

Pharmaceuticals and additives in personal care products as environmental pollutants

– Faroe Island, Iceland and Greenland





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ISBN 978-92-893-2561-5

<http://dx.doi.org/10.6027/TN2013-541>

TemaNord 2013:541

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Layout: Hanne Lebech

Cover photo: Maria Dam

This publication has been published with financial support by the Nordic Council of Ministers. However, the contents of this publication do not necessarily reflect the views, policies or recommendations of the Nordic Council of Ministers.

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Preface

Recently, a plan emerged to prepare an overview report of the present knowledge of pharmaceuticals and compounds used in personal care products in the Nordic Countries. It turned out however, that such an overview report would be more or less void on information for the area west of Norway, as only sporadic information was available on such compounds in Faroe Islands, Iceland and Greenland. Experience from earlier studies in Faroe Islands and Iceland on “new” contaminants (www.nordicscreening.org) indicated that local pollution could not be ruled out, but explicit data were lacking. Thus, it was decided to try to fill this knowledge-gap in a co-operative effort, and with leverage from experts in Scandinavia. The present report describes the result of this co-operation; a first snap-shot of the environmental concentrations of pharmaceuticals and compounds used in personal care products in hot-spot areas in Faroe Islands, Iceland and Greenland.

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Summary

The report summarises the results of screening analyses of pharmaceuticals and additives in personal care products in presumed hotspots in Faroe Islands, Iceland and Greenland. The compounds analysed were human pharmaceuticals that is compounds that are administered to alleviate and cure symptoms and illnesses. Also, the study included analyses of compounds added to personal care products to increase their hygienic properties or shelf life. The selection of pharmaceuticals and personal care substances, PPCPs, for the study, was based on assessments of pharmaceutical use in Faroe Island, Iceland and Nuuk, Greenland in 2010. In addition, studies of administered volumes of pharmaceuticals in Nordic Countries and assessment of risk to the environment posed by these, as well as results of a recent screening and risk assessment study performed in Norway, founded the basis for selecting substances included for screening in the present study. Sampling was done in 2010 and supplementary sampling in 2011. In all 38 pharmaceuticals or metabolites of pharmaceuticals and 7 additives in personal care products were analysed. The analyses were done on a total of 44 samples, whereof some were analysed as parallel samples and some as duplicates.

Of the PPCPs analysed, a few, like diclofenac and ibuprofen, were detected in every or nearly every sample, and some, like simvastatin and sulfamethizole, were not detected in any. The synthetic oestrogen 17α ethinylestradiol was not detected in any sample, and the natural counterpart 17β estradiol was detected only in a few. This was mainly due to the high detection limit of the applied method, whereas estrone, which is also a natural oestrogen, was detected in most samples.

The PPCPs occurring in highest overall (median) concentrations were cetrimonium salts (ATAC-C16) >sodium lauryl ethersulphate (SDSE01-4) \approx cocoamidopropyl betaine (CAPB) >sodium laurylsulphate (SDS) and salicylic acid. Ethylenediaminetetraacetic acid (EDTA), metformin and citalopram occurred in similar though somewhat lower median concentrations, as did ibuprofen and metoprolol. All but one PPCP, with median concentration above the detection limit in both solid and liquid samples, occurred in higher concentration in solids than in liquids, when seen on a weight to weight basis, where the concentration in one kg of liquids were compared to one kg of solids. The sole exception was paracetamol,

which often was found in higher concentrations in liquid samples than in solids. A high concentration ratio in sludge to that in liquids indicates that the potential for removing the PPCP in a WWTP is high, and the potential for escaping to the recipient is low.

In general, only a few PPCPs were detected in sediments from recipients, but salicylic acid, a metabolite of acetylsalicylic acid, was found in every one of these. Also the surfactant ATAC-C16 was found in most sediment samples.

Preliminary environmental risk assessments were based on the ratio of measured PPCP concentrations in recipient water to the predicted no-effect concentration, PNEC. The calculations indicated that the largest risk was posed by CAPB and ATAC-C16. Unacceptable risk ratios were found for CAPB and ATAC-C16 in particular, with overall highest risk in recipient water near Iggia in Greenland, and next highest near Sersjantvíkin WWTP in Torshavn, Faroe Islands. Risk ratios above 1 were also found for SDSEO1-4. Summing up, risk ratios exceeding 1 were found in eight of the 11 samples of recipient waters analysed, most frequently due to CAPB, and then ATAC-C16, and in one sample also due to SDSEO1-4.

Risk ratios exceeding 1 was not observed for any pharmaceutical in these recipient water samples. However, this does not necessarily exclude risk from these compounds, because ecosystem toxicity data, on which such assessments are based, were only available for approx. 2/3 of the pharmaceuticals analysed. Lack of PNECs hindered risk assessment for 12 of the pharmaceuticals analysed: amiloride, atenolol, dipyridamole, enalapril, enalaprilat, estrone, gliclazide, paroxetine, perindopril, perindoprilat, sulfamethizole and zopiclone. Although the highest risk may not necessarily be posed by the contaminant occurring in highest concentrations, it is relevant to state that the pharmaceuticals for which no risk assessment could be made, are mainly the ones that occurred in low concentrations, although dipyridamole, atenolol and amiloride were among the 10 pharmaceuticals occurring in overall highest median concentration both in liquid and in solid samples.

The study comprises analyses of PPCPs in sewage lines from households and industry in general, and from hospitals.

The sampling was done as snap-shot sampling, which means that fluctuations which occur naturally in waste water lines are not taken heed of. In solid samples as sludge and sediment, similar fluctuations do not occur, and the results obtained for these samples are more robust. However, when assessing the results it should be kept in mind that there are differences in WWTP design, residence time and loading into the waste water

lines. This means that a label like sludge or even sediment may in fact have been applied on quite different material. Among the waste water lines sampled, some discharge waste water to sea without treatment, and some incorporate one or more steps of microbial sludge digestion and filtering. These differences should be kept in mind when comparisons between sites are done. However, the primary purpose of the screening was not comparison between sites, but to provide insight into the discharge of pharmaceuticals and additives in personal care products in areas where little or no information on this existed. The users of this information are assumed to be mainly the authorities responsible for waste water treatment and environmental pollution monitoring.

The present study has provided a first impression of the concentration levels of PPCPs in Faroe Islands, Iceland and in Nuuk, Greenland. However, the study was done on a limited number of samples, and there are still knowledge gaps. Further investigations are recommended in order to investigate for example daily and seasonal variations, variations in throughput of the WWTP, and removal capacity of the WWTP. In addition, recipient waters from Iceland remain to be analysed. Risk assessment for sediments was not performed due to the lack of PNEC data for the sediments. Future assessments would benefit immensely from having toxicity data for sediment-dwelling organisms available. Also, it is strongly recommended that the findings are scrutinised more closely for each sewage line/WWTP/recipient location separately by the local authorities responsible for the waste water handling, so that possible shortfalls in this may be identified and prioritized for amelioration.

1. Frame of the study

In recent years, focus has been on what happens to pharmaceuticals and compounds added to personal care products after they have “done their job” so to say, and have left the consumer via the sewage line. That the question is pertinent, has been shown in studies of hormone actions on for instance fish in recipient waters near larger cities, and in a wealth of reports on pharmaceuticals in waters, even in groundwater. The problem is not one that will go away on its own, as the use of pharmaceuticals, in particular, is assumed to increase with the ageing of the population and the increasing demand for medical treatment. The problem is emphasized by the fact that waste water treatment plants are generally designed to deal with solids and substances that stick to these, whereas pharmaceuticals and personal care substances are often water-soluble.

The present report describes the result of a study designed to obtain information on the discharge and potential harmful concentrations of pharmaceuticals and additives in personal care products to and in the marine environment of Faroe Island, Iceland and Greenland. The process started with a survey of the most commonly used pharmaceuticals in the countries involved, and with a literature study of relevant and recent literature. The list of pharmaceuticals and additives in personal care products thus established was presented to the highly skilled analytical chemists for evaluation and refining, and thus, a final analytical scheme was produced. In 2010 and 2011, samples were taken in waste water /sewage lines near presumed hotspots, like hospitals and capitals, but also in areas with somewhat lower populations/population densities. The sampling was done using guidelines provided by the laboratories that also provided advice on sample storage. Because of the difficulties in arranging sampling and restraints imposed by the necessary long distance shipping of samples and the resulting risk for breakage of glassware, backup samples were taken and kept in store locally, to be shipped upon demand. Chemical analyses of pharmaceuticals were performed by the Norwegian Institute for Air Research, and the personal care substances were analysed by the Swedish Environmental Research Institute.

The overall aim of acquiring information of this kind was to learn about the flow of this group of environmental pollutants to the aquatic environment, and to elucidate if there are important shortcomings in the present waste water treatment.

The present project was run by a steering group consisting of one representative from a governmental or scientific agency whose working area covered environmental pollution, and one representative from the pharmaceutical authorities, from each country. This group initiated and planned the study, implemented the sampling and took part in the reporting.

The steering group members were:

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The funding for the survey was graciously provided by the Nordic Council of Ministers Arctic Co-operation Programme, by the Nordic Chemicals Group, by the Aquatic Ecosystems Group and by the Working Group under the Nordic Committee of Senior Officials for the Environment, all under the umbrella of the Nordic Council of Ministers, in addition to the participating governmental agencies and institutes.

2. Background

In this chapter a general overview of the groups of investigated pharmaceuticals and additives in personal care products, as well as on the individual investigated substances, are given.

Pharmaceuticals are substances used in the diagnosis, treatment, or prevention of disease and for restoring, correcting, or modifying organic functions.

Personal care products are non-medicinal consumable products that are used in the topical care and grooming of the body and hair and that is rubbed, poured, sprinkled, or sprayed on, introduced into, or otherwise applied to a body, human or animal, for cleansing, beautifying, promoting attractiveness, or altering the appearance without affecting the body's structure or functions. Personal care products are used in such activities as cleansing, toning, moisturizing, hydrating, exfoliating, conditioning, anointing, massaging, colouring/decorating, soothing, deodorizing, perfuming and styling.

2.1 Non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs

2.1.1 *Scope and definition*

Analgesics are agents that decrease pain without resulting in loss of consciousness and are often referred to as painkillers.

Anti-inflammatories are agents that reduce inflammation.

Antipyretics are agents that reduce fever and drugs included in the class of antipyretic analgesics possess analgesic and antipyretic actions but lack anti-inflammatory effects.

Local anaesthetic agents are drugs intended for topical or parenteral administration and produce a state of local anaesthesia by reversibly blocking the nerve conductors that transmit the feeling of pain from this locus to the brain. The loss of sensation can be induced with or without loss of consciousness.

Non-steroidal anti-inflammatory drugs (NSAID's) include both antipyretic and analgesic agents, although some of these drugs may only cover either

of these properties. Most NSAID's act as non-selective inhibitors of the enzyme cyclooxygenase (COX) and interfere with the biosynthesis pathway of prostaglandins and thromboxane. Their specific characteristics have prompted their frequent use in the treatment of rheumatic symptoms.

2.1.2 Compounds analysed

Acetylsalicylic acid possesses antipyretic, anti-inflammatory, analgesic and anticoagulative properties. It was chosen for screening since it is used in quite high amounts at the investigated locations. Due to its chemical structure and physico-chemical properties it is unstable in aquatic environments and degrades easily to the main metabolites acetic and salicylic acids. Salicylic acid was semi-quantitatively analysed and included in the screening in order to get an impression of its environmental concentration levels.

Diclofenac possess structural characteristics of both the arylalkanoic acid and the anthranilic acid classes of anti-inflammatory drugs, and displays anti-inflammatory, analgesic, and antipyretic properties. It is rapidly and almost completely absorbed after oral administration, but is only 50–60% bioavailable due to extensive first-pass effect.


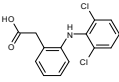
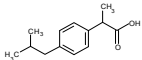
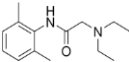
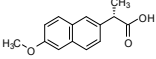
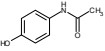
Ibuprofen is a racemic mixture with the S (+)-enantiomer being biologically active and exerts anti-inflammatory properties. The drug is rapidly absorbed following oral administration, metabolised rapidly and nearly completely excreted in the urine as unchanged drug and oxidative metabolites.

Naproxen has anti-inflammatory properties and is generally marketed as its S (+)-enantiomer. It is almost completely absorbed following oral administration and excreted as either unchanged drug (60%) or drug conjugates (10%).

Paracetamol (Acetaminophen) belongs to the antipyretics group and is indicated for use as an antipyretic/analgesic. Approximately 5% of the dose is excreted unchanged in the urine.

Lidocaine is a local anaesthetic which can be administered either parenterally or topically but is also frequently used as a class IB antiarrhythmic agent (see also chapter cardiovascular drugs). It is primarily metabolised in the liver followed by renal excretion of the unchanged drug (<10%) and metabolites.

Table1. Non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs selected for this study

Compound	Class	Structure	CAS1 No	ATC2 No
Acetylsalicylic acid	Non-steroidal anti-inflammatory		50-78-2	B01AC06 B01AC30 N02BA01
Diclofenac	Non-steroidal anti-inflammatory		15307-86-5	M01AB55 M01AB05 D11AX18 S01BC03 M02AA15
Ibuprofen	Non-steroidal anti-inflammatory		15687-27-1	M01AE01 C01EB16
Lidocaine	Local anaesthetic Anti-arrhythmic agent Class IB		137-58-6 73-78-9	N01BB20 N01BB02 QN01BB52 N01BB52 C05AA01
Naproxen	Non-steroidal anti-inflammatory		22204-53-1	M01AE02 M01AE52
Paracetamol (Acetaminophen)	Antipyretic		103-90-2	N02BE01 N02AA59

¹Chemical Abstracts Service Registry Number.

²Anatomical Therapeutic Chemical Classification System.

2.2 Antibiotics and antimicrobial agents

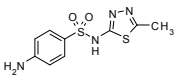
2.2.1 Scope and definition

Antibiotics are microbial metabolites or synthetic analogues, which inhibit the growth and survival of microorganisms without serious toxicity to the host. The many synthetic substances that are unrelated to natural products, but still inhibit or kill microorganisms, are referred to as *anti-microbial agents*.

2.2.2 Compounds analysed

Sulfamethizole is a sulfonamide antibiotic. The sulfonamides are synthetic bacteriostatic antibiotics with a wide spectrum against most gram-positive and many gram-negative organisms. It is rapidly excreted in the urine and mainly as unchanged drug (up to 90%).

Table 2. Antimicrobial agents selected for this study

Compound	Class	Structure	CAS No	ATC No
Sulfamethizole	antimicrobial		144-82-1	B05CA04 D06BA04 J01EB02 S01AB01

2.3 Antidepressants

2.3.1 Scope and definition

Antidepressant agents are used to counteract or treat depression and are classified according to their activity. The antidepressant analysed in the present study are classified as selective 5-HT reuptake inhibitors (SSRIs), and as selective norepinephrine reuptake inhibitors (SNRIs), Table 3.

2.3.2 Compounds analysed

Fluoxetine, paroxetine and citalopram are phenoxyphenylalkylamine SSRIs.

Fluoxetine is marketed as a racemic mixture of R- and S-fluoxetine. The oral bioavailability is approx. 70% and excretion via urine is between 25–50%.

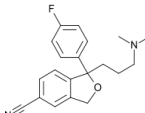
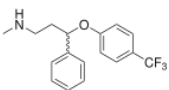
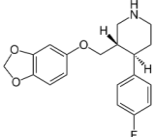
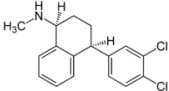
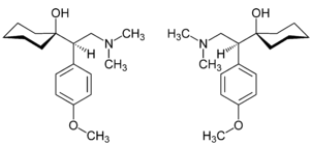
Paroxetine is a constrained analogue of fluoxetine. Its oral bioavailability is 50% and excretion occurs mainly via urine (51–60%).

(±) Citalopram can be viewed as a constrained analogue of paroxetine. It is around 80% orally available and excretion occurs mainly via feces (80–90%).

Sertraline is a phenylalkylamine SSRI with an oral bioavailability of 20 to 36%. Sertraline and its conjugates are excreted both via feces and urine, with less than 5% as the unchanged drug.

Venlafaxine belongs to the methoxyphenylethylamine NSRIs. It is rapidly absorbed with a bioavailability of 45% due to first-pass metabolism. Venlafaxine and its metabolites are primarily excreted in the urine (87%).

Table 3. Antidepressants selected for this study

Compound	Class	Structure	CAS No	ATC No
Citalopram	SSRI		59729-33-8	N06AB10 N06AB04
Fluoxetine	SSRI		54910-89-3	N06AB03
Paroxetine	SSRI		61869-08-7	N06AB05
Sertraline	SSRI		79617-96-2	N06AB06
Venlafaxine	SNRI		93413-69-5	N06AX16

2.4 Antidiabetics

2.4.1 Scope and definition

Antidiabetic medications are used in the treatment of diabetes mellitus by lowering glucose levels in the blood, and are available in several types as for instance insulin, sulfonylureas and biguanides. Many antidiabetics, though insulin is not among these, are orally administered and are thus often called oral hypo- or antihyperglycemic agents.

2.4.2 Compounds analysed

Metformin is an antihyperglycemic agent and belongs to the class of biguanides which are defined as insulin sensitizers by suppressing glucose production by the liver. Metformin is quickly absorbed and has a bioavailability from 50 to 60% and is excreted in the urine as unmetabolised drug.

Gliclazide belongs to the class of sulfonylureas and works by stimulating the pancreas to produce more insulin, which in turn reduces the blood glucose levels. Treatment applications are frequently combined with metformin or other agents to control diabetes. Gliclazide is extensively metabolised in the liver and only less than 1% of the orally administered dose appears unchanged in the urine.

Table 4. Antidiabetics selected for this study

Compound	Class	Structure	CAS No	ATC No
Metformin	Biguanide Antihyperglycemic agents		657-24-9	A10BD03 A10BD05 A10BD08 A10BA02 A10BD07
Gliclazide	Sulfonylurea		21187-98-4	A10BB09

2.5 Antiulcer drugs

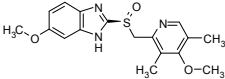
2.5.1 Scope and definition

Antiulcer drugs are used to treat ulcers in the stomach and the upper part of the small intestine. Recurrent gastric and duodenal ulcers are often caused by *Helicobacter pylori* infections, and treatments incorporate combined therapy with antibiotics and gastric acid suppressants. The primary class of drugs used for gastric acid suppression are the proton pump inhibitors.

2.5.2 Compounds analysed

Omeprazole is a proton pump inhibitor, inhibiting stimulated gastric acid secretion irrespective of the receptor stimulation process. Omeprazole is synthesised as a racemic mixture. However, *in vivo* the enantiomers interconvert, doubling the concentration of the more active (*S*)-enantiomer.

Table 5. Antiulcer drugs selected for this study

Compound	Class	Structure	CAS No	ATC No
Omeprazole	Proton pump inhibitor		119141-88-7	A02BC05 QA02BC01

Omeprazole was chosen for the screening since it is frequently administered in the investigated areas. Due to its chemical structure and physico-chemical properties it is unstable in the environment and was therefore not analysed quantitatively.

2.6 Cardiovascular drugs

2.6.1 Scope and definition

Cardiovascular drugs encompass a large number of prescription medications intended to affect the heart and blood vessels. It is a diverse group of drugs and many are used for multiple diseases.

Drugs affecting the cardiovascular system can be classified into six subgroups:

Cardiac glycosides, antianginal and antiarrhythmic agents

Cardiac glycosides occur mainly as secondary plant metabolites and are divided in positive inotropic and non-glycosidic positive inotropic agents. Positive inotropic drugs are often applied in treatment of congestive heart failure and associated oedema.

Antianginal drugs are used in the treatment of angina pectoris and are classified into organic nitrates, calcium channel blockers, β -adrenergic blocking agents, modulators of myocardial metabolism and coronary vasodilators.

Antiarrhythmic drugs suppress abnormal rhythms of the heart and are widely classified into categories based on their effects on the cardiac action potential and, consequently, on the electrophysiological properties of the heart. In Table 6 only mechanisms acting on the membranes are listed.

Table 6. Classification of antiarrhythmic drugs

Class	Mechanism of action	Primary sites of action
IA	Na ⁺ channel blocking intermediate association/dissociation	Atrial and ventricular tissue
IB	Na ⁺ channel blocking fast association/dissociation	Ventricular tissue
IC	Na ⁺ channel blocking slow association/dissociation	Ventricular tissue
II	β -adrenergic receptor blocking	SA and AV node
III	K ⁺ channel blocking	Atrial and ventricular tissue
IV	Ca ²⁺ channel blocking	SA and AV node

Diuretics

Diuretics are chemicals that elevate the rate of urination. Increased urine flow rate leads to increased excretion of electrolytes (especially Na⁺ and Cl⁻) and water from the body without affecting protein, vitamin, glucose, or amino acid reabsorption. These pharmacological properties have proven effective in the treatment of a wide range of clinical disorders, including oedematous conditions resulting from a variety of causes e.g. congestive heart failure, nephrotic syndrome and chronic liver disease, and in the management of hypertension. Diuretics include osmotic diuretics, carbonic anhydrase inhibitors, thiazide and thiazide-like diuretics, loop diuretics, potassium-sparing diuretics and aldosterone antagonists.

Angiotensin-converting enzyme inhibitors, angiotensin II receptor blockers and calcium channel blockers

Angiotensin-converting enzyme (ACE) inhibitors are primarily used for the treatment of hypertension (high blood pressure) and congestive heart failure. These compounds effectively block the conversion of angiotensin I to angiotensin II.

Calcium channel blockers are compounds with diverse chemical structures which block the inward movement of Ca²⁺ through slow cardiac calcium channels.

Central and peripheral sympatholytics and vasodilators

Sympatholytics are used to treat various conditions, including hypertension and different types of anxiety. Sympatholytic drugs can block the sympathetic adrenergic system at three different levels. Drugs that block sympathetic activity within the brain are called centrally acting sympatholytic drugs, like α_2 -adrenergic antagonists. Peripheral sympatholytic drugs, such as β -adrenergic receptor blockers, α_1 -adrenergic blockers and mixed α/β -blockers prevent the influence of norepinephrine at the effector organ (heart or blood vessel).

Vasodilator drugs relax the smooth muscle in blood vessels, causing the vessels to dilate and are used to treat hypertension, heart failure and angina.

Antihyperlipoproteinemics and inhibitors of cholesterol biosynthesis

Antihyperlipoproteinemics are agents that promote a reduction of lipoprotein levels in the blood by inhibiting the enzyme HMG-CoA Reductase, which is involved in the rate limiting step in the synthesis of cholesterol.

Anticoagulants

Anticoagulant drugs reduce the ability of the blood to form clots by blocking the action of clotting factors or platelets. Anticoagulant drugs fall into three categories: inhibitors of clotting factor synthesis, inhibitors of thrombin and antiplatelet drugs.

Coumarin derivatives and 1.3 indandiones are orally active anticoagulants and heparin-based anticoagulants are administered parenterally.

Antiplatelet drugs regulate blood coagulation and subsequent thrombus formation at the platelet level through a number of different mechanisms.

2.6.2 Compounds analysed

Amlodipine belongs to the group of antianginal drugs, is also a long term calcium channel blocker and belongs structurally to the dihydropyridins. Less than 4% of the unchanged drug is excreted in urine.

Lidocaine is primarily used as a local anaesthetic but can be used as an effective antiarrhythmic agent of the IB class if given parenterally. It is primarily subjected to rapid first pass metabolism in the liver followed by renal excretion of the unchanged drug (<10%) and metabolites.

Amiloride is a potassium sparing diuretic frequently combined with hydrochlorothiazide in a fixed-dose combination. Amiloride has the structural characteristics of an aminopyrazine and approximately 50% is excreted unchanged.

Bendroflumethiazide and *hydrochlorothiazide* are thiazide diuretics. They are not extensively metabolised and are primarily excreted unchanged in the urine.

Furosemide is a loop diuretic which may be regarded as a derivative of anthranilic acid or o-aminobenzoic acid and is excreted primarily unchanged.

Enalapril and *perindopril* are ACE inhibitor pro-drugs which are metabolised *in vivo* to their active forms *enalaprilat* and *perindoprilat*. The bioavailability of enalapril and perindopril are 60% and 60–95% respectively. Enalaprilat is excreted unchanged, in contrast to perindoprilat which is extensively metabolised.

Candesartan and *losartan* are angiotensin II receptor blockers. *Candesartan* is a structural biphenyl analogue of *losartan* and both are primarily (80%) excreted unchanged.

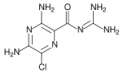
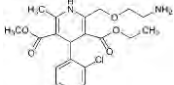
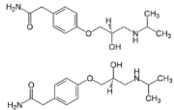
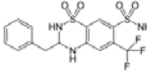
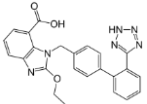
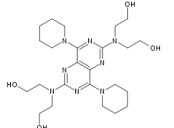
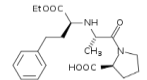
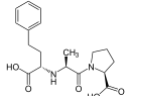
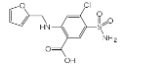
Atenolol and *metoprolol* belong to the group of peripherally acting sympatholytics and are classified as β -adrenergic receptor blockers. 50% of *atenolol* is excreted unchanged via the feces, whereas only less than 5% of *metoprolol* is excreted unchanged via urine.

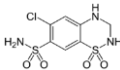
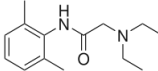
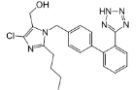
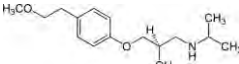
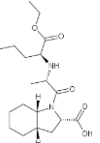
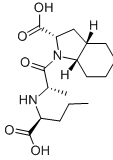
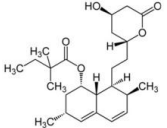

Dipyridamole is a pyrimidopyrimidine derivative with vasodilating and antiplatelet properties and works as a phosphodiesterase inhibitor. 20–30% of *dipyridamole* in plasma is present as metabolites, mainly as a monoglucuronide (www.fass.se).

Simvastatin is an inactive pro-drug that must undergo *in vivo* hydrolysis in order to produce its hypolipidemic effect. Major elimination routes are via feces and urine, with 60% and 13% excretion, respectively.

Warfarin is a coumarin derivative acting as an anticoagulant. It is highly metabolised and therefore almost no unchanged drug is excreted in the urine.

Table 7. Cardiovascular drugs selected for this study

Compound	Class	Structure	CAS No	ATC No
Amiloride	Diuretic Potassium-sparing diuretic		2016-88-8 (HCl) 2609-46-3	C03EA01
Amlodipine	Calcium channel blocker		88150-42-9	C08CA01 C09DB01 C09DX01 C09DB02
Atenolol	Peripherally acting sympatholytic β-adrenergic receptor blocker		29122-68-7	C07AB03
Bendroflu-methiazide	Diuretic Thiazide diuretic		73-48-3	C03AB01
Candesartan	Angiotensin II receptor blocker		139481-59-7	C09CA06
Dipyridamole	Antiplatelet drug Phosphodiesterase inhibitor Coronary vasodilator		58-32-2	B01AC07
Enalapril	ACE inhibitor Dicarboxylate-containing inhibitors		75847-73-3	C09BA02 C09AA02
Enalaprilat	ACE inhibitor Dicarboxylate-containing inhibitors Active metabolite of Enalapril		76420-72-9	
Furoseamide	Diuretic Loop diuretic		54-31-9	C03CA01

Compound	Class	Structure	CAS No	ATC No
Hydrochloro-thiazide	Diuretic (first –line) Thiazide diuretic		58-93-5	C03EA01 C09XA52
Lidocaine	Antiarrhythmic agent Class IB Local anaesthetic		73-78-9 137-58-6	N01BB20 N01BB02 QN01BB52 N01BB52 C05AA01
Losartan	Angiotensin II receptor blocker		114798-26-4	C09CA01 C09DA01
Metoprolol	Peripherally acting sympatholytic β-adrenergic receptor blocker		51384-51-1	C07AB02
Perindopril	ACE inhibitor Dicarboxylate-containing inhibitors		82834-16-0	C09AA04
Perindoprilat	ACE inhibitor Dicarboxylate-containing inhibitors Active metabolite of Perindopril		95153-31-4	
Simvastatin	Hypolipidemic		79902-63-9	C10AA01 C10BA02
Warfarin	Anticoagulant Coumarin derivative		81-81-2	B01AA03

2.7 Hormones

2.7.1 *Scope and definition*

Hormones are chemical substances released into the bloodstream by a cell or a gland in one part of the body and affects tissues or organs in other parts of the organism. Their effects appear slowly over time and affect many different processes, including growth and development, metabolism, sexual function, reproduction and mood. Extremely low concentrations can cause big changes in cells or even the whole body and unbalanced hormone levels can therefore have serious consequences. There are two major classes of hormones: (1) steroids (hydrophobic molecules) and proteins (2) peptides and modified amino acids (hydrophilic molecules).

The *sex hormones* are specific steroids necessary for reproduction as well as for the development of secondary sex characteristics in both sexes. The sex steroids are comprised of three classes: oestrogens, progestins and androgens.

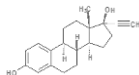
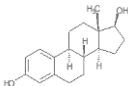
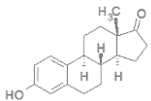
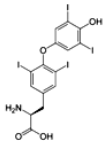
Thyroid hormones are iodinated amino acids derived from L-tyrosine in the thyroid gland and are primarily responsible for metabolism regulation. An under- or over- active thyroid gland results in hypo- or hyperthyroidism, which is treatable with naturally or synthetically produced thyroid hormones.

2.7.2 *Compounds analysed*

Four female sex hormones were investigated. These included the naturally produced oestrogens *estrone (E1)*, *estriol (E2)* and *17 β -estradiol (E3)*, and the synthetically produced estradiol derivative *17- α -ethinylestradiol (EE2)*. EE2 is used in almost all modern formulations of combined oral contraceptive pills and is one of the most commonly used medications.

Beside the sex hormones, *levothyroxine*, a synthetic iodinated amino acid thyroid drug for treatment of hypothyroidism was analysed.

Table 8. Hormones selected for this study

Compound	Class	Structure	CAS No	ATC No
17-a-Etinylestradiol (EE2)	Sex hormone Estrogen		57-63-6	
17-β-Estradiol (E2)	Sex hormone Estrogen		50-28-2	G03FA01 G03HB01 G03CA03
Estriol (E3)	Sex hormone Estrogen		50-27-1	QG03CA04 G03CA04
Estrone (E1)	Sex hormone Estrogen		53-16-7	
Levothyroxine	Thyroid hormone Synthetic thyroxine		51-48-9	H03AA01

2.8 Hypnotics

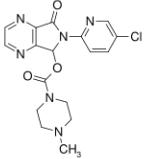
2.8.1 Scope and definition

Hypnotics are a class of drugs causing drowsiness and facilitate the initiation and maintenance of sleep. They are often referred to as sleeping pills and are applied to treat insomnia. The observed pharmacological effects of most drugs in this class are usually dose-related, step-wise inducing sedation, hypnosia and finally surgical anaesthesia. The hypnotic drugs are not characterised by common structural features but instead, a variety of chemical compounds have been used in clinical therapy.

2.8.2 Compounds analysed

Zopiclone is a non-benzodiazepine GABA_A agonist used as a short acting sedative hypnotic. The oral bioavailability is 80% and due to its fast metabolism only less than 10% is excreted as unchanged drug.

Table 9. Hypnotics selected for this study

Compound	Class	Structure	CAS No	ATC No
Zopiclone	nonbenzodiazepine GABA _A agonist		43200-80-2	N05CF01

2.9 Additives in personal care products

2.9.1 Scope and definition

Additives to personal care products are a class of new emerging contaminants that have raised concern in recent years. These compounds deserve attention because of their continuous introduction into the environment via effluents from sewage systems.

Personal care product additives are usually classified according to common properties as for instance surfactants, bactericides, UV-filters and antioxidants.

Bactericides are common additives used as preservatives of the product. The environmental concerns regarding additives in personal care products are due to their high-volume use and for several compounds due to their reported ecotoxicological effects.

One common feature of additives in personal care products and their metabolites are that they are transported with the sewage system and if they are not efficiently removed at a WWTP, they are discharged into receiving waters. The environmental risk these substances pose to the environment is not clear but could negatively impact the health of the ecosystem and humans.

Today there are numerous publications which show that the effluent from the WWTP as well as the surface water in the receiving water contains large number anthropogenic compounds, including additives in personal care products (Ternes, 1998; Stumpf et al., 1999; Kolpin et al., 2002).

Several studies have also identified these compounds in drinking water (Ternes et al., 2002).

2.9.2 Compounds analysed

Ethylene diamine tetraacetic acid (EDTA) is a strong complexing agent used in many cosmetic products as a stabiliser by chelating metal ions such as Fe. EDTA is also used in developer for photographic and X-ray film. In the Nordic countries the greatest use is in the pulp and paper industries. Free acid of EDTA do not exist in the environment. Under normal environmental conditions EDTA occurs in complexes with different metal ions depending on the equilibrium constant, exchange rate, pH and ion strength (Nowack, 2002; Hering et al., 1988). The different complexes have different configurations in space. Therefore, for simplicity, the chemical structure of the free acid of EDTA is presented in Table 10.

Diethyl phthalate (DEP) is a plasticizer added to plastics to increase their flexibility and is widely used in tools, automotive parts, tooth-brushes, food packaging, cosmetics and insecticide.

Phthalates have been shown to be endocrine disruptors (weak oestrogen mimics, inhibiting molting of *Daphnia magna*) (Jobling et al., 1995; Zou and Fingerman 1997). The most frequent use of DEP is in cosmetics and personal care products, principally as solubilizer in perfumes and as an alcohol denaturant. DEP is also used in hair preparations.

Butylparaben (BuP), with IUPAC name butyl 4-hydroxybenzoate, is a preservative agent used in personal care products. BuP is used as a flavouring agent or preservative in some foods (not EU), cosmetics and drugs. The preserving action of BuP stems from its ability to disrupt membrane transport properties and it is added to retard microbial growth (Toxnet <http://toxnet.nlm.nih.gov/>). In 2003, butylparaben was cleared to be used as a flavour additive in food by the FAO and the WHO, but butyl paraben is not among the food additives presently listed with acceptable uses in CODEX alimentarius (<http://www.codexalimentarius.net/gsfaonline/additives/index.html?lang=en#H>).

Sodium dodecyl sulphate (SDS) is a anionic detergent, used in soaps and shampoos as it is efficient for sebum removal (along with dead skin cells, dirt, and the bacteria living on it) (Emsley, 2007).

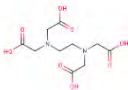
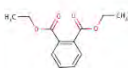
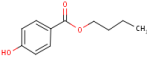
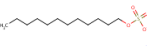
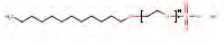
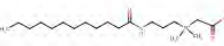
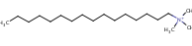
The detergent *sodium laureth sulphate* (SDEO1-4) is used in soaps and shampoos as a sebum removal along with dead skin cells, dirt, and the bacteria living on it (Emsley, 2007). It has a better water solubility than SDS at low temperatures and is therefore the preferred detergent in soaps and shampoos.

Cocoamidopropyl betaine (CAPB) is a cationic surfactant of the quaternary ammonium compound (QAC) kind, which form an important class of industrial chemicals. Because of their physical and chemical properties QAC are used as disinfectants, surfactants, anti-electrostatics

(e.g. in shampoo), and phase transfer catalysts. They have the capacity to attach themselves onto particle e.g. in the WWTP and sediment high concentration is therefore reported in these matrixes. QACs bioavailability is assumed to be low (e.g. Remberger et al., 2006).

Cetrimonium salts (ATAC -C16) belong to a group of compounds commonly known as alkyltrimethylammonium chlorides (ATAC), which is widely used as surfactant, bactericide, and algacide (Ding and Tsai, 2003).

Table 10. Additives in personal care products selected for this study

Compound	Class	Structure	CAS No
EDTA	Complexing agent		60-00-4
Diethyl phthalate (DEP)	Plasticizer		84-66-2
Butylparaben (BuP)	Biocide		94-26-8
Sodium dodecyl sulphate (SDS)	Surfactant		151-21-3
Sodium laureth sulphate (SDSEO-1-4)	Surfactant		9004-82-4
Cocoamidopropyl betaine (CAPB)	Surfactant		7292-10-8
Cetrimonium Br-salt (ATAC-C16)	Surfactant		57-09-0

2.10 Use of PPCPs in Faroe Island, Iceland and Greenland

The selection of pharmaceuticals included in the analytical scheme was based on a survey of the pharmaceuticals used in the three countries in 2010. The assessment was based on the number of defined daily doses, DDD (Table 11) which refers to the mass of defined substance administered per day to 70 kg adult according to WHO (2012). The data from Greenland is based on the volume of pharmaceuticals, bought to the National Pharmacy for distribution in Nuuk only, and thus not representative of the entire country. However, approximately one third of the population in Greenland resides in Nuuk, and the application of pharmaceuticals here is also most relevant for the present study which involves Greenlandic samples taken in Nuuk only. Though, as the application data is based on supplements to the pharmacy the Greenlandic data are somewhat less precise regarding actual use in 2010 than the Faroese and Icelandic data which are based on actually administered pharmaceuticals. The use of pharmaceuticals in relative number of DDD administered in 2010 does not say much about the volumes of pharmaceuticals used. However, it does tell us what compounds are being used frequently these days and as such was important for deciding which pharmaceuticals to include in the screening. In order to get a more precise quantitative view on the pharmaceuticals use, two more parameters are needed; first and foremost the volume of a DDD which may vary considerably between the pharmaceuticals, and of course, the actual and not just relative number of DDD used. To make a quantitative budget of the pharmaceuticals is outside the scope of the present work. For comparison purposes it may be useful to note that the volumes of DDD are very different, with DDD for paracetamol, ibuprofen, metformin and acetyl salicylic acid as painkiller in the range 2 to 3 g, whereas the DDD for citalopram is just approximately one hundredth of this, at 0.02 g. Also the DDD of the antidepressant venlafaxine is much lower than that of paracetamol, approximately 1/30, and the cardiovascular drugs atenolol and metoprolol are prescribed with comparatively small DDDs which are 0.075 and 0.15 g, respectively (WHO 2012).

