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REPORT

Screening of the fragrances OTNE, acetyl cedrene and diphenylether

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REPORT

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Summary

A screening study of the environmental occurrence of three fragrances has been performed. The studied compounds were:

- OTNE (CAS 54464-57-2)
- Diphenylether (CAS 101-84-8)
- Acetyl cedrene (CAS 32388-55-9)

As reference substances the fragrances galaxolide, tonalide, musk xylene, musk ketone and the metabolite galaxolide lactone were examined. The assignment is part of the Environmental Protection Agency's environmental monitoring programme, but also includes the participation of 13 county administrative boards. The sampling programme was designed according to the following key questions:

- To what degree are the substances released to and from municipal waste water treatment plants (WWTP)
- Do the WWTP effluents influence the levels in the receiving waters
- Do diffuse emissions occur in the urban environment
- Are the substances released to the environment via point sources: industries and landfills
- Are the substances accumulated in aquatic biota
- Do human exposure occur?

In total the study comprised 176 samples in the following media, listed in decreasing order of frequency: sewage sludge > WWTP effluents > surface water > incoming waste water > sediment > aquatic biota > stormwater and breast milk > landfill leachate. This report aims at giving a general overview of the results and to present overall interpretations.

OTNE and acetyl cedrene were commonly detected in incoming waste waters, effluents and sewage sludge from municipal WWTPs. DE was common in incoming waste water but was less frequently detected in sewage sludge and effluents. Levels of OTNE and acetyl cedrene in incoming waste water were reduced in the WWTPs with approximately 70 %, while levels of DE was reduced with more than 90 %. OTNE was the most abundant substance, occurring at higher concentrations that the reference substance galaxolide.

Surface water, sediment and biota (fish) were sampled from 16, 4 and 6 WWTP recipients, respectively. OTNE and acetyl cedrene were detected in surface water from 7 and 2 of these recipients, respectively. OTNE was also found in one fish sample, which show that this substance can bioaccumulate in biota. These results demonstrate that WWTPs can influence the levels of OTNE and acetyl cedrene in their recipients. Several of the examined reference substances were also detected in the recipients.

Human exposure was studied by examining the occurrence of fragrances in breast milk from 7 individual samples. OTNE was found in two samples and traces of acetyl cedrene in one sample. Levels of OTNE in breast milk were within the same range as found for the reference substances galaxolide and tonalide.

According to our judgement OTNE is the chemical of most concern in this study. This is motivated by the high levels found of the substance, that the substance is bioaccumulated and that human exposure is demonstrated. There appears to be no experimental data on the ecotoxicity of this chemical. The high abundance in waste water treatment plants, environment and human agree with the high exposure index predicted by the National Chemicals Inspectorate. Furthermore, the use of OTNE appears to increase.

Sammanfattning

En screening av tre doftämnens förekomst i miljön har genomförts. De studerade föroreningarna är:

- OTNE (CAS 54464-57-2)
- Difenyleter (CAS 101-84-8)
- Acetyl cedren (CAS 32388-55-9)

Som referenssubstanser undersöktes doftämnena galaxolid, tonalid, mysk xylen, mysk keton och metaboliten galaxolid lakton. Uppdraget ingår i Naturvårdsverkets miljöövervakning, men innefattar också deltagande av 13 länsstyrelser. Provtagningprogrammet har utformats för att belysa följande frågeställningar:

- I vilken mån sprids ämnena till och från kommunala reningsverk
- Kan utsläpp från kommunala reningsverk påverka halterna i recipienterna
- Sker diffus spridning i urban miljö
- Sprids ämnena till miljön från punktkällor: industri och deponier
- Bioackumuleras dessa ämnen i akvatisk biota
- Sker human exponering?

Totalt omfattade undersökningen 176 prov, fördelat på följande matriser i avtagande omfattning: slam > utgående avloppsvatten > ytvatten > inkommande avloppsvatten > sediment > akvatisk biota > dagvatten och bröstmjölk > lakvatten. Denna rapport syftar till att ge en allmän beskrivning av resultaten samt att presentera övergripande tolkningar.

OTNE och acetyl cedren förekom allmänt i inkommande och utgående avloppsvatten samt i slam från kommunala reningsverk. DE var allmänt förekommande i inkommande avloppsvatten men påträffades mer sällan i slam och utgående avloppsvatten. Halter av OTNE och acetyl cedren i inkommande avloppsvatten reducerades med ca 70 % i reningsverken, medan halter av DE reducerades med mer än 90 %. OTNE var det ämne som påträffades i högst halter i avloppsvatten, och dessa halter överskred motsvarande för referenssubstanserna.

Ytvatten, sediment och fisk provtogs från recipienterna till 16, 4 respektive 6 reningsverk. OTNE och acetyl cedren påträffades i ytvattnet från 7 respektive 2 av dessa recipienter. OTNE påträffades också i ett fiskprov, vilket visar att detta ämne kan bioackumuleras i biota. Detta visar att utsläpp från reningsverk kan påverka förekomsten av OTNE och acetyl cedren i deras recipienter. Flera av de undersökta referenssubstanserna detekterades också i recipienterna.

Human exponering studerades genom att undersöka doftämnenas förekomst i bröstmjölk från 7 individer. OTNE påträffades i 2 prov och spår av acetyl cedren i 1 prov. Halter av OTNE i bröstmjölk låg inom samma haltintervall som galaxolid och tonalid.

Enligt vår samlade bedömning av dessa tre ämnen är OTNE det ämne som förtjänar mest fortsatt uppmärksamhet. Detta motiveras av att OTNE uppträder i höga halter, bioackumuleras i akvatisk miljö och att human exponering sker. Det föreligger inga experimentella data kring OTNEs ekotoxikologiska egenskaper. Den omfattande förekomsten i reningsverk, miljö och människa stämmer väl överens med det exponeringsindex som Kemikalieinspektionen presenterat, och som bygger på förutsedd risk för spridning. Användningen av OTNE förefaller även att öka.

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1. Introduction

1.1. General

In environmental science and monitoring, the last 10-15 years have brought about a focus on relatively polar pollutants. Kolpin et al. (2002) demonstrated widespread pollution of rivers located downstream urban areas in the US. Pollutants with various intended use and of various origin were found in those rivers, including e.g. pharmaceuticals, stimulants, detergents, biocides, pesticides, plasticizers, flame retardants and fragrances. Similar findings have been drawn from numerous studies since then.

The Swedish Screening program, run by the Environmental Protection Agency, has also included a large number of samples from urban areas and waste water treatment plants (WWTPs). A review on these Swedish screening studies that encompasses urban areas and WWTPs can be found in WSP (2010). It appears that WWTPs can be major "sources[1]" of many current use polar pollutants. There is also a potential for direct diffuse emissions through e.g. urban and industrial stormwater.

As an assignment from the Swedish Environmental Protection Agency, WSP Environmental has during 2011-2012 performed a national screening investigation of three groups of relatively polar or volatile chemicals in the Swedish environment:

1. Fragrances: OTNE, acetyl cedrene and diphenylether
2. Complexing agents: EDTA, NTA, DTPA, 1,3-PDTA and ADA
3. Three polar pollutants: TPPO, TMDD and TCEP.

A number of regional screening studies of the same chemicals have also been performed by the county administrative boards, and are reported jointly with the national screening study in this and two other reports.

The goals of these studies are to investigate if:

- these chemicals are found in the Swedish environment
- diffuse releases appears to occur
- these chemicals are present in background lakes
- WWTP effluents may influence the chemical status of aquatic recipients
- industrial use may lead to a direct emission

For fragrances the goal is also to investigate if:

- these chemicals are bioaccumulated
- human exposure occurs

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[1] WWTP´s may not be considered as primary sources, because the chemicals emitted from wwtp´s are generally used upstream of the wwtp.

1.2. Fragrances

Fragrances are compounds used in toilet requisites such as soap, cosmetics, perfumes, shampoo and skin care products, but also in detergents and household chemicals, for their sweet smelling (COWI, 2011). Fragrances are also used in food, as additives in different material and products and as industrial chemicals. The occurrence of fragrances in the Swedish environment has earlier been studied and compounds like galaxolide (HHCB), tonalide (AHTN), musk ketone (MK), musk xylene (MX) and galaxolide lactone (HHCB LAC) have mainly been found in association with municipal sewage treatment plants (SWECO 2008 and 2010). Bioaccumulation and human exposure to musk compounds have also been found (Lignell 2004 and 2008; SWECO 2010). In the Great Lakes, sediment studies have revealed that several fragrances display increasing concentration trends over the 20th century (Peck et al., 2006).

This screening was focused on the chemical compounds OTNE (CAS 54464-57-2), diphenylether (DE, CAS 101-84-8) and acetyl cedrene (AC, 32388-55-9), which have not been studied in Sweden earlier. In addition HHCB, AHTN, MK, MX and HHCB lactone were included as reference compounds. The use of OTNE and AC are increasing in Sweden, mainly in perfume- and toilet requisites (product register, www.kemi.se). DE has a historical high use in Sweden, but until 2009 the use has decreased. However, in 2010 the use was increased again and the use is within heat transfer media.

OTNE, AC and DE have been found in WWTP in Europe and/or U.S (Simonich et al, 2002; Difrancesco et al, 2004; Harrison et al, 2006; Bester et al, 2008a). OTNE and AC have also been detected in anthropogenically influenced rivers in Europe (Balk and Rutten, 2000; Bester et al, 2008b).

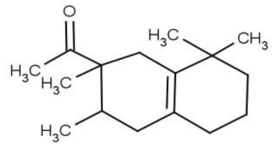
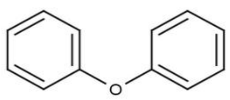
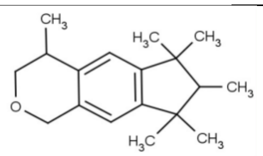
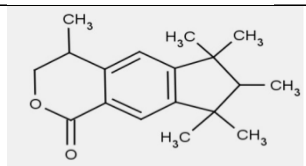
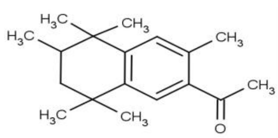
2. Properties

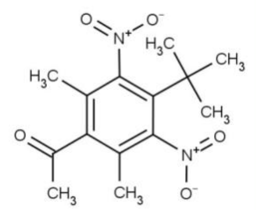
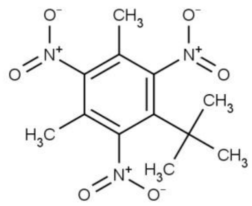
2.1. Physical and chemical properties

Eight different fragrances are included in this screening, whereof three are the main compounds, OTNE, DE and AC, and five reference compounds, HHCB, HHCB Lactone, AHTN, MK and MX, see Table 1. The reference compounds are fragrances that have been included within the screening program earlier (SWECO 2008, 2010).

OTNE is a synthetic terpene and DE is an ether. Acetyl cedrene (AC) exists as two different molecular structures with the same molecular weight and chemical formula (COWI, 2011). However it is only methyl cedryl ketone that is searchable in different chemical databases, and hence this study is performed on that particular compound.

Table 1. Molecular structure, cas.nr and the abbreviations used in this report of the studied compounds. HHCB; AHTN, HHCB LAC, MK and MX are reference substances in this study.

Name	CAS	Structure	Full name
OTNE	54464-57-2		*Isocyclemone E*
DE	101-84-8		Diphenyl ether
AC	32388-55-9		Methyl cedryl ketone
HHCB	1222-05-5		Galaxolide
HHCB LAC	N/A		Galaxolide lactone
AHTN	21145-77-7		Tonalide

Name	CAS	Structure	Full name
MK	81-14-1		Musk ketone
MX	81-15-2		Musk xylene

Physical and chemical properties of OTNE, AC and DE are summarised in Table 2. OTNE, AC and DE have a moderate water solubility and vapour pressure. All three compounds are semivolatile (Henry´s law constant similar to naphthalene) and lipophilic (relatively high log K_{ow}).

Table 2. Physical and chemical properties of the studied compounds. Reference were no other is given is COWI, 2011. Italicize data are model calculated.

Property	OTNE	AC	DE
M, g mol^{-1}	234[1]	246[3]	170[3]
log K_{ow}	5,23	5,6-5,9	4,2[5]
H, Pa-m^3 mol^{-1}	*47,6*[2]	*15,2*[2]	28,3[5]
Vapour pressure, Pa	0,2[1]	0,058[4]	20[3]
C, mg/l	*2,68*[1]	*1,28*[4]	21[3, 5]

[1]Chen et al. 2009. [2]Calculated with EPIWIN. [3]Prevent. [4]Dai, 2009. [5]USEPA, 2010.

2.2. Bioaccumulation and Toxicity

Toxicity data for OTNE, AC and DE are limited and none of the compounds have a binding classification in the database CLP/GHS. Most data are modeled or estimates. All three compounds however are considered, in varying degree, to have negative environmental properties: e.g. toxicity to aquatic organisms, persistence and/or bioaccumulation. Bioaccumulation and toxicity data for OTNE, AC and DE are summarised in Table 3.

Table 3. Bioaccumulation and toxicity of the studied compounds. Reference were no other is given is COWI, 2011. Possible classification as PBT is denoted with P, B or T. Italicized data are model calculated or estimates.

Property	OTNE	AC	DE
BCF	*1220*	867-3920	49-594[1]
Log K_{oc}	*4.64*[2]		*3.2*[1]
NOEC, mg/l, lowest reported value	n.d.	n.d.	0.4 (Dafnia)
$PNEC_{aqua}$, µg/l	*1-10*	*<1*	4.1
P (persistence)	*Probably*	*Probably*	*No*[1]
B (bioaccumulation)	Probably	*Probably*	*No*[1]
T (toxicity)	?	?	?

[1]USEPA, 2010; [2]Chen et al, 2009

The octanol-water partitioning coefficients (Log K_{ow}, see Table 2) for OTNE and AC indicate that there is a risk that these compounds are bioaccumulated in organisms. Bioconcentration factors (BCF) for OTNE (modeled) and AC (measured) indicates that these substances may fulfill the criteria regarding bioaccumulation for a PBT substance (COWI, 2011). Calculations and measurements on the persistence of OTNE and AC indicate that they also may fulfill the criteria of a persistent substance according to REACH appendix XIII (COWI, 2011). DE is not readily biodegradable and is expected to have moderate persistence and low bioaccumulation potential (USEPA, 2010).

Modeled or measured toxicity data indicates that OTNE, AC and DE should be classified as very toxic or toxic to aquatic organisms (COWI, 2011). According to ECHA guidance (2008) for PBT assessment a substance with a NOEC[1] (long-term) <0.01 mg/l for marine or freshwater organisms could be considered as toxic.

[1] NOEC = No observed effect concentration

3. Use and release

In Sweden OTNE and AC are mainly used in perfume- and toilet requisites, whereas DE mainly is used within heat transfer media (product register, www.kemi.se). In 2010 the registered use of OTNE, AC and DE in Sweden were 11.5, 1.8 and 25 tonnes, respectively. The same year the registered amount of products was 174, 50 and 59 for OTNE, AC and DE, respectively. In EU OTNE and AC are low volume products (i.e. 10-1000 tonnes per year is produced or imported), whereas DE is classified as a high volume product (>1000 tonnes/year) (COWI, 2011).

The use of OTNE and AC are increasing in Sweden, see Figure 1. DE has a historical high use in Sweden but until 2009 the use has decreased (Figure 1). However, in 2010 the use of DE was increased again.

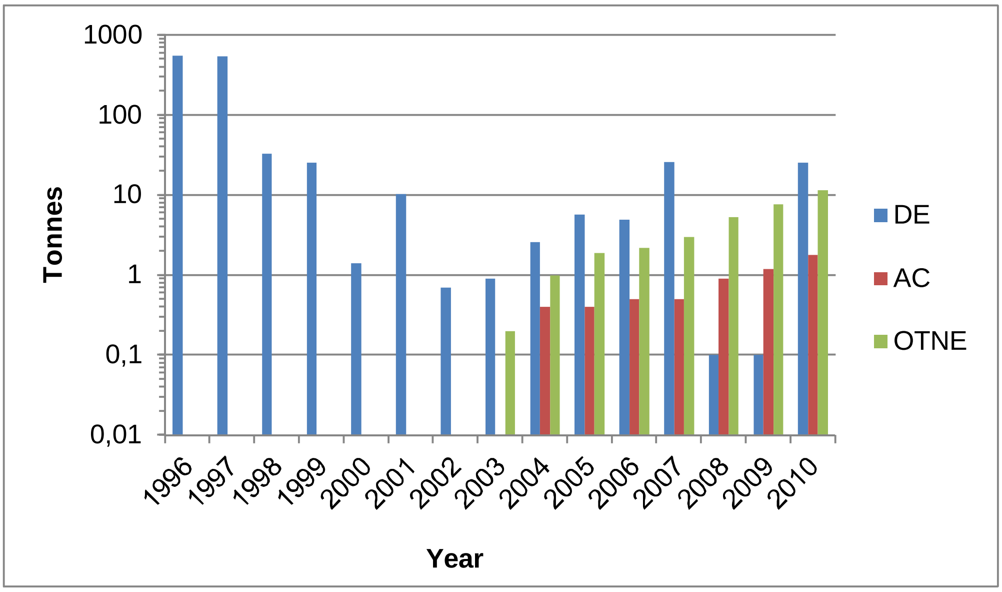


Figure 1. Registered amounts of OTNE, AC and DE used in Sweden. Data from product register, www.kemi.se. For DE amounts were registered since 1992, for AC since 1996 and for OTNE since 1999. Note the logarithmic axis.

