

Quaternary ammonium compounds

Analyses in a Nordic cooperation on screening





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Lennart Kaj, Petra Wallberg, Eva Brorström-Lundén

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Nordic Council of Ministers Ved Stranden 18

DK-1061 Copenhagen K Phone (+45) 3396 0200

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Preface

The aim of the Nordic environmental screening is to obtain a snapshot of the occurrence of potentially hazardous substances, both in regions most likely to be polluted and in some very pristine environments. The focus is on less known, anthropogenic substances and their derivatives, which are either used in high volumes or are likely to be persistent and hazardous to humans and other organisms. In this study the occurrence of quarternary ammonium compounds in environmental samples from the Nordic countries has been investigated. This was done to gain experience in environmental screening of this kind of substances and provide a better knowledge of quarternary ammonium compounds in the environment. If the substances subjected to screening are found in significant amounts this may result in further investigations or monitoring on national level and measures to reduce contamination.

Some of the samples were also analyzed for sodium dodecyl sulphate, sodium laureth sulphate and cocoamidopropylbetaine. These results are only briefly presented.

The Nordic screening project is run by a steering group with the following representatives:

- Susanne Boutrup, DCE Danish Centre for Environment and Energy, DCE – Nationalt Center for Miljø og Energi, Aarhus University, Denmark (sub@dmu.dk).
- Jaakko Mannio, Finnish Environment Institute (SYKE), Suomen ympäristökeskus, Finland (jaakko.mannio@ymparisto.fi)
- Maria Dam, Environment Agency, Umhvørvisstovan, the Faroe Islands (mariad@us.fo).
- Tina Mønster, Ministry of Housing, Nature and Environment, Ineqarnermut, Pinngortitamut Avatangiisinullu Naalakkersuisoqarfik, Greenland (tinm@nanoq.gl).
- Bergþóra Hlíðkvist Skúladóttir, The Environment Agency of Iceland, Umhverfisstofnun, Iceland (bergthoras@Umhverfisstofnun.is).
- Britta Hedlund, Swedish Environmental Protection Agency, Naturvårdsverket, Sweden (Britta.Hedlund@naturvardsverket.se).
- Linda Linderholm, Swedish Environmental Protection Agency, Naturvårdsverket, Sweden (linda.linderholm@naturvardsverket.se).

- Eivind Farmen, Norwegian Environment Agency, Miljødirektoratet, Norway (eivind.farmen@miljodir.no).
- Bård Nordbø, Norwegian Environment Agency, Miljødirektoratet, Norway (bard.nordbo@miljodir.no).

The project is financed and supported by the Nordic Council of Ministers through the Nordic Chemicals Group and the Aquatic Ecosystems Group as well as the participating institutions. The chemical analyses have been carried out by IVL Swedish Environmental Research Institute.

The respective participating Nordic countries organised sample selection, collection and transport of samples based on a sample manual provided by the analytical laboratory.

Summary

The overall aim of this screening study was to investigate the occurrence of quaternary ammonium compounds, QACs, in environmentally related samples from the Nordic countries. QACs are widely used as ingredients in industrial applications and find widespread use in household products, including fabric softeners, detergents, disinfectants, preservatives, and a range of personal care products.

Cations of the types alkyltrimethyl ammonium (ATAC), alkyl dimethyl benzyl (benzalkonium, BAC) and dialkyl dimethyl ammonium (DDAC), in total 17 compounds, were measured in effluents and sludges from waste water treatment plants, sediments and fish. Usually the participating countries (Denmark, Faroe Islands, Finland, Greenland, Iceland, Norway and Sweden) contributed three samples from each sample type. The relatively few samples and sample types were intended to give a "snap shot" of the situation.

In general there was a widespread occurrence of QACs in all included matrices.

Concentration in effluents varied over a large range. A majority of the "high concentration" effluents showed, even after ten times dilution, concentrations of ATACs and/or BACs that could pose a risk to the aquatic environment. Ecotoxicological data is lacking especially for long chained ATACs.

All the selected compounds were present in all sludges. Concentrations in sludge varied much less than in effluents. Patterns of individual compound concentrations were not clearly different among countries.

Behentrimonium (ATAC-C20 and ATAC-C22), recently described as a new emerging contaminant, was found in a majority of the samples, also in fish muscle and liver.

1. Frame of the study

The occurrence and the environmental risk of chemicals are prioritized issues in several international legislative acts (e.g. the EUs Water framework directive, Registration Evaluation Authorisation of Chemicals (REACH), Stockholm Convention on Persistent Organic Pollutants (POPs) and the Convention on long-range trans-boundary air pollution – LRTAP) but in addition to already well known substances included in different monitoring programs there is a focus on "emerging" chemicals in different research– and screening programs.

In this screening study concentrations of quaternary ammonium compounds were determined in wastewater effluents and sludge, sediments and fish collected at different locations representing dispersed use related to many activities and products. Background samples (fish) were also included in the study.

There are limitations and uncertainties in screening studies; measurements of a chemical are carried out in several media at several sites but only with a few samples at each site which gives only a "snap shot" of the situation. However this screening covers a limited number of matrices where it was expected to find the compounds investigated and with a harmonized sampling strategy among the different Nordic countries.

The matrices included were effluent and sludge from waste water treatment plants (WWTPs), sediment and biota (fish) with a wide geographical distribution. When possible, the samples were chosen to facilitate comparisons between areas/regions.

This screening study and selection of compounds were based on a literature survey by Johansson (2012).

2. Background

2.1 Investigated compounds

Quaternary ammonium compounds (QACs or Quats) share the common quaternary ammonium cation illustrated in Figure 1a. The nature of the substituents R1, R2, R3 and R4 are varied in order to influence the properties of the ion. In pure form the QACs will also contain an anion such as chloride, bromide or methosulfate. When in solution, like in a sewage system, the substances will be dissociated and the anion the QACs were once associated with will be irrelevant.

In this work QAC cations of the types alkyltrimethyl ammonium (ATAC, Figure 1c), alkyl dimethyl benzyl (benzalkonium, BAC, Figure 1b) and dialkyl dimethyl ammonium (DDAC, Figure 1d) were investigated. The QACs and the nature of the substituents R1, R2, R3 and R4 in each analyte are listed in Table 1.

Figure 1a: General formula of a quaternary ammonium cation



Figure 1b: Benzalkonium ion



Figure 1c: Examples of ATAC



Figure 1d: DDAC ions



Alternative abbreviations for ATAC used in the literature are ATMA, ATMAC and TMAC. Names referring to the origin of the alkyl chains are often used. "Tallow" in ditallowdimethyl ammonium chloride (DTDMAC) refers to alkyl chain lengths mainly C18 and C16, "coco" to C12 and C14. ATAC-C16 can also be called cetrimonium ion, ATAC-C18 DSDMAC (for distearyldimethyl ammonium chloride). The name behentrimonium, from behenic (docosanoic) acid, refers to products containing ATAC-C20 and ATAC-C22.

Table 1: Abbreviations and nature of substituents R1–R4 in ammonium cations (Figure 1a) included in this work

Name	R1	R2	R3	R4
ATAC-C12	n-C12	Methyl	Methyl	Methyl
ATAC-C14	n-C14	Methyl	Methyl	Methyl
ATAC-C16	n-C16	Methyl	Methyl	Methyl
ATAC-C18	n-C18	Methyl	Methyl	Methyl
ATAC-C20	n-C20	Methyl	Methyl	Methyl
ATAC-C22	n-C22	Methyl	Methyl	Methyl
BAC-C12	n-C12	Benzyl	Methyl	Methyl
BAC-C14	n-C14	Benzyl	Methyl	Methyl
BAC-C16	n-C16	Benzyl	Methyl	Methyl
BAC-C18	n-C18	Benzyl	Methyl	Methyl
DDAC-C10	n-C10	n-C10	Methyl	Methyl
DDAC-C12	n-C12	n-C12	Methyl	Methyl
DDAC-C14	n-C14	n-C14	Methyl	Methyl
DDAC-C14:16	n-C14	n-C16	Methyl	Methyl
DDAC-C16	n-C16	n-C16	Methyl	Methyl
DDAC-C16:18	n-C16	n-C18	Methyl	Methyl
DDAC-C18	n-C18	n-C18	Methyl	Methyl

BAC can also be abbreviated ADMBA or ADBAC. Commercial benzalkonium chloride is a mixture of alkylbenzyldimethylammonium chlorides of various even-numbered alkyl chain lengths. EPA (2006) lists twelve different CAS numbers for BACs where specifications of alkyl chain lengths are given. They tend to group into three categories:

```
    Dominated by C12: 50–70% C12
25–30% C14
5–10% C16
```

•	Dominated by C14:	14-40% C12
		50-60% C14
		10-28% C16
•	Almost only C14:	1-5% C12
		>90% C14
		1-5% C16

DDAC can also be abbreviated DADMA or DMAC. For DDACs with two identical chains the length is indicated only once (DDAC-C10), if the chain lengths are different they are both indicated (DDAC-C14:16). DDMAC refers to DDAC-C10. The abbreviation DDAC (DD for DiDecyl) can sometimes refer to DDAC-C10 only (DD for Dialkyl Dimethyl).

2.2 Properties and use

The compound properties vary with alkyl chain length. Water solubility decreases, and adsorptivity to surfaces increases, with increasing chain length. Some general properties of DDACs are listed in Table 2.

Table 2: Solubility and properties DDACs of different chain length (Porter, 1994)

Chain length	Solubility	Properties
2 C8 chains	Very soluble in water	Mild germicide
2 C10 chains	Soluble in water	Strong germicide
2 C12 chains	Poor solubility in water	Weak germicide
2 C14 chains	Low solubility in water	Antistatic
2 C16-18 chains	Practically insoluble in water	Softener and antistatic

Reorted values for log Kow and BCF (bioconcentration factor) are compiled in Table 3.

Table 3: Reported values for log Kow and BCF (EOSCA 2000)

	log Ko	ow		BCF		
ATAC-C8			2.4 ^b	0.5 ^b		
ATAC-C12			41 ^b	35°		
ATAC-C12, C14	1.56					
ATAC-C16	3.18	2.69 ^a				
ATAC-C16, C18			1,962 ^b	141 ^b		
ATAC-C18	4.26					
DDAC-C16:18			32 ^c	13 ^c		
DDAC-18	1.81 ^ª		38 ^b	104 ^b	3 ^b	32 ^b

^a: Ying, 2006.

^b:Fathead minnow, *Pimephales promelas*.

^c: Bluegill sunfish, *Lepomis macrochirus*.

QACs are widely used as ingredients in industrial applications and find widespread use in household products, including fabric softeners, detergents, disinfectants, preservatives, and a range of personal care products (Li *et al.*, 2010).

The major cationic surfactant used in fabric softeners worldwide DTDMAC (ATAC-C16, C18) is, due to its poor biodegradation kinetics, beeing replaced in Europe by the esterquat DEEDMAC (diethyl esterdimethyl ammonium chloride) (Ying, 2006, citing a source from 1995).

Behentrimonium chloride or methosulphate, containing ATAC-C20 and ATAC-C22 are used increasingly in personal care products, especially in hair care products (Lara-Martín *et al.*, 2010). The data on Log K_{ow} and BCF is limited (Table 3) and lacking for ATAC-C20 and ATAC-C22. However the available data suggests that bioaccumulation for ATACs might increase with increasing chain length.

The SPIN database consists of summarized information from the Nordic product registers on the use of chemical substances. (It should be noted that SPIN lists ingredients in chemical preparations, not in finished consumer articles and that the reporting to the SPIN database could vary between countries). Johansson (2012) made a search on substances containing the word "quaternary" in their names and got 104 hits of which 22 occurred in annual volumes exceeding 10 tonnes (2010). The total annual volume was 5,196 tonnes. We divided these substances into the groups ATACs, BACs, DDACs and "others". The annual volumes were 442 tonnes (9%), 1,218 tonnes (23%), 2,813 tonnes (54%) and 723 tonnes (12%) for the respective groups (Figure 2).



Figure 2: Total annual use 2010 (tonnes) of QACs extracted from the SPIN database (see text)

2.3 Environmental fate and ecotoxicity

QACs are cationic surfactants that strongly sorb onto suspended particulates and sludge, and therefore biodegradation in sediment and sludge is an important process for determination of their fate in the environment (Ying, 2006).

Under aerobic conditions, the biodegradability of QACs generally decreases with the number of non-methyl alkyl groups i.e. DDAC is less biodegradable than ATAC (Ying, 2006). Moreover, substitution of a methyl group with a benzyl group can decrease biodegradability further i.e. BAC is likely less biodegradable than ATAC.

In contrast, under anaerobic conditions, no or very poor primary biodegradation of QACs has been reported and no evidence of ultimate biodegradation.

Thus, except for ultrahydrophobic DTDMAC (DDAC-C16, DDAC-C18), aerobic biodegradation is an important process within WWTPs for QACs, such as DDAC-C10, ATAC-C12 – C16, and BAC-C12 – C18 (Clara *et al.*, 2007). However, the very strong sorption properties and resistance to desorption of even the most soluble QACs gives an explanation to that these compounds are found in sediments downstream WWTPs in appreciable quantities and imply that QACs can be relatively persistent in receiving waters (Martinez-Carballo *et al.*, 2007b; Li and Brownawell, 2010; Lara-Martin *et al.*, 2010). Moreover, QACs in sludge amended to soil are, also due to the adsorptive properties, not expected to contaminate surface and ground waters (US EPA, 2006).

QACs have disinfectant properties and, thus, high concentrations may inhibit the microbial processes in WWTPs.

Available toxicity data suggest that the substitution of a methyl group with a benzyl group increases the toxicity but that there is no difference in toxicity between homologues of different chain length (Ying, 2006). This could be attributed to a lower bioavailability of the longest chain homologues due to their decreasing solubility. For instance in WWTPs the toxicity to methanogenesis has been reported to decrease with increasing alkyl-chain length.

Utsunomiya *et al.* (1997) studied acute toxic effects of ATAC, BAC and DDAC on unicellar green alga *Dunaliella* sp. by measuring ¹³C-glycerol. The EC50 (24h; median effective concentrations) were 0.79 mg/l for ATAC, 1.3 mg/l for BAC, 18 mg/l for DDAC. Garcia *et al.* (2001) reported acute toxicity tests on *Daphnia magna* and *Photobacterium phosphoreum* for members of the groups ATAC and BAC. The EC50 on *D. magna* (24h; immobilization) ranged from 0.38 to 0.13 mg/l for ATAC-C12, ATAC-C14 and

ATAC-C16 and from 0.13 to 0.22 mg/l for BAC-C12, BAC-C14 and BAC-C16. EC50 on *P. phosphoreum* ranged from 0.15 to 0.63 mg/l. Sandbacka *et al.* (2000) reported EC50 values for ATAC ranging from 0.058 to 0.37 mg/l for *D. magna* and from 0.6 to 40 mg/l for rainbow trout.