Table 11. The relative volume of pharmaceuticals used in Greenland (Nuuk), Faroe Islands and Iceland in 2010 are shown as the ranked number of defined daily doses, DDD. The various pharmaceuticals have been given a colour to facilitate visual comparisons across countries

ATC*	Greenland (Nuuk)	Faroe Islands	Iceland
C09AA02	Enalapril	Amlodipine	Acidum acetylsalicylicum
C08CA01	Amlodipine	Enalapril	Simvastatin
C10AA01	Simvastatin	Acetylsalicylic acid	Zopiclone
A02BC01	Omeprazole	Simvastatin	Omeprazole and esomeprazole
N02BE01	Paracetamol	Paracetamol	Ibuprofen
C03AB01	Bendroflumethiazide and kalium	Bendroflumethiazide and kalium	Citalopram/escitalopram
B01AC06	Acetylsalicylic acid	Furosemide	Amlodipine
G03AC08	Etonandestrel	Omeprazole	Losartan
G03AA09	Desandestrel and estranden	Metoprolol	Enalapril
A11DA01	Thiamine (vitamin b1)	Candesartan	Hydrochlorothiazide
M01AE01	Ibuprofen	Gliclazide	Amiloride
N06AB04	Citalopram	Atorvastatin	Progestogen
A11AA03	Multivitamines and other minerals, incl. comb.	Citalopram	Paracetamol
C07AB02	Metoprolol	Esomeprazole	Furosemide
G03AA07	Levonorgestrel and estranden	Zopiclone	Levothyroxine natrium
D07AB02	Hydrocortisonbutyrat	Ibuprofen	Atenolol
R03BA02	Budesonide	Levothyroxine natrium	Levonorgestrel and estrogen
R03AC03	Terbutalin	Sertraline	Sertraline
C09CA01	Losartan	Isosorbidmononitrat	Diclofenac
C03CA01	Furosemide	Felodipin	Nicotine

Source: National Pharmacy Greenland, Chief Pharmaceutical Officer Faroe Islands and Icelandic Medicines Agency.

*ATC code refers to pharmaceutical listed in the Greenland (Nuuk) column.

3. Methodology

3.1 Sampling sites and sample selection

Because the study only includes pharmaceuticals used by humans and personal care substances, it was a natural choice to confine sampling to sites where sewage water from urban areas as well as hospitals is discharged to the recipient. Also, in areas where waste water treatment plants, WWTP's, are in place, it was chosen to analyse the sewage water on various sites in the treatment process. Thus, when sampling was done at WWTP's, samples were taken of waste water as it entered the WWTP that is influent water, and it was taken after purification in the WWTP, that is effluent water and this represents the water as it is discharged to the recipient. Also, samples were taken from sludge in the WWTP and, when possible, from sediments and water in the recipient. The rationale for sampling influent and effluent water was to get a glimpse of the effectivity of the purification process although the simplified nature of the sampling method does not allow rigorous conclusions.

In areas with no WWTP, as in Nuuk, Greenland, the sampling was done in the sewage line, SL, in sampling or maintenance wells. Also, in some cases, like with the waste water samples from Greenland and Fossvog Main Hospital in Iceland, sludge samples were taken from such wells. Waste water sampled in such wells with no subsequent purification step was classified as effluents to stress the facts that this water was discharged to the recipient without subsequent treatment.

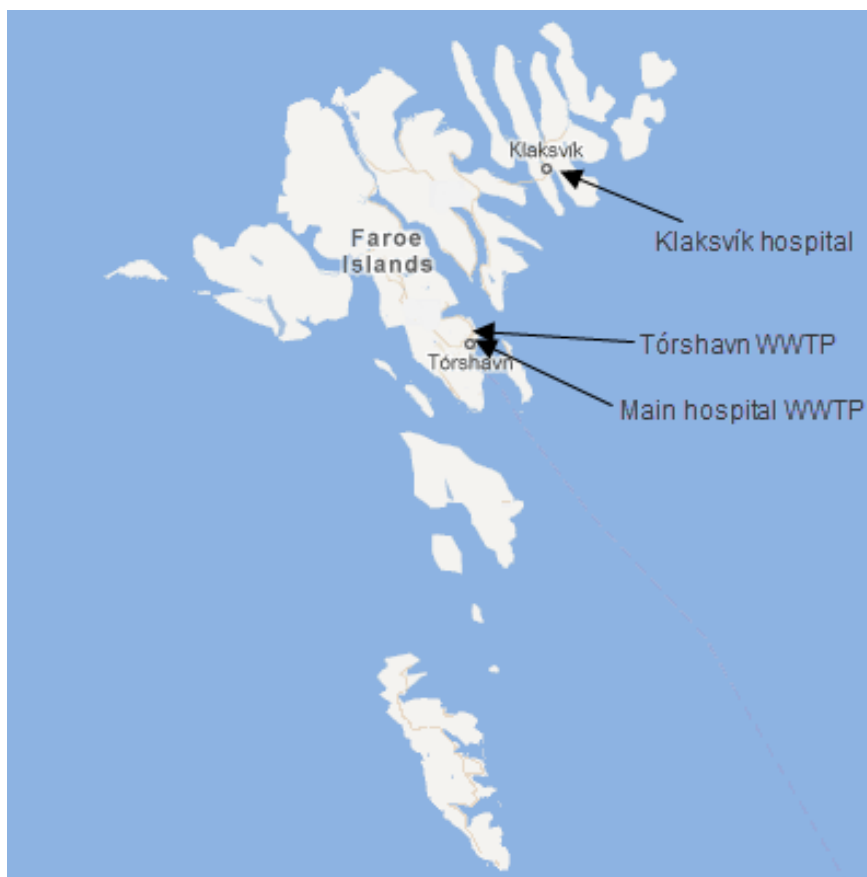
The project aim was to determine whether PPCP's are found in the environment, and therefore sampling were performed primarily in areas where such are most likely encountered, that is in the vicinity of capitals and/or other places with health care centres/hospitals (Table 12).

Table 12. Sampling sites overview

Country	Waste water treatment plants, WWTP, or sewage lines, SL.	Hospitals	Recipients
Faroe Islands	Torshavn WWTP (Sersjantvíkin)	Main Hospital Klaksvik Hospital	Torshavn Klaksvik
Iceland	Akureyri WWTP Reykjavik WWTP (Klettagørdum) Hveragerði WWTP	Fossvog Main Hospital	Akureyri Reykjavik Reykjavik: Fossvog Main Hospital
Greenland	Kolonihavnen SL	Sana (Queen Ingrid's) Main Hospital	Kolonihavnen Iggia Queen Ingrid's Hospital

3.1.1 Faroe Islands

Figure 1. Sampling sites in the Faroe Islands



Torshavn WWTP (Sersjantvíkin)

This sewage treatment plant is the main WWTP in Torshavn. It is situated in Sersjantvíkin, and is below ground. The treatment plant may be described as consisting of primary purification, where the treatment is composed of filtering followed by natural decay in a large tank, with residence time approx. ½ year. Samples were taken of influent to and effluent from the WWTP, in addition to surface sludge from the sedimentation tank. Samples of surface water in the recipient were taken approx. 10 m from the discharge site.

Main Hospital WWTP

The Main Hospital (Landssjúkrahúsið, www.lsh.fo) is situated in Torshavn and has a staff amounting to approx. 670 man-years. It provides 180 hospital beds, and performs approx. ½ million clinical chemical analyses per year, in addition to more than 30,000 x-ray diagnostic analyses. The hospital has its own sewage treatment plant. It is in principle of the same outline as the Torshavn (Sersjantvíkin) WWTP, but in addition it contains a bio filtering sprinkler system. Also, the main hospital WWTP sedimentation tank was open. Since the sampling in the main hospital WWTP in late September 2010, the old WWTP has been replaced with a new and closed one. Samples were taken of the influent and the effluent of the WWTP as well as of surface sludge in the sedimentation tank (Figure 2) Samples were taken of surface water in the recipient, approx. 10 m from the site of discharge.

Figure 2. The Main Hospital (Faroe Islands) WWTP



Upper left: The open air sludge tank. Upper right: Intake into WWTP near arrow, this is where influent water was sampled. Lower left: The sprinkler adds water to the bio filter following the passage of the sludge sedimentation tank. Lower right: The well where effluent was sampled. Water enters the well from the bio filter to the right in the picture, and runs to the recipient to the left.

Klaksvik Hospital SL

Klaksvik hospital is situated in Klaksvik in the northern part of the Faroe Islands with staff amounting to approx. 100 man-years. It has 36 hospital beds, and performs approx. 135,000 clinical chemical analyses per year, in addition to approx. 5,500 x-ray diagnostic analyses. The hospital does not have its own sewage treatment plant, and thus the sampling was done in the sewage line and in the recipient Klaksvik harbour, but at a site representing the inner harbour at large and not in the close proximity to the discharge site.

Figure 3. Klaksvik Hospital. The sewage line was accessed through a manhole at the parking lot



Recipient Torshavn harbour

Torshavn harbour and nearby areas are recipient for domestic and hospital waste waters, for shipyard activity, some food-processing waste waters and it harbours a marina. In addition to the sampling of surface water from the recipient of the Main Hospital WWTP and Torshavn WWTP (Sersjantvíkin) described above, sampling of surface water and sediments were done in Torshavn harbour near the shipyard and near the marina.

Recipient Klaksvik harbour

Klaksvik harbour is recipient for domestic waste water, hospital wastewater, food-processing waste water and a small shipyard. In addition, there is a marina in the harbour. Sediment samples were taken at the site of Stongina. Surface water of the recipient was taken near the marina close to the foot of the bay.

3.1.2 Iceland

Figure 4. Sampling sites in Iceland



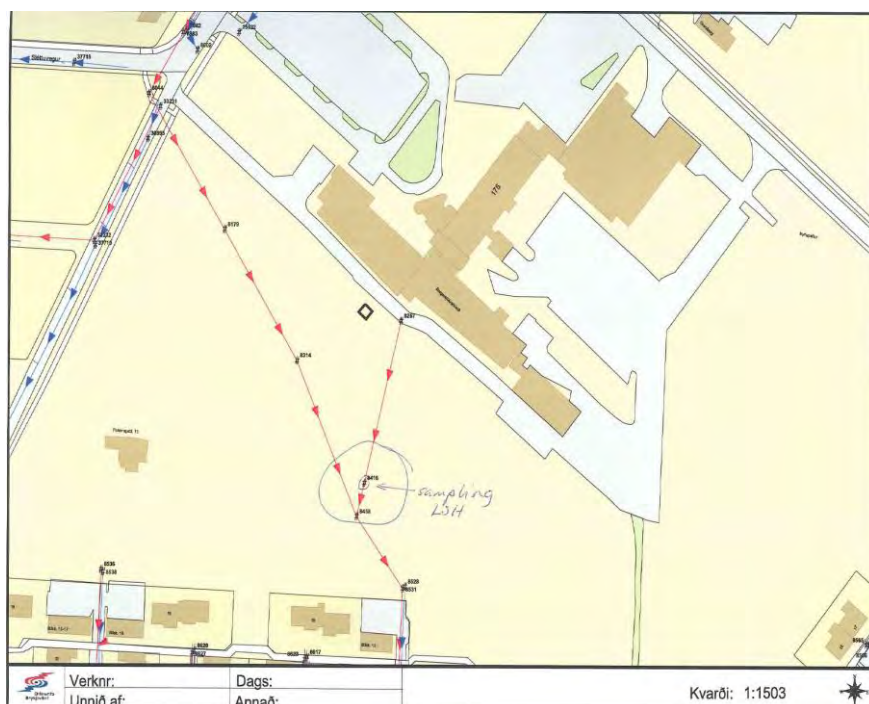
Reykjavik WWTP (Klettagörðum)

The WWTP receives sewage from Reykjavik, Kópavogur and Álftarnes, in all approx. 160,000 inhabitants. The WWTP includes two pumping stations, and sewage is filtered in several steps before discharged to Faxaflói at a depth of approx. 30 m. The WWTP receives waste waters from one major hospital, several health clinics, industry, production and households. Samples were taken of influents (two parallel samples), of effluents and of sludge in the WWTP.

Main Hospital SL (Fossvog)

The Main Hospital Iceland (www.lsh.is, data for 2010) consists of two units situated in Reykjavik, one in Hringbraut and one in Fossvog (the smaller). Combined in these two units are staff equivalent to approx. 3,650 man-years. The laboratories provide 1.2 mill. clinical chemical analyses per year, and 120,000 diagnostic imaging procedures are performed. Samples were taken of waste water in a maintenance well (Figure 5).

Figure 5. Sampling from the sewage line at Main Hospital (Fossvog) Iceland was done in manhole. This sample is classified as influent as it feeds into a WWTP



Hveragerði WWTP

The Hveragerði WWTP receives sewage from Hveragerði and whereabouts with approx. 2,000 inhabitants. The WWTP consists of one pumping stations, several filter steps, biodegradation, followed by filtration on outdoor gravel-bed before discharge of effluent into the Varmá river. The WWTP receives waste waters from a health clinic and spa facilities in addition to domestic sewage. The sampling included influent, effluent, sludge from the pumping station and sludge from the outside gravel beds.

Figure 6. The snow-covered gravel bed outside the Hveragerði WWTP where the canals are periodically flooded and sludge accumulates



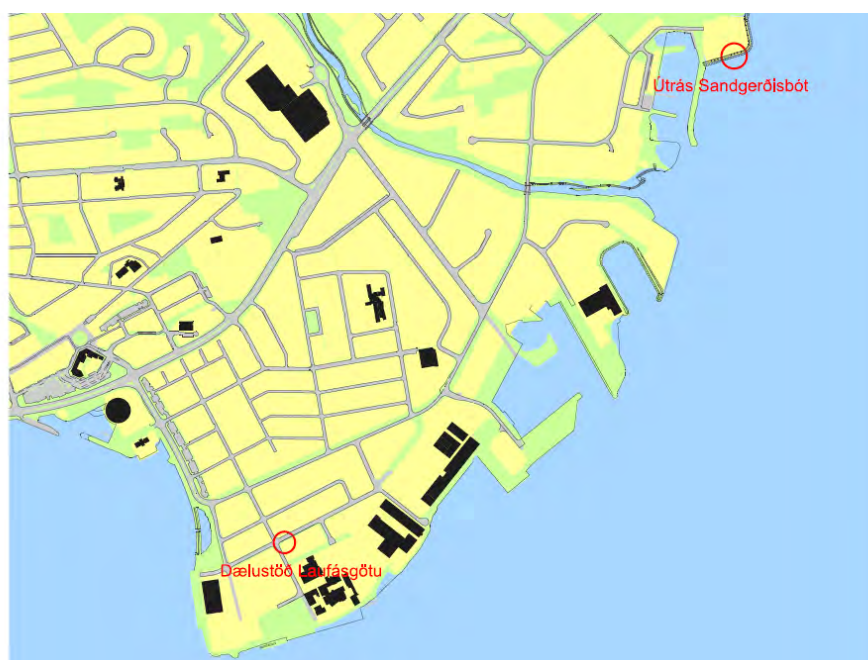
Figur 7. Hveragerði WWTP



Akureyri WWTP

The WWTP receives sewage from Akureyri with approx. 20,000 inhabitants. The WWTP consists of one pumping stations and filtering steps. The effluents are discharged to the sea. The WWTP receives waste water from a hospital, a health clinic, industry, production as well as domestic sewage. Samples included influent and effluent water, as well as sludge from the pumping station (Laufásgata). Sludge/sediments (referred to as sediments) were taken at the shore near the discharge point from the WWTP (Útrás Sandgerðisbót, Figure 8). Due to the small sample size from this location, all personal care substances but only a selection of pharmaceuticals were analysed in this.

Figure 8. Sampling sites in Akureyri waste water treatment system are shown.



3.1.3 Greenland

Kolonihavnen SL

Waste water stems mainly from households and in minor degree from hotels, restaurants, shops and office buildings. The sewage line (U11) serves in all approx. 4,351 person equivalents.

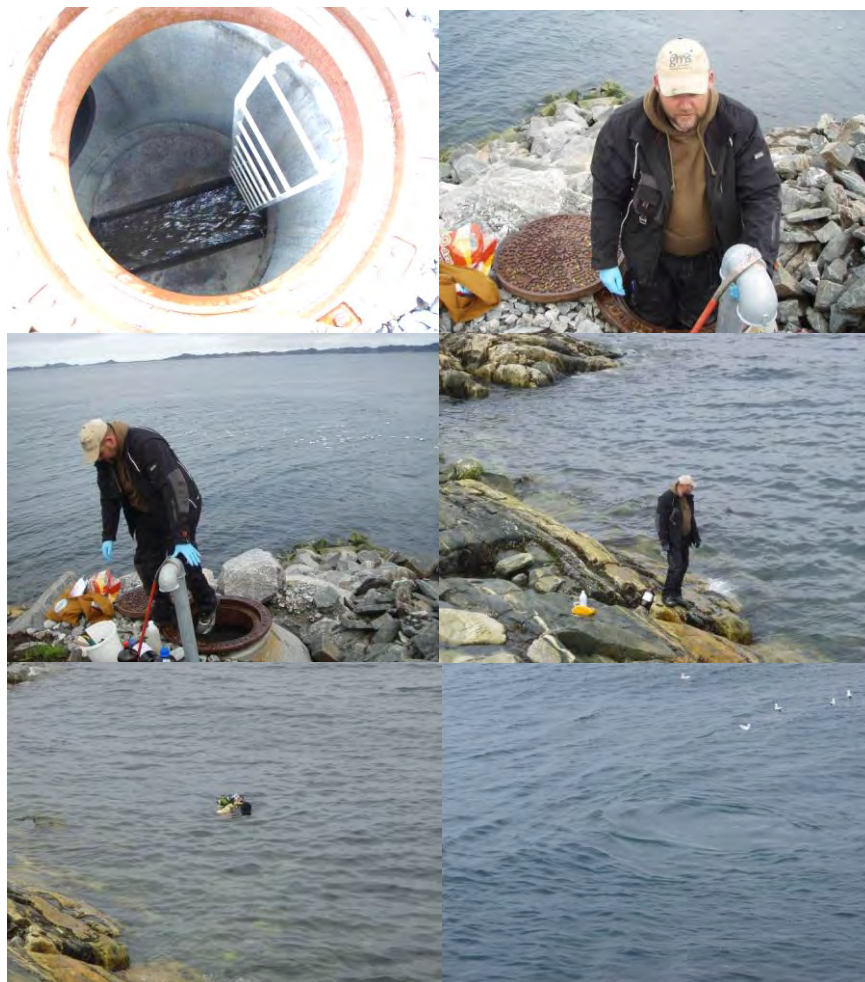
Samples of sludge and waste water in a maintenance well, and of water and sediments in the recipient were taken in July 2011 around 9–10 am.

Recipient water was taken approx. 2 m from the discharge site, at 6 m depth. Samples in the recipient were taken by a diver, who collected the samples directly into the sample containers provided by the laboratories.

Figure 9. Sampling sites in Nuuk, Greenland



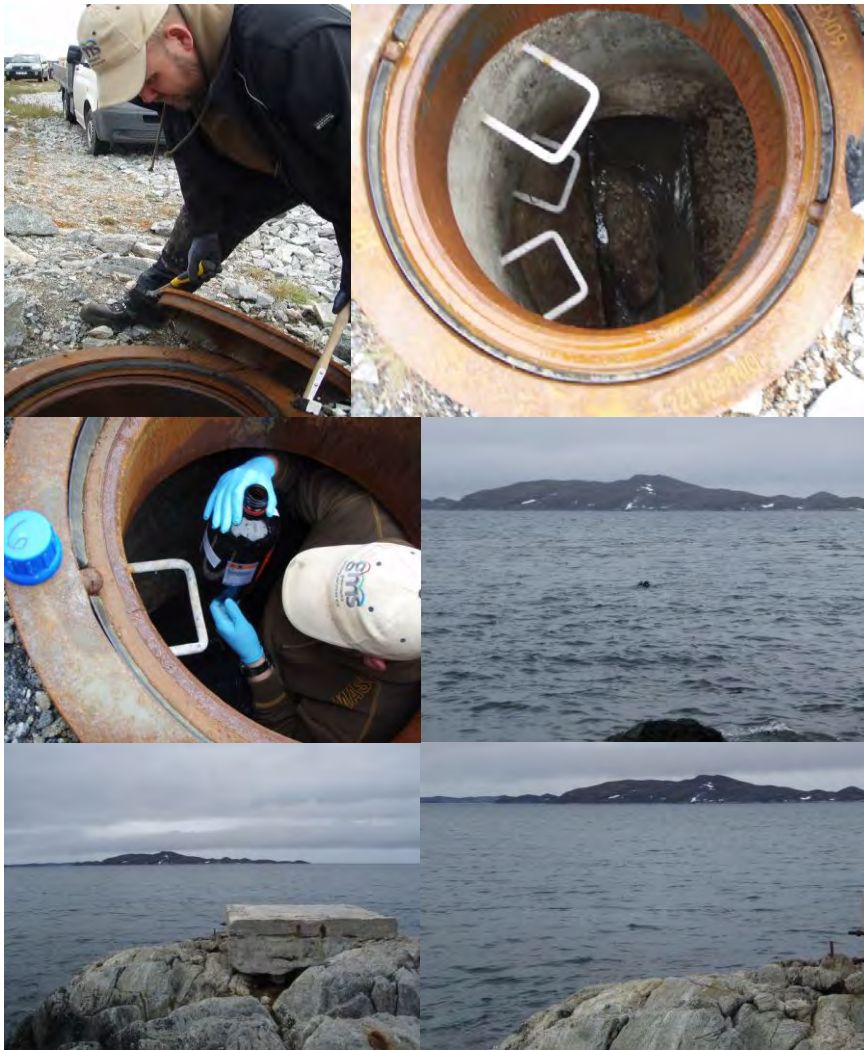
Figure 10. Sampling in the Kolonihavnen SL U11. The picture in the lower right corner shows where the waste water discharge below the water surface



Queen Ingrid's Hospital SL (Sana)

Queen Ingrid's Hospital (Sana) has staff amounting to 467 man-years. The hospital has 191 beds and performs 780,000 clinical chemical analyses and 12,000 x-ray analyses per year. Sewage from the hospital is led in a sewage line U7 which serves in all 2,935 PE. Samples were taken in the recipient at approx. 3.5 m depth and approx. 50 m from the discharge site in November 2010 using the water sampler as shown in Figure 14. In July 2011, samples of waste water and sludge were taken in a maintenance well of the SL (U7). In the recipient, samples of water and sediments were taken approx. 2–3 m from the discharge site, at a depth of approx. 9 m.

Figure 11. Sampling in the Queen Ingrid's Hospital SL (U7) in July 2011

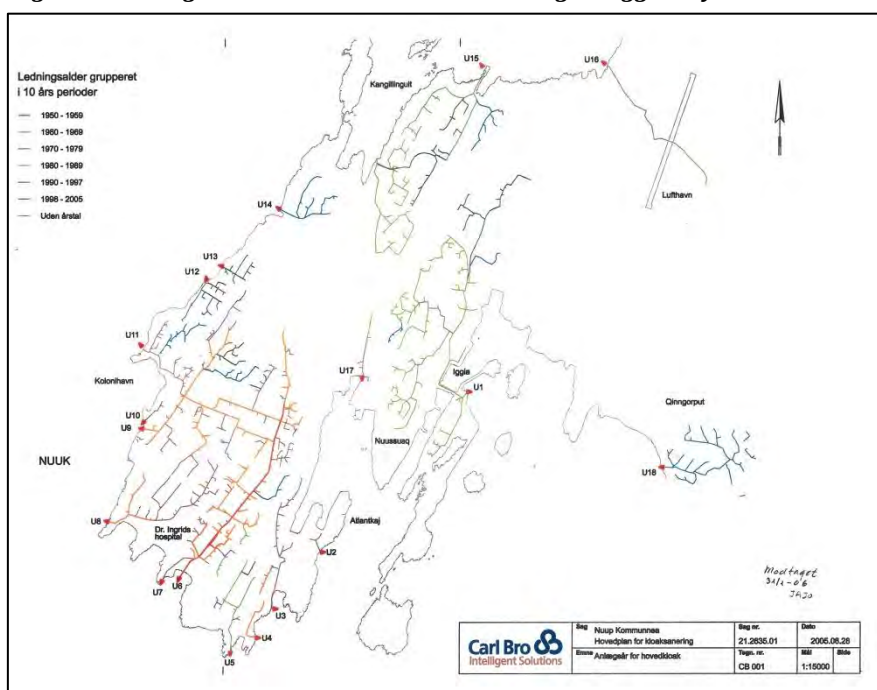


Iggia SL

Iggia SL (U1 Figure 12) combines waste water mainly from households and a brewery/bottling facility and serves a total of 4,640 PE. The waste water is discharged to sea in the tidal zone of the bay.

Samples were taken in the recipient at approx. 3.0 m depth and close to the discharge site in November 2010. In 2011, surface water samples were taken in the recipient at a distance from the discharge site of approx. 10 m (Figure 15).

Figure 12. Sewage lines in Nuuk. The SL U1 discharge to Iggia bay



3.2 Sampling methods

3.2.1 *Influent*

Influent water was sampled by immersing the provided bottles directly into the water stream, wearing disposable laboratory (nitrile or latex no-powder) gloves.

3.2.2 *Effluent*

Effluent water was sampled by immersing the provided bottles directly into the water stream, wearing disposable laboratory (nitrile or latex no-powder) gloves.

Samples taken in sewage lines with no waste water treatment are labelled effluent, even though these have not been subject to purification. The term effluent is used to stress that this is the quality of the water as it enters the recipient.

3.2.3 *Surface water recipient*

In Faroe Islands, the bottles were immersed by glove-clad hand or by string into the water.

Figure 13. Surface water in recipient sampling in Torshavn harbour



Figure 14. Recipient water sampling tool used in Greenland in the 2010 sampling



Figure 15. Recipient water sampling in Iggia U1 recipient in July 2010



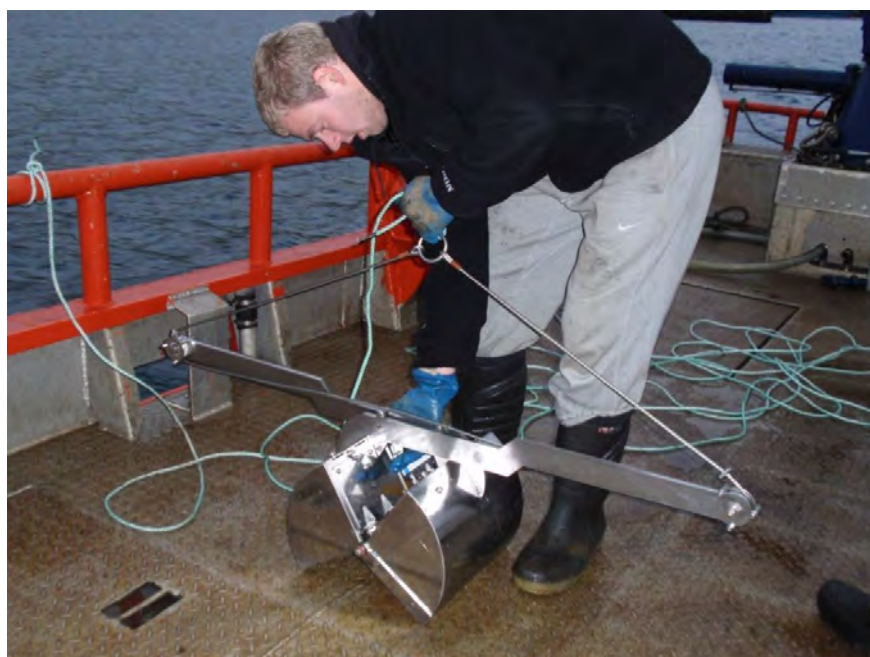
3.2.4 Sludge

Sludge in Faroe Island was sampled using clean (washed in dishwasher detergent and rinsed with acetone and finally with purified water) stainless steel spoons. In Iceland, the sludge was scooped up directly into the sample jar. In Greenland, sludge was sampled by glove clad hand, using the latex gloves provided by the laboratories.

3.2.5 Sediment

Sediment was sampled in Faroe Islands only, in Torshavn and Klaksvik harbours, using a van Veen grab. The samples consist of the uppermost approx. 2 cm of sediments. In Greenland, sediment samples were scooped directly into the sample containers provided by the laboratories.

Figure 16. The van Veen grab is prepared for sediment sampling in Klaksvik harbour. A similar grab was used in Torshavn harbour



4. Analysis methods

4.1 Pharmaceuticals

4.1.1 Chemicals

Native acetylsalicylic acid, amiloride hydrochloride, amlodipine, atenolol, rac bendroflumethiazide, candesartan, S-citalopram oxalate, diclofenac sodium salt, dipyridamole, (S,S,S)-enalapril maleate, enalaprilat dihydrate, omeprazole, 17 β -estradiol, estriol, estrone, 17 α -ethinylestradiol, fluoxetine hydrochloride, furosemide, gliclazide, hydrochlorothiazide, rac ibuprofen, thyroxine sodium salt, lidocaine hydrochloride monohydrate, losartan potassium salt, metformin hydrochloride, rac metoprolol hemi (+)-tartrate, rac naproxen, paracetamol (acetaminophen), paroxetine hydrochloride, perindopril t-butylamine salt, perindoprilat, sertraline hydrochloride, simvastatin, D,L-venlafaxine hydrochloride, warfarin and zopiclone were purchased from TRC and sulfamethizole from Sigma, all of 98% purity. Isotope-labelled compounds used as surrogate standard mixes and some as well as volumetric standards were purchased from TRC: d₄-amlodipine maleic acid salt, d₅-candesartan, d₆-citalopram oxalate, d₄-diclofenac, d₅-(S,S,S)-enalapril maleate, d₃-estriol, d₄-estrone, d₅-fluoxetine hydrochloride, d₃-rac ibuprofen, d₆-metformin hydrochloride, d₇-rac metoprolol, d₄-paracetamol (acetaminophen), defluoro paroxetine hydrochloride, d₃-rac sertraline hydrochloride, d₆-simvastatin, d₆-(D,L)-venlafaxine and d₈-zopiclone with 98% chemical purity and 99% isotopic purity; d₅-furosemide with 98.8% chemical purity and 98% isotopic purity and ¹³C,₂-hydrochlorothiazide with 97% chemical purity and 99% isotopic purity.

All solvents and reagents used in this work were of suprasolv, lichrosolv or pro analysis grade, and were purchased from Merck-Schuchardt (Hohenbrunn, Germany). Diethylhexylether (DHE), ammoniumformiate (NH₄HCO₂) and ammoniumacetate (NH₄OAc) were from Sigma Aldrich (Germany), ethanol from Arcus (Oslo, Norway). Ultra high purity water was delivered by a MilliQ Advantage A10 water purification system (Millipore, MA, USA).

4.1.2 Equipment

All not one-way equipment was base washed and rinsed with methanol before use.

4.1.3 Methods

Pre-treatment of water samples

The water samples were filtered through a pre-cleaned glass-fibre filter (GF/C, 8 h, 450°C), split up for the different analysis and stored in polypropylen bottles at -18°C until sample preparation.

Pre-treatment of sediment and sludge samples

The sediment and sludge samples were dried at 40°C until constant weight was reached, homogenised, sieved (2 mm, DIN 4,188), split up for the different analysis, packed in alumina foil and stored at -18°C until sample preparation.

Liquid phase micro extraction for acidic pharmaceuticals

Preparation of water samples

An aliquot of 240 ml sample was transferred to a glass bottle and fortified with an internal standard mixture (d_5 -furosemide, $^{13}C, d_2$ -hydrochlorothiazide and d_3 -ibuprofen). Adjustment to pH 2 was done by addition of 12M hydrochloric acid (HCl). The holofiber (Membrane, Wuppertal, Germany) was filled with an acceptor solution consisting of water-methanol 9:1, where the water was pH adjusted to pH 12 with aqueous ammonium hydroxide (NH_4OH). After transfer of the holofiber to the sample (donor) solution, the sample was stirred on a magnetic stirrer for 2 hours. Then the acceptor solution was quantitatively transferred into a total recovery vial and recovery standard (d_5 -candesartan) and 2 mM aqueous NH_4OAC solution were added.

Preparation of sediment and sludge samples

An aliquot of 1 g sample was transferred to a polypropylene tube and spiked with an internal standard mixture (d_5 -furosemide, $^{13}C, d_2$ -hydrochlorothiazide and d_3 -ibuprofen). One ml water pH 12 and 7 ml methanol were added to the sample, vortexed and sonicated for 15 min. After the centrifugation step the methanolic phase was carefully decant off, transferred to a glass bottle and diluted with 235 ml water. Adjustment to pH 2 was done by addition of 12M HCl. Following sample preparation steps were performed according the procedure for water samples.

Instrumental determination

The acidic pharmaceuticals (salicylic acid, furosemide, naproxen, warfarin, bendroflumethiazide, diclofenac and ibuprofen) were analysed by ultra-high pressure liquid chromatography triple-quadrupole mass spectrometry (UHPLC-MS/MS). Analysis was performed on a Thermo Scientific quaternary Accela 1250 pump with a PAL Sample Manager coupled to a Thermo Scientific Vantage MS/MS (Vantage TSQ). An injection volume of 10 μ L was used for sample separation on a Waters Acquity UPLC BEH C₁₈ column (2.1 \times 100 mm, 1.7 μ m) equipped with a Waters Van guard BEH C₁₈ guard column (2.1 \times 5 mm, 1.7 μ m). Separation was achieved using 2 mM aqueous NH₄OAc (A) and 2 mM NH₄OAc in methanol/acetonitrile 7:3 (B) as the mobile phase. The following gradient programme with a flow rate of 0.3 ml/min was applied: initial conditions 95% A/5% B for 30 sec, increased over 30 sec to 60% A/40% B, increased linearly over 180 sec to 40% A/60% B which was kept for 180 sec, then increased to 100% B over 240 sec and kept at 100% B for 120 sec, reduced to 95% A/ 5% B in 30 sec and finally equilibrated for 180 sec in a total run time of 16.50 min. Ionisation was performed in negative electrospray ionisation mode (ESI⁻). The MS was run in selected reaction mode (SRM) where two transitions of precursor ions from deprotonated molecular ions were monitored. For ibuprofen only one transition was available.

Liquid phase micro extraction for basic pharmaceuticals

Preparation of water samples

An aliquot of 240 ml sample was transferred to a glass bottle and fortified with an internal standard mixture (d₇-metoprolol, defluoro paroxetine, d₃-sertraline and d₆-venlafaxine). Adjustment to pH>12 was done by addition of 5M NaOH. The holofiber (Membrane, Wuppertal, Germany) was filled with an acceptor solution consisting of water/methanol 9:1, where the water was adjusted to pH 2 with formic acid. After transfer of the holo fibre to the sample (donor) solution, the sample was stirred on a magnetic stirrer for 2 hours. The acceptor solution was quantitatively transferred into a total recovery vial and recovery standard (d₄-paracetamol and d₅-candesartan) and 2 mM aqueous NH₄HCO₂ solution were added.

Preparation of sediment and sludge samples

An aliquot of 1 g sample was transferred to a polypropylene tube and fortified with an internal standard mixture (d₇-metoprolol, defluoro paroxetine, d₃-sertraline and d₆-venlafaxine). One ml of water pH 2 and

7 ml methanol were added to the sample, vortexed and sonicated for 15 min. After the centrifugation step the methanolic phase was carefully decant off, transferred to a glass bottle and diluted with 235 ml water. Adjustment to pH>12 was done by addition of 5M NaOH. Following sample preparation steps were performed according the procedure for water samples.

Instrumental determination

The basic pharmaceuticals (lidocaine, metoprolol, venlafaxine, citalopram, amlodipine, dipyridamole, paroxetine, fluoxetine and sertraline) were analysed by ultra-high pressure liquid chromatography triple-quadrupole mass-spectrometry (UHPLC-MS/MS). Analysis was performed on a Thermo Scientific quaternary Accela 1250 pump with a PAL Sample Manager coupled to a Thermo Scientific Vantage MS/MS (Vantage TSQ). An injection volume of 10 µL was used for sample separation on a Waters Acquity UPLC BEH C₁₈ column (2.1 × 100 mm, 1.7 µm) equipped with a Waters Van guard BEH C₁₈ guard column (2.1 × 5 mm, 1.7 µm). Separation was achieved using 2 mM aqueous NH₄HCO₂ (pH 2.5) (A) and 0.1% HCOOH in methanol (B) as the mobile phase. The following gradient programme was applied: initial conditions 95% A/5% B for 30 sec and a flow rate of 0.3ml/min, increased over 90 sec to 25% A/75% B, increased over 60 sec to 20% A/80% B which was kept for 180 sec, then increased to 100% B and 0.35 ml/min flow over 30 sec and kept for 180 sec, then reduced to 95% A/5% B, 0.3 ml/min in 30 sec, and finally equilibrated for 150 sec, in a total run time of 11.50 min. Ionisation was performed in positive electrospray ionisation mode (ESI⁺). The MS was run in SRM where two transitions of precursor ions from protonated molecular ions were monitored.