Earlier studies show that fragrances are mainly spread to the environment from the use of products containing these compounds via effluents from municipal waste water treatment plants (COWI, 2011). Other possible identified pathways are industrial effluents and the addition to soil from landfills and sludge treated arable land. DE may also be spread via air.

To give a rough estimate of the potential for diffuse release of individual chemicals, the National Chemicals Inspectorate has developed an "exposure index". This index gives a value from 1 to 7, on relative terms, for the potential for release to e.g. WWTPs and for human exposure. The index considers both the amounts used and the way the chemical is handled and used. As an example, equal amounts used give a higher index if the use is as a solvent rather than if it was used as a raw product for polymerisation.

The exposure index for OTNE, DE and AC based on data from 2008 are shown in Table 4. These three chemicals have the highest exposure index (7) for air, WWTP and humans indicating a high release via air and WWTP and a high human exposure. Exposure index for soil and surface water is also high ranging from 4 to 6. In chapter 8, these predictions will be compared to the actual levels measured in this study. A former review of screening data from urban areas and WWTPs showed a fairly good agreement between the exposure indexes and measured levels, although the scatter was wide (WSP, 2010).

Table 4. Exposure index (KemI, 2008). The relative scale goes from 1 to 7.

	Exposure index					***Trend***	
Substance	Surface water	Air	Soil	WWTP	Human	*Human (-2 - +2)*	*Environment (-2 - +2)*
DE	6	7	6	7	7	2	2
OTNE	5	7	5	7	7	2	2
AC	4	7	6	7	7	2	2

4. Previous environmental studies

OTNE, AC and DE have been analysed in environmental samples previously. All three are poorly examined. A selection of data is presented in Table 5. Data are mainly from waste water treatment plants from US and Europe. No previous investigation from Sweden has been found.

All three compounds have been found in sewage sludge from municipal waste water treatment plants in the range of thousand to ten thousands µg/kg dw. OTNE and AC are also found in incoming waste water and effluents with levels in effluents ranging from hundreds to thousands of ng/l. OTNE and AC are also found in anthropogenically influenced rivers in levels from ten to hundreds of ng/l.

Studies of DE performed 30 to 40 years ago have detected DE in industrial waste water (10 000-200 000 ng/l), surface water (e.g. >100 ng/l in Great Britain and 48 ng/l in Switzerland), ground water (1000 ng/l) and drinking water (3 ng/l) (COWI, 2011). DE has also been detected in landfill leachate and human adipose tissue (COWI, 2011).

No previous findings of levels in biota or human tissues have been found for OTNE and AC. Other more commonly studied fragrances such as HHCB and AHTN have been found to accumulate in both aquatic biota (fish) and human breast milk in Sweden (SLV, 2003; Lignell et al, 2004 and 2008; SWECO, 2008 and 2010).

Table 5. Environmental levels of OTNE, AC and DE determined in some previous studies. Levels given as intervals (min-max), single values or mean ± standard deviation. All water samples in ng/l.

Substance	Sewage sludge µg/kg	Incoming waste water	Effluents	Surface water	Study site	Reference
TNE	2000 - 4000	4000 - 13000	500 - 6900		Europe	Bester et al, 2008a
		3550 ± 1930	159 ± 117[1]		US	Simonich et al, 2002
		9000 ± 3770	3190[1]		Europe	Simonich et al, 2002
	7300 ± 1400; 30700 ± 3700				US (Wilmington and Georgetown)	Difrancesco et al, 2004
				30-100	Ruhr river catch-ment basin	Bester et al, 2008b
				29-810	Danube, Hungary	Bester et al, 2008b
AC	9000 ± 1600; 31300 ± 5100				US (Wilmington and Georgetown)	Difrancesco et al, 2004
		4970 ± 2270	176 ± 150[1]		US	Simonich et al, 2002
		7150 ± 4320	680[1]		Europe	Simonich et al, 2002
				50 - 100	Rhine, Meuse	Balk and Rutten, 2000
DE	ND 99600 ± 55100				US	Harrison et al, 2006; Difrancesco et al, 2004

[1]Effluent from WWTP with primary gravitational settling and activated sludge treatment.

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5. Sampling strategy and study areas

The study consists of a national programme, financed by the Swedish EPA, and regional programmes for 13 counties (Dalarna, Gotland, Jönköping, Kalmar, Kronoberg, Norbotten, Skåne, Södermanland, Uppsala, Värmland, Västernorrland , Örebro and Östergötland). The national and regional programmes are shown in Table 6 and Table 7. All sample details are listed in Appendix 1.

WSP developed a general strategy for the investigations, and this strategy was communicated and discussed with all participating county administrative boards. In each county, the regional sampling programme was setup and implemented by the county administrative boards. The strategy of the national programme is outlined below:

- A possible influence from long-range atmospheric transport was assessed by sampling in two national background lakes (Limmingssjön in the Örebro county and Remmarsjön in Västernorrland county).
- A possible urban influence, resulting from diffuse emissions, was investigated by sampling in three urban regions (Stockholm, Eskilstuna and Uppsala). This includes both local background and city centre. Diffuse emissions were also assessed by analysing samples from waste water treatment plants (see below).
- The role of waste water on contamination of the aquatic environment was investigated at two municipal sewage treatment plants and at the recipients of these WWTPs.
- To illustrate point source emissions, samples were taken in a WWTP and the recipient of waste water from an industry of personal care products. This includes both local background and close to the source. The recipient also represents a WWTP recipient.
- As indicators of bioaccumulation and human exposure, fish in downstream surface waters of urban areas/WWTPs/industry and breast milk from urban residents (Stockholm and Gothenburg) were analysed.

Table 6. National programme. The total number of samples is 54.

Site	Surface water	Storm-water	WWTP incoming	WWTP eff	Sewage sludge	Sedi-ment	Biota (fish)	Breastmilk
Background	2					2	2	
Local back-ground	4					2	1	
Urban	3	2				2	2	7
WWTP			4	4	4			
WWTP reci-pients	4					3	3	
Industry	1					1	1	
Total	*14*	*2*	*4*	*4*	*4*	*10*	*9*	*7*

The regional programmes were mainly focused on municipal sewage treatment plants and their recipients.

Table 7. Regional programmes. The total number of samples is 122.

Site	Surface water	Stormwater	Landfill leachate	WWTP incoming	WWTP eff	Sewage sludge	Sediment	Biota (fish)
Urban	2	5					1	
WWTP				13	38	40		
WWTP reci-pients	12						2	4
Landfills	1		1					
Diffuse	2						1	
Total	*17*	*5*	*1*	*13*	*38*	*40*	*4*	*4*

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6. Methods

6.1. Sampling

WSP developed general recommendations for sampling which was also communicated with the county administration boards. This protocol for sampling was sent to all personal involved in sampling, to assure similar treatment. Samples were stored dark and cold until transport to the laboratory. Water samples were treated with a small amount of hydrochloric acid before stored to stop any biological activity in the samples.

The national screening of water and sediments in urban and industrial sites were performed by WSP, local contractors or county administration boards. Samples of surface water and sediment from background lakes were sampled by the county administration board in which samples were collected. Water samples were generally taken as grab samples and surface sediments by gravity corers. Waste water and sewage sludge were sampled by staff at the waste water treatment plants.

Perch from the Stockholm, Eskilstuna and Uppsala region were catch during autumn 2011 by Stockholms Stad, Åfiske and Upplandsstiftelsen respectively, who kindly supplied us with these samples. Perch from background lakes and breast milk samples were kindly supplied by the Swedish Museum of Natural History (Miljöprovbanken/MPB).

6.2. Chemical analysis

Chemical analyses were performed by ALS Scandinavia in cooperation with GBA Germany. The analyses were performed according to the methods outlined below.

Water

- Sample amount: 50- 1000 mL (depends on matrix)
- Daily blank samples
- Internal standards: deuterated AHTN and deuterated musk xylene
  - AHTN-D3 Dr. Ehrenstorfer, Code XA10048600I0, 100 µg/ml
  - musk-Xylene-D15 Dr. Ehrenstorfer, Code XA15360100AC, 100 µg/ml
- Liquid/liquid-extraction with MTBE (1 x)
- Liquid/liquid-extraction with Hexane (1 x)
- Clean-up with silicagel / silicagel - silver nitrate
- fraction heptan/dichloromethane/ethylacetate
- Concentration down to 0.2 mL (nonane as keeper)

Soil, sediment and sludge

- Sample amount: 0,5- 2g (depends on matrix)
- Daily blank samples
- Internal standards: deuterated AHTN and deuterated musk xylene
- Liquid/liquid-extraction with Acetone/hexane (1 x)

- Liquid/liquid-extraction with MTBE (3 x)
- Clean-up with silicagel / silicagel - silver nitrate
- fraction heptan/dichloromethane/ethylacetate
- Concentration down to 0.5 mL (nonane as keeper)
- if necessary additional clean- up with GPC

Biota samples

- Sample amount: 0,5- 2g (depends on matrix)
- Daily blank samples
- Internal standards: deuterated AHTN and deuterated musk xylene
- Liquid/liquid-extraction with Acetone/hexane (1 x)
- Liquid/liquid-extraction with MTBE (3 x)
- 1. Clean-up: silicagel / silicagel - silver nitrate fraction heptan/dichloromethane/ethylacetate
- Concentration down to 0.5 mL (nonane as keeper)
- 2. Clean- up: GPC

Breast milk

- According to “Temporal trends of synthetic musk compounds in mothers milk and association with personal use of perfumed products”; S. Lignell et al. Environ. Sci. Technol. 2008, 42, 6743-6748
- Internal standards: deuterated AHTN and deuterated musk xylene

Measurement

- Analysis with GC/MS(EI), equipped with 30 m DB5ms column
- Daily 4-6 point-calibration
- Components out of linear working area: dilution

Reporting limits

Reporting limits for the different media are presented in Table 8 and Table 9. In water samples the reporting limits in general varied between 2-10 ng/l for most compounds. For a few water samples, reporting limits were higher due to high concentrations of solids. The reporting limits in sediment and sewage sludge varied between 2 - 330 µg/kg and 2 - 1000 µg/kg dw, respectively. In biota reporting limits varied between 2 - 25 µg/kg fw and in breast milk between 0,2 – 1,5 µg/kg fw.

Table 8. *Reporting limits (LOQs) of fragrances in different water media.*

Substance	*WWTP IN* ng/l	*WWTP EFF* ng/l	*Surface water* ng/l	*Stormwater* ng/l	*Landfill leachate* ng/l
OTNE	2	2	3 - 25	10 - 20	150
DE	30	2 - 15	1 - 5	2 - 10	50
AC	2	5 - 40	1 - 10	2 - 10	50
HHCB	2	2	2 - 10	3 - 5	75
HHCB lactone	2	5	2 - 20	3 - 5	250
AHTN	2	2	2 - 5	2 - 3	75
MK	5 - 20	2 - 5	1 - 3	2 - 3	50
MX	2 - 20	2 - 5	1 - 3	10 - 25	50

Table 9. *Reporting limits (LOQs) of fragrances in sediment, sewage sludge, biota and breastmilk.*

Substance	*Sediment* µg/kg dw	*Sludge* µg/kg dw	*Biota* µg/kg fw	*Breastmilk* µg/kg fw
OTNE	10 - 200	2	20	1,5
DE	2 - 50	2 - 140	2 - 10	0,5
AC	10 - 110	2 - 1000	5 - 10	0,5
HHCB	5 - 80	2	5	0,5
HHCB lactone	10 - 330	810	10 - 25	1,5
AHTN	5 - 80	85	5	0,5
MK	2 - 35	2 - 140	2 - 5	0,2
MX	2 - 35	2 - 140	2 - 5	0,2

7. Results

A general overview of the levels of the studied compounds is presented for each media in this section. Sample details are given in Appendix 1 and all data are presented in Appendix 2. A discussion on spatial trends, emission sources, environmental partitioning and possible risks to the health and environment is given in chapter 8.6.

In total, the national and regional programmes covered 176 samples, more than half of which were from waste water treatment plants (WWTPs) (Table 10). Fragrances were detected in all analysed media except of landfill leachate (n=1). Leachates will therefore not be presented further. Reporting limits are presented in Table 8 and Table 9.

Fragrances were most frequently detected in waste water and sludge from sewage treatment plants, followed by breast milk and surface water (Table 10). The most commonly found compounds in sewage treatment plants were OTNE, AC, HHCB, HHCB lactone and AHTN. These compounds were also the ones that were found in surface water and breast milk. DE, musk ketone and musk xylene were only detected in samples from sewage treatment plants. Fragrances were only rarely detected in stormwater, sediment and aquatic biota (fish). In all of these three media HHCB lactone were found. In sediment and biota HHCB were also found and in biota OTNE. A summary of the levels found in different media are given in Table 11.

Table 10. The occurrence of examined fragrance compounds in different media, where n-tot is the number of samples analysed. When n < 10 the reporting frequency is given as a ratio instead of percentage.

Media	n-tot	OTNE	DE	AC	HHCB	HHCB lactone	AHTN	MK	MX
Incoming ww	17	100%	94%	100%	100%	100%	100%	35%	6%
Effluent	42	100%	17%	88%	100%	98%	100%	71%	7%
Surface water	31	26%	0%	10%	39%	55%	19%	0%	0%
Stormwater	7	0 / 7	0 / 7	0 / 7	0 / 7	1 / 7	0 / 7	0 / 7	0 / 7
Landfill leachate	1	0 / 1	0 / 1	0 / 1	0 / 1	0 / 1	0 / 1	0 / 1	0 / 1
Sediment	14	0%	0%	0%	7%	7%	0%	0%	0%
Sewage sludge	44	100%	41%	91%	100%	95%	98%	5%	0%
Breastmilk	7	2 / 7	0 / 7	1 / 7	4 / 7	5 / 7	2 / 7	0 / 7	0 / 7
Aquatic biota	13	8%	0%	0%	8%	15%	0%	0%	0%

Table 11. A statistic summary of the levels of fragrances found in media where the compounds were detected above the reporting limit. Unit for water media ng/l, for sludge and sediment µg/kg dw and for biota and breast milk µg/kg ww.

Media and parameter	OTNE	DE	AC	HHCB	HHCB LAC	AHTN	MK	MX
Incoming ww, n=17								
min	1800	<30	150	760	13	65	<5 - <20	<2 - <20
max	27000	380	5800	16000	2700	2000	64	18
median	3800	34	560	1900	1100	160		
mean	5800	59	1000	3000	1200	310		
stdev	6000	87	1400	3500	760	460		
Effluent, n=42								
min	130	<2 - <15	<5 - <40	190	<5	9	<2 - <5	<2 - <5
max	4900	12	390	1100	4800	100	11	9,2
median	1000		130	550	1700	41	4,1	
medel	1200		150	590	1800	47	4,4	
stdev	800		99	220	1000	23	2,6	
Surface water, n=31								
min	<3 - <25	<1 - <5	<1 - <10	<2 - <10	<2 - <20	<2 - <5	<1 - <3	<1 - <3
max	460		130	620	4000	32		
Stormwater, n=7								
min	<10 - <20	<2 - <10	<2 - <10	<3 - <5	<3 - <5	<2 - <3	<2 - <3	<10 - <25
max					140			
Sediment, n=14								
min	<10 - <200	<2 - <50	< 10 - < 110	< 5 - < 80	< 10 - < 330	< 5 - < 80	< 2 - < 35	< 2 - < 35
max				32	130			
Sewage sludge, n=44								
min	640	<2 - <140	<2 - <1000	960	<810	<85	<2 - <140	<2 - <140
max	23000	270	3400	11000	14000	1400	62	
median	9500		1200	5900	3300	420		
mean	8500		1200	5600	4000	530		
stdev	5400		810	2600	3100	320		
Biota, n=13								
min	<20	<2-<10	<5- <10	<5	<10- <25	<5	<2-<5	<2- <5
max	46			25	110			
Breast milk, n=7								
min	< 1.5	< 0.5	< 0.5	< 0.5	<1.5	< 0.5	< 0.2	< 0.2
max	2.1		0.50	3.2	9.0	5.6		

7.1. Surface water

Surface water was sampled from 30 sites including:

- 2 national background lakes
- 14 recipients of WWTP:s (16 samples)
- 5 urban recipients and local background samples of 3 of these recipients
- one industrial point source recipient and a local background in the same recipient
- one recipient of a landfill
- 2 diffusely influenced rivers

Fragrances were found in half of the analysed samples of surface water (Table 10) and mainly in the WWTP:s recipients (14 of 16 samples). OTNE, AC and the reference compounds HHCB, HHCB lactone and AHTN were the compounds detected. Most abundant was HHCB lactone, followed by HHCB, OTNE, AHTN and last AC (Table 10). The most frequently detected compounds were also the ones found at the highest concentrations (Table 11). Highest measured concentration of OTNE were 460 ng/l and of AC 130 ng/l.

7.2. Stormwater

Storwater was sampled at 7 sites in the urban environment. OTNE, AC and DE were never detected in any of the samples (Table 10). One of the reference substances, HHCB lactone, was found in a sample from Stockholm. The sample was collected after rainfall after a longer period of dry climate in June 2012. The concentration of HHCB lactone was 140 ng/l.