US EPA (2006) reported lower acute effect values for BAC that was categorized as very highly toxic to *D. magna* (LC50 = 5.9 µg/l) and highly toxic to fish (LC50 = 280 µg/l). Chronic effects were seen in fish at a concentration of 32 µg/l and a no observable adverse effect concentration (NOAEC) of 4.2 µg/l for *Daphnia*. A 21-day growth NOEC value on *D. magna* have been reported to be 0.38 mg/l for DTDMAC (Lewis and Wee, 1983). Ferk *et al.* (2007) reported that BAC induced a moderate but significant genotoxic effect at 1.0 mg/l, and DDAC-C18 bromide (DDAB) caused damage at 0.3 mg/l in mammalian and plant-derived cells.

In the European legislation, REACH, the aquatic criterion for toxicity is based on a long-term no-observed effect concentration (NOEC) for chronic tests on fish, crustacean (daphnids) and algae (ECHA, 2012). A substance is considered to fulfil the toxicity criterion (T) when the long-term no-observed effect concentration (NOEC) for marine or freshwater organisms is less than 0.01 mg/l. If only data from acute toxic test are available a substance is considered to potentially meet the criteria for T when an acute E(L)C50 value is less than 0.1 mg/l. Thus, according to these criteria and the data from US EPA, ATACs with moderate chain lengths can be considered as potentially toxic and BAC as toxic. Available data suggest that DDACs is less toxic than ATACs and BAC.

2.4 Previous measurements

A compilation from the literature of some previous measurements is presented in Table 4. This data summary shows that QACs are present in samples from WWTPs, that these substances are measured in sediments downstream WWTPs and that they are found in sediments from urban sites. These data will be used for comparison in the results section.

		3																			
						АТА	<u>u</u>				BAC						DDAC				
Type			Unit	C12	C14	C16	C18	C20	C22	C12	C14	C16	C18	C10	C10	C14 (:14:16	C16 (C16:18	C18	Reference
Influent	3 WWTPS in Austria, 2004	n=5, median	l/gn	290	300	5,900				51,000	25,000	1,900	1,000	18,000	170	120		l,100		3,500	Martínez-Carballo <i>et</i> <i>al.</i> , 2007a
Effluent	3 WWTPS in Austria, 2004	n=8, median	l/gn	v	v	30				420	210	68	43	87	∞	26.5		42		61	Martínez-Carballo <i>et</i> <i>al.</i> , 2007a
Sludge	3 WWTPS in Austria, 2004	n=6, median	g/gn	32	80	720				5,800	5,400	670	1,500	7,300	096	350		2,800		9,800	Martínez-Carballo <i>et</i> <i>al.</i> , 2007b, from Fig 3.
Sludge	5 WWTPs in Sweden, 2008	n=5, median	ng/g dw			9,200								1,200							Kaj 2010, Reg Screen 2008
Sediment	Jamaica bay, New York, 1998	n=15, median	g/gn			170	390			140	210	200	440	150		390	, 730	005,t	14,000	20,000	Li, Brownawell, 2010
Sediment	Jamaica bay, New York, 1998	n=7, median	g/gu			170	430	21	200												Lara-Martín <i>et al.,</i> 2010
Sediment	Jamaica bay, New York, 2003	n=7, median	g/gn			86	160	67	700												Lara-Martín <i>et al.,</i> 2010
Sediment	Jamaica bay, New York, 2008	n=7, median	g/gu			210	210	100	1,800												Lara-Martín <i>et al.,</i> 2010
Sediment	rivers down- stream WWTPs in Austria, 2004	n ca 10, median	g/gu		6.8					130	66	12	12	76	3.3	3.6		100		310	Martínez-Carballo <i>et</i> <i>al.</i> , 2007b

Table 4: Previous measurements

3. Methodology

3.1 Sampling sites and sample selection

The purpose of this screening was to cover the matrices effluent and sludge from waste water treatment plants (WWTPs), sediment and biota (fish) with a wide geographical distribution at locations where it's expected to find the compounds investigated. Also a sediment sample and some fish samples from background areas were included. When possible, samples were chosen so as to facilitate comparisons between areas/regions.

Each country made their own selection of sample sites according to the strategy previously agreed upon in the steering group.

Sampling was done according to a sampling manual provided by the analysing laboratory (Appendix 1).

All samples collected are listed in Appendix 2, where also the sampling characteristics are given in detail. The sampling sites in each country are presented below and their locations are shown in maps.

3.1.1 Denmark

WWTP effluent and sludge

Effluent and sludge were sampled at tree waste water treatment plants (WWTP), Avedøre Wastewater Services, Køgeegnens Renseanlæg, Lynetten Renseanlæg. Table 5 shows the total annual effluent of nutrient and water from each of the waste water plants is shown and the number of the population served by the waste water plants.

Table 5: Annual effluent of nutrient and water and population served by the waste water plants (Naturstyrelsen, 2012; Naturstyrelsen, 2013)

	Total-N (kg/year)	Total-P (kg/year)	BI5 mod. (kg/year)	Water (m3/year)	Population load (pe)
Avedøre Wastewater Services ¹⁾	Not calculated	Not calculated	Not calculated	28,986,329	238,536
Køgeegnens Renseanlæg ²⁾	16,530	2,984	11,071	6,031,939	93,077
Lynetten Renseanlæg ²⁾	465,617	166,775	800,757	58,301,604	532,339

¹: data for 2011.

²: data for 2012.

The three waste water treatments plants receive waste water from households as well as industry. It is tree modern treatment plants with mechanical and biological treatment, nitrification and denitrification as well as chemical sedimentation. They are all placed in Copenhagen or in the vicinity of Copenhagen. Avedøre Wastewater Services receives waste water from ten suburban municipalities west of Copenhagen, Køgeegnens Renseanlæg receives waste water from the city Køge and surroundings, which is about 40 km south west of Copenhagen and Lynetten Renseanlæg receives waste water from Copenhagen city.

The samples from Lynetten Renseanlæg were collected shortly after a rain storm, which meant that the waste water passed untreated through the treatment plant. Therefore the waste water samples from Lynetten are more representative for influent than effluent.

Sediment

Sediment samples were collected from a lake, Thors Sø and two marine areas close to the coast, Langerak and Nivå.

Thors Sø is in Mid Jutland close to Silkeborg. The lake is surrounded by trees and housing and is used for recreative purposes such as e.g. bathing. The catchment is relatively big and is mostly covered by forest and ungrown fields. Thors Sø is an oligothrophic to mesotrophic lake according to the criteria defined in the habitat directive. The lake receives water from brook, springs and groundwater but also rainwater and waste water from sparsely built-up areas.

Langerak is in the eastern part of Limfjorden between Aalborg Østhavn and Mou and about 20 km west of the outlet to Skagerak. Aalborg is a city with about 100,000 citizens and Mou is a small town with about 1,100 citizens. The station has a sandy bottom with scattered eelgrass vegetation.

Nivå Bugt is situated in the northern part of The Sound on the Danish side of the Island of Ven, around 15 nautical miles north of Copenhagen on the Danish coast and Malmö a bit further down on the Swedish side. Helsinor-Helsingborg is 7 miles north of the Nivå Bugt, and the area is part of the major shipping route to the Baltic and ferry crossings at Helsinor-Helsingborg. The current is usually northerly from the Baltic Sea, but changes to southern when the wind is in coming from the west.

The samples were collected and handled according to the sampling manual and the Danish requirements on sediment sampling for hazardous substances.

Fish

Fish were sampled at the same locations as the sediment samples.

Fish from Thors Sø were collected in a fishing net by the Danish Nature Agency coordinated with sampling for the Danish monitoring programme. Fish from Langerak were collected in a fish trap by a local fisherman using bottom trawl. The samples were stored on ice aboard the ship and collected directly upon arrival to the harbour of Skodsborg and transported to DCE, AU, Roskilde on ice, before freezing and packaging for final transport to the analytical laboratory. The samples were collected and handled according to the sampling manual and the Danish requirements on fish sampling for hazardous substances.

3.1.2 Faroe Islands

WWTP effluent and sludge

Effluents were sampled at the Sersjantvikin WWTP, Torshavn, and in the Main Hospital WWTP, also in Torshavn. A sample of influent to the Main Hospital WWTP was taken from the side arm collecting discharge from the service buildings of the hospital, like that of the laundry, as well as parts of the hospital wards, including the maternity and psyciatric wards. Samples of effluents were also taken from Klaksvik Hospital.

Sludge was sampled in the Sersjantvikin WWTP, Torshavn and in the Main Hospital WWTP, Torshavn. The Sersjantvikin WWTP, Torshavn, receives domestic wastewater only and from approx. 1,000 pe. This WWTP may be described as consisting of a primary purification step. The Main Hospital has 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, of basically the same outline as the Sersjantvíkin WWTP, but in addition it contains a bio-filtering sprinkler system. Also, the main hospital WWTP sedimentation tank was in open-air. Klaksvík hospital is a small hospital with 36 hospital beds, and performes clinical chemical analyses and x-ray diagnostic analyses.

Sediment

Sediment was sampled in Torshavn harbour, in the inner areas near the marina and the shipyard (site BÁ), and near the discharge site of effluents from the Main Hospital WWTP (site ÁL). Sediment was also sampled in Klaksvik harbour (site á Stongum).

Sediment from Skálafjord, at station SK05 of the Faroe Marine Research Institute (62°07,126 N; 6°44,626 V), were sampled on 26/8/2013, by a Haps sediment sampler. The uppermost 2 cm of the sediment core were taken for QAS analyses. The sediment sample was placed in a heat-treated glass jar with heat-treated Al-foil between jar and lid, and shipped to the laboratory.

Fish

Cod (n=10), Gadus morhua, were taken on tackle in the harbour area of Runavík, Skálafjord, in November 2013. Cod (n=8) were taken on tackle in Klaksvik harbour within 300 m from the Klaksvik hospital waste water discharge site and the sediment sampling site á Stongum. Cod (n=2) was also taken in Torshavn harbour, at the western quay.

Cod, Gadus morhua, taken from Mýlingsgrunnur north-west on the Faroe shelf of approx. 45 cm fork length taken in September 2013 by the research vessel M7T Magnus Heinason (at 62° 27 N; 7° 16 W). The fish (n=6) were wrapped in heat-treated Al-foil and shipped intact to the laboratory for dissection and subsampling.

3.1.3 Finland

WWTP effluent and sludge

Effluent and sludge samples (single, dewatered) were collected at three municipal WWTPs: Kakolanmäki, Turku (population equivalent 890,000), Viinikanlahti, Tampere (pe 300,000) and Viikki, Helsinki (pe 1,220,000). All these plants collect small or medium enterprise (SME) industrial wastewaters as well.

Sediment

Sediment was sampled at urban locations in the same cities as was used for WWTP sampling; Turku, Tampere and Helsinki.

Fish

Fish (Perch) were sampled in Airisto in the Turku archipelago, Pirkkalan Pyhäjärvi in Tampere and Vanhankaupunginlahti in Helsinki.

3.1.4 Greenland

WWTP effluent and sludge

Effluent were sampled from four sewage drains placed in Nuuk: Quinngorput (5,500 pe), Sana Bay (2,100 pe), Kolonihavnen (4,900 pe) and Nuuk Imeq (5,200 pe). Greenland does not treat wastewater, which is why Greenland has not provided sludge samples, but only effluent. All sites receive waste water from households as well as from industry. In addition, Sana Bay does receive waste water from the Queen Ingrid's Hospital, the national hospital of Greenland.

Sediment

Surface sediments were sampled from the vicinity of three sewage discharge points: Sana Bay, Quinngorput and Nuuk Imeq, in addition to the background area Narsaq Ilua.

Fish

Fish were caught and sampled using net close to the sewage discharge points: Sana Bay (Cod, *Gadus morhua*), Kolonihavnen/Vildmandsnæs (Greenland cod, *Gadus ogac*), and Nuuk Imeq (Greenland cod, *Gadus ogac* and shorthorn sculpin, *Myoxocephalus scorpius*). While shorthorn sculpin is a stationary bottom dwelling species and therefore convenient

for monitoring hotspot contamination and gradients, the two cod species are more mobile but top predators with a higher potential for biomagnifications.

3.1.5 Iceland

WWTP effluent and sludge

Effluent samples were sampled at three locations in Iceland, at two WWTPs in Reykjavík (Ánanaust and Klettagarðar) as well as from the WWTP in Hafnarfjörður. Samples from Reykjavík were taken at 17:00 and the sample from Hafnarfjörður was taken at 16:30. Sludge samples were only taken from the WWTP in Hafnarfjörður. Samples were taken at the same occasion as effluent samples.

Sediment

Iceland did not contribute sediment samples.

Fish

Cod (Gadus morhua) was collected as a part of the national monitoring program according to the OSPAR guidelines. 25 individuals were sampled N–NW of Iceland (662142°N, 253977°W) and 25 individuals were sampled NE of Iceland (651787°N, 122593°W). The fish was bled and frozen at sea. The fish was thawed up at the laboratory, both fillets removed and homogenized into two separate pooled samples representing N–NW and NE of Iceland, respectively. Pooled sample was stored frozen in pre-cleaned glass jars until analyses.

3.1.6 Norway

WWTP effluent and sludge

Effluent and sludge were sampled at Veas WWTP, Oslo (700,000 pe). At Veas waste water is treated both with chemical and biological methods. Sludge is stabilized with mesofile anaerobe treatment and vacuum dried. Samples were taken from treated, ready to use, sludge.

Sediment

Surface sediments were sampled from the vicinity of Veas WTTP discharge point in the Inner Oslofjord.

Fish

Fish (Cod, *gadus morhua*) was sampled in vicinity of Veas WTTP discharge point in the Inner Oslofjord. The samples were delivered as frozen liver.

3.1.7 Sweden

WWTP effluent and sludge

Effluent and sludge were sampled at Henriksdal WWTP, Stockholm (660,000 pe), Gässlösa WWTP, Borås (110,000 pe) and Bollebygd WWTP (2,200 pe). All three WWTPs use activated sludge treatment processes. In Henriksdal and Gässlösas the sludge is anaerobically digested. Dewatered sludge from the anaerobic chamber was sampled. In Bollebygd the sludge is treated in a reed bed from which a combined sample was taken.

Sediment

Surface sediments were sampled from the vicinity of the discharge points of the three WWTPs.

Fish

Fish were caught using net close to the discharge points of the three WWTPs.