Solid phase extraction

Preparation of water samples

An aliquot of 240 ml sample was transferred to a beaker and pH adjusted to pH 6–7, if necessary. The internal standard mixture (d₅-candesartan, d₅-enalapril, d₃-estriol, d₄-estrone, d₆-metformin, ¹³C,₂-hydrochlorothiazide, d₆-simvastatin and d₈-zopiclone) was added and the beaker was put into an ultrasonic bath for some min. Samples were prepared on a vacuum manifold through an Oasis HLB-solid phase extraction column (Waters, USA). The SPE-column was pre-cleaned and conditioned before loading the sample. After drying the SPE-column by passing through air, the analytes were slowly eluted with methanol/acetone 2:1 and the extract was up-concentrated to 1 ml. Recovery

standard 1 (d₇-metoprolol) was added. 40 µL aliquots of the extracts were taken off into total recovery vials for SPE step 1 analysis and diluted with an aqueous 0.1% HCOOH solution.

For further clean-up an Oasis MAX column (Waters, USA) was used. After the pre-clean and condition steps the extract was applied at gravity speed and eluted with methanol/acetone 2:1. Both, sample and wash eluates were collected and reduced in volume to 0.5 ml. Recovery standard 2 (d₄-paracetamol) and 2 mM NH₄OAC water/acetonitrile (9:1) solution were added.

Preparation of sediment and sludge samples

An aliquot of 1 g sample was transferred to a polypropylene tube and spiked with an internal standard mixture (d₅-candesartan, d₅-enalapril, d₃-estriol, d₄-estrone, d₆-metformin, ¹³C,₂-hydrochlorothiazide, d₆-simvastatin and d₈-zopiclone). One ml of water pH 7 and 7 ml methanol were added to the sample, vortexed and sonicated for 15 min. After the centrifugation step the methanolic phase was carefully decant off, transferred to a beaker and diluted with 235 ml water. Adjustment of the pH to 6–7 was done if necessary. Following sample preparation steps were performed according the procedure for water samples.

Instrumental determination

The pharmaceuticals prepared with the SPE method were analysed by ultra-high pressure liquid chromatography triple-quadrupole mass spectrometry (UHPLC-MS/MS). Analysis was performed on a Thermo Scientific quaternary Accela 1,250 pump with a PAL Sample Manager coupled to a Thermo Scientific Vantage MS/MS (Vantage TSQ).

Compounds extracted within SPE step 1 were analysed with two different methods. For metformin, atenolol, hydrochlorothiazide, paracetamol, amiloride, enalapril, losartan, candesartan, gliclazide, levothyroxine and zopiclone an injection volume of 10 µL was used for sample separation on a Waters Acquity UPLC HSS C₁₈ column (2.1 × 100 mm, 1.8 µm) equipped with a Waters Van guard HSS C₁₈ guard column (2.1 × 5 mm, 1.8 µm). Separation was achieved using 0.1% HCOOH in water (A) and 0.1% HCOOH in methanol (B) as the mobile phase. The following gradient programme was applied: initial conditions 90% A/10% B, 0.2 ml/min for 55 sec, increased over 5 sec to 90% A/10% B 0.3 ml/min, increased over 120 sec to 60% A/40% B, which was then further increased in 250 sec to 100% B and a flow rate of 0.35 ml/min and kept for 180 sec, reduced in 90 sec to 90% A/10% B and 0.3 ml/min and hold for equilibration for 90 sec at 0.2 ml/min in a total run time of 13.20 min. Ionisation was performed in ESI⁺ mode. The MS was run in SRM

where two transitions of precursor ions from protonated molecular ions were monitored.

For enalaprilat, perindopril and perindoprilat an injection volume of 10 μ L was used for sample separation on a Thermo Scientific Hypersil Gold C₈ column (2.1 \times 50 mm, 1.9 μ m). Separation was achieved using 0.1% acetic acid in water (A) and methanol (B) as the mobile phase. The following gradient programme was applied: initial conditions 95% A/5% B for 55 sec at a flow rate of 0.2ml/min, increased over 5 sec to a flow rate of 0.5mL/min, increased in 130 sec to 25% A/75% B, increased in 60 sec to 100% B and with reduced flow rate of 0.4 ml/min, reduced to 95%A/5% B and 0.3 ml/min in 60 sec and further reduced to 0.2 ml flow rate in 60 sec in a total run time of 7.2 min. Ionisation was performed in ESI⁺ mode. The MS was run in SRM where two transitions of precursor ions from protonated molecular ions were monitored.

Compounds which need further clean-up within the second SPE step were 17 β -estradiol, estriol, estrone, 17 α -ethinylestradiol, simvastatin and sulfamethizole. An injection volume of 10 μ L was used for sample separation on a Waters Acquity UPLC BEH C₁₈ column (2.1 \times 100 mm, 1.7 μ m) equipped with a Waters Van guard BEH C₁₈ guard column (2.1 \times 5 mm, 1.7 μ m). Separation was achieved by 2 mM NH₄OAc water /acetonitrile 9:1 (A) and 2 mM methanolic NH₄OAc (B) as the mobile phase. The following gradient programme was applied: initial conditions 90% A/10% B, 0.2ml/min for 55 sec, increased flow rate in 5 sec to 0.35 ml/min, increased over 200 sec to 60% A/40% B, and further increased in 150 sec to 100% B, where then in 60 sec only the flow rate was increased to 0.4 ml/min, in 60 sec reduction to 90% A/10% B and 0.3 ml/min and to 0.2 ml/min in 90 sec for equilibration in a total run time of 10.80 min. Ionisation was performed in negative atmospheric pressure chemical ionisation mode (APCI⁻). The MS was run in SRM where two transitions of precursor ions from deprotonated molecular ions were monitored.

4.1.4 Quantification and quality control

Quantification was done by using the internal standard method with isotope labelled pharmaceuticals. For quantification of the liquid phase micro extraction (LPME) samples an eight point calibration curve (0, 0.1, 1, 10, 100, 500, 1,000 and 2,000 ng) was applied. Ultra-high purity water was fortified with native and isotope labelled standard mixtures and were treated in the same way as the samples during the extraction processes. For quantification of the SPE extracts, a solvent standard calibra-

tion curve with eleven points (0, 0.25, 0.5, 1, 5, 50, 500, 2,500, 5,000, 10,000 and 20,000 pg on column) was applied. During analysis, solvent injections were done regularly in order to monitor instrument background and carry-over effects. Procedure blank samples were prepared for quality assurance in each sample preparation batch. Field blank samples for sludge and sediment were only available from one sampling site from Greenland and were prepared with the two LPME extraction methods. Individual compound dependent method detection limits (MDLs) were set as the lowest concentration in the calibration curve which was within the linearity of the detector and had a greater signal to noise (S/N) of 3. Where blank contamination was detected, MDLs were estimated as three times the average blank value. If the MDL calculated from the blank contamination was higher as the MDL calculated from the calibration curve, MDL based on calculations from the blank samples was used.

For verification of correct identification and quantification of the target compounds quality criteria as retention time shift (± 0.05 min), detectable quantifier- and qualifier-ions and a greater S/N ratio of 3 were used.

4.2 Additives in Personal Care Products

4.2.1 Chemicals

The following chemicals used in the analytical work were purchased from Sigma-Aldrich: ethylenediaminetetraacetic acid (EDTA), 1,2-diaminopropan-N,N,N',N'-tetra acetic acid (PDTA), diethyl phthalate, butylparaben, 4-octylbenzene sulfonic acid, n-C₈-LAS, Sodium dodecyl sulphate (SDS), Trimethylhexadecylammonium chloride (ATAC-C16).

Trifluralin was delivered by Dr Ehrenstorfer GmbH.

Biphenyl (99%) came from an unknown source and was used as a volumetric standard.

As individual ethoxylated compounds were not available a technical product (Chemos, GmbH) was used. The sensitivity for the MRM transition molecular ion to m/z 97 [HSO₄] was assumed to be the same for the different ethoxylate chain lengths. By this assumption the following composition was found for the technical blend: SDS 21%, SDSEO₁ 27%, SDSEO₂ 31%, SDSEO₃ 15%, SDSEO₄ 6%. Sodium laureth sulphate concentration was calculated as the sum of SDSEO₁, SDSEO₂, SDSEO₃ and SDSEO₄.

Cocoamidopropyl betaine was obtained as a 30% solution (Chemos).

3-F-diethylphthalate, 3-F-propylparaben were synthesized at IVL laboratory Stockholm (>97% GC).

Solvents used for extraction were delivered from Rathburn Chemical Ltd. (Peeblesshire, Scotland).

Supelclean ENVI-Carb was obtained from Supelco.

4.2.2 Equipment

All equipment made of glass was acid washed and heat-cleaned (400°C, 4 h) before use. All other equipment was rinsed with hexane or acetone before use.

4.2.3 Methods

EDTA

Preparation of water samples

Water sample (50 ml) was filtrated through pre-cleaned GF/C-filter and spiked with surrogate standard (PDTA). The concentration (extraction) was performed with the aid of a Solid-Phase-Extraction (SPE) column fitted on a vacuum manifold. After passing the sample through the column it was rinsed with HCl (0.01 M) and dried for approximately 15 min under full vacuum. The column was eluted with an organic solvent and the extract evaporated to dryness under a stream of N₂.

Preparation of sediment and sludge samples

Freeze-dried sample (~0.5 g) was spiked with surrogate standards. Zinc sulphate and ultra-pure water were added and the sample was treated in an ultra-sonic bath for 15 min. Phosphate solution (KH₂PO₄) was added and the sample was again treated in the ultrasonic bath (5 min) followed by agitation on a shaking board (30 min). After centrifugation the extract was safeguarded. The extraction cycle was repeated twice with ultra-pure water. The combined extract was concentrated on SPE in the same way as water samples.

Derivatisation

The acids in the extract were esterified to the corresponding propyl esters using the reagent propanol/HCl at 90°C for 1 hour. The reaction was terminated by adding carbonate buffer. The derivatives were extracted with hexane, dried over sodium sulphate and concentrated under nitrogen gas. Volumetric standard (trifluralin) was added prior to gas chromatography determination.

Instrumental determination

The analysis was carried out with a HP 5890 Series II GC-NPD system, on-column injector and a HP 7376 auto sampler, all from Hewlett-Packard. The column consisted of two parts: (a) a methyl deactivated megabore pre-column (0.53 μm , 10–15 cm) needed for the auto on-column injector, (b) an analytical fused silica capillary column (15 m) with an ID of 0.25 mm and a film thickness of 0.25 μm (RTX-5 MS; Restek). After 50–100 injections, or when peak tailing appeared, the megabore column was exchanged. The following temperature program was used: 1 min isothermal at 100°C followed by an increase of 25°C/min to 200°C and then 10°C/min to 300°C, hold for 20 min. The detector signal from the gas chromatograph was acquired and processed with the chromatography data program Turbochrom™. The compounds were identified and quantified by comparison of their retention time and peak area to authentic reference compounds. The recovery of the analyte was estimated by means of the added surrogate standard.

Diethyl phthalate and butylparaben

Preparation of water samples

The water samples (200–800 ml) were filtrated (pre-heated GF/C-filter) prior to solid phase extraction. The filtrated sample was spiked with surrogate standards (3-F-diethylphthalate, 3-F-propylparaben), acidified and concentrated on a SPE-column (~15 ml/min). After the sample had passed through the SPE, the column was rinsed with HCl (0.01M) and dried. The analytes were eluted with methanol and a mixture of hexane: MTBE. The extracts were combined and the methanol was washed away by shaking the extract with HCl (0.01M). The extract was dried over sodium sulphate, concentrated and derivatised. After derivatisation the extract was cleaned-up on a silica SPE-column.

Preparation of sediment and sludge samples

Sediment (10 g f.w.) or sludge (2 g f.w.) was acidified with phosphorus acid and extracted twice with acetone: hexane (1:1) first in an ultrasonic bath (5 min) and then on a shaking board (25 min). The acetone was removed from the combined organic extract by shaking it with HCl (0.01M) water. The extract was dried over sodium sulphate, concentrated and derivatised. After derivatisation the extract was cleaned-up on a silica SPE-column.

Derivatisation and clean-up

Derivatisation was performed according to Remberger (2006). In brief, the phenol was acetylated with the reagent acetic acid anhydride with sodium acetate as base at 85°C in 45 min. The reaction was terminated by adding a carbonate buffer. The derivative in the organic solvent was withdrawn and used for the determination of butylparaben and diethyl phthalate.

The derivatised extract was applied onto a silica gel column. Two fractions were collected with eluents: (a) hexane and (b) hexane: MTBE (9:1). The former fraction was discarded and the latter was used for the determination of butylparaben and diethyl phthalate. Prior to GC-MS determination a volumetric standard (biphenyl) was added.

Instrumental determination

The sample extracts were analysed on a 6,890N gas chromatograph coupled to a 7,000N mass selective detector (Agilent). The injection, 1 μ L, was done in splitless mode at 240°C. The fused silica capillary column (VF-5MS 30 m \times 0.25 mm i.d. \times 0.25 μ m film thickness, Varian) was held at 45°C for 1 min., ramped 15°C/min to 200°C, 5°C/min until 300°C and held at 300°C for 5 min. Helium was used as carrier gas. The detector was used in Multiple Reaction Monitoring (MRM). The analytes were identified by their characteristic retention time and two product ions from selected precursor ions. Precursor ion for DEP was m/z 149 and the product ions were m/z 93 and m/z 65. Precursor ion for butylparaben was m/z 194 and the resulting product ions were m/z 138 and m/z 121.

Quantification was based on comparison of peak abundance to the known response of a surrogate standard mix. The reported analyte concentrations were corrected according to the determined surrogate standard losses.

Sodium dodecyl sulphate (SDS), Sodium laureth sulphate (SDSEO) and Cocoamidopropyl betaine (CAPB)

Preparation of water samples

Surrogate standard mix (4-Octylbenzene sulfonic acid, n-C8-LAS) was added to all samples. Water sample was without previous filtration, extracted on a graphitized carbon black SPE column (Supelclean ENVI-Carb), washed with methanol and eluted with dichloromethane/methanol containing tetramethylammoniumhydroxide (Di Corcia, et al., 1994). After evaporation the extract was redissolved in equal parts 10 mM NH₄OAc in water and methanol and analysed by LC-MS/MS.

Preparation of sediment and sludge samples

Freeze dried sample was extracted with methanol. After centrifugation the sediment extract was treated on a graphitized carbon black SPE column in the same way as described for water samples. The extract of sludge was diluted with equal parts 10 mM NH₄Ac in water and methanol. The extracts were analysed by LC-MS/MS.

Cetrimonium salt

Preparation of water samples

Water (25 ml) was acidified and 50 µg C₁₂LAS was added. The sample was extracted with chloroform which was evaporated to dryness (Martinez-Carballo et al., 2007). The residue was re-dissolved in methanol and analysed by LC-MS/MS.

Preparation of sediment and sludge samples

Freeze dried sediment or sludge was extracted with concentrated HCl diluted with methanol to a concentration of 1M in an ultrasonic bath (3 min) and then at 85°C (10 min). The extraction was repeated twice, the extracts were combined and the volume reduced to a few millilitres. After washing with hexane/MTBE (1:1) the extract was further evaporated to dryness (Remberger et al., 2006). The residue was dissolved in water (5 ml) containing 50 µg C₁₂LAS. The solution was extracted with chloroform which was evaporated to dryness, the residue re-dissolved in methanol and analysed by LC-MS/MS.

Instrumental determination

Liquid chromatography was performed using a Prominence UFLC system (Shimadzu) with two pumps LC-20AD, degasser DGU-20A5, auto sampler SIL-20AHT and column oven CTO-20AC. A column (Ascentis

C8 50 × 2.1 mm, particle size 5 µm, Supelco) was installed in the eluent flow line immediately upstream the auto sampler. This made analyte peaks originating from the solvent/solvent system elute later than peaks from the sample. The analytical column was a Thermo HyPurity C8 50 mm × 3 mm, particle size 5 µm (Dalco Chromtech). The solvent was 10 mM NH₄OAc in water mixed with methanol in a linear gradient from 30% to 100%. The column temperature was 50°C and the flow rate 0.5 ml/min. The effluent was directed to an API 4000 triple quadrupole mass spectrometer (Applied Biosystems). ESI⁻ mode was used for SDS, SDSEO and CAPB. Precursor ion was the deprotonated molecular ion. Product ions were *m/z* 170 for [n-C₈-LAS] and *m/z* 97 [SO₄H] and 80 [SO₃] for SDS. For CAP precursor ion was *m/z* 341 [C₁₂-CAPB H] and the product ion was *m/z* 102 [(CH₃)₂NCH₂COO] (Levine et al., 2002).

The determination of cetrimonium salt was performed with ESI⁺ mode. Trimethylhexadecylammonium chloride (ATAC-C16) was used as standard. Precursor ion was *m/z* 284, and product ion was *m/z* 60 [(CH₃)₃NH]⁺.

Quality control

The following quality criteria were used to ensure correct identification and quantification of the target compound: (a) the retention time should match those of the standard compounds within ± 0.05 min, (b) the intensity ratios of the selected ions (target- and qualifier-ions) are within ± 15% of expected / theoretical value (c) the signal-to-noise ratios are greater than 3:1.

Field blanks were collected at several sampling stations. An analytical method blank was included for each sample batch analysed to assess background interferences and possible contamination of the samples.

Concentrations below field blank levels were treated as not detected.

Possible background levels of analytes were subtracted from measured sample values.

Spiked (with the analytes) authentic background samples and parallel samples were also included to determine the recovery precision and trueness of the analytical results.

In this investigation limit of quantification (LOQ) was used and is defined as a signal 10 times the standard deviation of the blank values.

Surrogate standard mix was added to the sample at the start of the working-up procedure of the sample. The surrogate standard mix has similar chemical and physical properties to the compounds to be analysed.

4.3 Uncertainty of the study

When performing environmental screening or monitoring all steps in the study starting with the design of the study, selection of sampling sites and sampling frequency, time of sampling, performing of sampling, transport and storage of samples, chemical analysis and data treatment are generating some degree of uncertainty. To quantitatively estimate the contribution of all steps is an extreme difficult task or not possible at all. However, we will discuss the relevance of the different contributors in a qualitative way.

One important question is whether a sample is representative for a given time period or a given region. Many of the selected compounds are intermittently emitted to the environment and a constant concentration of these compounds in the environment is not expected. In this screening, the samples were collected within a narrow time frame and at only a few different geographical locations. The results obtained here are therefore only a snapshot of the reality at those places at the given time. The study was also designed as a screening/snap shot, e.g. discussion of the time of the day for sampling, most people use the bathroom in the mornings early etc. In the Faroe Islands, samples in WWTP were taken shortly after 7 am, but in Iceland for instance, samples in the main hospital SL and WWTP were taken around 9–10 am and in Hveragerði WWTP they were taken in the period 8:30 to 10:30 am.

Factors which influence sampling uncertainty are analyte loss due to adsorption to sample containers, wastewater flow and particle content, tidal water current, selection of sample type (water with or without particle phase), and degradation during transport and storage. Loss due to adsorption on vessel walls were kept to a minimum by careful selection of containers and container treatment, and loss due to degradation was limited by storing samples frozen until analyses.

The uncertainty of the chemical analysis is governed by loss during extraction and clean-up, interference from other compounds, trueness of analytical standards, instrumental parameters, and contamination. A normal approach to estimate and quantify these factors is the participation in laboratory intercalibration. However, at this stage the analysis of these compounds in environmental samples is not done routinely and intercalibration studies have not been available. The uncertainty is expected to be larger for compounds which are analysed the first time (26 of 37 pharmaceuticals) than for compounds which have been analysed previously or where similar compounds have been analysed earlier (14 of 37 pharmaceuticals). That means that most compounds will probably have analytical uncertainties in the range of 20 to 40%. For all analytes we consider the analytical uncertainty as fit-for-purpose, that means adequate for a first screening study. However, the results cannot be implemented uncritically in time-trend studies.

5. Results and discussion

Details on samples and sampling locations and individual concentrations are listed in the appendix in Table 28–Table 32. All measured concentrations of pharmaceuticals are given in ng/l in water samples and µg/kg dry weight in sediment and sludge samples. Concentrations of additives in personal care products are given in µg/l and mostly as µg/kg dry weight for water and solid samples respectively.

5.1 Non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs

The six drugs belonging to the afore-mentioned groups of agents (Table 1) were detected in all of the five different matrices (Table 13). Paracetamol, naproxen and ibuprofen were found in highest concentrations in waste water from the main hospital Fossvog at Iceland (Figure 17 C) at concentrations of 25–1,000 ng/l, 109,000 ng/l and 48,800 ng/l, respectively. Salicylic acid, diclofenac and lidocaine were also found in high concentrations in WWTP influents (Table 14); salicylic acid and lidocaine concentrations were 38,400 ng/l and 183 ng/l in influents to the main hospital Faroe Island WWTP, respectively, and the concentration of diclofenac was 697 ng/l in influents to WWTP Hveragerði, Iceland (Figure 17). Diclofenac occurred above the LOD in all matrices, at all locations. Paracetamol was not detectable in sediments (Table 13 and Table 14).

Table 13. Detection frequency of non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs. N number of samples d number of detections

	Influent			Effluent			Sludge			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
Salicylic acid	89	(9 ; 8)		91	(11 ; 10)		100	(8 ; 8)		40	(10 ; 4)		100	(6 ; 6)	
Diclofenac	100	(9 ; 9)		100	(11 ; 11)		100	(8 ; 8)		100	(10 ; 10)		100	(6 ; 6)	
Ibuprofen	100	(9 ; 9)		91	(11 ; 10)		100	(8 ; 8)		90	(10 ; 9)		50	(6 ; 3)	
Lidocaine	100	(9 ; 9)		73	(11 ; 8)		83	(6 ; 5)		50	(10 ; 5)		17	(6 ; 1)	
Naproxen	100	(9 ; 9)		91	(11 ; 10)		100	(8 ; 8)		20	(10 ; 2)		17	(6 ; 1)	
Paracetamol	89	(9 ; 8)		92	(12 ; 11)		50	(6 ; 3)		50	(10 ; 5)		0	(6 ; 0)	

Table 14. Maximum and minimum concentrations of non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs in ng/l for water samples and µg/kg dw for sludge and sediment samples

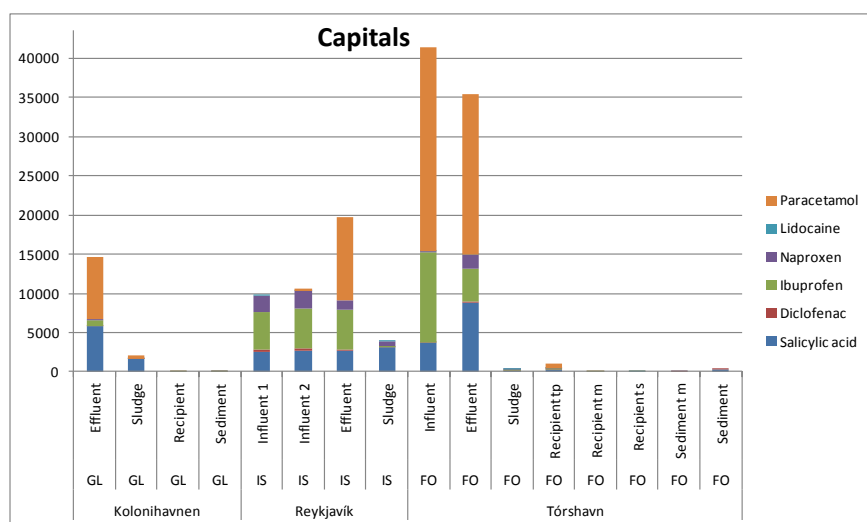
	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
Salicylic acid	38,400	1,110	24,600	<41.7	3,090	104	6,050	<41.7	222	7.69
Diclofenac	697	24.4	597	14.3	28.5	1.65	30.1	1.45	1.04	0.18
Ibuprofen	48,800	1.62	5,080	<0.42	210	15.8	872	<0.42	2.57	<0.10
Lidocaine	183	1.00	61	<0.42	46.5	<0.25	8.4	<0.42	0.73	<0.25
Naproxen	109,000	273	1,920	<1.05	640	0.32	45.9	<1.05	0.85	<0.01
Paracetamol	251,000	<20.8	71,500	<20.8	447	<5.0	931	<20.8	nd	<5.0

WWTP influent and effluent

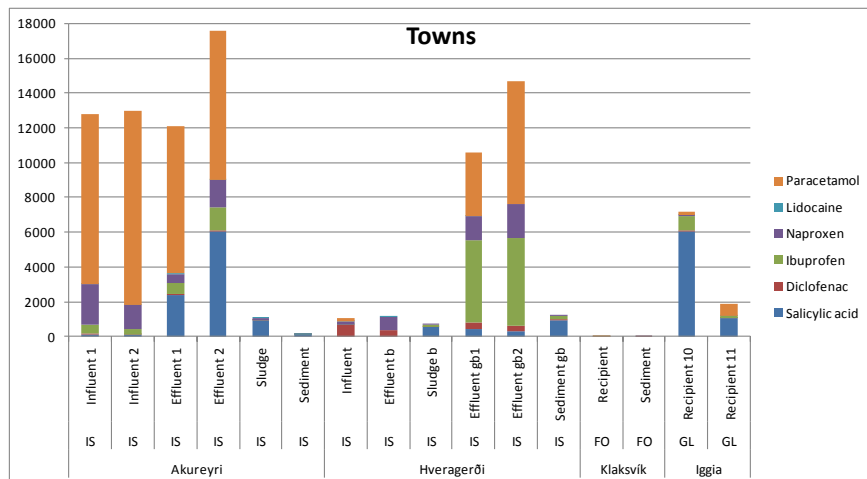
Salicylic acid was detected in almost all WWTP influent and effluent samples with the exception of some samples from Hveragerði WWTP. Generally, higher concentrations were measured in WWTP effluents compared to the WWTP influents (Figure 17), which shows a low removal capacity within the WWTP at Akureyri and Torshavn for this compound. In Reykjavik WWTP approximately the same levels of salicylic acid was found in influent as in effluent samples (Figure 17 A). Salicylic acid was previously reported in concentrations up to 170,000 ng/l and 18,000 ng/l in WWTP influents and effluents, respectively, in a Danish study (Mogensen et al., 2008). Acetylsalicylic acid, the mother substance of salicylic acid, has been monitored in Danish studies (Mogensen et al., 2008; Kjølft et al., 2003) and in a Swedish study (Remberger et al., 2009), where the compound was only detected in the WWTP influent samples in Denmark and then at average concentrations of 1,600 ng/l (Mogensen et al., 2009). The low detection frequency of acetylsalicylic acid is not surprising since this drug is hydrolysed to salicylic acid and other metabolites at humid conditions.

Diclofenac was detected in all influent and effluent samples (Table 13). It was detected in approximately the same levels in influents and effluents (Table 14, Table 29 and Figure 17).

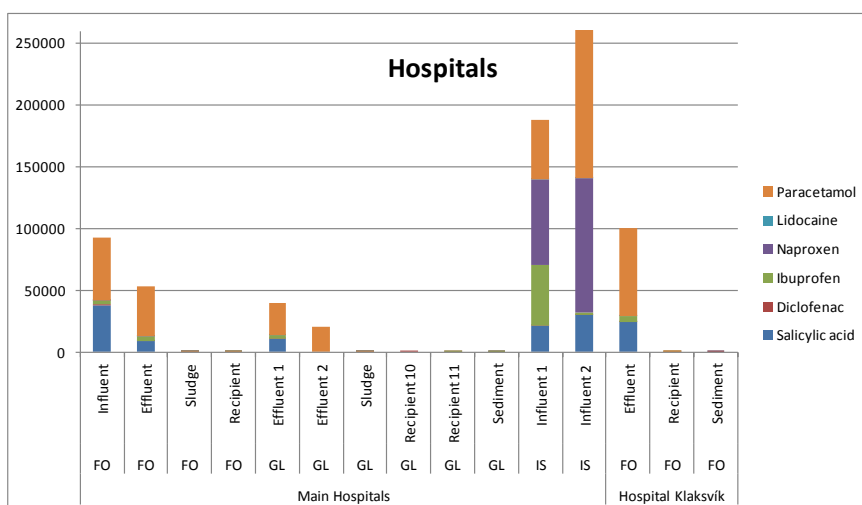
Figure 17. Non-steroidal anti-inflammatory and antipyretic analgesics and local anaesthetic drugs in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and µg/kg for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

Diclofenac has been reported from WWTP effluents in Finland (Vieno et al., 2008), Norway (Thomas et al., 2007) and Sweden (Andersson et al., 2006; Fick et al., 2011; Lilja et al., 2010) with the highest concentration of 3,900 ng/l in Sweden, followed by Iceland (this study, 390 ng/l) and Norway (370 ng/l). Swedish WWTPs exhibited the highest levels of 9 700 ng/l (Andersson et al., 2006; Fick et al., 2011; Lilja et al., 2010). Another Norwegian study reported diclofenac ranging from not detected to 44,700 ng/l, with the highest concentration observed in sewers from the hospital in Tromsø (Weigel et al., 2004).

Ibuprofen was detected in all WWTP influent samples, with the highest concentration of 48,800 ng/l being from the main hospital Fossvog (IS; Figure 17 C). Regarding WWTP effluents, Ibuprofen ranged from < 0.42–5,080 ng/l (Table 14) with the highest levels detected in Reykjavik WWTP. Ibuprofen was not detected in the effluent from the biological part of Hveragerði WWTP (Figure 17). Similar concentration levels were found in WWTP influent and effluent samples in other studies from Nordic countries (Mogensen et al., 2008; Mørskeland et al., 2006; Thomas et al., 2007; Fick et al., 2011; Remberger et al., 2009; Andersson et al., 2006; Lilja et al., 2010; Kjølholt et al., 2003; Vieno, 2008).

Naproxen was detected in all WWTP influent samples in concentration ranges from 2.73 ng/l (main hospital, FO) to 109,000 ng/l (main hospital Fossvog, IS; Table 14 and Figure 17) and in almost all WWTP effluent samples with the exception of the hospital in Klaksvik. The highest effluent concentration of 1,920 ng/l was found at Hveragerði (Figure 17). Other studies from Sweden (Fick et al., 2011; Remberger et al.,

2009; Andersson et al., 2006; Lilja et al., 2010) detected naproxen continuously in the different matrices in increased concentrations compared to the present study and other studies from Finland (with non-detects; Vieno, 2008) and Norway (Schlabach et al., 2009). The Norwegian environmental concentration levels of naproxen were approximately in the same ranges as in the present study.

Paracetamol exhibited the highest concentrations in WWTP effluents ranging from < 20.8–71,500 ng/l (Table 14) and was detected in 11 out of 12 samples (Table 13). The highest concentrations were found at the hospitals at Faroe Islands (Figure 17 and Table 29). Paracetamol was detected in eight out of nine WWTP influent samples (Table 13) at concentrations ranging from < 20.8 to 251,000 ng/l (Table 14). The highest concentration was observed at the STP main hospital at the Faroe Islands (Figure 17 C). WWTP influents from Denmark (Mogensen et al., 2008) and Sweden (Fick et al., 2011; Remberger et al., 2009) showed substantially lower levels, namely 1,200 µg/l and 540 µg/l, respectively, whereas for WWTP effluents, significantly higher concentrations were found in Sweden (280 µg/l).

It is the first time that lidocaine was analysed in Nordic samples. The measured concentrations were quite low (1–183 ng/l in influent and < 0.42–61 ng/l in effluent waters) compared to the other compounds summarised in this chapter (Table 14).

WWTP sludge

Salicylic acid seems to bind to particles and was therefore found also in all samples of WWTP sludge in concentrations ranging from 104–3 090 µg/kg (Table 13, Table 14 and Figure 17). In a Danish study, concentrations of 59–2 800 µg/kg were reported, while the mother compound acetylsalicylic acid was not detected in sewage sludge samples (Mogensen et al., 2008).

Similarly, diclofenac was also found in WWTP sludge (Table 13 and Table 14). Several Swedish studies reported diclofenac concentrations in WWTP sludge samples approximately 20 times higher (560 µg/kg, Andersson et al., 2006; Fick et al., 2011; Lilja et al., 2010), than in Norway (Thomas et al., 2007), Faroe Islands and Iceland. In Greenland, only low ng/kg levels were measured.

Ibuprofen was found in all WWTP sludge samples in concentration ranges from 2.57 µg/kg in Akureyri to 169 µg/kg Torshavn (Table 13, Table 14 and Figure 17). WWTP sludge samples from Sweden showed significantly higher concentrations (22,000 µg/kg; Andersson et al., 2006) compared to countries of the present study as well as Denmark

(not detected; Mogensen et al., 2008) and Norway (17 µg/kg; Møskeland et al., 2006; Thomas et al., 2007).

Naproxen was detected in all WWTP sludge samples (Table 13) in concentrations ranging from 0.32 µg/kg (main hospital, FO) to 640 µg/kg (capital Reykjavik, IS; Table 14 and Figure 17). Higher concentrations of naproxen in sludge samples were reported in other studies from Sweden (Fick et al., 2011; Remberger et al., 2009; Andersson et al., 2006; Lilja et al., 2010), whereas non detects and low concentrations were found in related studies from Finland (non detects; Vieno, 2008) and Norway (Schlabach et al., 2009). The Norwegian environmental concentration levels of naproxen were approximately in the same ranges as in the present study.

Paracetamol in WWTP sludge was detected in three out of seven samples (Table 13) in concentrations from < 5.0 to 447 µg/kg (Table 14), with the highest concentration found at Kolonihavnen, Greenland (Figure 17). In Denmark, sewage sludge concentrations exceeded 2,000 µg/kg (Mogensen et al., 2008) which is approximately five times higher than the highest concentrations found in the present study. Paracetamol was not detected in Norway (Schlabach et al., 2009; Thomas et al., 2007), and the highest concentration reported from a Swedish study on sewage sludge was 73 µg/kg (Remberger et al., 2009).

It is the first time that lidocaine was analysed in Nordic sludge samples. The measured concentrations were quite low (<0.42–8.40 µg/kg) compared to the other compounds summarised in this chapter (Table 14).

Recipient water and sediment

Salicylic acid and diclofenac were detected in recipient waters in levels from < LOD to 6,050 ng/l and 1.45–30.1 ng/l, in four out of 10 and in all samples, respectively (Table 13 and Table 14). These compounds tend to bind to particles and consequently were found in sediments of recipient waters. Diclofenac data for recipient waters and sediments are available only from Sweden (Andersson et al., 2006; Fick et al., 2011; Lilja et al., 2010) where values exceeding those in the present study were reported.

Ibuprofen was detected in 9 out of 10 recipient water samples, (Table 13) with the highest concentrations in Iggia (GL) with 872 ng/l (Table 14 and Figure 17). Ibuprofen was detectable only in three sediment samples (Table 13). From Sweden, recipient water ibuprofen levels exceeding 200 µg/l have been reported (Fick et al., 2011; Remberger et al., 2009; Andersson et al., 2006). Data on ibuprofen in sediments are available from Norway (Møskeland et al., 2006) and Sweden (Remberger et al., 2009; Andersson et al., 2006) with maximum concentrations of 2.8 and 6 µg/kg respectively.

Naproxen was found in two recipient water samples from Iggia (GL) and Torshavn (FO) with concentrations of 45.9 ng/l and 5.76 ng/l, respectively (Figure 17). Naproxen was detected in one sediment sample (Table 13 and Table 14). Other studies from Sweden (Fick et al., 2011; Remberger et al., 2009; Andersson et al., 2006; Lilja et al., 2010) reported naproxen in all matrixes, in higher concentrations compared to the present and other relevant studies from Finland (with non-detects; Vieno, 2008) and Norway (Schlabach et al., 2009). In Norway, environmental concentrations of naproxen were approximately within the same ranges as in the present study and with similar non-detects in sediment samples. Naproxen in surface waters is short-lived, due to biodegradation and photolysis occurring in uppermost layers (Straub and Steward 2007).

Paracetamol was detected in recipient waters in five out of 10 samples (Table 13) at concentration levels from < 20.8–931 ng/l (Table 14), whereas it was not detected in sediments in the present study. The removal efficiency for paracetamol in the investigated WWTPs was low since the compound was detected in effluents and in recipient waters in minor amounts due to dilution effects. Paracetamol was not detected in recipient waters from Denmark, Norway and Sweden and it has only been detected in sediments from Sweden, in concentrations as high as 69 µg/kg (Remberger et al., 2009).

Lidocaine was analysed for the first time in Nordic recipient water and sediment samples. Lidocaine concentrations were quite low < 0.42–8.4 ng/l in recipient waters with detectable levels in five only out of 10 samples (Table 13 and Table 14). Lidocaine was detected in one sediment sample at low concentration.

5.2 Antibiotics and antimicrobial agent

Sulfamethizole was the only drug analysed from this group of pharmaceuticals and it was always below LOD (20.8 ng/l and 5 µg/kg, Table 15 and Table 18). This is an interesting result, since 90% of the drug is excreted unmetabolised from the body via urine. Other studies performed in Norway (Møskeland et al., 2006) and Sweden (Kjølholt et al., 2003) also reported sulfamethizole below the LOD. It has been detected in Denmark however, at concentrations up to 3.2 µg/l and 110 µg/kg in WWTP effluents and sludge, respectively (Mogensen et al., 2008).

Table 15. Detection frequency of antimicrobial agents, antidiabetics and hypnotic drugs. N number of samples d number of detections

	Influent			Effluent			Sludge			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
Sulfamethizole	0	(9 ; 0)	0	0	(12 ; 0)	0	0	(6 ; 0)	0	0	(10 ; 0)	0	0	0	(6 ; 0)
Metformin	100	(9 ; 9)	100	100	(12 ; 12)	100	100	(6 ; 6)	50	50	(10 ; 5)	66	66	66	(6 ; 4)
Glicazide	44	(9 ; 4)	25	25	(12 ; 3)	17	17	(6 ; 1)	0	0	(10 ; 0)	0	0	0	(6 ; 0)
Zopiclone	0	(9 ; 0)	0	0	(12 ; 0)	0	0	(6 ; 0)	0	0	(10 ; 0)	0	0	0	(6 ; 0)

5.2.1 Antidepressants

WWTP influent and effluent

The detection frequency of citalopram, paroxetine, sertraline and venlafaxine in WWTP influent and effluent samples was generally high (78–100%; Table 16). The highest concentrations in influents of all the analysed antidepressants were found in Icelandic samples (Figure 18).

