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Office of Pollution Prevention and Toxics
U.S. Environmental Protection Agency
Attn: TSCA Section 8(e) Coordinator
Ariel Rios Building
1200 Pennsylvania Avenue, NW
Washington, DC 20460



Re: Siloxanes Monitoring Data

Dear TSCA Section 8(e) Coordinator:

On behalf of its member companies, the Silicones Environmental, Health and Safety Council (SEHSC)¹ submits the enclosed copies of the following two reports: *Siloxanes in the Nordic Environment (TernaNord 2005:593)* (the "Nordic Monitoring Report") and *Results from the Swedish National Screening Programme 2004. Subreport 4: Siloxanes* (the "Swedish Screening Report").² Although SEHSC members have not concluded that reporting of this information is required under section 8(e) of the Toxic Substances Control Act (TSCA), as interpreted in "TSCA Section 8(e); Notification of Substantial Risk; Policy Clarification and Reporting Guidance" (68 Fed. Reg. 33129, June 3, 2003), the referenced reports are nonetheless being provided to the U.S. Environmental Protection Agency (EPA or Agency) as they contain new monitoring data and other information that may be of interest to the Agency.

Chemical Substances

The enclosed reports present monitoring data on a variety of siloxanes, including octamethylcyclotetrasiloxane (D4) (CAS No. 556-67-2), decamethylcyclopentasiloxane (D5) (CAS No. 541-02-6), and dodecamethylcyclohexasiloxane (D6) (CAS No. 540-97-6).

¹ SEHSC is a not-for-profit trade association whose mission is to promote the safe use of silicones through product stewardship and environmental, health and safety research. The Council is comprised of North American silicone chemical producers and importers. SEHSC's members include Clariant LSM (Florida), Inc.; Degussa Corporation; Dow Corning Corporation; GE Advanced Materials-Silicones; Rhodia Inc.; Shin-Etsu Silicones of America; and Wacker Silicones, A Division of Wacker Chemical Corporation.

² We note that these reports were prepared with the involvement of various governmental authorities in the Nordic region. While recognizing that EPA regularly exchanges information with its regulatory counterparts around the world, because SEHSC members are not certain at this time whether the foreign authorities involved in the preparation of these reports have shared the reports or the information contained therein with the U.S. EPA, complete copies of the referenced reports are attached.



/ 293060

Summary of Results

The Swedish Screening Report provides the results of a screening study of siloxanes. Air, water, sludge, sediment, and biota samples were collected from various locations throughout Sweden and analyzed for the presence of certain siloxanes. In addition, samples of human breast milk, which were primarily collected for analysis of another chemical, were analyzed for the presence of the siloxanes. Various siloxanes were reported in some environmental samples, but not in biota. Very low levels of one or more of D4, D5, and D6 were detected in 11 out of 49 samples of human breast milk. However, we note that the addition of phosphoric acid prior to the storage and subsequent analysis of the breast milk samples confounds an interpretation of the sample results. Because siloxanes undergo rapid hydrolysis under acidic conditions, their presence in breast milk cannot be accurately determined using the sampling and storage methodology described in the report. As a result, the validity of the siloxane measurements may be questionable. Detection of the siloxanes could have resulted from contamination during analysis of the samples, as we would expect no siloxanes to be present in the sample after the addition of the phosphoric acid.

The Nordic Monitoring Report, which includes the environmental sampling results reported in the Swedish Screening Report, also provides the results of a screening study on the environmental occurrence and distribution of volatile methyl siloxanes. Air, biota, sediment, sludge, soil, and water were collected and analyzed from locations in Denmark, Faroe Islands, Finland, Iceland, Norway, and Sweden.³ Siloxanes were reported to be found in all analyzed sample types except soils, though not in all regions. The authors of the report indicate that there was great variation in concentrations and that cyclic siloxanes occurred at higher concentrations than linear siloxanes. In addition, the reported concentrations were generally elevated in urban areas and in areas close to sewage treatment plants.

The Nordic Monitoring Report shows that volatile methyl siloxanes appear to be detectable in biota, although a number of detections were below the limit of quantitation. However, there is insufficient quality control data in either reports to evaluate the accuracy of the results. Finally, the information provided in the reports suggests a high level of uncertainty associated with most of the reported concentrations in biota.

The stated aim of the screening programs described in the referenced reports was to obtain a "snapshot" of the occurrence of the targeted substances in the environment. As a general principle, monitoring data from screening programs are difficult to interpret unless samples are collected from well-defined environmental ecosystems.

A description of all of the results is available in the enclosed reports. Neither SEHSC, nor any member company, has made a determination at this time that any significant risk of injury to human health or the environment is presented by the findings in the referenced reports.⁴

³ The environmental sampling results of the Swedish Screening Report are repeated in the Nordic Monitoring Report.

⁴ Neither SEHSC nor any of its members participated in the gathering or review of the data or the preparation of the reports. SEHSC and its members obtained these reports when they were issued to the general public.

If you have any questions regarding this submission, please contact me at (703) 788-6570, rmenning@sehsc.com, or at the address provided herein.

Sincerely,

Nancy Hill for Reo Menning

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Siloxanes in the Nordic Environment

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Siloxanes in the Nordic Environment

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Nordic Environmental Co-operation

The Nordic Environmental Action Plan 2005-2008 forms the framework for the Nordic countries' environmental co-operation both within the Nordic region and in relation to the adjacent areas, the Arctic, the EU and other international forums. The programme aims for results that will consolidate the position of the Nordic region as the leader in the environmental field. One of the overall goals is to create a healthier living environment for the Nordic people.

Nordic co-operation

Nordic co-operation, one of the oldest and most wide-ranging regional partnerships in the world, involves Denmark, Finland, Iceland, Norway, Sweden, the Faroe Islands, Greenland and Åland. Co-operation reinforces the sense of Nordic community while respecting national differences and similarities, makes it possible to uphold Nordic interests in the world at large and promotes positive relations between neighbouring peoples.

Co-operation was formalised in 1952 when *the Nordic Council* was set up as a forum for parliamentarians and governments. The Helsinki Treaty of 1962 has formed the framework for Nordic partnership ever since. The *Nordic Council of Ministers* was set up in 1971 as the formal forum for co-operation between the governments of the Nordic countries and the political leadership of the autonomous areas, i.e. the Faroe Islands, Greenland and Åland.

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Preface

Since 2001 the Nordic countries have systematically been screening the environment for potentially hazardous substances.

The aim of the Nordic environmental screening of substances is to obtain a snapshot of the occurrence of potentially hazardous substances in the environment both in regions most likely to be polluted as well as in some very pristine environments. The focus is on little known, antropogenic substances and their derivatives, which are either used in high volumes or are likely to be persistent and hazardous to humans and other organisms. If substances being screened are found in significant amounts this may result in further investigations or monitoring on national level.

The Nordic screening project is run by a project group with representatives from the National Environmental Research Institute of Denmark, the Finnish Environment Institute, the Environment and Food Agency of Iceland, the Food-, Veterinary and Environmental Agency of the Faroe Islands, the Norwegian Pollution Control Authority and the Swedish Environmental Protection Agency.

The project is financed and supported by the Nordic Council of Ministers through the Nordic Chemicals Group and the Nordic Monitoring and Data Group as well as the participating institutions. The chemical analyses have been carried out jointly by the Norwegian Institute for Air Research (NILU) and the Swedish Environmental Research Institute (IVL).

The respective participating Nordic countries organised sample selection and collection and transport of samples based on a sample protocol and manuals provided by the analytical laboratories.

Summary

The here presented screening study on environmental occurrence and distribution of volatile methylated siloxanes in the Nordic environment involved six countries: Denmark, Faroe Islands, Finland, Iceland, Norway and Sweden.

Siloxanes belong to a group of substances used in a number of industrial applications and in consumer products such as additives in fuel, car polish, cleaners, anti foamers and car waxes. Besides this, they are widely used in e.g. personal care and biomedical products. The widespread use of siloxanes, their broad application as well as their high volatility has raised the concern for these compounds within various disciplines of environmental science.

As a result of their wide use, siloxanes are presumably spread into the environment both via point sources and via diffuse sources and may be found everywhere in the environment. Recent studies have suggested that siloxanes may have direct or indirect toxic effects on various biological processes.

The screening included the following substances: the linear siloxanes hexamethyl-disiloxane (MM or HMDS), octamethyltrisiloxane (MDM), decamethyltetrasiloxane (MD2M), dodecamethylpentasiloxane (MD3M) and the cyclic siloxanes octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5) and dodeca-methylcyclohexasiloxane (D6). In addition, hexamethylcyclotrisiloxane (D3) was analysed in biota. This substance is very volatile and subject to analytical difficulties, which is why it was not analysed in any of the other matrices.

Sampled media types were air, biota, sediment, sludge, soil and water. Siloxanes were found in all the analysed samples types except soils. The results indicate that there is a general pollution of siloxanes in the Nordic environment. There was, however, a great variation in concentrations. The cyclic siloxanes occurred in all media in significantly higher concentrations than the linear siloxanes. The table below shows the observed concentration ranges in different matrices.

Sub- stance	Air ($\mu\text{g}/\text{m}^3$)	Water ($\mu\text{g}/\text{L}$)		Sludge ($\text{ng}/\text{g dw}$)	Soil ($\text{ng}/\text{g dw}$)	Sediment ($\text{ng}/\text{g dw}$)	Biota ($\text{ng}/\text{g ww}$)
		Sew- age/Industrial*	Coastal/ Watercourse				
MM	<0.004	<0.0005-0.14	<0.0005-<0.0006	<0.5 - <3	<0.1	<0.02-<0.7	<0.4
MDM	<0.008	<0.0005-0.014	<0.0005-<0.0006	<1-64	<0.1	<0.02-<0.7	<0.3
MD2M	<0.006	<0.0005-0.078	<0.0005-<0.0006	1-450	<0.1	<0.02-29	<0.4 - 1.1
MD3M	<0.02	<0.004-0.23	<0.002-<0.004	3-550	<0.1	<0.02-57	<0.5
D3	n.a**	n.a	n.a	n.a	n.a	n.a	<50-90.4***
D4	0.08-4.0	<0.06-3.7	<0.04-<0.09	96-960	<6-<10	<3-84	<5-70
D5	0.05-19	<0.04-26	<0.02-<0.05	1100-89000	<3-<5	<2-2000	<5-2200
D6	0.02-2.1	<0.04-3.8	<0.02-<0.05	220-11000	<2-<4	<1-170	<5-74

* Samples represent influent and effluents to and from sewage treatment plants, landfill leachate and industrial storm water
 ** n.a = not analysed *** Detected levels were below limit of quantification

D5 was the dominating siloxane in all matrices but air, where D4 dominated. This is not in agreement with data on use in the Nordic countries, which indicates that the consumption of D5 and D4 is fairly equal. The results of air measurements indicate a regional variation, with highest concentrations in Norway and lowest in Sweden. Air concentrations of D5 detected inside sewage treatment plants were substantially elevated, and also D5 concentrations measured in other matrices surrounding such plants.

Diffuse sources seem to be most important for the observed concentrations of siloxanes. The concentrations were generally elevated in urban areas and in areas close to sewage treatment plants. The mean concentration of D5 in sludge is comparable to that of the widespread contaminant 4-nonylphenol, but this does not necessarily imply that the effects are the same.

The concentrations in fish liver were fairly variable. Siloxanes were mainly detected in fish samples from sites representing urban/diffuse sources and only a few background samples showed detectable levels. One pooled sample of cod liver from Inner Oslofjord showed highly elevated concentrations. On the whole, biota data indicate that siloxanes may bioaccumulate.

No Observed Effect Concentrations (NOECs) for D4 and estimated Chronic Values (ChV) for D5 were only exceeded in samples representing incoming sewage water to sewage treatment plants. These levels were, however, significantly reduced in the outgoing water from the same treatment plants. Since only little amounts of toxicity data are available for other siloxanes than D4 and No Observed Effect Levels and Chronic Values are estimated from a limited amount of data, the possibility of effects in the local environment close to emissions should not be excluded.

Conclusively, siloxanes are present as common pollutants in the Nordic environment and in many different matrices. They seem to be emitted through diffuse pathways and they enter the aquatic food chain. At present, the observed concentrations are not alarmingly high, and many

background sites seem to be non-contaminated. However, the use of siloxanes is extensive and it is possible that continued use will lead to increased environmental levels, eventually reaching effect concentrations.

1. Frame of the study

The aim of this screening programme was to investigate the occurrence and distribution of some siloxanes in environmental samples from the Nordic countries. The analytical results produced in this screening project will be a part of scientific measures to estimate the environmental risk posed by siloxanes in the vulnerable Nordic ecosystems.

Siloxanes are widely used chemicals and the selection of the substances to be included in the study was based on their use as well as on indications of their occurrence in the environment. The results from this study will indicate the level of contamination and give valuable information about the spatial distribution of siloxanes in the Nordic environment.

Initially, octamethylcyclotetrasiloxane (D4) and decamethylcyclopentasiloxane (D5) were selected for the Nordic screening study, due to their properties and widespread use in the Nordic countries. Additionally, dodecamethylcyclohexasiloxane (D6) and the non-cyclic analogues hexamethyldisiloxane (MM), octamethyltrisiloxane (MDM), decamethyltetrasiloxane (MD2M) and dodecamethylpentasiloxane (MD3M) have also been analysed. In addition, hexamethylcyclotrisiloxane (D3) was analysed in biota. This substance is very volatile and subject to analytical difficulties, which is why it was not analysed in any of the other matrices.

Siloxanes are referred to by their full names or abbreviations according to Table 1. The abbreviated names for siloxanes are taken from the General Electric's siloxane notation (Hurd, 1946).

Table 1. Siloxane chemicals selected for the Nordic screening programme.

Abbreviation	Name	CAS #	Structure
D3	Hexamethylcyclotrisiloxane	541-05-9	
D4	Octamethylcyclotetrasiloxane	556-67-2	
D5	Decamethylcyclopentasiloxane	541-02-6	
D6	Dodecamethylcyclohexasiloxane	540-97-6	
MM (or HMDS)	Hexamethyldisiloxane	107-46-0	
MDM	Octamethyltrisiloxane	107-51-7	
MD2M	Decamethyltetrasiloxane	141-62-8	
MD3M	Dodecamethylpentasiloxane	141-63-9	

2. Background

2.1 Chemical properties, fate and toxicity

Siloxanes form a large group of chemicals with molecular weights from a few hundreds to several hundred thousands. This study is limited to cyclic and linear polydimethylsiloxanes of low molecular weight. They occur as clear viscous liquids at room temperature and have varying physical-chemical properties according to Table 2.

Table 2. Chemical and physical data for siloxanes

Substance	MW (g/mol)	W_{soil} (mg/l)	V_p (mm Hg)	H (Atm m ³ /mol)	Log K_{ow}	BCF (L/kg)	K_{oc}
D3	222.5						
D4 ^a	296.6	0.9 (25°C)	1 (25°C)	0.42	-	12400	2.85×10^4
D5 ^b	370.8	0.24 ^b ; 0.017 ^c (25°C)	0.2 (25°C) ^{b,c}	0.4 ^b ; 0.3 ^c	5.7 ^b ; 5.2 ^c	5300 ^a	1.6×10^{4b}
D6 ^a	444.9						
MM ^a	162.4	2 (25°C)	42 (25°C)	4.5	4.2	900	4.6×10^3
MDM ^a	236.5						
MD2M ^a	310.7						
MD3M ^a	384.8	3.1×10^{-4c}	0.102 ^c	0.79 ^c	6 ^c		

^aHSDB, 2004; ^bECB, 2005; ^cSRC, 2005

In the atmosphere, siloxanes may exist both in the vapour and particle phases, the most volatile mainly in the vapour phase. When in the particle phase, siloxanes are removed from the air mainly through wet and dry deposition whereas in the vapour phase they may also react with hydroxyl radicals (HSDB, 2004). Half-lives for reaction with hydroxyl radicals in air are given in Table 3.

Table 3. Half-lives for reaction with hydroxyl radicals in air

Substance	Half-life	References
MM	12 days	HSDB, 2004
D4	16 days	HSDB, 2004
D5	10 days	HSDB, 2004

MM, D4, and D5, have high vapour pressures and high Henry's law constants and presumably vaporise both from wet and dry soils as well as from water. Siloxanes have high K_{oc} (Table 3) and are expected to be immobile in soil. They adsorb to particles in water and are likely to be enriched in sediments (HSDB, 2004).

Siloxanes are resistant to chemical reactions such as oxidation, reduction, and photodegradation (HSDB, 2004). Varying information exists on the possibility of siloxanes to undergo hydrolysis. While siloxanes in general are considered to be resistant to hydrolysis it was shown in a study of hydrolysis kinetics of D4 that in the pH range of 5-9 (25°C) D4 was degraded with a rate that was considered environmentally significant. Intermediate and final hydrolysis products were not established in the study (Durham et al., 2004).

A modelling exercise was performed using the Equilibrium Criterion (EQC) model (Mackay et al., 1996) in order to highlight the likely fate and partitioning behaviour of siloxanes. D5 and MD3M were selected as model substances for fate assessment. Physical-chemical properties were taken from Table 2. The degradation half-lives used were as follows; air: 170 h, water 550 h, soil 1700 h and sediment 5500 hours. The data was based on degradation data obtained from HSDB (2004), as well as estimated data using the EPIWIN software (Meylan, 1999) and classified according to Mackay (2001). Emission rates were set to 1000 kg/h, only for illustrative purposes. The outcome of the modelling exercise is shown in Table 4. The numbers given in the table should be regarded as indicative, as they are dependent on model structure as well as chemical property data.

Table 4. Results from EQC modelling of D5 and MD3M, using emission rates of 1000 kg/h

Emission-medium	Percentage in air		Percentage in water		Percentage in soil		Percentage in sediment		Persistence (h)	
	D5	MD3M	D5	MD3M	D5	MD3M	D5	MD3M	D5	MD3M
Air	99.9	100	<0.001	<0.001	0.1	<0.1	<0.01	<0.001	71	71
Water	4	1	26	8	<0.01	<0.001	70	91	778	2271
Soil	50	81	<0.01	<0.001	50	19	<0.01	<0.01	138	87
All three	17	6.5	21	8	7	0.5	55	85	329	810

The overall residence time in the system of both substances is predicted to be fairly low, and generally lower for D5 (<14 days when emitted to all media) than for MD3M (\approx 30 days). It should be emphasised, however, that advective processes contribute significantly to this low residence time, and it does not necessarily imply that the chemical is ultimately removed from the environment. On the contrary, the atmospheric half-lives of 1-2 weeks (Table 3) and their general resistance to chemical reactions as mentioned above, imply that siloxanes are persistent enough to undergo long-range atmospheric transport.

The model results emphasise the high volatility of siloxanes, showing significant partitioning to air when emitted to air or soil. When emitted to water, however, a large amount is also expected to deposit to sediments, as a result of a relatively high LogK_{OW}-value and a fair amount of D5 is predicted to remain in water. The primary receiving media are likely to be

air and water, based on the high volatility and the areas of use of siloxanes. Based on the model results, siloxanes are thus likely to be found in most environmental matrices.

Bioconcentration factors calculated from the apparent octanol/water partition coefficients are generally low for siloxanes with low molecular weight and high for siloxanes with high molecular weight. Laboratory experiments have also shown high BCF for D4 (HSDB, 2004). In practice, the bioconcentration of heavier siloxanes can be restricted because of limited absorbance through cell membrane due to their large size. High vaporisation of siloxanes from water as a result of their high volatility, combined with high sedimentation rates further reduce the actual concentrations available for uptake in biota (HSDB, 2004).

MM is irritant to skin and D4 is classified as R62 "possible risk of impaired fertility" and as R53 "may cause long-term adverse effects in the aquatic environment" in the EU (KemI, 2004). Some evidence exists on the potential carcinogenicity of siloxane D5 (U.S. EPA, 2003).

Table 5. Ecotox data for D4 (US EPA, 2005) IMBL: Immobilisation, GRO: growth, MOR: mortality, REP: Reproduction, ITX: intoxication, NOC, MULT: Multiple effects recorded as one result

Common name,Scientific name	Endpoint	Effect	Duration	Conc (ug/L)
Opposum Shrimp (<i>Americamysis bahia</i>)	NOEC	IMBL	14 d	9.1
Midge (<i>Chironomus tentans</i>)	NOEC	GRO	14 d	>15
Midge (<i>Chironomus tentans</i>)	NOEC	MOR	14 d	>15
Sheepshead minnow (<i>Cyprinodon variegatus</i>)	NOEC	MOR	14 d	6.3
Water flea (<i>Daphnia magna</i>)	NOEC	IMBL	48 h	>15
Water flea (<i>Daphnia magna</i>)	NOEC	REP	21 d	1.7 - 15
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	LC50	MOR	14 d	10, 8.5 - 13
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	LOEC	MOR	14 d	6.9
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	NOEC	MOR	14 d	≤4.4
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	NOEC	NOC, MULT	93 d	4.4
Water flea (<i>Daphnia magna</i>)		ITX, IMBL	21 d	1.7 - 15

The lowest value for No Observed Effect Concentrations (NOEC) was obtained for water fleas (*Daphnia magna*), a zooplankton that is an important grazer in many limnic ecosystems. In addition to the ecotoxicity data presented for D4 (Table 5), Lassen et al., (2005) have derived so-called Chronic Values (ChV) for fish for a number of siloxanes by using the U.S. EPA PBT Profiler software (U.S. EPA, 2005). ChV is the same as the chronic no effect concentration and shows at what concentration no long-term effects are expected. The results are listed in Table 6.

Table 6. Chronic values for fish derived by Lassen et al., (2005)

Chemical abbreviation	Fish ChV (mg/L)
D4	0.058
D5	0.021
MM	0.062
MDM	0.028

3.Applications and use of siloxanes

Siloxanes are widely used over the world. D4, D5, and MM are chemicals of high production volume within the European Union. In the Nordic countries there is a limited use of MD2M, MDM and MM, and more extensive use of D4 and D5 (Figure 1). The Norwegian and Finnish use of D6 is confidential, whereas Sweden reports 1 tonne in each of the years 2001 to 2003. Denmark reports occurrences of D6 in more than ten products, but no figures are given (SPIN, 2005). The consumption of MD3M is confidential (too few users) and restricted to Sweden. Only Denmark and Sweden have reported use of D3. Sweden and Denmark report occurrence in more than 15 products but no figures are given. In Norway the use of D3 is confidential (SPIN, 2005). Figure 2 shows a comparison of the D4 and D5 uses in the Nordic countries.