7.3. Sediment

Sediment was sampled at 14 sites of which 5 represented WWTP recipients, 3 urban recipients, one industrial point source, 2 national background lakes, 2 local backgrounds and one diffusively influenced river. OTNE, AC and DE were never detected in any of the samples (Table 10), however the reporting limits varied widely. The reference compounds were only detected in one sample, in a wetland downstream of a WWTP. The compounds found were HHCB and HHCB lactone and the concentrations were 32 and 130 µg/kg dw, respectively.

Previous studies have detected fragrances at concentrations that generally are lower than the current reporting limits (e.g. Peck et al., 2006). Low sedimentary concentrations may partly be due to the apparently weak partitioning to solids in surface waters (Peck and Hornbuckle, 2004).

7.4. Waste water and sewage sludge

Waste water treatment plants (WWTPs) were the main study objects in this study, comprising 17 incoming waste waters, 42 effluents and 44 sewage sludge samples. OTNE, AC and DE were detected in almost all incoming waste waters (Table 10). OTNE and AC were also detected in almost all effluents and sewage sludge, whereas DE was clearly less frequent in these media. All the reference compounds were also found in wastewater and sewage sludge, except of musk xylene which was rarely detected in waste water and never in sludge (Table

10). Musk ketone was also less commonly found in waste water compared to the other fragrances (Table 10).

In incoming waste water levels of OTNE varied between 1800 to 27 000 ng/l, of AC between 150 to 5 800 ng/l and of DE between <30 to 380 ng/l (Table 11,

Figure 2). The levels of the different compounds in incoming waste water decreased in the following order:

OTNE > HHCB > HHCB lactone > AC > AHTN > DE > MK > MX.

In sewage sludge and effluents the levels of the different compounds followed the same order as in incoming waste water, except that MX was not detected in sewage sludge and that HHCB lactone was found at the highest levels in effluents (Table 11,

Figure 2 and Figure 3).

In general, concentrations were higher in incoming waste water than in effluents, except for HHCB lactone (

Figure 2). The concentrations of OTNE, AC and DE varied widely between the different WWTPs, see appendix 3.This will be discussed further in chapter 8.3.2.

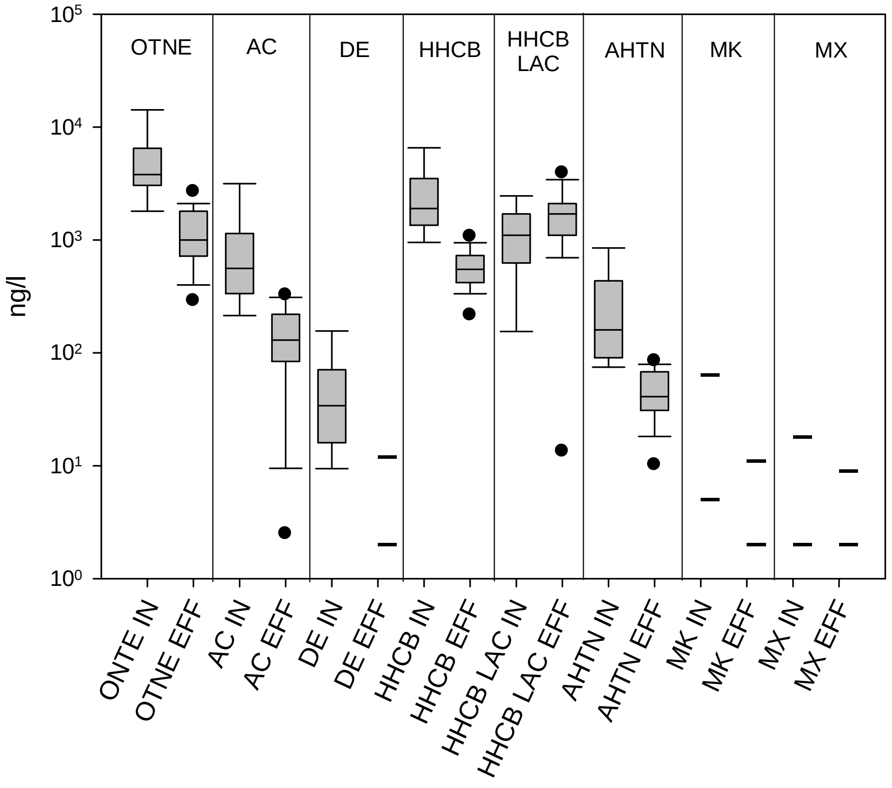

Figure 2. Levels of fragrances in incoming waste water (IN, n=17) and effluent (EFF, n=42) from municipal WWTPs. Levels of some fragrances in waste water are only given as an interval of lowest and highest concentrations, since the detection frequency in these media was low. Note the logarithmic concentration axis. The reporting limits varied between 2-40 ng/l for the different compounds.

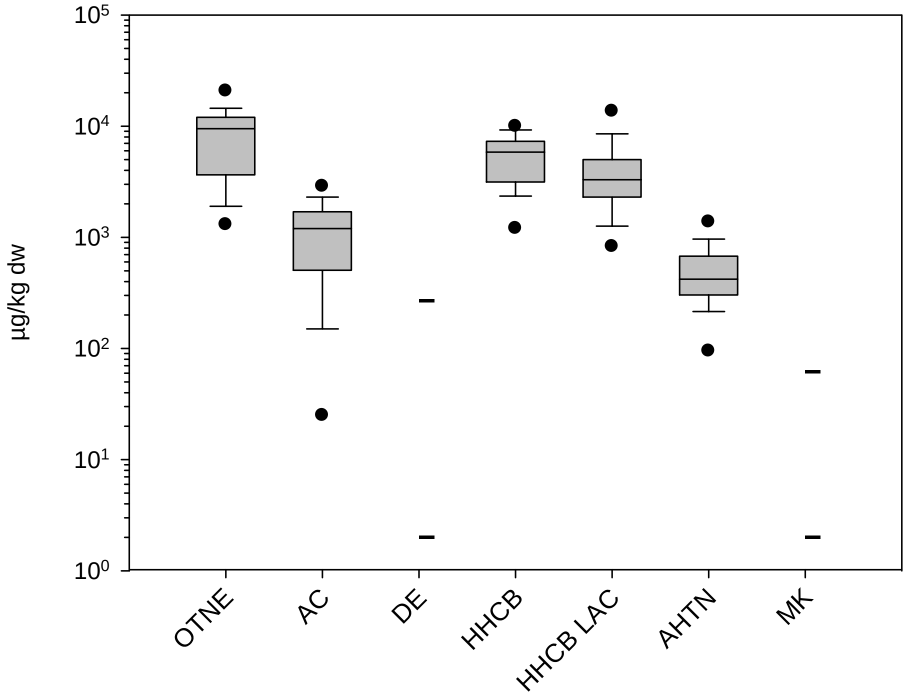

Figure 3. Levels of fragrances in sewage sludge (n=44) from municipal WWTPs (n=40). DE and MK levels are only given as an interval of lowest and highest concentrations, since the detection frequency in this media was low. Note the logarithmic concentration axis. The reporting limits varied between 2-140 µg/kg dw for the different compounds.

7.5. Aquatic biota

The occurrence of fragrances in aquatic biota were examined in fish from 11 sites, 6 of which represented recipients to WWTPs, 2 national background lakes, 2 urban and 1 point source/WWTP recipient. All fish samples were pooled muscle samples, whereof 12 were from Perch and one from Fourhorn sculpin.

Fragrances were detected in fish from two recipients of WWTPs, one Perch sample and one Fourhorn sculpin sample. The Perch sample was from a wetland downstream a WWTP. In the perch sample HHCB and HHCB lactone were found in levels of 25 and 110 µg/kg ww, respectively. In the Fourhorn sculpin sample OTNE and HHCB lactone were found in levels of 46 and 18 µg/kg ww, respectively. Reporting limits for the different compounds were in general between 2-10 µg/kg ww, however for OTNE it was 20 µg/kg ww and for HHCB lactone 10-25 µg/kg ww.

7.6. Breast milk

Fragrances were examined in 7 breast milk samples from residents in two larger citys (Stockholm and Gothenburg). Fragrances were found in 5 samples and most abundant was galaxolide lactone, followed by HHCB, OTNE and AHTN (Table 10,

Figure 4). Traces of AC were also found in one of the samples. DE, MX and MK were not found in any of the samples. The levels of the different compounds ranged from 0.5 to 9 µg/kg ww (

Figure 4). OTNE was found in similar concentration range as HHCB and AHTN. AC was only found in one sample at a low level.

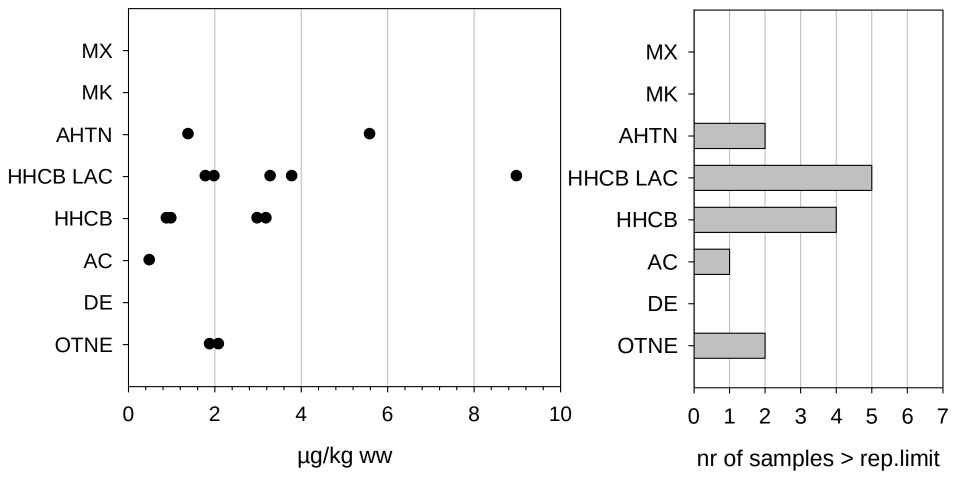

Figure 4. Levels of fragrances in breast milk (n=7) to the left and detection frequency to the right. Reporting limits varied between 0.2-1.5 µg/kg ww.

8. Discussion

8.1. Background areas

No fragrance was found in surface water, sediment or fish in the two national background lakes examined (Remmarsjön and Limmingssjön), which indicates that the studied fragrances not are long range transported. This is in accordance with the findings of one earlier screening of musk compounds (SWECO, 2008) and with conclusions from fugacity modelling of OTNE and AC (COWI, 2011). However, other results indicate that certain musk compounds may be subject to long range transport (Tema Nord, 2004; SWECO, 2010). HHCB and HHCB lactone have been found in arctic char in 2009 from the national background lake Abiskojaure (SWECO, 2010) and HHCB in 4 out of 23 rainwater samples within a Nordic screening study (Tema Nord, 2004).

Fragrance compounds were not found in local background sites in the present study, with the exception of one surface water sample from lake Mälaren at Klubbensborg where HHCB lactone were found (17 ng/l).

8.2. Urban areas

Many chemicals used in typical domestic products are released by diffuse processes. This may result in elevated levels in the urban aquatic environment, which has been recognized in many studies of the Swedish national screening programme (see a review in WSP, 2010). In this report, we try to distinguish between this direct result of diffuse emissions and the impact that is caused by releases from WWTPs.

Several samples of stormwater (n=7), sediment (n=4), surface water (n=5) and fish (n=2) were sampled in urban areas in order to investigate whether there was a general diffuse influence on their environmental occurrence.

OTNE, AC, DE and the reference compounds studied were rarely or never detected in the urban environment, which is in accordance with previous findings of HHCB and AHTN (SWECO, 2008). Neither were OTNE, AC nor DE detected in the urban stormwater samples (n=7), indicating that this is not a major transport pathway for these compounds to the environment.

Fragrances were found in surface water in 2 out of 5 studied cities. In Årstaviken in central Stockholm HHCB lactone were found in slightly higher concentrations as compared to the local background site at Klubbenborg. When also considering that HHCB lactone was found in a single urban stormwater sample, it seems that an urban influence of this chemical cannot be ruled out.

In Fyrisån downstream Uppsala OTNE, AC, HHCB, HHCB lactone and AHTN were found in similar concentrations as in the WWTP recipients. The level of OTNE in that particular sample, 460 ng/l, was the highest found in all surface water sampled within the screening. However, it is unclear whether this sample reflects direct urban emissions or if it is influenced by a WWTP.

8.3. Waste water treatment plants

OTNE, DE and AC were common in WWTPs. OTNE and AC were also found in surface water and OTNE in fish of WWTP recipients.

8.3.1. Levels compared to earlier findings

OTNE, AC and DE were commonly found in waste water and sewage sludge, showing that WWTPs are potentially sources of these compounds to the environment. These findings are in accordance with the use of these compounds and with earlier findings (Bester et al, 2008; Simonich et al., 2002, Difranceso et al, 2004). Levels found in the present study are in general within the same range as previously found (Table 12).

Table 12. Levels of OTNE, AC and DE in sewage sludge (SS), incoming waste water (IN WW), effluents (EFF) and receiving surface waters (SW) of WWTP:s compared to previous studies. Concentrations are given as mean or min-max.

Substance	Present study		Previous studies		Reference
	mean	min-max	mean	min-max	
OTNE					
SS (µg/kg)	8500	640-23000		2000-4000	Bester et al. 2008a
IN WW (ng/l)	5800	1800-27000		4000-13000	Bester et al. 2008a
EFF (ng/l)	1200	130-4900		500-6900	Bester et al. 2008a
AC					
SS (µg/kg)	1200	<2-3400	9000 resp. 31300		Difrancesco et al. 2004
IN WW (ng/l)	1000	150-5800	7150		Simonich et al, 2002
EFF (ng/l)	150	<5-390	680[1]		Simonich et al, 2002
DE					
SS (µg/kg)		<2-270		ND-99600	Difrancesco et al. 2004, Harrison et al, 2006
IN WW (ng/l)	59	<30-380		No recent data	
EFF (ng/l)		<2-12		No recent data	

[1]Single values on effluent from WWTP in Europe with primary gravitational settling and activated sludge treatment. Levels of AC in effluents with other treatments were also presented ranging from approximately 12 to 1400 ng/l.

8.3.2. Variations between different WWTPs

The degree to which concentrations of chemical compounds vary in waste waters and sludge is indicative of the chemicals sources to waste water. High variability or the existences of anomalously high values are indicative of point sources; low variability is indicative of a diffuse input. Table 13 gives CV and skewness as two measures of variability. CV is calculated as standard deviation divided by arithmetic mean. A CV-value larger than ca 50% indicates non-normal distribution. A skewness close to zero would indicate normal distribution. OTNE, AC and DE display significant variability between the samples in waste water and sludge, with highest variability in incoming waste water. In effluents and sludge the variability is clearly lower. Testing for outliers in effluents and sludge, indicated a potential outlier of OTNE in effluents. Excluding this value reduced the variability of OTNE.

Concentration variability was not apparently related to WWTP parameters such as size, domestic or mixed load, stormwater etc. Certain WWTPs were sampled twice, with a few months in between. Concentrations of OTNE, AC and DE in waste water within each of these WWTPs varied at the two occasions, see Figure 5 to Figure 7. The differences were generally within a factor 2 for OTNE and a factor of 3 for AC, but occasionally up to a factor of 40 for AC in effluents. Because most WWTPs were only sampled once, there is no specific information on the temporal variability. Moderate differences between the WWTPs should therefore not automatically be interpreted as true differences in the load of OTNE, AC and DE.

In conclusion, the variability in load of OTNE, AC and DE to many of the WWTPs studied here may be lower than indicated by the concentrations in the spot samples. Indication of influence from point source for OTNE was revealed. The general occurrence of these fragrances in municipal waste waters is thus likely to have a diffuse origin.

Table 13. Statistical description on variability of OTNE, AC, DE, HHCB and AHTN in waste waters and sewage sludge.

Chemical	***n***	***CV***	***Skewness***
OTNE incoming	17	102%	3,1
OTNE effluent	43	69%	2,2
OTNE eff, excl 1 outlier	42	55%	0,73
OTNE sludge	44	63%	0,57
AC incoming	17	132%	3,0
AC effluent	43	67%	0,49
AC sludge	44	66%	0,49
DE incoming	17	148%	3,5
HHCB incoming	17	117%	3,5
HHCB effluent	43	38%	0,53
HHCB sludge	44	45%	0,092
AHTN incoming	17	147	3,4
AHTN effluent	43	48%	0,37
AHTN sludge	44	61%	1,2

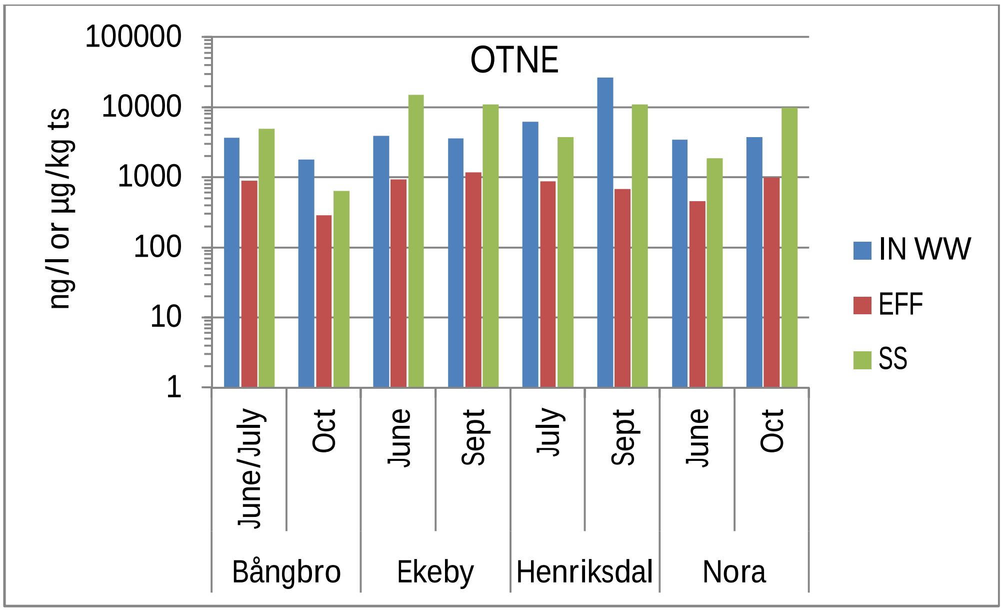

Figure 5. Levels of OTNE in incoming waste water (IN WW), effluents (EFF) and sewage sludge (SS) from 4 different WWTPs sampled once in the summer and once in the autumn 2011.

