10	48	90	**61**
Caffeine	38	5.2	5.4	2.3	17	84	340	380	140	5300	1800	**2000**	48	2300	44	5300	30	28	**2200**
Clotrimazole	1.7	0.5	0.5	0.5	0.6	0.5	2.2	2	0.5	0.5	0.5	0.83	0.5	0.5	0.5	0.5	0.5	0.5	0
Daidzein	2.1	2.1	2.1	2.1	0	2.1	300	760	16	170	31	**290**	5.3	2.1	4.3	66	7.1	2.1	**25**
DEET	630	41	1500	1100	**630**	31	67	64	32	100	130	39	130	710	130	870	59	82	**360**
Ethylparaben	61	10	7.5	15	**25**	7.5	7.5	7.5	13	23	40	13	7.5	16	11	12	7.5	7.5	3.4
Imazalil	66	0.53	0.53	0.53	**33**	0.53	0.53	0.53	0.53	0.53	0.53	0	0.53	0.53	0.53	0.53	0.53	0.53	0
Metconazole	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0
Methylparaben	3.7	35	23	18	13	8.8	3.7	3.7	3.7	3.7	10	3	6	5.5	3.7	3.7	5	3.7	1
Miconazole	0.5	0.5	0.5	0.5	0	0.5	5.8	6.6	0.5	0.5	0.5	3	0.5	0.5	0.5	0.5	0.5	0.5	0
Nicotine	16	6.2	7.7	6	4.7	6.6	600	990	130	1200	1300	**550**	7.9	7.4	6.1	600	110	140	**230**
Penconazole	1.1	1.1	1.1	1.1	0	1.1	1.1	1.1	1.1	1.1	1.1	0	1.1	3.8	1.1	1.1	1.1	1.1	1.1
Prochloraz	3.6	0.5	0.5	0.5	1.6	0.5	0.5	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0
Propylparaben	2.1	4.6	2.1	2.1	1.3	2.1	2.1	2.1	3.7	2.1	2.1	0.65	2.1	3.8	3.5	15	2.1	2.1	5.1
Tebuconazole	7.5	1.1	0.7	0.7	3.3	5.9	0.7	18	4.6	4.4	3.9	6	2.9	7.8	3	3.3	0.7	0.7	2.6
Tetraconazole	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0	0.5	0.5	0.5	0.5	0.5	0.5	0
Tolytriazole	620	32	16	27	**300**	70	360	730	140	1700	330	**610**	50	51	42	64	31	17	17
∑mean concentration	1500	150	1800	1400		370	3800	6300	1600	8700	4200		270	3300	260	6900	300	380	
Mean	86	8.9	100	84		22	220	370	93	510	250		16	190	15	410	18	22	
Median	3.7	2.1	2.1	2.1		2.1	3.7	6.6	3.7	3.7	3.9		2.9	3.8	3.5	3.7	2.1	2.1	
Detection frequency	9	6	6	6		7	8	9	7	8	9		6	8	6	7	6	5	

Table A8: ∑mean concentrations, mean concentrations, means, medians and standard deviations (STDs) (bolded ones are the 10 most significant in each area) (ng/g dw) of other chemicals of emerging concern (CECs) in the solid sewage samples from the sludge cell with belonging detection frequencies. If the concentration of the CEC was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A CEC was not detected if its mean concentration was ≤ LOQ (red box).