Figure 1. Registered use of D4 (left) and D5 (right) in the Nordic countries (SPIN, 2005).

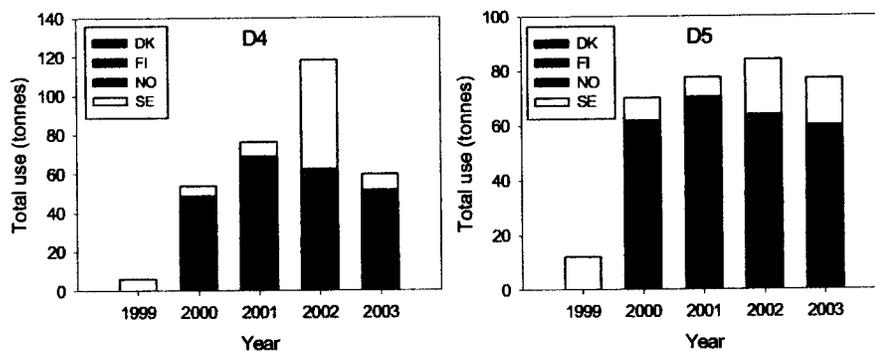
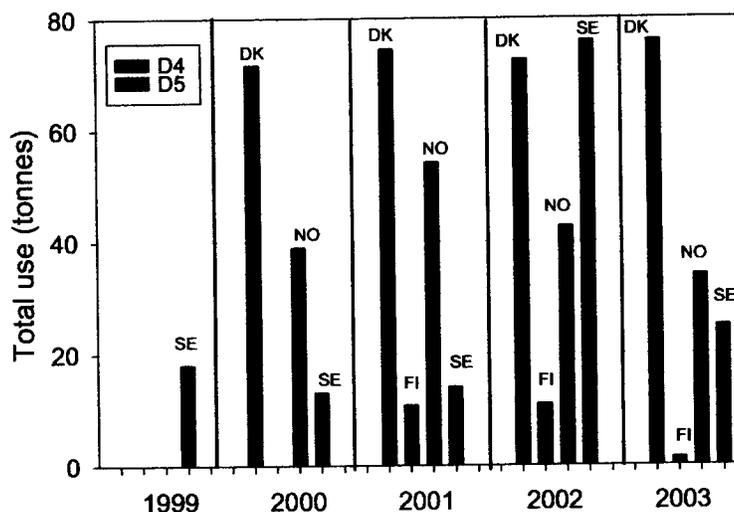


Figure 2. Comparison of use of D4 and D5 in the Nordic countries.



Hexamethyldisiloxane (MM) is included on the OSPAR candidate list for dangerous substances. D4 is classified as a PBT/vPvB chemical and hence as a phase-out substance in the priority database (PRIO) of the Swedish Chemicals Inspectorate. Thus, it is not supposed to be used in any new chemical applications within Sweden. In Denmark, the Association of Danish Cosmetics, Toiletries, Soap and Detergent Industries (SPT) is planning to introduce substitution of D4 (Danish EPA, 2004). Table 7 shows the fields of application of siloxanes in the Nordic countries.

Table 7. Areas of application of siloxanes in the Nordic countries (SPIN, 2005).

Substance	Area of application
D3	Industry for perfume, raw material and intermediaries for cosmetic production, manufacture of chemicals and chemical products. Sale, maintenance and repair of motor vehicles and motorcycles; retail sale of automotive fuel
D4	Fuel additives, Cleaning/washing agents, Impregnation materials, Adhesives, Binding agents, Surface treatment, Construction materials, Paints, laquers, varnishes, Fillers, Reprographic agents, Process regulators, Anti-set-off, Anti adhesive agents, Cosmetics
D5	Fuel additives, surface treatment, cleaning/washing agents, filler, impregnation material, adhesives, binding agents, paints, laquers, varnishes, reprographic agents, softeners, surface active agents, process regulators
D6	Surface treatment, paint, laquers, varnishes
MD	
M	Paint, laquers and varnishes
MD	
2M	

The figures from the SPIN database only represent the registered use in the Nordic countries. For most products, importers are not obliged to register the full content of chemicals. It is therefore difficult to estimate the true use of siloxanes in the Nordic countries. Judging from the many applications for siloxanes (Table 7) it can be assumed that the total use is larger than implied by the SPIN database.

In the cosmetics industry, the name cyclomethicone is used for the cyclic dimethylsiloxanes and blends thereof. In the cosmetic database of the Danish EPA cyclomethicone is found in 64 out of 766 products covered in the database. Product categories are body lotion, hair styling products, creams, lipstick, make-up for children and deodorants (Danish EPA, 2004).

3.1 Environmental levels and exposure

Data on the occurrence of siloxanes in the environment are scarce. In order to extend reference data, Table 8 shows results from previous measurements of polydimethylsiloxanes found in the literature, although the specific compounds were not specified in the study, and it is thus unclear whether the reported concentrations include chemicals investigated in the current study. Table 9 shows previously detected concentrations of MM and D4 in the environment.

Table 8. Concentrations of polydimethylsiloxane in varying matrices (HSDB, 2004)

Matrix	Concentration	Country	Year
Surface water near sewage treatment plant	0.8-5 µg/L	Australia, Japan and USA	1997
Surface water near industrial area	2.8 -54.2 µg/L	Japan	1997
Water from a sewage water treatment plant	Barely detected	USA and Japan	1997
Sludge	20-50100 mg/kg dw	Australia, Canada, Germany, Japan and USA	1997
Sediment	Nd - 314 mg/kg dw	Australia, Germany, Japan and USA	1997
Fish	0.6-0.7 mg/kg	HSDB	2004

Table 9. Concentrations of siloxanes in various matrices.

Country	Location	Matrix	MM	D4	Reference
Sweden	Landfill	Percolate water (µg/l)	2-106	1-2	Paxéus, 2000
Germany	Landfill 1	Biogas (mg/m ³)	1.04-1.31	7.97-8.84	Schweigkopfer 1999
Germany	Landfill 2	Biogas (mg/m ³)	0.38-0.77	4.24-5.03	Schweigkopfer 1999
Germany	STP 1	Biogas (mg/m ³)	0.05	6.40-6.98	Schweigkopfer 1999
Germany	STP 2	Biogas (mg/m ³)	0.01	2.87-3.02	Schweigkopfer 1999

As part of an interdisciplinary field study (DBH: Dampness in buildings and health) indoor air measurements in 400 Swedish homes has recently been carried out. NILU was responsible for the analysis of VOCs in children's bedrooms. The results from this study are summarised in Table 10.

Table 10. Concentration of siloxanes in indoor air in Sweden.

Siloxane	Number of homes with siloxane detected	Mean($\mu\text{g}/\text{m}^3$)	Min($\mu\text{g}/\text{m}^3$)	Max($\mu\text{g}/\text{m}^3$)
D5	250	9.7	0.5	79.4
D6	142	7.9	0.6	164
D4	73	9.0	0.6	51.2
D7	8	6.4	1.2	35.5
MD2M	5	20	5.3	73.2
MDM	2	7.4	2.5	12.3
MM	1	1.5	-	-
D3	1	7.3	-	-

4. Methodology

4.1 Sample selection: Criteria and priorities

All samples collected in this screening study are listed in Appendix 1, where also the sampling characteristics are given in detail. An overview of the sampling sites and their spatial distribution in the Nordic countries are shown for biota, sediment & soil, sludge, water and air in Figure 3 to Figure 7.

Each country made a selection of samples based on the knowledge of use and expected occurrence of siloxanes for that particular country. In all cases, samples were chosen to represent point sources as well as diffuse sources and in some cases also background areas. The goal was also to cover all the matrices air, water, sediment, sludge and biota. The strategies and samples selected for the different countries are outlined in the following sections.

4.1.1 Denmark

Abiotic samples

Landfill leachate was collected from one old landfill at Uggeløse, which has received mixed waste including household waste in the past, and from one landfill at Avedøre, which is still in use. The latter does not receive household waste.

Sewage water and sewage sludge samples were collected from Lynetten (Copenhagen) as bulk samples. At Bjergmarken (Roskilde) one week integrated samples of sewage water were collected.

Air samples were taken in the background area of Sepstrup Sande, at street level in Copenhagen (Jagtvej), 22 m above the street level (HCØ, Copenhagen), and from Bjergmarken sewage treatment plant close to the aeration basins.

Surface water and sediment samples were collected at Roskilde inderfjord (near Roskilde), in Øresund near Lynetten sewage treatment plant (Copenhagen) and in Kattegat (no known point sources).

Biotic samples

Common seal, *Phoca vitulina* were found dead at the Danish shores after an epidemic in 2002. After dissection the samples have been kept frozen at -20°C . Each sample consists of five subsamples of blubber from five individuals from each of four seal colonies. Marine fish were collected in the same area as water and sediment samples from Roskilde fjord, in

Øresund (Nivå bugt), which is influenced by cities along the coast of Denmark and Sweden and at two background areas at the Wadden Sea and the North Sea.

4.1.2 Faroe Islands

Abiotic samples

Sludge and effluent water was collected at the Sersjantvíkin sewage treatment plant in Torshavn. Air samples were taken inside this same sewage treatment plant as well as in a downtown intersection during rush hours. Soil samples were taken from an abandoned landfill in Havnardalur and from Húsarhaga which is the operating landfill for the Torshavn area. From Húsarhaga also runoff water was sampled, with sampling in the well where water draining a larger part of the landfill is led.

Sediment samples were taken in the Kaldbaksfjord, at one of the stations that are used by the Fiskeris Laboratory in their monitoring activities, at site KA05. The fjord is known to be influenced to some degree by pollution, but the sources have yet to be identified.

Biotic samples

Eggs from the seabird species fulmar (*Fulmarus glacialis*) and black guillemot (*Cepphus grylle*) were gathered in connection with environmental monitoring in various programs in the period 2000 to 2004, and stored at -20°C in PC jars. Land-locked Arctic char and brown trout were taken in 2004 in the lake á Mýranar, a site used also for sampling to the Arctic Monitoring and Assessment Program, AMAP. Marine fish species sculpin (*Myoxocephalus scorpius*) and dab (*Limanda limanda*) were taken near the bottom of the Kaldbaksfjord in 2004. Cod (*Gadus morhua*) were taken north at the Faroe plateau at Mýlingsgrunnurin in October 2004, by the research vessel "Magnus Heinason". These fish samples are also part of other monitoring programs like OSPAR CEMP and AMAP.

Two species of toothed whales have been included in the sample set from the Faroe Islands, these are long-finned pilot whale (*Globicephala melas*) and whitesided dolphins (*Lagenorhynchus actutus*). Samples of blubber from both species were taken in 2004 in connection with the traditional drive hunt, the pilot whales in Torshavn (Sandagerði) and the dolphins in Gøtu (Syðrugøtu).

4.1.3 Finland

Abiotic samples

Leachate water was collected from the solid waste treatment centre Ämmässuo in Espoo. The total amount of waste stored and dumped into the landfill was estimated to 8.5 millions m^3 in 2004. Extensive water collec-

tion systems have been built to drain the waters from the landfill area to a municipal wastewater treatment plant in Espoo City. The landfill leachate water is led to a pool of the size 150 x 150 m and a depth of 5 m. Water samples from this pool were taken in August 2004 and in January 2005. Wastewater and dehydrated sludge were collected from the main sewage treatment plants of Helsinki (population equivalent, pe 900 000), Espoo (pe 240 000), Porvoo (pe 30 000) and Nokia (pe 22 000). Samples of incoming wastewater from tyre and floor material industry in Nokia were included in the sampling programme. These are treated in the municipal treatment plant, but samples of incoming water were collected from a separate pipeline. As reference samples, sludge was collected from a small treatment plant of the Pornainen commune (pe <1000), where the main part of sewage originates from households.

One sediment sample (0-6 cm) was taken in the shallow bay near downtown Helsinki, which is also an outlet of the river Vantaanjoki. The bay has been target for pollution from several sources for decades. The other sediment sample is from area between Espoo and Kirkkonummi, about 10 – 15 km west of Helsinki City. This sea site is regarded as fairly polluted by strong yachting and shipping activities.

Air samples were collected at the sewage treatment plant in Nokia City and in the Ämmässuo waste treatment centre in Espoo. An air pump was placed near the exterior effluent pools in Nokia and near the large leachate water pool in the Ämmässuo landfill area. The sites were controlled so that the wind direction was adequate for operation and that the weather was good (no rain).

Biotic samples

Pikes (*Esox lucius*) from three separate sea bays of the Helsinki coast were caught. The study areas are shallow and eutrophic. The sea bay nearest downtown is the same where the sediment samples were taken (3 pooled liver samples). The other two inlet bays are slightly polluted by intensive boating and yachting (1 pooled liver sample per area).

4.1.4 Iceland

Abiotic samples

All abiotic samples were collected in the vicinity of Reykjavik. Sewage sludge samples were collected on 13th of December 2004 from the two main sewage-pumping stations in Reykjavik, Ananaust and Klettagardar. Waste landfill water was collected the same day from the runoff of a nearby waste dump at Alfsnes. Sediment samples and sea water were collected on the 3rd of February 2005 from the sea just outside the Reykjavik municipal waste landfill at Gufunes. Air was collected in the park-

ing lot of Kringlan mall on 26th February 2005 and in the Grensas traffic junction 28th February 2005.

Biotic samples

Common porpoise samples are from the year 2000. The whales were caught accidentally in fishing nets Northwest of Iceland and subsequently brought to the Maritime Research Institute storage in specimen bank.

4.1.5 Norway

Abiotic samples

In order to represent background areas (without known local input), sediment and water samples were collected from Lake Bergsjøen and Lake Røgden. A coastal background water sample, representing mainly long-range pollution, was collected at Færder in the Outer Oslofjord.

Representing possible point sources air samples from one sewage treatment plant plus runoff water from three waste deposits and one sewage treatment plant were taken.

As diffuse source samples, one sample was collected at NILUs air-pollution control site at Manglerud, a quite trafficked site in Oslo. Two of the air samples were taken at the Bekkelaget sewage treatment plant – about 2 m above incoming untreated water and mechanically treated water. The last sample was taken at the main hall of the Oslo Central station (railway station) on a Friday evening.

Sediment samples were collected from Leangbukta and Vrengensundet in Oslofjord and from the Ålesund harbour area on the West Coast. One water sample was taken at Steilene in the inner part of Oslofjord.

Biotic samples

Marine fish samples representing approximate background areas were collected in Farsund near Lista on the Norwegian South coast.

As diffuse source samples, marine and freshwater fish was collected from Lake Mjøsa, Inner Oslofjord (city), Inner Sørfjord and Ulsteinvik at the West Coast.

4.1.6 Sweden

Abiotic samples

In order to represent background areas (without known local input) two sediment samples from the Baltic Sea (Ö Gotlandsdjupet, Ö Landsorts djupet) were collected.

Representing possible point sources, air, water and sediment samples were collected in the proximity of a former rubber industry area (Gislaved) and from a municipal landfill (Högbytorp).

As diffuse source samples, air was collected in the centres of Stockholm and Gothenburg and sediment samples were collected from Essingen and Riddarfjärden in Stockholm. In addition sludge samples were collected from municipal wastewater treatment plants geographically distributed over Sweden (Skellefteå, Lerum, Eslöv and Kiruna).

Biotic samples

Eggs from herring gull (*Larus argentatus*) were collected and used as samples representing background conditions.

Freshwater fish (pike, *Esox lucius*) from the river Nissan (in the proximity of a rubber industry) were sampled to represent a point source area.

Figure 3. Nordic sampling sites for biota.

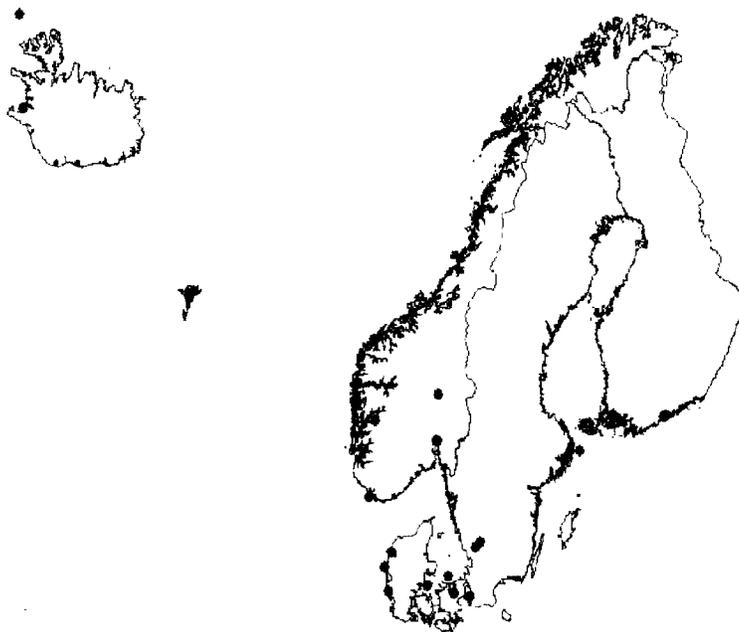


Figure 4. Nordic sampling sites for sediment & soil.

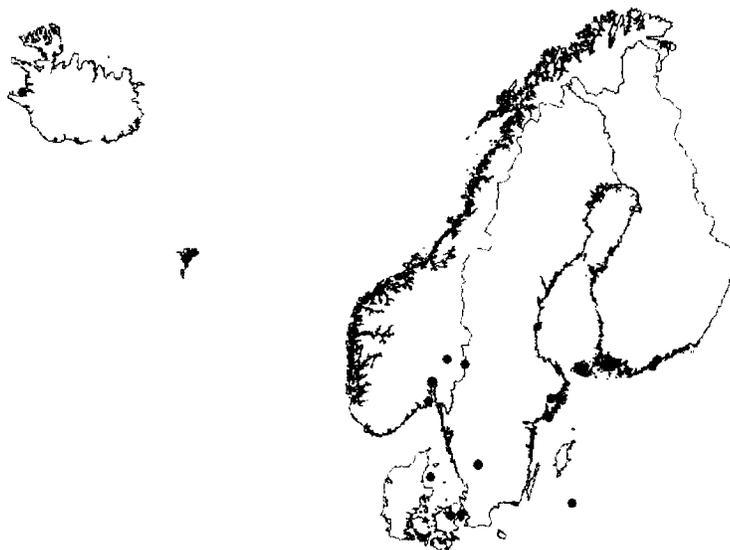


Figure 5. Nordic sampling sites for sludge.

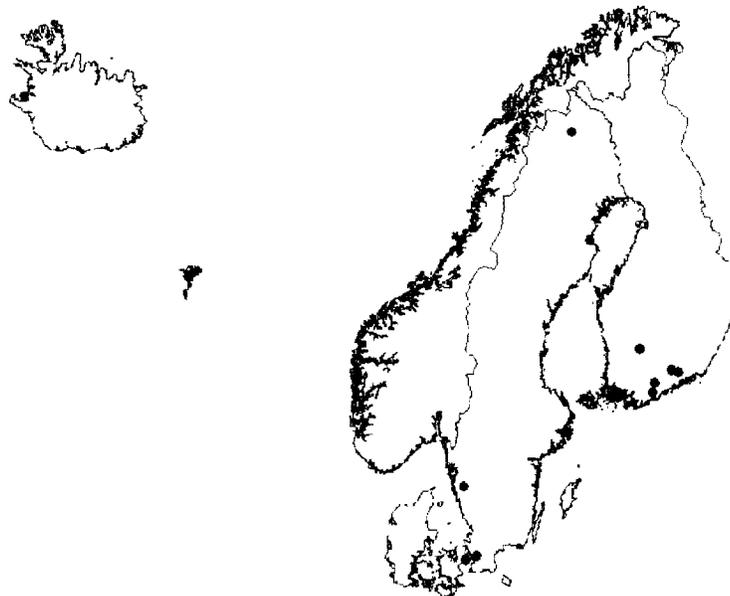


Figure 6. Nordic sampling sites for water.

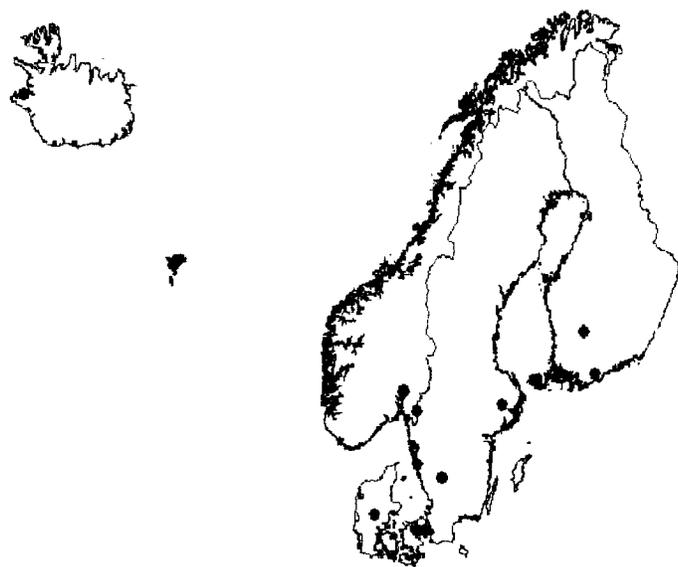
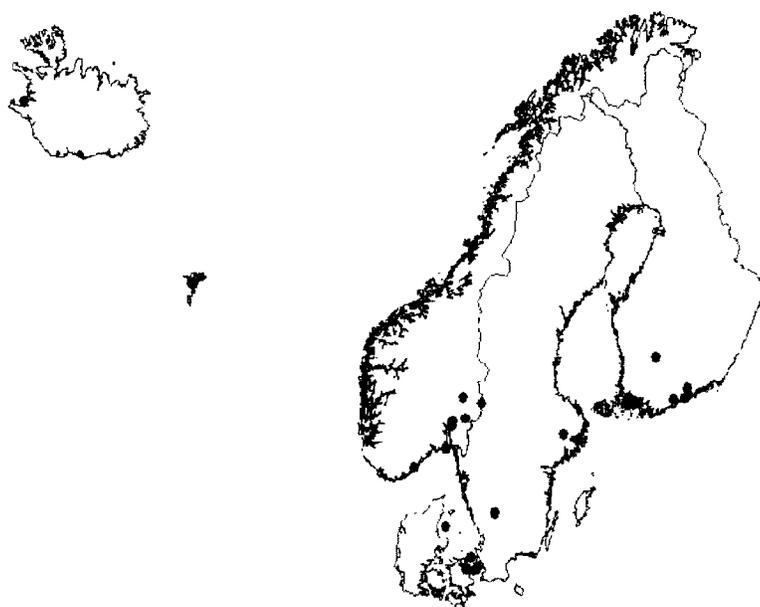


Figure 7. Nordic sampling sites for air



4.2 Sampling

As a guideline for adequate and consequent sampling, the laboratories in charge provided a manual (Appendix 3) for the sampling personnel in the Nordic countries participating in the screening. Detailed instructions for sampling, storing and transport were given. Sampling protocols for all sample types were included in the manual. The aim of the sampling protocol was to

1. Guide the personnel responsible for sampling on how to avoid contamination of the samples.
2. Ensure documentation of the sampling procedure, quality of the sample and environmental and physical circumstances during the sampling.