Figure 4: Sampling sites Finland



Sample type

- Effluent
- Sediment
- Sludge

Figure 5: Sampling sites Faroe Islands



Figure 6: Sampling sites Greenland



Sample type

- Effluent
- Fish
- Sediment
- Sludge

Figure 7: Sampling sites Iceland



Sample type

Sediment

Sludge

Figure 8: Sampling sites Norway



Figure 9: Sampling sites Sweden



3.2 Sampling methods

Effluents were sampled as grab samples directly into 0.5 litre heat treated (400 °C) glass bottles containing 2 ml 6M H_3PO_4 and kept refrigerated. Effluents were also sampled in 1 litre pe-bottles which were frozen and used for analysis of anions.

3.3 Analytical method, cations

3.3.1 Chemicals

Substances to be used as analytical standards (Table 6) were purchased from Sigma-Aldrich. EADAC-C12 and EADAC-C16 was used as internal standards.

Table 6: Substances used as analytical standards

Abbreviation	CAS #	Name	Purity
DDAC-C10	2390-68-3	Didecyldimethylammonium bromide	98%
DDAC-C12	3282-73-3	Didodecyldimethylammonium bromide	98%
DDAC-C14	68105-02-2	Dimethylditetradecylammonium bromide	>=97%
DDAC-C16	70755-47-4	Dimethyldihexadecylammonium bromide	97%
DDAC-C18	3700-67-2	Dimethyldioctadecylammonium bromide	>=98.0%
BAC-C12	139-07-1	Benzyldimethyldodecylammonium chloride	>=99.0%
BAC-C14	139-08-2	Benzyldimethyltetradecylammonium chloride	>=99.0%
BAC-C16	122-18-9	Benzyldimethylhexadecylammonium chloride	
BAC-C18	206752-43-4	Benzyldimethylstearylammonium chloride monohydrate	90%
ATAC-C12	112-00-5	Dodecyltrimethylammonium chloride	>=99.0%
ATAC-C14	1119-97-7	Myristyltrimethylammonium bromide	>=99%
ATAC-C16	57-09-0	Hexadecyltrimethylammonium bromide	99%
ATAC-C18	112-03-8	Trimethyloctadecylammonium chloride	>95%
AEDAC-C12	68207-00-1	Dodecylethyldimethylammonium bromide	>=98%
AEDAC-C16	124-03-8	Ethylhexadecyldimethylammonium bromide	>=98%

3.3.2 Sample preparation

The extraction procedure was combined from methods discibed by Martínez-Carballo *et al.* (2007b) and Li & Brownawell (2009) and was designed to minimize analyte loss by avoiding unnecessary transfer of extracts to new glassware.

For effluent water a filled 500 ml sample bottle was weighed and the content transferred to a 1,000 ml separatory funnel. LAS (sodium dodecylbenzene sulfonic acid, tech. Aldrich) (0.4 mg in 1 ml chloroform) and internal strandard (EADAC-C12) was added. The sample bottle was rinsed with chloroform (50 ml) which was transferred to the separatory funnel. The empty sampling bottle was weighed and the extracted sample amount calculated. The separatory funnel was shaken and the contents allowed to separate. The aquous phase was emptied into the sampling bottle and the organic phase to a 50 ml glass tube. The extraction
was repeated twice and the organic phase successively evaporated to dryness using a stream of nitrogen.

A freeze dried sample of sludge or sediment, LAS (0.4 mg in 40 μ l MeOH) and internal standard (EADAC-C12) was extracted with methanol containing 8% concentrated hydrochloric acid in a glass tube in a microwave bath at approx. 60 °C for 30 min. The extraction was repeated twice. The extract, or for sludge a part of it, was evaporated to dryness in a glass tube.

Fish tissue was freeze dried and homogenized by stirring. A subsample (0.2 g eqvivalent to approx. 1 g fresh weight) was weiged into a teflon tube. Internal standard and acetonitrile containing 4% concentrated hydrochloric acid (freshly prepared) was added. After microwave assisted extraction (Ethos One, Milestone, Sorisole, Italy) for 15 min at 100 °C the extract was transferred to a glass tube and the solvent evaporated.

A common extract cleanup procedure was applied. The glass tube was extracted four times with water (4 ml) and the combined water extract finally extracted three times with chloroform (8 ml). The organic phases were transferred to a 10 ml glass tube and succesively evaporated to dryness. The residue was dissolved in methanol (1 ml).

3.3.3 Instrumental analysis

Liquid chromatography was performed on a Prominence UFLC system (Shimadzu) with two pumps LC-20AD, degasser DGU-20A5, autosampler SIL-20ACHT and column oven CTO-20AC. The analytical column was a Thermo HyPurity C8 50 mm x 3 mm, particle size 5 μ m (Dalco Chromtech). The column temperature was 35 °C and the flow rate 0.5 ml/min. The mobile phase A was 5 mM ammonium acetate in water, the mobile phase B was 0.1% formic acid in methanol. A gradient from 30% to 100% B was run. The run time was 12 min, followed by 4.1 min reequilibration of the column to initial conditions.

The effluent was directed to an API 4,000 triple quadrupole mass spectrometer (Applied Biosystems). Electrospray ionisation in positive MRM mode was used. Instrumental parameters are listed in Table 7.

	Q1	Q3	DP	CE	СХР
ATAC C12	228.4	60.1	87	52	10
ATAC C14	256.4	60.1	94	52	11
ATAC C16	284.3	60.1	100	52	12
ATAC C18	312.6	60.1	106	55	12
ATAC C20	340.6	60.1	108	65	12
ATAC C22	368.4	60.1	108	65	12
BAC C12	304.4	91.0	59	52	8
BAC C14	332.4	91.0	87	55	8
BAC C16	360.4	91.0	115	57	8
BAC C18	388.7	91.0	115	61	9
DDAC C10	326.4	186.3	110	40	12
DDAC C12	382.4	214.3	125	45	12
DDAC C14	438.7	242.3	146	49	12
DDAC C14:C16	466.5	242.3	146	52	12
DDAC C16	494.6	270.4	155	54	12
DDAC C16:C18	522.6	270.4	162	56	12
DDAC C18	550.6	298.4	170	59	12
EADAC-C12	242.0	74.0	120	52	12
EADAC-C16	299.0	74.0	120	52	12

Table 7: Precursor ion (Q1) product ion (Q3) and the instrumental parameters declustering potential (DP), collision energy (CE) and collision cell exit potential (CXP)

A five point standard curve was run were EADAC-C12 was used as an internal standard.

For DDAC C14:C16 the average response for DDAC C14 and DDAC 16 was used. For DDAC C16:C18 the average response for DDAC C16 and DDAC 18 was used (Li & Brownawell, 2009).

For ATAC-C20 and ATAC C22 no pure standard substance was available. Instead a solution of a hair care product containing behentrimonium chloride was used as a standard for the retention times, and the response for ATAC-C18 was used also for ATAC-C20 and ATAC C22 (Lara-Martín *et al.*, 2010).

3.3.4 Quality control

The efficiency for extraction from water was evaluated by making a fourth chloroform extract which was analysed separately. The increase in summed concentration of all analytes was 3.0% and 5.5% for two different effluents. This was judged not to necessitate a fourth extraction.

Examples of recoveries when real samples were spiked with low amounts (100 ng for effluent sediment and fish, 500 ng for sludge) of analytes are presented in Table 8. The results were adjusted according to the internal standard. Analytical results were not further adjusted according to these recoveries.

	Effluent	Sludge	Sediment	Fish
ATAC-C12	130%	121%	63%	95%
ATAC-C14	88%	72%	58%	92%
ATAC-C16	86%	47%	39%	79%
ATAC-C18	81%	55%	31%	78%
BAC-C12	146%	69%	92%	126%
BAC-C14	111%	55%	45%	89%
BAC-C16	72%	59%	29%	78%
BAC-C18	89%	65%	30%	74%
DDAC-C10	102%	61%	44%	90%
DDAC-C12	79%	62%	27%	69%
DDAC-C14	77%	59%	38%	69%
DDAC-C16	98%	47%	45%	68%
DDAC-C18	126%	31%	53%	58%

Table 8: Recoveries when real samples were spiked with low amounts of analytes

The coefficent of variation from duplicate or triplicate analysis of the same sample is shown in Table 9. In the rightmost column an average value (root mean square) for all samples are given. The average CV (root mean square) for all averages was 17%. An approximate 95% confidence interval for the analytical results could be obtained by multiplying the CVs with a "coverage factor" of 2. A reasonable estimate of the overall analytical uncertainty would be $\pm 40\%$ but one should bear in mind that a complete extraction of these very adsorptive substances are not always obtained in practice.

Table 9: Coefficent of variation from duplicate or triplicate analysis of the same sample

	SE-1-Eff	SE-3-Eff	SE-2-Eff	DK-6-Slu	FI-4-Slu	GL-9-Sed	NO-9-Sed	Average (RMS)
DDAC-C10	6.6%	1.0%	1.0%	13%	1.2%	18%	32%	15%
DDAC-C12			1.1%	18%	5.4%	2.8%		9.4%
DDAC-C14			13%	9.6%	26%	6.7%	12%	15%
DDAC-C14:16			0.8%	17%	23%	25%	5.0%	17%
DDAC-C16	14%		4.8%	3.2%	13%	14%	11%	11%
DDAC-C16:18	7.7%		2.2%	3.0%	10%	18%	12%	10%
DDAC-C18	12%		1.1%	3.9%	10%	15%	11%	10%
BAC-C12	2.1%	3.2%	2.4%	4.7%	0.8%	1.4%	12%	5.0%
BAC-C14	6.4%	16%	5.7%	9.3%	3.9%	10%	39%	17%
BAC-C16			0.4%	8.3%	15%	26%	69%	34%
BAC-C18	59%		1.8%	18%	1.8%	6.1%	44%	31%
ATAC-C12		4.1%	2.5%	20%	15%	4.4%		12%
ATAC-C14			8.8%	27%	19%	0.9%		17%
ATAC-C16	21%	2.8%	2.6%	8.1%	2.0%	4.7%		9.5%
ATAC-C18	32.8%		1.3%	10.3%	5.8%	10.1%	73.2%	33.4%
ATAC-C20	7.0%	2.6%	2.2%	8.5%	1.5%	4.7%		5.1%
ATAC-C22	14%	8.9%	1.5%	5.7%	3.1%	12%	7.6%	8.7%

LOQs were calculated from variation of laboratory extraction blanks (10 \times SD) for each sample type.

3.4 Analytical method, anions

500 mg graphitised carbon (Supelclean ENVI-Carb 120/200, Sigma) preheated to 400 °C for 4h was packed in SPE columns and washed with 35 ml 10 mM tetramethyl ammonium hydoxide in methanol. After washing with methanol and MilliQ water, sample was applied to the column together with the internal standard C8-LAS (4-octylbenzenesulfonic acid, sodium salt, Aldrich). The analytes were eluted with 7 ml 10 mM tetramethyl ammonium hydoxide in methanol. The extract was evaporated to dryness under a stream of nitrogen and the residues dissolved i methanol. Instrumental analysis was used according to Kaj *et al.* (2008) with the method adjusted to also include MRMs for SDS, SDSEO and CAPB.

4. Results

4.1 Measured concentrations, cations

All measured QAC concentrations are listed in Appendix 3. An overview of the detection frequencies (fraction of samples where a substance was found in concentrations above the limit of quantification, LOQ) is shown in Table 10.

This overveiw shows that the QACs frequently occurred both in the samples from WWTPs and in environmental samples. All analytes were detected in all sludge samples, in the majority of the sediments and (with one exception) in the majority of the effluents. Most analytes were also found in one or more of the fish samples (Table 10).

	Effluent (n=23)	Sludge (n=17)	Sediment (n=20)	Fish, muscle (n=19)	Fish, liver (n=3)
ATAC-C12	78%	100%	65%	0%	0%
ATAC-C14	83%	100%	95%	21%	0%
ATAC-C16	100%	100%	90%	53%	0%
ATAC-C18	96%	100%	100%	11%	33%
ATAC-C20	100%	100%	100%	11%	100%
ATAC-C22	100%	100%	90%	16%	100%
BAC-C12	74%	100%	85%	0%	0%
BAC-C14	65%	100%	85%	5%	0%
BAC-C16	83%	100%	100%	47%	33%
BAC-C18	87%	100%	95%	26%	67%
DDAC-C10	83%	100%	80%	16%	100%
DDAC-C12	70%	100%	80%	11%	0%
DDAC-C14	35%	100%	75%	5%	0%
DDAC-C14:16	74%	100%	90%	5%	33%
DDAC-C16	78%	100%	100%	5%	33%
DDAC-C16:18	91%	100%	100%	5%	33%
DDAC-C18	87%	100%	100%	5%	33%

Table 10: Detection frequencies (%) of the analytes in effluent, sludge, sediment, fish muscle and fish liver

A quick graphic overview of the results (Figure 10) reveals that some of the substances occured in higer concentrations than others; ATAC-C16, ATAC-C22, BAC-C12, BAC-14 and DDAC-C10 were dominating compounds in effluents and also in sludge. In sludges the relative importance of the long chained DDACs increased, and in sediments they often dominated.



Figure 10: Overview of all results for (from top to bottom) effluents, sludges and sediments. Each dot represents a single measurement

Below, the results are presented in more detail, by sample type.

4.1.1 WWTP effluent

In Table 11 detection frequency, minimum, median and maximum concentrations of the analytes in the 23 effluents samples are given. The red colour is proportional to the values in one column with the maximum value in that column shown with full colour intensity. The Table illustrates high detection frequencies and a great variation in the concentrations among the substances.

	Det freq	min	median	max
ATAC-C12	78%	<1	39	12,000
ATAC-C14	83%	<1	23	510
ATAC-C16	100%	1.3	290	4,500
ATAC-C18	96%	<1	42	730
ATAC-C20	100%	2.6	110	1,200
ATAC-C22	100%	15	510	6,800
BAC-C12	74%	<6.8	400	60,000
BAC-C14	65%	<6.6	67	17,000
BAC-C16	83%	<1.2	12	790
BAC-C18	87%	<3.5	26	760
DDAC-C10	83%	<8.4	59	15,000
DDAC-C12	70%	<1.5	2.6	29
DDAC-C14	35%	<1.9	<2.1	16
DDAC-C14:16	74%	<1	2.1	26
DDAC-C16	78%	<5.9	25	480
DDAC-C16:18	91%	<4	34	350
DDAC-C18	87%	<18	63	830

 Table 11: Detection frequency, minimum, median and maximum concentrations

 (ng/l) in effluents, n=23

The summed concentrations of ATACs, BACs and DDACs in the individual effluent samples are shown in Figure 11 and for samples with summed concentrations less than 5,000 ng/l also in Figure 12. The total concentrations varied between 64 ng/l (SE-3-Eff, Bollebygd WWTP) and 100,000 ng/l (GL-3-Eff, Sana Bay).