PFASs	**Feb 21**	**March 21**	**April 21**	**May 21**	**June 21**	**July 21**	**Aug 21**	**Sep 21**	**Storvreta**	**STD**
FOSA	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFBS	0.5	0.5	0.5	0.5	2.7	0.5	0.5	0.5	0.5	**0.73**
PFDA	0.5	0.5	0.5	2.6	0.5	0.5	1.6	0.5	0.5	**0.75**
PFDoDA	0.5	0.5	0.5	1.5	0.5	0.5	0.5	0.5	0.5	0.33
PFHpA	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFHxA	0.5	0.5	0.5	0.5	11	3	3.9	2.4	0.5	**3.4**
PFHxS	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFNA	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0
PFOA	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFOS_linear	3.9	3.2	3.2	5.5	11	4.7	5	2.8	0.5	**2.9**
PFPeA	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0
PFTeDA	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
PFUnDA	0.85	0.85	0.85	1.7	0.85	0.85	0.85	0.85	0.85	**0.28**
∑mean concentration	11	10	10	16	30	14	16	11	7.3	
Mean	0.82	0.77	0.77	1.2	2.3	1.1	1.3	0.88	0.56	
Median	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	
Detection frequency	1	1	1	3	3	2	3	2	0	
Remaining CECs	**Feb 21**	**March 21**	**April 21**	**May 21**	**June 21**	**July 21**	**Aug 21**	**Sep 21**	**Storvreta**	**STD**
BAM	3.7	16	3.7	3.7	3.7	3.7	3.7	3.7	3.7	4.1
Caffeine	5.3	3.8	4.5	21	26	12	25	11	2.6	**9.4**
Clotrimazole	33	59	31	38	58	42	86	37	21	**20**
Daidzein	3.7	4.4	0.9	3.1	4.5	3.9	4.3	3.6	0.9	1.4
DEET	0.5	0.5	0.5	0.5	4.8	10	4.6	1.4	2.2	3.2
Ethylparaben	2.2	2.2	2.2	2.2	5.5	3.8	2.2	2.2	2.2	1.2
Imazalil	190	290	290	440	480	330	420	230	120	**120**
Metconazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Methylparaben	2.9	1.5	1.5	7.3	7.5	7.3	14	4.1	1.5	4.2
Miconazole	46	74	48	55	80	47	60	41	33	**15**
Nicotine	7.3	0.5	0.5	7.1	7.2	24	22	23	5.8	**9.5**
Penconazole	6.5	6.5	6.5	6.5	6.5	6.5	6.5	6.5	6.5	0
Prochloraz	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Propylparaben	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Tebuconazole	18	14	21	29	41	22	37	28	37	**9.4**
Tetraconazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0
Tolytriazole	10	11	11	11	20	18	23	17	19	4.9
∑mean concentration	330	490	420	630	750	530	710	410	260	
Mean	19	29	25	37	44	31	42	24	15	
Median	3.7	3.8	2.2	6.5	6.5	7.3	6.5	4.1	2.6	
Detection frequency	8	8	6	9	11	10	10	10	8	

Table A9: ∑mean concentrations, mean concentrations, means and medians (ng/L) of pharmaceuticals in the time-integrated (TI) and flow-proportional sampling (FPS) point(s) within Hovgården's facility with belonging detection frequencies. If the concentration of the pharmaceutical was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A pharmaceutical was not detected if its mean concentration was ≤ LOQ (red box).