All samples were sent directly to the analytical laboratories by the national institutions responsible for sampling. Water, sludge and sediment/soil samples were sent to IVL in Sweden and the air and biota samples to NILU in Norway.

4.3 Method of analysis

4.3.1 Analysis of sludge, sediment, water and soil samples

In short a sample was diluted with water and purged with a gas stream passing through an adsorbent trap from which the analytes were later thermally desorbed and analysed by GC-MS.

Approximately 2 g of wet sludge were diluted to 20 ml with MilliQ water and homogenised with a high frequency mixer (Polytron). Approximately 1 ml of the slurry was weighed in to the purge & trap vessel and diluted to 10 ml. Sediment was diluted in a similar way, but homogenised by shaking only. Water samples were homogenised by shaking. In all cases 0.5 ml buffer solution (2M K_2HPO_4 , 0.4M HCl, 80g $Na_2EDTA \cdot 2H_2O$ per litre) was added to the purge & trap vessel.

The purge & trap apparatus for sludge, sediment, and waste water samples consisted of a 25 ml graduated glass test tube with an adapter with one inlet for a Pasteur pipette extending to the bottom of the tube and one side arm to which an empty adsorbent tube was connected using flexible tubing (Viton). This tube acted as a short cooler and water trap. An adsorbent tube containing 0.25 g Tenax TA was connected to the empty tube, the Pasteur pipette was connected to the purge gas (nitrogen, 50 ml/min) and the tube was immersed in a thermostated water bath held

at 70°C. An electric fan facilitated air cooling of the upper part of the apparatus. Samples were purged for 20 min (analysis of MM) and for 2h (all other siloxanes). For low contaminated water samples gas washing bottles with glas frit gas inlet was used as purge vessels. Water volume was 60 - 150 ml, the other conditions were the same as above.

The adsorbent tube was transferred to a thermal desorber (Unity, Markes) connected to a GC-MS instrument (6890N, 5973N, Agilent). Pre-purge time was 2 min, tube desorption time 5 min at 225°C, the trap was held at 3°C and heated at 32°C/s to 250°C. The desorbing flow was 30 ml/min and the split flow 10 ml/min. The flow path temperature was 150 °C and it connected directly to the GC-column, which was a CP-Sil 8CB 30 m × 0.25 mm id, film thickness 0.5µm (Varian). The column temperature was 40°C for 3 min, programmed to 200°C at 12°C/min and to 260°C at 30°C/min. The carrier gas was helium held at constant pressure 10 psi measured at 40°C. The massspectrometric detector was used in electron impact single ion recording mode.

500 mg each of D3, D4, D5, MM, MDM, MD2M, MD3M (Aldrich) and D6 (Gelest) were mixed in a test tube. The chemicals were of 97% declared purity or better. 200 mg of the mixture was dissolved in methanol and diluted to 25 ml. This made a stock solution of 1 mg/ml for each component. This solution was further diluted with methanol. Different amounts of this solution were added to 10 ml MilliQ water and 0.5 ml buffer solution in the purge & trap apparatus and analysed as samples. In this way a seven-point linear calibration curve was constructed and used for quantification of the samples. The blank level and calibration was regularly checked by running water blanks and one or more of the calibration points together with the samples.

The two soil samples from Faroe Islands were analysed according to the procedure described for sediments.

4.3.2 Analysis of biota samples

Different methods for determination of D4 and D5 have been described in literature (Flassbeck et al., 2001; Flassbeck et al., 2003; Kala et al., 1997; and Varaprath et al., 2000). However, none of the described methods were using high resolution mass spectrometry for quantification.

Due to the ubiquitary nature of the cyclic siloxanes great care was given to reduce the risk for contamination of the samples with siloxanes through direct contact with the lab staff, the equipment used for sample storage, preparation, and extraction. To avoid evaporation loss of the volatile siloxanes and to reduce the contamination risk a very short and comprehensive sample preparation and quantification method was developed and validated.

Sample preparation

Typically, the sample material was thawed and homogenised with a household mixer. A 0.30 g aliquot of the sample was mixed thoroughly with 1 mL n-hexane on a whirl mixer for 5 min. The mixture was separated by centrifugation at 10000 rpm and the clear solution was carefully removed with a Pasteur pipette and transferred without any further treatment into a GC/MS vial. There was no significant difference in the results from sample extracts which were dried with sodium sulfate according to the procedure published by Dow Corning (Varaprath et al., 2000) and the results from undried extracts.

GC/MS quantification

For method testing and calibration a solution was prepared containing D3, D4, D5, D6, MM, MDM, MD2M, and MD3M at concentrations of about 3 ng/ μ L and about 30 ng/ μ L in n-hexane.

The sample extracts were analysed on a GC/HRMS system (GC: 6890 Agilent, MS: Micromass Ultra Autospec) using the following parameters:

- **Gas chromatograph:**
Splitless injection (injector temp: 200°C), Helium as carrier gas (1 mL/min; constant flow), 25m \times 0.2mm \times 0.11 μ m Agilent Ultra2 capillary; Temperature program: 35°C, 3min, 7°/min, 130°C, 0 min, 30°/min, 325° C, 5 min. Interface temperature: 250 ° C.
- **Mass spectrometer:**
Ion source temperature: 200° C; Electron impact mode with accelerating voltage 8000 V, Resolution: 10000 at 5 %; Single ion monitoring mode acquiring the following masses (m/z): 147.0661 (MM), 207.0329 (D3), 221.0849 (MDM), 281.0517 (D4), 295.1037 (MD2M), 355.0705 (D5), 369.1225

Before and after a series of 10 samples including a complete method blank the calibration solution was injected in 2 parallels. Quantification was performed as external standard quantification.

4.3.3 Analysis of air samples

Perkin Elmer adsorption tubes filled with 200 mg Tenax TA were used for sampling. The tubes were plugged on both ends with brass swagelock caps with PTFE ferrules. The sampling pumps were adjusted to slightly below 100 mL/min. Air samples were sucked through the tubes with the pumps at the back end. Metal Bellow Ultra Clean air-pumps were used for all sampling except the samples taken in Sweden (SKC Pocket Pump 210-1002). Field blank tubes were in each shipment of samples to the different sampling sites.

The samples were analysed on an Automatic Thermo Desorption Unit ATD 400 (Perkin Elmer) coupled to a Hewlett Packard 1800 A GCD GC/MS-system. Tubes were desorbed at 275°C for 20 min, preconcentrated at -30°C and separated on J&W DB1701 capillary column (30 m×0.22 mm×1.0 µm. The mass spectrometer was used in single ion monitoring mode (SIM) acquiring the following masses (m/z): 207.208 (D3); 281.282 (D4); 267.355 (D5); 341.147 (D6); 147.148 (MM); 221.222 (MDM); 207.295 (M2DM); 281.369 (MD3M); 355.221 (MD4M); 355.295 (MD5M). The quantification was performed with external standards on pre-cleaned tubes.

4.4 Quality control and method comparison

Adequate quality control measures and documentation were introduced covering the entire analytical procedure: sampling, storage, transport, sample preparation, analysis and quantification. To assure a correct sampling procedure and reduce the risk of contamination as well as to assure documentation of possible deviations during sampling and transport, sampling protocol was developed in close co-operation between the different analytical laboratories and the screening group's steering committee (see section 4.2).

4.4.1 Limit of detection/limit of quantification

Limit of detection (LoD) and limit of quantification (LoQ) are considered as two priority parameters, describing the quality of a quantitative analytical method. According to IUPAC (McNaught and Wilkinson 1997, Thomsen et al. 2003), the LoD, expressed as the concentration, cL, or the quantity, qL, is derived from the smallest measure, xL, that can be detected with reasonable certainty for a given analytical procedure. The value of xL is given by the equation: $xL = \bar{x}_{bi} - k s_{bi}$ where \bar{x}_{bi} is the mean of the blank measures, s_{bi} is the standard deviation of the blank measures, and k is a numerical factor chosen according to the confidence level desired. For abiotic and biotic samples $k = 3$ (3 x signal/noise) was chosen for the present screening study. In the last Nordic screening report (Kallenborn et al., 2004) the following definition of the limit of quantification was used: the lowest concentration of an analyte that produces a signal/response that is sufficiently greater than the signal/response of lab reagent blanks to enable reliable detection (and thus quantification) during routine laboratory operating conditions. The analyte response at the limit of quantification (LoQ) should be at least 5 times the response compared to the blank response. LoD and LoQ determination was performed in accordance to the guidelines given in the above described documents.

For air samples there are only reported concentrations of D4, D5, and D6. Due to a very high background level, D3 results were omitted from

the report (see also 4.4.3). The lowest concentration reported for air samples is 0.02 ng/L of D4 in one of the Swedish samples. Concentrations below that do not give good enough signal-to-noise on the equipment used. None of the air samples had significant peaks in order to quantify linear siloxanes.

4.4.2 Laboratory and field blanks

Based on measurements of water blanks, LoD expressed in ng for sludge, sediment, and water samples were calculated (Table 11). As different sample amounts were analysed, individual samples will have varying LoDs in units related to sample mass. Individual LoDs are given in Appendix 2.

Table 11. Limit of detection, LoD, ng/sample (see above).

	D4	D5	D6	MM	MDM	MD2M	MD3M
Sludge, sediment	3.9	1.9	1.5	0.04	0.04	0.04	0.04
Water	4.7	2.5	2.6	0.03	0.04	0.03	0.25

Twelve water field blanks were analysed. All results were below LoD except in one case where MM was 17% of the measured concentration in the corresponding run off water sample.

For the biological samples no relevant surrogate for field blanks could be found and therefore only laboratory blanks are performed as given in Table 12. In addition, the LoD and LoQ calculated according to the formula given in 4.4.1 and the LoD which is used in the results table in Appendix 2 is listed.

Table 12. Laboratory blanks (complete method blanks) for the analysis of biological material (ng/g).

Chemical	1	2	3	4	5	6	7	8	9	10	Average	Stddev	LoD calc	LoQ calc	LoD used	LoQ used
D3	34.9	15.2	22.5	26.2	18.1	18.0	17.4	15.9	16.7	18.1	20.3	6.1	38.6	102	50	150
D4	1.4	1.6	3.8	4.7	1.7	1.5	1.5	1.4	1.4	1.3	2.0	1.2	5.6	10.2	5	15
D5	1.3	1.1	1.9	3.5	1.9	ND	1.4	1.8	0.5	ND	1.7	0.9	4.3	8.4	5	15
D6	1.5	0.6	ND	ND	3.0	ND	ND	1.8	ND	ND	1.7	1.0	4.7	8.6	5	15

For air samples, all blank values were below 5% of the actually measured compounds in the air samples. An exception was D3, where blank samples compared to real air samples showed much higher values – in some

cases values as high or even higher than those actually measured at the sampling sites. It was therefore decided not to report D3-values from air samples – due to these contamination problems.

4.4.3 Performance tests

To give an estimate of the coefficient of variation, a sludge sample were analysed in triplicate and one wastewater sample four times. The sludge subsamples were taken from the same homogenised predilution (see 4.3.1). Comparison of different homogenates would give higher CV reflecting sample inhomogenities. The results are given in Table 13.

Table 13. Coefficient of variation (CV%) for repeated analysis of the same sample.

	D4	D5	D6	MM	MDM	MD2M	MD3M
Sludge n=3	14	12	14		12	8.8	
Waste water n=4	16	10	13	4.6	17	13	14

The sampling method for air has been tested. Tubes coupled in series were used in order to control the sampling efficiency of the adsorbents. For this test the normally used flow rate of 100 mL/min was applied. For air samples with a sample volume of more than 20 L there was a break trough of compounds in the order of 10%. Therefore it was decided to keep the actual sampling time to less than 3 hours.

4.4.4 Laboratory intercomparison

IVL prepared two sets of adsorption tubes using their own standard solution containing 25 ng of each compound. The sample tubes were shipped to NILU and analysed at NILU using their own standard solutions.

Table 14. Results of a laboratory intercomparison. Samples spiked at IVL and quantified at NILU.

Compound	Spiking level (IVL) in ng	GC/MS results (NILU) in ng
D4	25	25.33
D5	25	25.83
D6	25	31.50

5. Results and discussion

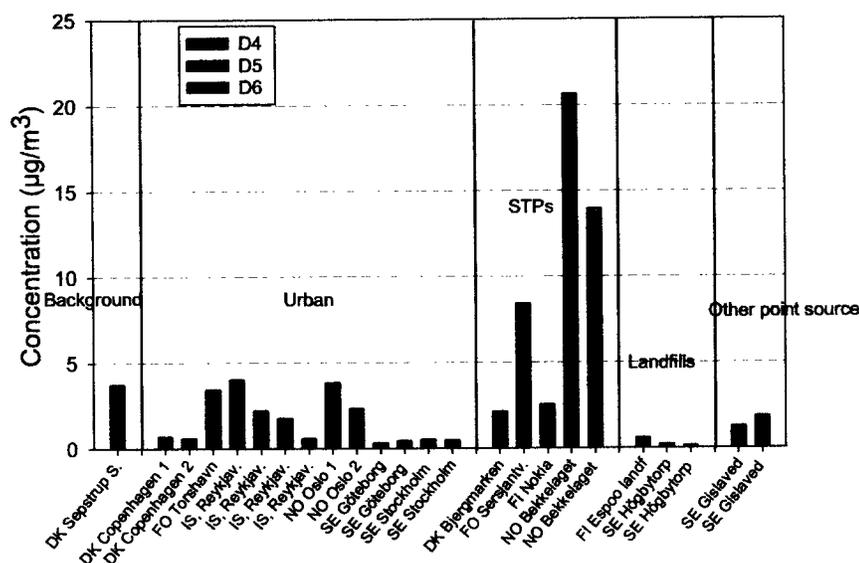
5.1 Environmental concentrations

The concentrations of siloxanes found in sediment, soil, sludge, water, biota, and air are given in Appendix 2.

5.1.1 Air

Concentrations of cyclic siloxanes found in air are shown in Figure 8 where the total (sum of D4, D5 and D6) concentrations as well as the relative distribution between D4, D5 and D6 are illustrated.

Figure 8. Concentration of siloxanes in air, divided by source types



The measured concentrations of the sum D4, D5 and D6 in air were generally within the range 0.1-5 $\mu\text{g}/\text{m}^3$, with the exception of samples taken inside sewage treatment plants (STPs), which were higher. The samples from Bekkelaget STP in Norway were taken indoors, about 2 m above incoming, untreated water and mechanically treated water and show concentrations as high as 14 and 21 $\mu\text{g}/\text{m}^3$ respectively. This is comparable to measured concentrations of D5 in children's bedrooms in Sweden (mean: 9 $\mu\text{g}/\text{m}^3$, n=250; Table 10). The sample from Sersjantviki STP,

showing the third highest concentration, was also taken inside the plant. The outdoor samples from Bjergmarken STP and Nokia STP did not show elevated concentrations in comparison to other sites.

No increased concentrations of siloxanes in air were found close to the other point sources. The concentration of individual siloxanes in urban air was between 0.1 and 2 $\mu\text{g}/\text{m}^3$ with the highest concentrations in urban areas towards the northwest (Norway, Faroe Islands and Iceland) and the lowest in the south (Sweden and Denmark). No urban air samples were collected in Finland. As only one background air sample was taken (Sepstrup Sande), it is difficult to speculate whether the geographical variations are due to differences in local releases or whether there might be an influence of long-range atmospheric transport.

A difference in relative proportions of individual siloxanes was found. The Norwegian samples contained higher concentration of D5 relative to the other siloxanes, whereas D4 dominated in most of the samples from the other countries. This is not in agreement with data on use, where the relative consumption of D4, D5 and D6 is fairly similar in the different Nordic countries (see Figure 2).

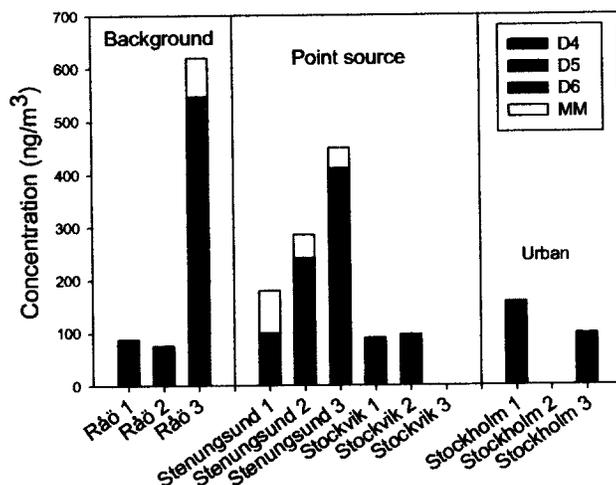
The concentrations of linear siloxanes were below the detection limits in all samples analysed, these being shown in Table 15.

Table 15. Detection limits of siloxanes in air.

Siloxane	D3	D4	D5	D6	MM	MDM	MD2M	MD3M
LoD ($\mu\text{g}/\text{m}^3$)	0.01	0.006	0.02	0.03	0.004	0.008	0.006	0.02

As an assignment from the Swedish Environmental Protection Agency, IVL has performed a National Screening Study of siloxanes during 2004-2005 (Kaj et al 2005). This screening was carried out in parallel to this Nordic Screening. The results of the air measurements from the Swedish screening are shown in Figure 9.

Figure 9. Results from measurements of siloxanes in air in the Swedish screening study (Kaj et al., 2005). Note that the concentration is expressed in ng/m^3 .



The concentrations in the air samples collected within the Nordic screening were generally higher than concentrations of siloxanes in Swedish air, where levels varied between 0.1 and $0.7 \mu\text{g}/\text{m}^3$ ($100 - 700 \text{ ng}/\text{m}^3$). D4 dominated in most of the Swedish samples. In contrast to the Nordic study, the linear MM was found in some of the Swedish air samples, in concentrations similar to D6 (see Figure 9). The background samples from Råö show largely varying concentrations, with 6 times higher concentrations at the last occasion. This higher concentration is still as low as the lowest concentrations observed in the Nordic study. All Swedish background samples were taken in November 2004, thus the observed difference cannot be a result of seasonal variations. Back trajectories using the HYSPLIT model (NOAA, 2005) does not indicate any clear correlation between wind direction and observed concentration. In all three cases, the predominant wind direction was from the west and on occasion 1 and 3 the wind package originated from the Atlantic, passing over Norway (case 1) or Denmark (case 3). On occasion 2 the wind package originated from the Norway area but passed over the inner parts of Sweden before reaching the sampling station. It is therefore difficult to state the reason for the elevated siloxane concentrations observed at Råö on sampling occasion 3. Contamination of the sample cannot be ruled out.

The results from the siloxane measurements in air do not give any clear clues about the atmospheric dispersion pattern in air of these compounds. It seems that sewage treatment plants and certain other diffuse/urban sources may generate slightly elevated concentrations of si-

loxanes in air. However, the air outside sewage treatment plants did not contain elevated levels, and about 50 % of the urban samples showed concentrations lower than background concentrations. Conclusively, more extensive air monitoring, e.g. along urban-rural transects or with increasing distance from sewage treatment plants would be needed to fully answer these questions.

5.1.2 Sludge

The cyclic siloxanes D4, D5, and D6 were found in all the sludge samples analysed (see Appendix 2). D5 was the dominant species in all cases, making up 78 - 94 % of the total amount. The range of concentrations found is illustrated in the "box plots" in Figure 10. Results for individual samples including relative distribution between different siloxanes are illustrated in Figure 11.

Figure 10. Measured concentrations of cyclic siloxanes in sludge in the Nordic screening programme. The lower and upper boundaries of the box represent the 25- and 75-percentiles, and the line within the box is the median concentration. The whiskers represent the 10- and 90-percentiles, and the dots are outliers.

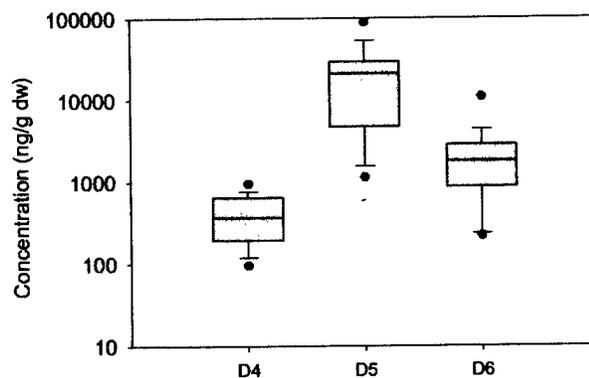
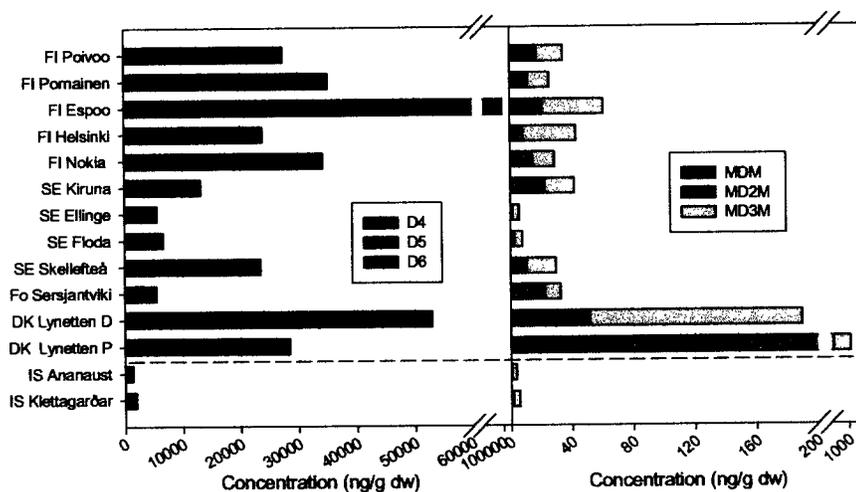


Figure 11.. Concentration of cyclic (left) and linear (right) siloxanes in sludge. The samples from Iceland (IS Ananaust and Klettagarðar) are separated because they represent a different type of sludge.