Citalopram were found in the range 82.2 ng/l (Reykjavik) to 2 040 ng/l (main hospital Fossvog) and from 12.2 ng/l (Hveragerði biol.) to 540 ng/l (hospital Klaksvik) in WWTP influent and effluent samples, respectively (Table 17).

Paroxetine was detected in concentrations ranging from < 1.51–783 ng/l in influent samples and < 1.51–149 ng/l in effluent samples (Table 17) with highest concentrations in Hveragerði and Torshavn (Figure 18). Paroxetine was found in lower concentrations in effluents than in influents in Torshavn and Hveragerði, but not in Reykjavik (Figure 17).

Sertraline was found in slightly lower concentrations than citalopram, paroxetine and venlafaxine (Table 17), with highest concentrations in influents from the main hospital Fossvog (382 ng/l) and in effluents from Hveragerði (33 ng/l), (Figure 18).

Venlafaxine was detected in concentration ranges from 29.3–30,200 ng/l and 21.3–1,020 ng/l in influents and effluents, respectively (Table 17). Highest concentrations were found in WWTP influents coming from the main hospital Fossvog in Iceland and in effluents from the main hospital in Greenland (Figure 18).

Fluoxetine was detected in one out of nine influent and one out of 11 effluent samples (Table 17), both from the Akureyri WWTP (Figure 18).

Citalopram, paroxetine, sertraline and fluoxetine have been analysed in a study from Tromsø, Norway. Citalopram was found in concentrations from 13–612 ng/l in influent samples, thus, approximately 6 times lower than in the present study. Concentrations in effluent samples in the Tromsø study ranged from 9.2 to 382 ng/l, thus lower than the levels found in the present study. Paroxetine, sertraline and fluoxetine were detected in low ng/l concentrations (Vasskog et al., 2006). In a follow up study, similar results were reported for Tromsø, whereas in Longyearbyen, Spitsber-

gen, citalopram was below the quantification limit (Vasskog et al., 2008). Hospital wastewaters and WWTP influent in Oslo (Norway) also had low ng/l concentrations of fluoxetine, sertraline and paroxetine (Langford and Thomas, 2009). The Swedish National Screening Programme 2010 (Fick et al., 2011) reported generally higher concentrations of fluoxetine (ca. 9–110 ng/l) in both WWTP influents and effluents and similar concentrations for sertraline (ca. 80–140 ng/l). WWTP effluent concentrations were higher in both the Norwegian and the Swedish study. For paroxetine, lower concentrations (similarly to the present work) were measured in Swedish WWTP influents (ca. 10–100 ng/l), whereas in effluents paroxetine was not detectable. Venlafaxine were found in lower concentrations (ca. 110–1 500 ng/l) in both WWTP influents and effluents than in the present study, where 20-fold higher concentrations in WWTP effluent samples were measured.

Table 16. Detection frequency of antidepressants. N number of samples d number of detections

	Influent			Effluent			Sludge%			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
Citalopram	100	(9; 9)	9	100	(11; 11)	11	100	(6; 6)	6	70	(10; 7)	7	100	(6; 6)	6
Fluoxetine	11	(9; 1)	1	9	(11; 1)	1	83	(6; 6)	6	0	(10; 0)	0	17	(6; 1)	1
Paroxetine	89	(9; 8)	8	91	(11; 10)	10	83	(6; 5)	5	10	(10; 1)	1	33	(6; 2)	2
Sertraline	78	(9; 7)	7	91	(11; 10)	10	83	(6; 5)	5	0	(10; 0)	0	82	(6; 5)	5
Venlafaxine	100	(9; 9)	9	100	(11; 11)	11	100	(6; 6)	6	60	(10; 6)	6	100	(6; 6)	6

Table 17. Maximum and minimum concentrations of antidepressants in ng/l for water samples and µg/kg for sludge and sediment samples

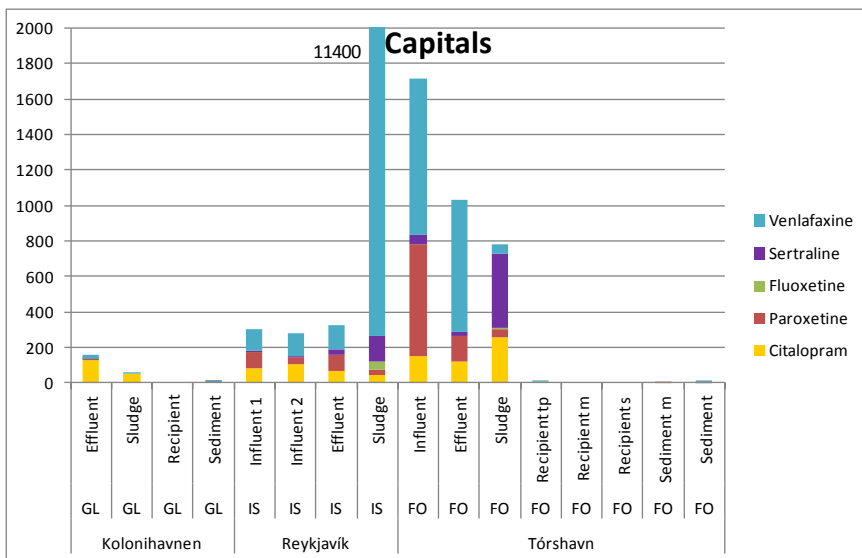
	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
Citalopram	2,040	82.2	540	12.2	382	46.1	6.25	<0.42	44.2	0.10
Fluoxetine	17.0	<4.17	5.00	<4.17	49.4	<0.10	nd	<4.17	10.8	<0.10
Paroxetine	783	<1.51	149	<1.51	120	<0.19	1.76	<1.51	6.91	<0.19
Sertraline	382	<0.42	33	<0.42	1,070	<0.10	nd	<0.42	28.1	<0.42
Venlafaxine	30,200	29.3	1,020	21.3	11,400	7.01	7.92	<0.42	73.6	<0.10

WWTP sludge

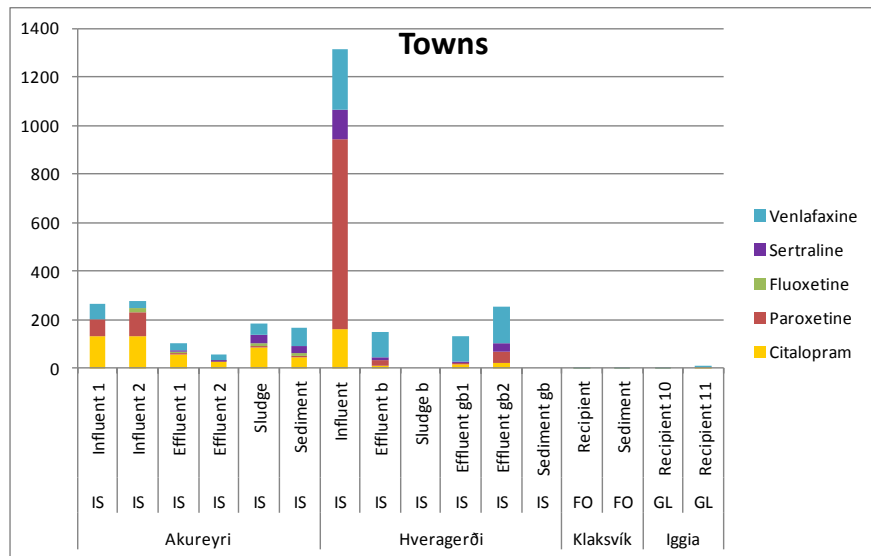
All five antidepressants analysed had a high frequency of occurrence (83–100%) in WWTP sludge samples (Table 16). The highest concentrations were found for citalopram, paroxetine and sertraline with 328,120 and 1,070 µg/kg, respectively at the main hospital in the Faroe Islands (Table 17 and Figure 18 C). Fluoxetine and venlafaxine were detected at concentrations of 49.4 and 11,400 µg/kg in Reykjavik (Table 17 and Figure 18 A), where venlafaxine in particular was found in high concentration at this location.

Previously reported concentrations of citalopram and fluoxetine from the Swedish National Screening Programme in 2010 (Fick et al., 2011) were considerably higher than in the present study. This was not the case for paroxetine, sertraline and venlafaxine though, where the highest concentrations were found in the present study, in Faroe Islands and Iceland. In Norway, concentration ranges of fluoxetine from 31 to 41 µg/kg were reported (Møskeland et al., 2006).

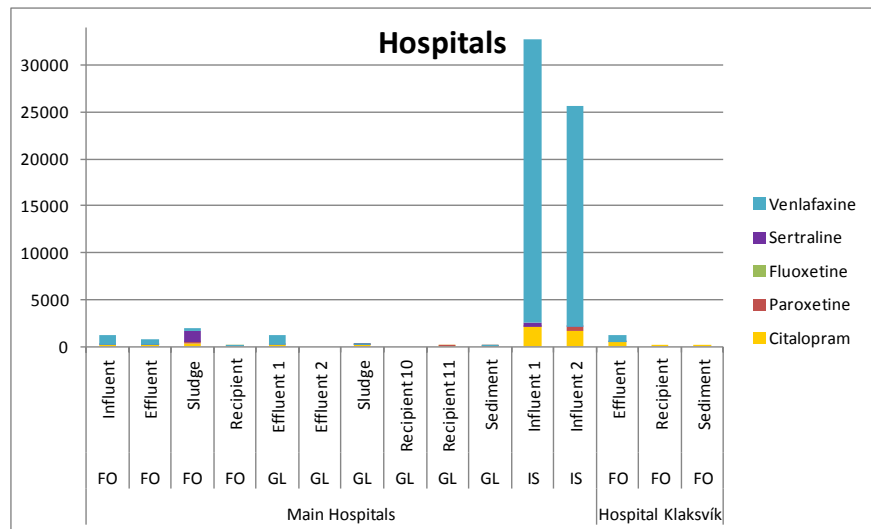
Figure 18. Antidepressant drugs in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and µg/kg for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

Recipient water and sediment

Citalopram was detected in seven out of 10 recipient samples and in all sediment samples (Table 16) at concentration ranges from < 0.42–6.25 ng/l and from 0.14–44.2 µg/kg, respectively (Table 17).

Paroxetine was detected in one out of 10 recipient waters and one out of six sediment samples (Table 17 and Figure 18).

Fluoxetine and sertraline were not detected in recipient water samples. Previously reported results from the Swedish National Screening

Programme 2011 (Fick et al., 2011) showed considerably higher concentrations in recipient waters, which were rivers in this case.

Fluoxetine and sertraline were detected in one and in five out of six sediment samples, respectively. Fluoxetine was found in the sediment sample from Akureyri at a concentration of 10.8 ng/l (Table 17 and Figure 18 B). Sertraline was found in the range from < 0.42–28.1 ng/l (Table 17) with the highest concentration in Akureyri.

There are no other data published on these two pharmaceuticals in sediments. The concentrations of fluoxetine in settleable particulate material were found to be from low ng/l up to approximately 100 ng/l in a Finnish study (Lathi and Oikari, 2001).

5.3 Antidiabetics

WWTP influent and effluent

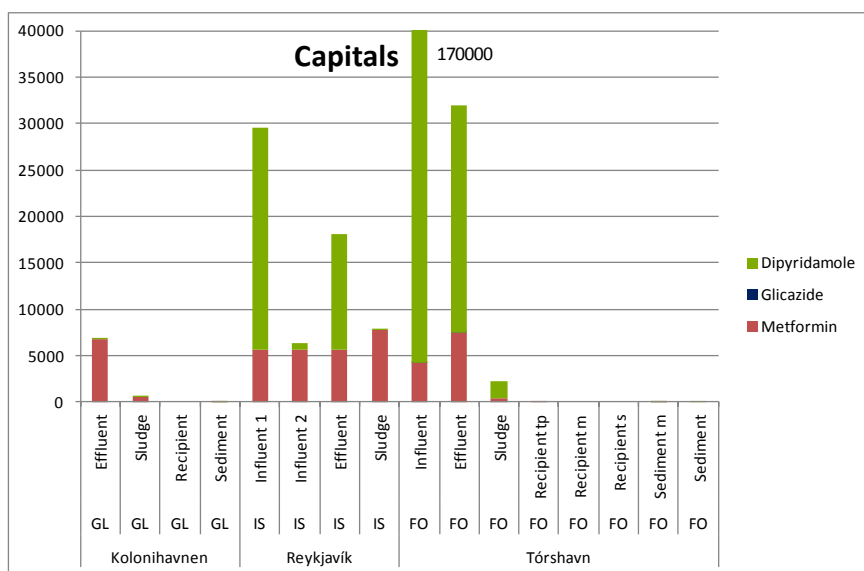
Metformin was detected in all WWTP influent samples in concentration ranges from 1,780 ng/l–59,000 ng/l with the lowest level from Akureyri and the highest from the main hospital Fossvog, both in Iceland (Table 15, Table 18 and Figure 19). Metformin was also detected in WWTP effluents at 100% frequency. The highest effluent concentration (7 950 ng/l) was found at the hospital in Klaksvik (Figure 19 and Table 29). Previously, concentration ranges of approximately 500 to 15,000 ng/l were reported for Swedish WWTP influents, while metformin was not detected in WWTP effluents in the same study (Fick et al., 2011).

Table 18. Maximum and minimum concentrations of antimicrobial agents, antidiabetics and hypnotic drugs in ng/l for water samples and µg/kg for sludge and sediment samples

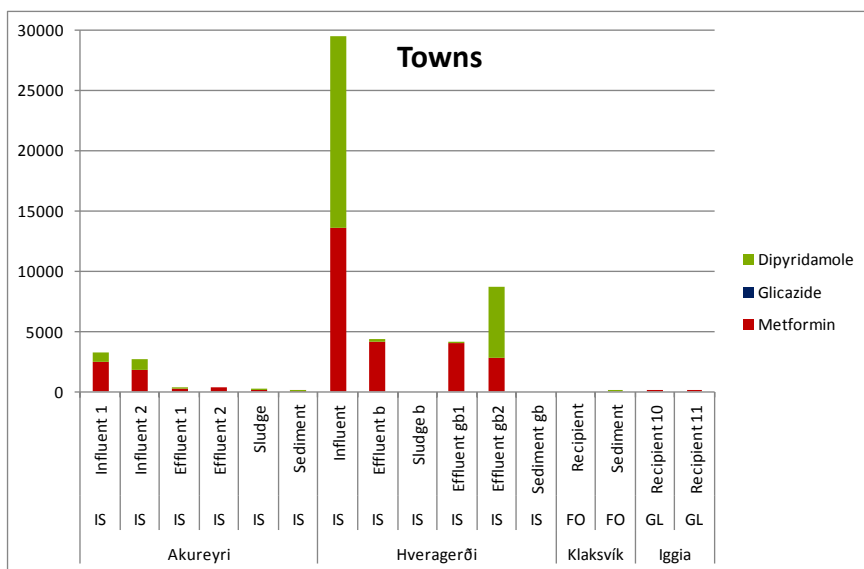
	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
Sulfamethizole	nd	<20.8	nd	<20.8	nd	<5.0	nd	<20.8	nd	<5
Metformin	59,000	1,780	7,950	238	7,810	149	748	<2.08	56.6	<0.05
Gliclazide	538	<2.08	30	<2.08	0.56	<0.50	nd	<20.8	nd	<0.50
Zopiclone	nd	<20.8	nd	<20.8	nd	<5.00	nd	<20.8	nd	<5.00

Gliclazide was analysed for the first time in samples from the Nordic environment, and was detected in four out of nine WWTP influent samples (Table 15). Very high concentrations (538 and 116 ng/l) were detected in influent samples from the main hospital Fossvog (Table 18 and Figure 19). Gliclazide was detected in three out of 12 WWTP effluent samples. The highest concentrations were measured at the main hospital in the Faroe Islands (30 ng/l; Table 15, Table 18 and Figure 19).

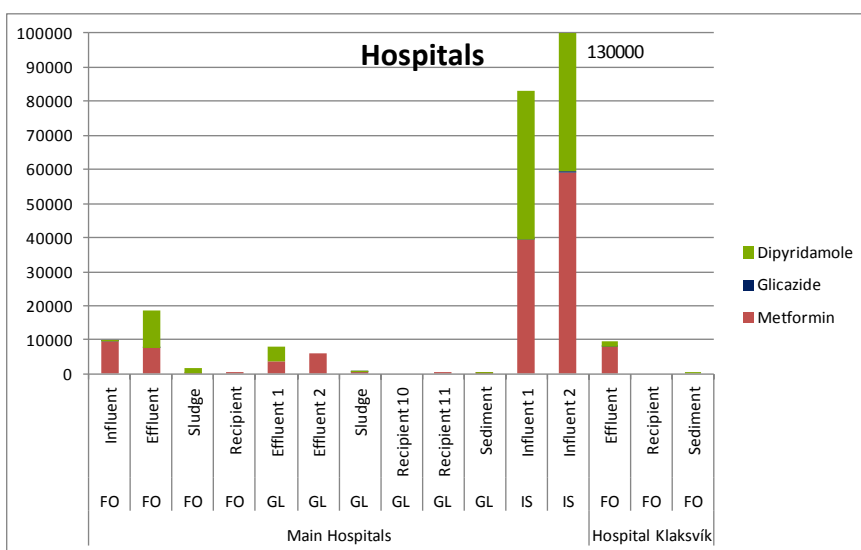
Figure 19. Antidiabetic drugs and a phosphodiesterase inhibitor (cardiovascular drug) in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and $\mu\text{g}/\text{kg}$ for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

WWTP sludge

Metformin was detected at a frequency of 100% in WWTP sludge samples (Table 15). The highest concentration in sludge (7,810 µg/kg; Table 18) was found in Reykjavik (Figure 18). Other data on metformin in WWTP sludge from Nordic countries are not available.

Gliclazide was detected in one WWTP sludge sample only, and then close to the LOD (Table 15 and Table 16). Other data for gliclazide in WWTP sludge from Nordic countries are not available.

Recipient water and sediment

Metformin was detected in five out of 10 recipient water samples with highest concentration (748 ng/l) at the Queen Ingrid's Hospital in Greenland in 2011 (Table 15 and Table 18), while in the sample from 2010, metformin was not detected (Figure 19). In the sediments, metformin was detectable in four out of six samples (Table 18) at relatively low concentrations in both Kolonihavnen and Torshavn, as well as at the main hospital in Iceland (Figure 19). Other data on metformin in recipient waters and sediments from Nordic countries are not available.

Gliclazide was not detected in recipient waters and sediments (Table 15). Other data for Gliclazide in recipient waters and sediments from Nordic countries are not available.

5.4 Antiulcer drugs

Omeprazole was analysed only qualitatively, but it was not detected. This might be due to the fact that omeprazole is not stable in the environment due to its chemical structure and physio-chemical properties. Stability studies have been conducted and have revealed that it is acid labile (pH < 7) and sensitive to light and heat (Wallmark and Lindberg, 1987; DellaGraca et al., 2006).

Omeprazole was included in previous Norwegian and Danish surveys but was not detected (Møskeland et al., 2006; Mogensen et al., 2008).

5.5 Cardiovascular drugs

WWTP influent and effluent

The *antianginal drug* amlodipine was detected in two out of nine WWTP influent samples and in nine out of 11 WWTP effluent samples (Table 19) at concentration ranges from < 4.17 to 247 ng/l and < 4.17 to 448 ng/l, respectively. The highest influent concentration was observed at the main hospital in Faroe Islands, while the highest effluent concentration was from the hospital in Klaksvik (Table 20 and Figure 21). A previous study from Denmark did not report amlodipine above the LOD in WWTP influent or effluent samples (Mogensen et al., 2008).

The *diuretics* amiloride, furosemide and hydrochlorothiazide showed high detection frequency in WWTP influents and effluents (100%), while bendroflumethiazide was only detected in four out of 11 WWTP effluent samples (Table 19).

Amiloride was detected in WWTP influents in concentrations that ranged from 18.9 ng/l at the main hospital in the Faroe Islands to 1 260 ng/l in samples from the main hospital Fossvog (Table 20 and Figure 21). Concentration levels in WWTP effluents ranged from < 3.03–217 ng/l with the highest in Reykjavik (Table 20 and Figure 21). There are no other published data on amiloride in the Nordic countries.

Bendroflumethiazide was not detected in WWTP influents and was found in relatively low concentrations (< 0.42–7 ng/l) in WWTP effluents (Table 20, Table 30 and Figure 21). Earlier surveys from Denmark reported bendroflumethiazide in WWTP influent samples with concentrations up to 112 ng/l (Mogensen et al., 2008) and up to 70 ng/l in effluent samples, thus, much higher concentrations compared to the present study (Mogensen et al., 2008; Kjølholt et al., 2003).

Furosemide was detected in concentration ranges from 71.8–13,900 ng/l in WWTP influent samples and 47.3–11,400 ng/l in WWTP effluent samples, with the highest concentrations observed at the main hospital Fossvog and the hospital in Klaksvik (Table 20, Table 30 and Figure 20). These results can be considered as exceptions, because concentrations in all other samples were much lower. Previous studies of WWTP influents in Denmark and Norway, reported lower maximum concentrations at 6.3 µg/l and 5.75 µg/l, respectively (Mogensen et al., 2008; Møskeland et al., 2006). Similar observations can be done for furosemide levels in WWTP effluent samples where lower concentrations than in the present study were found in Denmark (4.6 µg/l; Mogensen et al., 2008; Kjølholt et al., 2003) and Norway (1.9 µg/l; Møskeland et al., 2006).

Hydrochlorothiazide was detected in concentration ranges of 87.9–1960 ng/l and 6.26–984 ng/l in WWTP influents and effluents, respectively (Table 20). The highest concentrations were detected at the main hospital Fossvog (Figure 20 C) and in Hveragerði (Figure 20 B), both in Iceland. There are no other data on hydrochlorothiazide in Nordic countries available.

The ACE inhibitors enalapril and its active form enalaprilat were both detected in eight out of nine WWTP influents and in all WWTP effluents (Table 19). Concentrations of enalapril were generally higher compared to enalaprilat in both matrices and corresponding samples (Figure 22). Only in two influent and effluent samples were the levels of enalaprilat higher (Akureyri and Hveragerði; Figure 22 B). Previously, a study from Denmark reported enalapril concentration levels from 170–680 ng/l in WWTP influent samples (Mogensen et al., 2008) and 50–230 ng/l in WWTP effluent samples (Mogensen et al., 2008; Kjølholt et al., 2003), which are higher than in the present study. There are no other published Nordic studies on enalaprilat in the environment available.

The ACE inhibitors perindopril and its active form perindoprilat showed low detection frequencies. Perindopril was detected in three out of nine and in three out of 12 WWTP influent and effluent samples, respectively. Perindoprilat was only detected in five out of 12 WWTP effluents (Table 19). Generally, concentrations of few ng/l were found. An exception was the WWTP influent sample from the main hospital Fossvog in Iceland where the concentration was 190 ng/l (Table 20 and Figure 22). There are no other data on perindopril and perindoprilat in the Nordic countries available.

The *angiotensin II receptor blocker* candesartan was detected in seven out of nine and six out of 12 WWTP influent and effluent samples, respectively, while losartan was detected in all WWTP samples (Table 19). Can-

desartan were found in the concentration range < 2.08–1,040 ng/l with highest influent concentration at Fossvog main hospital (IS, Figure 22). The effluent candesartan concentrations range was <2.08–251 ng/l, with the highest concentration found at the hospital in Klaksvik in the Faroe Islands (Table 20, Figure 20 and Figure 22). Losartan showed concentration ranges from 25.8–8,700 ng/l, with highest influent concentrations at the hospital Fossvog (IS; Figure 20) and highest effluent concentration at 327 ng/l in Hveragerði (IS; Table 20 and Figure 20) There are no other data on candesartan and losartan in the Nordic countries available.

The *β-adrenergic receptor blocker* atenolol was detected in eight out of nine and nine out of 12 WWTP influent and effluent samples, respectively, while metoprolol was detected in all wastewater samples (Table 19).

Atenolol was present at concentrations ranging from <20.8 to 12,700 ng/l with highest influent concentration at Fossvog main hospital (IS; Figure 20 C). In effluents, atenolol ranged between < 20.8 and 1,730 ng/l with the highest concentrations in Hveragerði (Table 20 and Figure 20). In previous studies from Finland atenolol was detected in WWTP influent and effluent samples at average concentrations of 800 and 300 ng/l, respectively (Vieno, 2008). In Sweden, atenolol was detected in higher concentrations in influent samples (330–4,900 ng/l; Fick et al. 2011) than in the present study but at lower levels in effluents (130–920 ng/l). Similarly, atenolol was detected in lower concentrations in effluents from Oslo, Norway and Uppsala, Sweden (Langford and Thomas, 2009; (Daneshvar et al., 2010).

Metoprolol was found in the range 14.4–404 ng/l, with the highest influent concentration found at the main hospital in the Faroe Islands (Figure 20 C). In WWTP effluents, metoprolol ranged between 51.2 and 810 ng/l with the highest concentration observed at the hospital in Klaksvik (Table 20 and Table 20 C). Earlier, Finnish and Swedish surveys reported much higher concentrations of metoprolol in WWTP influent samples with an average concentration of 1,060 ng/l and 2,580 ng/l, respectively (Vieno, 2008; Fick et al., 2011). Concentrations in WWTP effluents from Finland (Vieno, 2008) and Norway (Thomas et al., 2007) were lower than in the present study. The Swedish study reported higher concentrations in WWTP effluents (130–2,800 ng/l; Fick et al. 2011).

The *phosphodiesterase inhibitor* dipyridamole was detected in all WWTP influent samples and in 10 out of 11 effluent samples (Table 19). Concentration levels ranged between 422 and 166,000 ng/l and from < 16.5 to 24,600 ng/l in WWTP influents and effluents, respectively (Table 20). Highest concentrations for both matrices were found in Torshavn (Figure 19 and Appendix Table 30). There are no other published Nordic studies on dipyridamole in WWTP waters available.

The *hypolipidemic* simvastatin was not detected in WWTP waters (Table 19 and Figure 21). Previously, low ng/l concentrations of simvastatin were reported in Norwegian influent and effluent wastewaters (Langford and Thomas, 2009).

The *anticoagulant* warfarin was detected in two out of nine influent samples only and in one out of 11 WWTP effluent samples (Table 19). Warfarin was found in WWTP influents in Hveragerði (1.48 ng/l) and in Torshavn (3.21 ng/l; Figure 21 and Appendix Table 30). In WWTP effluents it was only detected in Hveragerði, and then close to the LOD (Table 20 and Figure 21). Previously, low ng/l concentrations of warfarin were reported for Norwegian wastewaters (Langford and Thomas, 2009).

Table 19. Detection frequency of cardiovascular drugs. N number of samples d number of detections

	Influent			Effluent			Sludge			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
Amiloride	100	(9 ; 9)		67	(12 ; 8)		100	(6 ; 6)		90	(10 ; 9)		83	(6 ; 5)	
Amlodipine	22	(9 ; 2)		82	(11 ; 9)		100	(6 ; 6)		0	(10 ; 0)		33	(6 ; 2)	
Atenolol	89	(9 ; 8)		75	(12 ; 9)		67	(6 ; 4)		0	(10 ; 0)		17	(6 ; 1)	
Bendroflumethiazide	0	(9 ; 0)		36	(11 ; 4)		38	(8 ; 3)		20	(10 ; 2)		17	(6 ; 1)	
Candesartan	78	(9 ; 7)		50	(12 ; 6)		17	(6 ; 1)		20	(10 ; 2)		0	(6 ; 0)	
Dipyridamole	100	(9 ; 9)		91	(11 ; 10)		100	(6 ; 6)		0	(10 ; 0)		100	(6 ; 6)	
Enalapril	89	(9 ; 8)		100	(12 ; 12)		100	(6 ; 6)		60	(10 ; 6)		0	(6 ; 0)	
Enalaprilat	89	(9 ; 8)		100	(12 ; 12)		0	(6 ; 0)		40	(10 ; 4)		17	(6 ; 1)	
Furosemide	100	(9 ; 9)		100	(11 ; 11)		100	(8 ; 8)		30	(10 ; 3)		17	(6 ; 1)	
Hydrochlorothiazide	100	(9 ; 9)		100	(12 ; 12)		50	(6 ; 3)		0	(10 ; 0)		0	(6 ; 0)	
Losartan	100	(9 ; 9)		100	(12 ; 12)		100	(6 ; 6)		30	(10 ; 3)		17	(6 ; 1)	
Metoprolol	100	(9 ; 9)		100	(11 ; 11)		100	(6 ; 6)		60	(10 ; 6)		50	(6 ; 3)	
Perindopril	33	(9 ; 3)		25	(12 ; 3)		0	(6 ; 0)		0	(10 ; 0)		0	(6 ; 0)	
Perindoprilat	0	(9 ; 0)		42	(12 ; 5)		0	(6 ; 0)		40	(10 ; 4)		0	(6 ; 0)	
Simvastatin	0	(9 ; 0)		0	(12 ; 0)		0	(6 ; 0)		0	(10 ; 0)		17	(6 ; 1)	
Warfarin	22	(9 ; 2)		9	(11 ; 1)		38	(8 ; 3)		0	(10 ; 0)		0	(6 ; 0)	

Table 20. Maximum and minimum concentrations of cardiovascular drugs in ng/l for water samples and µg/kg for sludge and sediment samples

	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
Amiloride	1,260	18.9	217	<3.03	93.6	0.93	552	<3.03	21.0	<0.10
Amlodipine	247	<4.17	448	<4.17	286	13.1	nd	<4.17	9.58	<1.43
Atenolol	12,700	<20.8	1,730	<20.8	1,650	<5.00	nd	<20.8	58.6	<5.00
Bendroflumethiazide	nd	<0.42	7.00	<0.42	3.23	<0.89	1.00	<0.42	1.39	<0.89
Candesartan	1,040	<2.08	251	<2.08	49.7	<0.50	5.34	<2.08	nd	<0.50
Dipyridamole	166,000	422	24,600	<16.5	1,880	3.62	nd	<16.0	14.2	1.86
Enalapril	522	<0.10	322	1.58	2.38	0.12	5.87	<0.10	nd	<0.03
Enalaprilat	178	<2.34	73	10.1	nd	<2.08	10.7	<2.34	2.12	<2.08
Furosemide	13,900	71.8	11,400	47.3	686	2.23	48.6	<4.17	2.70	<0.17
Hydrochlorothiazide	1,960	87.9	984	6.26	168	<5.00	nd	<2.08	nd	<5.00
Losartan	8,700	25.8	327	21.5	74.8	1.80	5.03	<2.08	392	<0.50
Metoprolol	404	14.4	810	51.2	549	14.7	23.7	<0.50	62.8	<0.10
Perindopril	190	<2.08	18	<2.08	nd	<2.08	nd	<2.08	nd	<2.08
Perindoprilat	nd	<2.08	13.0	<2.08	nd	<2.08	2.91	<2.08	nd	<2.08
Simvastatin	nd	<20.8	nd	<20.8	nd	<5.00	nd	<20.8	31.9	<5.00
Warfarin	3.00	<0.80	1.00	<0.80	0.18	<0.10	nd	<0.80	nd	<0.10

WWTP sludge

The *antianginal drug* amlodipine was detected in 100% of the WWTP sludge samples (Table 19) with concentrations ranging from 13.1 to 286 µg/kg (Table 20). The highest concentrations were found in Torshavn WWTP and the WWTP of the main hospital in the Faroe Islands (Figure 21). Amlodipine has been found in concentrations up to 310 µg/kg in an earlier study in Denmark (Mogensen et al., 2008).

The *diuretics* amiloride and furosemide appeared in 100% of the sludge samples, while bendroflumethiazide and hydrochlorothiazide were detected only in three out of eight and three out of six of these, respectively (Table 19).

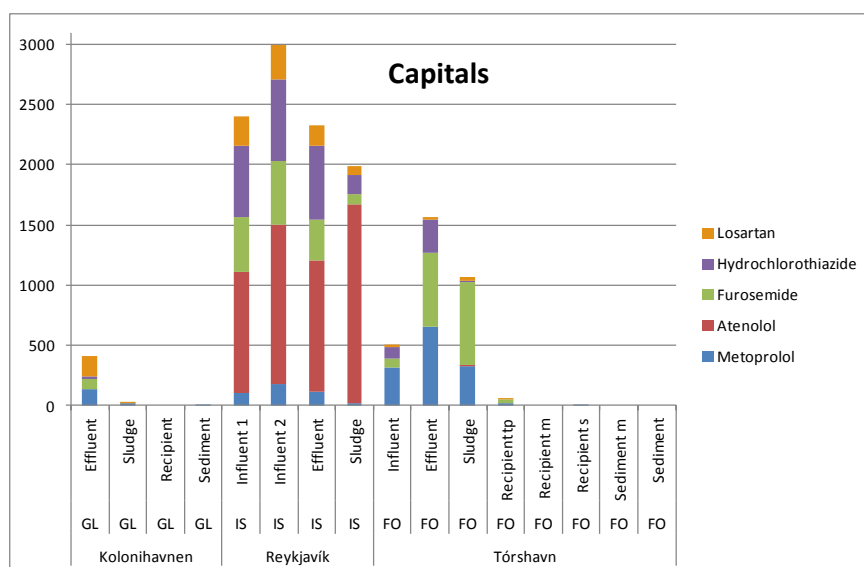
The highest concentration of amiloride at 93.6 µg/kg was found in Akureyri (Table 20 and Figure 21), whereas concentrations measured at the other locations were much lower. There are no other data on amiloride in Nordic countries available.

Bendroflumethiazide was detected in low and sub µg/kg concentrations (Appendix Table 30) in Kolonihavnen and the main hospitals of Greenland and Faroe Islands (Figure 21). A previous study from Denmark reported quite high concentrations of bendroflumethiazide in WWTP sludge samples, with the highest concentration being 1 200 µg/kg (Mogensen et al., 2008).

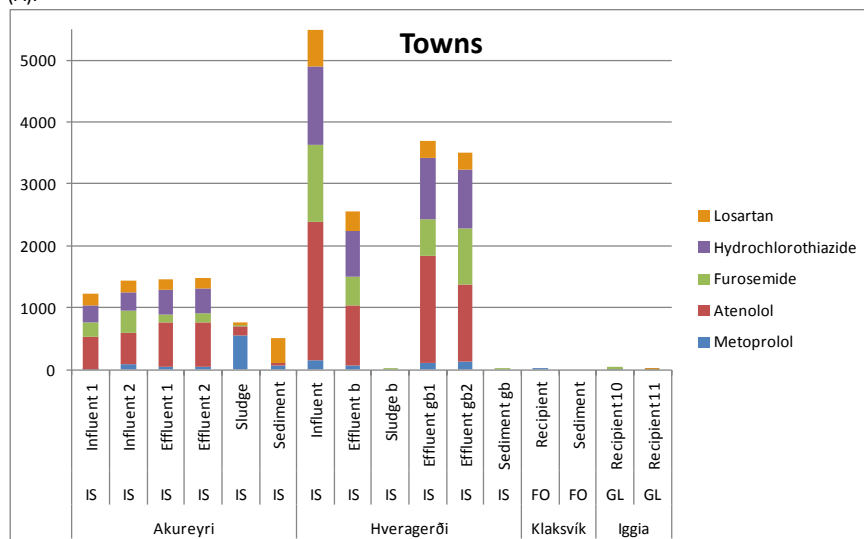
Furosemide ranged between 2.23 and 686 µg/kg (Table 20); by far, the highest concentration was detected in Torshavn (Figure 20). Earlier studies from Denmark and Norway found lower concentrations levels than those found in the Faroe Islands in the present study (Mogensen et al., 2008; Møskeland et al., 2006).

Hydrochlorothiazide appeared in the concentration range from < 5.00 to 1 686 µg/kg (Table 20). In this case, the highest concentration was in Reykjavik (Figure 20 A). There are no other data on hydrochlorothiazide from the Nordic countries available.

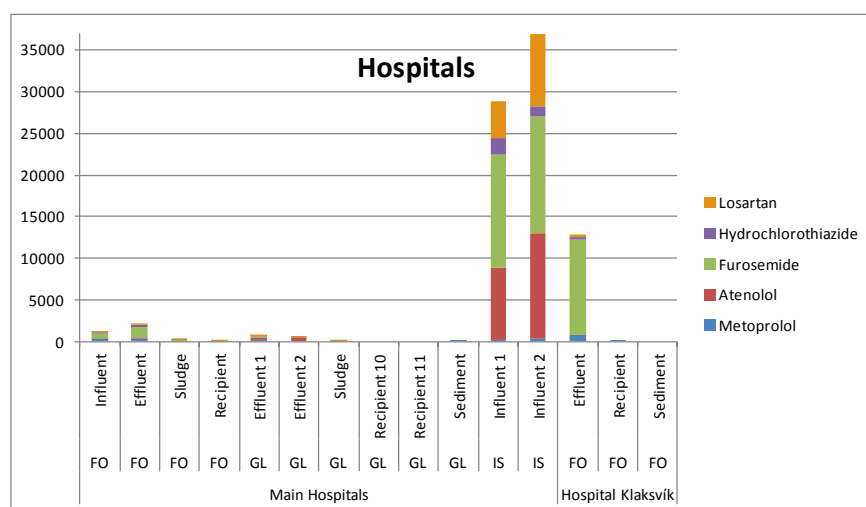
Figure 20. Selected cardiovascular drugs (β -adrenergic receptor blockers, diuretics and angiotensin II receptor blocker) in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and $\mu\text{g}/\text{kg}$ for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

The *ACE inhibitors* enalapril was detected in all WWTP sludge samples, while its active form enalaprilat, as well as perindopril and its active form perindoprilat were not detected in any of the WWTP sludge samples (Table 19). Enalapril was found at the low and sub $\mu\text{g}/\text{kg}$ level, ranging from 0.12 to 2.38 $\mu\text{g}/\text{kg}$ (Table 20 and Figure 22). In an earlier study from Denmark, enalapril was not detected in sewage sludge samples (Mogensen et al., 2008). There are no other data from Nordic studies on enalaprilat, perindopril and perindoprilat available.