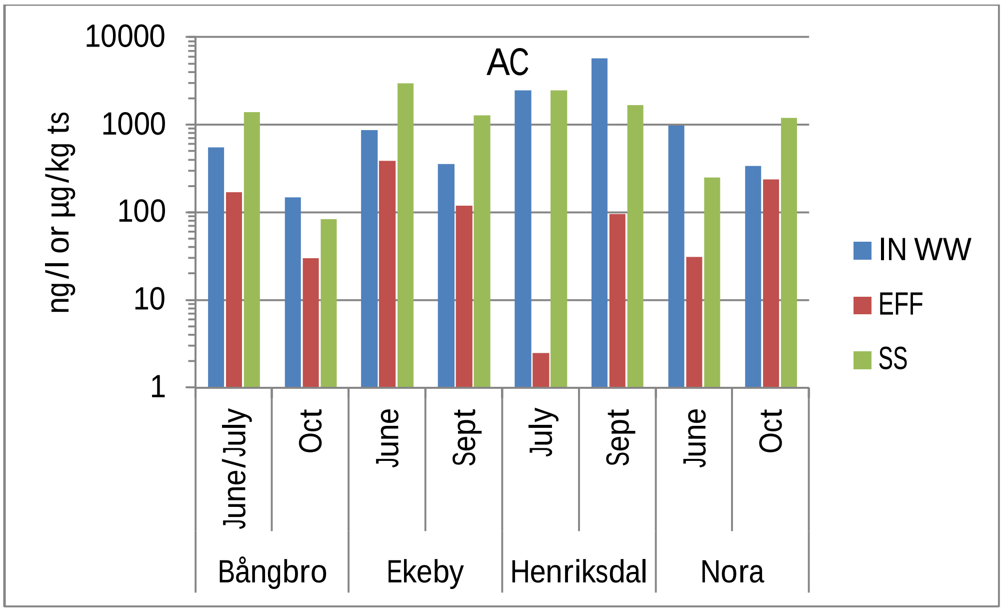

Figure 6. Levels of AC in incoming waste water (IN WW), effluents (EFF) and sewage sludge (SS) from 4 different WWTP:s sampled once in the summer and once in the autumn 2011. Unit for waste water and effluents ng/l and for sewage sludge µg/kg dw.

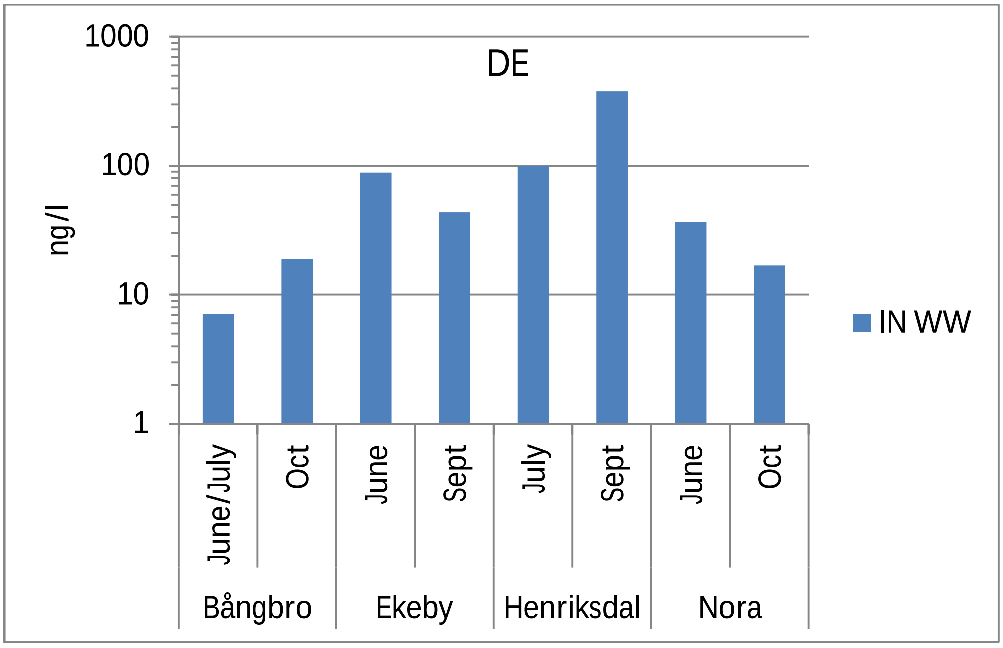

Figure 7. Levels of DE in incoming waste water (IN WW) from 4 different WWTP:s sampled once in the summer and once in the autumn 2011. Unit ng/l.

8.3.3. Correlations between compounds

The correlation between OTNE, AC, HHCB, HHCB lactone and AHTN in incoming waste water, sewage sludge and effluents has been tested with a Spearman rank order correlation test (significant relationships at $p<0.05$). DE was also included in the test of incoming waste water. The following have been found:

- **Incoming waste water** (n=17) **–** all compounds except HHCB lactone correlate positively with each other. Thus, the original compounds seem to have a similar source.
- **Effluents** (n=42) – all compounds except of AHTN correlate with each other, however AHTN correlate with levels of HHCB.
- **Sewage sludge** (n=44) – all compounds, except AHTN with OTNE and HHCB, correlate with each other.

8.3.4. Correlations between different media

The correlation in levels of OTNE, AC, HHCB, HHCB lactone and AHTN between effluents and incoming waste water or sewage sludge has also been tested with a Spearman rank order correlation test (significant relationships at $p<0.05$). The following have been found:

- **Incoming waste water versus effluents** (n=17) **–** no correlations found for any compounds. This indicates that levels of fragrances in effluents are influenced by treatment in the WWTPs rather than levels in incoming waste water.
- **Effluents versus sewage sludge** (n=32) – correlations for HHCB, AHTN and HHCB lactone were found. Thus, for these compounds levels in sludge seems to vary with levels in effluents.

8.3.1. Removal rate

The change in levels of fragrances in incoming waste water and effluents have been calculated for the WWTP were these two matrices were sampled simultaneously and for OTNE, AC, HHCB and AHTN the relative decrease is in general around 70 % and for DE >90 %, see Table 14. Levels of HHCB lactone instead tend to increase in the effluents probably as a result of HHCB being metabolized.

Table 14. Relative change in levels of fragrances from incoming waste water (IN WW) to effluents (EFF) expressed as median values from 17 sampling times at 13 different WWTP.

Substance	Changes in levels from IN WW to EFF (%)
OTNE	-76
DE	>-90
AC	-70
HHCB	-65
HHCB LAC	+12
AHTN	- 65

Bester et al (2008a) calculated a removal rate of OTNE during waste water treatment of 56-64 % in three large scale sewage treatment plants in Europe. Simonich et al (2002) calculated the overall removal (primary and secondary treatment) of fragrances for different types of waste water treatment. The removal of OTNE and AC with primary gravitational settling was approximately 30 % and with primary and secondary treatments between 70 to 99 %. Highest removal rate for all fragrances was with lagoon treatment. Simonich et al (2002) concluded that the profile of fragrances in final effluent was a function of the design of the waste water treatment plant and that in general the removal of sorptive, none biodegradable compounds was correlated with the removal of total suspended solids in the plant, while the removal of nonsorptive biodegradable compounds was correlated with 5-day biological oxidation demand removal in the plant.

8.3.2. Influence on recipients

Levels of OTNE, AC and especially DE are reduced in the WWTPs so that lower levels reach the aquatic environment (Figure 2). However, OTNE and AC were found in surface water of WWTPs recipients in 7 respectively 2 out of 15 samples, showing that WWTPs are potential sources of these substances to recipients. The levels found are within the same range as previously found in anthropogenically influenced rivers in Europe (Table 12). The levels found of OTNE are also within the same range as found for the reference substance HHCB (Table 11).

Table 15. Levels of OTNE, AC and DE in receiving surface waters of WWTPs compared to previous studies. Concentrations are given as min-max. Unit ng/l.

Substance	Present study	Previous studies	Reference
OTNE	<10-290	30-800	Bester et al. 2008b
AC	<2-130	50-100	Balk and Rutten, 2000
DE	<5	No recent data	

Within the national program sampling was performed in a gradient from local background to urban influence and finally to WWTP influence in two cities – Eskilstuna and Stockholm. In both places there were clear influences on the levels of certain fragrances in the surface water downstream the WWTP (Figure 8 and Figure 9). Largest influence in both cities can be seen for the metabolite galaxolide lactone (HHCB LAC). In Stockholm there was also an influence of this compound in the local background site in lake Mälaren, possibly caused by other WWTPs upstream this point. Galaxolide lactone was also found in a sample of stormwater from Stockholm showing that this compound also is spread via this media. In Stockholm sampling was also performed at two depths in Saltsjön, 0.5 and 10 m. Levels of the detected fragrances were clearly higher in the deeper sample.

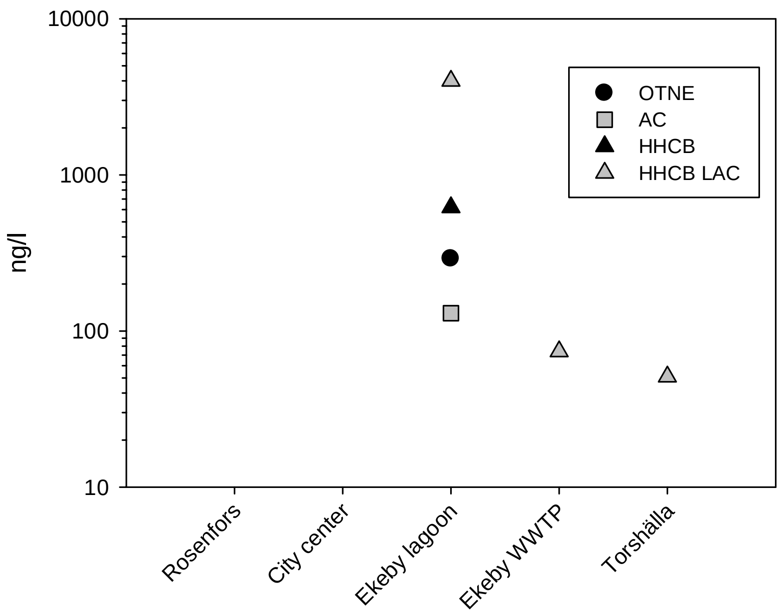

Figure 8. Levels of fragrances in Eskilstuna river and the lagoon at Ekeby WWTP (Regional program). Rosenfors represents a local background and Torshälla is approximately 3 km downstream the WWTP. Note the logarithmic concentration axis.

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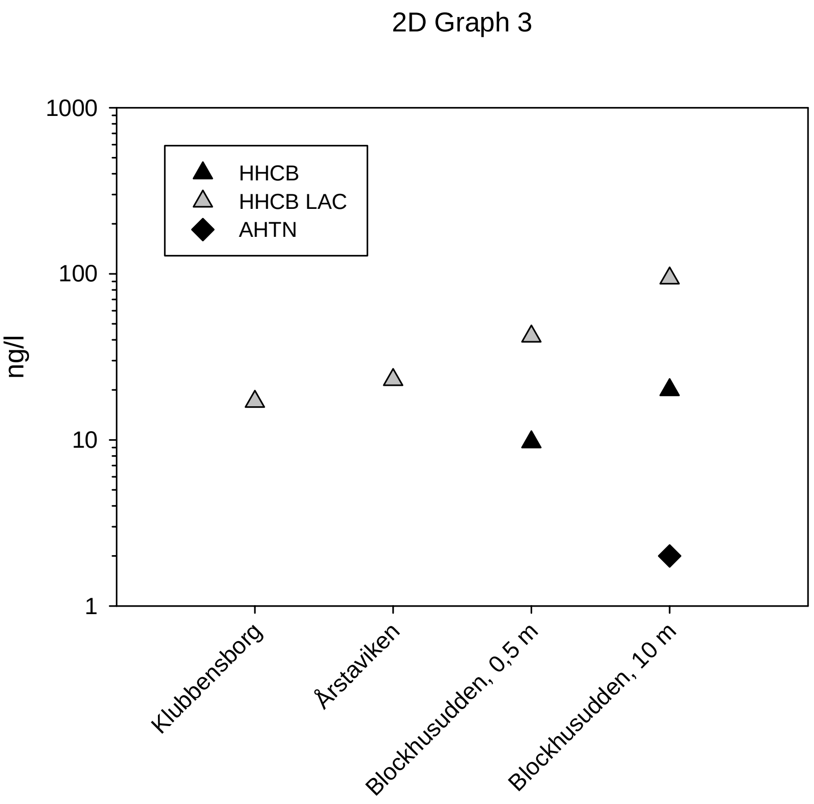


Figure 9. Levels of fragrances in lake Mälaren (Klubbensborg, Årstaviken) and Saltsjön (Blockhusudden). Klubbensborg represents a local background, Årstaviken urban influence and Blockhusudden WWTP influence at two depths (0.5 and 10 m). Note the logarithmic concentration axis.

8.4. Point sources

The environmental impact of an industry producing personal care products were examined by sampling waste water and sludge at the WWTP that receives waste water from the industry. The study also encompasses surface water, sediment and fish from the recipient to the WWTP.

The receiving WWTP displayed higher sludge levels of OTNE and AC than in any of the other 40 examined WWTPs (Figure 10). In effluents levels of OTNE and AC were also one of the highest of 38 examined WWTPs (Figure 11). Levels of AHTN in sewage sludge and effluents were also high. These results indicate that levels of fragrances in the WWTP are impacted by the waste water from the industry. However, no fragrances were found in levels above the reporting limit in the recipient (surface water, sediment and fish) of the WWTP, implying that the environmental influence of the industry is low.

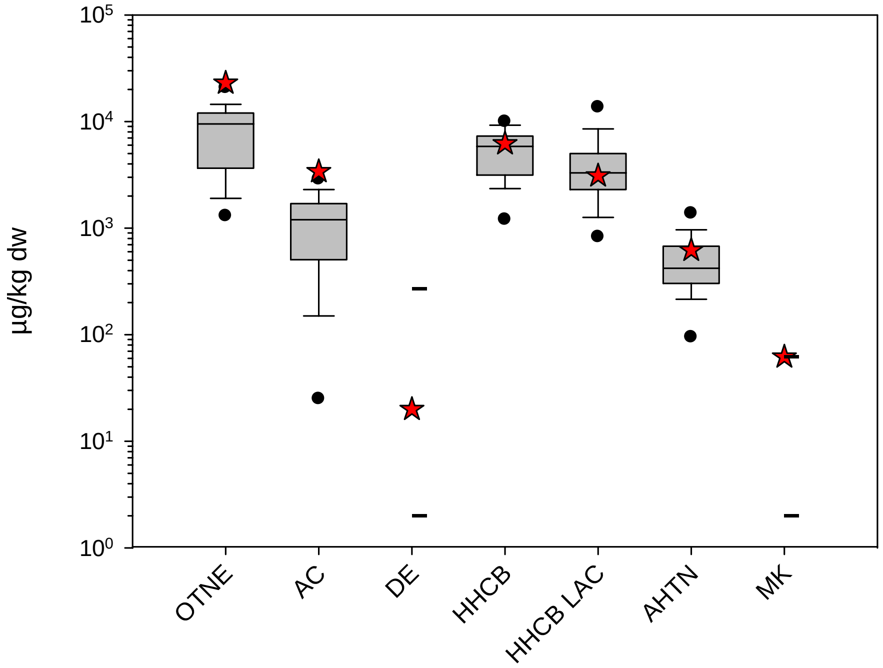


Figure 10. Levels of fragrances in sewage sludge from 40 WWTP (boxplot or concentration interval) compared to levels in a WWTP receiving waste water from an industry producing personal care products (red star).