Effluents from Faroe Islands, Greenland and Iceland generally showed higher concentrations than effluents from Denmark (with the exception DK-3-Eff, Lynetten WWTP), Finland, Norway and Sweden. DK-3-Eff (Lynetten WWTP) was from a period when the waste water passed untreated through the treatment plant. In this effluent BACs dominated.



Figure 11: Summed concentrations of ATACs, BACs and DDACs in all effluents, ng/l



Figure 12: Summed concentrations of ATACs, BACs and DDACs in effluents with summed concentrations below 5 000 ng/l

The concentrations of the individual compound in the groups ATACs, BACs and DDACs are shown in Figure 13, Figure 14, and Figure 15 respectively. ATACs were dominated by ATAC-C16 and ATAC-C22. An exception was FO-3-Eff (Klaksviks Hospital) where ATAC-C12 dominated and showed a higher concentration than in any other effluent. BACs were dominated by the short chain compounds BAC-C12 and BAC-C14 which is in agreement with what was tabulated for commercial bensalkonium products (2.1). However, in IS-1-Eff (Ánanaust WWTP) the concentration of BAC-C18 exceeded that of BAC-C14 (Figure 14). A usual pattern for DDACs was that DDAC-C10 dominated, there was a minimum for DDAC-C12 or C14 and increasing concentration towards DDAC-C18 (Figure 15).





Figure 14: Concentrations of BACs in effluents, ng/l



Figure 15: Concentrations of DDACs in effluents, ng/l



4.1.2 WWTP sludge

In Table 12 detection frequency, minimum, median and maximum concentrations of the analytes in the 17 sludge samples are given.

Table 12: Detection frequency, minimum, median and maximum concentrations (ng/g dw) in sludge, n=17 $\,$

	Det freq	min	median	max
ATAC-C12	100%	99	1,300	40,000
ATAC-C14	100%	130	930	7,600
ATAC-C16	100%	1,400	13,000	26,000
ATAC-C18	100%	820	2,400	6,300
ATAC-C20	100%	1,600	5,000	13,000
ATAC-C22	100%	8,700	30,000	71,000
BAC-C12	100%	1,500	29,000	83,000
BAC-C14	100%	1,200	9,900	21,000
BAC-C16	100%	110	1,100	2,000
BAC-C18	100%	660	2,200	3,900
DDAC-C10	100%	1,600	12,000	50,000
DDAC-C12	100%	67	240	1,200
DDAC-C14	100%	20	93	300
DDAC-C14:16	100%	36	140	280
DDAC-C16	100%	270	1,300	3,000
DDAC-C16:18	100%	480	2,000	5,000
DDAC-C18	100%	1,900	5,900	18,000

The summed concentrations of ATACs, BACs and DDACs in the individual sludge samples are shown in Figure 16. The total concentrations varied between 42,000 ng/g dw (DK-6-Slu, Lynetten WWTP) and 230,000 ng/g dw (DK-5-Slu). In general the concentrations in sludge varied less than in effluents. ATACs occurred in the highest concentrations in nine cases, BACs in five and DDACs in three with no apperent correlation to the contry of origin.

No sludge samples from Greenland were included.



Figure 16: Summed concentrations of ATACs, BACs and DDACs in sludge, ng/g dw

The concentrations of the individual compound in the groups ATACs, BACs and DDACs are shown in Figure 17, Figure 18 and Figure 19, respectively.

ATACs were dominated by ATAC-C22 and ATAC-C16. The only exception was DK-5-Slu (Køge) where ATAC-C12 had the highest concentration. The dominating BACs were BAC-C12 and BAC-C14. BAC-C18 were almost always higher than BAC-C16.

For DDACs the common pattern was the same as was seen for effluents: DDAC-C10 dominated, there was a minimum for DDAC-C12 or C14 and increasing concentration towards DDAC-C18. In the Swedish samples and DK-6-Slu (Lynetten WWTP) the concentration of DDAC-C18 was higher than for DDAC-C10.

120000 100000 ATAC-C22 80000 ATAC-C20 60000 ATAC-C18 ATAC-C16 40000 ATAC-C14 20000 ATAC-C12 0 OKASIU OK-5-51U FLASIN 04-6-510 FI-5-SIU F1-6-51U NO.A.SIU N0.5.514 NO-6-514 F0-5-51 F0-6511 15-4-51 15-6-51 SEA.SH 55-6-514 15:5:51 stisi

Figure 17: Concentrations of ATACs in sludge, ng/g dw







Figure 19: Concentration of DDACs in sludge, ng/g dw

4.1.3 Sediment

In Table 13 detection frequency, minimum, median and maximum concentrations of the analytes in the 23 sediment samples are given. The Table shows high detection frequencies for QACs in sediments.

Table 13: Detection frequency, minimum, median and maximum concentrations (ng/g dw) in sediment, n=23

	Det freq	min	median	max
ATAC-C12	65%	<0.71	2	110
ATAC-C14	95%	<0.35	2.35	400
ATAC-C16	90%	<2	17	450
ATAC-C18	100%	0.8	10.1	380
ATAC-C20	100%	0.39	2.25	150
ATAC-C22	90%	<0.79	8.85	560
BAC-C12	85%	<8.2	26.5	1,200
BAC-C14	85%	<4.2	15	490
BAC-C16	100%	0.17	3.3	340
BAC-C18	95%	<0.25	4.95	480
DDAC-C10	80%	<2.7	9.4	340
DDAC-C12	80%	<0.2	0.875	540
DDAC-C14	75%	<0.28	1.05	120
DDAC-C14:16	90%	<0.055	4.75	290
DDAC-C16	100%	0.33	47	2,300
DDAC-C16:18	100%	0.25	85.5	5,600
DDAC-C18	100%	0.36	200	12,000

The summed concentrations of ATACs, BACs and DDACs in the individual sediment samples are shown in Figure 20.

The lowest total concentrations were measured in SE-9-Sed, Bollebygd WWTP, (<20 ng/g dw), DK-9-Sed, Nivå (63 ng/g dw) and GL-14-Sed, Narsaq Ilua, which is a background area (84 ng/g dw).

The highest summed concentrations were measured in SE-7-Sed, Henriksdal WWTP (25,000 ng/g dw), GL-9-Sed, Nuuk imeq (5,900 ng/g dw) and FO-7-Sed, Tórshavn harbour, ship yard (4,300 ng/g dw).

The dominating compound group was DDACs. These are also the most adsorptive compounds due to the two alkyl chains in the molecule.



Figure 20: Summed concentrations of ATACs, BACs and DDACs in sediment, ng/g dw

The concentrations of the individual compound in the groups ATACs, BACs and DDACs are shown in Figure 21, Figure 22 and Figure 23, respectively. ATACs were dominated by ATAC-C16, C18 and C22. FI-8-Sed (Tampere) was an exception where ATAC-C14 dominated. For BACs usually the short chain compounds dominated, but FO-7-Sed (Tórshavn harbour, ship yard) showed highest concentrations for BAC-C16 and BAC-C18. The usual pattern for DDACs was an increase from DDAC-C12 towards the dominating DDAC-C18. The relative concentration of DDAC-C10 varied. For FO-8-Sed (Tórshavn harbour, main hospital discharge) and FO-9-Sed (Klaksvik harbour) DDAC-C10 dominated.





Figure 22: Concentrations of BACs in sediment, ng/g dw



Figure 23: Concentrations of DDACs in sediment, ng/g dw



4.1.4 Fish

In Table 14 detection frequency, minimum, median and maximum concentrations of the analytes in the 19 fish muscle samples are given. With the exception of ATAC-16 and BAC-16 the Table shows rather low detection frequencies.

Table 14: Detection frequency, minimum, median and maximum concentrations (ng/g) in fish muscle. n=19

	Det freq	min	median	max
ATAC-C12	5%	<0.43	<0.56	0.44
ATAC-C14	21%	<0.29	<0.33	0.78
ATAC-C16	53%	<2.4	4.7	13
ATAC-C18	11%	<1.4	<1.8	4.7
ATAC-C20	11%	<0.32	<0.42	1.8
ATAC-C22	16%	<1.4	<1.8	30
BAC-C12	0%	<0.77	<1	<1.2
BAC-C14	5%	<0.59	<0.78	1.1
BAC-C16	47%	<0.62	<0.76	2.5
BAC-C18	26%	<0.61	<0.80	4.8
DDAC-C10	16%	<0.19	<0.24	1.3
DDAC-C12	11%	<0.15	<0.2	0.67
DDAC-C14	5%	<0.26	<0.35	0.95
DDAC-C14:16	5%	<0.14	<0.18	0.63
DDAC-C16	5%	<2.4	<3.1	5.6
DDAC-C16:18	5%	<3.0	<3.8	8.2
DDAC-C18	5%	<5.8	<7.5	13

The summed concentrations of measured ATACs, BACs and DDACs in the individual fish muscle samples are shown in Figure 24.



Figure 24: Summed concentrations of ATACs BACs and DDACs in fish muscle, ng/g

QACs were not detected in FO-13-Fis, IS-10-Fis and IS-11-Fis, all representing background conditions at the Faroe and Icelandic shelf respectively, FO-10-Fis (Tórshavn harbour) and GL-11-Fis (Vildmandsnæs). Highest concentrations were found in SE-11-Fis (Borås) and SE-10-Fis (Stockholm), 43 and 42 ng/g, respectively. ATACs dominated in most of the samples with one clear exception: SE-10-Fis (Stockholm), instead dominated by DDACs.

The concentrations of the individual compound in the groups ATACs, BACs and DDACs are shown in Figure 25, Figure 26 and Figure 27, respectively. It can be noticed that the BACs found tend to be long chained opposite to what was found in sludge and also in the commercial product.

Figure 25: Concentrations of ATACs in fish muscle ng/g



Figure 26: Concentrations of BACs in fish muscle ng/g



Figure 27: Concentrations of DDACs in fish muscle ng/g



Fish liver was not planned to be part of the investigation but three composite samples of cod liver (NO-10-Fis, NO-12-Fis, NO-13Fis) from the vicinity of VEAS WWTP, Oslo were included. In Table 15 detection frequency, minimum, median and maximum concentrations of the analytes in the three fish liver samples are given.

Table 15: Detection frequency, minimum, median and maximum concentrations (ng/g) in fish liver, n=3

	Det freq	min	median	max
ATAC-C12	0%	<3.2	<3.4	<3.5
ATAC-C14	0%	<1.9	<2.1	<2.1
ATAC-C16	0%	<14	<15	<16
ATAC-C18	33%	<11	<11	16
ATAC-C20	100%	11	23	160
ATAC-C22	100%	250	460	5,400
BAC-C12	0%	<5.7	<6.2	<6.3
BAC-C14	0%	<4.4	<4.8	<4.9
BAC-C16	33%	<3.9	<4.3	6.5
BAC-C18	67%	<4.9	12	12
DDAC-C10	100%	5.1	5.1	6.8
DDAC-C12	0%	<1.1	<1.2	<1.3
DDAC-C14	0%	<2.0	<2.1	<2.2
DDAC-C14:16	33%	<1.1	<1.1	8.9
DDAC-C16	33%	<20	<20	51
DDAC-C16:18	33%	<24	<24	59
DDAC-C18	33%	<47	<48	70

The summed concentrations of ATACs, BACs and DDACs in the individual fish liver samples are shown in Figure 28. The summed concentration varied from 290 to 5,800 ng/g. Compared to the maximum concentration found in fish muscle (43 ng/g) this was 7 to 130 times higher. The dominating compound class was ATACs.



Figure 28: Summed concentrations of ATACs BACs and DDACs in fish liver, ng/g

The concentrations of the individual compound in the groups ATACs, BACs and DDACs are shown in Figure 29, Figure 30 and Figure 31 respectively. ATAC-C22 dominated totally. Long chained BACs and DDACs were also found but in much lower concentrations.



Figure 29: Concentrations of ATACs in fish liver, ng/g

Figure 30: Concentrations of BACs in fish liver, ng/g



Figure 31: Concentrations of DDACs in fish liver, ng/g



4.2 Measured concentrations, anions

Effluents (19 samples) were analysed for LAS (linear alkylbenzene sulfonate), sodium dodecyl sulphate (SDS), sodium laureth sulphate (SDSEO) and cocamidopropyl betaine (CAPB). The results are shown for all samples in Figure 32 and for samples with summed concentration less than 120 μ g/l in Figure 33. Please note that the unit in these figures is μ g/l.

Figure 32: Concentration of LAS, SDS, SDSEO, CAPB in effluents, µg/l



Figure 33: Concentration of LAS, SDS, SDSEO, CAPB in effluents were the summed concentration was less than 120, $\mu g/l$



The over all concentration pattern of anions in effluents was similar to the one for QACs, with highest concentrations in samples from Greenland, Faeroe Islands and Iceland. As for QACs a realive increase in concentration was seen for the sample DK-3-Eff (Lynetten).

5. Discussion

5.1 Occurence of QACs

Many of the organic chemicals used in society will eventually accumulate in sludge from waste water treatment. All 17 substances from the three substance groups ATACs, BACs and DDACs included in this work were found in all sludge samples. This shows that the substances are used throughout the Nordic countries.

The actual concentrations in sludge varies due to differences in ease of degradation, adsorption efficiency and other parameters specific to each substance, but if the use pattern was clearly different in different countries, this would be reflected in the sludge. No such clear differences were observed.

The sampled sludge types were rather different: some samples were taken from the solid part of untreated sewage, but most were taken from the active sluge after anaerobic treatment. In spite of this, the summed QAC concentrations in the sludges were fairly uniform. The ratio of the highest to the lowest summed concentration was 5.5 on a dry weight basis.

The concentrations varied between effluents to a much larger extent than between sluges. The ratio of the highest to the lowest summed QAC concentrations in effluents was 1,600. High values were found in effluents from Faroe Islands, Greenland and Iceland and low values from Denmark, Finland, Norway and Sweden probably mostly reflecting differences in waste water treatment. High concentrations were in most cases associated with relatively small WWTPs so high concentrations are not always related to large emitted amounts.

According to the SPIN database, the use of QACs is dominated by DDACs (Chapter 2.2). This was not reflected in the concentration patterns in the effluents or sludges. Perhaps it would be if influents to WWTPs was analysed.

The effluent FO-3-Eff (Klaksviks Hospital) contained the highest concentrations of ATAC-C12, which were a minor component in all other effluents. This may reflect a special use in hospitals.