The *angiotensin II receptor blocker* candesartan was detected in one out of six sewage sludge samples, while losartan was detected in all such samples (Table 19). Candesartan was found in Torshavn in a concentration of 49.7 $\mu\text{g}/\text{kg}$ (Figure 22 A). Losartan appeared in the concentration range from 1.80 to 74.8 $\mu\text{g}/\text{kg}$ (Table 20) with the highest concentration in Reykjavik WWTP (Figure 21 A). There are no other data on candesartan and losartan in the Nordic countries available.

The *β -adrenergic receptor blocker* atenolol was detected in four out of six sewage sludge samples, while metoprolol was detected in all these (Table 19).

Atenolol was found in levels of < 5.00–1 650 $\mu\text{g}/\text{kg}$ (Table 20) with the particularly high concentration of 1 650 $\mu\text{g}/\text{kg}$ found in sewage sludge from Reykjavik (Figure 21 A). In an earlier Swedish survey, sewage sludge samples contained atenolol concentrations from 12 to 38 $\mu\text{g}/\text{kg}$ (Fick et al., 2011).

Metoprolol were found in concentrations ranging from 14.7 to 549 $\mu\text{g}/\text{kg}$ (Table 20) with highest concentrations in Akureyri (Figure 20 B)

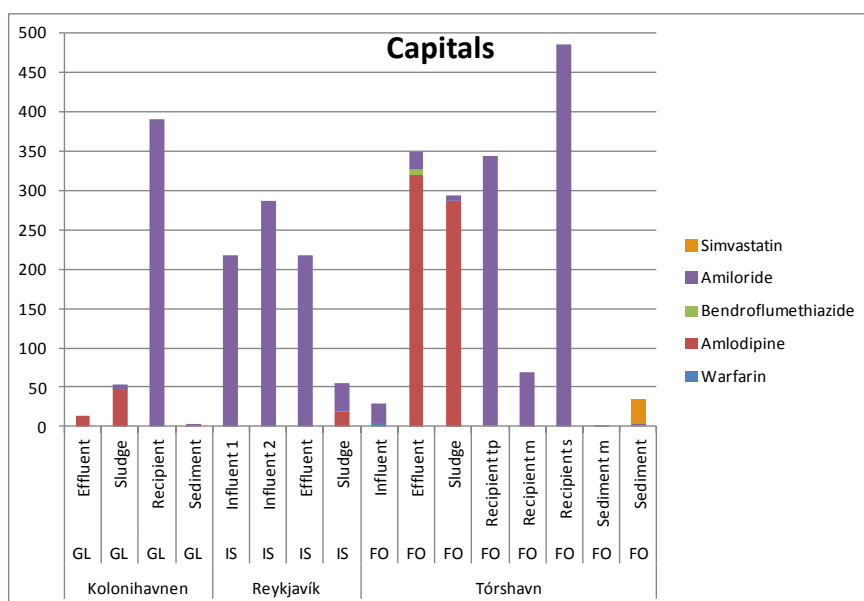
and in Torshavn (Figure 20 A). Previous studies in Norway and Sweden reported lower concentrations of metoprolol in sewage sludge samples; the highest concentration in the former was 21 µg/kg (Thomas et al., 2007) and 410 µg/kg (Fick et al., 2011) in the latter.

The *phosphodiesterase inhibitor* dipyridamole was detected in 100% of the WWTP sludge samples (Table 19). Concentrations ranged from 3.62–1 880 µg/kg (Table 20) with highest concentrations in Torshavn and the main hospital in Faroe Island (Figure 19). There are no other data on dipyridamole in WWTP sludge samples from the Nordic countries available.

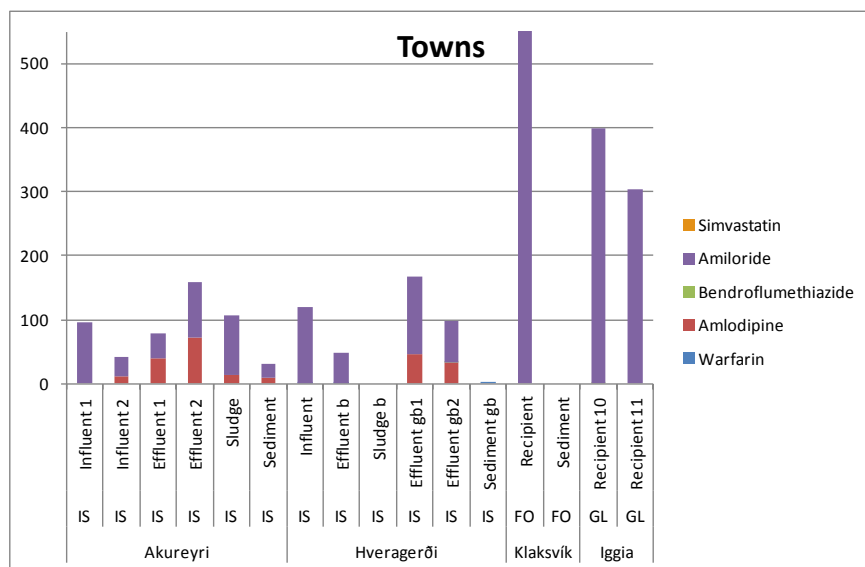
The *hypolipidemic* simvastatin was not detected in WWTP sludge (Table 19 and Figure 21), and there are no data available for this compound in any other Nordic study.

The *anticoagulant* warfarin was detected in three out of eight WWTP sludge samples (Table 19) at concentrations close to the LOD (Table 20 and Figure 21). As with simvastatin, there are no other published data from Nordic studies.

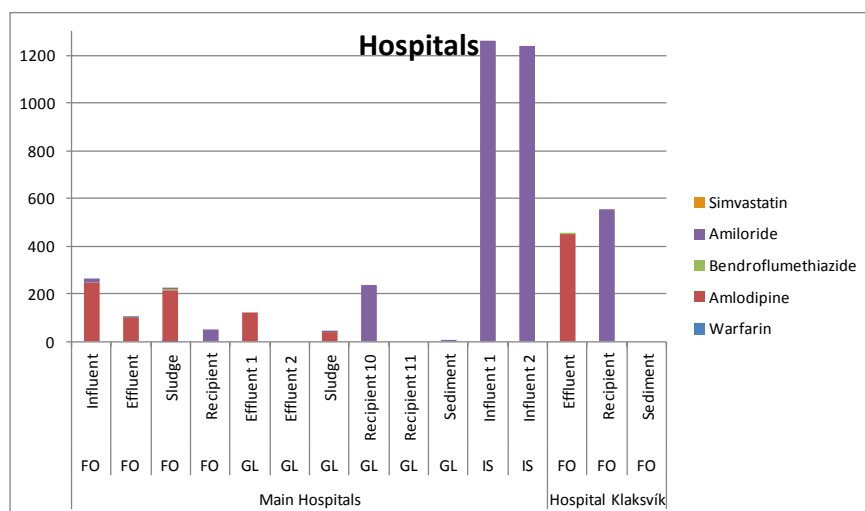
Figure 21. Selected cardiovascular drugs (anticoagulant, calcium channel blocker, diuretics and hypolipidemic) in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and µg/kg for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

Recipient water and sediment

The *antianginal drug* amlodipine was not detected in recipient waters, whereas it was detected in two out of six sediment samples, (Table 19) where it was found in low concentrations in Akureyri (Figure 21 B) and in Kolonihavnen (Figure 21 A). In an earlier study in Denmark, amlodipine was not detected in surface waters (Mogensen et al., 2008).

The *diuretic* amiloride appeared with a relatively high detection frequency in nine out of 10 recipient water samples and five out of six sediment samples, while bendroflumethiazide and furosemide had low detection frequencies. Hydrochlorothiazide was not detected in any recipient water or sediment sample (Table 19). There are no other data available on hydrochlorothiazide in Nordic countries.

Amiloride was present at concentrations ranging from < 3.03 to 552 ng/l in recipient waters and from < 0.10 to 21.0 µg/kg in sediment samples (Table 20), with the highest concentrations in Klaksvik harbour and in Akureyri, respectively (Figure 21). There are no other data available on amiloride in the Nordic countries.

Bendroflumethiazide was detected at low concentrations only in recipient waters in Torshavn (Figure 20 A) and Iggia (Figure 20 B) and in one sediment sample from Torshavn (Figure 20 A). It was not detectable in an earlier investigation of recipient waters from Denmark (Mogensen et al., 2008).

Furosemide ranged from < 4.17 to 48.6 ng/l (Table 20) in recipient water samples, with the highest concentration found in Iggia in 2010 (Figure 20). Furosemide in sediments was only detected in Akureyri, and then at a concentration of 2.7 µg/kg (Figure 20). Previous studies have reported similar concentrations for recipient waters in Norway (Møskeland et al., 2006), but not for Denmark, where it has not been detected (Mogensen et al., 2008). Furosemide was not detected in sediment samples from Norway (Møskeland et al., 2006).

The *ACE inhibitors* enalapril and its active form enalaprilat were detected in six and four out of 10 recipient waters, respectively (Table 19), at concentration ranges from < 0.10 to 5.87 ng/l for enalapril and < 2.34–10.7 ng/l for enalaprilat (Table 20). Highest concentrations were found in surface waters in Torshavn both those taken outside the Sersjantvîkin WWTP and those off the main hospital (Figure 22). Enalaprilat was detected in one sediment sample only, one from Akureyri, and then close to the LOD. In a previous study from Denmark enalapril was not found in recipient water samples (Mogensen et al., 2008). There are no other published Nordic studies dealing with enalaprilat in the environment.

Perindopril was not detected in recipient waters or sediment samples. Its active form perindoprilat was detected in four out of 10 recipient water samples, but not in sediment samples (Table 19). The concentrations of perindoprilat were low and close to the LOD (Appendix Table 30) at all locations. No other data on perindopril and perindoprilat in the environment of the Nordic countries are available.

The *angiotensin II receptor blocker* candesartan was detected in two out of 10 recipient water samples but not in sediment samples, while losartan was detected in three out of 10 recipient waters and in one out of six sediment samples (Table 19). Candesartan and losartan were found in Torshavn also at the site receiving effluents from the main hospital in the Faroe Islands with low concentrations in recipient waters (Figure 21, Figure 22 and Appendix Table 30). Losartan was also detected in recipient waters from Iggia (5 ng/l) and in surprisingly high concentration in the sediment from Akureyri (392 µg/kg; Figure 21 B). There are no other published Nordic studies on candesartan and losartan available.

The *β-adrenergic receptor blocker* atenolol was not detected in recipient water samples, but only in one sediment sample (Table 19) from Akureyri (Figure 20 B). Previously, atenolol had been detected in Sweden in recipient waters with the highest concentration being 390 ng/l (Fick et al., 2011) and with a mean concentration 38 ng/l (Daneshvar et al., 2010).

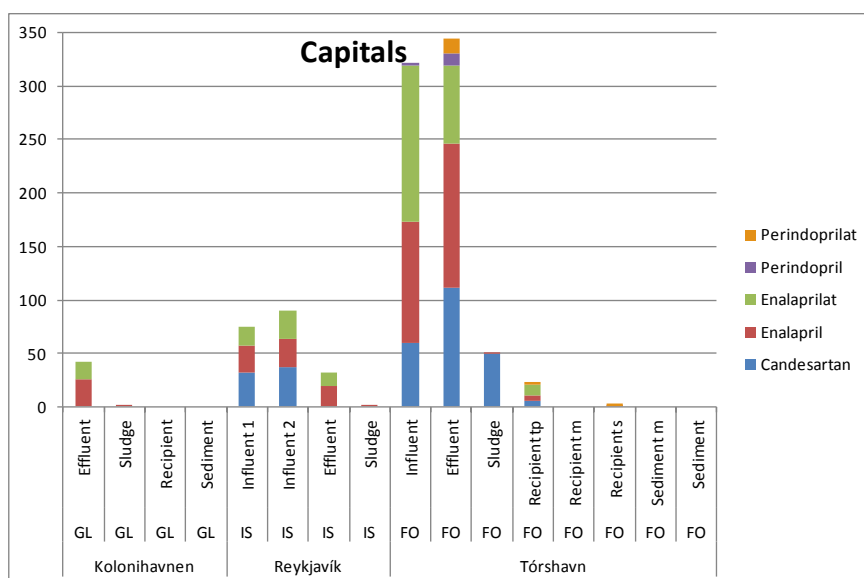
Metoprolol was detected in six out of 10 recipient water samples and in three out of six sediment samples (Table 19). Concentrations ranged between < 0.50 and 23.7 ng/l and 0.10–62.8 µg/kg (Table 20), with highest concentrations at the main hospital in Greenland and in Torshavn Faroe Islands, respectively (Figure 20). Earlier Swedish studies had reported higher concentrations of metoprolol in recipient water samples (Fick et al., 2011; Daneshvar et al., 2010).

The *phosphodiesterase inhibitor* dipyridamole was not detected in recipient water samples, but occurred in all sediment samples (Table 19). Concentrations from 1.86 to 14.2 µg/kg were measured in sediment samples with highest concentration in Akureyri, followed by Kolonihavnen with 4.04 µg/kg (Figure 20). There are no other published Nordic studies on dipyridamole in recipient water and sediment samples available.

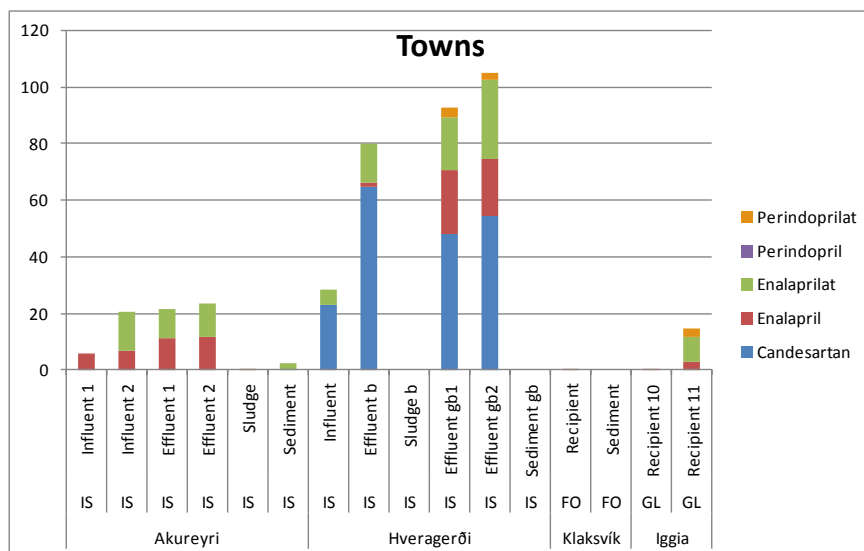
The *hypolipidemic* simvastatin was not detected in recipient water samples but occurred in one sediment sample (Table 19), that from Torshavn at a concentration of 31.9 µg/kg (Figure 20). There are no other published Nordic studies on simvastatin in recipient water and sediment samples available.

The *anticoagulant* warfarin was not detected in recipient water and sediment samples (Table 19). There are no other published data available on warfarin in recipient water and sediment samples in a Nordic country.

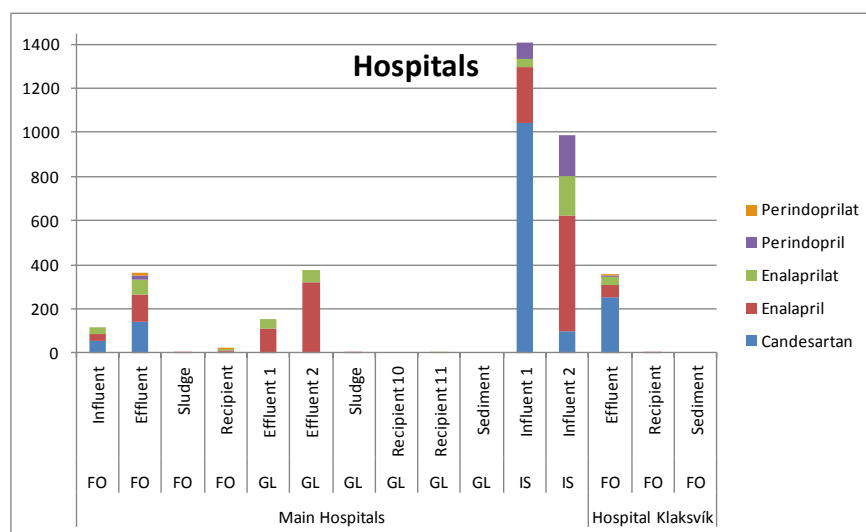
Figure 22. Selected cardiovascular drugs (angiotensin II receptor blocker and ACE inhibitors) in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and µg/kg for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

5.6 Hormones

17 α -Ethinylestradiol was not detected in any of the samples in this study (Table 21). In previous surveys it was detected in WWTP influents and effluents from Denmark (Ingerslev et al., 2003; Kjølholt et al., 2003; Stuer-Lauridsen et al., 2005), Norway (Møskeland et al., 2006; Thomas et al., 2007) and Sweden (Andersson et al., 2006; Fick et al., 2011) in quite high concentrations. In WWTP sludge, 17 α -Ethinylestradiol was only investigated and detected in Norway (Møskeland et al., 2006; Thomas et al., 2007). Surface waters were characterized by low ng/l levels in Denmark (Ingerslev et al., 2003; Stuer-Lauridsen et al., 2005) and Sweden and non-detects in Norway (Møskeland et al., 2006). For sediment samples, concentrations in Norway (Møskeland et al., 2006) and Sweden (Andersson et al., 2006) were below the LOD. Analysis of 17 α -Ethinylestradiol is a challenge, since there are limitations in achieving low LOD's even when using the newest generation of analytical instrumentation.

Table 21. Detection frequency of hormones. N number of samples d number of detections

	Influent			Effluent			Sludge			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
17 α Ethinylestradiol	0	(9 ; 0)	0	0	(12 ; 0)	0	0	(6 ; 0)	0	0	(10 ; 0)	0	0	0	(6 ; 0)
17 β Estradiol	33	(9 ; 3)	3	33	(12 ; 4)	4	17	(6 ; 1)	1	0	(10 ; 0)	0	0	0	(6 ; 0)
Estriol	67	(9 ; 6)	6	86	(12 ; 10)	10	100	(6 ; 6)	6	0	(10 ; 0)	0	0	0	(6 ; 0)
Estrone	67	(9 ; 6)	6	83	(12 ; 10)	10	100	(6 ; 6)	6	80	(10 ; 8)	8	50	(6 ; 3)	3
Levothyroxine	22	(9 ; 2)	2	17	(12 ; 2)	2	83	(6 ; 5)	5	0	(10 ; 0)	0	0	0	(6 ; 0)

WWTP influent and effluent

17 β -Estradiol was detected in three out of nine WWTP influent samples and in four out of 12 WWTP effluent samples with highest concentrations of 473 ng/l at the hospital Fossvog (IS) and 375 ng/l at the Queen Ingrid's Hospital in Greenland, respectively (Table 21, Table 22 and Figure 23). Previous studies undertaken in Denmark (Stuer-Lauridsen et al., 2006; Kjøl t et al., 2003; Ingerslev et al., 2003), Norway (Møskeland et al., 2006; Thomas et al., 2007) and Sweden (Andersson et al., 2006; Fick et al. 2011) reported much lower concentrations in WWTP influents and effluents, with maximum values in Sweden of 25.5 ng/l and 67.7 ng/l, respectively.

Estriol was detected in six out of nine WWTP influent samples (Table 21) at concentration levels from <20.8 to 98.7 ng/l (Table 22) with the highest concentration observed in Reykjavik (Figure 23). In WWTP effluent samples, estriol was detected in 10 out of 12 samples (Table 21) with the highest concentration of 198 ng/l in the WWTP of the main hospital in the Faroe Islands (Table 22 and Figure 23). Previous studies performed in Norway (Thomas et al., 2007) and Sweden (Andersson et al., 2006; Fick et al., 2011) reported estriol in lower concentrations in WWTP influents and effluents.

Estrone was detected in six out of nine WWTP influent samples and 10 out of 12 WWTP effluent samples (Table 21). The measured concentration levels ranged from < 3.73 to 141 ng/l and from < 3.73 to 21 ng/l, respectively (Table 22). The highest concentrations were found in the two WWTP influent samples from the hospital Fossvog (IS) and the sewage line effluent from main hospital in Greenland (Figure 26). Other national surveys detected estrone in Denmark (Stuer-Lauridsen et al., 2005; Kjøl t et al., 2003; Ingerslev et al., 2003) in higher concentrations than in the present study, whereas in Norway, concentrations were higher in influent (Møskeland et al., 2006) and lower in the effluent samples (Møskeland et al., 2006; Thomas et al., 2007).

Levothyroxine was detected in two out of nine WWTP influent samples, and in two out of 12 effluent samples (Table 21). The highest concentration was 2.7 ng/l and was observed in the two samples from the main hospital in Fossvog in Iceland (Table 22, Figure 23 and Appendix Table 31). Previously, a Finnish study detected 64 ng/l in the WWTP influent and 22 ng/l in the WWTP effluent sample from Turku (Svanfeldt et al., 2010).

Table 22. Maximum and minimum concentrations of hormones in ng/l for water samples and µg/kg for sludge and sediment samples

	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
17 α Ethinylestradiol	nd	<208	nd	<208	nd	<50.0	nd	<208	nd	<50.0
17 β Estradiol	473	<208	375	<208	77.7	<50.0	nd	<208	nd	<50.0
Estriol	98.7	<20.8	198	<20.8	210	5.59	nd	<20.8	nd	<5.00
Estrone	141	<3.73	21	<3.73	64.4	6.89	2.09	<3.73	1.11	<0.66
Levothyroxine	2.70	<1.61	2.00	<1.61	14.3	<1.04	0.00	<1.61	nd	<1.04

WWTP sludge

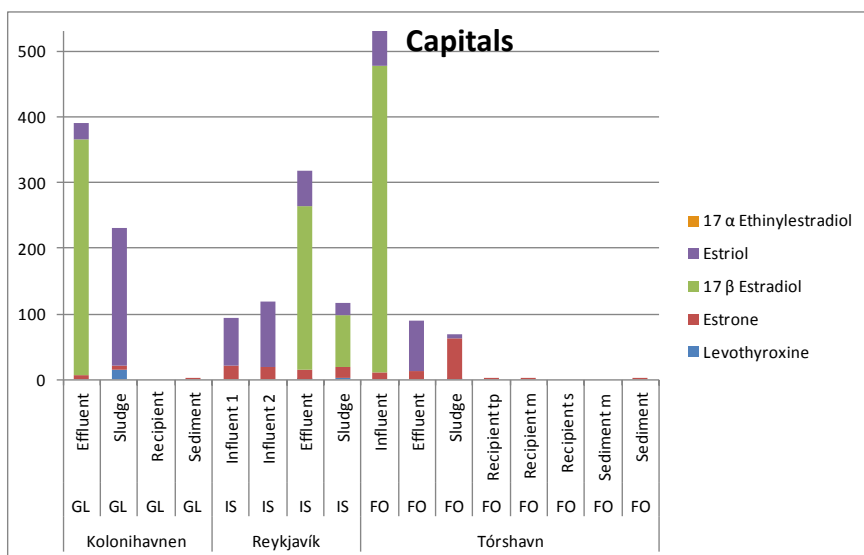
17β-Estradiol was detected in one out of six WWTP sludge samples at a concentration of 77.7 µg/kg in Reykjavik (Table 21, Table 22 and Figure 23). Previous national surveys performed in Norway (Møskeland et al., 2003) and Sweden (Andersson et al., 2006; Fick et al., 2011) reported approximately 10-fold lower concentrations in Norway, but higher maximum concentration, at 310 µg/kg, in Sweden.

Estriol was detected in all WWTP sludge samples (Table 21) showing concentration levels from 5.59 to 210 µg/kg (Table 22). The highest concentration was observed in Kolonihavnen (Figure 23). A previous study from Norway (Thomas et al., 2007) reported concentrations similar to the lowest concentrations found in the present study. In contrast, much higher concentrations were detected in Sweden (Andersson et al., 2006; Fick et al., 2011).

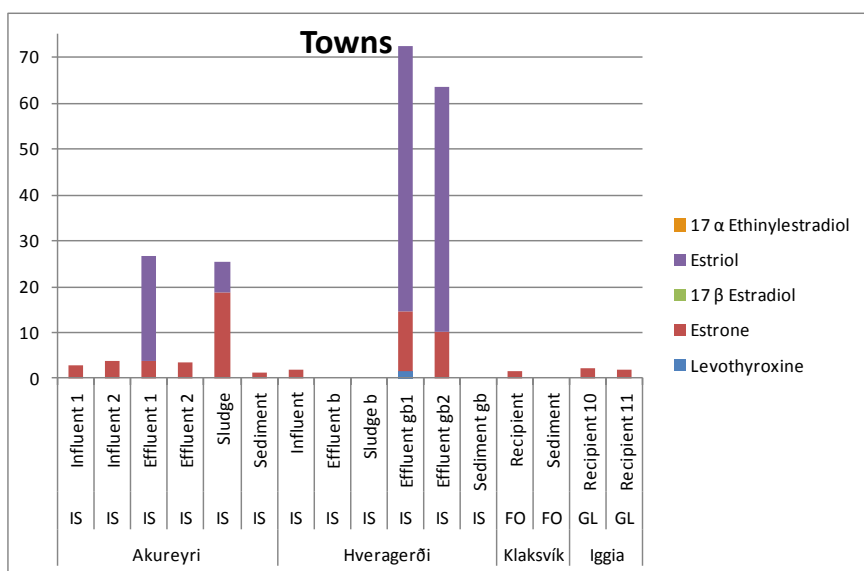
Estrone was detected in all WWTP sludge samples (Table 21) at concentration levels from 6.89 to 64.4 µg/kg (Table 22) with the highest concentration found in Torshavn (Figure 23 A). In previous Norwegian surveys estrone was found in lower concentrations (Møskeland et al., 2006; Thomas et al., 2007).

Levothyroxine was detected in five out of six WWTP sludge samples (Table 21) at concentration levels from <1.04–14.2 µg/kg (Table 22) with the highest concentration found in Kolonihavnen (Figure 23 A). Other data for WWTP sludge from Nordic countries are not available.

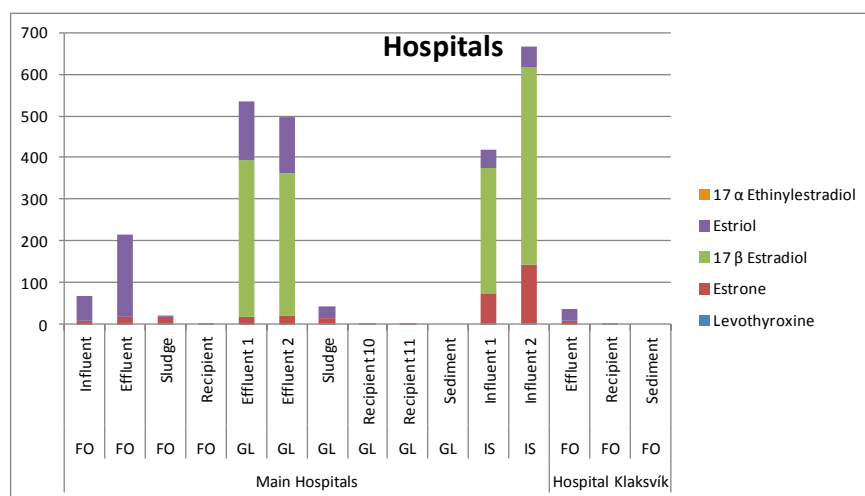
Figure 23. Hormones in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and µg/kg for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

Recipient water and sediment

Hormones were not detected in recipient waters. Only estrone was detected in eight out of 10 recipient water samples and in three out of six sediment samples (Table 21) with the highest concentration at 2.09 ng/l in Iggia Kolonihavnen and 1.4 µg/kg in Torshavn respectively (Table 22 and Figure 23). Estrone was reported in low ng/l concentrations in recipient waters from Denmark (Stuer-Lauridsen et al., 2005) and Norway (Møskeland et al., 2006; Thomas et al., 2007) and in sediments from Norway (Møskeland et al., 2006).

17β-Estradiol was reported in low to sub ng/l concentrations in recipient waters from other national surveys in Denmark, Norway and Sweden (Møskeland et al., 2006; Stuer-Lauridsen et al., 2005; Andersson et al., 2006; Fick et al., 2011) and in low µg/kg concentrations in sediments from Denmark (Møskeland et al., 2006).

Estriol was detected in low ng/l levels in recipient waters from Sweden, but not in sediments (Andersson et al., 2006; Fick et al., 2011). Levothyroxine was studied in surface waters near Turku (Finland) but was below LOD (Svanfelt et al., 2010). Other data for sediment samples from Nordic countries are not available.

5.7 Hypnotics

Zopiclone was the only drug analysed among this group of pharmaceuticals and it was not detected in any of the samples (Table 15, Table 18). In a previous Norwegian survey zopiclone was detected in quite high concentrations in WWTP influents (0.3–2.8 µg/l), WWTP effluents (< LOD–2.4 µg/l) and WWTP sludge (< LOD–1 100 µg/kg; Møskeland et al., 2006).

5.8 Additives in personal care products

5.8.1 Ethylenediaminetetraacetic acid (EDTA)

WWTP influent and effluent

EDTA was found in all WWTP water and sludge samples, in 50% of the recipient waters and in 33% of the sediments (Table 23). The high detection frequency of EDTA is probably due to its ubiquitous use and persistence in the environment (Allard et al., 1996; Remberger et al., 1997). EDTA was detected in influents and effluents mostly at low concentrations (9.0–49 µg/l; Table 24 and Figure 24) but in three hospital effluents the concentrations were an order of magnitude higher (420–630 µg/l; Figure 24 B). Comparison of the concentration in influent and effluent water gave few indications of reduction of EDTA in the WWTPs. Previous studies have shown that EDTA is persistent in WWTP (Alder et al., 1990; Reemtsma et al., 2006).

The detected concentrations in WWTP effluents in the present study (11–630 µg/l) are in the same range (79–310 µg/l) as reported previously from Norway (Schlabach et al., 2009) but the detection frequency was higher in present investigation.

WWTP sludge

All de-watered sludge samples contained detectable amounts of EDTA. The concentrations were generally low but one sample from the main hospital in Torshavn contained 750 µg/kg wet weights (ww). The EDTA in the sludge was probably dissolved in remaining pore water (Remberger, 2001). Sludge is considered not to be a sink for EDTA (Allard et al., 1996). The concentrations of EDTA in sludge in the present report also agreed with the results from Norway (Schlabach et al., 2009).

Recipient water and sediment

EDTA was detected in five out of ten recipient waters at low concentrations (1.3–1.8 µg/l; Tabel 24, Figure 24 and Figure 25). The low concen-

trations may be attributed to effective dilution. UV-degradation may occur but is not likely since the photoactive complex Fe-EDTA is not likely to exist under the conditions in question (Hering et al., 1988; Hudson et al., 1992; Deacon et al., 1994). The detected concentrations of EDTA in Oslo fjord and Tromsø sound were lower than reported in a previous survey (Schlabach et al., 2009).

EDTA in sediments was determined after removing most of the pore water by centrifugation. Low concentrations, between 1.3 and 1.8 µg/kg ww, were detected but could be attributed to some remaining pore water in the sediment which means that EDTA is not adsorbed to the solid phase in the sediments. This finding is in agreement with Allard et al., (1996) and Remberger et al., (2001).

EDTA was also detected in sediments affected by sewage from WWTP (1.3–40 µg/kg dw). EDTA was not detected in sediments in a previous study (Schlabach et al., 2009).

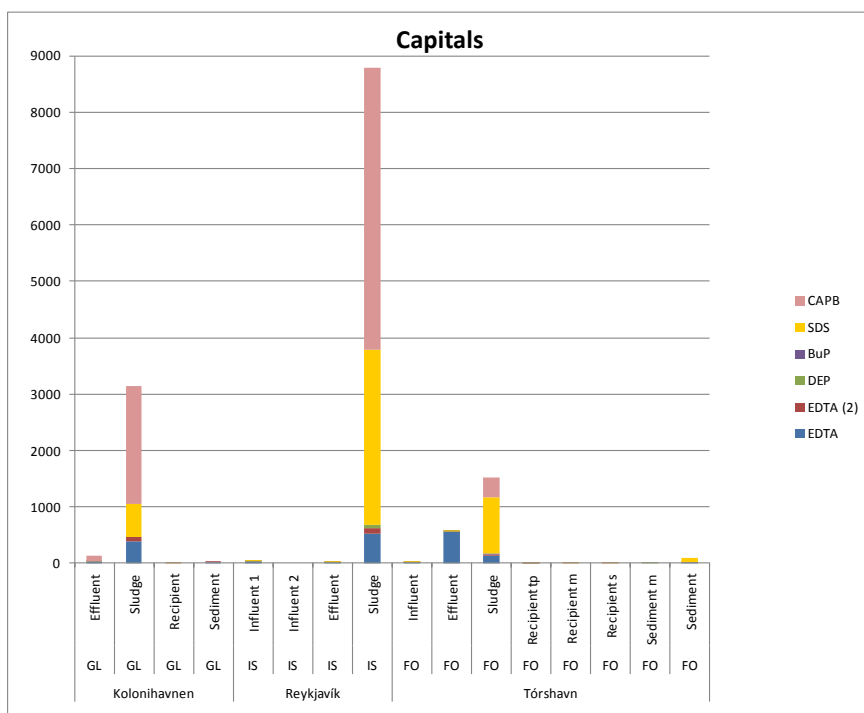
Table 23. Detection frequency of additives in personal care products. N number of samples d number of detections

	Influent			Effluent			Sludge			Recipient water			Sediment		
	%	N	d	%	N	d	%	N	d	%	N	d	%	N	d
EDTA	100	(6 ; 6)		100	(9 ; 9)		100	(8 ; 8)		50	(10 ; 5)		33	(6 ; 2)	
EDTA (2)							100	(8 ; 8)					33	(6 ; 2)	
Diethylphthalate	100	(6 ; 6)		100	(9 ; 9)		25	(8 ; 2)		20	(10 ; 2)		17	(6 ; 1)	
Butylparaben	100	(6 ; 6)		100	(9 ; 9)		50	(8 ; 4)		20	(10 ; 2)		0	(6 ; 0)	
SDS	100	(6 ; 6)		100	(9 ; 9)		100	(8 ; 8)		100	(10 ; 10)		33	(6 ; 2)	
SDSE01-4	100	(6 ; 6)		100	(9 ; 9)		100	(8 ; 8)		80	(10 ; 8)		17	(6 ; 1)	
CAPB	33	(6 ; 2)		67	(9 ; 6)		100	(8 ; 8)		80	(10 ; 8)		17	(6 ; 1)	
ATAC-C16	100	(6 ; 6)		100	(9 ; 9)		100	(8 ; 8)		40	(10 ; 4)		100	(6 ; 6)	

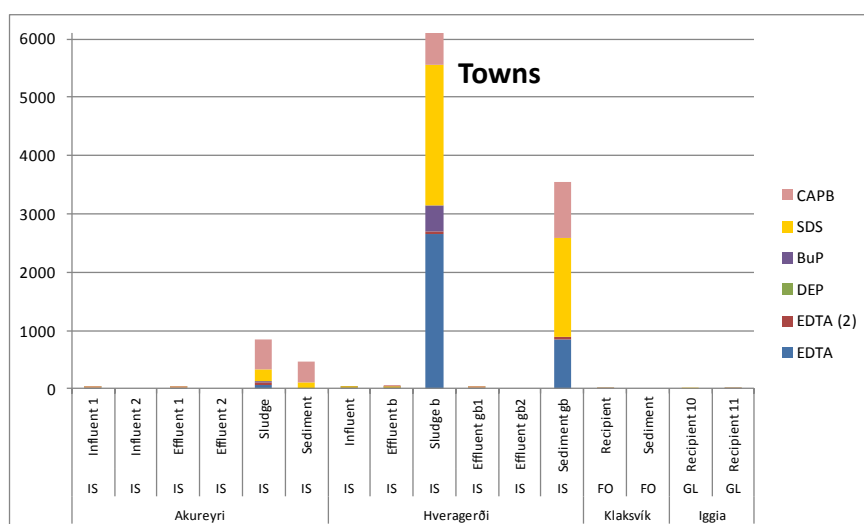
Table 24. Maximum and minimum concentrations of additives in personal care products in µg/l for water samples and µg/kg for sludge and sediment sample

	Influent ng/l		Effluent ng/l		Sludge µg/kg		Recipient water ng/l		Sediment µg/kg	
	max	min	max	min	max	min	max	min	max	min
EDTA (1)	49.0	9.00	630	11.0	6,800	62.4	1.80	<LOQ	4.85	<4.00
EDTA (2)	na	na	na	na	748	15.5	na	na	1.70	<2.00
Diethylphthalate	3.00	0.44	2.10	0.17	78.0	<LOQ	0.14	<LOQ	10.0	<2.00
Butylparaben	0.05	0.02	nd	0.01	440	<LOQ	0.01	<LOQ	nd	<2.00
SDS	8.00	0.85	6.00	0.79	3,100	210	0.41	<LOQ	110	<40.0
SDSE01-4	970	2.70	450	0.84	180,000	510	19.0	<LOQ	360	<80.0
CAPB	22.0	<LOQ	89.0	<LOQ	5,000	350	7.50	<LOQ	360	<20.0
ATAC-C16	87.0	1.28	31.0	1.55	680,000	1,700	1.64	<LOQ	1,500	<20.0

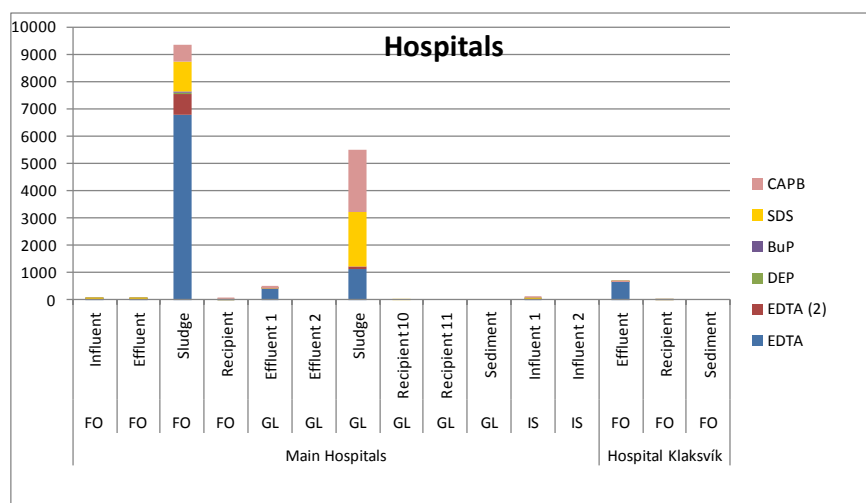
Figure 24. Selected additives in personal care products in different matrixes of capitals (A), towns (B) and hospitals (C) in ng/l for water samples and $\mu\text{g}/\text{kg}$ for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

5.8.2 Diethyl phthalate (DEP)

DEP was found in all WWTP influent and effluent water samples. The detection frequency in sludge was 25%, in recipient water 20% and in sediment 17% (Table 23).