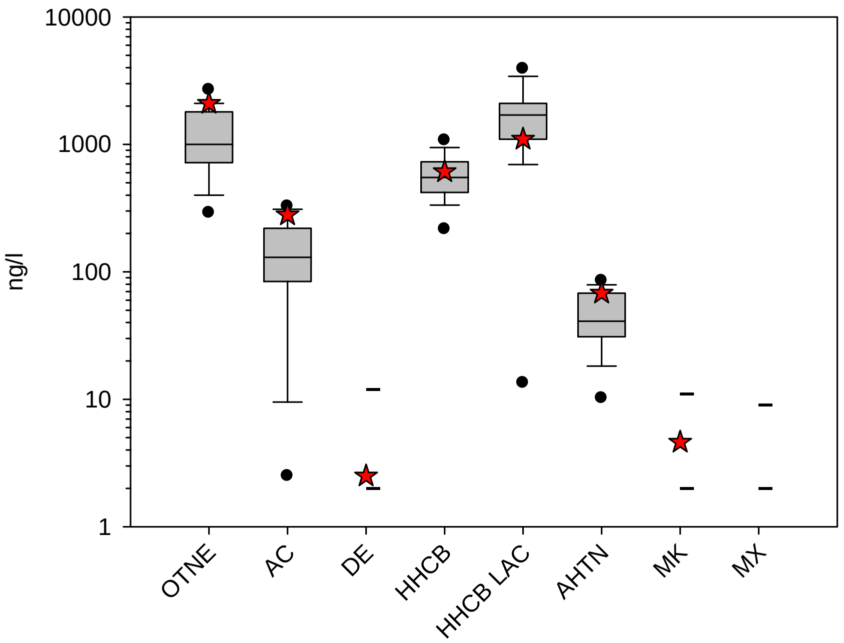


Figure 11. Levels of fragrances in effluents from 38 WWTP:s (boxplot or concentration interval) compared to levels in a WWTP receiving waste water from an industry producing personal care products (red star).

8.5. Bioaccumulation and human exposure

OTNE was found in one out of six fish samples from WWTP recipients, which indicates that this substance accumulates in biota. This finding is consistent with the lipophilic property of the substance (high Log K_{ow}) and conclusions drawn about bioaccumulation of OTNE in the literature study (COWI, 2011). No earlier measurements of OTNE in fish have been found.

In addition, the reference compounds HHCB and HHCB lactone were found in 1 respectively 2 fish samples from WWTP recipients. Levels found of HHCB and HHCB lactone is consistent with previous findings in Sweden, see Table 16. The level found of OTNE is within the same range as previous findings of HHCB and HHCB lactone.

One of the recipients where HHCB and HHCB lactone were found in perch is a wetland of a WWTP. In the receiving surface water of the wetland these compounds were not detected in perch above the reporting limit.

Table 16. Concentrations of detected fragrances in fish from recipients of WWTP compared to earlier studies (SWECO, 2010).

Substance	Species	Present study (µg/kg ww)	SWECO, 2010 (µg/kg)
OTNE	Fourhorn sculpin	46	
OTNE	Perch	<20	
HHCB	Fourhorn sculpin	18	
HHCB	Perch	<5-25	3,4-48
HHCB lactone	Perch	<10-110	<10-260

Musk compounds have earlier been found in human breast milk showing that human exposure occurs and that these compounds are accumulated in human fat (Lignell 2004 and 2008, Reiner 2007). However, the fragrances OTNE, AC and DE have to our best knowledge not been studied in breast milk earlier. In Table 17 breast milk levels found are compared to results from earlier performed studies. To be able to compare the results the levels of the present study has been converted to a lipid weight basis assuming a general lipid weight of 4,6 % in breast milk (Livsmedeldatabasen, www.slv.se). In reality the lipid content may vary between the samples.

OTNE and traces of AC were found in human breast milk samples, indicating that these fragrances also are accumulated in human fat. The more commonly studied musk compounds HHCB, HHCB lactone and AHTN were also found in breast milk in similar concentrations as earlier found in Sweden (SLV, 2003; Lignell et al, 2004 and 2008) and USA (Reiner et al, 2007). However, musk xylene and musk ketone where not found in levels above the reporting limit. Musk xylene has been found in higher levels in breast milk in Sweden in 2002-2003 (Lignell et al, 2004). In the same study they also found a decreasing time trend in levels of musk xylene, which may explain the result of the present study. They also found a decreasing time trend of AHTN.

Table 17. Concentrations of detected fragrances in human breast milk converted to lipid weight basis assuming a general lipid weight of 4,6 % (Livsmedeldatabasen, www.slv.se) compared to earlier studies. Concentrations from the present study are given as min-max, and for the other studies as mean (min-max). Unit, µg/kg lw.

Reference	Present study	Lignell et al, 2004 and 2008	Reiner et al, 2007
Location	Sweden	Sweden	United states
Sample date	2009, 2010	1996-2003	2004
Number of samples	7	101	39
OTNE (µg/kg lw)	<33-46		
DE (µg/kg lw)	<11		
AC (µg/kg lw)	<11-11		
HHCB (µg/kg lw)	<11-70	78 (2,8-268)	220 (<5-917)
HHCB LAC (µg/kg lw)	<33-196		47 (<10-88,0)
AHTN (µg/kg lw)	<11-122	13 (<3-53)	53 (<5-144)
MK (µg/kg lw)	<4	4,2 (<5-24)	74 (<2-238)
MX (µg/kg lw)	<4	15 (<6-84)	30 (<2-150)

8.6. Environmental significance of the observations

The three chemicals studied (OTNE, AC and DE) are pollutants with varying degree of negative environmental properties, such as toxicity to aquatic organisms, persistence and/or bioaccumulation. Especially OTNE and AC have properties indicating that they are bioaccumulative and persistent.

According to the results of the present study and previous studies, the most obvious and generally occurring environmental exposure pathway of these chemicals is from WWTPs to recipients, which is well in line with the exposure index that the National Chemicals Inspectorate has developed (see Table 4). In the WWTP these substances are removed via sorption to sludge or degradation/transformation so that lower levels reach the aquatic environment. This is especially the case for DE where more than 90 % is removed in the WWTP. In the recipient the substances are diluted. However, OTNE and AC are found in surface water.

In Table 18 a risk assessment is made by comparing the maximum measured concentration in surface water of the detected fragrances with the predicted no effect concentration (PNEC). With a limited data set a risk ratio > 0.1 is assumed to indicate a risk. Thus, for mainly OTNE and galaxolide lactone risk for the aquatic environment cannot be excluded. However, the PNEC values are very unsafe since experimental measures are lacking, making it impossible to draw any final conclusions.

The relatively high log K_{ow} value of OTNE and that it is found in biota indicates that this substance may cause secondary poisoning in the recipients.

One exposure pathways not examined in this study and that can be of importance is sludge application on arable land. In a previous study of musk compounds in arable soil several fragrances were found a few weeks after application of sludge (SWECO, 2010). However, after one year no fragrances could be detected.

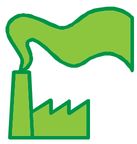
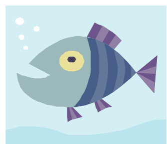
Table 18. Risk assessment of detected musk compounds in surface water. Reference of PNEC were no other is given is COWI, 2011.

Substance	Maximum measured concentration (MEC)	PNEC	MEC/PNEC ratio
OTNE	460	1 000-10 000	0,46 – 0,046
AC	130	<1 000	>0,13
HHCB	620	4 400[1]	0,14
HHCB lactone	4000	100[1]	40
AHTN	32	350[1]	0,09

[1]SWECO, 2010.

9. Conclusions

- OTNE, AC and DE do not seem to be long range transported.
- Diffuse spreading from urban environment via stormwater does not seem to be an important source of these compounds.
- Diffuse spreading via WWTPs seems to be the main source of OTNE, AC and DE to the environment.
- DE is mainly removed or decomposed in the WWTP and were never found in surface water.
- OTNE and AC reach the aquatic environment through WWTP effluents. The removal rate in WWTP is around 70 %. Both substances are found in surface water.
- Industries of personal care products seem to contribute to the spreading of OTNE and AC to the environment via their waste water.
- OTNE can be accumulated in biota and human exposure occurs.
- Human exposure of AC also seems to occur.

Sub-stance					
	Long range transport	**Diffuse emissions**	**Point sources**	**Bioaccumulation**	**Human exposure**
OTNE	No	Yes	Yes	Yes	Yes
AC	No	Yes	Yes	No	Yes?
DE	No	Yes	?	No	No

10. Acknowledgments

The study was funded by the Swedish Environmental Protection Agency together with the Swedish county administrative boards, which are all thanked for good cooperation during the planning of this study. We thank staff members at the waste water treatment plants included in this study for help with sampling waste water and sludge as well as Stockholm Stad, Åfiske, Upplands stiftelsen and NRM for providing with fish samples. We also thank the Swedish county administrative boards for help with sampling.

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11. References

Balk F and Rutten A.L.M., 2000. Fragrance ingredients. Association of river waterworks (RIWA)

Bester K, Klasmeier J and Kupper T., 2008a. Emissions of OTNE (ISO_E-super) – Mass flows in sewage treatment plants. Chemosphere 71, 2003-2010.

Bester K, Hüffmeyer N, Schaub E and Klasmeier J, 2008b. Surface water concentrations of the fragrance compound OTNE in Germany – A comparison between data from measurements and models. Chemosphere 73, 1366-1372.

Chen X, Pauly U, Rehfus S and Bester K, 2009. Personal care compound in a reed bed sludge treatment system. Chemosphere 76, 1094-1101.

COWI, 2011. Litteraturstudie 2011 – Doftämnen, komplexbildande ämnen, urval av föreslagna nya prioriterade ämnen in om WFD. Naturvårdsverket.

Dai L, 2009. Fate and effects of sediment-associated acetyl cedrene in sediments inhabited by the deposit feeder, *Capitella* sp. I. Integrated master thesis, Roskilde University.

DiFrancesco A, Chiu P, Standley L, Allen H and Salvito D, 2004.Dissipation of fragrance materials in sludge amended soils. Environ. Sci. Technol. 38, 194-201.

Harrison E, Oakes S, Hysell M and Hay A, 2006. Sci. Tot. Environm. 367, 481-497.

Kolpin D.W., Furlong E.T., Meyer M.T., Thurman E.M., Zaugg C.D., Barber L. and Buxton H.T., 2002. Pharmaceuticals, hormones, and other organic waste water contaminants in U.S. streams, 1999−2000: A National Reconnaissance. Environ. Sci. Technol., 36, 1202–1211.

Lignell S, Aune M, Darnerud PO and Glynn A, 2004. Kompletterande mätningar av syntetiska myskföreningar i bröstmjölk från förstföderskor i Uppsala 1996-2003. Rapport till Naturvårdsverket.

Lignell S, Darnerud PO, Aune M, Cnattingius S, Hajslova J, Setkova L and Glynn A, 2008. Temporal trends of synthetic musk compounds in mother′s milk and associations with personal use of perfumed products. Environ. Sci. Technol. 42, 6743-6748.

Peck A.M., Linebaugh E.K. and Hornbuckle K.C., 2006. Synthetic musk fragrances in Lake Erie and Lake Ontario sediment cores. Environ. Sci. Technol. 40, 5629-5635.

Peck A.M. and Hornbuckle K.C., 2004. Synthetic musk fragrances in Lake Michigan. Environ. Sci. Technol. 40, 5629-5635.

Reiner J, Wong C, Arcaro K and Kannan K, 2007. Synthetic musk fragrances in human milk from the United States. Environ Sci Technol 41, 3815-3820.

Simonich S, Federle T, Eckhoff W, Rottiers A, Webb S, Sabaliunas D and Wolf W, 2002. Removal of fragrance materials during U.S. and European waste water treatment. Environ. Sci. Technol. 36, 2839-2847.

SLV, 2003. Syntetiska myskföreningar i bröstmjölk och fisk. Resultatrapport till Naturvårdverket, avtals.nr. 219021.

SWECO, 2008. Screening of musk compounds. Project nr: 1270248000.

SWECO, 2010. Screening of musk compounds and metabolites. Project nr: 1270415000.

TemaNord, 2004. Musk compounds in the Nordic environment. TemaNord 2004:503. Nordic Council of Ministers, Copenhagen.

USEPA, 2010. Screening-level hazard characterization – Diphenyl oxide (casnr 101-84-8).

WSP, 2010. Miljöövervakning av miljögifter i urbana områden – sammanställning och analys. Rapport till Naturvårdsverket.

WSP Environmental, 2012-07-06

Ann Helén Österås

John Sternbeck

Appendix 1. Sample details

This appendix shows sample details for all samples. The corresponding analytical results are shown in appendix 2.

Appendix 1.1 Leachate, stormwater and surface water sample details

Sample no	Program	County	Municipality	Media	Site name	SWEREF 99, N	SWEREF 99, E	Category	Sampling date	Sample details
D154	Regional	Södermanland	Eskilstuna	Leachate	Lilla Nyby	6579430	587524	Landfill	2011-09-06	
D156	Regional	Södermanland	Katrineholm	Stormwater	Katrineholm	6539624	569996	URBAN	2011-09-28	huvudledning
D161	Regional	Södermanland	Flen	Stormwater	Flen	6547196	590130	URBAN	2011-09-28	huvudledning
C305	Regional	Uppsala	Uppsala	Stormwater	Libroback	6640771	1600349	URBAN	2011-10-04	
C306	Regional	Uppsala	Uppsala	Stormwater	Bärby	6641071	1600882	URBAN	2011-10-04	
C307	Regional	Uppsala	Uppsala	Stormwater	Industristaden	6638043	1603365	URBAN	2011-10-04	
WSP431	National	Södermanland	Eskilstuna	Stormwater	Eskilstuna	6583283	585485	URBAN	2011-09-21	Large stormwater well at Nordwallsgatan.
WSP432	National	Stockholm	Stockholm	Stormwater	Stockholm, Södermalm			URBAN	2012-06-01	
									2011-09-29	
W08	National	Dalarna	Falun	Surface water	Runn	6707902	1490933	Industry background	2011-09-29	
W16	National	Dalarna	Falun	Surface water	Runn	6718507	1491918	Industry	2011-10-05	
F41	Regional	Jönköping	Gislaved	Surface water	Anderstorpsån	6346442	415224	URBAN	2011-11-21	
F52	Regional	Jönköping	Eksjö	Surface water	Torsjöån	6387131	499681	WWTP REC	2011-11-22	
F491	Regional	Jönköping	Vetlanda	Surface water	Pauliströmsån	6368905	531285	Diffuse	2011-11-22	Paulistföms pappersbruk
F492	Regional	Kalmar	Hultsfred	Surface water	Storgölen	6364673	549171	Landfill	2011-09-26	Hultsfred deponi
I61	Regional	Gotland	Gotland	Surface water	Åminne	6391826	724271	Diffuse	2011-09-06	
D151	Regional	Södermanland	Eskilstuna	Surface water	Ekeby våtmark	6583995	583069	WWTP REC	2011-09-28	
D155	Regional	Södermanland	Katrineholm	Surface water	Djulösjön	6537300	570920	WWTP REC	2011-09-28	
D160	Regional	Södermanland	Flen	Surface water	Gårdsjön	6546818	590520	WWTP REC	2011-09-28	
C301	Regional	Uppsala	Uppsala	Surface water	Vindbron, Fyrisån	6636136	1604114	WWTP REC	2011-10-04	
C308	Regional	Uppsala	Östhammar	Surface water	Krutuddens WWTP	6685572	1643415	WWTP REC	2011-09-28	
C312	Regional	Uppsala	Enköping	Surface water	Enköpings WWTP	6612069	1571592	WWTP REC	2011-12-13	
S331	Regional	Värmland	Årjäng	Surface water	Kyrkbruds WWTP	6584925	335558	WWTP REC	jan-12	spot sample
S335	Regional	Värmland	Storfors	Surface water	Storforsälven	6601914	1412306	WWTP REC	2011-12-14	spot sample
S339	Regional	Värmland	Kristinehamn	Surface water	Kristinehamn WWTP	6577984	1400628	WWTP REC	2012-01-04	

Sample no	Program	County	Municipality	Media	Site name	SWEREF 99, N	SWEREF 99, E	Category	Sampling date	Sample details
S343	Regional	Värmland	Sunne	Surface water	Kolsnäs	6634409	129770	WWTP REC	2011-10-25	spot sample
E375	Regional	Östergötland	Linköping	Surface water	Stångån			WWTP REC	2011-10-25	downstream Nykvarns WWTP
E376	Regional	Östergötland	Linköping	Surface water	Stångån			URBAN	2011-07-12	upstream Nykvarns WWTP
WSP391	National	Västernorrland	Örnsköldsvik	Surface water	Remmarsjön			Background	2011-08-17	
WSP394	National	Örebro	Hällefors	Surface water	Limmingssjön			Background	2011-09-21	
WSP413	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån	6584822	583205	WWTP REC	2011-09-21	Close too E 20, downstream WWTP
WSP414	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån, Torshälla	6587467	583397	WWTP REC	sep-11	I Torshälla
WSP415	National	Stockholm	Stockholm	Surface water	Blockhusudden, 0,5 m			WWTP REC	sep-11	Influence from Stockholm
WSP416	National	Stockholm	Stockholm	Surface water	blockhusudden, 10 m			WWTP REC	2011-09-21	Influence from WWTP
WSP425	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån, Rosenfors	6577910	584015	URBAN background	2011-09-21	
WSP426	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån	6583857	583152	URBAN	sep-11	below bridge, ca 200 m upstream WWTP
WSP427	National	Stockholm	Stockholm	Surface water	Klubbensborg			URBAN background	sep-11	
WSP428	National	Stockholm	Stockholm	Surface water	Årstaviken			URBAN	sep-11	
WSP429	National	Uppsala	Uppsala	Surface water	Fyrisån, central	6641569	644307	URBAN	sep-11	upstream WWTP
WSP430	National	Uppsala	Uppsala	Surface water	Fyrisån, Klastorp	6637143	648594	URBAN background	2011-09-06	