Pharmaceutical	L5TI1	L6TI	L8TI	L11TI	L12TI	W1TI	W2TI	W5TI	W9FP
Albuterol	0.5	0.5	37	0.5	0.5	0.5	0.5	0.5	0.5
Amitriptyline	0.5	0.5	13	2.4	4.1	0.5	0.5	0.5	0.5
Atenolol	0.5	0.5	0.5	32	34	0.5	0.5	0.5	0.5
Atorvastatin	0.5	4.4	71	26	6.3	0.5	0.5	0.5	0.5
Azithromycin	0.55	0.55	110	0.55	15	0.55	0.55	0.55	0.55
Bezafibrate	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5	4.5
Bicalutamide	0.5	31	610	130	100	0.5	0.5	0.5	3.8
Bisoprolol	0.5	2.4	50	12	12	0.5	0.5	0.5	0.5
Carbamazepine	40	120	600	260	230	44	45	51	50
Cetirizine	0.5	3.3	120	16	26	2.3	1.4	0.5	3.5
Chloramphenicol	7.3	4.5	39	4.5	19	26	4.5	4.5	9.3
Chlorzoxazone	0.55	0.55	0.55	1.7	2.3	0.55	0.55	0.55	0.55
Citalopram	0.5	0.5	34	0.5	4.5	0.5	0.5	0.5	0.5
Clarithromycin	0.5	0.5	8.1	0.5	0.5	0.5	0.5	0.5	0.5
Climbazole	0.5	0.5	1.3	0.5	0.5	0.5	0.5	0.5	0.5
Clindamycin	0.5	11	12	16	31	0.5	0.5	2	31
Codeine	1.2	1.2	8.6	19	8.6	1.2	1.2	1.2	1.2
Desvenlafaxine	1.8	68	1900	410	630	4.6	4.3	8.7	20
Diazepam	0.5	0.5	0.5	2	0.5	0.5	0.5	0.5	0.5
Diclofenac	61	97	300	100	90	3.4	3.4	3.4	3.4
Diltiazem	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Erythromycin	50	46	220	95	170	65	30	85	100
Fexofenadine	6.4	20	1100	150	200	22	21	16	12
Fluconazole	3.8	9.1	66	14	20	0.5	1.4	2.1	4
Fluoxetine	0.5	0.5	1.1	0.5	0.5	0.5	0.5	0.5	0.5
Furosemide	61	40	130	160	2.9	95	59	2.9	66
Gemfibrozil	59	17	280	18	75	30	29	10	150
HCTZ	0.95	3.4	71	290	77	2.5	0.95	0.95	0.95
Irbesartan	0.55	12	75	23	14	0.55	0.55	0.55	0.55
Lamotrigine	1.3	78	740	280	270	1.3	1.3	1.3	41
Lidocaine	35	25	70	1500	67	38	43	25	30
Loperamide	0.5	0.5	1.6	0.5	0.5	0.5	0.5	0.5	0.5
Losartan	0.7	390	1700	580	470	19	16	8.2	4.2
Memantine	0.5	3.5	0.5	2	10	0.5	0.5	0.5	0.5
Metformin	44	74	290	270	150	18	13	13	14
Metoprolol	0.5	52	540	590	430	6.9	6.6	4.4	8.1
Metronidazole	0.5	0.5	0.5	6.3	9.6	0.5	0.5	0.5	0.5
Mirtazapine	0.5	0.5	24	4.7	2.9	0.5	1.7	0.5	0.5
Norsertraline	10	10	10	10	10	10	10	10	10
Oxazepam	3.1	3.1	3.1	3.1	3.1	3.1	3.1	3.1	3.1
Oxycodone	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Panthenol	1.3	1.3	1.3	1.3	1.3	1.3	1.3	1.3	1.3
Paroxetine	0.5	0.5	8.3	0.5	0.5	0.5	0.5	0.5	0.5
Propranolol	0.5	0.5	25	4.5	4.4	0.5	0.5	0.5	0.5
Ranitidine	0.55	0.55	0.55	0.55	0.55	0.55	0.55	0.55	0.55
Roxithromycin	0.55	0.55	0.55	0.55	0.55	0.55	0.55	0.55	0.55
Salicylic acid	16	18	51	74	98	3.8	5.3	5.4	7.6
Sertraline	0.75	0.75	48	2.6	4.1	0.75	0.75	0.75	0.75
Simvastatin	2.9	2.9	170	2.9	2.9	2.9	2.9	2.9	2.9
Sotalol	3.7	3.7	3.7	44	94	3.7	3.7	3.7	89
Sulfamethoxazole	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
Terbutaline	5.2	0.5	31	0.5	0.5	0.5	0.5	0.5	0.5
Tramadol	0.5	13	290	110	290	4.8	4.3	6.3	6.5
Trimethoprim	23	23	23	23	23	23	23	23	23
Valsartan	0.95	190	600	150	300	0.95	0.95	0.95	0.95
Venlafaxine	0.5	530	87	94	0.5	0.5	0.5	0.5	0.5
∑mean concentration	460	1400	11000	5500	4100	450	360	320	720
Mean	8.2	25	200	99	74	8.1	6.4	5.6	13
Median	0.55	3.2	36	11	10	0.65	0.88	0.78	0.95
Detection frequency	10	24	40	35	35	15	14	13	19

Table A10: ∑mean concentrations, mean concentrations, means and medians (ng/L) of other chemicals of emerging concern (CECs) in the time-integrated (TI) and flow-proportional sampling (FPS) point(s) within Hovgården's facility with belonging detection frequencies. If the concentration of the CEC was below its limit of quantification (LOQ), it was given the value LOQ/2. LOQ was used a detection parameter, by comparing it to the corresponding mean concentration. A CEC was not detected if its mean concentration was ≤ LOQ (red box).