WWTP influent and effluent

DEP was detected in all influents and effluent waters in the concentrations range 170–3,100 ng/l (Table 24). A comparison of influent and effluent waters in five WWTPs (Island and Faroe Islands) showed a reduction rate of 36–75%. The detected concentrations in WWTP effluents in the present investigation are in the same range as reported by Schlabach et al., (2009) but the detection frequency was higher in the present investigation.

WWTP sludge

De-watered sludge samples contained low concentrations of DEP in two out of eight cases (68–78 µg/kg dw; LOQ 8 µg/kg dw; Table 23 and Table 24). The low detection frequency in sludge is probably a result of the high water solubility of DEP (1,080 mg/l; ChemIDplus). The concentrations of DEP in sludge in this study agreed with the results from a previous study in Sweden and Norway (Schlabach et al., 2009).

Recipient water and sediment

Two out of 10 recipient water samples contained DEP in quantifiable concentrations (LOQ 30 ng/l) in the range 45 to 140 ng/l (Table 24). The

highest concentration was detected in Torshavn, Sersjantvíkin WWTP (140 ng/l; Figure 24 A).

DEP was determined in a previously study in water samples from Oslo fjord and Tromsø sound. The reported concentrations were 14–22 ng/l and 17–140 ng/l respectively. The low concentrations in environmental samples may be a result of dilution and bio- or UV- degradation.

DEP was detected in one out of six sediments (17%; Table 23) at a low concentration (9.9 µg/kg dw; LOQ 2 µg/kg dw; Table 24).

Schlabach et al., (2009) reported DEP in sediments from the recipient in Norway at higher frequency and concentration than in the present study.

5.8.3 Butylparaben (BuP)

BuP was found in all WWTP influent and effluent water samples. The detection frequency in sludge was 50% and in recipient water 20%. BuP was not detected in any sediment sample (<5 µg/kg dw; Table 23).

WWTP influent and effluent

BuP was detected in influents and effluents in the concentration range 11–110 ng/l (Table 24). The concentration in Greenland influent and effluent waters (81–109 ng/l) seems to be somewhat elevated compared to Iceland (11–54 ng/l) and the Faroe Islands (12–53 ng/l). A comparison of influent and effluent waters from WWTPs (Iceland and the Faroe Islands) showed an apparent reduction of the compound in the treatment process (9–78%) but in two out of five WWTPs (Hveragerði and Akureyri, Iceland) the concentration was higher in effluent compared to influent. The latter observation may be a result of the timing of the sampling of influent and effluent (Figure 24).

The detected concentrations in WWTP effluents in the present investigation are in the same range as reported previously from the Nordic countries (Schlabach et al., 2009). In a Swedish screening survey BuP was detected in one out of 14 effluents (100 ng/l; Remberger et al., 2005).

WWTP sludge

Four out of eight de-watered sludge samples (50%; Table 23) contained BuP above LOQ (5 µg/kg dw) in a concentration range of 5.3–440 µg/kg dw (Table 24). The high value was found in sludge from Hveragerði, Iceland (Figure 24). The low detection rate in sludge is probably a result of high water solubility (207 mg/l; ChemIDplus) and biodegradation. The detection frequency of BuP in sludge in the present study was higher than in a recent Swedish screening (4%; Remberger et al., 2005). The reported

concentration was 63 µg/kg dw (Remberger et al., 2005). No BuP was detected in sludge in a Norwegian study (Schlabach et al., 2009).

Recipient water and sediment

BuP could be quantified in two out of 10 recipient water samples (two from the Faroe Islands and one from Iceland; Table 23), with concentrations in the range 8–9.9 ng/l (LOQ 6 ng/l; Table 24). As with DEP, the low concentrations in environmental samples may be result of dilution and bio and UV-degradation.

BuP was not detected in any of the sediment samples (Table 23; LOQ 2 µg/kg dw).

Schlabach et al., (2009), detected BuP in receiving water in Oslo fjord (2–4 ng/l) and Tromsø sound (3 and 900 ng/l) in the same range as was detected in the present study but with one exception, one sample from Tromsø sound that showed a much higher level. BuP was not detected in sediments influenced of WWTP which agrees with Schlabach et al., (2009).

5.8.4 Sodium dodecyl sulphate (SDS)

SDS was found in all WWTP influent, effluents, sludge and recipient samples. The detection frequency in sediments was 33% (Table 23).

WWTP influent and effluent

SDS was detected in concentration range 0.79–7.9 µg/l (Table 24). Two samples were somewhat elevated compared to the others and both came from the Faroe Islands (Sersjantvíkin WWTP Torshavn influent 7.9 µg/l and WWTP Main Hospital effluent 5.6 µg/l; Figure 24). The concentrations in influent and effluent waters from five WWTPs were compared. The data are not conclusive. In two WWTP's an apparent reduction (46–90%) was observed, in one was the concentration unchanged and in two the concentration was higher in effluent compared to the influent. The detected concentrations in WWTP effluents in the present investigation are in the same range (0.3–9.6 µg/l) as reported previously from Norway (Schlabach et al., 2009) but the detection frequency was higher in the present investigation.

WWTP sludge

All eight de-watered sludge samples contained SDS in a concentration range of 210–3,100 µg/kg dw (Table 24). The highest concentration was found in sludge from Klettagørðum, Reykjavik (3,100 µg/kg dw; Figure 24 A) and Hveragerði (2,400 µg/kg dw; Figure 24 B). The lowest concentration was detected in Akureyri (210 µg/kg dw; Figure 24 B). The gen-

erally quite high concentrations suggest that the sludge may act as a sink for SDS. The concentrations of SDS in sludge in the present study agreed with previous results from Norway (Schlabach et al., 2009).

Recipient water and sediment

Recipient water samples contained SDS above the LOD in the range 0.04–0.41 µg/l (Table 23 and Table 24).

SDS was detected in two out of six sediment samples (Table 23). The two positive samples came from Iceland (Akureyri 110 µg/kg dw; Figure 24 B) and one from Faroe Islands (Torshavn harbour shipyard 93 µg/kg dw; Figure 24 A).

Schlabach et al., (2009) reported SDS in recipient water samples from Oslo fjord and Tromsø sound. The concentration was lower (<0.040–0.50 µg/l) than in the present investigation (Table 24). SDS not detected in Oslo fjord and Tromsø sound sediments.

5.8.5 Sodium laureth sulphate (SDSE01-4)

SDSE01-4 was found in all WWTP influent, effluent and sludge samples. The detection frequency in recipient water and sediments was 80% and 17% respectively (Table 23).

WWTP influent and effluent

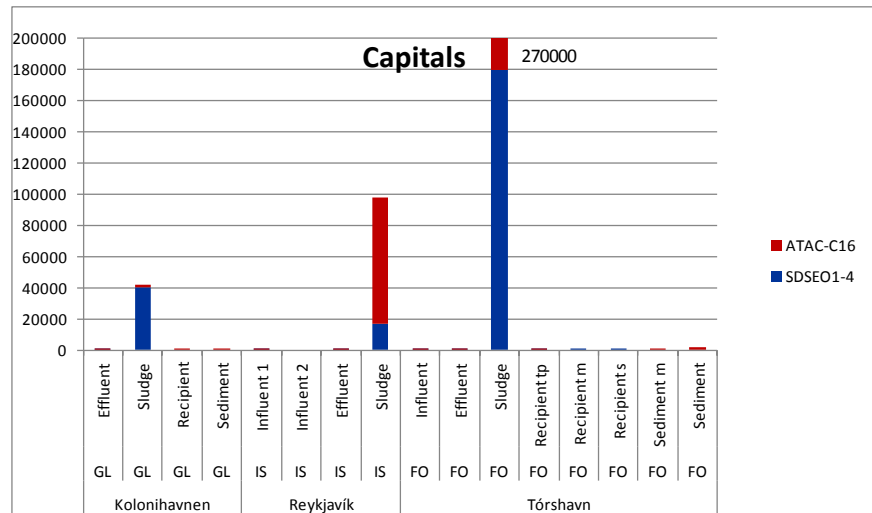
SDSE01-4 was detected in the concentration range 0.84–970 µg/l (Table 24) The highest concentrations were detected in the sample Landspítali Hospital Fossvog (970 µg/l; IS; Figure 25 C), Sersjantvíkin WWTP, Torshavn (510 µg/l; FO; Figure 25 C), Queen Ingrid's Hospital (330 µg/l; GL; Figure 25 C) and Kolonihavnen (350 µg/l; Figure 25 A). A comparison of the concentrations in influent and effluent water from five WWTP's (Island and Faroe Islands) showed an apparent reduction of SDSE01-4 (51–93%) in three out of five WWTPs but in Hveragerði and Akureyri the concentration was higher in effluent than in influent (Table 24). The detected concentrations in WWTP effluents in the present study are in the same range as reported previously (0.60–320 µg/l; Schlabach et al., 2009).

WWTP sludge

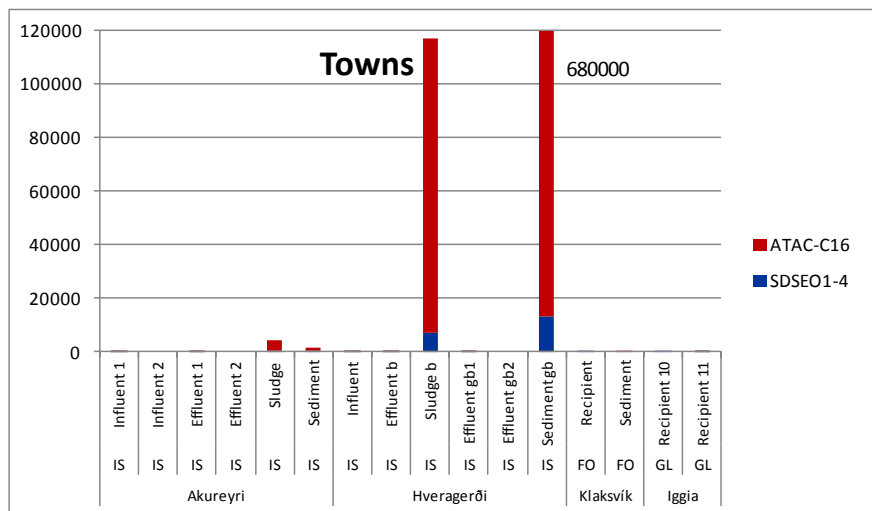
All eight de-watered sludge samples contained SDSE01-4 in a concentration range of 510–180,000 µg/kg dw (Table 24). The lowest concentrations were detected in Akureyri (510 µg/kg dw; Figure 25 B). The highest concentration was found in the WWTP sludge from Sersjantvíkin Torshavn (180,000 µg/kg dw; Figure 25 A) and differs significantly from

the other WWTP sludge. The high concentrations in sludge suggest that this matrix may act as a sink for this surfactant. The concentrations of SDSE01-4 in sludge in the present report agree well with previously reported concentrations (Schlabach et al., 2009).

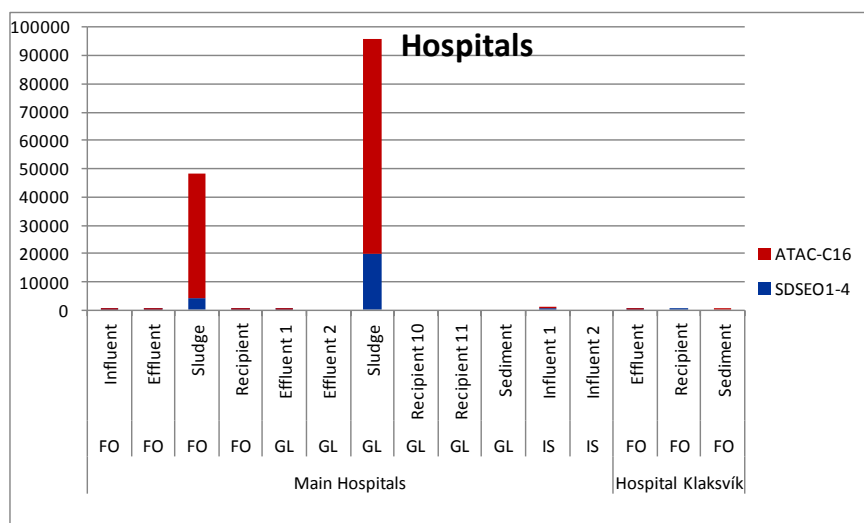
Figure 25. Selected additives in personal care products (SDSE01-4 and ATAC-C16) in different matrixes of capitals (A), towns (B) and hospitals (C) in $\mu\text{g/l}$ for water samples and $\mu\text{g/kg}$ for sludge and sediment samples. Please note, Iggia is a sampling site in Nuuk but is presented in the "Towns" figure due to space constraints



(A).



(B).



(C).

Recipient water and sediment

Eight out of 10 (80%) recipient water samples (Table 23) contained detectable concentrations of SDSEO1-4 in the range 0.28–19 µg/l (Table 24) and with a median concentration of 0.4 µg/l. The sample from Iggja harbour deviated significantly from the others and contained 19 µg/l (Figure 25).

SDSEO1-4 was detected in one out of six sediment samples (17%; Table 23). The positive samples came from Iceland (Akureyri 360 µg/kg dw; Figure 25 B).

In a previous study SDSEO1-4 was detected in recipient water in Norway (Oslo fjord and Tromsø sound). The concentrations detected was somewhat lower (<0.040–1.6 µg/l) than in the present study (<0.16–19 µg/l) and SDSEO1-4 was not detected in sediment (Schlabach et al., 2009).

5.8.6 Cocoamidopropyl betaine (CAPB)

CAPB was found in 33% of WWTP influent and 67% in effluent samples and in all sludge samples. The detection frequency in recipient waters and sediments were 80% and 17%, respectively (Table 23).

WWTP influent and effluent

CAPB was detected in the concentration range 0.44–89 µg/l (LOQ 0.08 µg/l; Table 24). The highest concentrations were detected in Greenland sewage line effluents (Kolonihavnen 85 and 89 µg/l; Figure 24). Indications of a reduction of CAPB in a WWTP was found in one WWTP only

(Akureyri) and was 75%. In the other plants the concentrations in influent and effluent were below the LOQ (Table 24). Schlabach et al., (2009), reported no detectable concentrations of CAPB in WWTP effluents.

WWTP sludge

All eight de-watered sludge samples contained CAPB in a concentration range of 350–5,000 µg/kg dw (Table 24). The sample from Klettag-ørðum Reykjavik (5,000 µg/kg dw) deviated significantly (Figure 24 A). The lowest concentration was detected in Torshavn Sersjantvíkin WWTP (350 µg/kg dw; Figure 24). The quite high concentrations indicate that sludge may be a sink for CAPB. This is not unlikely since it is recognised that quaternary amines have a high affinity to organic matter in the environment (Fernández et al., 1996). The concentrations of CAPB in sludge in the present study agreed with the results from a Norwegian study (Schlabach et al., 2009).

Recipient water and sediment

CAPB could be quantified in eight out of 10 recipient water samples (Table 23) and were found in the concentration range 0.10–7.5 µg/l (Table 24). All but one sample contained less than 1 µg/l. The highest concentration was detected in Iggia (7.5 µg/l; Figure 24).

CAPB was detected in one out of six sediment samples (17%; Table 24). The positive sample came from Iceland (Akureyri 360 µg/kg dw; Figure 24 B). The quite high concentrations in sediment suggest that sediment, like sludge, may act as a sink for CAPB.

In a previous Norwegian study CAPB was not detected in receiving water (Oslo fjord and Tromsø sound, <10 ng/l) nor in sediments (Schlabach et al., 2009).

5.8.7 Cetrimonium salt (ATAC-C16)

ATAC-C16 was found in all WWTP influents, effluents, sludge and sediment samples. The detection frequencies in recipient waters were 40% (Table 23).

WWTP effluent and influent

ATAC-C16 was detected in the concentrations range 1.3–87 µg/l (Table 24). A comparison of the concentrations in influent and effluent samples from WWTPs (Iceland and Faroe Islands) showed a reduction in four out of five WWTPs (49–94%), whereas in the WWTP at the main hospital in the Faroe Islands the opposite situation was detected (Table 24, Figure 25).

Schlabach et al., (2009), detected ATAC-C16 in the concentration range <0.04–3.6 µg/l in WWTP effluents in Norway.

WWTP sludge

All de-watered sludge samples contained ATAC-C16 in a wide concentration range, 1,700–680,000 µg/kg dw (Table 24). The highest value was found in sludge from Hveragerði (680,000 and 110,000 µg/kg dw; Figure 25 B). Two sludge samples deviated significantly from the others and showed relatively low concentrations: Akureyri contained (4,000 µg/kg dw; Figure 25 B) and Kolonihavnen (1,700 µg/kg dw; Figure 25 A). The generally high concentrations in sludge strongly suggest that the sludge act as a sink for ATAC-C16. The concentrations of ATAC-C16 in sludge in the present report were higher than found in Norway in 2008 (3,300–15,000 µg/kg dw; Schlabach et al., 2009).

Recipient water and sediment

Four out of 10 recipient water samples contained ATAC-C16 (Table 23) in the range 0.35–1.6 µg/l (Table 24). The highest concentration was detected in Torshavn in Sersjantvíkin (1.6 µg/l; Figure 25).

ATAC-C16 was detected in all sediment samples (Table 23) in a concentration range of 190–1,500 µg/kg dw (Table 24). The highest concentration was found in the sample from Torshavn harbour shipyard (1,500 µg/kg dw; Table 30 and Figure 25).

In a previous study in Norway, ATAC-C16 could not be detected in recipient water samples from Oslo fjord and Tromsø sound (<0.04 µg/l) but in the sediments at low concentrations (0.04–17 µg/kg dw; Schlabach et al., 2009).

6. Concentration patterns

In this chapter pharmaceuticals which were detected repeatedly in high concentrations are discussed.

The following pharmaceuticals were found to be highly abundant in this study: paracetamol (antipyretic), metformin (antidiabetic), salicylic acid and ibuprofen (both non-steroidal anti-inflammatory), citalopram and venlafaxine (both anti-depressants), atenolol and metoprolol (cardiovascular drugs) and dipyridamole (cardiovascular drug).

The relative occurrence depended on matrix, and most often the highest concentration was found in a solid matrix from one sampling site and most often in sludge. Paracetamol formed an exception, in that it occurred in higher concentrations in the water phase than in the solids. On the other extreme, with a factor of 1,000 or more, higher concentrations in a solid sample than in liquids were CAPB, sertraline and amlodipine (calculated from data in Table 29 to Table 32).

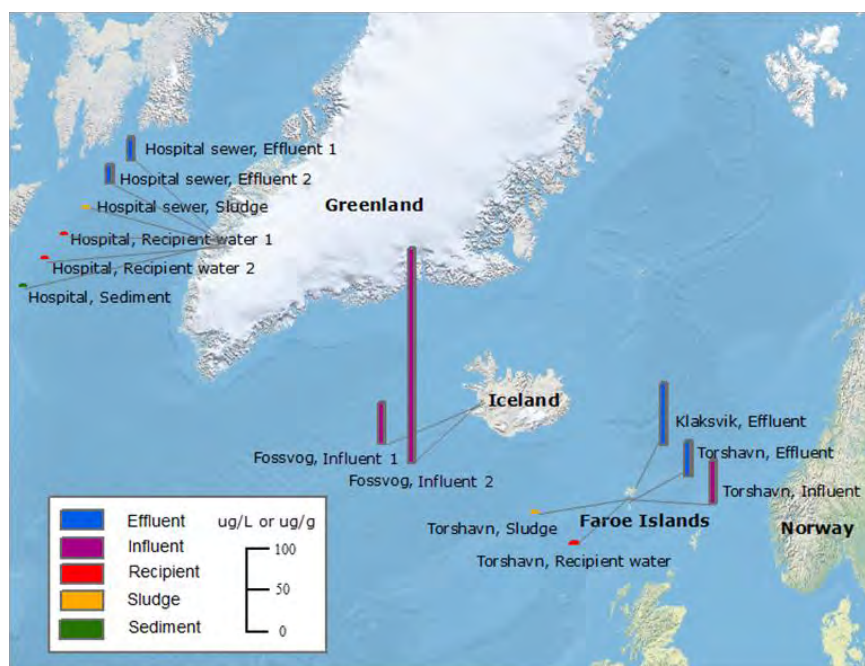
The additives in personal care products occurred in overall highest concentrations, where especially ATAC-C16, SDSEO 1-4, CAPB and SDS were found in high concentrations particularly in solids.

Comparing the findings in environmental samples to the drug use information in Table 11, indicates that there is a general agreement between drug use and occurrence in environmental samples in and around the sewage line. However, the information on drug use is relative and thus a drug like amlodipine which ranks number seven among the most frequently used pharmaceuticals in Iceland and number one in the Faroe Islands, may well be used in larger volumes in a population of approx. 300,000 (Iceland) than in one counting approx. 50,000 (Faroe Islands and Greenland). The relative data available in this report do not facilitate quantitative comparison between countries. Albeit these restraints in comparisons between countries, it is possible to assess the within country data with the proper information on the masses of drug in a defined daily dose (DDD). For the Faroe Islands for instance, the pharmaceutical used most frequently was amlodipine. The DDD for amlodipine is 0.005 g (WHO 2012), whereas DDDs for paracetamol and acetylsalicylic acid (as painkiller) are 3 g. This means that one DDD of paracetamol is equivalent to a mass of 600 DDDs of amlodipine, and thus it is not that surprising that amlodipine was not found in high concentrations in efflu-

ents from the Sersjantvíkin WWTP in Torshavn, whereas paracetamol was among the pharmaceuticals which were. The pharmaceutical occurring in highest concentration in this sample was dipyridamole, with a DDD resembling those of the above mentioned painkillers. This was not expected, because in contrast to these painkillers, dipyridamole was not listed among the 20 most frequently used pharmaceuticals but occurred as no. 40 on the list over the most frequently used pharmaceuticals in the Faroe Islands in 2010 (and therefore not shown in Table 11). As this comparison is done on effluents and thus could be influenced by the removal action of the WWTP it is pertinent to consider the concentration of these compounds in sludge also. Comparing the tendency to stick to solids and thus be caught in the WWTP among these compounds indicate that paracetamol and dipyridamole are not very different in this respect; paracetamol does not stick to solids and passes the WWTP unscaled, whereas dipyridamole to a somewhat larger extent stays with the sludge, but more modestly compared to for instance metformin. However, when taking into account that only 5% of paracetamol is excreted in the original form, the apparent “loss” is at least partly accounted for. Similarly, as in the Sersjantvíkin, Torshavn WWTP effluents, the pharmaceutical occurring in next highest concentration at the Reykjavik, Klettagørðum WWTP, after dipyridamole, was paracetamol. In the Kolonihavnen sewage line, however, dipyridamole was not prominent, but again paracetamol was the pharmaceutical occurring in highest concentrations, with metformin and salicylic acid at similar concentrations. In some instances the concentration of PPCP was actually higher in the water leaving the WWTP than in the inflowing water. This was seen for instance with salicylic acid, furosemide and naproxen but not as a general observation throughout all WWTPs, but at one or more. This does not necessarily mean that the WWTP actually acts as a source for PPCP rather than as a sink, but is likely an artefact stemming from the fact that sampling was done simultaneously of influent and effluent. By this, it is not the same water volumes which are sampled before and after treatment. Depending on the volume of the WWTP and the actual load of waste-water fed into it, the residence time of the waste water may be for instance a day, and thus when effluent water is sampled it is from the previous day, whereas the influent taken simultaneously is “fresh”.

Paracetamol was also the pharmaceutical which was found in highest concentrations in WWTP influent and effluent samples at hospital sites (Figure 26 and Appendix Table 29). The main hospital Fossvog in Iceland contained a factor 5 higher concentration of paracetamol in one WWTP influent sample (251,000 ng/l, sample 2) than a parallel sample from the same location (48,500 ng/l, sample 1), where sample 1 and 2 have been taken in succession. The different concentration levels of paracetamol in these two influent samples demonstrate clearly the variations related to the sampling times and that each sample represents only a snapshot and not the actual state of pollution. The main hospital in Nuuk, Greenland showed the lowest concentrations of paracetamol in sewage line effluents, recipient waters and sediments, while the sludge concentration was approximately 4 times higher than in the WWTP at the main hospital Torshavn in the Faroe Islands (Figure 26).

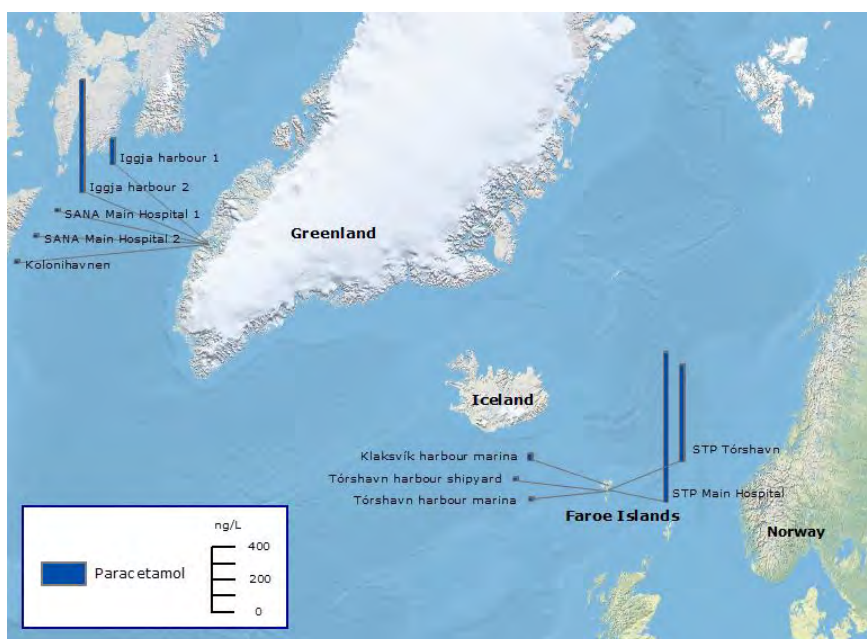
Figure 26. Concentrations of paracetamol in different matrix samples at hospital sites.



Paracetamol was detected in three out of five recipient water samples from the Faroe Islands, where the highest concentration was found at the main hospital (931 ng/l), followed by Sersjantvíkin WWTPs in Torshavn (599 ng/l) and Klaksvik harbour (42 ng/l; Figure 27). In the Torshavn harbour, in

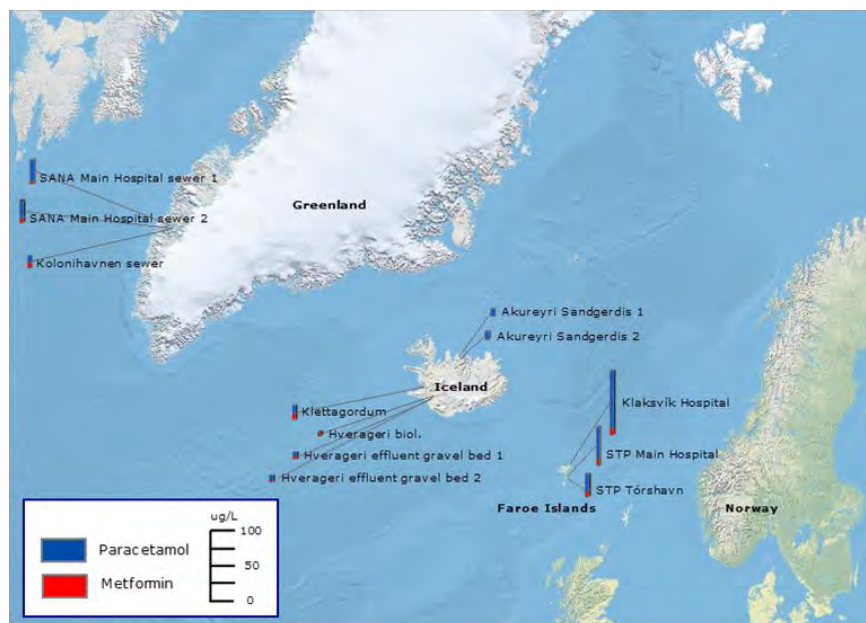
samples taken near the marina and shipyard, paracetamol was below the LOD. Paracetamol was found in recipient water samples from Iggja in Nuuk in 2010 and 2011 at 164 and 698 ng/l), respectively. In contrast to this, paracetamol could not be detected in the vicinity of the sewage outlets from the main hospital and U11 which runs into Kolonihavnen, both Nuuk. No recipient waters from Island were analysed in the present study.

Figure 27. Concentrations of paracetamol in recipient water samples at all sites.



Concentrations of paracetamol and metformin in WWTP effluents are shown in Figure 28 and in the Appendix Table 29. Paracetamol was generally detected in higher concentrations than metformin with one exception in Hveragerði (site biol. which represent effluent from the biological purification step), where paracetamol was below the LOD.

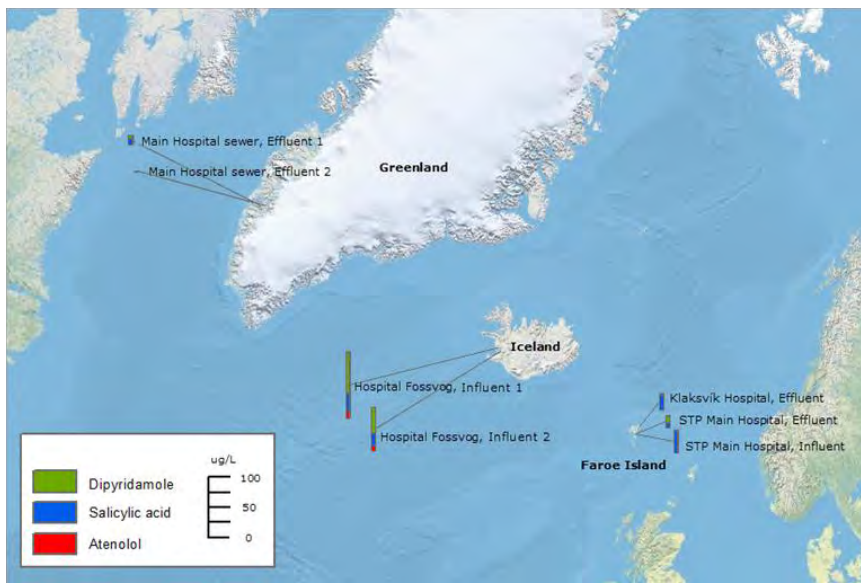
Figure 28. Concentrations of paracetamol and metformin in effluent waters



Paracetamol was found in higher concentrations in effluents from hospitals than in domestic effluents in the Faroe Islands and in Greenland.

Other pharmaceuticals which were found in relatively speaking high concentrations in the present study, were salicylic acid, atenolol and dipyridamole (Figure 29, Appendix Table 29 and Table 30). Salicylic acid was found in similar concentrations in influent of the main hospital and effluent of the Klaksvik hospital in the Faroe Islands, and in the influents from the main hospital Fossvog in Iceland. Somewhat lower concentrations were found in effluents from the main hospitals in the Faroe Islands and in Greenland. Atenolol and dipyridamole were detected in highest levels in influents from the main hospital in Iceland, followed by the effluents from the main hospitals in Greenland and the Faroe Islands. Interestingly, the concentrations of atenolol and dipyridamole measured in influent to the main hospital WWTP in the Faroe Islands, were much lower than the corresponding effluent concentrations. Effluent samples from the Klaksvik hospital in the Faroe Islands had low concentrations of both atenolol and dipyridamole.

Figure 29. Concentrations of salicylic acid, atenolol and dipyridamole in WWTP influent and effluent samples at hospital sites



Antidepressants were generally found in highest concentrations in Iceland in all sample matrixes (Figure 18 and Appendix Table 31). Venlafaxine was the dominating antidepressant in WWTP influents from the main hospital and sludge from WWTP Klettagørðum, Reykjavik. This WWTP sludge sample from Reykjavik was showing extremely high concentrations and was not comparable to the WWTP sludge sample from Torshavn, which also had quite a high load of antidepressants, with sertraline as the dominating compound. However, although the concentration of venlafaxine in sludge from Reykjavik was higher, the concentrations of venlafaxine in all effluent samples from Faroe Islands were higher than those from Iceland. Venlafaxine and citalopram were the most prominent antidepressant in the survey.

Concentrations of PPCP in sediments from Akureyri were overall high, and much higher than in other sediment samples. The reason for this is likely that the sediments were taken in the immediate vicinity of the effluent outlet at the shore and thus—as also indicated by their appearance—were highly influenced by this.

7. Preliminary ecotoxicological risk assessment

To assess the risk associated with release of PPCP from the WWTP to the surrounding environment, the most important pathway from the WWTP to the surroundings is the water phase. In those cases where waste water is discharged directly to the recipient without any treatment steps, more solids are deposited in the recipient, and the analyses of sediments in these become important to elucidate possible influence.

7.1.1 *Pharmaceuticals*

In the present study, preliminary ecotoxicological risk assessment for the investigated recipient waters (Table 26) and effluent samples was (Table 27) performed.

The predicted no-effect concentrations (PNEC) of measured no effect levels of pharmaceuticals were collected from the fass-database (www.fass.se) and literature. In those cases where PNEC values were not available in the above mentioned sources, no ecotoxicological risk assessments were undertaken. Thus, the preliminary ecotoxicological risk assessment does not include 12 of the 37 analysed pharmaceuticals; amiloride, atenolol, dipyridamole, enalapril, enalaprilat, estrone, gliclazide, paroxetine, perindopril, perindoprilat, sulfamethizole and zopiclone. The sediment samples were not included in this initial assessment since PNEC data are only available for water. Previously published studies have applied PNECs for sediments estimated from PNEC for water. However, such rough estimates might result in inaccurate and also incorrect risk assessments.

Ratios between measured environmental concentrations (MEC) in recipient waters and PNECs are calculated and used as an indicator of risk (Table 26). For WWTP effluents a dilution factor of 10 was applied. If the MEC or MEC/10 are greater than the PNEC (i.e. the MEC/PNEC ratio >1 or MEC/10/PNEC >1), then it can be assumed that there is a risk of toxic effects in the environment.

For the investigated pharmaceuticals in recipient waters, the ratios were lower or much lower than 1 (Table 26), which indicates that there are no

risks to the aquatic environment. Caution should be taken when interpreting the MEC/PNEC data for 17β -estradiol, estriol and 17α -ethinylestradiol since the PNECs were below the limits of detections for these compounds. Analysis of hormones is a challenging task, since there are analytical limitations in achieving low LODs, even when state-of-the-art analytical instrumentation is used. No recipient waters from Iceland were investigated in the present study and therefore no initial ecotoxicological risk assessment could be performed.

Also, the risk ratios calculated from the diluted effluent samples were lower or much lower than 1 (Table 27). This indicates that these concentrations of pharmaceuticals in general pose no risk to the environment. For candesartan however, the ratios at two locations in the Faroe Islands (the hospital Klaksvik and the main hospital) were >1 . Samples from the recipients of these effluents were also analysed and the risk assessment on these indicate that there is a small margin only to a level where unacceptable risk from candesartan is expected. Risk ratios for 17β -estradiol at 31 and higher were found in diluted effluents from Queen Ingrid's hospital, Kolonihavnen U11 SL and WWTP Klettagørðum, Reykjavik. Estriol risk ratios were from 3 to 7.7 at six locations; Akureyri, 2 \times Hveragerði, Reykjavik, Kolonihavnen and the Klaksvik hospital). Even higher ratios, from 10.1 up to 26.4, were calculated for samples taken at the four locations; Torshavn, the Klaksvik hospital and the main hospital in the Faroe Islands and in two samples from Queen Ingrid's hospital in Greenland). These findings indicate a chronic risk for aquatic organisms staying and/or living around WWTP effluent pipe-outlets.