The two samples from Iceland (Ananaust and Klettagarðar) represent material from mechanical treatment only, while the other samples represent biologically digested sewage sludge. Thus, the Icelandic samples are likely to be less concentrated than the others and concentrations are therefore not directly comparable. The difference in concentration between the two samples from Lynetten, Denmark may be related to the fact that one represents primary and the other represents digested sludge.

Excluding the Icelandic samples yields an average concentration of the sum of D4, D5, and D6 (D456) of 30 000 ng/g dw. Including them gives an average D456 concentration of 26 000 ng/g dw. The sample from Espoo sewage treatment plant in Finland showed the highest concentration of D456, 100 000 ng/g dw. Apart from the Icelandic samples, the lowest concentrations were found in sludge from two small Swedish sewage treatment plants, Ellinge and Floda, and in the sample from Sersjantvíkin in Faroe Islands, with concentrations of 5 600, 6 700 and 5 500 ng/g dw. No Norwegian sludge samples were included in the study.

D5 and D4 are chemicals of high production volume in Europe. D5 was the dominating siloxane in all sludge samples. This is not explained by data on use in the Nordic countries, where the consumption of D5 and D4 is fairly equal (Figure 1, Figure 2).

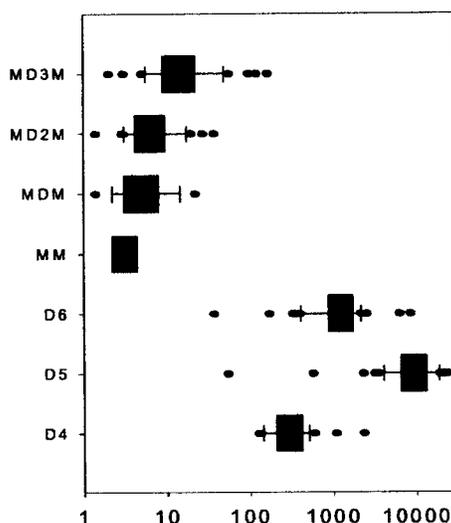
The linear dimethylsiloxanes measured here (MM, MDM, MD2M and MD3M) occurred in significantly lower concentrations than the cyclic analogues, with an average concentration of 110 ng/g dw (sum of 4). The concentrations are illustrated in Figure 9. (Please note the different scales

on the concentration axes in the left and right part of the figure). The highest concentrations were observed in two Danish samples, primary and digested sludge from Lynetten sewage treatment plant, Copenhagen, 1 060 and 190 ng/g dw respectively. Apart from the Icelandic samples, the lowest concentrations were found in Ellinge and Floda sewage treatment plants, Sweden with 6 and 8 ng/g dw respectively.

In all samples the relative concentration increased in the order MM, MDM, MD2M, MD3M. MM, which is a high production volume (HPV) chemical, was not detected in any of the sludge samples. MM may be too volatile to be able to accumulate in sludge.

The concentration of D5 observed in the current study are comparable to those found in the national sampling programme within the recent Swedish screening study of siloxanes (Kaj et al., 2005), where an average concentration of 11 000 ng/g dw (D5) was obtained. D6 and D4 followed with concentrations close to 3000 and 300 ng/g dw respectively. The concentrations of the linear analogues were substantially lower: MM and MDM <2, MD2M 8-16 and MD3M 24-46 ng/g dw. In addition to the national sampling programme, regional samples were also collected by different county administrations adding up to a total sum of 51 sludge samples collected at municipal sewage treatment plants with regional distribution and varying size. As a comparison to results in this study, the Swedish data are summarised as box plots in Figure 12. D5 and D6 were detected in all samples and the linear siloxanes were detected in 47 of the samples.

Figure 12. Concentration of siloxanes (ng/g dw, logarithmic scale) in sludge from Swedish municipal sewage treatment plants. The lower and upper boundaries of the box represent the 25- and 75-percentiles, and the line within the box is the median concentration. The whiskers represent the 10- and 90-percentiles, and the dots are outliers.

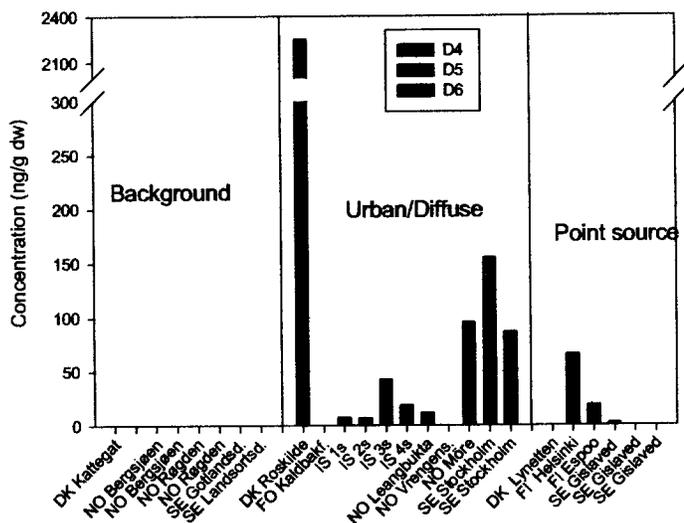


5.1.3 Soil and sediment

Two soil samples from Faroese landfills were analysed for siloxanes. The concentrations of all siloxanes were below the detection limits, which varied from 0.1 to 10 ng/g dw for different siloxanes.

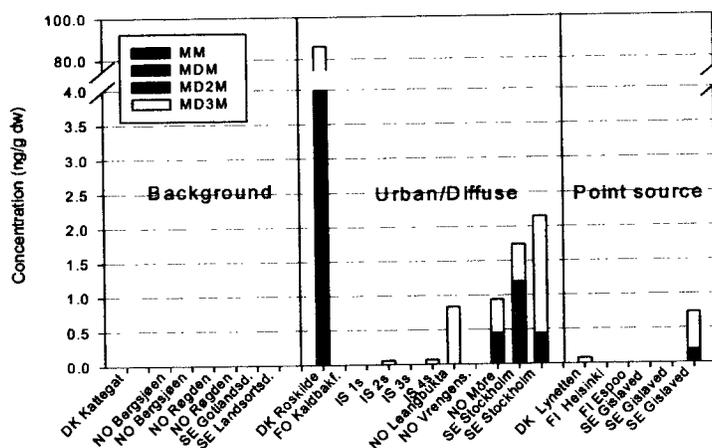
The results of the siloxane measurements in sediment are shown in Figure 13 (cyclic siloxanes) and in Figure 14 (linear siloxanes), where the total concentrations as well as the relative distribution between the different siloxanes are illustrated. As for the sludge samples, there was a great variation in sediment concentrations. The highest concentrations occurred in sediments collected close to urban areas.

Figure 13. Concentrations of cyclic siloxanes in sediment.



The cyclic siloxanes were not detected in any of the samples collected at the background sites, but in all but one sample from urban areas. The sample collected near Roskilde in Denmark contained the highest concentrations (2 300 ng/g dw), which was almost 15 times higher than the second highest concentration from Essingen in Stockholm, Sweden (160 ng/g dw). D5, which dominated in all sludge samples, was the dominating siloxane also in the sediments.

Figure 14. Concentrations of linear siloxanes in sediment samples. MD3M dominates all samples but one from Stockholm. The inserted break hides the MD3M contribution to the sample from Roskilde.

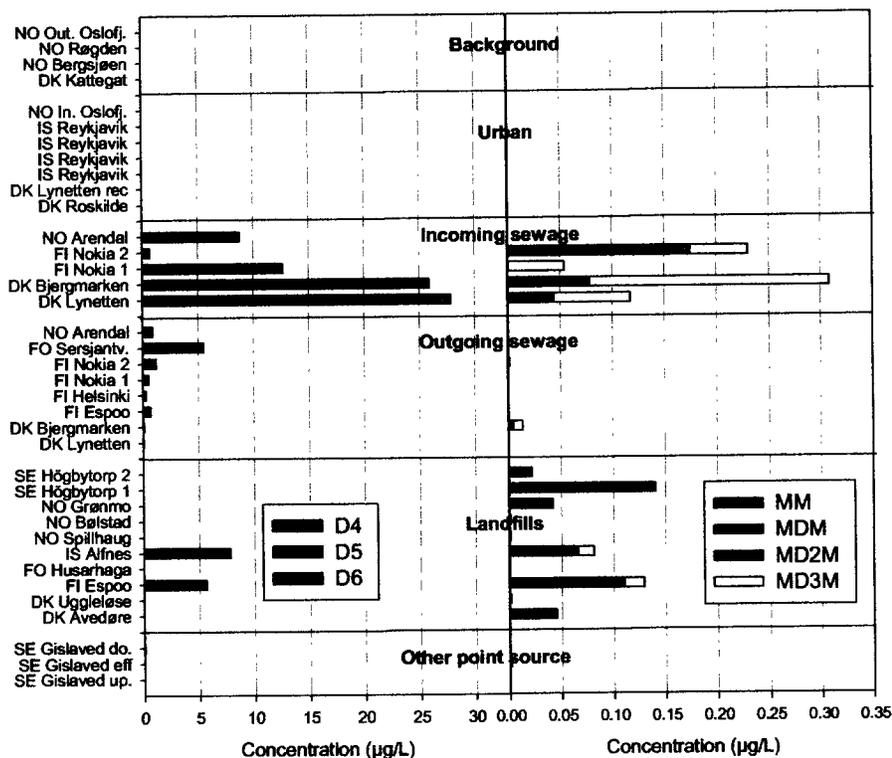


Similar to the cyclic siloxanes, the linear siloxanes were only detected in urban/diffuse samples and in samples close to potential point sources, and generally in those samples that showed the highest concentration of cyclic siloxanes. Like the cyclic siloxanes, the sediment sample from Roskilde contained the highest concentrations, 87 ng/g dw, which was about 40 times higher than the second highest concentration (2.2 ng/g dw) in sediments from Stockholm (Figure 14). MD3M was the dominating linear siloxane in all sediment samples except for one sample from Stockholm, Sweden.

5.1.4 Water

The concentrations of siloxanes in water are given in Appendix 2 and shown in Figure 15, where the total concentrations as well as the relative distribution between the different siloxanes are illustrated.

Figure 15. Concentrations of cyclic (left) and linear (right) siloxanes in water. Note the different scales on the x-axes.



There were no detectable amounts of siloxanes in the water samples collected at the background or urban sites. Neither were they found in the three water samples from Swedish point sources. The detection limit was below $0.1 \mu\text{g/l}$ for the individual cyclic siloxanes and below $0.006 \mu\text{g/L}$ for the linear siloxanes.

Substantial amounts of both cyclic and linear siloxanes occurred in samples from incoming water to sewage treatment plants. There was, however, a great variation in concentrations among the different samples. The concentrations of the cyclic siloxanes were about 100 times higher than the concentrations of the linear analogues.

In most of the samples of incoming water the relative distribution of D4, D5 and D6 was similar to that in sludge with D5 as the dominating siloxane. A different distribution was found in one of the samples from Nokia, Finland, representing wastewater from a tyre manufactory, where D4 and D6 occurred in substantial amounts. The distribution of the linear siloxanes varied for the different samples, MD3M was highest in the samples from Denmark and one of the Finnish samples (tyre wastewater),

while MM was present in highest concentration Finnish sample from Nokia, representing wastewater from the floor manufactory. Significantly lower concentrations were detected in outgoing water from sewage treatment plants. Linear siloxanes were only detected in one of these samples.

Cyclic siloxanes were only detected in two of the samples from landfills, while the linear siloxanes were found in 6 of the samples. Alfnes landfill in Iceland, which contained relatively high concentrations of cyclic siloxanes, receives all the waste from the Reykjavik area, as well as dry material from the two plants Ananaust and Klettagarðar. In contrast to water samples from sewage treatment plants, MM dominated in the landfill samples, although the absolute concentrations were lower than concentrations of cyclic compounds in the samples where both were detected.

The shift from cyclic to linear (especially MM) dominance in the water samples from landfills relative to sewage treatment plant samples is noteworthy. The physical-chemical properties of MM indicates that it is more water soluble than other siloxanes, but also more volatile, thus the properties cannot explain the observed pattern. Further measurements in leachate and exploration of the degradation route of other siloxanes under landfill-like conditions could help addressing this issue.

5.1.5 Biota

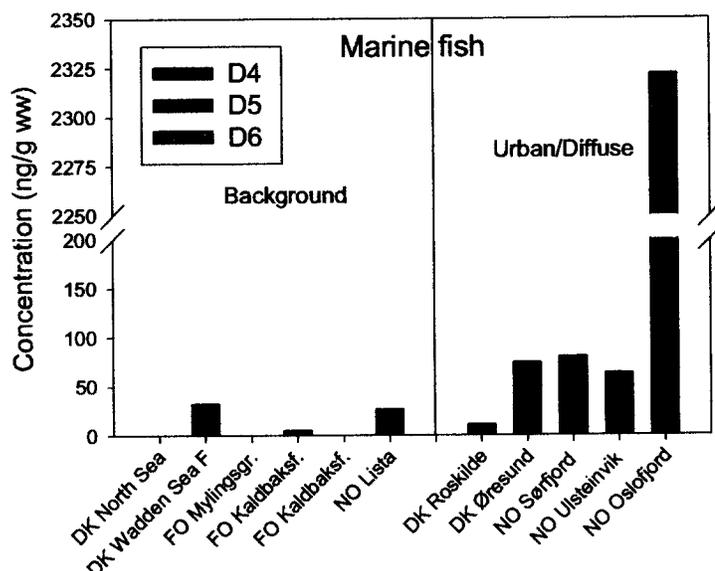
The results from the measurements in biota are presented below. The results have been divided into the subgroups marine and freshwater fish, seabird eggs and marine mammals, and results are illustrated by source type in the respective group. All results and detection limits are also presented in Appendix 2.

The cyclic siloxanes D4, D5 and D6 were detected in fish samples and marine mammals. D3 was detected in a few samples, but as the concentrations were below the limit of quantification in all cases they are not included in the figures. The concentrations of other siloxanes were between the limit of detection and the limit of quantification in a number of samples. For details on the individual samples, see Appendix 2.

Marine fish

Figure 16 shows the measured concentrations of cyclic siloxanes in liver from marine fish species. The species are not specified in the figure, but include eelpout, flounder, cod, sculpin and dab (Appendix 2). Results are presented based on type of sampling site. With one exception (see below) no linear siloxanes were detected in marine fish samples, LoD varying between 0.3 and 0.5 ng/g ww.

Figure 16. Concentrations and distribution of cyclic siloxanes in marine fish liver. Some of the reported concentrations were between the limits of detection and quantification and thus have a larger uncertainty. See Appendix 2 for details.



The concentrations in marine fish liver were fairly variable, typically in the range of <math><5 - 100 \text{ ng/g ww}</math>. Only one sample of cod liver (9 livers pooled) from Inner Oslofjord, close to the city center, exceeded this range with a concentration of

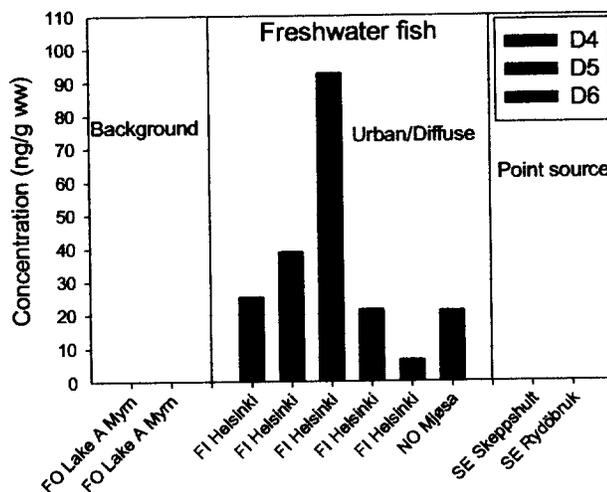
Siloxanes were mainly detected in marine fish liver samples from sites representing urban/diffuse sources and only a few background samples showed detectable levels. The concentrations are not directly comparable as they represent different species of varying age and gender. However, diffuse sources may contribute to the observed concentrations in marine fish.

Freshwater fish

Figure 17 shows the measured concentrations of cyclic siloxanes in liver from freshwater fish species. The species are not specified in the figure, but include arctic char, brown trout, pike and vendace (Appendix 2). Results are presented based on type of sampling site. No linear siloxanes

were detected in freshwater fish samples, LoD varying between 0.3 and 0.5 ng/g ww.

Figure 17. Concentrations and distribution of cyclic siloxanes in freshwater fish liver. Some of the reported concentrations were between the limits of detection and quantification and thus have a larger uncertainty. See Appendix 2 for details.



Just like the marine fish liver samples, the concentrations in freshwater fish varied in the range <5-100 ng/g ww. D5 was the dominating siloxane in all freshwater fish liver samples and D6 was not at all detected. Siloxanes were only detected in freshwater fish liver samples from sites representing urban/diffuse sources and not at all in the background samples from Faroe Islands or in samples representing point sources in Sweden. The detected concentrations are comparable, as all the Finnish samples are pikes, however the Norwegian sample is vendace. Urban and diffuse sources appear to contribute significantly to the observed concentrations in freshwater fish.

Seabird eggs

No siloxanes were detected in samples of seabird eggs (Fulmar, black guillemot and herring gull) from Sweden and Faroe Islands. The detection limits for the seabird eggs are given in Table 16 and in Appendix 2.

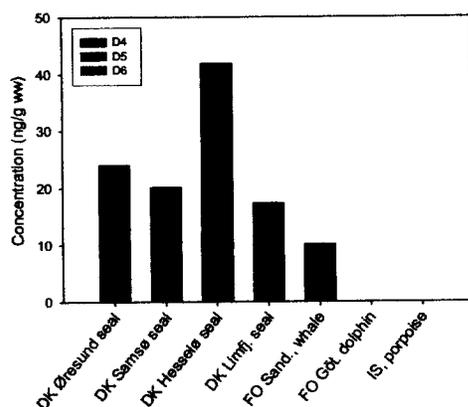
Table 16. Limit of detection for seabird eggs (ng/g wet weight)

D3	D4	D5	D6	MM	MDM	MD2M	MD3M
50	5	5	5	0.4	0.3	0.4	0.5

Marine mammals

Figure 18 shows the measured concentrations of cyclic siloxanes in marine mammals. No linear siloxanes were detected in mammals, LoD varying between 0.3 and 0.5 ng/g ww.

Figure 18. Concentrations of siloxanes in marine mammals. Some of the reported concentrations were between the limits of detection and quantification and thus have a larger uncertainty. See Appendix 2 for details



Among the mammal samples, seal and pilot whale showed detectable concentrations of siloxanes, and D5 was the dominating compound. In fact, other cyclic siloxanes were only detected in one seal sample from Hesselø in Denmark. All the Danish seal samples were taken away from cities. Some of the reported concentrations were between the limit of detection and the limit of quantification, see Appendix 2 for details. D3 was detected in one sample, but it has not been included in the figure as it was below the limit of quantification (see Appendix 2).

5.2 Comparison of biota and water concentrations

A comparison of observed concentrations in water and biota can give useful information on the potential for biotic uptake of siloxanes. The results in the current study show water concentrations of D5 that vary between limit of detection (<0.02 µg/L) and 26 µg/L. Excluding incoming sewage water (to which aquatic organisms are not likely to be exposed) and non-detected concentrations gives a range of 0.06-5.4 µg/L. Using a BCF of 5300 (Table 2), this would represent an expected range of 300 – 28000 ng/g ww in fish. The observed range for detected concentrations in biota is 6 – 2200 ng/g ww, including values below the limit of

quantification. Consequently, the observed concentrations in biota are 10 to 100 times lower than would be expected from the observed concentrations in water (neglecting the influence of uptake via the food chain). This can not be regarded as a high discrepancy considering that most of the detected water concentrations were related to effluent water. Water samples from seawater and freshwater were generally below detection limits. Biota and water was in most cases not sampled in the same region, but also at those locations (Lynetten, Bjergmarken) where fish and recipient water was sampled in the same area, the water concentrations were generally below the detection limit. Regarding this, the levels we find in biota may be a confirmation of the expected bioconcentration.

On the whole, data show that siloxanes have the potential to bioaccumulate. It is, however, not possible to state whether the observed occurrence in aquatic organisms is due to uptake via water, via the food chain or a combination of these processes. In order to draw such conclusions, it would be necessary to analyse surface water and organisms belonging to different trophic levels at the same sampling location.

5.3 Concentration patterns at geographically related sites

In some Nordic countries several samples, representing different matrices, were collected in the same area. Examples of sites following this sampling strategy are given below.

5.3.1 Copenhagen, Lynetten area

In the area surrounding Lynetten sewage treatment plant (STP) in Denmark, samples of water, sludge, sediment and fish were collected and included in the study. Elevated concentrations of all siloxanes except MM were found in incoming water and sludge in the STP. In effluent water, only D5 was detected and in significantly lower concentrations, 0.2 % of that in the influent. The cyclic siloxanes were below the detection limits in the recipient water and sediment samples from this area but they were detected in fish collected in the recipient area (Table 17).

Table 17. Detected concentrations in geographically related samples from the Lynetten area, Denmark. Biota values written in italic letters represent concentrations below the limit of quantification.