PFASs	L5TI	L6TI	L8TI	L11TI	L12TI	W1TI	W2TI	W5TI	W9FP
FOSA	1.3	1.3	1.3	1.3	2.8	1.3	1.3	1.3	1.3
PFBS	210	27	16	7.9	35	220	210	260	170
PFDA	6.3	6	0.5	0.5	6.9	3.5	0.5	0.5	3.5
PFDoDA	0.85	0.85	0.85	0.85	6.4	1.5	2	2.8	1.8
PFHpA	220	14	0.5	48	38	230	200	170	180
PFHxA	420	63	78	88	100	480	440	550	340
PFHxS	160	5.8	19	23	47	70	87	45	110
PFNA	12	5.8	0.5	0.5	5.3	25	30	23	0.5
PFOA	530	17	20	87	110	250	210	75	210
PFOS_linear	160	23	23	30	100	110	72	26	41
PFPeA	300	43	24	70	69	320	180	210	330
PFTeDA	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
PFUnDA	0.7	0.7	0.7	0.7	0.7	16	0.7	0.7	0.7
∑mean concentration	2000	210	180	360	520	1700	1400	1400	1400
Mean	160	16	14	28	40	130	110	100	110
Median	160	6	1.3	7.9	35	70	72	26	41
Detection frequency	9	9	6	7	11	10	9	9	9

Remaining CECs	L5TI	L6TI	L8TI	L11TI	L12TI	W1TI	W2TI	W5TI	W9FP
BAM	85	120	10	10	10	85	180	170	10
Caffeine	17	80	330	5300	1800	420	380	38	55
Clotrimazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Daidzein	2.1	36	300	91	51	2.1	2.1	2.1	2.1
DEET	800	30	57	88	120	650	630	150	100
Ethylparaben	27	7.5	7.5	28	7.5	7.5	7.5	18	7.5
Imazalil	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53
Metconazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Methylparaben	3.7	3.7	3.7	3.7	34	3.7	3.7	3.7	3.7
Miconazole	0.5	0.5	7	0.5	0.5	0.5	0.5	0.5	0.5
Nicotine	39	31	580	1100	1300	9.3	11	7.4	12
Penconazole	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1
Prochloraz	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Propylparaben	5.3	2.1	2.1	2.1	2.1	2.1	2.1	2.1	2.1
Tebuconazole	0.7	0.7	0.7	3.7	3.5	0.7	0.7	0.7	0.7
Tetraconazole	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Tolytriazole	23	96	420	1200	260	27	25	29	39
∑mean concentration	1000	410	1700	7800	3600	1200	1200	430	240
Mean	59	24	100	460	210	71	73	25	14
Median	2.1	2.1	3.7	3.7	3.5	2.1	2.1	2.1	2.1
Detection frequency	7	6	6	7	7	5	5	6	4

SUPPLEMENTARY INFORMATION: RESULTS AND DISCUSSION

S4.1: Detected pharmaceuticals at Hovgården.

The 42 pharmaceuticals that were detected in at least one sampling point (SP) included: albuterol, amitriptyline, atenolol, atorvastatin, azithromycin, bezafibrate, bicalutamide, bisoprolol, cetirizine, chloramphenicol, chlorzoxazone, citalopram, clarithromycin, climbazole, clindamycin, codeine, desvenlafaxine, diazepam, diclofenac, erythromycin, fluconazole, fluoxetine, furosemide, gemfibrozil, HCTZ, irbesartan, lamotrigine, loperamide, memantine, metronidazole, mirtazapine, oxazepam, paroxetine, propranolol, ranitidine, salicylic acid, sertraline, sotalol, terbutaline, tramadol, valsartan and venlafaxine (Table A5).