The QACs were found at varying concentrations in almost all sediments. The most adsorptive compound group, DDAC, dominated. Increased concentrations were seen in sediments also close to WWTPs where the effluent concentrations were low, illustrating the effect of accumulation zones, where hydrodynamic conditions are favourable for sedimentation. The most striking example is the related samples SE-1-Eff (Henriksdal WWTP), an effluent showing comparatively very low concentrations, and SE-7-Sed being by far the most polluted sediment. SE-7-Sed was from Stockholm inner archipelago close to the discharge point of Henriksdal WWTP.

The sediment SE-8-Sed from a medium sized river close to the discharge point of Gässlösa WWTP showed 58 times lower total QAC concentration than SE-7-Sed. These two sediments were in another project analysed for PAHs. The summed concentration of PAHs were 31 times higher in the sediment from Stockholm. Thus the QAC to PAH ratios in the two sediments were fairly equal. The concentration differences mainly reflects that the sediments represent more or less efficient accumulation zones.

High and also rapidly increasing concentrations of QACs in sediments downstream WWTPs have been reported in other studies (Martinez-Carballo *et al.*, 2007b; Li and Brownawell, 2010; Lara-Martin *et al.*, 2010). Our findings are in agreement with Martinez-Carballo, 2007b, who stated that the strong sorption properties of QACs may explain why these compounds are found in sediments downstream WWTPs in appreciable quantities and implied that QACs can be relatively persistent in receiving waters.

In Figure 34 the concentration profile for SE-7-Sed is compared to median results for sediments from the highly urbanized estuary Jamaica Bay, New York from 1998 (Table 4). The concentration profiles are very similar with highest concentrations for long chained DDACs.



Figure 34: Results of SE-7-Sed compared to results from Jamaica Bay, New York

Earlier studies have often focused on ATAC-C16 and ATAC-C18, which are active ingredients in the surfactant DTDMAC (Chapter 2.2), and also ATACs with shorter chain lengts (Table 4). Recently, behentrimonium, in which the main ingredients are ATAC-C20 and ATAC-C22, has been described as a new emerging contaminant (Lara-Martín *et al.*, 2010). ATAC-C20 and ATAC-C22 were in the present study found in all effluents

constituting up to 78% of the summed concentration of all measured QACs. In sludge, ATAC-C20 and ATAC C22 constituted 12 – 66 (median 24) percent of the summed concentration of all measured QACs. ATAC-C20 and ATAC-C22 were also found in almost all sediments but constituted only a few percent of the total QAC content.

In Figure 35 concentrations of ATAC-C16, ATAC-C18, ATAC-C20 and ATAC-C22 in SE-7-Sed is compared to results from Jamaica Bay, New York for the years 1998, 2003 and 2008 were the concentrations of ATAC-C20 and ATAC-C22 were seen to increase with time. In 2008 ATAC-C22 clearly dominated. In SE-7-Sed the relative dominance of ATAC-C22 was less profound.



Figure 35: Results of SE-7-Sed compared to results from Jamaica Bay, New York

5.2 Ecotoxicity, cations

The available ecotoxicity data is limited for the pelagic environment and even less data has been found for sediment dwelling organisms. Based on the ecotoxicity data found, ATAC and BAC are reported to be more toxic than DDAC, and Daphnia are reported to be the most sensitive species to ATAC and BAC.

A preliminary assessment of environmental risk for the aquatic phase was made based on the MEC/PNEC ratio (Measured Environmental Concentration / Predicted No Effect Concentration) for the maximum, the median and minimum MEC of ATAC and BAC (Table 16). The MEC values used were the concentrations measured in effluents divided by 10 to account for dilution of waste water by surface water flow (ECHA, 2008). For ATAC the lowest ecotoxicity value on acute tests (EC50) were used and an assessment factor of 1,000 based on the assumption that Daphnia is the most sensitive species (Garcia *et al.*, 2001; Sandbacka *et al.*, 2000; see 2.3). For BAC chronic data for Daphnia was used and an assessment factor of 10 (US EPA, 2006; see 2.3).

Parameter	ATAC	BAC	Reference
EC50	58 μ/l		Garcia <i>et al</i> . (2001)
NOAC		4.15 μg/l	US EPA (2006)
Assessment Factor	1,000	10	ECHA 2008
MEC median	0.091 μg/l	0.049 μg/l	This study
MEC max	1.4 μg/l	7.8 μg/l	This study
MEC min	0.0032 μg/l	0	This study
MEC/PNEC median	1.6	0.1	
MEC/PNEC max	24	19	
MEC/PNEC min	0.05	0	

Table 16: Values used for calculation of risk ratio for ATAC och BAC. MEC (n=23) is corrected for dilution by surface water by a factor of 10

For ATAC the MEC/PNEC risk ratios for the maximum and also the median MEC (24 and 1.6 respectively) were larger than unity which indicates a risk for the aquatic environment. For BAC this was the case for the maximum, but not for the median MEC (MEC/PNEC 19 and 0.1 respectively).

Individual effluents that (after 10 times dilution) gave MEC/PNEC ratios larger than one for summed concentrations of both ATACs and BACs were DK-3-Eff (Lynetten), FO-3-Eff (Klaksviks hospital), GL-3-Eff (Sana Bay), GL-4-Eff (Kolonihavnen) and IS-3-Eff (Hraunavik WWTP). Effluents with MEC/PNEC ratios larger than one for summed concentrations of ATACs only were the remaining effluents from FO, GL, IS, NO and SE-2-Eff (Gässlösa WWTP). (For ATAC a high assessment factor was used due to limited ecotoxicity data. If more data should be available a lower assessment factor could be used and it is possible that MEC/PNEC ratios would be lower).

There was not sufficient ecotoxicity data to make a risk assessment for DDAC but based on the available data the risk ratio can be assumed to be lower, based on the median MEC (230 ng/l), due to lower toxicity and lower median MEC and similar maximum MEC as for ATAC.

Based on the limited data and the available information on the circumstances of the tests the risk assessment should be regarded as preliminary. For instance, in risk assessment for QACs the concentration of the active ingredient should be taken into account as the solubility decreases with increasing alkyl chain length. For instance DDACs with chain lengths of C16-18 are practically insoluble in water (Table 2). Furthermore, environmental factors, such as the presence of clay mineral and humic acid in suspended sediment may reduce the toxicity of quaternary ammonium salts in the water column (van Wijk *et al.*, 2009).

For DDAC-C10 in sediment Thomas *et al.* (2009) reported a NOEC for *Caenohabditis elegans* (Nematoda) of 3,344 mg/kg for natural sediments. The highest concentration of DDAC-C10 measured in the present study equals 0.065 mg/kg (wet weight). This is more than four decades lower than the NOEC-value.

According to the criteria from REACH, ATACs with moderate chain lengths can be considered as potentially toxic. For long chained ATACs data is lacking while BAC meet the criteria to be classified as toxic. For DDAC available data suggest that DDAC is less toxic than ATAC and BAC (Chapter 2.3). A more in depth assessment of these substances regarding toxicity as well as persistence and bioaccumulation would be valuable in order to asses the the potential impact in accumulation zones. In that case also other surfactants such as LAS should be considered to be included as they show similar environmental behaviour.

6. Conclusions

In general there was a widespread occurence of QACs in all included matrices. Total concentrations and distribution among specific compounds varied to a large extent.

In effluents there was a clear difference in QAC-concentrations with high values from Faroe Islands, Greenland and Iceland on one hand and low values from Denmark, Finland, Norway and Sweden probably mostly reflecting differences in waste water treatment. A majority of the "high concentration" effluents showed, even after ten times dilution, concentrations of ATACs and/or BACs that could pose a risk to the aquatic environment. High effluent concentrations from a (Danish) WWTP that overflowed at the time of sampling also illustrates that effective treatment substantially reduces QAC levels in effluents.

Concentrations in sludge varied much less than in effluents. Patterns of individual compound concentrations were not clearly different among countries.

Behentrimonium (ATAC-C20 and ATAC-C22), recently described as a new emerging contaminant, was found in all effluents, sludges, almost all sediments and, as described below, in fish muscle and liver.

QACs were detected in most fish muscle samples. The summed concentration range was 1–43 ng/g, mostly dominated by ATACs and specifically ATAC-C16 or ATAC-C22. QACs were not detected in fish muscle from backround areas.

Three fish liver samles were analysed, all from Oslo fjord influenced by effluents from a large WWTP. The summed concentration range was 290–5,800 ng/g, totaly dominated by ATAC-C22.

Ecotoxicological data is lacking, especially for long chained ATACs. A more in depth assessment of these substances regarding toxicity as well as persistence and bioaccumulation would be valuable in order to asses the potential impact in accumulation zones.

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Sweden

Tomas Viktor, IVL, conducted the field sampling. Hendrik Braun, IVL, prepared the maps.

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9. Sammanfattning

Det övergripande syftet med denna screeningstudie var att undersöka förekomsten av kvartenära ammoniumföreningar i miljörelaterade prover från de nordiska länderna. Kvartenära ammoniumföreningar har stor användning i industriella tillämpningar och i hushållsprodukter såsom sköljmedel, detergenter, desinfektionsmedel, konserveringsmedel och ett stort antal produkter för kroppsvård. Katjoner av typerna alkyltrimetylammonium (ATAC), alkyldimetylbensylammonium (bensalkonium, BAC) och dialkyldimetylammonium (DDAC), totalt 17 olika, mättes i utgående vatten och slam från avloppsreningsverk, sediment och fisk. De deltagande länderna (Danmark, Färöarna, Island, Finland, Grönland, Norge och Sverige) bidrog vanligen med tre prov av varje provtyp. De relativt få proven och provtyperna avsåg att ge en ögonblicksbild av situationen.

De analyserade föreningarna var vanligt förekommande i alla provmatriserna.

Halter i utgående avloppsvatten varierade kraftigt. Många av proven höll koncentrationer som även efter tio gångers utspädning kan innebära en risk för vattenmiljön. Ekotoxikologiska data saknas speciellt för långkedjiga föreningar i gruppen ATAC.

Alla föreningar påvisades i alla slamprover. Koncentrationerna i slam varierade betydligt mindre än koncentrationerna i utgående vatten. Några tydliga skillnader i fördelningsmönstret mellan de olika föreningarna i prover från olika länder kunde inte ses.

Behentrimonium (ATAC-C20 och ATAC-C22) har nyligen uppmärksammats som en "emerging contaminant", en tidigare inte uppmärksammad miljöförorening. Den påvisades i en majoritet av proven, också i fiskmuskel och fisklever.

10. Appendices

10.1 Appendix 1, Sampling manual

Each country is asked to contribute:

- 3 WWTP effluent waters.
- 3 WWTP sludges.
- 3 sediments.
- 3 fish samples.

All samples will be analysed for quaternary ammonium compounds.

The 3 efffluent waters (2 only from FO and IS) will also be analysed for anions: SDS, SLS and cocoamidopropylbetaine.

Equipment provided

- 6 x 0.5 L glass bottles (2 x 0.5 L for each effluent water sample).
- 3 x 1 L plastic (pe) bottles (effluent water, anions).
- 3 x 250 ml plastic jars (sludge).
- 3 x 250 ml glass jars (sediment).
- Heat-treated Al-foil (small, extra for sediment jars).
- Heat-treated Al-foil (large, fish).
- Labels.
- Plastic gloves.
- Sampling protocol.

Planning the sampling

Start by opening the provided "Sampling protocol" in Excel format. Fill in data for each planned sample, one sample on each row. Each sample is given a Sample identification in the format *CO-no-Mat* where *CO* indicates country, *no* is a consecutive number and *Mat* indicates matrix. The samples should be labelled with their respective Sample identification. Example: DK-4-Slu, the fourth Danish sample which is a sludge sample. Use a printout of the sample list to record sample date and other relevant observations and type it in later. The sheet Notes can also be used.

Please give coordinates as LAT and LONG in the WGS84 reference system expressed as degrees and decimal minutes. See example in the sampling protocol. We strongly suggest to cross-check the registered coordinates with an independent geographic tool like Google Earth or similar. Select all six Excel cells containing the coordinates, copy the content with Ctrl-C and paste it into the "fly to" window in the upper-left corner of Google Earth. By clicking onto the magnifier symbol Google Earth should show you the correct sample position.

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 hair spray, or skin lotions on the day of sampling. Only the specially cleaned sampling containers provided by the laboratory should be used.

Blanks water will be stored at the laboratory. Transport blanks will be used during the sampling in Sweden, but will not be sent to the other participating countries.

A. Sampling of water

As the analytes are prone to adsorb to surfaces grab samples are preferred over composite samples. Two 0.5 L glass bottles should be filled at each site. The bottles contains a preservative (2 ml 6M H_3PO_4) and *should not be emptied*.

- Use the supplied gloves (or similar, nitrile is preferred).
- Open the lid carefully and fill the bottle once. Close the lid again. *Do not rinse*.
- Fill also one 1 L plastic bottle.
- Mark bottles with Sample identification.
- Make notes on the sample protocol.
- Store the glass bottles in a refrigerator and the plastic bottles in a freezer.

B. Sampling of sludge

- Put on the supplied gloves.
- Fill the jar with sludge and close the lid.
- Mark the sample with Sample identification.
- Put each sample in its plastic bag.
- Make notes on the sample protocol.
- Store the samples in a freezer.
C. Sampling of sediment

- Put on the supplied gloves.
- Open the lid of the jar.
- Fill sediment in the jar and close the lid. If the Al-foil protecting the lid of the jar is ruptured replace it with new Al-foil (supplied).
- Mark the sample with Sample identification.
- Put the sample in its plastic bag.
- Make notes on the sample protocol.
- Store the samples in a fridge. If the sample has low water content it can be stored in a freezer (risk of breaking the glass).

D. Sampling of fish

The number of fish per sample should be 5–10 or more depending on weight. The quantity should be sufficient to prepare a composite muscle sample of at least 20 g.

- Put on the supplied gloves.
- Wrap each fish individually in Al-foil provided.
- Put the Al-foil packages making up one sample in one common plastic bag, or, if needed, mark additional bags accordingly.
- Mark the samples with Sample identification.
- Make notes on the sample protocol.
- Store the samples in a freezer (-18°C).

Storage and transport

Send all samples to IVL, Stockholm (address, see below) in such a way that the samples will reach the laboratory preferably over night or as soon as possible (TNT or equivalent courier service). Send samples in the same containers used for providing sampling material. Make sure to insulate glass bottles properly to avoid breakage during transport. Include a printout of the Sampling protocol. Send the samples to the address below.

Address

IVL Swedish Environmental Research Institute, Lennart Kaj Valhallavägen 81, SE-114 27 Stockholm, Sweden. tel +46 (0)8-598 563 00

When sending the samples please send also an e-mail with the filled in Sample protocol attached to Lennart Kaj (lennart.kaj@ivl.se).