Sample ID	Sample type	Unit	D3	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3915	Waste water influent	µg/L		0.28	26	1.6	<0.01	0.0034	0.041	0.073
MR-3917	Primary sludge	ng/g DW		740	27000	1100	<3	64	450	550
MR-3918	Digested sludge	ng/g DW		470	50000	2800	<1	8	44	140
MR-3916	Waste water, effluent	µg/L			0.063			<0.001	<0.02	
MR-3717	Water, recipient	µg/L				<0.0005	<0.04			
MR-3718	Sediment, recipient	ng/g DW				<0.05	<5			0.08
MR-3714	Sediment, distant	ng/g DW				<0.03	<3			
05/400	Fish, recipient area	ng/g ww	90.4	13.5	52.3	8.73		<0.3	<0.5	

5.3.2 Roskilde, Bjergmarken sewage treatment plant

At the Bjergmarken STP in Roskilde, samples of wastewater, recipient water, sediment and fish as well as air were collected. D4, D6, D5, MD2M, and MD3M were detected in influent water and the latter three also in the effluent. The concentration of D5 in the effluent was 0.4 % of that in the influent while the concentrations of MD2M and MD3M were 6 and 4 % of that in the influent, respectively. Siloxanes were not found in recipient water, but the sediment was polluted. The relative concentrations of D4, D5, and D6 in the sediment were similar to the pattern in the influent water. D4 was also found in the fish sample. The air samples contained elevated concentrations compared to background areas (Table 18).

Table 18. Detected concentrations in geographically related samples from the Bjergmarken area, Denmark.

Sample ID	Sample type	Unit	D3	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3920	Waste water influent	µg/L		0.60	24	1.3	<0.01	<0.01	0.078	0.23
MR-3921	Waste water, effluent	µg/L		<0.06	0.092		<0.04	<0.0005	0.0045	0.0087
458-nilu238	Air at STP	µg/m ³		0.66	1.33	0.17	<	<	<	<
MR-3716	Water, recipient	µg/L					<0.0005	<0.04		
MR-3723	Sediment, recipient	ng/g DW		84	2000	170	<0.3	<0.71	29	57
05/399	Fish, recipient	ng/g ww	<50	10.7			<0.3	<5		
MR-3713	Water, distant	µg/L					<0.0005	<0.04		

5.3.3 Finland Nokia

At Nokia in Finland, wastewater and sludge samples from the industry as well as from STPs were included. One air sample was also collected. The sample from the floor industry contained all the siloxanes analysed. The concentration in water after treatment was 16% of that in untreated water and concentrations of cyclic siloxanes in STP sludge were the third highest in among all of the Nordic samples. The air concentration at the STP

was in the same order of magnitude as the Danish background sample but about ten times higher than background levels found in the Swedish screening study (Kaj et al., 2005) (Table 19).

Table 19. Detected concentrations in geographically related samples from the Nokia area, Finland.

Sample ID	Location	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3940	Tyre industry waste water pipeline	Water	µg/L	3.7	5.3	3.7		<0.007		0.054
MR-3941	Floor industry 1B	Water	µg/L	0.25	0.33	0.12	0.12	0.014	0.041	0.055
MR-3942	Kullonvuori STP effluent 1	Water	µg/L	<0.06	0.48	0.03		<0.001	<0.004	
MR-3943	Kullonvuori STP Effluent 2	Water	µg/L	0.11	0.98	0.045	<0.001	<0.001	0.0012	<0.005
MR3932	Nokia Kullonvuori	Sludge	ng/g DW	960	30000	3500	<2	3	12	14
344-nilu106	Nokia City ww-treatment	Air	µg/m ³	1.08	1.27	0.2	<	<	<	<

Conclusively, it seems that STPs and some industrial sources (floor, tyre industry) are important sources for the occurrence of siloxanes in the environment. Possibly, sewage treatment plants located near certain industry types receive a larger amount of siloxanes than others.

5.4 Concentrations of D5 in sludge compared to other contaminants

Sludge is a common matrix for analysis of organic pollutants as it receives a fair amount of chemicals used in households and consumer products, as well as in industrial processes. Numerous substances have been detected in sludge in concentrations high enough to raise concern, as sludge has been proposed to be used as a source of nutrients within agriculture. The average concentration of D5 in biological sludge from STPs in the current study (12 samples) was 27 000 ng/g dw and in the similar study covering 54 STPs in Sweden (Kaj et al., 2005), the average concentration of D5 was 11 000 ng/g dw. These rather high concentration levels can be compared to those for other widespread organic contaminants found in sludge. Due to physico-chemical properties this does not necessarily imply that the effects are the same. In a recent study concerning sludge from 20 Swedish STPs di-(ethylhexyl)-phthalate (DEHP) averaged to 49 000 and 4-nonylphenol to 9 000 ng/g dw (Bignert and Remberger, 2005). In another study including 23 Swedish STPs the average concentration of 4-nonylphenol was 15 000 ng/g dw (Remberger et al., 2004). Thus the concentration level of D5 found in the present study is similar to that for 4-nonylphenol and somewhat lower than that for DEHP. Siloxanes therefore add to the list of organic pollutants that may make it problematic to use municipal sewage sludge in a sustainable way, i.e. as a source of nutrients in agriculture.

5.5 Toxicity and ecotoxicity

No Observed Effect Concentrations (NOECs) for D4 (Table 5) were only exceeded in samples representing incoming sewage water to sewage treatment plants (STPs). These levels were, however, significantly reduced in the outgoing water from the same STPs. In the latter, D4 levels were about one order of magnitude lower than any NOECs found in literature. In landfill leachate from Iceland, the concentration of D4 (1.1 µg/L) was in the lower end of the NOEC range for *Daphnia Magna* (1.7 – 15 µg/L). The estimated chronic value (ChV, see Table 6) for D5 was exceeded in two samples of incoming sewage water, but in no other water samples. The ChV was not exceeded for any other siloxanes.

Detailed ecotoxicity data was only obtained for D4. D5 is, however, a suspected carcinogen and may also have other toxic effects in ecosystems. Toxic effects of D6 are less known but its similarity to the other cyclic siloxanes still makes it interesting. Uptake in biota is known to decrease with increasing molecular size due to difficulties when passing through the cell membrane, which may reduce the toxic effects of D5 and D6 relative to D4. The concentrations of D5 and D6 were highest in incoming water to sewage treatment plants and, as in the case of D4, substantially reduced in outgoing water. The concentrations of D5 and D6 in leachate water as well as in sewage water were higher than the concentrations of D4. In two leachate water samples (Alfnes and Ämmässuo) D5 and D6 reached levels that were in the same range as several NOECs for D4. In some effluent samples D5 and D6 were detected in concentrations slightly lower than NOECs for D4. Since only limited amounts of toxicity data are available for other siloxanes than D4 and No Observed Effect Levels and Chronic Values are estimated based on a limited amount of data (and as such objects of some uncertainty) the possibility of effects in the local environment close to emissions should not be excluded. No detailed toxicity data could be found for linear siloxanes and measured concentrations were also lower than for cyclic siloxanes.

Siloxanes were not detected in any background samples or in the recipient water outside Lynetten sewage treatment plant. Siloxanes reaching the aquatic environment are probably quickly diluted in the receiving water. Acute and possibly also chronic toxic effects are hence probably limited to locations in very close proximity of outlets from STPs, landfills or other point sources. Observed levels in fish and sediment still show that siloxanes are present in several aquatic systems, and are likely to occur also in the water phase, even though in concentrations lower than the limit of detection.

6. Conclusions

Siloxanes were found in all the analysed media but soil: air, water, sediment, sludge and biota. The results indicate that there is a general pollution of siloxanes in the Nordic countries. There was, however, a great variation in concentrations. The cyclic siloxanes occurred in all media in significantly higher concentrations than the linear siloxanes.

D5 was the dominating siloxane in most samples, which is not in agreement with data on use in the Nordic countries, where the consumption of D5 and D4 is fairly equal. D4 occurred in the highest concentration in the air samples with the exception of all Norwegian samples (4), one Icelandic sample, one Finnish sample, two Danish samples and one Swedish sample.

The results of air measurements in urban areas indicate a regional variation, with the highest concentration in Norway and the lowest in Sweden. The air concentrations were highest inside sewage treatment plants, and elevated levels were commonly observed in other matrices surrounding STPs. No other obvious point sources were found.

Diffuse sources seem to be most important for the observed concentrations of siloxanes in the Nordic environment. The concentrations were generally elevated in urban areas and in areas close to sewage treatment plants. The mean concentration of D5 in biological sludge from STPs (27 000 ng/g dw) is in the lower range of what has been found elsewhere (see Table 8) and comparable to that of the widespread contaminant 4-nonylphenol.

The concentrations in fish liver were fairly variable. Siloxanes were mainly detected in fish liver samples from sites representing urban/diffuse sources and only a few background samples showed detectable levels. One pooled sample of cod liver from Inner Oslofjord showed highly elevated concentrations. On the whole, biota data indicate that siloxanes may bioaccumulate.

No Observed Effect Concentrations for D4 and estimated Chronic Values for D5 were only exceeded in samples representing incoming sewage water to sewage treatment plants. These levels were, however, significantly reduced in the outgoing water from the same treatment plants. Since only limited amounts of toxicity data are available for other siloxanes than D4 and No Observed Effect Levels and Chronic Values are estimated, the possibility of effects in the local environment close to emissions should therefore not be excluded.

Conclusively, siloxanes are present as a common pollutant in the Nordic environment and in many different matrices. They seem to be emitted through diffuse pathways and they enter the aquatic food chain. At pre-

sent, the observed concentrations are not alarmingly high, and many background sites seem to be non-contaminated. However, the use of siloxanes is extensive and it is possible that continued use will lead to increased environmental levels, eventually reaching effect concentrations.

7. Acknowledgements

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Sammanfattning

Den här presenterade "screeningstudien" avseende spridning och förekomst av flyktiga metylerade siloxaner i den nordiska miljön involverar sex länder: Danmark, Färöarna, Finland, Island, Norge och Sverige.

Siloxaner hör till en grupp ämnen som används i ett stort antal olika industriella applikationer samt i konsumtionsprodukter såsom additiv i bränsle, bilvaxer, rengöringsmedel samt antiskummedel. Dessutom förekommer de i olika hygien- och kosmetikaprodukter. Den omfattande användningen, de många applikationsområdena samt den höga flyktigheten har orsakat intresse för siloxaner inom flera olika miljövetenskapliga discipliner.

Till följd av deras omfattande användning sprids siloxaner sannolikt både via punktkällor samt via diffusa källor, och förväntas kunna återfinnas överallt i miljön. Nyligen utförda studier har pekat på att siloxaner kan ha direkta eller indirekta toxiska effekter på olika biologiska processer.

Screeningstudien inkluderade följande ämnen: de linjära siloxanerna hexametyl-disiloxan (MM eller HMDS), oktametyltrisiloxan (MDM), dekametyltetrasiloxan (MD2M), dodekametylpentasiloxan (MD3M) och de cykliska siloxanerna oktametylcyclotetrasiloxan (D4), dekametylcyclopentasiloxan (D5) och dodekametyl-cyklohexasiloxan (D6). Dessutom analyserades hexametylcyclotrisiloxan (D3) i biota. Detta ämne är starkt flyktigt och förorsakar analytiska svårigheter, varför den ej analyserades i övriga matriser.

Siloxaner hittades i alla matriser som studerats förutom jord: luft, vatten, sediment, slam och biota. Resultaten tyder på att siloxanes är allmänt förekommande föroreningar i den nordiska miljön. Det fanns dock en stor variation i observerade koncentrationer. De cykliska siloxanerna förekom i avsevärt högre koncentrationer än de linjära i alla matriser som analyserats. Tabellen nedan visar de uppmätta intervallen av olika siloxaner i olika matriser.

Ämne	Luft ($\mu\text{g}/\text{m}^3$)	Vatten ($\mu\text{g}/\text{L}$)		Slam ($\text{ng}/\text{g dw}$)	Jord ($\text{ng}/\text{g dw}$)	Sediment ($\text{ng}/\text{g dw}$)	Biota ($\text{ng}/\text{g ww}$)
		Av- lopp/industriellt*	Kust/ vattendrag				
MM	<0.004	<0.0005-0.14	<0.0005-<0.0006	<0.5 - <3	<0.1	<0.02-<0.7	<0.4
MDM	<0.008	<0.0005-0.014	<0.0005-<0.0006	<1-64	<0.1	<0.02-<0.7	<0.3
MD2M	<0.006	<0.0005-0.078	<0.0005-<0.0006	1-450	<0.1	<0.02-29	<0.4 - 1.1
MD3M	<0.02	<0.004-0.23	<0.002-<0.004	3-550	<0.1	<0.02-57	<0.5
D3	n.a**	n.a	n.a	n.a	n.a	n.a	<50-90.4***
D4	0.08-4.0	<0.06-3.7	<0.04-<0.09	96-960	<6-<10	<3-84	<5-70
D5	0.05-19	<0.04-26	<0.02-<0.05	1100-89000	<3-<5	<2-2000	<5-2200
D6	0.02-2.1	<0.04-3.8	<0.02-<0.05	220-11000	<2-<4	<1-170	<5-74

*Proverna representerar in- och utgående avloppsvatten till och från reningsverk, lekavatten från deponier samt industriellt dagvatten **n.a = ej analyserad ***Detekterade halter låg under kvantifieringsgränsen

D5 var den dominerande siloxanen i samtliga matriser utom luft, där D4 generellt förekom i högre halter. Detta överensstämmer inte med konsumtionsdata för Norden, där användningen av D5 och D4 uppges vara ganska lika. Resultaten från luftmätningarna indikerar en regional variation, med högst koncentrationer i Norge och lägst i Sverige. Uppmätta koncentrationer av D5 i luft inne i reningsverk var markant förhöjda i jämförelse med andra luftprover, likaså var koncentrationer av D5 i andra matriser i anslutning till reningsverk förhöjda.

Diffusa källor tycks vara av störst betydelse för de observerade siloxan-koncentrationerna. Halterna var allmänt sett förhöjda i urbana områden samt i områden nära reningsverk. Medelhalten av D5 i slam är jämförbar med vad som observerats för 4-nonylfenol, men detta innebär inte nödvändigtvis att det er likedana effekter som kan observeras.

Halterna i fisklever var varierande. Siloxaner hittades främst i fiskprover från lokaler som representerar urbana/diffusa källor och endast ett fåtal bakgrundsprover uppvisade detekterbara halter. Ett poolat prov av torsklever från inre Oslofjorden hade starkt förhöjda halter. Totalt sett indikerar biota data att siloxaner kan bioackumuleras.

No Observed Effect Concentrations (NOECs) för D4 samt uppskattade sk "Chronic Values" (ChV) för D5 överskreds endast i prover av inkommande avloppsvatten till reningsverk. Dessa koncentrationer reducerades avsevärt i utgående vatten från samma reningsverk. Då tillgången på toxicitetsdata är ytterst begränsad för andra siloxaner än D4 och NOECs samt ChV är uppskattade värden, kan det inte helt uteslutas att effekter kan uppkomma lokalt i direkt anslutning till emissionspunkter.

Sammanfattningsvis kan konstateras att siloxaner är allmänt förekommande miljöföroreningar i den nordiska miljön och hittas i många olika matriser. De verkar släppas ut framför allt genom diffusa spridningsvägar och de tas upp i den akvatiska näringskedjan. I dagsläget är halterna inte alarmerande höga, och många bakkgrundsstationer verkar vara fria från föroreningar. Användningen av siloxaner är dock omfattan-

de och det går inte att utesluta att fortsatt användning skulle kunna leda till ökande halter som på lång sikt kan nå effektnivåer.

Appendix 1: Sample characteristics

Table A 1. Sample characteristics for air samples.

Sample ID	Country	City	Location	X coord.	Y coord.	Sampling date	Sampling time
345-nilu237	Denmark		Jagtvejen	55 40.5 N	12 33.8 E	04.01.05	1.05-2.05 pm
458-nilu238	Denmark		Bjergmarken STP	55°39N	12°03E	31.12.04	10.45-11.45 am
337-nilu239	Denmark		Sepstrup Sande	56 05 N	9 25 E	12.01.05	2.30-3.57 pm
132-nilu240	Denmark		H.C. Ørsted Institute	55 42N	12 34 E	21.12.04	11.37 am-1.37 pm
150-nilu241	Faroe Islands	Torshavn	Downtown, Steinatun	62 00,7 N	6 46,5 E	15.12.04	3.50-4.34 pm
225-nilu242	Faroe Islands	Torshavn	Sersjantvikin STP	62 00,5 N	6 45,7 E	16.12.04	3.16-4.10 pm
344-nilu106	Finland		Nokia City STP	59 10,8 N	25 57,45 E	28.01.05	2.00-3.00 pm
142-nilu105	Finland		Espoo landfill	60 11,58 N	24 74,84 E	24.05.05	2.00-3.00 pm
69-nilu232	Iceland	Reykjavik	Urban	64 09 00	21 58 00		30 minutes
209-nilu233	Iceland	Reykjavik	Urban	64 09 00	21 58 00		30 minutes
340-nilu234	Iceland	Reykjavik	Urban	64 09 00	21 58 00		30 minutes
451-nilu451	Iceland	Reykjavik	Urban	64 09 00	21 58 00		30 minutes
150-nilu108	Norway		Bekkelaget STP	59 53 53,12	10 48 54,96	13.05.05	1.24-1.55 pm
416-nilu109	Norway		Bekkelaget STP	59 53 53,12	10 48 54,96	13.05.05	12.50 am-1.20 pm
149-nilu111	Norway	Manglerud	Manglerud	59 52 57,87	10 46 02,10	13.05.05	5.50-6.35 pm
50-nilu110	Norway	Oslo	Oslo Central Station	59 54 33,70	10 45 17,00	27.05.05	7.30-8.00 pm
419-nilu206	Sweden	Högbytorp	Högbytorp landfill windside	6603702	1603007	16.02.05	12.41 am-2.41 pm
204-nilu208	Sweden	Högbytorp	Högbytorp landfill windside	6603702	1603007	16.02.05	10.40-12.40 am
6-nilu229	Sweden	Gislaved	Mossarps recycling site	6356021	1364021	16.11.04	08.00-10.00 am
453-nilu230	Sweden	Gislaved	Mossarps recycling site	6356021	1364021	16.11.04	08.00-10.00 am
415-nilu210	Sweden	Göteborg	Kapellplatsen	57° 41N	11° 58E	03.02.05	2.08-4.08 pm
228-nilu216	Sweden	Göteborg	Kapellplatsen	57° 41N	11° 58E	04.02.05	8.32-10.54 am
502-nilu214	Sweden	Stockholm	Hudiksvallsgratan	59° 21N	18° 2E	02.12.04	3.40-5.40 pm
509-nilu218	Sweden	Stockholm	Hudiksvallsgratan	59° 21N	18° 2E	02.12.04	09.30 am-3.30 pm

Table A 2. Sample characteristics for soil, sediment, sludge and water samples.

Sample ID	Country	City	Site	Notes	Sampling date	Matrix	DW (%)	Co-ordinates
MR-3895	Faroe islands		Havnardalur	Abandoned landfill	20.12.04	Soil	54.8	62°00.884'N, 06°51.049'W
MR-3899	Faroe islands	Torshavn	Húsarhaga Landfill	Operating landfill for the Torshavn area	20.12.04	Soil	66.1	62°01.692'N, 06°48.308'W
MR-3714	Denmark		Coast area Kattegat		02.10.04	Sediment	77.4	56.51.4N, 10.47.6E
MR-3718	Denmark	Kobenhavn	Coast area Øresund, Lynetten		26.10.04	Sediment	65.3	55.43.60N 12.37.86E
MR-3723	Denmark	Roskilde	Coast area	Innerford	19.10.04	Sediment	12.5	55.39.55, 12.03.79
MR-3901	Faroe islands		Kaldbaksfjørður, KA05	Influenced by pollution from unidentified sources	30.08.04	Sediment	39.8	62°03.347'N, 6°49.301'W
MR3945	Finland	Helsinki	Vanhankaupunginlahti (Vakal), 0-6 cm	Old City Bay in front of downtown. Historical pollution by former burning plant for hazardous wastes etc.	21.10.04	Sediment	31.2	60° 11,75'N, 24° 59,59' E
MR3946	Finland	Espoo	Espoo-Kirkkonummi coastal sea area 10-15 km west of Helsinki City, 0-6 cm	Monitoring point UYK2. Background polluted by strong yachting and shipping activities	13.10.04	Sediment	21.4	60° 01,57' N, 24° 36,42' E
MR-3909	Iceland		Sediment 1s, 050203		03.02.05	Sediment	78.5	64.09.00, 21.58.00
MR-3910	Iceland		Sediment 2s, 050203		03.02.05	Sediment	58.8	64.09.00, 21.58.00
MR-3912	Iceland		Sediment 3s, 050203		03.02.05	Sediment	53.8	64.09.00, 21.58.00
MR-3913	Iceland		Sediment 4s, 050203		03.02.05	Sediment	59	64.09.00, 21.58.00
MR-3740	Norway		Lake Bergsjøen	Background area		Sediment	6	N:60,534 E: 11,350
MR-3741	Norway		Lake Bergsjøen	Background area		Sediment	6	N:60,534 E: 11,350
MR-3746	Norway		Lake Røgden	Background area		Sediment	7.1	N:60,401 E: 12,555
MR-3747	Norway		Lake Røgden	Background area		Sediment	8	N:60,401 E: 12,555
MR-3869	Norway		Leangbukta	2-4 m (04/989)	07.10.04	Sediment	27.3	N: 59,835 E:10,473
MR-3870	Norway		Vrengansundet	37 m 0-2 cm (04/1424)	26.09.04	Sediment	42.1	N:56,169 E: 10,388
MR-3871	Norway	Møre, Romsdal	Brødrene Sunde Verft	0-1 cm (04/1427)	16.09.04	Sediment	47.3	N: 62,458 E: 6,350
MR-3774	Sweden	Gislaved	Nissan	storm water effluent (sand)	16.11.04	Sediment	76.4	6353937,1364076
MR-3776	Sweden	Gislaved	Nissan	Downstreams storm water effluent (sand)	16.11.04	Sediment	81.8	6352791,1363001
MR-3772	Sweden	Gislaved	Nissan	Upstreams storm water effluent (sand)	16.11.04	Sediment	42.8	6356021,1364021
MR-3314	Sweden	Stockholm	Easingen	0-2 cm	20.12.03	Sediment	16.5	6579997,1625005
MR-3316	Sweden	Stockholm	Riddarfjärden	0-2 cm	20.12.03	Sediment	14.3	6580235,1627362
MR-3750	Sweden		Ö Gotlandsdjupet	0-2 cm 121m SGU 04-0058	2004	Sediment	7.2	6216805,1697299
MR-3753	Sweden		Ö Landsortsdjupet	0-2cm 403m SGU 09j-0841	2004	Sediment	4.2	6513868,1619918
MR-3917	Denmark	Kobenhavn	Lynetten STP	Primary		Sludge	6.9	55°41.7N, 12°37'E