S4.1.1: Detected pharmaceuticals in specific sampling points and the mean composition profile of pharmaceuticals in stream 1.

The 11 pharmaceuticals that were detected in all sampling points (SPs) included: amitriptyline, carbamazepine, fexofenadine, erythromycin, lidocaine, losartan, metformin, metoprolol, salicylic acid, sertraline and terbutaline. The 34 pharmaceuticals that were detected in at least one SP included: albuterol, atenolol, atorvastatin, azithromycin, bicalutamide, bisoprolol, cetirizine, chloramphenicol, citalopram, clarithromycin, climbazole, clindamycin, codeine, desvenlafaxine, diazepam, diclofenac, fluconazole, fluoxetine, furosemide, gemfibrozil, HCTZ, irbesartan, lamotrigine, loperamide, memantine, metronidazole, mirtazapine, oxazepam, paroxetine, propranolol, ranitidine, sotalol, tramadol and venlafaxine. Six of the 34 pharmaceuticals were detected in all SPs except in one. This included chloramphenicol, furosemide and gemfibrozil in all SPs except in L3, as well as azithromycin, lamotrigine and tramadol in all SPs except in L5, ranging from 1.1 ng/L (tramadol) to 2100 ng/L (gemfibrozil). The 17 pharmaceuticals that were only detected in L2 included: albuterol, atorvastatin, citalopram, clarithromycin, climbazole, clindamycin, diazepam, fluconazole, fluoxetine, HCTZ, loperamide, memantine, metronidazole, oxazepam, paroxetine, propranolol and venlafaxine (Table A5).

The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in stream 1 included lidocaine, metoprolol, carbamazepine, gemfibrozil, losartan, desvenlafaxine and erythromycin (5.2, 5.7, 8.0, 8.9, 11, 13 and 21 %, respectively). This was mainly caused by L2's high mean compositions of these pharmaceuticals (Figure 5).

S4.1.2: Detected pharmaceuticals in specific sampling points and the mean composition profile of pharmaceuticals in stream 2.

The 29 pharmaceuticals that were detected in all SPs included: amitriptyline, azithromycin, bicalutamide, bisoprolol, carbamazepine, cetirizine, chloramphenicol, clindamycin, desvenlafaxine, diclofenac, erythromycin, fexofenadine, fluconazole, furosemide, gemfibrozil, irbesartan, lamotrigine, lidocaine, losartan, memantine, metformin, metoprolol, mirtazapine, propranolol, salicylic acid, sertraline, tramadol, valsartan and venlafaxine. The 19 pharmaceuticals that were detected in at least one SP included: albuterol, atenolol, atorvastatin, bezafibrate, chlorzoxazone, citalopram, clarithromycin, climbazole, codeine, diazepam, fluoxetine, HCTZ, loperamide, metronidazole, oxazepam, paroxetine, ranitidine, sotalol and terbutaline. Four of the 19 pharmaceuticals were detected in all SPs except in one. This included atorvastatin,

codeine and HCTZ in all SPs except in L6 and citalopram in all SPs except in L11, ranging from 5.0 ng/L (atorvastatin) to 320 ng/L (HCTZ) (Table A5).

The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in stream 2 included erythromycin, carbamazepine, lamotrigine, valsartan, metoprolol, fexofenadine, losartan and desvenlafaxine (5.0, 5.4, 5.6, 6.0, 7.1, 7.4, 13 and 16 %, respectively) (Figure 7).