Appendix 1.2 Sediment sample details

Sample no	Program	County	Municipality	Media	Site name	SWEREF 99, N	SWEREF 99, E	Category	Sampling date	Level (cm)
W10	National	Dalarna	Falun	SED	Runn	6707902	1490933	Industry background	2011-09-29	0 - 2
W18	National	Dalarna	Falun	SED	Runn	6718507	1491918	Industry	2011-09-29	0 - 2
F58	Regional	Jönköping	Vetlanda	SED	Vetlandabäcken	6364104	506065	URBAN	2011-11-22	
F60	Regional	Jönköping	Vetlanda	SED	Emån ned Vetlanda, Sjunnendammen	6365959	509961	WWTP REC	2011-11-22	
I63	Regional	Gotland	Gotland	SED	Åminne	6391826	724271	Diffuse	2011-09-26	0 - 2
D152	Regional	Södermanland	Eskilstuna	SED	Ekeby våtmark	6583995	583069	WWTP REC	2011-09-06	
WSP392	National	Västernorrland	Örnsköldsvik	SED	Remmarsjön			Background	2011-07-12	0 - 2
WSP395	National	Örebro	Hällefors	SED	Limmingssjön			Background	2011-08-17	0 - 2
WSP417	National	Södermanland	Eskilstuna	SED	Eskilstunaån	6584822	583205	WWTP REC	2011-09-21	0 - 4
WSP418	National	Södermanland	Eskilstuna	SED	Eskilstunaån, Torshälla	6587467	583397	WWTP REC	2011-09-21	0 - 4
WSP419	National	Stockholm	Stockholm	SED	Klubbensborg			WWTP background	sep-11	
WSP420	National	Stockholm	Stockholm	SED	Saltsjön			WWTP REC	sep-11	
WSP434	National	Stockholm	Stockholm	SED	Årstaviken			URBAN	sep-11	
WSP433	National	Södermanland	Eskilstuna	SED	Eskilstunaån	6583857	583152	URBAN	2011-09-21	0 - 4

Appendix 1.3 WWTP sample details

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Size WWTP (pe)	Load	Active sludge step	Chemical precip.	Sample
W02	Regional	Dalarna	Borlänge	IN WW	Borlänge WWTP	WWTP	2011-10-06	34 000	mix	yes	yes	
W03	Regional	Dalarna	Borlänge	SLUDGE	Borlänge WWTP	WWTP	2011-10-06	34 000	mix	yes	yes	spot sample
W04	Regional	Dalarna	Borlänge	EFF	Borlänge WWTP	WWTP	2011-10-06	34 000	mix	yes	yes	
W06	Regional	Dalarna	Falun	SLUDGE	Främby WWTP	WWTP	2011-10-11	45 000	mix	yes	yes	
W07	Regional	Dalarna	Falun	EFF	Främby WWTP	WWTP	2011-10-11	45 000	mix	yes	yes	
F31	Regional	Jönköping	Jönköping	EFF	Simsholmen WWTP	WWTP	2011-10-10	61700	dom	yes	yes	spot sample
F32	Regional	Jönköping	Jönköping	SLUDGE	Simsholmen WWTP	WWTP	2011-10-10	61700	dom	yes	yes	spot sample
F33	Regional	Jönköping	Jönköping	EFF	Huskvarna WWTP	WWTP	2011-10-11	27543	dom	yes	yes	spot sample
F34	Regional	Jönköping	Jönköping	SLUDGE	Huskvarna WWTP	WWTP	2011-10-11	27543	dom	yes	yes	spot sample
F36	Regional	Jönköping	Jönköping	SLUDGE	Bankeryd WWTP	WWTP	2011-10-26	4143	dom	yes	yes	spot sample
F38	Regional	Jönköping	Jönköping	SLUDGE	Gränna WWTP	WWTP	2011-10-25	4083	dom	yes	yes	spot sample
F55	Regional	Jönköping	Vetlanda	EFF	Vetlanda WWTP	WWTP	2011-11-23	19300	mix	yes	yes	pooled (day)
F56	Regional	Jönköping	Vetlanda	SLUDGE	Vetlanda WWTP	WWTP	2011-11-23	19 300	mix	yes	yes	spot sample
F495	Regional	Jönköping	Gislaved	EFF	Gislaved WWTP	WWTP	2011-10-11	17573	dom	no	yes	pooled
F496	Regional	Kalmar	Hultsfred	EFF	Hultsfred WWTP	WWTP	2011-09-27	8500	mix	yes	yes	pooled
F497	Regional	Jönköping	Eksjö	EFF	Eksjö WWTP	WWTP	2011-09-20	16200	mix	yes	yes	spot sample
F498	Regional	Jönköping	Nässjö	EFF	Nässjö WWTP	WWTP	2011-09-27	17079	dom	no	yes	pooled (day)
F499	Regional	Jönköping	Tranås	EFF	Tranås WWTP	WWTP	2011-10-17	18000	mix	yes	yes	spot sample
I62	Regional	Gotland	Gotland	SLUDGE	Visby AVR	WWTP	2011-09-27	60 000	mix	yes	yes	pooled
G91	Regional	Kronoberg	Alvesta	SLUDGE	Alvesta WWTP	WWTP	2011-12-12	12000	dom	Biobädd	yes	
G95	Regional	Kronoberg	Lessebo	SLUDGE	Lessebo WWTP	WWTP	2011-11-30	9000	mix	Fast bärarmtrl.	yes	
G99	Regional	Kronoberg	Ljungby	SLUDGE	Ljungby WWTP	WWTP	2011-11-22	33000	mix	yes	yes	
G103	Regional	Kronoberg	Markaryd	SLUDGE	Ribersdals WWTP	WWTP	2012-03-05	10000	dom	yes	yes	

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Size WWTP (pe)	Load	Active sludge step	Chemical precip.	Sample
G107	Regional	Kronoberg	Tingsryd	SLUDGE	Tingsryds WWTP	WWTP	2011-11-30	42000	dom	yes	yes	
G110	Regional	Kronoberg	Uppvidinge	SLUDGE	Åseda WWTP	WWTP	2011-12-06	6000	mix	yes	yes	
G113	Regional	Kronoberg	Växjö	SLUDGE	Sundet WWTP	WWTP	2011-11-30	80000	mix	yes	yes	
G117	Regional	Kronoberg	Älmhult	SLUDGE	Älmhult WWTP	WWTP	2011-11-23	22700	dom	yes	yes	
T121	Regional	Örebro	Ljusnarsberg	IN WW	Bångbro WWTP	WWTP	2011-06-29	33 000	ind	yes	yes	spot sample
T122	Regional	Örebro	Ljusnarsberg	EFF	Bångbro WWTP	WWTP	2011-06-29	33 000	ind	yes	yes	spot sample
T123	Regional	Örebro	Ljusnarsberg	SLUDGE	Bångbro WWTP	WWTP	2011-07-08	33 000	ind	yes	yes	spot sample
T124	Regional	Örebro	Nora	IN WW	Nora WWTP	WWTP	2011-06-29	8 500	dom	yes	yes	spot sample
T125	Regional	Örebro	Nora	EFF	Nora WWTP	WWTP	2011-06-29	8 500	dom	yes	yes	spot sample
T126	Regional	Örebro	Nora	SLUDGE	Nora WWTP	WWTP	2011-07-08	8 500	dom	yes	yes	spot sample
T127	Regional	Örebro	Ljusnarsberg	IN WW	Bångbro WWTP	WWTP	2011-10-27	33 000	ind	yes	yes	spot sample
T128	Regional	Örebro	Ljusnarsberg	EFF	Bångbro WWTP	WWTP	2011-10-27	33 000	ind	yes	yes	spot sample
T129	Regional	Örebro	Ljusnarsberg	SLUDGE	Bångbro WWTP	WWTP	2011-10-27	33 000	ind	yes	yes	spot sample
T130	Regional	Örebro	Nora	IN WW	Nora WWTP	WWTP	2011-10-27	8 500	dom	yes	yes	spot sample
T131	Regional	Örebro	Nora	EFF	Nora WWTP	WWTP	2011-10-27	8 500	dom	yes	yes	spot sample
T132	Regional	Örebro	Nora	SLUDGE	Nora WWTP	WWTP	2011-10-27	8 500	dom	yes	yes	spot sample
D157	Regional	Södermanland	Katrineholm	EFF	Rosenholms WWTP	WWTP	2011-10-11	53000	mix	no	yes	
D158	Regional	Södermanland	Katrineholm	SLUDGE	Rosenholms WWTP	WWTP	2011-10-11	53000	mix	no	yes	
D162	Regional	Södermanland	Flen	EFF	Flens WWTP	WWTP	2011-10-11	19900	mix	yes	yes	
D163	Regional	Södermanland	Flen	SLUDGE	Flens WWTP	WWTP	2011-10-11	19900	mix	yes	yes	
D165	Regional	Södermanland	Vingåker	SLUDGE	Vingåkers WWTP	WWTP	2011-10-11	9600	mix	no	yes	
Y181	Regional	Västernorrland	Sundsvall	EFF	Fillanverket	WWTP	2011-10-19	21600	mix	yes	yes	spot sample
Y182	Regional	Västernorrland	Sundsvall	EFF	Tivoliverket	WWTP	2011-10-19	53000	mix	yes	yes	spot sample
Y183	Regional	Västernorrland	Örnsköldsvik	EFF	Knorthems WWTP	WWTP	2011-10-04	12500	dom	yes	yes	pooled (day)

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Size WWTP (pe)	Load	Active sludge step	Chemical precip.	Sample
Y184	Regional	Västernorrland	Sollefteå	EFF	Hågesta WWTP	WWTP	2011-09-01	13150	mix	no	yes	pooled (day)
BD211	Regional	Norrbotten	Luleå	EFF	Uddebo WWTP	WWTP	2012-02-09	ca 60 000 personer	dom	no (moving bed)	yes	spot sample
BD212	Regional	Norrbotten	Luleå	SLUDGE	Uddebo WWTP	WWTP	2012-02-09	ca 60 000 personer	dom	no (moving bed)	yes	spot sample
BD213	Regional	Norrbotten	Piteå	EFF	Sandholmens WWTP	WWTP	2011-10-28	ca 30 500 personer.	dom	no	yes	pooled
BD214	Regional	Norrbotten	Piteå	SLUDGE	Sandholmens WWTP	WWTP	2011-10-28	ca 30 500 personer.	dom	no	yes	pooled
BD215	Regional	Norrbotten	Gällivare	EFF	Kavahedens WWTP	WWTP		Ca 18 000-20 000 personer.	mix	biobädd	yes	spot sample
BD216	Regional	Norrbotten	Gällivare	SLUDGE	Kavahedens WWTP	WWTP		ca 18 000-20 000 personer.	mix	biobädd	yes	spot sample
M271	Regional	Skåne	Helsingborg	IN WW	Öresundsverket	WWTP	2011-12-13	200000	dom	yes	no	pooled (week)
M272	Regional	Skåne	Helsingborg	EFF	Öresundsverket	WWTP	2011-12-13	200000	dom	yes	no	pooled (week)
M273	Regional	Skåne	Helsingborg	SLUDGE	Öresundsverket	WWTP	2011-12-13	200000	dom	yes	no	pooled
C302	Regional	Uppsala	Uppsala	IN WW	Kungsängsverket	WWTP	2011-09-28	200 000	dom	yes	yes	
C303	Regional	Uppsala	Uppsala	EFF	Kungsängsverket	WWTP	2011-09-28	200 000	dom	yes	yes	
C304	Regional	Uppsala	Uppsala	SLUDGE	Kungsängsverket	WWTP	2011-09-28	200 000	dom	yes	yes	
C309	Regional	Uppsala	Östhammar	IN WW	Krutuddens WWTP	WWTP	2011-10-04	4700	dom	Förfällning samt bioreaktor med bärarmaterial	yes	
C310	Regional	Uppsala	Östhammar	EFF	Krutuddens WWTP	WWTP	2011-10-04	4700 pe	dom	Förfällning samt bioreaktor med bärarmaterial	yes	
C311	Regional	Uppsala	Östhammar	SLUDGE	Krutuddens WWTP	WWTP	2011-10-04	4700	dom	Förfällning samt bioreaktor med bärarmaterial	yes	
C313	Regional	Uppsala	Enköping	IN WW	Enköpings WWTP	WWTP	2011-09-28	30 000	mix	yes	yes	
C314	Regional	Uppsala	Enköping	EFF	Enköpings WWTP	WWTP	2011-09-28	30 000	mix	yes	yes	

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Size WWTP (pe)	Load	Active sludge step	Chemical precip.	Sample
S332	Regional	Värmland	Årjäng	IN WW	Kyrkbruds RV	WWTP	2011-12-13	5000	dom	yes	yes	spot sample
S333	Regional	Värmland	Årjäng	EFF	Kyrkbruds RV	WWTP	2011-12-13	5000	dom	yes	yes	spot sample
S334	Regional	Värmland	Årjäng	SLUDGE	Kyrkbruds RV	WWTP	2011-12-13	5000	dom	yes	yes	spot sample
S336	Regional	Värmland	Storfors	IN WW	Storfors WWTP	WWTP	jan-12	4500	dom	no	yes	spot sample
S337	Regional	Värmland	Storfors	EFF	Storfors WWTP	WWTP	jan-12	4500	dom	no	yes	spot sample
S338	Regional	Värmland	Storfors	SLUDGE	Storfors WWTP	WWTP	jan-12	4500	dom	no	yes	spot sample
S340	Regional	Värmland	Kristinehamn	IN WW	Kristinehamn WWTP	WWTP	2011-12-14	12081	mix	yes	yes	
S341	Regional	Värmland	Kristinehamn	EFF	Kristinehamn WWTP	WWTP	2011-12-14	12081	mix	yes	yes	
S342	Regional	Värmland	Kristinehamn	SLUDGE	Kristinehamn WWTP	WWTP	2011-12-14	12081	mix	yes	yes	spot sample
S344	Regional	Värmland	Sunne	SLUDGE	Sunne WWTP	WWTP	2012-01-04	7500	dom	no	yes	spot sample
S345	Regional	Värmland	Sunne	EFF	Sunne WWTP	WWTP	2012-01-04	7500	dom	no	yes	spot sample
S346	Regional	Värmland	Sunne	IN WW	Sunne WWTP	WWTP	2012-01-04	7500	dom	no	yes	spot sample
E361	Regional	Östergötland	Åtvidaberg	EFF	Håckla WWTP	WWTP	2011-09-26	7700	dom	no	yes	pooled (day)
E362	Regional	Östergötland	Åtvidaberg	SLUDGE	Håckla WWTP	WWTP		7700	dom	no	yes	spot sample
E363	Regional	Östergötland	Mjölby	EFF	Gudhem WWTP	WWTP	2011-10-19	6 000	dom	yes	yes	
E364	Regional	Östergötland	Mjölby	SLUDGE	Gudhem WWTP	WWTP	2011-10-19	6 000	dom	yes	yes	
E365	Regional	Östergötland	Mjölby	EFF	Mjölkulla WWTP	WWTP	2011-10-19	55 000	dom	yes	yes	
E366	Regional	Östergötland	Mjölby	SLUDGE	Mjölkulla WWTP	WWTP	2011-10-19	55 000	dom	yes	yes	
E367	Regional	Östergötland	Motala	EFF	Karshult WWTP	WWTP	10-16 okt 2011	40 000	dom	yes	yes	pooled (day)
E368	Regional	Östergötland	Motala	SLUDGE	Karshult WWTP	WWTP	2011-10-16	40 000	dom	yes	yes	spot sample
E369	Regional	Östergötland	Vadstena	EFF	Vadstena WWTP	WWTP	10-16 okt 2011	9 500	dom	no	yes	pooled (day)
E370	Regional	Östergötland	Vadstena	SLUDGE	Vadstena WWTP	WWTP	2011-10-16	9 500	dom	no	yes	spot sample
E371	Regional	Östergötland	Norrköping	EFF	Slottshagens WWTP	WWTP	2011-10-12	200 000	dom	yes	yes	
E372	Regional	Östergötland	Norrköping	SLUDGE	Slottshagens WWTP	WWTP		200 000	dom	yes	yes	

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Size WWTP (pe)	Load	Active sludge step	Chemical precip.	Sample
E373	Regional	Östergötland	Linköping	EFF	Nykvarns WWTP	WWTP	2011-10-25	235 000	dom	yes	yes	pooled (day)
E374	Regional	Östergötland	Linköping	SLUDGE	Nykvarns WWTP	WWTP	2011-10-25	235 000	dom	yes	yes	spot sample
WSP399	National	Södermanland	Eskilstuna	IN WW	Ekeby	WWTP	2011-06-28	94000	dom	yes	yes	
WSP400	National	Södermanland	Eskilstuna	EFF	Ekeby	WWTP	2011-06-28	94000	dom	yes	yes	
WSP401	National	Södermanland	Eskilstuna	SLUDGE	Ekeby WWTP	WWTP	2011-06-28	94000	dom	yes	yes	
WSP402	National	Stockholm	Stockholm	IN WW	Henriksdal	WWTP	2011-07-26	ca 700 000	dom	yes	yes	spot sample
WSP403	National	Stockholm	Stockholm	EFF	Henriksdal	WWTP	2011-07-26	ca 700 000	dom	yes	yes	
WSP404	National	Stockholm	Stockholm	SLUDGE	Henriksdal WWTP	WWTP	2011-07-26	ca 700 000	dom	yes	yes	spot sample
WSP405	National	Södermanland	Eskilstuna	IN WW	Ekeby	WWTP	sep-11	94000	dom	yes	yes	
WSP406	National	Södermanland	Eskilstuna	EFF	Ekeby	WWTP	sep-11	94000	dom	yes	yes	
WSP407	National	Södermanland	Eskilstuna	SLUDGE	Ekeby WWTP	WWTP	sep-11	94000	dom	yes	yes	
WSP408	National	Stockholm	Stockholm	IN WW	Henriksdal	WWTP	2011-09-19	ca 700 000	dom	yes	yes	spot sample
WSP409	National	Stockholm	Stockholm	EFF	Henriksdal	WWTP	2011-09-19	ca 700 000	dom	yes	yes	
WSP410	National	Stockholm	Stockholm	SLUDGE	Henriksdal WWTP	WWTP	2011-09-19	ca 700 000	dom	yes	yes	spot sample