Sample ID	Country	City	Site	Notes	Sampling date	Matrix	DW (%)	Co-ordinates
MR-3918	Denmark	Kobenhavn	Lynetten STP	Digested		Sludge	20.5	55°41.7N, 12°37E
MR-3897	Faroe Islands	Torshavn	Sersjantvikin STP		25.01.05	Sludge	23.2	62°00.495'N, 06°45.714'W
MR3932	Finland	Nokia	Nokia City STP Kulloonvuori	Sources: Wastewaters from several industries e.g. tyre, floor material and textile manufacturing. 24 000 pe.	02.02.05	Sludge	12.5	59° 10,80' N, 25° 57,45' E
MR3935	Finland	Helsinki	Helsinki City, Viikinkaari STP	Municipal+urban+industrial sewage water. 960 000 pe.	27.01.05	Sludge	31.0	60° 27,95' N, 24° 87,35' E
MR3936	Finland	Espoo	Espoo City, Suomenoja STP	Potential Siloxane sources: Waste (influent) water from perfume manufacturing and leachate water from the Ämmässuo landfill area. 240 000 pe.	28.01.05	Sludge	27.6	60° 11,58' N, 24° 74,84' E
MR3937	Finland	Pornainen	Pornainen municipal STP	Rural, 700 pe	26.01.05	Sludge	13.6	60° 46,08' N, 25° 39,89' E
MR3938	Finland	Porvoo	Porvoo City STP	Urban industrial, 20 000 pe.	26.01.05	Sludge	18.2	60° 37,74' N, 25° 61,42' E
MR-3862	Iceland	Reykjavik	Klettegardar STP	Not biological sludge	13.12.04	Sludge	26.6	64 09 00, 21 58 00
MR-3865	Iceland	Reykjavik	Ananaust STP	Not biological sludge	13.12.04	Sludge	12.5	64 09 00, 21 58 00
MR-3821	Sweden	Skellefteå	Skellefteå STP	Digested, 59 000 pe, no industrial impact	26.10.04	Sludge	13.9	7190298,1750209
MR-3701	Sweden	Lerum	Floda STP			Sludge	29	6414015,1294677
MR-3848	Sweden	Esöv	Ellinge STP	Digested, 100 000 pe, food industry	07.12.04	Sludge	18.9	6189961,1342090
MR-3812	Sweden	Kiruna	Tekniska verket		29.11.04	Sludge	17.8	7534822,1691614
MR-3713	Denmark		Coast area Kattegat		02.10.04	Water	-	56.51.4, 10.47.5
MR-3716	Denmark	Roskilde	Coast area	Innerfjord	19.10.04	Water	-	55.39.55, 12.03.78
MR-3717	Denmark	Kobenhavn	Coast area Øresund Lynetten		26.10.04	Water	-	12.37.86, 55.43.59
MR-3915	Denmark	Kobenhavn	Lynetten STP	Influent		Water	-	55°41.7N, 12°37E
MR-3916	Denmark	Kobenhavn	Lynetten STP	Effluent		Water	-	55°41.7N, 12°37E
MR-3920	Denmark	Roskilde	Bjergmarken STP	Influent		Water	-	55°39N, 12°03E
MR-3921	Denmark	Roskilde	Bjergmarken STP	Effluent		Water	-	55°39N, 12°03E
MR-3923	Denmark		Avedøre Landfill	Leachate		Water	-	55°36N, 12°29E
MR-3924	Denmark		Uggeløse Landfill	Leachate		Water	-	55°50N, 12°16E
MR-3902	Faroe Islands	Torshavn	Sersjantvikin STP	Effluent	25.01.05	Water	-	62 00,495 N, 06 45,712 E
MR-3903	Faroe Islands	Torshavn	Húsarhaga landfill	Leachate, sample from well on site	26.01.05	Water	-	62 01,692 N, 06 48,308 E
MR-3939	Finland	Espoo	Landfill + waste tip + waste treatment centre Ämmässuo,	Leachate water	28.01.05	Water	-	60° 12,53' N, 24° 32,90' E
MR-3940	Finland	Nokia	"Nokia 1" - Tyre manufacturing wastewater	Influent to the Nokia City sewage treatment plant	01.09.04	Water	-	59° 10,80' N, 25° 57,45' E
MR-3941	Finland	Nokia	"Nokia 2" - Floor material manufacturing wastewater	Influent to the Nokia City sewage treatment plant	01.09.04	Water	-	59° 10,80' N, 25° 57,45' E
MR-3942	Finland	Nokia	"Nokia 1"- Kulloonvuori STP	Treated effluent	01.09.04	Water	-	59° 10,80' N, 25° 57,45' E

Sample ID	Country	City	Site	Notes	Sampling date	Matrix	DW (%)	Co-ordinates
MR-3943	Finland	Nokia	"Nokia 2" - Kullonvuori STP	Effluent	02.02.05	Water	-	59° 10,80' N, 25° 57,45' E
MR-4075	Finland	Espoo	Suomenoja STP	Effluent	24.05.05	Water	-	60° 11,58' N, 24° 74,84' E
MR-4076	Finland	Helsinki	Viihkimäki STP	Effluent	24.05.05	Water	-	60° 27,95' N, 24° 87,35' E
MR-3863	Iceland	Reykjavik	Alfnas Landfill	Runoff water	13.12.04	Water	-	64 09 00, 21 58 00
MR-3905	Iceland	Reykjavik	1w, 1l	Sea water	03.02.05	Water	-	64 09 00, 21 58 00
MR-3906	Iceland	Reykjavik	2w, 0.25l	Sea water	03.02.05	Water	-	64 09 00, 21 58 00
MR-3907	Iceland	Reykjavik	3w, 1l	Sea water	03.02.05	Water	-	64 09 00, 21 58 00
MR-3908	Iceland	Reykjavik	4w, 0.25l	Sea water	03.02.05	Water	-	64 09 00, 21 58 00
MR-3617	Norway	Arendal	Arendal STP	Influent	03.02.05	Water	-	N:58,424 E: 8,743
MR-3618	Norway	Arendal	Arendal STP	Effluent		Water	-	N:58,424 E: 8,743
MR-3738	Norway		Lake Bergsjøen	Background area, surface water		Water	-	N: 60,534 E: 11,350
MR-3743	Norway		Lake Røgden	Background area, surface water		Water	-	N: 60,401 E: 12,555
MR-3858	Norway	Færder	Outer Oslofjord	Surface water, coastal background	29.11.04	Water	-	59 01.61N, 10 31.71E
MR-3860	Norway	Stellene	Inner Oslofjord	Urban surface water	13.12.04	Water	-	59 48.9N, 10 34.057E
MR-3884	Norway	Spillhaug	Landfill	Runoff water	11.01.05	Water	-	6646149, 645418
MR-3886	Norway	Bølstad	Landfill	Runoff water	11.01.05	Water	-	6618417, 599763
MR-3888	Norway	Grønmo	Landfill	Runoff water	11.01.05	Water	-	6634850, 603995
MR-3768	Sweden	Gislaved	Nissan	Upstreams storm water effluent	16.11.04	Water	-	6356021,1364021
MR-3770	Sweden	Gislaved	Nissan	storm water effluent	16.11.04	Water	-	6353937,1364076
MR-3766	Sweden	Gislaved	Nissan	Downstreams storm water effluent	16.11.04	Water	-	6352791,1363001
MR-3928	Sweden	Upplandsbro	Högbytorp	Untreated percolate water	16.02.05	Water	-	6603702,1603008
MR-3926	Sweden	Upplandsbro	Högbytorp	Treated percolate water	16.02.05	Water	-	6603702,1603007

Table A 3. Sample characteristics for biota samples.

Sample ID	Country	Location	Notes	Sampling date	Matrix	Coordinates
05/399	Denmark	Roskildeifjord	3 Eelput, liver	11.11.04	Marine fish	55°40.4 N, 12° 03.5E
05/400	Denmark	Øresund at Nivå bugt	3 Flounders, liver		Marine fish	55°48.17 N, 12°32,1
05/401	Denmark	North Sea, Hvide sande	3 Flounders, liver	2002	Marine fish	56°14 N, 7°58 E
05/402	Denmark	Wadden Sea	3 Flounders, liver	02.10.04	Marine fish	55°35,16 N, 8°18.42E,
05/389	Faroe Islands	Mýlingsgrunnurin	9 Cod liver, liver	Oct. 04	Marine fish	62°20'N, 7°40'W
05/390	Faroe Islands	Kaldbaksfjørður	10 Sculpin, liver	2004	Marine fish	62°03.87'N, 6°54.30'W
05/391	Faroe Islands	Kaldbaksfjørður	19 flatfish (gab), liver	2004	Marine fish	62°03.87'N, 6°54.30'W
05/411	Norway	15B Lista/Farsund	6 Cod, liver	19.10.04	Marine fish	N: 58,050, E: 6,717

Sample ID	Country	Location	Notes	Sampling date	Matrix	Coordinates
05/560, DL-04-19:41	Faroe Islands	Vidareidi	1 Fulmar egg, FG 0085	May 03	Seabird eggs	62°21'N, 6°31'W
05/637, C2005/7001	Sweden	Söderskäret	1 egg, Herring Gull (Larus Argentatus)	28.04.05	Seabird eggs	6607046, 1711685
05/638, C2005/7002	Sweden	Svartlögaiforden, Tråsskär	1 egg, Herring Gull	26.04.05	Seabird eggs	6609275, 1679066
05/639, C2005/7003	Sweden	Svartlögaiforden, Tråsskär	1 egg, Herring Gull	26.04.05	Seabird eggs	6609275, 1679066
05/640, C2005/7004	Sweden	Svartlögaiforden, Tråsskär	1 egg, Herring Gull	26.04.05	Seabird eggs	6609275, 1679066
05/641, C2005/7005	Sweden	Svartlögaiforden, Tråsskär	1 egg, Herring Gull	26.04.05	Seabird eggs	6609275, 1679066
05/642, C2005/7006	Sweden	Svartlögaiforden, Tråsskär	1 egg, Herring Gull	26.04.05	Seabird eggs	6609275, 1679066

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Appendix 2: Results tables

Table A 4. Results from siloxane measurements in air

Sample ID	Country	City	Location	Sampling date	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
345-nilu237	Denmark		Jagtvejen	04.01.05	µg/m ³	0.32	0.19	0.07	<0.004	<0.008	<0.006	<0.02
458-nilu238	Denmark		Bjergmarken STP	31.12.04	µg/m ³	0.66	1.3	0.17	<0.004	<0.008	<0.006	<0.02
337-nilu239	Denmark		Sepstrup Sande	12.01.05	µg/m ³	2.4	0.95	0.44	<0.004	<0.008	<0.006	<0.02
132-nilu240	Denmark		H.C. Ørsted Institute	21.12.04	µg/m ³	0.26	0.31	0.14	<0.004	<0.008	<0.006	<0.02
150-nilu241	Faroe Islands	Torshavn	Downtown	15.12.04	µg/m ³	2.1	0.93	0.39	<0.004	<0.008	<0.006	<0.02
225-nilu242	Faroe Islands	Torshavn	Sensjantivikin STP	15.12.04	µg/m ³	4.0	2.4	2.1	<0.004	<0.008	<0.006	<0.02
344-nilu106	Finland		Nokia City STP	28.01.05	µg/m ³	1.1	1.3	0.20	<0.004	<0.008	<0.006	<0.02
142-nilu105	Finland		Espoo landfill	24.05.05	µg/m ³	0.29	0.23	0.09	<0.004	<0.008	<0.006	<0.02
69-nilu232	Iceland	Reykjavik	Urban		µg/m ³	2.1	1.5	0.42	<0.004	<0.008	<0.006	<0.02
209-nilu233	Iceland	Reykjavik	Urban		µg/m ³	0.32	1.6	0.34	<0.004	<0.008	<0.006	<0.02
340-nilu234	Iceland	Reykjavik	Urban		µg/m ³	0.76	0.73	0.29	<0.004	<0.008	<0.006	<0.02
451-nilu451	Iceland	Reykjavik	Urban		µg/m ³	0.4	0.13	0.08	<0.004	<0.008	<0.006	<0.02
150-nilu108	Norway		Bekkelaget STP	13.05.05	µg/m ³	1.0	1.9	0.57	<0.004	<0.008	<0.006	<0.02
416-nilu109	Norway		Bekkelaget STP	13.05.05	µg/m ³	0.85	12	1.0	<0.004	<0.008	<0.006	<0.02
149-nilu111	Norway	Manglerud	Manglerud	13.05.05	µg/m ³	0.55	2.5	0.82	<0.004	<0.008	<0.006	<0.02
50-nilu110	Norway	Oslo	Oslo Central Station	27.05.05	µg/m ³	0.58	0.89	0.87	<0.004	<0.008	<0.006	<0.02
419-nilu206	Sweden	Högbytorp	Högbytorp landfill windside	16.02.05	µg/m ³	0.09	0.06	0.06	<0.004	<0.008	<0.006	<0.02
204-nilu208	Sweden	Högbytorp	Högbytorp landfill windside	16.02.05	µg/m ³	0.08	0.05	0.02	<0.004	<0.008	<0.006	<0.02
6-nilu229	Sweden	Gislaved	Mossarps recycling site	16.11.04	µg/m ³	0.79	0.29	0.17	<0.004	<0.008	<0.006	<0.02
453-nilu230	Sweden	Gislaved	Mossarps recycling site	16.11.04	µg/m ³	1.1	0.52	0.24	<0.004	<0.008	<0.006	<0.02
415-nilu210	Sweden	Göteborg	Kapellplatsen	03.02.05	µg/m ³	0.14	0.10	0.08	<0.004	<0.008	<0.006	<0.02
228-nilu216	Sweden	Göteborg	Kapellplatsen	04.02.05	µg/m ³	0.35	0.05	0.05	<0.004	<0.008	<0.006	<0.02
502-nilu214	Sweden	Stockholm	Hudiksvallsgatan	02.12.04	µg/m ³	0.24	0.18	0.10	<0.004	<0.008	<0.006	<0.02
509-nilu218	Sweden	Stockholm	Hudiksvallsgatan	02.12.04	µg/m ³	0.18	0.20	0.11	<0.004	<0.008	<0.006	<0.02

Table A 5. Results from measurements in soil, sediment, sludge and water

Sample ID	Country	City	Location	Sampling date	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3895	Faroe Islands		Havnardalur	20.12.04	Soil	ng/g DW	<10	<5	<4	<0.1	<0.1	<0.1	<0.1
MR-3899	Faroe Islands		Husarhaga landfill	20.12.04	Soil	ng/g DW	<6	<3	<2	<0.1	<0.1	<0.1	<0.1
MR-3714	Denmark		Coast area Kattegat	02.10.04	Sediment	ng/g DW	<3	<2	<1	<0.03	<0.03	<0.03	<0.03
MR-3718	Denmark		Øresund, Lynetten	26.10.04	Sediment	ng/g DW	<5	<3	<2	<0.05	<0.05	<0.05	0.08
MR-3723	Denmark	Roskilde	Coast area	19.10.04	Sediment	ng/g DW	84	2000	170	<0.3	<0.71	29	57
MR-3901	Faroe Islands		Kaldbakfjordur	30.08.04	Sediment	ng/g DW	<11	<5	<4	<0.1	<0.1	<0.1	<0.1
MR3945	Finland	Helsinki	Vakal (old city bay)	21.10.04	Sediment	ng/g DW	<20	58.0	8.6	<0.2	<0.2	<0.15	<0.2
MR3946	Finland	Espoo	Espoo coastal sea	13.10.04	Sediment	ng/g DW	<20	19.0	<8	<0.2	<0.2	<0.2	<0.2
MR-3909	Iceland		Sediment 1s	03.02.05	Sediment	ng/g DW	<4	7.6	<2	<0.04	<0.04	<0.04	<0.04
MR-3910	Iceland		Sediment 2s	03.02.05	Sediment	ng/g DW	<3	7.4	<1	<0.03	<0.03	<0.03	0.060
MR-3912	Iceland		Sediment 3s	03.02.05	Sediment	ng/g DW	<10	39	4.0	<0.1	<0.1	<0.1	<0.1
MR-3913	Iceland		Sediment 4s	03.02.05	Sediment	ng/g DW	<7	19	<3	<0.07	<0.07	<0.07	0.070
MR-3740	Norway		Bergsjøen		Sediment	ng/g DW	<60	<30	<23	<0.6	<0.6	<0.6	<0.6
MR-3741	Norway		Bergsjøen		Sediment	ng/g DW	<65	<30	<25	<0.7	<0.7	<0.7	<0.7
MR-3746	Norway		Røgden		Sediment	ng/g DW	<50	<30	<19	<0.5	<0.5	<0.5	<0.5
MR-3747	Norway		Røgden	07.10.04	Sediment	ng/g DW	<50	<30	<17	<0.5	<0.5	<0.5	<0.5
MR-3869	Norway		Leangbukta	26.09.04	Sediment	ng/g DW	<20	12	<8	<0.2	<0.2	<0.2	0.83
MR-3870	Norway		Vrengansundet	16.09.04	Sediment	ng/g DW	<10	<5	<4	<0.1	<0.1	<0.1	<0.1
MR-3871	Norway	Møre och Romsd.	Brødrene Sunde Verft	16.09.04	Sediment	ng/g DW	<11	96	<13	<0.1	<0.1	0.46	0.48
MR-3774	Sweden	Gislaved	Nissan	16.11.04	Sediment	ng/g DW	<2	1.8	0.89	<0.06	<0.02	<0.02	<0.02
MR-3776	Sweden	Gislaved	Nissan	16.11.04	Sediment	ng/g DW	<2	<1	<0.6	<0.02	<0.02	<0.02	<0.02
MR-3772	Sweden	Gislaved	Nissan	16.11.04	Sediment	ng/g DW	<3	<1	<1	<0.03	<0.03	0.20	0.53
MR-3314	Sweden	Stockholm	Essingen	20.12.03	Sediment	ng/g DW	<13	130	25	<0.13	<0.13	1.2	0.54
MR-3316	Sweden	Stockholm	Riddarfjärden	20.12.03	Sediment	ng/g DW	<9	77	10	<0.09	<0.09	0.45	1.7
MR-3750	Sweden		Ö Gotlandsdjupet 0-2 cm	2004	Sediment	ng/g DW	<30	<10	<9	<0.2	<0.3	<0.2	<0.2
MR-3753	Sweden		Ö Landsortsdjupet 0-2cm	2004	Sediment	ng/g DW	<30	<20	<12	<0.3	<0.3	<0.3	<0.3

Sample ID	Country	City	Location	Sampling date	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3917	Denmark	Kobenhavn	Lynetten		Sludge	ng/g DW	740	27000	1100	<3	64	450	550
MR-3918	Denmark	Kobenhavn	Lynetten		Sludge	ng/g DW	470	50000	2800	<1	8	44	140
MR-3897	Faroe Islands	Torshavn	Sersjantivikin	16.12.04	Sludge	ng/g DW	190	4300	1000	<1	<1	23	10
MR3932	Finland	Nokia	Nokia Kullonvuori	02.02.05	Sludge	ng/g DW	960	30000	3500	<2	3	12	14
MR3935	Finland	Helsinki	Helsinki Viikinkaari	27.01.05	Sludge	ng/g DW	230	21000	2200	<1	1	8	34
MR3936	Finland	Espoo	Espoo Suomenoja	28.01.05	Sludge	ng/g DW	530	89000	11000	<0.5	4	17	40
MR3937	Finland	Pori	Pori STP	26.01.05	Sludge	ng/g DW	740	31000	3600	<0.5	4	8	14
MR3938	Finland	Pori	Kokkonlehti STP	26.01.05	Sludge	ng/g DW	660	25000	1900	<0.5	10	8	17
MR-3862	Iceland	Reykjavik	Klettgardar STP	13.12.04	Sludge	ng/g DW	120	1600	240	<1	<1	2	4
MR-3865	Iceland	Reykjavik	Ananaust STP	13.12.04	Sludge	ng/g DW	96	1100	220	<1	<1	1	3
MR-3621	Sweden	Skellefteå	Skellefteå STP	26.10.04	Sludge	ng/g DW	370	21000	2400	<1	3	8	19
MR-3701	Sweden	Lerum	Floda STP		Sludge	ng/g DW	120	5800	870	<1	<1	3	5
MR-3848	Sweden	Eslöv	Ellinge STP	07.12.04	Sludge	ng/g DW	200	4500	700	<1	<1	2	4
MR-3812	Sweden	Kiruna	Tekniska verket	29.11.04	Sludge	ng/g DW	380	11000	1800	<1	<1	23	19

Sample ID	Country	City	Location	Sampling date	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3713	Denmark		Coast area Kattegat	02.10.04	Water	µg/L	<0.04	<0.02	<0.02	<0.0005	<0.0005	<0.0005	<0.002
MR-3716	Denmark	Roskilde	Coast area innerfjord	19.10.04	Water	µg/L	<0.04	<0.02	<0.02	<0.0005	<0.0005	<0.0005	<0.002
MR-3717	Denmark	Kobenhavn	Øresund Lynetten	26.10.04	Water	µg/L	<0.04	<0.02	<0.02	<0.0005	<0.0005	<0.0005	<0.002
MR-3915	Denmark	Kobenhavn	Lynetten STP, influent		Water	µg/L	0.28	26	1.6	<0.01	0.0034	0.041	0.073
MR-3916	Denmark	Kobenhavn	Lynetten STP, effluent		Water	µg/L	<0.06	0.063	<0.02	<0.001	<0.001	<0.001	<0.004
MR-3920	Denmark	Roskilde	Bjergmarken STP, influent		Water	µg/L	0.60	24	1.3	<0.01	<0.01	0.078	0.23
MR-3921	Denmark	Roskilde	Bjergmarken STP, effluent		Water	µg/L	<0.06	0.092	<0.04	<0.0005	<0.0005	0.0045	0.0087
MR-3923	Denmark		Avedøre Landfill		Water	µg/L	<0.08	<0.04	<0.04	0.045	<0.001	<0.0005	<0.005
MR-3924	Denmark		Uggeløse Landfill		Water	µg/L	<0.08	<0.04	<0.04	<0.0005	<0.001	0.00087	<0.004
MR-3902	Faroe Islands	Torshavn	Serejartvíkin STP	26.01.05	Water	µg/L	<0.08	5.2	0.33	<0.0005	<0.001	<0.0005	<0.004
MR-3903	Faroe Islands	Torshavn	Húsarhaga landfill	26.01.05	Water	µg/L	<0.08	<0.05	<0.05	<0.0005	<0.001	<0.0005	<0.005
MR-3939	Finland	Espoo	Landfill & waste tip Ámmässuo	28.01.05	Water	µg/L	<0.4	3.9	1.7	0.11	<0.004	<0.004	0.019
MR-3940	Finland	Nokia	"Nokia 1" - Tyre industry wastewater, influent to STP	01.09.04	Water	µg/L	3.7	5.3	3.7	<0.007	<0.007	<0.007	0.054
MR-3941	Finland	Nokia	"Nokia 2" - Floor industry wastewater, influent to STP	01.09.04	Water	µg/L	0.25	0.33	0.12	0.12	0.014	0.041	0.055
MR-3942	Finland	Nokia	"Nokia 1" - Kulkonvuori STP, Treated effluent	01.09.04	Water	µg/L	<0.06	0.48	0.03	<0.004	<0.001	<0.001	<0.004
MR-3943	Finland	Nokia	"Nokia 2" - Kulkonvuori STP, Effluent	02.02.05	Water	µg/L	0.11	0.98	0.045	<0.001	<0.001	0.0012	<0.005
MR-4075	Finland	Espoo	Espoo City Suomenoja STP, Effluent	24.05.05	Water	µg/L	<0.08	0.62	0.07	<0.0006	<0.0007	<0.001	<0.005
MR-4076	Finland	Helsinki	Helsinki City Viikinnmäki STP, Effluent	24.05.05	Water	µg/L	<0.08	0.29	<0.04	0.0005	<0.0007	<0.001	<0.004