S4.1.3: Detected pharmaceuticals in specific sampling points and the mean composition profile of pharmaceuticals in the mixed stream and in the river.
The nine pharmaceuticals that were detected in all SPs included: carbamazepine, clindamycin, fexofenadine, furosemide, lidocaine, losartan, metformin, metoprolol and tramadol. The 29 pharmaceuticals that were detected in at least one SP included: albuterol, amitriptyline, azithromycin, bicalutamide, bisoprolol, chloramphenicol, chlorzoxazone, cetirizine, citalopram, codeine, desvenlafaxine, diazepam, diclofenac, erythromycin, fluconazole, gemfibrozil, HCTZ, irbesartan, lamotrigine, memantine, paroxetine, propranolol, ranitidine, sertraline, sotalol, terbutaline, valsartan and venlafaxine. Four of the 29 pharmaceuticals were detected in all SPs except in one. This included salicylic acid in all SPs except in W1 as well as cetirizine, diclofenac and fluconazole in all SPs except in R2, ranging from 1.7 ng/L (fluconazole) and 50 ng/L (diclofenac). The 11 pharmaceuticals that were only detected in W2 included: albuterol, amitriptyline, bisoprolol, chlorzoxazone, citalopram, codeine, diazepam, irbesartan, propranolol, ranitidine and sertraline (Table A5).

The dominating contributors (≥ 5.0 % in mean composition) of pharmaceuticals in the mixed stream and in the river included lidocaine (5.8 and 5.0 %), sotalol (6.4 and 8.9 %), carbamazepine (9.2 and 11 %), erythromycin (11 and 8.5 %) and furosemide (16 and 18 %). Also, diclofenac and fexofenadine in the mixed stream (5.6 and 5.7 %) and trimethoprim in the river (6.5 %) (Figure 9).

S4.2: Detected pharmaceuticals in specific sampling points and the mean composition profile of pharmaceuticals in sewage sludge.
The 24 pharmaceuticals that were detected in all SPs included: amitriptyline, atorvastatin, bicalutamide, carbamazepine, cetirizine, citalopram, climbazole, desvenlafaxine, diclofenac, fexofenadine, fluoxetine, irbesartan, lamotrigine, loperamide, losartan, memantine, metoprolol, mirtazapine, norsertraline, oxazepam, paroxetine, propranolol, sertraline and venlafaxine. The 11 pharmaceuticals that were detected in at least one SP included: azithromycin, bisoprolol, clindamycin, erythromycin, furosemide, gemfibrozil, HCTZ, metformin, panthenol, salicylic acid and valsartan. Two of the 11 pharmaceuticals were detected in all SPs except in one. This included bisoprolol in all SPs except in May 21 and valsartan in all SPs except in Storvreta, ranging from 1.3 ng/g dw (bisoprolol) to 3.5 ng/g dw (valsartan) (Table A6).

The dominating contributors (≥ 5.0 %) of pharmaceuticals in the sludge cell included desvenlafaxine, amitriptyline, losartan, fexofenadine, citalopram, norsertraline and sertraline (5.8, 7.2, 7.3, 7.6, 7.7, 13 and 15 %, respectively) (Figure 11).

S4.4: Other detected CECs at Hovgården.
The 19 other CECs that were detected in at least one SP included: FOSA, PFDA, PFDoDA, PFHpA, PFHxS, PFNA, PFTeDA, PFUnDA, BAM, clotrimazole, daidzein, ethylparaben, imazalil, methylparaben, miconazole, penconazole, prochloraz, propylparaben and tebuconazole (Table A7).

S4.4.1: Other detected CECs in specific sampling points and the mean composition profile of other CECs in stream 1.
The seven PFASs that were detected in all SPs included: PFBS, PFHpA, PFHxA, PFHxS, PFOA, PFOS_linear and PFPeA. The five PFASs that were detected in at least one SP included: PFDA, PFDoDA, PFNA, PFTeDA and PFUnDA. PFDA and PFNA were detected in all SPs except in L2 (Table A7).

The dominating contributors (≥ 5.0 % in mean composition) of PFASs in stream 1 was the highest for PFHxS, PFOS_linear, PFBS, PFHpA, PFPeA, PFHxA and PFOA (6.5, 8.2, 12, 13, 16, 20 and 23 %, respectively) (Figure A7).