Appendix 1.4 Biota and breast milk sample details

Sample no	Program	County	Municipality	Media	Site name	Category	Sampling date	Species	Number in pooled sample	Length (cm)	Fresh weight (g)
W09	National	Dalarna	Falun	BIOTA	Runn	Industry background	2011-09-30	Perch *(perca fluvialis)*	12	16.2 - 20.2	total = 784
W17	National	Dalarna	Falun	BIOTA	Runn	Industry	2011-09-23	Perch *(perca fluvialis)*	12	17.2 - 20.4	total = 784
D153	Regional	Södermanland	Eskilstuna	BIOTA	Ekeby våtmark	WWTP REC	2011-09-07	Perch *(perca fluvialis)*			
D159	Regional	Södermanland	Katrineholm	BIOTA	Djulösjön	WWTP REC	2011-10-18	Perch *(perca fluvialis)*	2	20 resp 16.3	mean = 155
D164	Regional	Södermanland	Flen	BIOTA	Gårdsjön	WWTP REC	2011-09-21	Perch *(perca fluvialis)*			
Y185	Regional	Västernorrland	Härnösand	BIOTA	Kattastrand	WWTP REC	2011-10-26	Fourhorn sculpin (*Myoxocephalus quadricornis*)	5	mean = 24.2	total = 855
WSP393	National	Västernorrland	Örnsköldsvik	BIOTA	Remmarsjön	Background	2011-10-13	Perch *(perca fluvialis)*	10	mean = 20.1	mean = 78.2
WSP396	National	Örebro	Hällefors	BIOTA	Limmingssjön	Background	2011-08-31	Perch *(perca fluvialis)*	10	mean = 19.2	mean = 71.0
WSP421	National	Södermanland	Eskilstuna	BIOTA	Eskilstunaån	WWTP REC	sep-11	Perch *(perca fluvialis)*	5	mean = 22.9	mean = 166
WSP422	National	Södermanland	Eskilstuna	BIOTA	Eskilstunaån	WWTP REC	sep-11	Perch *(perca fluvialis)*	5	mean = 25.3	mean = 234
WSP423	National	Stockholm	Stockholm	BIOTA	Saltsjön, Djur-gårds-brunnsviken	WWTP REC	2011-08-27	Perch *(perca fluvialis)*			
WSP435	National	Uppsala	Uppsala	BIOTA	Fyrisån	Urban	2011-09-01	Perch *(perca fluvialis)*			
WSP436	National	Stockholm	Uppsala	BIOTA	Årstaviken	Urban	2011-10-23	Perch *(perca fluvialis)*			
WSP443	National	Stockholm	Stockholm	Breast milk	Stockholm	Urban	2009				
WSP444	National	Stockholm	Stockholm	Breast milk	Stockholm	Urban	2009				
WSP445	National	Stockholm	Stockholm	Breast milk	Stockholm	Urban	2009				
WSP446	National	Stockholm	Stockholm	Breast milk	Stockholm	Urban	2009				
WSP447	National	Västra Götaland	Göteborg	Breast milk	Göteborg	Urban	2010				
WSP448	National	Västra Götaland	Göteborg	Breast milk	Göteborg	Urban	2010				
WSP449	National	Västra Götaland	Göteborg	Breast milk	Göteborg	Urban	2010				

Appendix 2. Analytical data.

Table A1. Concentrations in waters (all data in ng/l).

Sample no	Media	Category	Sampling depth (m)	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
D154	Leachate	Landfill		<150	<50	<50	<75	<75	<50	<50	<250
D156	Stormwater	URBAN		<10	<5.0	<10	<5.0	<5.0	<3.0	<3.0	<25
D161	Stormwater	URBAN		<10	<5.0	<10	<5.0	<5.0	<3.0	<3.0	<25
C305	Stormwater	URBAN		<20	<5.0	<10	<5.0	<5.0	<2.0	<2.0	<20
C306	Stormwater	URBAN		<20	<5.0	<10	<5.0	<5.0	<2.0	<2.0	<20
C307	Stormwater	URBAN		<10	<2.0	<2.0	<3.0	<3.0	<2.0	<2.0	<10
WSP431	Stormwater	URBAN		<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	<20
WSP432	Stormwater	URBAN		<20	<10	<5.0	<5.0	<2.0	<2.0	<2.0	140
W08	Surface water	Industry background	0.5	<20	<2.0	<5.0	<4.0	<4.0	<2.0	<2.0	<20
W16	Surface water	Industry	0.5	<10	<5.0	<3.0	<3.0	<3.0	<2.0	<2.0	<10
F41	Surface water	URBAN	0.5	<10	<5.0	<5.0	<3.0	<3.0	<2.0	<2.0	<20
F52	Surface water	WWTP REC	0.3	<25	<2.0	<5.0	17	<5.0	<2.0	<2.0	67
F491	Surface water	Diffuse	0.5	<25	<2.0	<5.0	<4.0	<4.0	<2.0	<2.0	<15
F492	Surface water	Landfill	0.5	<25	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<15
I61	Surface water	Diffuse	0.2-0.4	<10	<1.0	<2.0	<2.0	<2.0	<2.0	<2.0	<5.0
D151	Surface water	WWTP REC		290	<5.0	130	620	<5.0	<5.0	<5.0	4000
D155	Surface water	WWTP REC	0.5	34	<2.0	<5.0	29	3.6	<2.0	<2.0	110
D160	Surface water	WWTP REC	0.5	14	<5.0	<5.0	19	<5.0	<3.0	<3.0	100
C301	Surface water	WWTP REC	0.5	280	<5.0	<5.0	260	24	<3.0	<3.0	860
C308	Surface water	WWTP REC	0.5	<10	<3.0	<3.0	<3.0	<3.0	<2.0	<2.0	<10
C312	Surface water	WWTP REC	0.5	290	<5.0	46	120	<5.0	<3.0	<3.0	250
S331	Surface water	WWTP REC	0	<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	56
S335	Surface water	WWTP REC	0.95	220	<5.0	<5.0	58	12	<2.0	<2.0	61
S339	Surface water	WWTP REC		<20	<2.0	<5.0	18	<5.0	<2.0	<2.0	95
S343	Surface water	WWTP REC	0.3	<20	<2.0	<5.0	<10	<5.0	<2.0	<2.0	<20
E375	Surface water	WWTP REC		100	<5.0	<10	70	10	<2.0	<2.0	230
E376	Surface water	URBAN		<25	<5.0	<10	<5.0	<5.0	<2.0	<2.0	<15

Sample no	Media	Category	Sampling depth (m)	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
WSP391	Surface water	Background	0.1	<5.0	<1.0	<1.0	<2.0	<2.0	<1.0	<1.0	<2.0
WSP394	Surface water	Background	0.5	<3.0	<3.0	<3.0	<3.0	<3.0	<3.0	<3.0	<3.0
WSP413	Surface water	WWTP REC	0.5	<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	74
WSP414	Surface water	WWTP REC	0.5	<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	51
WSP415	Surface water	WWTP REC	0,5	<10	<2.0	<2.0	9.7	<2.0	<2.0	<2.0	42
WSP416	Surface water	WWTP REC	10	<10	<2.0	<2.0	20	2	<2.0	<2.0	94
WSP425	Surface water	URBAN background	0.5	<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	<10
WSP426	Surface water	URBAN	0.5	<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	<10
WSP427	Surface water	URBAN background		<10	<5.0	<10	<5.0	<5.0	<2.0	<2.0	17
WSP428	Surface water	URBAN		<20	<5.0	<10	<5.0	<5.0	<2.0	<2.0	23
WSP429	Surface water	URBAN		460	<2.0	42	330	32	<2.0	<2.0	1100
WSP430	Surface water	URBAN background		<10	<2.0	<2.0	<5.0	<5.0	<2.0	<2.0	<10

Table A2. Concentrations in sediment (all data in µg/kg dry weight).

Sample no	Media	Category	Level (cm)	TS, %	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
W10	SED	Industry back-ground	0 - 2	12	<85	<17	<42	<25	<17	<17	<17	<42
W18	SED	Industry	0 - 2	7,6	<130	<30	<70	<40	<30	<30	<30	<70
F58	SED	URBAN		19,2	<130	<11	<110	<27	<27	<11	<11	<80
F60	SED	WWTP REC		12,7	<200	<16	<80	<40	<40	<16	<16	<80
I63	SED	Diffuse	0 - 2	45,2	<20	<10	<10	<10	<10	<5.0	<5.0	<25
D152	SED	WWTP REC		55,8	<40	<20	<20	32	<20	<10	<10	130
WSP417	SED	WWTP REC	0 - 4	34,5	<60	<30	<30	<30	<30	<15	<15	utgår
WSP418	SED	WWTP REC	0 - 4	29,7	<60	<30	<30	<30	<30	<15	<15	<75
WSP419	SED	WWTP back-ground		6,2	<160	<50	<80	<80	<80	<35	<35	<330
WSP420	SED	WWTP REC		8,4	<10	<2.0	<10	<5.0	<5.0	<2.0	<2.0	<10
WSP392	SED	Background	0 - 2	8,2	<120	<25	<61	<25	<25	<25	<25	<250
WSP395	SED	Background	0 - 2	6,6	<150	<30	<76	<31	<31	<31	<3.0	<300
WSP434	SED	URBAN		10,1	<100	<30	<50	<50	<50	<20	<20	<200
WSP433	SED	URBAN	0 - 4	34,8	<60	<30	<30	<30	<30	<15	<15	<75

Table A3. Concentrations in incoming waste water (IN WW) and effluents (EFF), unit ng/l.

Sample no	Media	Category	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
W02	IN WW	WWTP	3800	28	420	1900	190	<5.0	<5.0	1700
T121	IN WW	WWTP	3700	7.1	560	1900	110	<10	<10	360
T124	IN WW	WWTP	3500	37	980	3100	150	<10	<10	690
T127	IN WW	WWTP	1800	19	150	760	94	<5.0	<5.0	720
T130	IN WW	WWTP	3800	17	340	1800	160	<5.0	<5.0	1400
M271	IN WW	WWTP	5100	34	720	2000	160	2.4	<2.0	1700
C302	IN WW	WWTP	8700	76	1300	3900	470	<20	<20	2400
C309	IN WW	WWTP	11000	41	1600	4100	450	<10	<10	2700
C313	IN WW	WWTP	6700	66	720	2900	420	<5.0	<5.0	1200
S332	IN WW	WWTP	2600	25	330	1000	65	3.6	<2.0	1100
S336	IN WW	WWTP	3600	<30	450	1300	100	7	<3.0	1900
S340	IN WW	WWTP	1800	13	230	1100	83	5.4	<2.0	710
S346	IN WW	WWTP	2300	10	300	1400	77	4.8	<2.0	1600
WSP399	IN WW	WWTP	3900	89	880	2400	160	<10	<10	190
WSP402	IN WW	WWTP	6300	100	2500	4200	560	<10	18	560
WSP405	IN WW	WWTP	3600	44	360	1400	87	<5.0	<5.0	750
WSP408	IN WW	WWTP	27000	380	5800	16000	2000	64	<10	13
W04	EFF	WWTP	1000	<5.0	180	510	67	11	<3.0	1600
W07	EFF	WWTP	2100	<5.0	280	610	68	4.6	<3.0	1100
F31	EFF	WWTP	840	<5.0	150	420	39	4.6	<2.0	1400
F33	EFF	WWTP	1100	<5.0	160	660	68	9	<2.0	1800
F55	EFF	WWTP	130	<2.0	<5.0	200	9	<2.0	<2.0	1900
T122	EFF	WWTP	900	3.2	170	750	51	<5.0	<5.0	<5.0
T125	EFF	WWTP	460	1.6	31	340	20	<5.0	<5.0	56
T128	EFF	WWTP	290	<2.0	30	190	15	<2.0	<2.0	1000
T131	EFF	WWTP	1000	<5.0	240	630	80	<2.0	9.2	1800
D157	EFF	WWTP	1600	<5.0	130	810	100	4.6	<3.0	2000
D162	EFF	WWTP	820	<5.0	130	490	31	7.1	<3.0	1900
Y181	EFF	WWTP	1900	<5.0	220	650	43	3.5	<3.0	1200
Y182	EFF	WWTP	2000	<10	330	620	60	5	<2.0	1100
Y183	EFF	WWTP	940	<5.0	120	280	26	3.7	<3.0	1100

Sample no	Media	Category	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
Y184	EFF	WWTP	2000	<5.0	310	1100	55	<5.0	<5.0	2500
BD211	EFF	WWTP	2000	6.1	240	550	43	3.5	<2.0	810
BD213	EFF	WWTP	1000	2.9	90	500	29	5.1	<2.0	1200
BD215	EFF	WWTP	1800	7.4	150	610	17	5.5	<2.0	720
M272	EFF	WWTP	630	<2.0	<5.0	360	24	5	<2.0	1900
C303	EFF	WWTP	770	<10	<40	650	30	5.6	<5.0	3200
C310	EFF	WWTP	1200	<5.0	140	330	31	5.2	<2.0	1100
C314	EFF	WWTP	4900	<5.0	260	1100	68	11	<5.0	4800
S333	EFF	WWTP	2800	12	310	910	38	6.1	<2.0	1200
S337	EFF	WWTP	1600	<15	210	490	42	6.3	<2.0	1800
S341	EFF	WWTP	640	<3.0	<5.0	730	73	2.6	<2.0	1700
S345	EFF	WWTP	2100	-------	200	630	40	4.1	<2.0	1200
E361	EFF	WWTP	720	<5.0	84	400	58	<2.0	<2.0	1300
E363	EFF	WWTP	360	<5.0	77	410	36	<2.0	6.4	2100
E365	EFF	WWTP	910	<5.0	77	450	38	3.7	<2.0	1700
E367	EFF	WWTP	1200	<5.0	110	530	40	3.5	<3.0	3600
E369	EFF	WWTP	1000	<5.0	100	380	38	4.8	<3.0	1700
E371	EFF	WWTP	660	<5.0	86	550	36	4.3	<2.0	2200
E373	EFF	WWTP	750	<10	65	470	38	4	<2.0	1600
WSP400	EFF	WWTP	950	5.7	390	980	77	<5.0	<5.0	2200
WSP403	EFF	WWTP	890	<5.0	<5.0	900	78	<5.0	7.3	3500
WSP406	EFF	WWTP	1200	<5.0	120	800	76	7.4	<2.0	2000
WSP409	EFF	WWTP	680	<2.0	97	760	82	11	<2.0	2.8
F495	EFF	WWTP	1000	<5.0	110	430	41	4.2	<2.0	1700
F496	EFF	WWTP	2200	<10	240	620	42	<5.0	<5.0	3300
F497	EFF	WWTP	960	<5.0	180	390	32	<5.0	<5.0	680
F498	EFF	WWTP	2100	<10	310	970	86	5.9	<5.0	3100
F499	EFF	WWTP	550	<5.0	100	550	42	2.8	<2.0	1800

Table A4. Concentrations in sludge. unit µg/kg dry weight.