Sample ID	Country	City	Location	Sampling date	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR-3863	Iceland	Reykjavik	Alfnes Landfill	13.12.04	Water	µg/L	1.1	5.4	1.3	0.039	<0.0005	0.027	0.015
MR-3905	Iceland	Reykjavik	Sea water, 1w, 1l	03.02.05	Water	µg/L	<0.05	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.003
MR-3906	Iceland	Reykjavik	Sea water, 2w, 0.25l	03.02.05	Water	µg/L	<0.05	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.003
MR-3907	Iceland	Reykjavik	Sea water, 3w, 1l	03.02.05	Water	µg/L	<0.05	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.003
MR-3908	Iceland	Reykjavik	Sea water, 4w, 0.25l	03.02.05	Water	µg/L	<0.05	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.003
MR-3617	Norway	Arendal	Arendal STP		Water	µg/L	<0.3	5.0	3.8	<0.004	<0.004	<0.004	<0.004
MR-3618	Norway	Arendal	Arendal STP		Water	µg/L	<0.07	0.72	0.16	<0.0005	<0.0006	<0.0005	<0.004
MR-3738	Norway		Lake Bergsjøen		Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0006	<0.0005	<0.004
MR-3743	Norway		Lake Røgdalen		Water	µg/L	<0.09	<0.05	<0.05	<0.0005	<0.0008	<0.0006	<0.005
MR-3858	Norway	Farder	yttre Oslofjord	29.11.04	Water	µg/L	<0.07	<0.04	<0.04	<0.0005	<0.0006	<0.0005	<0.004
MR-3860	Norway	Steilene	indre Oslofjord	13.12.04	Water	µg/L	<0.07	<0.04	<0.04	<0.0005	<0.0007	<0.0005	<0.004
MR-3884	Norway	Spillhaug	Landfill	11.01.05	Water	µg/L	<0.08	<0.05	<0.05	0.0009	<0.0007	<0.0006	<0.005
MR-3886	Norway	Bøistad	Landfill	11.01.05	Water	µg/L	<0.07	<0.04	<0.04	0.0008	<0.0007	<0.0005	<0.004
MR-3888	Norway	Grønmo	Landfill	11.01.05	Water	µg/L	<0.07	<0.04	<0.04	0.042	<0.0006	<0.0005	<0.004
MR-3768	Sweden	Gislaved	River Nissan	16.11.04	Water	µg/L	<0.07	<0.04	<0.05	<0.0006	<0.0006	<0.0005	<0.004
MR-3770	Sweden	Gislaved	River Nissan (Storm water)	16.11.04	Water	µg/L	<0.07	<0.04	<0.04	0.0008	<0.0006	<0.0005	<0.004
MR-3766	Sweden	Gislaved	River Nissan	16.11.04	Water	µg/L	<0.07	<0.04	0.064	<0.0006	<0.0006	<0.0005	<0.004
MR-3928	Sweden	Upplandsbro	Högbytorp landfill	16.02.05	Water	µg/L	<0.12	<0.07	<0.07	0.14	<0.0010	<0.0008	<0.006
MR-3926	Sweden	Upplandsbro	Högbytorp landfill	16.02.05	Water	µg/L	<0.09	<0.05	<0.05	0.022	<0.0007	<0.0006	<0.005

Table A 6. Results from siloxane measurements in biota (concentrations in bold italic figures are above limit of detection but below limit of quantification)

Sample ID	Country	Location	Matrix	Unit	D3	D4	D5	D6	MM	MDM	MD2M	MD3M
05/399	Denmark	Roskilde-fjord	Marine fish	ng/g ww	<50	11	<5	<5	<0.4	<0.3	<0.4	<0.5
05/400	Denmark	Øresund at Nivå bugt	Marine fish	ng/g ww	90	13	52	8.7	<0.4	<0.3	<0.4	<0.5
05/401	Denmark	North Sea, Hvide sande	Marine fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/402	Denmark	Wadden Sea	Marine fish	ng/g ww	<50	<5	33	<5	<0.4	<0.3	<0.4	<0.5
05/399	Faroe Islands	Mylingsgrunnurin	Marine fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/390	Faroe Islands	Kaldbaksfjørður	Marine fish	ng/g ww	<50	<5	<5	5.2	<0.4	<0.3	<0.4	<0.5
05/391	Faroe Islands	Kaldbaksfjørður	Marine fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/411	Norway	15B ListaFarsund	Marine fish	ng/g ww	<50	<5	26	<5	<0.4	<0.3	<0.4	<0.5
05/412	Norway	53B Indre Sørfjord	Marine fish	ng/g ww	<50	12	61	6.7	<0.4	<0.3	<0.4	<0.5
05/413	Norway	Ulleinvik	Marine fish	ng/g ww	<50	11	46	6.1	<0.4	<0.3	<0.4	<0.5
05/414	Norway	30B Indre Oslofjord	Marine fish	ng/g ww	<50	70	2 200	74	<0.4	<0.3	1.1	<0.5
05/397	Faroe Islands	Lake A Myranar	Freshwater fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/398	Faroe Islands	Lake A Myranar	Freshwater fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/479	Finland	Old City Bay, Helsinki	Freshwater fish	ng/g ww	<50	5.8	20	<5	<0.4	<0.3	<0.4	<0.5
05/480	Finland	Old City Bay, Helsinki	Freshwater fish	ng/g ww	<50	5.7	33	<5	<0.4	<0.3	<0.4	<0.5
05/481	Finland	Old City Bay, Helsinki	Freshwater fish	ng/g ww	<50	8.9	84	<5	<0.4	<0.3	<0.4	<0.5
05/482	Finland	Cold water bay, Helsinki	Freshwater fish	ng/g ww	<50	<5	22	<5	<0.4	<0.3	<0.4	<0.5
05/483	Finland	Guard Village Bay, Helsinki	Freshwater fish	ng/g ww	<50	<5	6.5	<5	<0.4	<0.3	<0.4	<0.5
05/415	Norway	Mjøsa	Freshwater fish	ng/g ww	<50	<5	21	<5	<0.4	<0.3	<0.4	<0.5
05/408 MR-3892	Sweden	Nissan, Skapshuit	Freshwater fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/409 MR-3894	Sweden	Nissan, Rydöbruk	Freshwater fish	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/404	Denmark	Coastal area, Øresund	Marine mammals	ng/g ww	<50	<5	24	<5	<0.4	<0.3	<0.4	<0.5
05/405	Denmark	Samsø	Marine mammals	ng/g ww	<50	<5	20	<5	<0.4	<0.3	<0.4	<0.5
05/406	Denmark	Limfjorden	Marine mammals	ng/g ww	<50	<5	17	<5	<0.4	<0.3	<0.4	<0.5
05/407	Denmark	Hesselø	Marine mammals	ng/g ww	68	12	22	7.9	<0.4	<0.3	<0.4	<0.5
05/392	Faroe Islands	Sandangerði	Marine mammals	ng/g ww	<50	<5	10	<5	<0.4	<0.3	<0.4	<0.5
05/393	Faroe Islands	Gøtu	Marine mammals	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/403	Iceland		Marine mammals	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5

Sample ID	Country	Location	Matrix	Unit	D3	D4	D5	D6	MM	MDM	MD2M	MD3M
05/394	Faroe Islands	Skúvoy	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/396	Faroe Islands	Koltur/Skúvoy	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/552 DL-04-019:33	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/553 DL-04-019:34	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/554 DL-04-019:35	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/555 DL-04-19:36	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/556 DL-04-19:37	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/557 DL-04-19:38	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/558 DL-04-19:39	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/559 DL-04-19:40	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/560 DL-04-19:41	Faroe Islands	Víkareidi	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/637 C2005/7001	Sweden	Söderskäret skan	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/638 C2005/7002	Sweden	Svartögafljorden	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/639 C2005/7003	Sweden	Svartögafljorden	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/640 C2005/7004	Sweden	Svartögafljorden	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/641 C2005/7005	Sweden	Svartögafljorden	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5
05/642 C2005/7006	Sweden	Svartögafljorden	Seabird eggs	ng/g ww	<50	<5	<5	<5	<0.4	<0.3	<0.4	<0.5

Appendix 3: Sampling manual

Analysis of siloxanes in a Nordic cooperation on screening
Sampling and sample handling manual

IVL/NILU

1. Air samples
2. Sediment, sludge, soil, and water samples
3. Biological samples
4. Sampling forms

Air sampling

General remarks

Sampling of Siloxanes in air is a challenging task since Siloxanes are widely used within personal care products like deodorants, skin lotions and perfumes and other household products. A lot of flexible tubing, often used together with air sampling equipment are also source of siloxanes.

It is therefore necessary to avoid contact between the sampling tube and bare skin. Together with the sampling equipment we will provide you with disposable gloves. In order to avoid contamination of the sampling tubes by the persons taking the samples – there should be as little as possible contact time. While sampling in closed indoor environment the sampling personnel should not stay close to the pump and adsorption tube. Sampling outdoors the personnel should stay down wind the sampling equipment.

- Please don't use any kind of water-resistant permanent markers to write information onto the tube or the pen-clip.
- Each tube has its own id-number which should be used together with the sampling-form for adsorption tubes.
- Please do not use any type of glue or tape in connection with the sampling.

Sampling procedure

Use disposable gloves.

The adsorption-tubes are sealed with a unit of a brass nut and brass cap at both ends with a teflon –sealing inside. On each tube is a pen-clip with an ID-number and arrows showing the correct direction of the air-flow.

You need two spanners or a set of 1/2 inch and 9/16 inch tools to loosen the unit with about half a turn and then remove the brass unit as one piece from the adsorption tube. At first remove the brass unit from the tube at the side to where the arrows on the ID-clip is pointing. This is the side, which is connected to the Inlet Port of the pump. This port is equipped with a Swagelock-type brass unit with teflon sealing inside. Push the end of the tube into it – tighten the nut with your fingers and use a 9/16 inch tool to tighten (maximum ¼ of a full turn). Right before starting sampling the other brass unit has to be removed. Fill inn the sampling form.

The sampling time will be 1 hour for indoor, industrial or urban sites and 2 hours for rural or background sites.

After sampling the adsorption tube has to be reassembled with the brass caps. Please start to tighten the unit with your fingers and use the 9/16 inch tool to tighten (maximum $\frac{1}{4}$ of a full turn).

Both empty and exposed adsorption tubes can be stored at normal indoor or outdoor temperature conditions.

Address

NILU v/Schmidbauer eller Lunder
P.b. 100
2027 Kjeller Tf 63898000

Sampling of sediment, sludge, soil, and water

General remarks

Siloxanes are frequently used in cosmetic formulations. Do not use products such as antiperspirant, eye shadow, hair spray, or skin lotions on the day of sampling. Only specially cleaned sampling containers provided by the laboratory should be used.

To check for contamination sampling blanks are used. The sampling blank to be used for sediment, sludge, and soil sampling contains silica particles, the one to be used for water contains MilliQ-water. The sampling blanks should not be emptied or filled. They shall only be opened and closed at the time of sampling.

The number of sampling blanks is limited. If the number of samples per sample type (water, sediment, sludge, soil) and country is one to four the number of sampling blanks is one. If the number of samples per sample type and country is five or more the number of sampling blanks is two. The sites used for blank sampling should be selected at random before the start of sampling.

Sampling

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

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

Fill the sample container, if required using the enclosed spoon. If the Al-foil protecting the lids is ruptured replace it with new Al-foil and close the lid on the sample bottles (and sample blank). Label the sample containers. Put each container in a plastic bag.

Fill in the sample protocol.

Storage and transport

Store the samples refrigerated (or in a freezer - be aware of the risk for damage to glass containers!) and send to the laboratory as soon as possible in such a way that the samples will reach the laboratory within one day. When sending the samples please send an e-mail including a list specifying the samples to Lennart Kaj (lennart.kaj@ivl.se).

Address

IVL Swedish Environmental Research Institute
Lennart Kaj
Hälsingegatan 43
SE-113 31 Stockholm.
Sweden.

Biological samples

General remarks

Sampling should be performed in accordance with general sampling strategies for chemical trace analysis. In case of questions about the practicability of procedures or usability of special material and equipment NILU must be contacted (Biological samples: Martin Schlabach, Tel: +47 6389 8213, Fax: +47 6389 8050, msc@nilu.no). The sampling strategy should take into account the specific objectives of the monitoring programme, including the quantitative objectives. Natural variability within the samples should be reduced by an appropriate sampling design. The sampling strategy is an intrinsic component of the data, and may limit their use and interpretation.

Nothing is known about the homogeneity of the distribution of siloxanes in the Nordic environment, since no comprehensive screening has been performed in these countries yet. Therefore, it is difficult to give recommendations about the choice of representative sampling sites. Only a relatively high number of samples taken from as many different places as possible can overcome this challenge and will add to our knowledge about distribution patterns of PFAS in the environment. The detailed sampling site selection lies within the responsibility of the sampling institutes. Sampling sites must be indicated on the sampling protocols as accurate as possible (preferably with latitude/longitude data).

Risk of contamination / Risk of evaporation loss

Siloxanes are widely used in cosmetics (especially deodorants and creams) and a row of technical products (paints and plastics), so great care has to be taken to avoid contamination of the samples. When handling the samples gloves are mandatory, and measures should be taken to exclude that the personnel involved are using cosmetics containing siloxanes during the sample-handling period. Even cleaned laboratory air can contain vapors of siloxanes; so prolonged direct contact with the laboratory air should be avoided. Furthermore all utensils coming in contact with the samples should be solvent rinsed with 3 times acetone and 3 times n-pentane following the normal cleaning. Glass and metal utensils should finally be heated for 2 hours at 450 °C. Direct contact with polymer utensils should be kept at a minimum.

In contrast to the compounds of the two preceding NMR-screening studies siloxanes can be characterized as typical volatile organic compounds (VOC) and thus evaporation loss during sample handling is a risk which we must try to hold at a minimum. We propose therefore to keep

the biological samples as intact as possible. This means that whole fishes, complete livers, whole eggs, or pieces of seal/whale meat/blubber are preferred instead of homogenized small samples.

All samples of one kind from each country should be sent together to NILU as soon as possible after the sampling. NILU should be notified about the arrival of samples in order to ensure that the samples are received and handled properly.

NILU must be contacted in case of questions about the usability of certain materials in contact with samples.

Field sampling and required sample size

All biological samples must be frozen immediately after catch and preparation.

Fish and fish liver

Fish should be dissected immediately after collection. Small fishes shall be packed and sent as whole fish. From bigger fishes at least a 10 cm chop from the middle part of each fish shall be sent. Liver samples shall be packed and sent as whole pieces.

Bird eggs

Only newly laid eggs are usable for this study, since the siloxanes will truly be volatilized and lost during breeding several weeks at 37 °C. Fresh eggs shall be packed separately, frozen and sent.

Seal and whale

The seal or whale should be dissected immediately after collection. From each individual a 5 * 5 cm slice of meat or blubber shall be taken, packed and sent.

Storage and shipping of samples

All samples must be collected, stored and shipped in clean, siloxane-free packaging. We suggest to wrap the samples tightly with aluminium foil, and pack these packages in a polyethylene zip-bag

Biological samples must be kept frozen during storage (< -18 °C). During transportation, an insulated box should be used to ensure that the temperature does not exceed thawing temperature (< 0 °C). All samples must be clearly and unmistakably marked with a sample name and sent

together with their sampling protocols by an express delivery service (TNT, DHL, EMS or similar) to the following address:

Norwegian Institute for Air Research (NILU)
Att Martin Schlabach
Instituttveien 18
NO-2027 Kjeller
Norway

To assure that samples reach the destination within short time (usually within the same day), they should be sent early in the morning and not on a Friday (preferably Monday to Wednesday). When sending the samples a notice including the airway bill number (AWB) of the package must be sent to NILU to the following fax number: ++47 6389 8050 addressed to Martin Schlabach or e-mail to msc@nilu.no. The delivery should be marked with "samples NMR-siloxane study" to avoid unnecessary delays during the registration procedure at the analysing institute.

Sampling forms

Sampling form – adsorption tubes

Sampling with pump

Country			
Sampling site			
Coordinates			
Sample taken by : (name)			
Pump ID			

Tube ID	Start/Stop Date, time (precise)	Place (e.g on a table or at the floor - approximately height above ground)	Remarks

Questions about sampling, call NILU at + 47 63 89 82 15

Sent to NILU:	
Received at NILU:	

Address:
 NILU Attn: Schmidbauer / Lunder
 P.b 100
 2027 Kjeller

When sending the samples a list of the samples together with the airway bill number (AWB) of the package must be sent to NILU to the following fax number: ++47 6389 8050 or e-mail to ns@nilu.no.

Sampling form – Sediment, sludge, soil and water samples

Sample type:

- | | |
|--|--|
| <input type="checkbox"/> Sewage water | <input type="checkbox"/> Digested sludge |
| <input type="checkbox"/> Surface water | <input type="checkbox"/> Sediment |
| <input type="checkbox"/> Soil | |
| <input type="checkbox"/> Sampling blank included | |

Sample name / identity:

Coordinates for the sample site:

Sampling day:

Shipped to IVL:

Received at IVL:

Used sampling equipment:

Responsible person:

Sample storage:

-
- Freezer
-
- Fridge
-
- Other

Sample preservation:

-
- NaN
- ₃
-
- phosphoric acid
-
- Other

Address:

IVL Swedish Environmental Research Institute
Lennart Kaj
Hälsingegatan 43
SE-113 31 Stockholm

When sending the samples please send an e-mail including a list specifying the samples to Lennart Kaj (lennart.kaj@ivl.se).

Sampling form -Biological samples

Sample name:	Sample material:
---------------------	-------------------------

Sampling		Comments
Date:		
Site (description, preferably with latitude/longitude data):		
Number and size of individuals used for the pooled sample:		
Age and sex of the individuals		
Storage temp. after sampling:		
Date and site of dissection:		
Total sample amount:		
Special observations:		
Storage		Comments
Storage time:		
Storage temperature:		
Special observations:		
Shipping		Comments
Date sample sent:		
Shipping temperature:		
Date sample received*:		
Condition upon receipt*:		
Special observations*:		

* To be filled in by NILU

Send to:
 Norwegian Institute for Air Research (NILU)
 Att Martin Schlabach
 Instituttveien 18
 NO-2027 Kjeller
 Norway

When sending the samples a list of the samples together with the airway bill number (AWB) of the package must be sent to NILU to the following fax number: ++47 6389 8050 or e-mail to msc@nilu.no.

Results from the Swedish
National Screening Programme
2004

Subreport 4: Siloxanes

Lennart Kaj, Jeanette Andersson, Anna Palm Cousins,
Mikael Remberger, Eva Brorström-Lundén, IVL
Ingemar Cato, SGU
B1643
October 2005

IVL Swedish Environmental
Research Institute

IVL Svenska
Miljöinstitutet
Swedish Environmental Research Institute

<p>Organization IVL Swedish Environmental Research Institute Ltd.</p>	<p>Report Summary</p>
<p>Address P.O. Box 21060 SE-100 31 Stockholm</p>	<p>Project title</p>
<p>Telephone +46 (0)8-598 563 00</p>	<p>Project sponsor</p>
<p>Author Lennart Kaj, Jeanette Andersson, Anna Palm Cousins, Mikael Remberger, Ylva Ekheden. Brita Dusan, Eva Brorström-Lundén, IVL, Ingemar Cato, SGU</p>	
<p>Title and subtitle of the report Results from the Swedish National Screening Programme 2004. Subreport 4: Siloxanes</p>	
<p>Summary As an assignment from the Swedish Environmental Protection Agency, IVL has performed a screening study of siloxanes. The substances included were three cyclic polydimethylsiloxanes (D4, D5, and D6) and four linear analogues (MM, MDM, MD2M and MD3M). The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden. A total of 138 samples of air, water, sludge, sediment and fish were analysed. D4 that is classified as a phase out substance was found in 37 out of 54 municipal sludge samples in concentrations from 130 to 2 300 ng/g DW and in various air samples in concentrations up to 300 ng/m³. D4 was not found in any of the water, sediment or fish samples. MM, which is on the OSPAR candidate list for dangerous substances, was found in leachate water from landfills and in low concentrations in air in the Stenungsund chemical industrial area. D5 was the dominating siloxane in most samples. It was found in all sludge samples from municipal treatment plants. The average concentration was 11 000 ng/g DW. Siloxanes were not found in aquatic biota (fish muscle). One or more of D4, D5 and D6 were found in 11 out of 49 samples of human breast milk. The maximum concentration of D4 was 10 µg/L, of D5 4.5 µg/L and of D6 4.8 µg/L.</p>	
<p>Keyword Screening siloxanes D4 D5 D6 MM MDM MD2M MD3M</p>	
<p>Bibliographic data IVL Report B1643</p>	
<p>The report can be ordered via Homepage: www.ivl.se, e-mail: publicationservice@ivl.se, fax+46 (0)8-598 563 90, or via IVL, P.O. Box 21060, SE-100 31 Stockholm Sweden</p>	

Sammanfattning

IVL Svenska Miljöinstitutet har på uppdrag av Naturvårdsverket utfört en screeningstudie av siloxaner. Studien omfattade tre cykliska polydimetylsiloxaner (D4, D5, D6) och fyra linjära (MM, MDM, MD2M, MD3M). Det huvudsakliga syftet med studien var att bestämma koncentrationen i ett antal matriser i den svenska miljön, att undersöka viktiga transportvägar samt att utröna om pågående emissioner kan tänkas förekomma i Sverige. Vidare var syftet att undersöka betydelsen av atmosfärisk transport samt upptag i biota.