The four of the remaining CECs that were detected in all SPs included: caffeine, DEET, nicotine and tolytriazole. The eight of the remaining CECs that were detected in at least one SP included: BAM, clotrimazole, ethylparaben, imazalil, methylparaben, prochloraz, propylparaben and tebuconazole. Methylparaben was detected in all SPs except in L2 (Table A7).

The dominating contributors of the remaining CECs in stream 1 included BAM (9.6 %), tolytriazole (15 %) and DEET (68 %) (Figure A9).

S4.4.2: The mean composition profile of other CECs in stream 2.
The six PFASs that were detected in all SPs included: PFBS, PFDA, PFHxA, PFOA, PFOS_linear and PFPeA. The six PFASs that were detected in at least on SP included: PFNA, PFDoDA, PFHxS, PFTeDA and PFUnDA. PFDoDA and PFHxS were detected in all SPs except in L6, as well as PFHpA except in L8 (Table A7).

The dominating contributors (≥ 5.0 % in mean composition) of PFASs in stream 2 included PFHxS, PFHpA, PFBS, PFOS_linear, PFPeA, PFOA and PFHxA (6.0, 7.0, 9.4, 12, 13, 15 and 27 %, respectively) (Figure A11).

The five of the remaining CECs that were detected in all SPs included: BAM, caffeine, DEET, nicotine and tolytriazole. The six of the remaining CECs that were detected in at least one SP included: daidzein, clotrimazole, ethylparaben, methylparaben, miconazole and tebuconazole. Daidzein was detected in all SPs except in L6, as well as tebuconazole except in L7 (Table A7).

The dominating contributors of the remaining CECs in stream 2 included daidzein, tolytriazole, nicotine, BAM and caffeine (5.1, 13, 17, 30 and 32 %, respectively) (Figure A13).

S4.4.3: The mean composition profile of other CECs in the mixed stream and in the river.

The eight PFASs that were detected in all SPs included: PFBS, PFHpA, PFHxA, PFHxS, PFNA, PFOA, PFOS_linear. The three PFASs that were detected in at least one SP included: FOSA, PFDA, and PFDoDA. PFDoDA was detected in all SPs except in W9 and R2. PFDA was only detected in W1, W2 and W9 (Table A7).

The dominating contributors (≥ 5.0 % in mean composition) of PFASs in the mixed stream and in the river included PFBS, PFPeA, PFDA, PFUnDA, PFHxA and PFOS_linear (5.3, 5.7, 6.1, 6.7, 18 and 32 %, respectively) (Figure A15).

The four of the remaining CECs that were detected in all SPs included: caffeine, DEET, nicotine and tolytriazole. The six of the remaining CECs that were detected in at least one SP included: BAM, daidzein, ethylparaben, penconazole, propylparaben and tebuconazole (Table A7).

The dominating contributors of remaining CECs in the mixed stream and in the river included nicotine (5.8 and 37 %, respectively), DEET (17 and 21 %, respectively) and caffeine (72 and 8.5 %, respectively). Tolytriazole (7.0 %) and BAM (20 %) were also dominating contributors in the river (Figure A17).

S4.5: The mean composition profile of other CECs in the sewage sludge.

The four PFASs that were detected in at least one SP included: PFBS, PFDA, PFDoDA and PFHxA (Table A8).

The dominating contributors (≥ 5.0 % in mean composition) of PFASs in the sludge cell included PFBS, PFPeA, PFDA, PFUnDA, PFHxA and PFOS_linear (5.3, 5.7, 6.1, 6.7, 18 and 32 %, respectively) (Figure A19).

The six of the remaining CECs that were detected in all SPs included: caffeine, clotrimazole, imazalil, miconazole, tebuconazole and tolytriazole. The six of the remaining CECs that were detected in at least one SP included: BAM, daidzein, DEET, ethylparaben, methylparaben and nicotine (Table A8).

The dominating contributors of the remaining CECs in the sludge cell included tebuconazole, clotrimazole, miconazole and imazalil (5.5, 9.0, 11 and 62 %, respectively) (Figure A21).