Thank you for your cooperation!

MR#	Sample ident.	Location	Site / Water area	Species etc	Sampling date	۲	T WGS84	2	DNG WGS84
						Deg,	decimal min	De	t, decimal min
2529	DK-1-Eff	Avedøre, Sjælland	Spildevandscenter Avedøre	Effluent, grab sample	203-09-18	55	36.51 N	12	26.85 E
2530	DK-2-Eff	Køge, Sjælland	Køgeegnens Renseanlæg	Effluent, grab sample	2013-09-18	55	29.307 N	12	11.423 E
2531	DK-3-Eff	København, Sjælland	Lynetten Renseanlæg	Effluent ¹ , grab sample	2013-09-18	55	41.711 N	12	36.889 E
2532	DK-4-Slu	Avedøre, Sjælland	Spildevandscenter Avedøre	Drained sludge	2013-09-18	55	36.51 N	12	26.85 E
2533	DK-5-Slu	Køge, Sjælland	Køgeegnens Renseanlæg	Centrifuged sludge	2013-09-18	55	29.307 N	12	11.423 E
2534	DK-6-Slu	København, Sjælland	Lynetten Renseanlæg	Centrifuged sludge	2013-09-18	55	45.711 N	12	36.889 E
2653	DK-7-Sed	Thors Sø	Randers Fjord		2013-10-31	56	7.5 N	6	31.59 E
2654	DK-8-Sed	Langerak	Limfjorden	Sand-silt sediment	2013-10-24	57	1.85 N	10	6.475 E
2683	DK-9-Sed	Nivå	The Sound		2013-10-10	55	54.937 N	12	32.477 E
2656	DK-10-Fis	Thors Sø	Randers Fjord	Perch, <i>Perca fluviatilis</i> , muscle	2013-09-11	56	7.5 N	6	31.59 E
2657	DK-11-Fis	Langerak	Limfjorden	Cod, <i>Gadus morhua</i> , muscle	2013-10-23	57	1.8 N	10	6.4 E
2684	DK-12-Fis	Nivå	The Sound	Flounder, muscle	2013-10-31	55	54.937 N	12	32.477 E
2621	FI-1-Eff	Turku	Kakolanmäki WWTP		2013-10-01	60	26.712 N	22	14.204 E
2622	FI-2-Eff	Tampere	Viinikanlahti WWTP		2013-10-02	61	29.319 N	23	46.043 E
2623	FI-3-Eff	Helsinki	Viikki WWTP		2013-10-03	60	13.544 N	24	59.612 E
2624	FI-4-Slu	Turku	Kakolanmäki WWTP		2013-10-01	60	26.712 N	22	14.204 E
2625	FI-5-Slu	Tampere	Viinikanlahti WWTP		2013-10-02	61	29.319 N	23	46.043 E
2626	FI-6-Slu	Helsinki	Viikki WWTP		2013-10-03	60	13.544 N	24	59.612 E
2627	FI-7-Sed	Turku	Turku Harbour		2013-10-01	60	26.371 N	22	13.103 E
2628	FI-8-Sed	Tampere	Viinikanlahti		2013-10-02	61	29.374 N	23	45.808 E
2629	FI-9-Sed	Helsinki	Vanhankaupunginlahti		2013-10-03	60	11.550 N	24	59.575 E
2833	FI-10-Fis	Turku	Airisto Seili	Perch <i>, Perca fluviatilis,</i> muscle	Nov-13	60	14.568 N	21	58.5448 E
2630	FI-11-Fis	Tampere	Pirkkalan Pyhäjärvi	Perch <i>, Perca fluviatilis,</i> muscle	Oct-13	61	28.220 N	23	38.683 E
2631	FI-12-Fis	Helsinki	Vanhankaupunginlahti	Perch, <i>Perca fluviatilis</i> , muscle	Sep-13	60	11.807 N	24	59.108 E
2658	FO-1-Eff	Sersjantvíkin STP	Effluent well		2013-11-04	62	0.495 N	9	45.714 W
2659	FO-2-Eff	Main Hospital STP	Effluent well		2013-11-01	62	0.6 N	9	46 W
2660	FO-3-Eff	Klaksvik Hospital	Manhole		2013-10-31	62	13.5 N	9	35.5 W
2661	FO-4-Eff	Main Hospital Laundry	Laundry, materiny and psyciatric ward	Infl	2013-11-01	62	0.6 N	9	46 W
2662	FO-5-Slu	Sersjantvíkin STP	The STP will be emptied later in nov. 2013		2013-11-04	62	0.495 N	9	45.714 W
2663	FO-6-Slu	Main Hospital STP	The STP has been emptied some months ago		2013-11-01	62	0.6 N	9	46 W
2664	FO-7-Sed	Tórshavn harbour	Ship yard (BÁ)		2013-11-04	62	0.43 N	9	46.439 W
2665	FO-8-Sed	Tórshavn harbour	Main Hospital (ÁL) effl discharge		2013-11-04	62	0.131 N	9	46.351 W
2666	FO-9-Sed	Klaksvik harbour	Á Stongum		2013-10-31	62	13.844 N	9	35.531 W
2885	FO-10-Fis	Tórshavn harbour		Cod, <i>Gadus morhua</i> , muscle	2014-01-05	62	0.334 N	9	46.183 W
2886	FO-11-Fis	Runavik		Cod, <i>Gadus morhua</i> , muscle	2013-11-23	62	6.739 N	9	43.641 W
2887	FO-12-Fis	Klaksvik harbour		Cod, <i>Gadus morhua</i> , muscle	2013-11-25	62	13.909 N	9	35.742 W
2668	FO-13-Fis	Faroe Shelf	Mýlingsgrunnur	Cod, <i>Gadus morhua</i> , muscle	2013-09-24	62	27 N	7	16 W
2667	FO-14-Sed	Skálafjord	SK05	upper two cm	2013-08-26	62	07.126 N	9	44.626 W
2669	GL-1-Eff	Quinngorput	sewage/Nuuk		2013-10-30	64	10.345 N	51	40.367 W
2670	GL-2-Eff	Nuuk imeq	sewage/Nuuk		2013-10-30	64	10.646 N	51	42.28 W

10.2 Appendix 2, Sample list

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						Deg	, decimal m	.Ľ	Ğ	, decimal n	i
2671	GL-3-Eff	Sana Bay	sewage/Nuuk		2013-10-30	64	10.026	z	51	44.642	≥
2672	GL-4-Eff	Kolonihavnen	sewage/Nuuk		2013-10-30	64	10.527	z	51	44.911	≥
2673	GL-7-Sed	Sana Bay	Nuuk		2013-10-29	64	9.9766	z	51	44.595	≥
2674	GL-8-Sed	Quinngorput	Nuuk		2013-10-29	64	10.276	z	51	40.654	≥
2675	GL-9-Sed	Nuuk imeq	Nuuk		2013-10-29	64	10.6583	z	51	42.2166	≥
2676	GL-10-Fis	Sana Bay	Nuuk	Cod, <i>Gadus morhua</i> , muscle	2013-10-27	64	9.9766	z	51	44.595	≥
2677	GL-11-Fis	Vildmandsnæs	Nuuk	Greenland cod, <i>Gadus ogac, ,</i> muscle	2013-10-27	64	10.2083	z	51	45.2066	≥
2678	GL-12-Fis	Nuuk imeq	Nuuk	Greenland cod, <i>Gadus ogac</i> , muscle	2013-10-27	64	10.6583	z	51	42.2166	≥
2679	GL-13-Fis	Nuuk imeq	Nuuk	Shorthorn sculpin, Myoxocephalus scorpius, muscle	2013-10-18	64	10.6583	z	51	42.2166	≥
2655	GL-14-Sed	Narsaq Ilua		Marine sediment	2010-09-09	60	55.7	z	46	3.6	≥
2692	IS-1-Eff	Reykjavik	Ánanaust WWTP		2013-11-07	64	9.184	z	21	49.38	≥
2690	IS-2-Eff	Reykjavik	Klettagarðar WWTP		2013-11-07	64	9.33	z	21	52.364	≥
2685	IS-3-Eff	Hafnarfjörð ur	Hraunavik WWTP		2013-11-04	64	3.026	z	22	0.667	≥
2693	IS-4-Slu	Reykjavik	Ánanaust WWTP		2013-11-07	64	9.184	z	21	49.38	≥
2691	IS-5-Slu	Reykjavik	Klettagarðar WWTP		2013-11-07	64	9.33	z	21	52.364	≥
2686	IS-6-Slu	Hafnarfjörð ur	Hraunavik WWTP		2013-11-04	64	3.026	z	22	0.667	≥
2694	IS-10-Fis	Icelandic shelf	50 km off N coast	Cod, <i>Gadus morhua</i> , muscle	2013-03-09	99	29.59	z	20	55.015	≥
2695	IS-11-Fis	Icelandic shelf	100 km off W coast	Cod, <i>Gadus morhua</i> , muscle	2013-03-12	64	58.72	z	26	12.49	≥
2564	NO-1-Eff	Oslo	VEAS WTW		2013-09-25	59	47.61	z	10	29.96	ш
2565	NO-2-Eff	Oslo	VEAS WTW		2013-09-26	59	47.61	z	10	29.96	ш
2566	NO-3-Eff	Oslo	VEAS WTW		2013-09-27	59	47.61	z	10	29.96	ш
2568	NO-4-Slu	Oslo	VEAS WTW		2013-09-25	59	47.61	z	10	29.96	ш
2569	NO-5-Slu	Oslo	VEAS WTW		2013-09-26	59	47.61	z	10	29.96	ш
2570	NO-6-Slu	Oslo	VEAS WTW		2013-09-27	59	47.61	z	10	29.96	ш
2571	NO-7-Sed	Indre Oslofjord	Oslofjord		2013-08-06	59	47.411	z	10	31.153	ш
2572	NO-8-Sed	Indre Oslofjord	Oslofjord		2013-08-06	59	47.291	z	10	31.149	ш
2573	NO-9-Sed	Indre Oslofjord	Oslofjord		2013-08-06	59	46.696	z	10	31.407	ш
2574	NO-10-Fis	Indre Oslofjord	Oslofjord	Cod, Gadus morhua, liver	2013-08-05	59	48.184	z	10	33.428	ш
2575	NO-11-Fis	Indre Oslofjord	Oslofjord	Cod, Gadus morhua, liver	2013-08-05	59	48.184	z	10	33.428	ш
2576	NO-12-Fis	Indre Oslofjord	Oslofjord	Cod, Gadus morhua, liver	2013-08-05	59	48.184	z	10	33.428	ш
2552	SE-1-Eff	Stockholm	Henriksdal WWTP		2013-10-07	59	18.585	z	18	6.458	ш
2496	SE-2-Eff	Borås	Gässlösa WWTP		2013-08-27	57	42.295	z	12	55.599	ш
2489	SE-3-Eff	Bollebygd	Bollebygd WWTP		2013-08-29	57	39.690	z	12	31.336	ш
2555	SE-4-Slu	Stockholm	Henriksdal WWTP		2013-10-08	59	18.585	z	18	6.458	ш
2497	SE-5-Slu	Borås	Gässlösa WWTP		2013-08-30	57	42.295	z	12	55.599	ш
2490	SE-6-Slu	Bollebygd	Bollebygd WWTP		2013-08-29	57	39.776	z	12	32.187	ш
2598	SE-7-Sed	Stockholm	Henriksdal WWTP		2013-09-25	59	19.086	z	18	7.022	ш
2502	SE-8-Sed	Borås	Gässlösa WWTP		2013-08-28	57	41.510	z	12	54.532	ш
2491	SE-9-Sed	Bollebygd	Bollebygd WWTP		2013-08-27	57	39.706	z	12	32.082	ш
2778	SE-10-Fis	Stockholm	Henriksdal WWTP	Perch, <i>Perca fluviatilis</i> , muscle	2013-09-25	59	18.993	z	18	7.189	ш
2506	SE-11-Fis	Borås	Gässlösa WWTP	Perch, <i>Perca fluviatilis</i> , muscle	2013-08-29	57	41.510	z	12	54.532	ш
2492	SE-12-Fis	Bollehvad	Rollehvød W/M/TP	Perch. Percn fluvintilis . muscle		57	30 706	N	;	32 087	ш

Sample ident. suffix: Eff=Effluent, Inf=Influent, Slu=Sludge, Sed=sediment, Fis=Fish. ¹: Untreated effluent due to overflow after stormwater.