Sample no	Media	Category	TS. %	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
W03	SLUDGE	WWTP	33.5	7800	8.4	1200	5700	300	<6.0	<6.0	1500
W06	SLUDGE	WWTP	29.1	23000	20	3400	6200	620	62	<5.0	3100
F32	SLUDGE	WWTP	29.2	10000	<5.0	1400	4100	410	<5.0	<5.0	4100
F34	SLUDGE	WWTP	27.5	10000	<3.0	1500	5800	510	<3.0	<3.0	4700
F36	SLUDGE	WWTP	29.5	12000	92	1700	4400	370	<5.0	<5.0	2600
F38	SLUDGE	WWTP	15.9	3600	<5.0	520	2800	130	<3.0	<3.0	2100
F56	SLUDGE	WWTP	19.2	2100	<2.0	120	2300	320	<2.0	<2.0	3300
I62	SLUDGE	WWTP	19.7	12000	<20	1600	7100	450	<20	<20	2800
G91	SLUDGE	WWTP	33	7000	24	420	4800	300	<15	<15	2300
G95	SLUDGE	WWTP	14.3	3200	<35	180	2800	390	<35	<35	3700
G99	SLUDGE	WWTP	16.1	22000	<10	930	9900	930	<2.0	<2.0	5800
G103	SLUDGE	WWTP	14.6	12000	<35	580	6700	420	<14	<14	2700
G107	SLUDGE	WWTP	16.4	4600	<31	460	2400	220	<31	<31	2000
G110	SLUDGE	WWTP	16.4	2700	<30	270	3000	210	<30	<30	3500
G113	SLUDGE	WWTP	5.9	7800	<85	1700	7600	610	<85	<85	3700
G117	SLUDGE	WWTP	1.5	13000	<140	<1000	11000	1300	<70	<70	8000
T123	SLUDGE	WWTP	6.2	5000	<33	1400	7300	420	<81	<81	<810
T126	SLUDGE	WWTP	10.4	1900	<20	250	960	83	<20	<20	820
T129	SLUDGE	WWTP	6.1	640	<85	<170	970	<85	<35	<35	1600
T132	SLUDGE	WWTP	3.6	10000	<140	1200	9200	830	<140	<140	5000
D158	SLUDGE	WWTP	6.6	12000	<5.0	1100	6500	530	<5.0	<5.0	2000
D163	SLUDGE	WWTP	12.1	2000	<2.0	<2.0	1900	290	27	<2.0	4000
D165	SLUDGE	WWTP	2	14000	120	2300	8500	1400	<2.0	<2.0	3200
BD212	SLUDGE	WWTP	21.7	17000	74	2300	8300	510	<24	<24	2800
BD214	SLUDGE	WWTP	3.3	11000	<5.0	<10	6700	820	<3.0	<3.0	2800
BD216	SLUDGE	WWTP	15.5	4300	<5.0	390	3100	230	<3.0	<3.0	1100
M273	SLUDGE	WWTP	22.2	2700	34	1000	5900	680	<25	<25	5900
C304	SLUDGE	WWTP	22.9	9200	<20	1100	6100	610	<20	<20	2700
C311	SLUDGE	WWTP	25.2	12000	<3.0	1700	5200	290	<3.0	<3.0	3800
S334	SLUDGE	WWTP	21	12000	45	1600	6200	420	<25	<25	2800
S338	SLUDGE	WWTP	18.4	5400	<27	540	3000	330	<27	<27	1800
S342	SLUDGE	WWTP	16.4	4500	46	1200	7300	790	<31	<31	4900
S344	SLUDGE	WWTP	25.8	12000	27	1400	5800	390	<20	<20	5000

Sample no	Media	Category	TS. %	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
E362	SLUDGE	WWTP	34.8	12000	<5.0	1600	4300	460	<5.0	<5.0	4300
E364	SLUDGE	WWTP	16.4	8500	67	1200	4700	310	<9.0	<9.0	850
E366	SLUDGE	WWTP	14.3	9800	<3.0	2300	9100	500	<20	<20	5000
E368	SLUDGE	WWTP	28	11000	28	1600	7500	280	<10	<10	5400
E370	SLUDGE	WWTP	16.8	1200	18	890	3300	360	<18	<18	8900
E372	SLUDGE	WWTP	27.2	1900	77	1800	4400	810	<11	<11	14000
E374	SLUDGE	WWTP	28.2	1600	<18	1900	2400	850	<8.0	<8.0	12000
WSP401	SLUDGE	WWTP	6.7	15000	270	3000	10000	670	<75	<75	-------
WSP404	SLUDGE	WWTP	28.9	3800	140	2500	5900	1400	<7.0	<7.0	14000
WSP407	SLUDGE	WWTP	20.3	11000	54	1300	6400	340	<15	<15	3900
WSP410	SLUDGE	WWTP	25.8	11000	85	1700	9300	1000	<80	<80	2900

Table A5. Concentrations in aquatic biota and breast milk (all data in µg/kg wet weight).

Sample no	Media	Category	fat, % fv.	OTNE	DE	AC	HHCB	AHTN	MK	MX	HHCB LAC
W09	BIOTA	Industry background		<20	<10	<10	<5.0	<5.0	<5.0	<5.0	<25
W17	BIOTA	Industry		<20	<10	<10	<5.0	<5.0	<5.0	<5.0	<25
D153	BIOTA	WWTP REC		<20	<10	<10	25	<5.0	<5.0	<5.0	110
D159	BIOTA	WWTP REC		<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<15
D164	BIOTA	WWTP REC		<20	<10	<10	<5.0	<5.0	<5.0	<5.0	<25
Y185	BIOTA	WWTP REC		46	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	18
WSP393	BIOTA	Background	0.3	<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<10
WSP396	BIOTA	Background	0.21	<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<10
WSP421	BIOTA	WWTP REC		<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<15
WSP422	BIOTA	WWTP REC		<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<15
WSP423	BIOTA	WWTP REC	0.86	<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<10
WSP435	BIOTA	Urban		<20	<10	<10	<5.0	<5.0	<5.0	<5.0	<25
WSP436	BIOTA	Urban	0.59	<20	<2.0	<5.0	<5.0	<5.0	<2.0	<2.0	<10
WSP443	Breast milk	Urban		<1.5	<0.50	<0.50	<0.50	<0.50	<0.20	<0.20	<1.5
WSP444	Breast milk	Urban		2.1	<0.50	<0.50	3	<0.50	<0.20	<0.20	3.8
WSP445	Breast milk	Urban		<1.5	<0.50	<0.50	1	1.4	<0.20	<0.20	9
WSP446	Breast milk	Urban		<1.5	<0.50	<0.50	3.2	5.6	<0.20	<0.20	3.3
WSP447	Breast milk	Urban		<1.5	<0.50	<0.50	<0.50	<0.50	<0.20	<0.20	1.8
WSP448	Breast milk	Urban		1.9	<0.50	0.5	0.9	<0.50	<0.20	<0.20	2
WSP449	Breast milk	Urban		<1.5	<0.50	<0.50	<0.50	<0.50	<0.20	<0.20	<1.5

Appendix 3. Levels of OTNE, AC and DE in different WWTPs

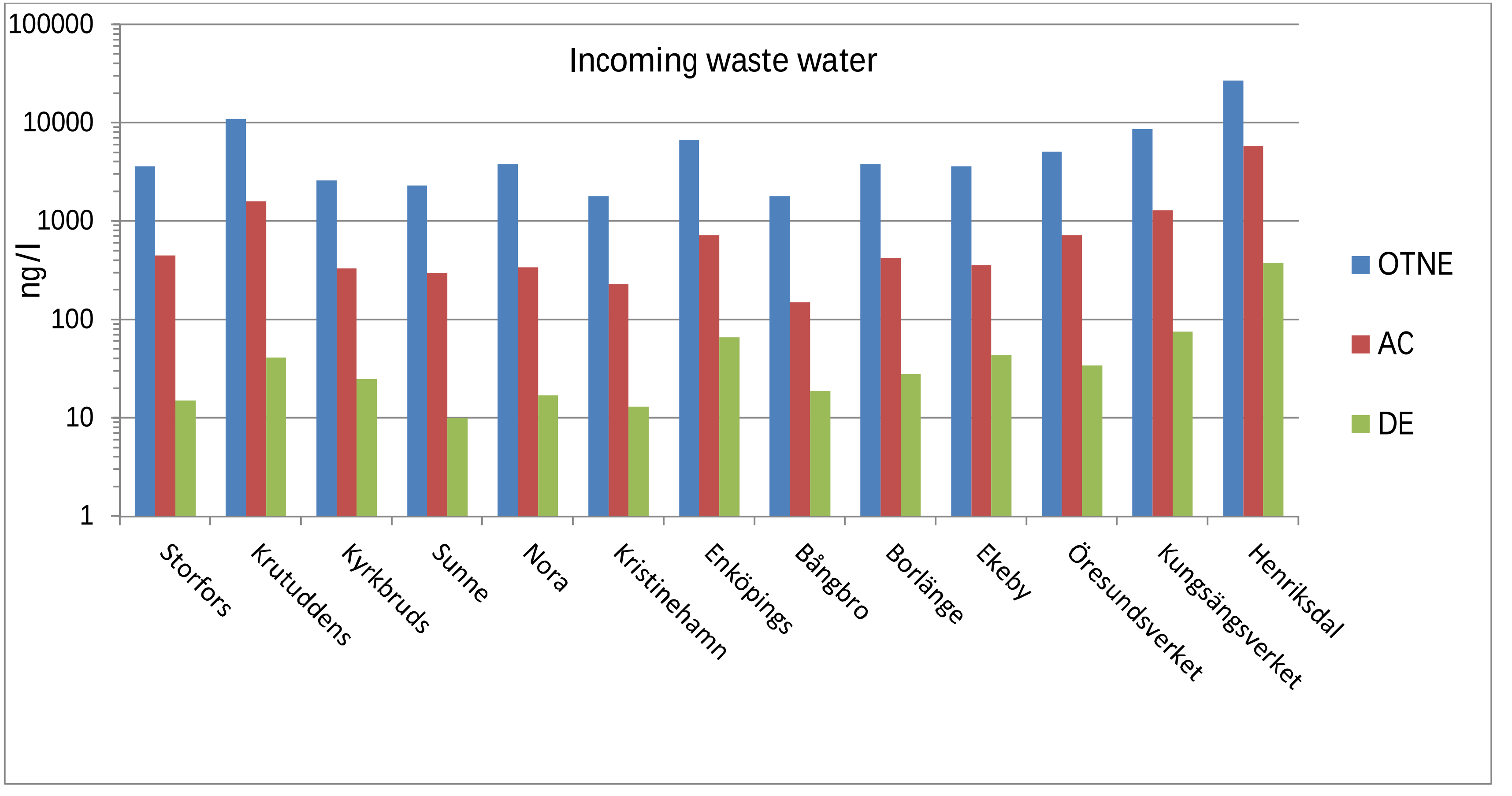

Figure 12. Levels of OTNE, AC and DE in incoming waste water of 13 different WWTP:s arranged from smallest to largest in size (pe). Note that the scale is logarithmic.

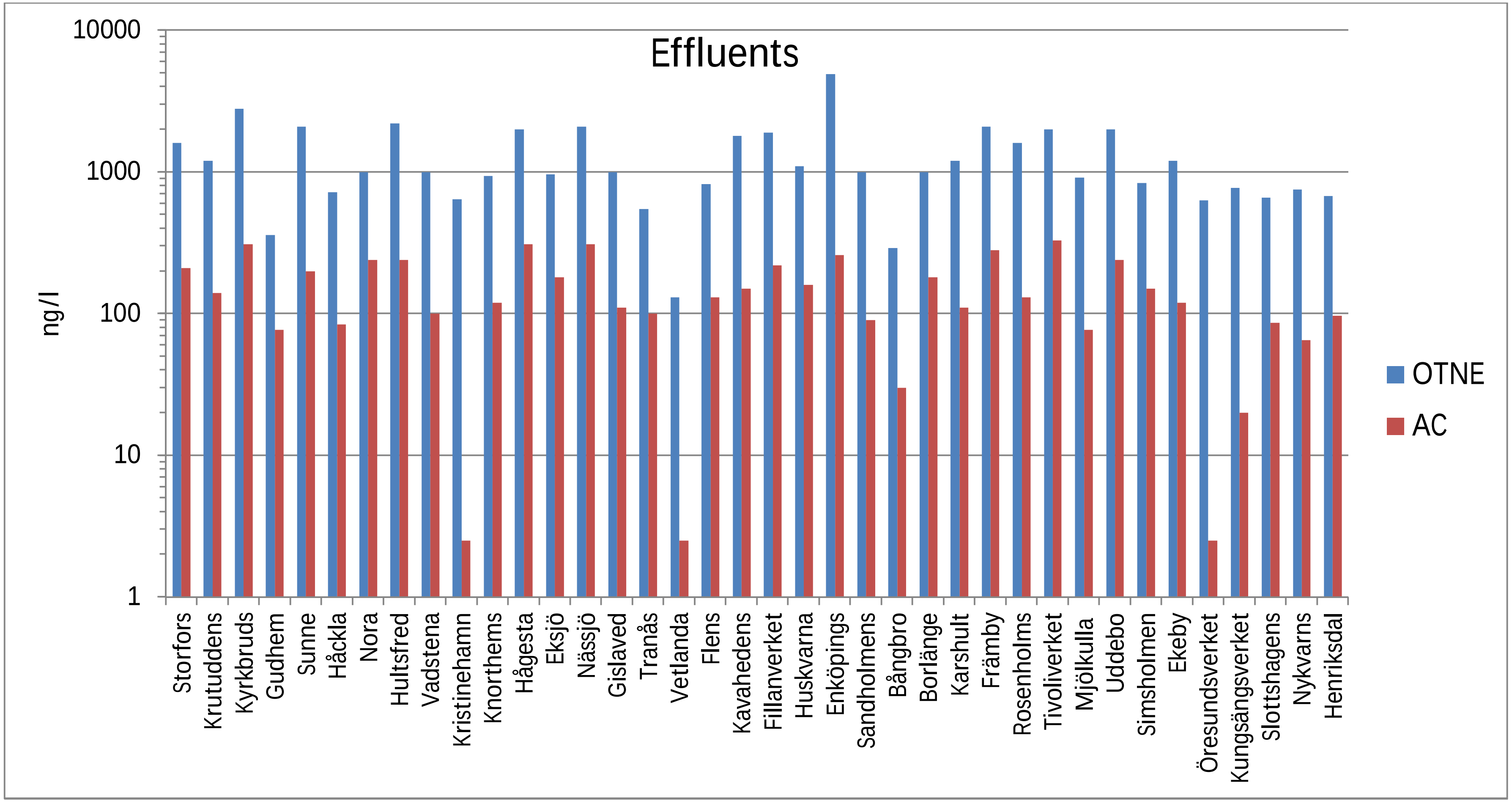

Figure 13. Levels of OTNE and AC in effluents of 38 different WWTP:s arranged from smallest to largest in size (pe). Note that the scale is logarithmic.

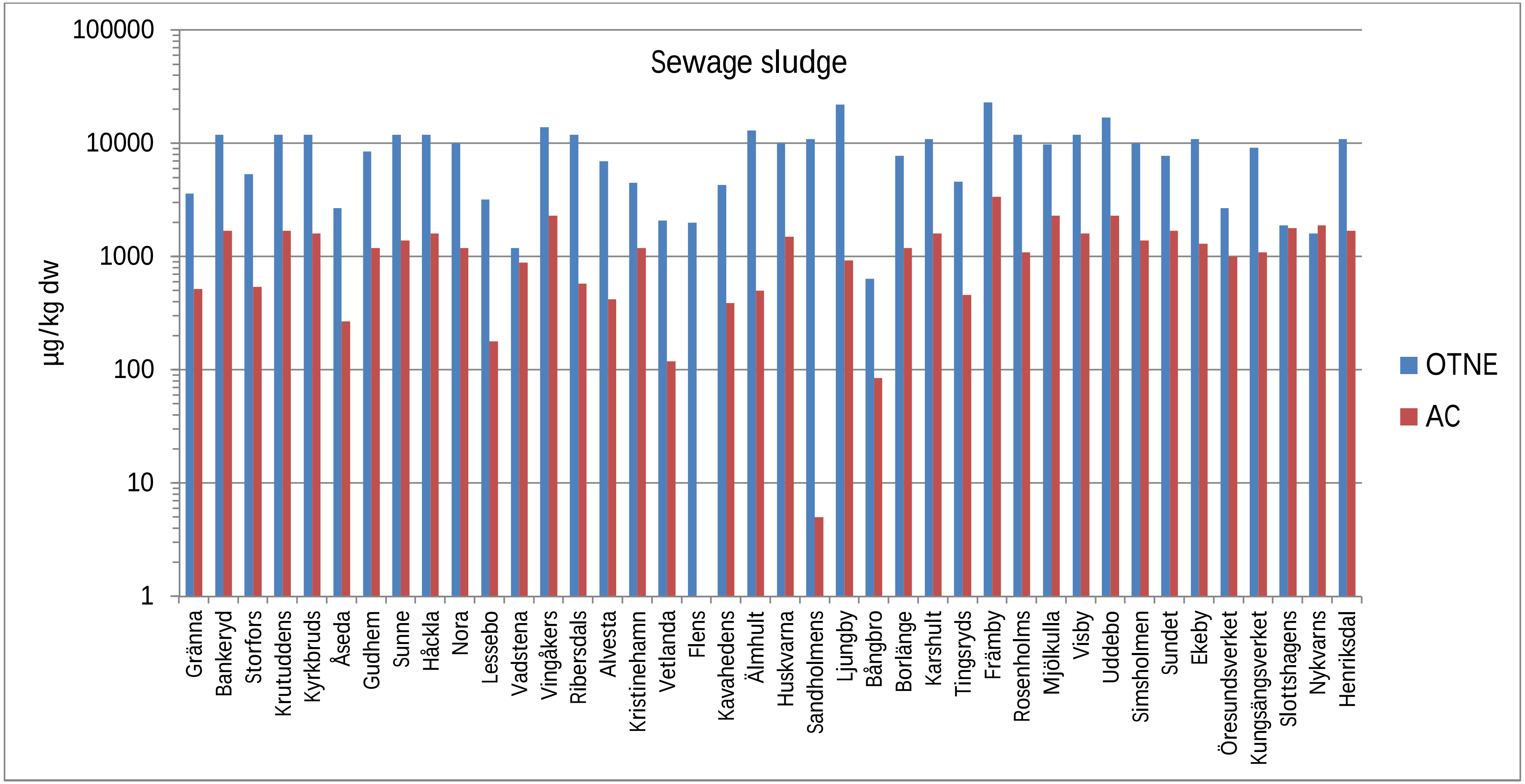

Figure 14. Levels of OTNE and AC in sewage sludge of 40 different WWTP:s arranged from smallest to largest in size (pe). Note that the scale is logarithmic.