Apparently, a high dilution of WWTP effluents discharged into recipient waters minimises the risks for the aquatic organisms. However, the results of the preliminary risk assessment should be taken with caution since it is based on snapshot samples and sometimes only one sample per location.

Additives in personal care products

Two MEC/PNEC ratios were calculated based on the maximum MEC (MEC_{max}/PNEC) detected and the median MEC (MEC_{median}/PNEC) in the receiving water in the vicinity of respective WWTP. The calculated MEC/PNEC are summarized in Table 25. The PNECs used for the MEC/PNEC calculations were retrieved from Schlabach et al. (2007).

PNEC data are only available for aquatic organisms, thus the risk assessment is made for water living organism and not for sediment dwelling ones. The MECs used were the detected concentrations in "recipient water" in the receiving water in the vicinity of respective WWTP.

Table 25. Calculated MEC/PNEC ratios for selected additives in personal care products

Ratio	DEP	BuP	EDTA	SDS	SDSEO1-4	CAP B	ATAC-C16
MECmax/PNEC	0.04	0.02	0.2	0.02	19	375	165
MECmedian/PNEC	0.02	0.02	0.2	0.01	0.4	14	112
PEC/PNEC (Sclabach et al 2007)	0.62	0.002	0.23	15	563	1773	360

MEC/PNEC (max): Calculation based on the highest MEC detected in recipient water. MEC/PNEC

(median): Calculation based on the median MEC detected in recipient water.

MEC/PNEC for DEP, BuP, SDS and EDTA in Table 26: all shows a risk ratio <1. Furthermore, MEC/PNEC for CAPB and ATAC-C16 are all >1. Thus it can be assumed that there is a risk of toxic effects in the aquatic environment for CAPB and ATAC-C16.

Table 27. Calculated risk factors based on measured environmental concentrations (MEC) in effluent waters divided by a factor 10 to account for dilution and predicted no effect concentrations (PNEC). Concentrations of pharmaceuticals are given in ng/l and of additives in personal care products in µg/l

Compounds	PNEC	Towns											Hospitals				Capitals				References					
		Akureyri IS		Akureyri IS		Hveragerði IS		Hveragerði IS		Hveragerði IS		Main Hospital FO	Main Hospital GL		Main Hospital GL	Hospital Klaksvík FO	Kolonihavnen GL		Reykjavík IS			Tórshavn FO				
ng/L	MEC/10	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC	MEC	MEC/PNEC				
Pharmaceuticals																										
Salicylic acid	90000	239	0.003	603	0.007	<41.7	-	46.0	5.11E-04	30.2	3.35E-04	870	0.010	1100	0.012	2460	0.027	583	6.48E-03	265	0.003	874	0.010	fass.se		
Amiloride	NA	4.09	NA	8.50	NA	4.78	NA	12.3	NA	6.44	NA	0.83	NA	<3.03	NA	<3.03	NA	<3.03	NA	216.81	NA	22.82	NA			
Amlodipine	280	3.86	0.014	7.29	0.026	<4.17	-	4.50	0.016	3.22	0.012	9.81	0.035	12.1	0.043	44.8	0.160	1.28	0.005	<4.17	-	31.9	0.114	fass.se		
Atenolol	NA	71.1	NA	70.7	NA	97.7	NA	173	NA	125	NA	18.8	NA	31.1	NA	47.0	NA	<20.8	NA	<20.8	NA	<20.8	NA			
Bendroflumethiazide	23000	<0.42	-	<0.42	-	<0.42	-	<0.42	-	0.13	5.48E-06	0.14	6.01E-06	<0.42	-	0.70	3.04E-05	<0.42	-	<0.42	-	0.65	2.81E-05	fass.se		
Candesartan	12	<2.08	-	<2.08	-	6.48	0.540	4.80	0.400	5.43	0.452	14.2	1.18	<2.08	-	<2.08	-	25.1	<2.08	-	<2.08	-	11.1	0.927	Kümmerer and Hempel	
Citalopram	8000	5.90	0.001	2.69	3.36E-04	1.22	1.52E-04	1.77	2.21E-04	2.20	2.76E-04	10.1	0.001	19.2	2.40E-03	54.0	0.007	13.0	0.002	6.92	0.001	11.7	0.001	Besse et al., 2008		
Diclofenac	100000	3.34	3.34E-05	6.34	6.34E-05	39.0	3.90E-04	34.2	3.42E-04	30.0	3.00E-04	59.7	5.97E-04	1.43	1.43E-05	13.8	1.38E-04	2.35	2.35E-05	21.5	2.15E-04	14.8	1.48E-04	fass.se		
Dipyridamol	NA	8.08	NA	<16.05	NA	20.4	NA	11.3	NA	588	NA	1110	NA	464	NA	170	NA	8.39	NA	1250	NA	2460	NA			
Enalapril	NA	1.14	NA	1.15	NA	0.16	NA	2.27	NA	2.02	NA	12.0	NA	11.3	NA	32.2	NA	2.61	NA	1.92	NA	13.5	NA			
Enalaprilat	NA	1.01	NA	1.22	NA	1.37	NA	1.88	NA	2.79	NA	7.13	NA	3.90	NA	5.03	NA	1.61	NA	1.26	NA	7.31	NA			
17β-Estradiol	0.80	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	37.5	46.9	34.2	42.7	<2.08	-	35.7	45	24.9	31	<2.08	-	
Estrinol	0.75	2.28	3.04	<2.08	-	<2.08	-	5.78	7.70	5.34	7.12	19.8	26.4	14.0	18.6	13.5	18.0	3.10	4.1	2.54	3.4	5.39	7.2	7.59	10.1	Schlabach et al., 2007
Estrone	NA	0.38	NA	<3.73	NA	<3.73	NA	1.29	NA	1.02	NA	1.85	NA	1.89	NA	2.10	NA	0.74	NA	0.78	NA	1.33	NA	1.32	NA	
17α-Ethinylestradiol	0.10	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	fass.se
Fluoxetine	110	0.53	0.005	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	<4.17	-	fass.se
Furosemide	45140	12.0	2.67E-04	15.2	3.36E-04	46.0	0.001	58.8	0.001	90.9	0.002	114	0.003	4.73	1.05E-04	1140	0.025	8.43	1.87E-04	33.7	0.001	61.2	0.001	0.001	fass.se	
Glicazide	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	2.96	NA	<2.08	NA	<2.08	NA	2.26	NA	<2.08	NA	2.40	NA			
Hydrochlorothiazide	100000	41.3	4.13E-04	41.0	4.10E-04	73.7	0.001	98.4	0.001	94.4	0.001	35.4	3.54E-04	0.64	6.42E-06	0.83	6.26E-06	34.5	3.45E-04	2.26	2.26E-05	61.7	0.001	27.8	2.78E-04	fass.se
Ibuprofen	7100	64.0	0.009	132	0.019	<0.42	-	473	0.067	508	0.072	338	0.048	281	0.040	450	0.063	70.0	0.010	506	0.071	419	0.059		fass.se	
Levothyroxine	497000	<1.61	-	<1.61	-	<1.61	-	0.16	3.29E-07	<1.61	-	<1.61	-	<1.61	-	<1.61	-	<1.61	-	0.17	3.46E-07	<1.61	<1.61	-	fass.se	
Lidocaine	106000	6.14	5.79E-05	0.53	4.95E-06	0.13	1.24E-06	1.42	1.34E-05	<0.42	-	0.16	1.55E-06	0.06	6.04E-07	0.30	2.80E-06	<0.42	-	<0.42	-	0.43	4.02E-06	0.001	0.001	Kümmerer and Hempel
Losartan	331000	17.5	5.30E-05	16.3	4.93E-05	32.7	9.87E-05	28.1	8.50E-05	27.6	8.35E-05	15.9	4.81E-05	7.43	2.25E-05	29.2	8.82E-05	16.5	4.98E-05	16.2	4.88E-05	2.29	6.93E-06	0.001	0.001	fass.se
Metformin	101000	23.4	2.32E-04	30.5	3.02E-04	414	0.004	403	0.004	283	0.003	756	0.007	358	0.004	590	0.006	680	0.007	559	0.006	742	0.007	0.007	Kümmerer, 2004	
Metoprolol	53300	5.22	9.80E-05	5.12	9.61E-05	6.65	1.25E-04	12.0	2.25E-04	13.5	2.54E-04	35.6	0.001	25.1	4.72E-04	81.0	0.002	13.6	2.54E-04	11.7	2.20E-04	65.3	0.001	0.001	fass.se	
Naproxen	640	52.5	0.082	162	0.254	73.2	0.114	140	0.218	192	0.301	0.79	0.001	0.36	0.001	<1.05	-	10.6	0.017	121	0.190	182	0.284		fass.se	
Paracetamol	50000	847	0.017	854	0.017	<2.08	-	366	0.007	705	0.014	4030	0.081	2580	0.052	2060	0.041	7150	0.016	1050	0.021	2050	0.041	0.041	fass.se	
	9200	847	0.092	854	0.093	<2.08	-	366	0.040	705	0.077	4030	0.438	2580	0.280	2060	0.224	7150	0.087	1050	0.114	2050	0.223	0.223	fass.se	
Paroxetine	NA	0.34	NA	0.20	NA	2.33	NA	0.32	NA	4.59	NA	1.34	NA	2.08	NA	NA	<1.51	NA	0.27	NA	8.95	NA	14.9	NA		
Perindopril	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	1.84	NA	<2.08	NA	<2.08	NA	0.66	NA	<2.08	NA	1.18	NA			
Perindoprilat	NA	<2.08	NA	<2.08	NA	<2.08	NA	0.30	NA	0.29	NA	0.99	NA	<2.08	NA	<2.08	NA	0.26	NA	<2.08	NA	1.33	NA			
Sertraline	56	0.60	0.011	0.30	0.005	0.94	0.017	0.46	0.008	3.35	0.060	2.27	0.041	<0.42	-	0.07	0.001	0.20	0.003	2.94	0.053	1.91	0.034		fass.se	
Simvastatin	9600	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	<2.08	-	fass.se
Sulfamethizole	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA		fass.se	
Venlafaxine	4800	2.62	5.45E-04	2.40	5.01E-04	10.4	0.002	10.8	0.002	14.9	0.003	64.7	0.013	102	0.021	66.1	0.014	2.13	4.43E-04	13.8	0.003	74.6	0.016	0.016	fass.se	
Warfarin	11000	<0.8	-	<0.8	-	0.83	7.52E-05	<0.8	-	<0.8	-	<0.8	-	<0.8	-	<0.8	-	<0.8	-	<0.8	-	<0.8	-	<0.8	-	fass.se
Zopiclone	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	-	<2.08	NA	<2.08	NA	<2.08	NA	<2.08	NA			
PCP																										
EDTA	9	1.80	0.200			2.90	0.322	2.50	0.278			1.10	0.122	42.0	4.7			63.0	7.0	3.70	0.411	1.40	0.156	56.0	6.2	Schlabach et al., 2007
EDTA(2)																										
Diethylphthalate	3.65	0.053	0.014			0.077	0.021	0.047	0.013			0.028	0.008	0.207	0.057			0.017	0.005	0.086	0.024	0.073	0.020	0.051	0.014	Schlabach et al., 2007
Butylparaben	0.404	0.002	0.005			0.005	0.013	0.004	0.009			0.001	0.003	0.008	0.020			0.002	0.006	0.011	0.027	0.001	0.003	0.005	0.012	Schlabach et al., 2007
SDS	20	0.420	0.021			0.200	0.010	0.180	0.009			0.560	0.028	0.240	0.012			0.130	0.007	0.096	0.005	0.220	0.011	0.079	0.004	Schlabach et al., 2007
SDSE01-4	1	6.700	6.7			0.550	0.550	0.084	0.084			3.300	3.3	33.00	33			12.00	12	45.00	45	0.130	0.130	3.600	3.6	Schlabach et al., 2007
CAPB	0.02	0.550	28			0.100	5.0	0.036	1.8			<0.085	-	8.500	425			4.700	235	8.900	445	<0.2	-	<0.084	-	Schlabach et al., 2007
ATAC-C16	0.01	0.299	30			0.155	15.5	0.690	69			1.056	106	0.340	34			2.879	288	0.376	38	3.1	310	0.520	52	Schlabach et al., 2007

8. Conclusions and recommendations

This screening study was the first assessment of concentrations of pharmaceuticals and additives in personal care products in wastewater and wastewater treatment plants as well as recipient waters from the Faroe Islands, Greenland and Iceland.

Thirty three substances of the 37 investigated pharmaceuticals and all seven analysed additives to personal care products were detected. 17 α -Ethinylestradiol, omeprazole, sulfamethizole and zopiclone were below the LODs in all samples.

To our knowledge, this was the first time in the Nordic countries that amiloride, candesartan, enalaprilat, gliclazide, hydrochlorothiazide, levothyroxine, lidocaine, losartan and perindoprilat were analysed in environmental samples.

The present study present for the first time data on amlodipine, bendroflumethiazide, enalapril, enalaprilat, fluoxetine, sertraline and zopiclone in sediment samples in the Nordic countries.

And to our knowledge is also the first time simvastatin and warfarin have been analysed in WWTP sludge, recipient waters and sediment samples from the Nordic countries.

The following pharmaceuticals were found to be highly abundant: paracetamol (antipyretic), metformin (antidiabetic), salicylic acid and ibuprofen (both non-steroidal anti-inflammatory), citalopram and venlafaxine (both anti-depressants), atenolol and metoprolol (cardiovascular drugs) and dipyridamole (cardiovascular drug).

The cardiovascular drugs atenolol and hydrochlorothiazide were found in much higher concentration in Iceland than in Faroe Islands and Greenland, whereas the opposite was true for amlodipine and metoprolol, which were found in highest concentrations in the Faroe Islands with one exception only. That amlodipine is a much used drug in the Faroe Islands is apparent also from the overview of volume of pharmaceuticals used in 2010 (Table 11). Overall, lower concentrations of PPCP were found in Greenlandic samples than in those from the Faroe Islands and Iceland, although the highest effluent butylparaben and DEP concentrations were found in the samples from Kolonihavnen U11 and Queen Ingrid's hospital,

respectively. Antidepressants were generally found in highest concentrations in WWTP sludge from Iceland, but in effluent samples, the highest concentrations were found in Faroe Islands. These differences are likely reflecting differences in population and population densities, but also differences in sewage treatment and differences in pharmaceutical preferences, which will vary with time and between countries.

Samples from hospital sites showed generally higher concentrations in all type of sample materials. The main hospital Fossvog in Iceland showed outstanding high concentrations of many of the pharmaceuticals in WWTP effluent samples, a factum which is linked to the much higher capacity of this hospital than those in the Faroe Islands and Greenland.

The surfactants SDSE01-4, ATAC-C16 and CAPB and the complexing agent EDTA were the additives in personal care products which were detected in highest concentrations in WWTP effluent waters, sludge and sediments. The sediment and sludge probably act as a sink for these substances. The more readily degradable personal care chemicals, DEP and butylparaben, were detected at low concentrations (low ng/l to low µg/l). DEP and butylparaben have low affinity to organic particles and are not concentrated in sludge and sediments.

The study provides insight into the discharge of pharmaceuticals and additives in personal care products in areas with very different waste water treatment policies. In some of the analysed areas, WWTP with several treatment steps are in place, whereas in others, the waste water is discharged to the recipient untreated. The design of the screening does not allow solid conclusions about the efficiency of the analysed WWTPs. Still, the study comprises some data sets which allow a preliminary estimate of the removal efficiency of the WWTP's and the potential threat to the ecosystems.

The low concentrations in surface water and the high concentrations detected in sediments indicate a quite efficient elimination of the surfactants from the water phase through adsorption to sediment. This is also true for the WWTP: the concentration of surfactants was high in sludge compared to the water concentrations. It was concluded that the sediment and the sludge act as a sink for these compounds.

Nonetheless, when the measured environmental concentrations are compared to the predicted no-effect level (MEC/PNEC ratios) for the surfactant SDSE01-4, CAPB and ATAC-C16 it indicates that there are risks for chronic toxicity in the environment. On the other hand, the MEC/PNEC ratios for the additives in personal care products DEP, butylparaben and SDS indicate that these compounds pose no risk for aquatic organisms in the investigated environments.

Calculated MEC/PNEC ratios for pharmaceuticals in recipient waters were lower or much lower than 1, which indicates that there was no risk to the environment at the time of sampling. The MEC/PNEC ratios for 17 β -estradiol, estriol and 17 α -ethinylestradiol are inconclusive, since the PNECs for these hormones were lower than the limits of detections for these compounds. Furthermore, no initial ecotoxicological risk assessment could be performed for the following compounds: amiloride, atenolol, dipyridamole, enalapril, enalaprilat, estrone, gliclazide, paroxetine, perindopril, perindoprilat, sulfamethizole and zopiclone due to the lack of available PNEC-data. In all, this means that of the 33 analysed pharmaceuticals—not counting the natural hormones—an ecological risk assessment was only possible for approx. 2/3 of these.

In assessments based on diluted WWTP effluents, the risk ratios were lower or much lower than 1 for most of the analysed pharmaceuticals. This indicates that most of the pharmaceuticals for which basic toxicity data were available (as PNEC's) pose no risk to the environment. However, candesartan, 17 β estradiol and estriol showed ratios >1 and >>1 at two or more locations, which indicates a risk for chronic toxicity in aquatic organisms staying around WWTP effluent pipe-outlets.

No recipient waters from Iceland were investigated in the present study and therefore no initial ecotoxicological risk assessment could be performed. Hence, it is recommended to analyse recipient waters from Iceland in future screening studies.

Another limitation was the lack of PNEC data for sediments and the assessment of risks based on contaminants in sediment samples, since there are only PNEC data available for water. Thus, risk assessment for sediments was not performed. Future assessments would benefit immensely from having toxicity data for sediment-dwelling organisms available.

The conclusions above are based on relatively few samples per site. Besides, not always complete sample sets were available. Nevertheless, the screening has identified the occurrence of a high number of pharmaceuticals and additives in personal care products in waste water, waste water treatment plants and recipients from Faroe Islands, Greenland and Iceland. A complete risk assessment could not be performed within this study due to limitations in sample material and availability of PNEC values. Therefore, further investigations are recommended in order to fill knowledge gaps as e.g. daily and sea-

sonal variations, variations in throughput of the WWTP, removal capacity of the WWTP, etc.

Also, it is strongly recommended that the findings are scrutinized more closely for each sewage line/WWTP/recipient location separately by the local authorities responsible for the waste water handling, so that possible shortfalls in this may be identified and prioritized for amelioration.

9. Acknowledgements

Faroe Islands

Thanks are due to Katrin Hoydal, Sanna í Túni Nielsen, Regin Olsen, Øssur Hentze, Martin Gregersen and Julian Dam for assistance in sampling, and to Rakul Mortensen for assistance with sampling and producing graphics for the report. Sanna í Túni Nielsen also assisted in proofreading.

Greenland

Thanks are due to Anette Hansen and Mathias Barfoed for assistance in planning and performing the sampling in 2010. For assistance with sampling in 2011 Kaare Winther Hansen, Kommuneqarfik Sermersooq is gratefully acknowledged, as is hired hand Kristian Nielsen, GMS, for climbing down the sewer for samples.

Iceland

Thanks are due to Gunnar Steinn Jónsson, for clarifications and pictures, and to staff at Orkuveita Reykjavíkur; Sigurður Skarphéðinsson, Kristján Tómasson, Egill Sölvason. Thanks also to Guðmundur Baldursson, Árni Svavarsson both staff at Hveragerði WWTP, and to Gunnþór Hákonarson at Akureyri WWTP for sampling.

Norway

Thanks to Inger-Christin Steen at NILU for help with sample preparation, Terje Vasskog (Norut), Morten K. Moe and Linda Hansen at NILU for help with initialising the LPME methods, Athanasios A. Katsogiannis (NILU) for proofreading and Silje Winnem (NILU) for assistance.

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11. Sammendrag

Denne rapporten beskriver resultater av en screeningundersøkelse av legemiddelrester og tilsetningsstoffer i personlig pleieprodukter i områder på Færøyene, Island og Grønland. Undersøkelsen omfattet analyser av prøver fra kilder der stoffene er forventet i relativt høye konsentrasjoner, som kloakkutslipp enten fra husholdninger og industri generelt, eller fra sykehus.

Studien omfattet legemidler som brukes for å lindre og behandle symptomer og sykdommer og kjemikalier som tilsettes personlig pleieprodukter for å bedre deres hygieniske egenskaper eller holdbarhet. Utvelgelsen av medisiner og personlig pleiekjemikalier (samlet referert til som PPCP) som skulle inngå i undersøkelsen ble basert på statistikk og dokumentasjon av medisinbruken fra 2010 på Færøyene, Island og i Nuuk, Grønland. I tillegg ble resultatene fra en nordisk studie som analyserte miljørisikoen som er forbundet med bruken av (human) medisin, samt en nylig utført screening og risikoanalyse av slike stoffer i Norge, brukt som grunnlag for utvelgelsen av stoffer.

Prøvetaking ble gjort i 2010, med noe tilleggsprøvetaking i 2011. I alt ble 38 medisiner og/eller deres omformingsprodukter, og 7 kjemikalier som brukes i personlig pleieprodukter analysert. I undersøkelsen inngikk analyser av totalt 44 prøver, hvorav noen ble analysert som parallelle prøver og noen som duplikater.

Noen stoffer, som diclofenac og ibuprofen, ble funnet i de fleste prøvene, og enkelte, som simvastatin og sulfametizole, ble ikke påvist i det hele tatt. Det syntetiske østroget 17 α ethinylestradiol ble ikke detektert i noen prøver, mens den naturlige analogen, 17 β estradiol, ble påvist i noen få. Manglende påvisning skyldtes i hovedsak høy deteksjonsgrensen for analysemetoden som ble brukt. Deteksjonsgrensene er en stor utfordring ved analyse av hormoner i prøver fra miljøet. Estrone, som også er et naturlig østroget, ble funnet i de fleste prøvene.

De PPCP-forbindelsene som ble påvist i høyest konsentrasjon hvis alle prøvene ses under ett, var cetrimonium salter (ATAC-C16) > natrium dodecylsulfat (SDSE01-4) \approx cocoamidopropylbetain (CAPB) > natrium dodecylsulfat (SDS) og salisylsyre.

Etylendiamintetraeddiksyre (EDTA), metformin, citalopram, ibuprofen og metoprolol viste sammenlignbare konsentrasjoner som var litt lavere

enn de dominerende PPCP. Alle PPCP, bortsett fra paracetamol, forekom i høyere konsentrasjoner i faste prøver enn i flytende når sammenligningen ble gjort på vektbasis. Paracetamol ble derimot ofte funnet i høyere konsentrasjoner i flytende prøver enn i faste. En høy konsentrasjonsbrøk for forekomsten av et gitt stoff i slam fra et renseverk sammenlignet med flytende prøver fra samme, indikerer at potensialet for renseverket til å fjerne forbindelsen er høyt, og muligheten for at forbindelsen skal unnsnippe til resipienten er lav.

Generelt ble det bare påvist noen få PPCPer i sedimenter, men salisylsyre, en metabolitt til acetylsalisylsyre, ble funnet i samtlige prøver. Også det overflateaktive stoffet ATAC-C16 ble funnet i de fleste sedimentprøvene.

Foreløpige miljørisikovurderinger ble utført på bakgrunn av målte PPCP-konsentrasjoner i vannprøver fra resipienter sett i forhold til anslåtte null-effektkonsentrasjoner, PNEC (*predicted no-effect concentrations*). Beregningene antydte at den største miljørisikoen var knyttet til CAPB og ATAC-C16, hvor den største miljøtrusselen synes å være i resipienten ved Iggia i Nuuk, Grønland, og dernest ved utslippet av renseverket Serskantvíkin i Torshavn, Færøyene. Risikokvotienter (= målt konsentrasjon/PNEC) større enn 1 ble også påvist for SDSEO1-4. Sammenlagt ble det funnet risikokvotienter større enn 1 i åtte av de 11 prøvene av resipientvann som ble analysert. Som oftest var det CAPB som ga de høye risikokvotientene, dernest ATAC-C16, og i én prøve var det også så høy konsentrasjon av SDSEO1-4 at miljørisiko er sannsynlig.

Det ble ikke funnet risikokvotienter større enn 1 for noen av legemiddelrestene i resipientprøvene. Dette utelukker imidlertid ikke miljørisiko fra disse forbindelsene, siden PNEC data som er helt nødvendige for å lage slike beregninger, kun var tilgjengelig for ca. 2/3 av de analyserte stoffene. Mangel på kjennskap til PNEC gjorde derved risikovurdering umulig for 12 av de legemidlene som ble analysert: amiloride, atenolol, dipyridamole, enalapril, enalaprilat, estrone, gliclazide, paroxetine, perindopril, perindoprilat, sulfamethizole og zopiclone. Selv om det ikke på noen måte er gitt at den høyeste miljørisiko utgjøres av det stoffet som forekommer i høyest konsentrasjon, er det allikevel relevant å vise til at de medisiner som ikke kunne risikovurderes stort sett var de som forekom i lave konsentrasjoner, selv om dipyridamole, atenolol og amiloride var blant de 10 medisinene som ble funnet i høyest median konsentrasjon når flytende og faste prøver ble sett under ett.

Prøvetakingen ble gjort som øyeblikksprøver, hvilket innebærer at det ikke tas høyde for fluktuasjoner som naturlig forekommer i kloakkledningene. Slike fluktuasjoner gjør seg imidlertid ikke gjeldende i for-

hold til faste prøver som slam og sediment, og derfor er analyseresultatene som er oppnådd for disse prøvene langt mere robuste. På den annen side, når man sammenligner og vurderer resultatene, er det er det viktig å huske på at det er forskjell på utformingen av og oppholdstiden i renseverkene, samt belastning på kloakkrørene. Det betyr for eksempel at en merkelapp som "slam" eller "sediment" kan ha vært brukt på nokså forskjellige typer prøver. Noen kloakkanlegg i denne undersøkelsen har flere rensetrinn med mikrobiell nedbrytning og filtrering, mens andre steder ledes kloakken direkte til resipient uten rensing. Disse forskjellene er det viktig å ta hensyn til når man sammenligner fra sted til sted. Imidlertid var ikke det primære formålet med undersøkelsen å lage sammenligninger mellom lokaliteter, men å gi innsikt i utslipp av legemidler og personlig pleie kjemikalier i områder der lite eller ingen informasjon om dette var tilgjengelig. Brukerne av denne informasjonen antas først og fremst å være forvaltningsinstanser som er ansvarlig for kloakkanlegg og miljøovervåkning.

Denne undersøkelsen har gitt et førsteinntrykk av PPCP nivåer på Færøyene, Island og i Nuuk, Grønland i et begrenset antall prøver. Det anbefales å følge opp med nærmere undersøkelser som kan belyse variasjoner gjennom døgnet og mellom årstider, samt variasjoner som skyldes belastning på renseanleggene og den rensekapasitet disse har. Dessuten bør resipientprøver fra Island undersøkes. På grunn av mangel på PNEC- data for sedimenter ble det ikke foretatt noen risikovurdering for disse. Fremtidige undersøkelser ville ha stor nytte av å ha tilgjengelig økotoksisitetsdata for sedimentlevende organismer. Dessuten anbefales det sterkt at resultatene av denne undersøkelsen analyseres nærmere for de enkelte renseanleggene/kloakutslippene av de myndigheter som har ansvaret for kloakkanleggene, slik at eventuelle mangler kan identifiseres og prioriteres for utbedring.

12. Appendices

12.1 Individual results

Table 28. Detailed information on samples

NILU-ID	IVL-ID	Name on samples as the project group prefers	Sample mark	Country	Site	Description	Matrix	Code
11-1960 1	9033	Akureyri Dalustod 1	Dalustiod, Landfasgota	Iceland	Town	Domestic	Influent	Influent 1
11-1960 2	9033	Akureyri Dalustod 2	Dalustiod, Landfasgota	Iceland	Town	Domestic	Influent	Influent 2
11-1961 1	9034	Akureyri Sandgerdis. 1	Utras, Sandgesdistotn	Iceland	Town	Domestic	Effluent	Effluent 1
11-1961 2	9034	Akureyri Sandgerdis. 2	Utras, Sandgesdistotn	Iceland	Town	Domestic	Effluent	Effluent 2
11-1964	9037	Akureyri Dalustod	Dalustiod	Iceland	Town	Domestic	Sludge	Sludge
11-1965	9038	Akureyri Sandgerdis.	Utras Sandgesdistotn	Iceland	Town	Domestic	Sludge	Sediment
11-1962	9089	Hveragerdi	Hveragerdi	Iceland	Town	Domestic	Influent	Influent
11-1963	9088	Hveragerdi, effluent biol.	Hveragerdi	Iceland	Town	Domestic	Effluent	Effluent b
11-2117	9090	Hveragerdi, sludge biol.	Hveragerdi effluent sludge	Iceland	Town	Domestic	Sludge	Sludge b
11-1958 1	9087	Hveragerdi, effluent gravel bed	Recipient, Hveragerdi	Iceland	Town	Domestic	Effluent	Effluent gb1
11-1958 2	9087	Hveragerdi, effluent gravel bed	Recipient, Hveragerdi	Iceland	Town	Domestic	Effluent	Effluent gb2
11-2118	9091	Hveragerdi, sludge gravel bed	Recipient, Hveragerdi	Iceland	Town	Domestic	Sludge	Sludge gb
11-1969	9137	Klaksvik harbour marina	Sjögur, Klaksvik, Batahulunin 28.09.10(2 av 10)	Faroe Islands	Town	Recipient	Recipient water	Recipient
11-1979	9144	Klaksvik harbour	Klaksvik Astongum 28-9-2010 (A+B+C)	Faroe Islands	Town	Recipient	Sediment	Sediment
11-1983	9134	Iggia harbour recipient 2010	IGGIA, (X)	Greenland	Town	Recipient	Recipient water	Recipient 10
11-1988	9336	Iggia harbour recipient 2011	IGGIA, U1 (recipient)	Greenland	Town	Recipient	Recipient water	Recipient 11
11-1976	9140	STP Main Hospital	LSH, STP (1 av 4) 29.9.2010	Faroe Islands	Hospital	Hospital	Influent	Influent
11-1972	9136	STP Main Hospital	LSH	Faroe Islands	Hospital	Hospital	Effluent	Effluent
11-1977	9145	STP Main Hospital	LSH STP	Faroe Islands	Hospital	Hospital	Sludge	Sludge
11-1975	9130	STP Main Hospital recipient	Sjögur, AL 21.09.10	Faroe Islands	Hospital	Recipient	Recipient water	Recipient
11-1986 1	9332	SANA Main Hospital sewer	SANA, U7 (kloak)	Greenland	Hospital	Hospital	Effluent	Effluent 1
11-1986 2	9332	SANA Main Hospital sewer	SANA, U7 (kloak)	Greenland	Hospital	Hospital	Effluent	Effluent 2
11-1990	9340	SANA Main Hospital sewer	SANA, U7, (kloak)	Greenland	Hospital	Hospital	Sludge	Sludge
11-1982	9133	SANA Main Hospital recipient 2010	SANA (X)	Greenland	Hospital	Recipient	Recipient water	Recipient 10
11-1987	9333	SANA Main Hospital recipient 2011	SANA, U7 (recipient)	Greenland	Hospital	Recipient	Recipient water	Recipient 11
11-1989	9339	SANA Main Hospital recipient	SANA, U7 (recipient)	Greenland	Hospital	Recipient	Sediment	Sediment
11-1956 1	9092	Landsspitali Hospital Fossvog	Landsspitala	Iceland	Hospital	Hospital	Influent	Influent 1
11-1956 2	9092	Landsspitali Hospital Fossvog	Landsspitala	Iceland	Hospital	Hospital	Influent	Influent 2
11-1974	9131	Klaksvik Hospital	Klaksvikar sjukrahús. 1. 28-09-10	Faroe Islands	Hospital	Hospital	Effluent	Effluent
11-1969	9137	Klaksvik harbour marina	Siøgur, Klaksvik, Batahulunin 28.09.10(2 av 10)	Faroe Islands	Town	Recipient	Recipient water	Recipient
NILU-ID	IVL-ID	Name on samples as the project group prefers	Sample mark	Country	Site	Description	Matrix	Code
11-1984	9334	Kolonihavnen sewer	Kolonihavnen, U11 (kloak)	Greenland	Capital	Domestic	Effluent	Effluent
11-1992	9337	Kolonihavnen sewer	Kolonihavnen,U11(kloak)	Greenland	Capital	Domestic	Sludge	Sludge
11-1985	9335	Kolonihavnen recipient	Kolonihavnen, U11 (recipient)	Greenland	Capital	Recipient	Recipient water	Recipient
11-1991	9338	Kolonihavnen recipient	Kolonihavnen, U11(recipient)	Greenland	Capital	Recipient	Sediment	Sediment
11-1959 1	9093	Reykjavik Klettagordum 1	Reykjavik	Iceland	Capital	Domestic	Influent	Influent 1
11-1959 2	9093	Reykjavik Klettagordum 2	Reykjavik	Iceland	Capital	Domestic	Influent	Influent 2
11-1957	9094	Reykjavik Klettagordum	RVK	Iceland	Capital	Domestic	Effluent	Effluent
11-1966	9095	Reykjavik Klettagordum	RVK Klettagordum Reykjavik	Iceland	Capital	Domestic	Sludge	Sludge
11-1971	9141	STP Sersjantvikin Torshavn	STP Sersjantviken	Faroe Islands	Capital	Domestic	Influent	Influent
11-1973	9135	STP Sersjantvikin Torshavn	Faroe Islands 28-9-2010 STP Sersjantviken (1 av 4)	Faroe Islands	Capital	Domestic	Effluent	Effluent
11-1981	9146	STP Sersjantvikin Torshavn	STP Sersjantviken 28.9.10 (A+B+C)	Faroe Islands	Capital	Domestic	Sludge	Sludge
11-1967	9139	STP Sersjantvikin Torshavn recipient	Sjögur, S. 21-9-10	Faroe Islands	Capital	Recipient	Recipient water	Recipient tp
11-1968	9132	Torshavn harbour marina	Sjögur, BAT. 21-9-10	Faroe Islands	Capital	Recipient	Recipient water	Recipient m
11-1970	9138	Torshavn harbour shipyard	Sjögur BA, 21.09.10	Faroe Islands	Capital	Recipient	Recipient water	Recipient s
11-1980	9142	Torshavn harbour marina	BAT 21.9.10 (A+B+C)	Faroe Islands	Capital	Recipient	Sediment	Sediment m
11-1978	9143	Torshavn harbour shipyard	BA21.9.10 (A+B+C)	Faroe Islands	Capital	Recipient	Sediment	Sediment

Table 29. Individual results of non-steroidal anti-inflammatory and antipyretic analgesics, local anaesthetic drugs, antibiotics, antidiabetics and hypnotics (Sludge and sediment concentrations on dry weight basis)

Name on samples	STP name	Country	Code	Unit	Salicylic acid	Diclofenac	Ibuprofen	Naproxen	Lidocaine	Paracetamol	Sulfamethizole	Metformin	Glicazide	Zopiclone
Akureyri Dalustod 1	Akureyri	IS	Influent 1	ng/L	132	41.8	521	2340	1.16	9790	< 20.8	2450	< 2.08	< 20.8
Akureyri Dalustod 2	Akureyri	IS	Influent 2	ng/L	111	24.3	321	1370	2.13	11200	< 20.8	1780	< 2.08	< 20.8
Akureyri Sandgerdis. 1	Akureyri	IS	Effluent 1	ng/L	2390	33.4	640	525	61.4	8470	< 20.8	234	< 2.08	< 20.8
Akureyri Sandgerdis. 2	Akureyri	IS	Effluent 2	ng/L	6030	63.4	1320	1620	5.25	8540	< 20.8	305	< 2.08	< 20.8
Akureyri Dalustod	Akureyri	IS	Sludge	µg/kg	929	1.65	15.8	120	0.85	< 5.0	< 5	149	< 0.50	< 5.0
Akureyri Sandgerdis.	Akureyri	IS	Sediment	µg/kg	110	1.04	2.57	0.85	0.73	< 5.0	< 5	56.7	< 0.50	< 5.0
Hveragerdi	Hveragerði	IS	Influent	ng/L	< 41.7	697	1.62	175	3.48	204	< 20.8	13600	< 2.08	< 20.8
Hveragerdi, effluent biol.	Hveragerði	IS	Effluent b	ng/L	< 41.7	390	< 0.42	732	1.31	< 20.8	< 20.8	4140	< 2.08	< 20.8
Hveragerdi, sludge biol.	Hveragerði	IS	Sludge b	µg/kg	535	19.7	130	10.3						
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb1	ng/L	460	342	4730	1400	14.2	3660	< 20.8	4000	< 2.08	< 20.8
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb2	ng/L	302	300	5080	1920	< 0.42	7050	< 20.8	4830	< 2.08	< 20.8
Hveragerdi, sludge gravel bed	Hveragerði	IS	Sludge gb	µg/kg	952	19.4	210	51.0						
Klaksvík harbour marina	Klaksvík	FO	Recipient	ng/L	< 41.7	1.84	10.2	< 1.05	< 0.42	42.3	< 20.8	< 2.08	< 2.08	< 20.8
Klaksvík harbour	Klaksvík	FO	Sediment	µg/kg	7.69	0.26	< 0.1	< 0.1	< 0.25	< 5.0	< 5	< 0.50	< 0.50	< 5.0
Iggja harbour recipient 2010	Iggja	GL	Recipient 10	ng/L	6050	30.1	872	45.9	0.43	164	< 20.8	33.1	< 2.08	< 20.8
Iggja harbour recipient 2011	Iggja	GL	Recipient 11	ng/L	1050	10.6	120	< 1.05	0.86	698	< 20.8	62.3	< 2.08	< 20.8
STP Main Hospital	Main Hospitals	FO	Influent	ng/L	38400	190	3530	2.73	183	50600	< 20.8	9660	3.68	< 20.8
STP Main Hospital	Main Hospitals	FO	Effluent	ng/L	8690	597	3380	7.88	1.64	40300	< 20.8	7560	29.6	< 20.8
STP Main Hospital	Main Hospitals	FO	Sludge	µg/kg	159	26.9	53.5	0.32	46.5	22.4	< 5	239	< 0.50	< 5.0
STP Main Hospital recipient	Main Hospitals	FO	Recipient	ng/L	88.8	8.41	70.33	< 1.05	8.40	931	< 20.8	61.4	< 2.08	< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 1	ng/L	11000	14.3	2810	3.55	0.64	25800	< 20.8	3580	< 2.08	< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 2	ng/L						20600	< 20.8	5900	< 2.08	< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Sludge	µg/kg	162	1.87	48.2	0.87	15.4	85.2	< 5	553	0.560	< 5.0
SANA Main Hospital recipient 2010	Main Hospitals	GL	Recipient 10	ng/L	< 41.7	2.42	< 0.42	< 1.05	< 0.42	< 20.8	< 20.8	< 2.08	< 2.08	< 20.8
SANA Main Hospital recipient 2011	Main Hospitals	GL	Recipient 11	ng/L	< 41.7	17.3	0.98	< 1.05	< 0.42	< 20.8	< 20.8	748	< 2.08	< 20.8
SANA Main Hospital recipient	Main Hospitals	GL	Sediment	µg/kg	14.4	0.19	0.18	< 0.1	< 0.25	< 5.0	< 5	2.45	< 0.50	< 5.0
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 1	ng/L	21700	58.8	48800	69100	87.3	48500	< 20.8	39300	116	< 20.8
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 2	ng/L	30500	119	1810	109000	144	251000	< 20.8	59000	538	< 20.8
Klaksvík Hospital	Hospital Klaksvík	FO	Effluent	ng/L	24600	138	4500	< 1.05	2.97	71500	< 20.8	7950	22.6	< 20.8
Klaksvík harbour marina	Hospital Klaksvík	FO	Recipient	ng/L	< 41.7	1.84	10.2	< 1.05	< 0.42	42.3	< 20.8	< 2.08	< 2.08	< 20.8
Klaksvík harbour	Hospital Klaksvík	FO	Sediment	µg/kg	7.69	0.26	< 0.1	< 0.1	< 0.25	< 5.0	< 5	< 0.50	< 0.50	< 5.0