MR	Sample	DW,	Unit	ATAC-	ATAC-	ATAC-	ATAC-	ATAC-	ATAC-	BAC-	BAC-	BAC-	BAC- D	DAC- D	DAC D	DAC- L	DDAC- [DDAC- I	DDAC- D	DAC-	Sum	Sum	Sum	Sum
	ident.	%		C12	C14	C16	C18	C20	C22	C12	C14	C16	C18	C10	-C12	C14 C1	4:16	C16 C	16:18	C18 /	VTACs	BACs I	DACs	
2529	DK-1-Eff		ng/l	4	1.8	14.0	6.2	7.4	38	<6.8	<6.5	<1.2	<3.4	<8.3	<1.4	<2.1	4	<5.8	4	<18	67			67
2530	DK-2-Eff		ng/l	2.5	2.2	5.9	8.0	2.6	15	<6.4	<6.1	<1.1	6.2	8.0	<1.3	<1.9	4	12	12	37	36	6.2	69	110
2531	DK-3-Eff		ng/l	89	92	2,100	280	600	3,800	11,000	2,200	140	280	1,600	13	16	26	260	350	830	7,000 1	3,000	3,100	23,000
2532	DK-4-Slu	23%	ng/g dw	860	810	12,000	2,000	3,100	22,000	64,000	17,000	1,700	3,900	9,100	120	72	140	1,600	2,800	3,000 4	1,000 8	36,000	22,000 1	50,000
2533	DK-5-Slu	26%	ng/g dw	40,000	7,600	14,000	4,400	2,900	25,000	83,000	21,000	1,100	970 1.	2,000	74	56	140	1,900	3,100	3,200 9	4,000 1	10,000	26,000 2	30,000
2534	DK-6-Slu	23%	ng/g dw	160	130	1,400	820	2,100	16,000	6,300	2,100	300	006	2,200	67	50	130	1,300	2,100	5,000 2	1,000	6,600	12,000	42,000
2653	DK-7-Sed	%9	ng/g dw	<0.71	1.8	8.8	6.2	1.4	2.0	74	15	0.99	1.3	<2.7	0.22	0.53	1.9	14	27	99	20	91	110	220
2654	DK-8-Sed	73%	ng/g dw	<0.28	2.7	17	17	0.8	0.76	4.8	2.5	2.9	3.8	1.2	<0.2	0.64	2.9	29	55	130	38	14	220	270
2683	DK-9-Sed	71%	ng/g dw	<0.31	0.42	\$	3.0	0.39	<0.77	<4.3	<2.2	0.55	0.55	<1.2	<0.2	<0.15	0.8	8.2	15	34	3.8	1.1	58	63
2656	DK-10-Fis		ng/g	<0.56	<0.33	5.1	<1.9	<0.42	<1.8	4	<0.78	2.4	1.4	<0.24	<0.2	<0.35	<0.18	<3.2	<3.9	<7.6	5.1	3.8		8.9
2657	DK-11-Fis		ng/g	<0.6	<0.36	<2.7	\$	<0.46	<1.9	<1.1	<0.84	2.5	<0.86	<0.26 <	0.22	<0.37	<0.2	<3.4	<4.2	<8.2		2.5		2.5
2684	DK-12-Fis		ng/g	<0.5	<0.35	<2.6	<1.9	0.71	4.2	4	<0.81	1.1	<0.83	<0.25 <	0.21	<0.36	<0.19	<3.3	4	<7.8	4.9	1.1		6.0
2621	FI-1-Eff		ng/l	1.2	1.3	16	9.2	18	93.0	<6.4	<6.1	1.8	4.5	8.9	1.5	\$	4	<5.4	4.6	<17	140	9	15	160
2622	FI-2-Eff		ng/l	1.6	2.8	130	40	58	260.0	56.0	26.0	5.0	13.0	81.0	3.0	∽	1.5	14	25	62	490	100	190	780
2623	FI-3-Eff		ng/l	5.3	3.5	19	12	20	100.0	400.0	67.0	15.0	13.0	9.8	<1.2	<1.9	4	<5.3	6.8	17.0	160	490	34	069
2624	FI-4-Slu	23%	ng/g dw	1,300	730	18,000	2,400	5,000	30,000	31,000	9,500	970	1,900 1.	5,000 1	,200	190	150	1,100	2,000	5,100 5	7,000 4	13,000	24,000 1	20,000
2625	FI-5-Slu	33%	ng/g dw	1,400	1,100	21,000	3,300	5,800	30,000	44,000	12,000	1,100	2,200 1	6,000	660	300	280	1,500	2,200	5,200 6	3,000 5	0000'69	28,000 1	50,000
2626	FI-6-Slu	30%	ng/g dw	1,700	066	16,000	2,900	6,200	32,000	59,000	14,000	1,300	2,300 1.	2,000	240	93	170	1,800	3,000	7,300 6	0,000 7	6,000	25,000 1	60,000
2627	FI-7-Sed	27%	ng/g dw	14	4.4	55	18	11	50	210	74	18	15	69	9	1.6	4.3	60	110	190	150	320	440	910
2628	FI-8-Sed	38%	ng/g dw	110	400	230	48	7.7	23	370	140	31	26	7.8	18	5.4	9.2	110	200	370	820	560	720	2,100
2629	FI-9-Sed	33%	ng/g dw	3.9	1.9	7.3	7.6	2.3	8.2	99	18	4.6	8.9	11	2.3	1.1	5.2	42	76	210	31	98	340	470
2833	FI-10-Fis		ng/g	<0.55	<0.33	6.3	<1.8	<0.42	<1.7	<0.99	<0.76	<0.67	<0.79	<0.24	<0.2	<0.34	<0.18	<3.1	<3.8	<7.4	6.3			6.3
2630	FI-11-Fis		ng/g	<0.51	0.78	13	<1.7	<0.38	<1.6	<0.91	<0.71	<0.62	<0.73	<0.22 <	0.18	<0.32	<0.16	<2.9	<3.5	<6.9	14			14
2631	FI-12-Fis		ng/g	<0.43	0.51	10	<1.4	<0.32	<1.4	<0.77	<0.59	0.72	<0.61	<0.19 <	0.15	<0.26	<0.14	<2.4	ų	<5.8	11	0.7		11
2658	FO-1-Eff		ng/l	39	23	800	180	460	2,500	200	57	6.2	11	57	2.2	<2.7	2.2	25	40	110	4,000	280	230	4,500
2659	FO-2-Eff		ng/l	65	75	290	17	37	370	096	180	12	14	250	1.5	<1.9	1.3	30	34	58	860	1,200	380	2,400
2660	FO-3-Eff		ng/l	12,000	510	1,300	540	20	220	18,000	3,500	84	26	1,300	4.8	5.3	4.2	59	99	91 1	4,000 2	2,000	1,500	38,000
2661	FO-4-Eff		ng/l	200	92	350	20.0	22	230	1,200	200	16	36	230	<1.5	<2.3	2.1	52	42	63	910	1,500	390	2,800
2662	FO-5-Slu	21%	ng/g dw	140	170	4,100	1,700	3,500	20,000	19,000	7,400	450	1,200 5	000′0	110	210	110	930	1,400	3,500 3	0,000 2	8,000	56,000 1	10,000
2663	FO-6-Slu	10%	ng/g dw	1,300	1,000	5,500	1,400	1,600	9,300	20,000	6,300	2,000	3,200	5,100	67	06	69	720	1,100	2,200 2	0,000 E	1,000	9,400	61,000
2664	FO-7-Sed	65%	ng/g dw	8.5	11	82	160	4.0	14	170	92	340	480	280	2.3	4.3	34	270	590	1,800	280	1,100	3,000	4,300
2665	FO-8-Sed	%69	ng/g dw	2.4	1.7	16	2	1.4	8.7	370	130	3.7	4.5	33	0.75	<0.1	<0.05	0.33	0.25	1.0	33	510	35	580
2666	FO-9-Sed	66%	ng/g dw	3.5	1.8	17	3.7	1.1	ŝ	330	75	5.4	8.5	220	0.51	<0.15	0.23	0.55	0.32	0.36	30	423	380	830
2885	FO-10-Fis		g/gu	<0.61	<0.36	<2.7	\$	<0.46	<1.9	<1.1	<0.85	<0.74	<0.87	<0.27 <	0.22	<0.38	<0.2	<3.4	<4.2	<8.2				
2886	FO-11-Fis		ng/g	<0.62	<0.37	5.6	4.7	<0.47	\$	<1.1	<0.87	1.8	4.8	1.3	0.67	0.95	<0.2	<3.5	<4.3	<8.4	10	4.8	2.9	18
2887	FO-12-Fis		g/gu	<0.65	<0.39	<2.9	<2.1	<0.49	<2.1	<1.2	<0.9	<0.79	<0.93	1 ~	0.23	<0.4	<0.21	<3.7	<4.5	<8.8			1.0	1.0
2668	FO-13-Fis		ng/g	<0.56	<0.33	<2.5	<1.8	<0.42	<1.8	4	<0.77	<0.68	<0.8	<0.24	<0.2	<0.35	<0.18	<3.1	<3.8	<7.5				
2667	FO-14-Sed	43%	ng/g dw	<0.52	2.4	20	6.3	2.9	7.6	7.8	5.7	2.4	3.2	19	0.34	0.68	0.7	10	26	85	40	19	140	200
2669	GL-1-Eff		ng/l	150	34	390	240	1,100	6,800	1,300	280	13	15	59	<1.3	<2.1	12	480	330	350	8,700	1,600	1,200	12,000
2670	GL-2-Eff		ng/l	130	49	1,300	81	150	1,600	3,400	390	21	26	150	2.4	6.5	26	240	250	400	3,300	3,900	1,100	8,300
2671	GL-3-Eff		ng/l	110	120	2,900	290	350	1,800	60,000	17,000	790	290 1	5,000	3.0	5.0	7.3	150	250	780	5,500 7	8,000	16,000 1	000'00
2672	GL-4-Eff		ng/l	200	220	4,500	730	1,200	6,700	13,000	9,200	220	180	1,700	2.6	2.9	16	190	250	480 1	4,000 2	3,000	2,600	39,000
2673	GL-7-Sed	79%	ng/g dw	0.52	1.4	13	7.2	1.3	5.4	ß	£	0.83	1.4	1.6	0.26	0.16	1.8	15	21	45	28	10	85	120

10.3 Appendix 3, Individual results, cations

C14 C14:161 C16 C1 2.2 51 520 1 9.2 51 520 1 9.2 51 520 1 6.33 6.117 53.1 520 1 6.33 6.017 $6.3.1$ $6.3.3$ 6.117 6.3 6.33 6.016 7.0 93.0 1 2.8 8.1 5.8 16.0 $12.0.0$ 23.3 6.11 6.2 8.1 3.1 11.0 74.0 120.0 23.3 6.70 93.0 12 2.0 3.1 11.0 74.0 120.0 23.3 60.35 60.19 63.3 270 93.3 20.2 21.1 11.00 11.1 7.3 9.7 9.1100 11.2 11.2 11.2 11.2 22.5 2.0 1.2 1.2 1.2 2.12	14 Clatic Cla Clatic Cla Clatic Cla Clatic Cla Clatic Cla Clatic Cla Clatic Clat <thclat< th=""> Clat</thclat<>	1:4:16 C.16 C.16 C.16 C.16 C.17 5:1 5:2 1 1 1 1 1 1 <th1< th=""> 1 <th1< th=""> <</th1<></th1<>	C14:161 C16 C1 51 520 1 51 520 1 52 1 52 1 51 520 3 <0.17 <3.1 <0.17 <3.1 <0.17 <3.1 <0.17 <3.1 <0.16 <3.3 <0.11 <3.1 <0.16 <3.16 $0.23.0$ $3.11.00$ $3.10.0$ $3.10.0 3.11.000 3.11.000 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 3.10.0 $	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	C14:16 C16 C1 C3 51 520 1 520 1 51 520 3.1 520 1 60.17 <3.1 <0.17 <3.1 <0.13 <0.13 <0.13 <0.13 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12 <0.12	CI4:16 C16	Ci6 Ci 52 3.1 52 3.1 52 3.1 52 3.1 53 3.1 53 53 19 53 58 11 1100 11,100 58 11 120.0 27 58 19 58 11 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 11,100 58 58 58 58 58 58 58 58 58 58 58 58 58	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	⁰ 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	$\forall \forall \forall \forall \forall \bullet \bullet$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\nabla \nabla $	$\begin{array}{c} \Diamond \\ \Diamond \\ \downarrow \\$	$\begin{array}{c} 4 \\ 4 \\ 4 \\ 6 \\ 6 \\ 6 \\ 7 \\ 7 \\ 7 \\ 7 \\ 7 \\ 7 \\ 7$	$\begin{array}{c} 111 \\ 112 \\ 111.0 \\ 111$	
0.34 0.34 0.19 0.19 0.19 0.29 0.21 1.10 0.21 1.70 0.21 1.70 0.21 1.70 0.21 0.21 0.21 0.21 0.2 0.2 0.2 0.2 0.2 0.2 0.2 0.2	0.34 0.34 0.19 0.19 0.19 0.19 0.19 0.19 0.20 0.2 0.2 0.2 0.2 0.2 0.2 0.	 L4 C) 24 C) 24 C) 19 C) 19 C) 13 C) 19 C) 29 C) 23 C) 29 C) 28 C) 29 C) 20 C) 29 C) 20 <li< th=""><th>0.14 0.14 0.14 0.19 0.13 0.33 0.19 0.33 0.31 0.19 0.33 0.33 0.19 0.33 0.33 0.19 0.33 0.31 0.10 5.8 0.31 170 44 2.5 170 73 2.15 170 73 2.3 60.2 0.35 0.35 60.2 0.35 2.10 550 100 550 100 550 100 5.2 2.1 7.4 7.5 1.5 4.1 7.4 7.5 1.10 5.8 650 100 5.2 2.1 5.8 4.1 2.1 2.1 5.8 4.1 2.1 2.1 5.8 4.19 2.1 2.1 5.8 4.19 2.1 2.1 5.8 4.19 2.1 2.1</th><th>$\begin{array}{cccccccccccccccccccccccccccccccccccc$</th><th>0.34 0.34 0.34 0.19 0.33 0.33 0.19 0.33 0.31 4.5 2.5 0.33 4.5 2.5 0.33 170 4.4 2.5 170 2.9 7.3 170 2.9 7.3 290 7.3 2.6 290 7.3 2.6 81.0 2.9 7.3 9.5 9.1 0.3 9.5 9.1 0.3 550 100 650 1.5 2.1 2.1 7.4 7.5 2.1 7.4 7.5 2.1 7.4 7.5 2.1 7.4 7.5 4.1 7.4 7.5 2.1 6.13 4.1 4.1 7.4 7.5 4.1 7.3 4.1 4.1 7.4 7.3 4.1 7.5 4.1 4.1 7.1 4.1 4.1 7.1 4.1 4.1 7.3 4.1 4.1 7.4 7.2 4.1 7.5 4.1 4.1 7.6 7.0 7.</th><th>$\begin{array}{cccccccccccccccccccccccccccccccccccc$</th><th>$\begin{array}{cccccccccccccccccccccccccccccccccccc$</th></li<>	0.14 0.14 0.14 0.19 0.13 0.33 0.19 0.33 0.31 0.19 0.33 0.33 0.19 0.33 0.33 0.19 0.33 0.31 0.10 5.8 0.31 170 44 2.5 170 73 2.15 170 73 2.3 60.2 0.35 0.35 60.2 0.35 2.10 550 100 550 100 550 100 5.2 2.1 7.4 7.5 1.5 4.1 7.4 7.5 1.10 5.8 650 100 5.2 2.1 5.8 4.1 2.1 2.1 5.8 4.1 2.1 2.1 5.8 4.19 2.1 2.1 5.8 4.19 2.1 2.1 5.8 4.19 2.1 2.1	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0.34 0.34 0.34 0.19 0.33 0.33 0.19 0.33 0.31 4.5 2.5 0.33 4.5 2.5 0.33 170 4.4 2.5 170 2.9 7.3 170 2.9 7.3 290 7.3 2.6 290 7.3 2.6 81.0 2.9 7.3 9.5 9.1 0.3 9.5 9.1 0.3 550 100 650 1.5 2.1 2.1 7.4 7.5 2.1 7.4 7.5 2.1 7.4 7.5 2.1 7.4 7.5 4.1 7.4 7.5 2.1 6.13 4.1 4.1 7.4 7.5 4.1 7.3 4.1 4.1 7.4 7.3 4.1 7.5 4.1 4.1 7.1 4.1 4.1 7.1 4.1 4.1 7.3 4.1 4.1 7.4 7.2 4.1 7.5 4.1 4.1 7.6 7.0 7.	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{ccccccc} & 0.24 & 0.19 & 0.27 \\ & 0.77 & 0.24 & 0.19 & 0.19 & 0.17 \\ & 0.72 & 0.22 & 0.18 & 0.19 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.12 & 0.110 & 25,000 & 110 & 27 & 0.12 & 0.23 & 0.25 & 0.02 & 0.23 & 0.25 & 0.21 & 0.33 & 0.25 & 0.21 & 0.21 & 0.22 & 0.22 & 0.21 & 0.22$	$\begin{array}{ccccccc} & 0.27 & 0.27 & 0.27 \\ < 0.77 & 0.24 & 0.19 & 6. \\ < 0.72 & 0.22 & 0.18 & 6. \\ < 0.72 & 0.22 & 0.18 & 6. \\ 760.0 & 8,900.0 & 29.0 & 390.0 & 1,600.0 & 4.5 & 0.12 & 6. \\ 390.0 & 1,600.0 & 1,00 & 290 & 87 & 2,200 & 1,100 & 25,000 & 110 & 25,000 & 110 & 25,000 & 110 & 23,000 & 1,000 & 2900 & 290 & 3,400 & 1,000 & 550 & 3,400 & 1,000 & 5,40 & 1,000 & 5,40 & 1,000 & 5,40 & 1,000 & 5,40 & 1,000 & 5,40$	$\begin{array}{ccccccc} & 0.27 & 0.27 & 0.27 \\ < 0.77 & 0.24 & 0.19 & 6. \\ < 0.72 & 0.22 & 0.18 & 6. \\ < 0.72 & 0.22 & 0.18 & 6. \\ 390.0 & 1,600.0 & 37 & 0.2 \\ 2,100 & 5,5000 & 87 & 0.2 \\ 2,100 & 25,000 & 87 & 0.2 \\ 2,200 & 11.0 & 25 & 0.2 & 6. \\ 3,000 & 19,000 & 290 & 37 & 6. \\ 3,000 & 19,000 & 290 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 33 & 0.25 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 32 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 32 & 0.21 & 6. \\ 3,000 & 11,000 & 550 & 5. & 6. & 7.4 & 4.5 & 1.5 \\ 2,1 & 4,2 & 1,2 & 6. & 7.4 & 6. \\ 3,2 & 6,9 & 7,4 & 6. & 1.1 & 6. & 6. \\ 3,40 & 1,000 & 550 & 290 & 1.70 & 600 & 1.70 & 540 & 5.0 \\ 1,300 & 9,300 & 7,500 & 290 & 1.70 & 660 & 1,600 & 1.70 & 540 & 5.0 \\ 1,600 & 1,700 & 520 & 340 & 5$	$\begin{array}{ccccccc} & 0.75 & 0.0.7 & 0.0.7 \\ & 0.75 & 0.42 & 0.19 & 0.1 \\ & 0.77 & 0.22 & 0.18 & 0.1 \\ & 0.72 & 0.22 & 0.13 & 0.1 \\ & 760.0 & 8,900.0 & 29.0 & 0.1 \\ & 740.0 & 6,500.0 & 11.0 & 2,1 \\ & 2,100 & 2,000 & 170 & 0.2 \\ & 2,100 & 2,000 & 170 & 0.2 \\ & 3,700 & 19,000 & 290 & 0.1 \\ & 3,700 & 19,000 & 290 & 0.2 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 3,700 & 19,000 & 290 & 0.1 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 0.83 & 0.25 & 0.21 & 0.2 \\ & 0.83 & 0.25 & 0.21 & 0.2 \\ & 0.83 & 0.25 & 0.21 & 0.2 \\ & 0.83 & 0.25 & 0.21 & 0.2 \\ & 0.8 & 0.11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 11,000 & 550 \\ & 3,400 & 12,000 & 0.170 \\ & 4,5 & 3,1 & 1.0 \\ & 4,2 & 3,1 & 1.0 \\ & 4,2 & 3,1 & 1.0 \\ & 4,2 & 3,1 & 1.0 \\ & 4,2 & 3,1 & 1.0 \\ & 4,1 & 4,2 \\ & 4,1 & 4,2 \\ & 4,1 & 4,2 \\ & 4,1 & 4,2 \\ & 5,8 & 4,1 & 4,1 \\ & 4,1 & 4,2 \\ & 5,8 & 4,1 & 4,1 \\ & 4,1 & 4,2 \\ & 5,1 & 4,2 \\ & 5,1 & 4,2 \\ & 5,1 & 4,1 & 4,2 \\ & 5,1 &$	$\begin{array}{cccccccc} & 0.25 & 0.27 & 0.27 \\ & 0.75 & 0.42 & 0.19 & 0.19 \\ & 0.72 & 0.22 & 0.18 & 0.12 \\ & 760.0 & 8,900.0 & 29.0 & 37 \\ & 760.0 & 8,900.0 & 29.0 & 37 \\ & 760.0 & 8,900.0 & 29.0 & 37 \\ & 740.0 & 6,500.0 & 11.0 & 29.0 \\ & 2,100 & 2,5000 & 11.0 & 29.0 \\ & 3,700 & 19,000 & 2900 & 290 \\ & 0.83 & 0.25 & 0.21 & 0.1 \\ & 2,200 & 2,5000 & 11.0 & 290 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 3,80 \\ & 3,400 & 11,000 & 560 & 170 \\ & 3,1 & 1.0 & 290 \\ & 4,2 & 3,1 & 1.0 \\ & $	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{ccccccc} & 0.25 & 0.02 & 0.03 & 0.03 & 0.03 & 0.03 & 0.01 & 0.02 & 0.13 & 0.13 & 0.12 & 0$	
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0.00 0.38 0.93 0.040 150.0 156 2000 350 040 150.0 82 940 330 240 240 150.0 82 940 230 220 $1/700$ 360 550 $8,100$ $1,900$ $2,200$ $2,200$ 380 390 $9,900$ $2,300$ $2,200$ $2,200$ 380 0.33 2.5 4.1 $2,200$ $2,200$ 41 11 11 11 11 110 1100 41 11 11 11 110 1100 1100 $1,400$ $1,100$ $23,000$ $5,000$ $10,000$ 1100 $1,400$ $1,100$ $23,000$ $5,000$ $10,000$ 1100 $1,400$ $1,100$ $23,000$ $5,000$ $10,000$ 1100 $1,400$ $1,100$ $23,000$ $5,000$	$\sigma_{0.0}$ 0.38 9.3 0.40 150.0 15 2000 350 640 150.0 82 940 330 220 240 160 160 3300 920 $1/700$ 240 360 550 $8,100$ $1,900$ $2,200$ 2900 380 390 $9,900$ $2,300$ $2,200$ $2,400$ 41 41 $1,900$ $2,200$ $2,419$ $2,440$ 41 41 110 $21,000$ $2,900$ $20,44$ 41 41 112 34 110 110 41 11 12 34 110 110 $1,400$ $1,100$ $23,000$ $5,200$ $10,000$ 110 $1,400$ $1,100$ $23,000$ $5,200$ $10,000$ 110 $1,400$ $1,100$ $23,000$ $5,200$ $10,000$ 110 1	$\sigma_{0.0}$ 0.38 9.9 0 0.40 150.0 35 2,000 350 640 150.0 82 940 230 280 160 160 3,300 920 1,700 360 550 8,100 1,900 2,200 380 390 9,900 2,300 2,200 0.55 8,100 1,900 2,200 0.55 8,100 1,900 2,200 0.55 0.33 2.5 $6.1.9$ $6.0.42$ 0.55 0.33 2.5 $6.1.9$ $6.0.42$ 0.140 1.10 $23,000$ $5,000$ $10,000$ $1,400$ $1,100$ $23,000$ $5,200$ $10,000$ $1,600$ $1,100$ $23,000$ $5,200$ $10,000$ $1,600$ $1,100$ $23,000$ $5,200$ $10,000$ $1,600$ $1,100$ $23,000$ $5,200$ $10,000$ $1,800$ </th <th>$\alpha_{0.0}$ 0.38 9.3 0.40 150.0 35 $5,000$ 350 640 150.0 82 940 230 280 160 160 3,300 920 1,700 360 550 8,100 1,900 2,200 360 550 8,100 1,900 2,200 380 390 9900 2,300 2,200 α_{11} α_{11} α_{11} α_{12} α_{14} α_{11} α_{11} α_{11} α_{12} α_{14} α_{110} α_{11} α_{11} α_{11} α_{12} α_{14} α_{110} α_{11} α_{11} α_{11} α_{11} α_{11} α_{11} α_{11} α_{11} α_{12} α_{11} α_{12} α_{12} α_{11} α_{11} α_{11} α_{12} α_{12} α_{12} α_{11} α_{11} α_{12} α_{12} α</th> <th>$\alpha_{0.0}$ 0.38 9.9 0.40 150.0 35 500 350 640 150.0 81 3300 920 1,700 360 550 8,100 1,900 2,200 360 550 8,100 1,900 2,200 380 390 9,900 2,300 2,900 41 41 16 3,300 2,900 41 41 16 42 110 41 41 12 3,4 110 41 41 12 3,4 110 41 11 2,3 41 14 41 11 2,3 11 2,3 41 110 2,3,000 5,000 10,000 1,600 1,100 2,3,000 5,200 10,000 1,600 1,100 2,3,000 5,200 10,000 1,600 1,100 2,3,000 5,200 10,000</th> <th>0.00 0.38 0.79 0.400 150.00 156 2000 350 0.400 150.00 82 940 330 220 2400 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 520 2300 2700 2900 110 110 110 2100 11000 110 110 $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,100$ $1,100$</th> <th>0.00 0.38 9.3 0.40 150.0 55 2000 350 640 150.0 82 940 230 240 3100 320 920 $1/700$ 2200 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 520 2300 2300 2900 110 110 110 2100 10000 $1,400$ $1,100$ $23,000$ $5,000$ $10,000$ $1,600$ $1,100$ $23,000$ $5,000$ $10,000$ $3,110$ $2,10$ $2,10$ <td< th=""><th>0.00 0.38 9.3 0.40 0.40 150.0 156 2000 350 940 240 150.0 82 940 230 220 $1/700$ 3100 550 $8,100$ $1,900$ $2,200$ $2,200$ 3100 550 $8,100$ $1,900$ $2,200$ $2,200$ 3100 $5,200$ $2,300$ $2,300$ 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41 41 12 3,4 110 41 11 2,3 41 14 41 11 2,3 11 2,3 41 110 2,3,000 5,000 10,000 1,600 1,100 2,3,000 5,200 10,000 1,600 1,100 2,3,000 5,200 10,000 1,600 1,100 2,3,000 5,200 10,000	0.00 0.38 0.79 0.400 150.00 156 2000 350 0.400 150.00 82 940 330 220 2400 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 520 2300 2700 2900 110 110 110 2100 11000 110 110 $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,000$ $1,100$ $23,000$ $5,000$ $10,000$ $1,100$ $1,100$	0.00 0.38 9.3 0.40 150.0 55 2000 350 640 150.0 82 940 230 240 3100 320 920 $1/700$ 2200 3100 550 8100 1900 2200 3100 550 8100 1900 2200 3100 520 2300 2300 2900 110 110 110 2100 10000 $1,400$ $1,100$ $23,000$ $5,000$ $10,000$ $1,600$ $1,100$ $23,000$ $5,000$ $10,000$ $1,600$ $1,100$ $23,000$ $5,000$ $10,000$ $1,600$ $1,100$ $23,000$ $5,000$ $10,000$ $1,600$ $1,100$ 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results, anions	
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Appendix 4	
10.4	

MR	Sample ident.	Location	Site / Water area	Unit	LAS	SDS	SDSEO	CAPB	Sum
2529	DK-1-Eff	Avedøre, Sjælland	Spildevan dscenter Avedøre	l/gu	10	<0.19	<0.14	<0.12	10
2530	DK-2-Eff	Køge, Sjælland	Køgeegnens Renseanlæg	ug/l	2.4	<0.17	<0.14	<0.11	2.4
2531	DK-3-Eff	København, Sjælland	Lynetten Renseanlæg	ug/l	240	1.20	5.7	<0.42	250
2621	FI-1-Eff	Turku	Kakolanmäki WWTP	ug/l	5.8	<0.22	0.24	<0.14	6.1
2622	FI-2-Eff	Tampere	Viinikanlahti WWTP	ug/l	5.2	<0.21	0.12	<0.13	5.3
2623	FI-3-Eff	Helsinki	Viikki WWTP	ug/l	15	<0.23	0.57	<0.14	16
2658	FO-1-Eff	Sersjantvíkin STP	Effluent well	ug/l	82	11	140	31	260
2659	FO-2-Eff	Main Hospital STP	Effluent well	ug/l	190	12	400	130	730
2660	FO-3-Eff	Klaksvik Hospital	Manhole, Infl	ng/l	1,800	390	1,000	680	3,900
2669	GL-1-Eff	Quinngorput	sewage/Nuuk	ug/l	5,800	7.2	4,000	069	10,000
2670	GL-2-Eff	Nuuk imeq	sewage/Nuuk	ug/l	2,000	7.2	1,800	400	4,200
2671	GL-3-Eff	Sana Bay	sewage/Nuuk	ng/l	3,000	6.3	2,200	830	6,100
2685	IS-3-Eff	Hafnarfjörður	Hraunavik WWTP	ug/l	640	15	170	44	860
2564	NO-1-Eff	Oslo	VEAS WTW	ug/l	2.9	<0.16	0.17	<0.10	3.0
2565	NO-2-Eff	Oslo	VEAS WTW	ug/l	4.3	<0.15	<0.14	<0.10	4.3
2566	NO-3-Eff	Oslo	VEAS WTW	ng/l	2.0	<0.16	0.19	<0.10	2.2
2552	SE-1-Eff	Stockholm	Henriksdal WWTP	ug/l	2.4	0.18	0.39	0.54	3.5
2496	SE-2-Eff	Borås	Gässlösa WWTP	ug/l	25	0.31	2.6	<0.15	28
2489	SE-3-Eff	Bollebygd	Bollebygd WWTP	l/gu	9.4	0.29	3.9	<0.13	14



Ved Stranden 18 DK-1061 Copenhagen K www.norden.org

Quaternary ammonium compounds

The report "Quaternary ammonium compounds Analyses in a Nordic cooperation on screening" describes the findings of a Nordic environmental study. The quaternary ammoniums included are compounds which are used in large volumes in a variety of industrial, health sector and domestic products. The quaternary ammoniums are used to provide antistatic, antibacterial, emulating and other properties in a range of formulations like hair conditioners, cosmetics, in fabric softeners and in cleansing and disinfecting products. Some quaternary ammoniums are poorly degraded and some are highly toxic to aquatic organisms. The samples analysed were taken mainly near assumed hot-spot areas as in sewage lines and in receiving waters, but also in background areas far from anthropogenic sources. Samples include water, sludge, sediment and fish.

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