Table 30. Individual results of cardiovascular drugs (Sludge and sediment concentrations on dry weight basis)

Name on samples	STP name	Country	Code	Unit	Warfarin	Metoprolol	Amlodipine	Atenolol	Bendroflumethiazide	Enalapril	Enalaprilat	Perindopril	Perindoprilat	Furosemide	Hydrochlorothiazide	Amloride	Losartan	Candesartan	Dipyridamole	Simvastatin
Akureyri Dalustod 1	Akureyri	IS	Influent 1	ng/L	< 0.8	14.3	< 4.17	522	< 0.42	5.88	< 2.34	< 2.08	< 2.08	237	258	94.9	189	< 2.08	836	< 20.8
Akureyri Dalustod 2	Akureyri	IS	Influent 2	ng/L	< 0.8	95.3	11.4	501	< 0.42	6.50	14.00	< 2.08	< 2.08	363	286	30.6	192	< 2.08	887	< 20.8
Akureyri Sandgerdis. 1	Akureyri	IS	Effluent 1	ng/L	< 0.8	52.2	38.6	711	< 0.42	11.4	10.1	< 2.08	< 2.08	120	413	40.9	175	< 2.08	80.8	< 20.8
Akureyri Sandgerdis. 2	Akureyri	IS	Effluent 2	ng/L	< 0.8	51.2	72.9	707	< 0.42	11.5	12.2	< 2.08	< 2.08	152	410	85.0	163	< 2.08	< 16.0	< 20.8
Akureyri Dalustod	Akureyri	IS	Sludge	µg/kg	< 0.1	549	13.1	161	< 0.89	0.12	< 2.08	< 2.08	< 2.08	18.1	< 5.0	93.6	39.9	< 0.50	55.6	< 5
Akureyri Sandgerdis.	Akureyri	IS	Sediment	µg/kg	< 0.1	62.8	9.58	58.6	< 0.89	< 0.03	2.13	< 2.08	< 2.08	2.75	< 5.0	21.0	392	< 0.50	14.2	< 5
Hveragerði	Hveragerði	IS	Influent	ng/L	1.48	158	< 4.17	2230	< 0.42	< 0.10	5.37	< 2.08	< 2.08	1250	1260	117	586	23.0	15916	< 20.8
Hveragerði, effluent biol.	Hveragerði	IS	Effluent b	ng/L	0.83	66.5	< 4.17	977	< 0.42	1.58	13.7	< 2.08	< 2.08	460	737	47.8	327	64.8	204	< 20.8
Hveragerði, sludge biol.	Hveragerði	IS	Sludge b	µg/kg	< 0.1				< 0.89					38.8						
Hveragerði, effluent gravel bed	Hveragerði	IS	Effluent gb1	ng/L	< 0.8	120	45.0	1730	< 0.42	22.7	18.8	< 2.08	2.98	588	984	123	281	48.0	113	< 20.8
Hveragerði, effluent gravel bed	Hveragerði	IS	Effluent gb2	ng/L	< 0.8	135	32.2	1250	1.26	20.2	27.9	< 2.08	2.90	909	944	64.4	276	54.3	5882	< 20.8
Hveragerði, sludge gravel bed	Hveragerði	IS	Sludge gb	µg/kg	0.11				< 0.89					2.23						
Klaksvík harbour marina	Klaksvík	FO	Recipient	ng/L	< 0.8	2.82	< 4.17	< 20.8	< 0.42	0.19	< 2.34	< 2.08	< 2.08	< 4.17	< 2.08	552	< 2.08	< 2.08	< 16.0	< 20.8
Klaksvík harbour	Klaksvík	FO	Sediment	µg/kg	< 0.1	< 0.1	< 1.43	< 5.0	< 0.89	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	< 0.10	< 0.50	< 0.50	1.86	< 5
Iggja harbour recipient 2010	Iggja	GL	Recipient 10	ng/L	< 0.8	2.16	< 4.17	< 20.8	1.23	0.25	< 2.34	< 2.08	< 2.08	48.6	< 2.08	398	< 2.08	< 2.08	< 16.0	< 20.8
Iggja harbour recipient 2011	Iggja	GL	Recipient 11	ng/L	< 0.8	10.1	< 4.17	< 20.8	< 0.42	2.98	8.59	< 2.08	2.91	< 4.17	< 2.08	303	5.03	< 2.08	< 16.0	< 20.8
STP Main Hospital	Main Hospitals	FO	Influent	ng/L	< 0.8	404	247	36.8	< 0.42	32.1	30.1	< 2.08	< 2.08	532	87.9	18.9	98.5	53.8	422	< 20.8
STP Main Hospital	Main Hospitals	FO	Effluent	ng/L	< 0.8	356	98.1	188	1.38	120	71.3	18.4	9.94	1140	354	8.27	159	142	11100	< 20.8
STP Main Hospital	Main Hospitals	FO	Sludge	µg/kg	0.18	108	214	9.82	3.23	0.13	< 2.08	< 2.08	< 2.08	97.7	7.52	0.93	14.4	< 0.50	1330	< 5
STP Main Hospital recipient	Main Hospitals	FO	Recipient	ng/L	< 0.8	11.2	< 4.17	< 20.8	< 0.42	4.40	10.7	< 2.08	2.36	6.69	< 2.08	53.0	4.06	3.92	< 16.05	< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 1	ng/L	< 0.8	251	121	311	< 0.42	113	39.0	< 2.08	< 2.08	47.3	6.42	< 3.03	74.3	< 2.08	4630	< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 2	ng/L				470		322	50.3	< 2.08	< 2.08		6.26	< 3.03	21.5	< 2.08		< 20.8
SANA Main Hospital sewer	Main Hospitals	GL	Sludge	µg/kg	< 0.1	41.4	37.5	< 5.0	0.96	0.37	< 2.08	< 2.08	< 2.08	6.18	< 5.0	1.83	1.80	< 0.50	326	< 5
SANA Main Hospital recipient 2010	Main Hospitals	GL	Recipient 10	ng/L	< 0.8	< 0.5	< 4.17	< 20.8	< 0.42	< 0.10	< 2.34	< 2.08	< 2.08	< 4.17	< 2.08	238	< 2.08	< 2.08	< 16.0	< 20.8
SANA Main Hospital recipient 2011	Main Hospitals	GL	Recipient 11	ng/L	< 0.8	< 0.5	< 4.17	< 20.8	< 0.42	< 0.10	3.27	< 2.08	< 2.08	< 4.17	< 2.08	< 3.03	< 2.08	< 2.08	< 16.0	< 20.8
SANA Main Hospital recipient	Main Hospitals	GL	Sediment	µg/kg	< 0.1	7.39	< 1.43	< 5.0	< 0.89	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	0.60	< 0.50	< 0.50	2.26	< 5
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 1	ng/L	< 0.8	277	< 4.17	8590	< 0.42	253	37.7	76.1	< 2.08	13600	1960	1260	4330	1040	43600	< 20.8
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 2	ng/L	< 0.8	350	< 4.17	12700	< 0.42	522	178	190	< 2.08	13900	1230	1240	8700	98.8	69600	< 20.8
Klaksvík Hospital	Hospital Klaksvík	FO	Effluent	ng/L	< 0.8	810	448	< 20.8	7.00	57.4	39.0	6.58	2.57	11400	345	< 3.03	292	251	1700	< 20.8
Klaksvík harbour marina	Hospital Klaksvík	FO	Recipient	ng/L	< 0.8	2.82	< 4.17	< 20.8	< 0.42	0.19	< 2.34	< 2.08	< 2.08	< 4.17	< 2.08	552	< 2.08	< 2.08	< 16.0	< 20.8
Klaksvík harbour	Hospital Klaksvík	FO	Sediment	µg/kg	< 0.1	< 0.1	< 1.43	< 5.0	< 0.89	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	< 0.10	< 0.50	< 0.50	1.86	< 5
Kolonihavnen sewer	Kolonihavnen	GL	Effluent	ng/L	< 0.8	136	12.8	< 20.8	< 0.42	26.1	16.1	< 2.08	< 2.08	84.3	22.6	< 3.03	165	< 2.08	83.9	< 20.8
Kolonihavnen sewer	Kolonihavnen	GL	Sludge	µg/kg	< 0.1	14.7	45.7	< 5.0	0.97	1.07	< 2.08	< 2.08	< 2.08	2.96	< 5.0	6.67	1.92	< 0.50	3.62	< 5
Kolonihavnen recipient	Kolonihavnen	GL	Recipient	ng/L	< 0.8	< 0.5	< 4.17	< 20.8	< 0.42	< 0.10	< 2.34	< 2.08	< 2.08	< 4.17	< 2.08	390	< 2.08	< 2.08	< 16.0	< 20.8
Kolonihavnen recipient	Kolonihavnen	GL	Sediment	µg/kg	< 0.1	0.67	1.47	< 5.0	< 0.89	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	0.79	< 0.50	< 0.50	4.04	< 5
Reykjavík Klettagordum 1	Reykjavík	IS	Influent 1	ng/L	< 0.8	98.4	< 4.17	1000	< 0.42	24.5	17.4	< 2.08	< 2.08	454	598	217	240	32.4	23900	< 20.8
Reykjavík Klettagordum 2	Reykjavík	IS	Influent 2	ng/L	< 0.8	181	< 4.17	1320	< 0.42	26.9	26.2	< 2.08	< 2.08	525	681	287	282	36.6	600	< 20.8
Reykjavík Klettagordum	Reykjavík	IS	Effluent	ng/L	< 0.8	117	< 4.17	1090	< 0.42	19.2	12.6	< 2.08	< 2.08	337	617	217	162	< 2.08	12500	< 20.8
Reykjavík Klettagordum	Reykjavík	IS	Sludge	µg/kg	< 0.1	19.2	18.5	1650	< 0.89	2.39	< 2.08	< 2.08	< 2.08	80.1	168	35.9	74.8	< 0.50	29.7	< 5
STP Sersjantvíkin Torshavn	Tórshavn	FO	Influent	ng/L	3.21	319	< 4.17	< 20.8	< 0.42	112	147	2.49	< 2.08	71.8	90.4	25.6	25.8	60.3	166000	< 20.8
STP Sersjantvíkin Torshavn	Tórshavn	FO	Effluent	ng/L	< 0.8	653	319	< 20.8	6.46	135	73.1	11.8	13.3	612	278	22.8	22.9	111	24600	< 20.8
STP Sersjantvíkin Torshavn	Tórshavn	FO	Sludge	µg/kg	0.10	324	286	13.4	< 0.89	1.05	< 2.08	< 2.08	< 2.08	686	15.3	6.56	33.0	49.7	1880	< 5
STP Sersjantvíkin Torshavn recipient	Tórshavn	FO	Recipient tp	ng/L	< 0.8	23.7	< 4.17	< 20.8	1.39	5.87	8.97	< 2.08	2.49	30.8	< 2.08	343	4.60	5.34	< 16.0	< 20.8
Torshavn harbour marina	Tórshavn	FO	Recipient m	ng/L	< 0.8	< 0.5	< 4.17	< 20.8	< 0.42	< 0.10	< 2.34	< 2.08	< 2.08	< 4.17	< 2.08	69.3	< 2.08	< 2.08	< 16.0	< 20.8
Torshavn harbour shipyard	Tórshavn	FO	Recipient s	ng/L	< 0.8	1.69	< 4.17	< 20.8	< 0.42	0.37	< 2.34	< 2.08	2.54	< 4.17	< 2.08	486	< 2.08	< 2.08	< 16.0	< 20.8
Torshavn harbour marina	Tórshavn	FO	Sediment m	µg/kg	< 0.1	< 0.1	< 1.43	< 5.0	< 0.89	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	0.59	< 0.50	< 0.50	1.62	< 5
Torshavn harbour shipyard	Tórshavn	FO	Sediment	µg/kg	< 0.1	< 0.1	< 1.43	< 5.0	1.32	< 0.03	< 2.08	< 2.08	< 2.08	< 0.17	< 5.0	1.3	< 0.50	< 0.50	1.52	31.9

Table 31. Individual results of antidepressants and hormones (Sludge and sediment concentrations on dry weight basis)

Name on samples	STP name	Country	Code	Unit	Citalopram	Paroxetine	Fluoxetine	Sertraline	Venlafaxine	Levothyroxine	Estrone	17β-Estradiol	Estriol	17α-Ethinylestradiol
Akureyri Dalustod 1	Akureyri	IS	Influent 1	ng/L	130	70.9	< 4.17	< 0.42	61.9	< 1.61	2.83	< 208	< 20.8	< 208
Akureyri Dalustod 2	Akureyri	IS	Influent 2	ng/L	132	96.3	16.5	< 0.42	29.3	< 1.61	3.66	< 208	< 20.8	< 208
Akureyri Sandgerdis. 1	Akureyri	IS	Effluent 1	ng/L	59.0	3.37	5.29	6.02	26.2	< 1.61	3.78	< 208	22.8	< 208
Akureyri Sandgerdis. 2	Akureyri	IS	Effluent 2	ng/L	26.9	2.04	< 4.17	2.99	24.0	< 1.61	3.57	< 208	< 20.8	< 208
Akureyri Dalustod	Akureyri	IS	Sludge	µg/kg	86.7	6.72	7.95	33.6	48.5	< 1.04	18.8	< 50	6.73	< 50
Akureyri Sandgerdis.	Akureyri	IS	Sediment	µg/kg	44.2	6.91	10.8	28.1	73.6	< 1.04	1.11	< 50	< 5	< 50
Hveragerdi	Hveragerði	IS	Influent	ng/L	163	783	< 4.17	121	245	< 1.61	2.04	< 208	< 20.8	< 208
Hveragerdi, effluent biol.	Hveragerði	IS	Effluent b	ng/L	12.2	23.3	< 4.17	9.42	104	< 1.61	< 3.73	< 208	< 20.8	< 208
Hveragerdi, sludge biol.	Hveragerði	IS	Sludge b	µg/kg										
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb1	ng/L	17.7	3.19	< 4.17	4.64	108	1.63	12.9	< 208	57.8	< 208
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb2	ng/L	22.0	45.9	< 4.17	33.5	149	< 1.61	10.2	< 208	53.4	< 208
Hveragerdi, sludge gravel bed	Hveragerði	IS	Sludge gb	µg/kg										
Klaksvik harbour marina	Klaksvík	FO	Recipient	ng/L	0.61	< 1.51	< 4.17	< 0.42	0.94	< 1.61	1.69	< 208	< 20.8	< 208
Klaksvik harbour	Klaksvík	FO	Sediment	µg/kg	0.68	< 0.19	< 0.1	< 0.42	0.30	< 1.04	< 0.66	< 50	< 5	< 50
Iggja harbour recipient 2010	Iggja	GL	Recipient 10	ng/L	1.13	< 1.51	< 4.17	< 0.42	0.54	< 1.61	2.09	< 208	< 20.8	< 208
Iggja harbour recipient 2011	Iggja	GL	Recipient 11	ng/L	6.25	< 1.51	< 4.17	< 0.42	5.58	< 1.61	2.00	< 208	< 20.8	< 208
STP Main Hospital	Main Hospitals	FO	Influent	ng/L	127	< 1.51	< 4.17	2.10	1071	< 1.61	8.59	< 208	58.8	< 208
STP Main Hospital	Main Hospitals	FO	Effluent	ng/L	101	13.4	< 4.17	22.7	647	< 1.61	18.5	< 208	198	< 208
STP Main Hospital	Main Hospitals	FO	Sludge	µg/kg	382	120	30.6	1070	282	1.32	15.4	< 50	5.59	< 50
STP Main Hospital recipient	Main Hospitals	FO	Recipient	ng/L	2.59	< 1.51	< 4.17	< 0.42	7.09	< 1.61	2.01	< 208	< 20.8	< 208
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 1	ng/L	192	20.8	< 4.17	< 0.42	1020	< 1.61	18.9	375	140	< 208
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 2	ng/L						< 1.61	21.0	342	135	< 208
SANA Main Hospital sewer	Main Hospitals	GL	Sludge	µg/kg	134	1.15	0.13	28.8	23.6	3.90	11.0	< 50	26.7	< 50
SANA Main Hospital recipient 2010	Main Hospitals	GL	Recipient 10	ng/L	< 0.42	< 1.51	< 4.17	< 0.42	< 0.42	< 1.61	1.67	< 208	< 20.8	< 208
SANA Main Hospital recipient 2011	Main Hospitals	GL	Recipient 11	ng/L	0.50	1.76	< 4.17	< 0.42	< 0.42	< 1.61	1.88	< 208	< 20.8	< 208
SANA Main Hospital recipient	Main Hospitals	GL	Sediment	µg/kg	1.20	< 0.19	< 0.1	0.16	1.68	< 1.04	< 0.66	< 50	< 5	< 50
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 1	ng/L	2040	130	< 4.17	382	30200	2.72	70.3	302	44.6	< 208
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 2	ng/L	1630	412	< 4.17	61.8	23500	2.74	141	473	50.0	< 208
Klaksvik Hospital	Hospital Klaksvík	FO	Effluent	ng/L	540	< 1.51	< 4.17	0.73	661	< 1.61	7.44	< 208	31.0	< 208
Klaksvik harbour marina	Hospital Klaksvík	FO	Recipient	ng/L	0.61	< 1.51	< 4.17	< 0.42	0.94	< 1.61	1.69	< 208	< 20.8	< 208
Name on samples	STP name	Country	Code	Unit	Citalopram	Paroxetine	Fluoxetine	Sertraline	Venlafaxine	Levothyroxine	Estrone	17 β Estradiol	Estriol	17 α Ethinylestradiol
Kolonihavnen sewer	Kolonihavnen	GL	Effluent	ng/L	130	2.70	< 4.17	1.96	21.3	< 1.61	7.82	357	25.4	< 208
Kolonihavnen sewer	Kolonihavnen	GL	Sludge	µg/kg	51.6	< 0.19	< 0.1	< 0.1	7.01	14.3	6.89	< 50	210	< 50
Kolonihavnen recipient	Kolonihavnen	GL	Recipient	ng/L	< 0.42	< 1.51	< 4.17	< 0.42	< 0.42	< 1.61	< 3.73	< 208	< 20.8	< 208
Kolonihavnen recipient	Kolonihavnen	GL	Sediment	µg/kg	3.69	< 0.19	< 0.1	0.27	2.97	< 1.04	0.80	< 50	< 5	< 50
Reykjavik Klettagordum 1	Reykjavík	IS	Influent 1	ng/L	82.2	91.5	< 4.17	9.63	116	< 1.61	20.9	< 208	73.1	< 208
Reykjavik Klettagordum 2	Reykjavík	IS	Influent 2	ng/L	104	37.8	< 4.17	12.2	124	< 1.61	20.1	< 208	98.7	< 208
Reykjavik Klettagordum	Reykjavík	IS	Effluent	ng/L	69.2	89.5	< 4.17	29.4	138	1.72	13.3	249	53.9	< 208
Reykjavik Klettagordum	Reykjavík	IS	Sludge	µg/kg	46.1	27.4	49.4	141	11400	2.26	17.9	77.7	19.1	< 50
STP Sersjantvíkin Torshavn	Tórshavn	FO	Influent	ng/L	151	631	< 4.17	53.1	880	< 1.61	11.6	465	58.8	< 208
STP Sersjantvíkin Torshavn	Tórshavn	FO	Effluent	ng/L	117	149	< 4.17	19.1	746	< 1.61	13.2	< 208	75.9	< 208
STP Sersjantvíkin Torshavn	Tórshavn	FO	Sludge	µg/kg	255	49.4	3.46	418	53.4	1.48	61.4	< 50	6.42	< 50
STP Sersjantvíkin Torshavn recipient	Tórshavn	FO	Recipient tp	ng/L	4.28	< 1.51	< 4.17	< 0.42	7.92	< 1.61	1.83	< 208	< 20.8	< 208
Torshavn harbour marina	Tórshavn	FO	Recipient m	ng/L	< 0.42	< 1.51	< 4.17	< 0.42	< 0.42	< 1.61	1.79	< 208	< 20.8	< 208
Torshavn harbour shipyard	Tórshavn	FO	Recipient s	ng/L	< 0.42	< 1.51	< 4.17	< 0.42	< 0.42	< 1.61	< 3.73	< 208	< 20.8	< 208
Torshavn harbour marina	Tórshavn	FO	Sediment m	µg/kg	0.14	< 0.19	< 0.1	0.20	< 0.1	< 1.04	< 0.66	< 50	< 5	< 50
Torshavn harbour shipyard	Tórshavn	FO	Sediment	µg/kg	0.82	0.34	< 0.1	3.70	0.16	< 1.04	1.42	< 50	< 5	< 50

Table 32. Individual results of additives in personal care products. (Sludge and sediment concentrations on dry weight basis;EDTA (2) on wet weight basis).

Name on samples	STP name	Country	Code	Unit	EDTA	EDTA (2)	Diethylphtalate	Butylparaben	SDS	SDSEO1-4	CAPB	ATAC-C16
Akureyri Dalustod 1	Akureyri	IS	Influent 1	µg/l	9		1.19	0.016	2.9	30	22	57
Akureyri Dalustod 2	Akureyri	IS	Influent 2									
Akureyri Sandgerdis. 1	Akureyri	IS	Effluent 1	µg/l	18		0.53	0.021	4.2	67	5.5	3.0
Akureyri Sandgerdis. 2	Akureyri	IS	Effluent 2									
Akureyri Dalustod	Akureyri	IS	Sludge	µg/kg	62	39	<8.00	26.0	210	510	510	4000
Akureyri Sandgerdis.	Akureyri	IS	Sediment	µg/kg	<4	<3	<5.00	<4.00	110	360	360	1000
Hveragerdi	Hveragerði	IS	Influent	µg/l	16		3.15	0.029	2	2.7	<0.27	4.1
Hveragerdi, effluent biol.	Hveragerði	IS	Effluent b	µg/l	29		0.77	0.054	2	5.5	1	1.6
Hveragerdi, sludge biol.	Hveragerði	IS	Sludge b	µg/kg	2660	50	<8.00	440	2400	6900	890	110000
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb1	µg/l	25		0.47	0.037	1.8	0.84	0.36	6.9
Hveragerdi, effluent gravel bed	Hveragerði	IS	Effluent gb2									
Hveragerdi, sludge gravel bed	Hveragerði	IS	Sludge gb	µg/kg	844	40	<8.00	<5.00	1700	13000	960	680000
Klaksvik harbour marina	Klaksvík	FO	Recipient	µg/l	1.6		<0.03	<0.007	0.13	0.28	0.27	<0.38
Klaksvik harbour	Klaksvík	FO	Sediment	µg/kg	<4	<2	<7.00	<2.00	<40	<80	<20	340
Iggia harbour recipient 2010	Iggia	GL	Recipient 10	µg/l	<0.3		<0.03	<0.006	0.038	0.34	<0.084	<0.36
Iggia harbour recipient 2011	Iggia	GL	Recipient 11	µg/l	1.3		<0.04	<0.008	0.41	19	7.5	0.88
STP Main Hospital	Main Hospitals	FO	Influent	µg/l	11		0.44	0.037	0.85	68	<0.21	2.7
STP Main Hospital	Main Hospitals	FO	Effluent	µg/l	11		0.28	0.012	5.6	33	<0.085	11
STP Main Hospital	Main Hospitals	FO	Sludge	µg/kg	6785	748	78.0	5.00	1100	4300	630	44000
STP Main Hospital recipient	Main Hospitals	FO	Recipient	µg/l	1.8		0.045	0.010	0.25	0.42	0.27	1.4
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 1	µg/l	420		2.1	0.081	2.4	330	85	3.4
SANA Main Hospital sewer	Main Hospitals	GL	Effluent 2									
SANA Main Hospital sewer	Main Hospitals	GL	Sludge	µg/kg	1116	99	<8.00	<5.00	2000	20000	2300	76000
SANA Main Hospital recipient 2010	Main Hospitals	GL	Recipient 10	µg/l	<0.3		<0.03	<0.006	0.04	<0.16	<0.082	<0.35
SANA Main Hospital recipient 2011	Main Hospitals	GL	Recipient 11	µg/l	<0.3		<0.04	<0.008	<0.035	<0.16	<0.083	<0.36
SANA Main Hospital recipient	Main Hospitals	GL	Sediment	µg/kg	<4	<3	<3.00	<2.00	<40	<80	<20	<20
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 1	µg/l	49		0.50	0.026	1.6	970	0.44	1.3
Landsspítali Hospital Fossvog	Main Hospitals	IS	Influent 2									
Klaksvik Hospital	Hospital Klaksvík	FO	Effluent	µg/l	630		0.170	0.025	1.3	120	47	29
Klaksvik harbour marina	Hospital Klaksvík	FO	Recipient	µg/l	1.6		<0.03	<0.007	0.13	0.28	0.27	<0.38
Klaksvik harbour	Hospital Klaksvík	FO	Sediment	µg/kg	<4	<2	<7.00	<2.00	<40	<80	<20	340
Kolonihavnen sewer	Kolonihavnen	GL	Effluent	µg/l	37		0.86	0.109	0.96	450	89	3.8
Kolonihavnen sewer	Kolonihavnen	GL	Sludge	µg/kg	382	80	<8.00	<5.00	580	40000	2100	1700
Kolonihavnen recipient	Kolonihavnen	GL	Recipient	µg/l	<0.3		<0.04	<0.008	0.082	<0.16	0.1	0.35
Kolonihavnen recipient	Kolonihavnen	GL	Sediment	µg/kg	4.9	1.7	<2.00	<2.00	<40	<80	<20	190
Reykjavík Klettagordum 1	Reykjavík	IS	Influent 1	µg/l	27		1.1	0.048	4.1	4.4	<0.14	61
Reykjavík Klettagordum 2	Reykjavík	IS	Influent 2									
Reykjavík Klettagordum	Reykjavík	IS	Effluent	µg/l	14		0.73	0.011	2.2	1.3	<0.2	31
Reykjavík Klettagordum	Reykjavík	IS	Sludge	µg/kg	525	94	68.0	<17.0	3100	17000	5000	81000
STP Sersjantvikin Torshavn	Tórshavn	FO	Influent	µg/l	16		1.5	0.053	7.9	510	<0.19	87
STP Sersjantvikin Torshavn	Tórshavn	FO	Effluent	µg/l	560		0.51	0.048	0.79	36	<0.084	5.2
STP Sersjantvikin Torshavn	Tórshavn	FO	Sludge	µg/kg	131	16	<50.0	22.8	1000	180000	350	79000
STP Sersjantvikin Torshavn recipient	Tórshavn	FO	Recipient tp	µg/l	1.3		0.14	0.008	0.19	2.2	0.58	1.6
Torshavn harbour marina	Tórshavn	FO	Recipient m	µg/l	<0.3		<0.03	<0.006	0.13	0.4	0.14	<0.37
Torshavn harbour shipyard	Tórshavn	FO	Recipient s	µg/l	<0.3		<0.03	<0.005	0.081	0.37	0.44	<0.37
Torshavn harbour marina	Tórshavn	FO	Sediment m	µg/kg	<4	<2	10.0	<2.00	<40	<80	<20	820
Torshavn harbour shipyard	Tórshavn	FO	Sediment	µg/kg	4.4	1.3	<7.00	<2.00	93	<80	<20	1500

12.2 Sampling manual NILU

12.2.1 *Sampling of sediment, sludge and sedimenting material*

Sample type

Table 33. The original sample matrix overview

Country/ Matrix	Waste water/ Sewage treatment plant influent	Sewage treatment plant effluent	Sewage treatment plant Sludge	Sediment recipient	Surface water recipient	Biota	Sum
Faroe Islands	3	2	2	3	5	0	15
Iceland	3	3	3	3	3	0	15
Greenland	4	-	-	4	4	4	16
Sum	10	5	5	10	12	4	46

Due to the water soluble nature of some of the compounds water and partly sediment or sludge are the most relevant sample types. Biological samples are difficult to handle in chemical analyses of these kinds and can not be included in the project without increasing the costs considerably.

Sample amount

Sample amount for sludge/sedimenting material should be decided by the size of the jars/containers received from the laboratories. Store samples in the freezer, and keep it frozen until it reaches the laboratory.

Sediment/sludge containers to be filled in each location: 2 glass + 1 plastic

Sample amount for water/liquid samples should be decided by the size and the jars/containers received from the laboratories, though such that *glass bottles are filled only ½ full, and plastic bottles 80% full.*

Sampling

If sample pooling will increase the risk for contamination, pooling should be avoided and spot samples shall be taken.

Arrange the sampling bottle to be used (and, if the site is selected for blank sampling, one sampling blank) on a clean spot on the sampling site. Put on the supplied gloves.

Immediately before sampling open the lid of the sampling container (and the sampling blank).

Fill the sample container as described above in “sample amount”, if required using a purified spoon. If the Al-foil protecting the lids is ruptured replace it with new Al-foil and close the lid on the sample jars (and sample blank). NB; if bottles are not provided with Al-foil, then don't use Al-foil, as foil tend to hinder the lid from sealing properly and thus bottles may leak when laid side down in the freezer. Label the sample containers. Put each container in a plastic bag.

Fill in the sample protocol and freeze as soon as possible.

NB; the bottles provided are treated differently and the material chosen with care to minimize adsorption/loss of analyte on container walls. Please fill bottles directly, i.e. do not use transfer vessel for water samples, and do not pour from one bottle of a kind over in a bottle of an other kind.

Sample container

Sediment/sludge samples: All samples shall be filled into plastic containers or cleaned glass jars as provided by the laboratories. Composite samples can be sent in one container. If it is necessary to use several containers for one sample, please, identify clearly that this is part of a composite sample.

Water/liquid samples: Three types of bottles are provided;

- 2 * sample number *silanized brown glass bottles* (from NILU) + 2 times sample number for backup.
- Clean *brown glass bottles* (IVL) + 1 time the sample number for backup.
- *Plastic bottles* (IVL).

From each sampling site all three kinds of bottles are used; and for the glass-bottles double samples are taken. Meaning that from each sampling site in all:

- 4 silanized glass bottles are filled ($\frac{1}{2}$ full).
- 2 glass bottles are filled ($\frac{1}{2}$ full).
- 1 plastic bottle is filled (80%).

Sampling blanks

Sampling blanks will be taken by each country at sites, for instance at the sites where the contamination is supposedly at it's highest. Containers will be provided by the laboratory. The sampling for the sampling blanks is done as follows:

During the sampling of the standard samples, a sample blank jar/bottle is carried into the field along with the other sample contain-

ers. When at the sampling site, the lid/cork on the sampling bland container is removed and replaced again immediatedly–this just to get an idea of how much “background” pollution the handling of the sample containers in the sampling procedure is introducing.

Samplings blanks will be taken at three sites by each country.

Storage and shipment of samples

All samples are stored at approx. -20C. When preparing to ship or transport samples, make contact with the contact persons beforehand to arrange for the shipment and arrival of samples.

All samples are shipped to NILU.

12.3 Sampling form –Water samples

Sample type: STP influent water STP effluent water
 Surface water recipient
 Sampling blank included

Sample name / identity:

Coordinates for the sample site:

Sampling day: Shipped to NILU:

Received at NILU:

Used sampling equipment:

Responsible person:

Sample storage: Freezer Fridge Other

Sample preservation: NaN₃ phosphoric acid Other

Address

Norwegian Institute for Air Research (NILU)

Att. Kirsten Davanger

Instituttvegen 18

NO-2027 Kjeller

Norway

E-mail: Kirsten.Davanger@nilu.no

When sending the samples please send an e-mail including a list specifying the samples to Kirsten.Davanger@nilu.no

12.4 Sampling manual IVL

12.4.1 *Sampling manual*

Precautions to be taken in advance of sampling to avoid contamination

Cosmetic formulations contain various chemicals that potentially can contaminate the samples. Do not use products such as antiperspirant, eye shadow, hair spray, or skin lotions on the day of sampling. Only specially cleaned sampling containers provided by the laboratory should be used.

Equipment provided per sampling site

- 20 x 1 l glass bottles.
- 10 x 1 l plastic bottles.
- 5 x 120 ml plastic jars.
- 5 x 120 ml glass jars.
- Plastic bags 2 l for jar and 3 l for glass bottles.
- Muffled Al-foil (packed in Al-foil).
- Etiquettes.
- Plastic glows.

Sampling of water

- Arrange the sampling bottles to be used on a clean spot on the sampling site. Put on the supplied gloves.
- Immediately before sampling open the lid of the sampling container.
- Fill the sample container: 2 glasses and 1 plastic bottle per sampling station. If the Al-foil protecting the lids of the glass bottles is ruptured replace it with new Al-foil. Close the lid on the sample bottles.
- Mark bottles with sample name/identity.
- Put each bottle in a plastic bag (3 l).
- Fill in the sample protocol.
- Store the samples in refrigerator (5–10°C).

Sludge samples

- Put on the supplied gloves.
- Open the lids of one plastic jar and one glass jar.
- Fill the jars (one glass plus one plastic) with sludge / sediment and close the lids.
- Put each sample in a plastic bag (2 l).
- Mark the sample with name/identity.

- Fill in the sample protocol.
- Store the samples in freezer (-18°C).

Storage and transport

Store sludge / sediment samples frozen and water samples refrigerated. Send to the laboratory as soon as possible in such a way that the samples will reach the laboratory within one day (DHL or equivalent courier services). Send samples in the same containers used for the provided sampling material. Make sure to insulate glass bottles properly to avoid breakage during transport. Label of content to be simply “water samples”.

When sending the samples please send also an e-mail including a list specifying the samples to Mikael Remberger (mikael.remberger@ivl.se).

Address

IVL Swedish Environmental Research Institute
 Mikael Remberger
 Vallhallavägen 81
 114 23 Stockholm; Sweden.

12.5 Analysis of selected substances – Sample protocol

Sample name/identity	Sample type (water/sludge)	Coordinates sampling site	Sampling date	Used sampling equipment	Comments

Responsible person

Shipped to IVL:

Received at IVL:

Address

IVL Swedish Environmental Research Institute
 Mikael Remberger
 Valhallavägen 81
 114 23 Stockholm
 Sweden

When sending the samples, please send an e-mail including a list specifying the samples to Mikael Remberger (mikael.remberger@ivl.se).

Sampling form–Sediment, sludge, and sedimenting material

Sample type: Sediment
 Digested sludge
 Sedimenting material
 Sampling blank

Sample name / identity:

UTM-Coordinates for the sample site:

Sampling day: Shipped to NILU:
Received at NILU:

Used sampling equipment:

Responsible person:

Sample storage: Freezer Fridge Other

Address

Norwegian Institute for Air Research (NILU)
Att. Kirsten Davanger
Instituttvegen 18
NO-2027 Kjeller
Norway
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TemaNord 2013:541

Pharmaceuticals and additives in personal care products as environmental pollutants

The application of pharmaceuticals and personal care products is substantial in industrialized and high-income north-western European societies. Faroe Island, Iceland and Greenland are part of this modern society, although some areas are more suffused by technology and modern living than others. This also pertains to the standards of the local solutions for waste water treatment systems, but not so much to the health services. The present report summarises the results of screening analyses of pharmaceuticals and additives in personal care products in presumed hotspots in Faroe Islands, Iceland and Greenland. The study focuses on sewage lines from households and industry in general, and from hospitals. In all 38 pharmaceuticals or metabolites of pharmaceuticals and 7 personal care products were analysed.



TemaNord 2013:541
ISBN 978-92-893-2561-5



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