En nationell provtagningsstrategi utarbetades. Provtagningsprogrammet baserades på identifierade möjliga utsläppskällor samt på ämnens egenskaper. Programmet inkluderade mätningar både i bakgrundsområden och nära potentiella punktkällor. Även mätningar av diffusa spridningsvägar i form av avloppssystem inkluderades. Mätningarna omfattade 39 prov fördelade på luft, vatten, slam, sediment och fisk. För att belysa humant upptag analyserades också 39 prov av bröstmjölk.

Utöver det nationella programmet bidrog tretton länsstyrelser med ytterligare 99 prov av samma provtyper från regionala program.

D4 som klassas som ett utfasningsämne återfanns i 37 av 54 slamprov från kommunala avloppsreningsverk i koncentrationer mellan 130 och 2 300 ng/g TS. D4 kunde inte detekteras i prov på vatten, sediment eller fiskmuskel.

MM som finns upptagen på OSPARs kandidatlista över farliga substanser förekom i lakvatten från deponier och i låga halter i luft från Stenungsundsområdet.

D5 var den dominerande siloxanen i de flesta proven. D5 fanns i alla slamprov från kommunala reningsverk. Medelkoncentrationen var 11 000 ng/g DW.

Siloxaner detekterades inte i något av de biologiska proven från den akvatiska miljön (fiskmuskel).

I elva av 49 analyserade prov av human bröstmjölk påvisades en eller flera av D4, D5 och D6. Den maximala koncentrationen av D4 var 10 µg/l, av D5 4.5 µg/l och av D6 4.8 µg/l. Kvantifieringsgränsen var 2 µg/l.

Summary

As an assignment from the Swedish Environmental Protection Agency, IVL has performed a screening study of siloxanes. The substances included were three cyclic polydimethylsiloxanes (D4, D5, and D6) and four linear analogues (MM, MDM, MD2M and MD3M). The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden.

A national sampling strategy was developed. The sampling programme was based on identified possible emission sources and use of the chemicals as well as on the behaviour of the substances in the environment. The programme included measurements in background areas and close to potential point sources. Measurements of diffuse pathways from the society including sewage systems were also included. This programme totalled 39 samples of air, water, sludge, sediment and fish. To elucidate human exposure of siloxanes 39 samples of human breast milk were included in the national program. Thirteen county administrative boards throughout Sweden contributed 99 additional samples of the same sample types.

D4 that is classified as a phase out substance was found in 37 out of 54 municipal sludge samples in concentrations from 130 to 2 300 ng/g DW and in various air samples in concentrations up to 300 ng/m³. D4 was not found in any of the water, sediment or fish samples.

MM, which is on the OSPAR candidate list for dangerous substances, was found in leachate water from landfills and in low concentrations in air collected in the Stenungsund chemical industrial area.

D5 was the dominating siloxane in most of the samples. It was found in all sludge samples from municipal treatment plants. The average concentration was 11 000 ng/g DW.

Siloxanes were not found in biota from aquatic environment (fish muscle).

One or more of D4, D5 and D6 were found in 11 out of 49 samples of human breast milk. The maximum concentration of D4 was 10 µg/l, of D5 4.5 µg/l and of D6 4.8 µg/l. The quantification limit was 2 µg/l.

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Appendix Information on samples and lists of results

1 Introduction

As an assignment from the Swedish Environmental Protection Agency, IVL has performed a "Screening Study". This screening, which was carried out during 2004/2005, includes the following substances: adipates, octachlorostyrene, limonene, siloxanes, mirex, endosulfan and isocyanates. The different substances or groups of substances are emitted to and spread in the environment via a variety of sources, e.g. point sources and use in products. Some of the chemicals are commonly used internationally and/or in Sweden.

The seven chemicals or chemical groups studied have been identified as potentially toxic, bioaccumulative and/or persistent. Some are also included on different international/national priority lists. Table 1 shows an overview of the chemicals included and the major reasons for their concern.

Table 1. Overview of chemicals included in the screening 2004 and the reason for their concern (Loh et al., 2003; Andersson, 2004; OSPAR, 2005; UNEP, 2005) The chemicals considered in the current report are written in bold/italic letters.

	Chemical type	Banned/ Restricted	HPV ^a	Indications of toxicity	Evidence for B/P ^b	International Priority List
Adipates	Additive	No	x	x		
Octachlorostyrene	Unintentional by-product	PRIO- substance ^c		x	x	Candidate for the Stockholm convention
Limonene	Cleaning agent (also naturally occurring)			x		
Siloxanes	Lubricant, industrial raw material, chemical additive	PRIO- substance^c	x	x	x	OSPAR (HMDS)
Mirex	Pesticide	Banned		x	x	Stockholm convention
Endosulfan	Pesticide	Banned		x	x	WFD, OSPAR, Candidate for the Stockholm convention
Isocyanates	Industrial raw material	Regulated	x	x		OSPAR ^d

^{a)} High Production Volume

^{b)} Bioaccumulation/Persistence

^{c)} The chemical is included on Swedish Chemicals Inspectorate's PRIO-list, and is identified as a "phase-out-chemical"

^{d)} Concerns 3,3'-(ureylenedimethylene)-bis-(3,5,5-trimethylcyclohexyl) diisocyanate

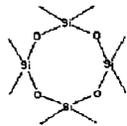
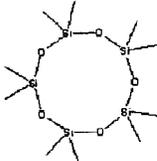
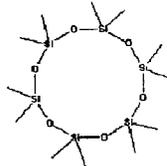
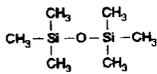
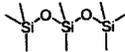
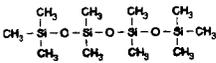
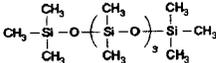
The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden. A further aim was to investigate the likelihood of atmospheric transport and uptake in biota. The outcome of the study is aimed to serve as a basis for decision-making regarding monitoring activities of these chemicals.

Due to the variety in emission sources and use as well as differences in chemical properties, the screening has been carried out in seven sub-projects. This report concerns the results of **siloxanes**. Results for the other chemicals are presented in subreports 1-3 and 5-6.

2 Chemical properties, fate, toxicity and regulation

Siloxanes form a large group of chemicals with molecular weights from a few hundreds to several hundred thousands. This study is limited to three cyclic and four linear polydimethylsiloxanes of low molecular weight. These compounds are listed in Table 2. They are referred to by their full names or the abbreviations given in this table. The abbreviated names are according to the General Electric's siloxane notation (Hurd, 1946).

Table 2. Siloxanes included in the screening programme.

Abbreviation	Name	CAS #	Structure
D4	Octamethylcyclotetrasiloxane	556-67-2	
D5	Decamethylcyclopentasiloxane	541-02-6	
D6	Dodecamethylcyclohexasiloxane	540-97-6	
MM (or HMDS)	Hexamethyldisiloxane	107-46-0	
MDM	Octamethyltrisiloxane	107-51-7	
MD2M	Decamethyltetrasiloxane	141-62-8	
MD3M	Dodecamethylpentasiloxane	141-63-9	

The siloxanes selected in this study occur as clear viscous liquids at room temperature and have varying physical-chemical properties according to Table 3.

Table 3. Chemical and physical data.

Substance	MW	W_{soil} (mg/l)	V_o (mm Hg)	H (Atm m ² /mol)	Log K_{ow}	BCF	K_{oc} (L/kg)
D4 ^a	296.6	0.9 (25°)	1 (25°)	0.42	-	12400	2.85×10^4
D5 ^b	370.8	0.24 ^b ; 0.017 ^c (25°)	0.2 (25°) ^{b,c}	0.4 ^b ; 0.3 ^c	5.7 ^b ; 5.2 ^c	5000 ^b	1.6×10^{6b}
D6	444.9						
MM ^a	162.4	2 (25°)	42 (25°)	4.5	4.2	5000	4.6×10^3
MDM	236.5						
MD2M	310.7						
MD3M ^c	384.8	3.1×10^{-4}	0.10	0.79	6		

^a HSDB, 2004; ^b ECB, 2005; ^c SRC, 2005

In the atmosphere siloxanes may exist both in the vapour and particle phases. However the most volatile siloxanes are mainly in the vapour phase. In particle phase, the siloxanes are mainly removed from the air through deposition whereas in the vapour phase they may also react with hydroxyl radicals. Half-lives for reaction with hydroxyl radicals in air are given in Table 4.

Table 4. Half-lives for reaction with hydroxyl radicals in air.

Substance	Half life	Reference
D4	16 days	HSDB, 2004
D5	10 days	HSDB, 2004
MM	12 days	HSDB, 2004

MM, D4 and D5, have high vapour pressures and high Henry's law constants and may vaporise both from soil and from water. Siloxanes have high K_{oc} (Table 3) and are considered not to be transported with water in soil. In water, they may sorb to particles and are likely to be enriched in sediment (HSDB, 2004). Siloxanes are resistant to chemical reactions such as oxidation, reduction, and direct photodegradation (HSDB, 2004). However, varying information exists on the possibility of siloxanes to undergo hydrolysis e.g in a study of hydrolysis kinetics of D4 the degradation rate was considered environmentally significant. Intermediate and final hydrolysis products were not established in the study (Durham, 2004).

A modelling exercise was performed using the Equilibrium Criterion (EQC) model (Mackay et al., 1996) in order to highlight the likely fate and partitioning behaviour of siloxanes. D5 and MD3M were selected as model substances for fate assessment. Physical-chemical properties were taken from Table 2. The degradation half-lives used were as follows; air: 170 h, water 550 h, soil 1700 h and sediment 5500 hours. The data was based on degradation data obtained from HSDB (2004), as well as estimated data using the EPIWIN software (Meylan, 1999) and classified according to Mackay (2001). Emission rates were set to 1000 kg/h, only for illustrative purposes. The outcome of the modelling exercise is shown in Table 5. The numbers given in the table should be regarded as indicative, as they are dependent on model structure as well as chemical property data.

The overall residence time in the system of both substances is predicted to be fairly low, and generally lower for D5 (<14 days when emitted to all media) than for MD3M (≈30 days). It should be emphasised, however, that advective processes contribute significantly to this low residence

time, and it does not necessarily imply that the chemical is ultimately removed from the environment. On the contrary, the atmospheric half-lives of 1-2 weeks (Table 4) and their general resistance to chemical reactions as mentioned above, imply that siloxanes are persistent enough to undergo long-range atmospheric transport.

The model results emphasize the high volatility of these siloxanes, showing significant partitioning to air regardless of emission media. When emitted to water, however, a large amount is also expected to deposit to sediments, as a result of a relatively high LogK_{ow}-value. The primary receiving media are likely to be air and water, based on the high volatility and the areas of use of siloxanes.

Table 5. Results from EQC modelling of D5 and MD3M, using emission rates of 1000 kg/h

Emission medium	% In air		% In water		% In soil		% In sediment		Persistence (h)	
	D5	MD3M	D5	MD3M	D5	MD3M	D5	MD3M	D5	MD3M
Air	99.9	100	<0.001	<0.001	0.1	<0.1	<0.01	<0.001	71	71
Water	4	1	26	8	<0.01	<0.001	70	91	778	2271
Soil	50	81	<0.01	<0.001	50	19	<0.01	<0.01	138	87
All three	17	6.5	21	8	7	0.5	55	85	329	810

Laboratory experiments have shown high BCF for D4 and somewhat lower for D5 and MM (Table 3). High vaporisation rate from water as a result of high volatility, combined with high tendency to bind to particles and thereby high sedimentation rates acts to reduce the actual concentrations available for uptake in biota (HSDB, 2004).

Ecotoxicity data are given in Table 6 below.

Table 6. Ecotox data for D4 (US EPA, 2005) IMBL: Immobilisation, GRO: growth, MOR: mortality, REP: Reproduction, ITX: intoxication, NOC, MULT: Multiple effects recorded as one result

Common name, Scientific name	Endpoint	Effect	Duration	Conc (ug/L)
Opposum Shrimp (<i>Americamysis bahia</i>)	NOEC	IMBL	14 d	9.1
Midge (<i>Chironomus tentans</i>)	NOEC	GRO	14 d	>15
Midge (<i>Chironomus tentans</i>)	NOEC	MOR	14 d	>15
Sheepshead minnow (<i>Cyprinodon variegatus</i>)	NOEC	MOR	14 d	6.3
Water flea (<i>Daphnia magna</i>)	NOEC	IMBL	48 h	>15
Water flea (<i>Daphnia magna</i>)	NOEC	REP	21 d	1.7 - 15
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	LC50	MOR	14 d	10, 8.5 - 13
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	LOEC	MOR	14 d	6.9
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	NOEC	MOR	14 d	<=4.4
Rainbow trout, donaldson trout (<i>Oncorhynchus mykiss</i>)	NOEC	NOC, MULT	93 d	4.4
Water flea (<i>Daphnia magna</i>)		ITX, IMBL	21 d	1.7 - 15

The lowest value for No Observed Effect Concentrations (NOEC) was obtained for water fleas (*Daphnia magna*), a zooplankton that is an important grazer in many limnic ecosystems. Lassen et al., (2005) have derived so-called Chronic Values (ChV) for fish for a number of siloxanes by using

the U.S. EPA PBT Profiler software (U.S. EPA, 2005). ChV is the same as the chronic no effect concentration and shows at what concentration no long-term effects are expected. The results are listed in Table 7.

Table 7. Chronic values for fish derived by Lassen et al., (2005)

Chemical abbreviation	Fish ChV (mg/L)
D4	0.058
D5	0.021
MM	0.062
MDM	0.028

Some evidence exists on the potential carcinogenicity of D5 (U.S. EPA, 2003).

D4 is classified as R62 "possible risk of impaired fertility" and as R53 "may cause long-term adverse effects in the aquatic environment" (KemI, 2004). D4 is also classified as a PBT/vPvB substance and hence as a phase-out substance in the priority data base of the Swedish Chemicals Inspectorate and as such not supposed to be used in any new chemical applications within Sweden. MM is included on the OSPAR candidate list for dangerous substances.

3 Production and use

Siloxanes are widely used over the world and D4, D5 and MM are chemicals of high production volume within the European Union. Table 8 shows the main fields of application for siloxanes within the Nordic countries.

Table 8. Fields of application of siloxanes in the Nordic countries (SPIN, 2005).

	Field of application
D4	Fuel additives, Cleaning/washing agents, Impregnation materials, Adhesives, Binding agents, Surface treatment, Construction materials, Paints, laquers and varnishes, Fillers, Reprographic agents, Process regulators, Anti-set-off, Anti adhesive agents
D5	Fuel additives, Surface treatment, Cleaning/washing agents, Fillers, Impregnation material, Adhesives, Binding agents, Paints, laquers and varnishes, Reprographic agents, Softeners, Surface active agents, Process regulators
D6	Surface treatment, Paint, laquers and varnishes
MDM, MD2M	Paint, laquers and varnishes

In Sweden, there is limited use of MD2M, MDM and MM, and more extensive use of D4, D5 and D6 (Figure 1). The use of MD3M is however confidential (too few users). In the cosmetics industry, the name cyclometicone is used for the cyclic dimethylsiloxanes. Examples of product categories are body lotion, hair styling products, creams, lipstick and deodorants.

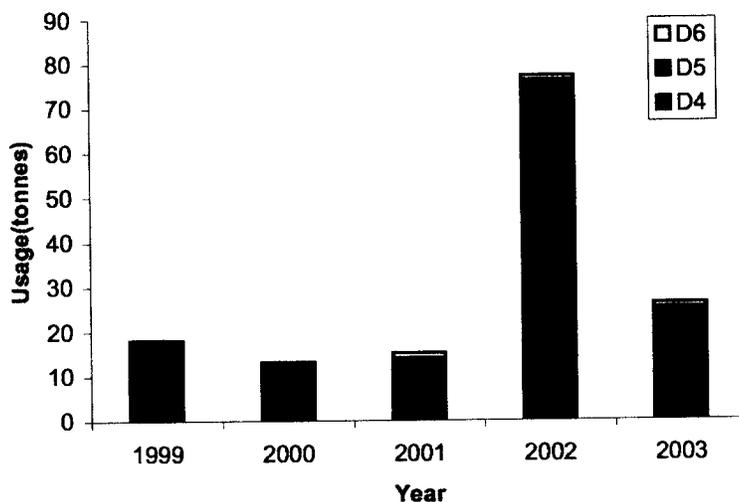


Figure 1. Swedish use of D4, D5, and D6 (SPIN, 2005)

4 Previous measurements in the environment

Data on the occurrence of siloxanes in the environment are scarce. Table 9 shows examples on previously detected concentrations of MM and D4 in the environment.

Table 9. Concentration levels of siloxanes in various matrices.

Country	Location	Matrix	MM	D4	Reference
Sweden	Landfill	Percolate water ($\mu\text{g/l}$)	2-106	1-2	Paxéus, 2000
Germany	Landfill 1	Biogas (mg/m^3)	1.04-1.31	7.97-8.84	Schweigkopfler & Niessner 1999
Germany	Landfill 2	Biogas (mg/m^3)	0.38-0.77	4.24-5.03	Schweigkopfler & Niessner 1999
Germany	STP 1	Biogas (mg/m^3)	0.05	6.40-6.98	Schweigkopfler & Niessner 1999
Germany	STP 2	Biogas (mg/m^3)	0.01	2.87-3.02	Schweigkopfler & Niessner 1999

In order to extend reference data, Table 10 shows results from previous measurements of polydimethylsiloxanes found in the literature, even though the specific compounds were not specified.

Table 10. Concentration levels of polydimethylsiloxane in different matrices (HSDB, 2004)

Matrix	Country	Concentration
Surface water near sewage treatment plant	Australia, Japan and USA, 1997	0.8-5 $\mu\text{g/l}$.
Surface water near industrial area	Japan, 1997	2.8 -54.2 $\mu\text{g/l}$
Water from a sewage water treatment plant	USA and Japan, 1997	Barely detected
Sludge	Australia, Canada, Germany, Japan and USA, 1997	20-50100 mg/kg dw
Sediment	Australia, Germany, Japan and USA, 1997	314 mg/kg dw
Fish		0.6-0.7 mg/kg

As part of an interdisciplinary field study (DBH: Dampness in buildings and health) indoor air measurements in 400 Swedish homes has recently been carried out. The results for siloxanes from a study of VOCs in air in children's bedrooms are summarised in Table 11 (Schmidbauer, 2005).

Table 11. Concentration of siloxanes in indoor air in Sweden.

Siloxane	Number of homes with siloxane detected	Mean ($\mu\text{g/m}^3$)	Min ($\mu\text{g/m}^3$)	Max ($\mu\text{g/m}^3$)
D5	250	9.7	0.5	79.4
D6	142	7.9	0.6	164
D4	73	9.0	0.6	51.2
D7	8	6.4	1.2	35.5
MD2M	5	20	5.3	73.2
MDM	2	7.4	2.5	12.3
MM	1	1.5	-	-
D3	1	7.3	-	-

5 Sampling strategy and study sites

5.1 National

A national sampling strategy was developed in order to determine the environmental concentrations of siloxanes and related substances in different environmental matrices in Sweden. An additional aim of the sampling programme was to identify major emission sources as well as important transport pathways. The sampling programme was therefore based on identified possible emission sources and use of the chemicals as well as on the behaviour of the substances in the environment. The programme included both measurements in background areas and close to potential point sources. Measurements of diffuse pathways from the society including sewage systems were also included. The programme is summarised in Table 12.

Table 12. National sampling strategy for siloxanes

Site	Air	Water	Sediment	Fish	Sludge	Total
Background						
Råö	3					3
Various			3	3		6
Point sources						
Stenungsund (industrial area)	3	3	3	3		12
Chemical production plant, Sundsvall	3	3	3	3		12
Diffuse sources						
STP Henriksdal					1	1
STP Borås					1	1
STP Göteborg					1	1
Stockholm	3					3
Total						39

Additionally 39 samples of human breast milk were analyzed.

5.2 Regional

Swedish county administrative boards have had the possibility during the sampling period to add regional samples to the national sampling programme. Different counties have chosen different strategies for their regional sampling scheme. One way of selecting has been to increase the number of samples for substances connected to their environmental programmes i.e. substances that have been regulated or included in the national priority database. Another strategy has been to choose substances where environmental levels are expected to differ from national levels because of intense use within the county i.e. local industrial areas or because of geographic proximity to European industrial areas.

Thirteen regional county administrative boards participated in the regional screening programme with in total 99 additional samples consisting of 10 fish samples, 20 water samples, 17 sediment samples and 52 sludge samples.

6 Methods

6.1 Sampling

As a guideline for adequate and consistent sampling, a manual for the sampling personnel in the national as well as the regional screening programmes was developed. Detailed instructions for sampling, storing and transport were given. Sampling protocols for all sample types were included in the manual. The overall aim of the sampling protocols was to:

1. Guide the personnel, responsible for sampling on how to avoid contamination of the samples
2. Ensure documentation of the sampling procedure, quality of the sample and environmental and physical circumstances during the sampling.

All samples from the regional county administrative boards were sent to IVL Swedish Environmental Research Institute for analysis.

6.1.1 Air

Air samples were collected by pumping air through two adsorbent tubes in series containing Tenax™. The airflow varied between 50 and 100 ml/min. The sampling duration was 2 hours. An additional unexposed adsorbent tube was used as a field blank.

6.1.2 Water

Water samples were collected with pre-heated (400°C) bottles fitted with aluminium foil lined screw cap and stored in a refrigerator until analysed. A bottle with milli-Q water, which was exposed to the surrounding environment during the sampling time, was used as a field blank.

6.1.3 Sediment

Sediment samples from lakes or sites close to the coast were collected by means of a Kajak sampler. The sediment core was sliced and transferred into pre-heated (400°C) glass jars fitted with aluminium foil lined screw caps and stored in a refrigerator until analysed. A glass jar filled with diatomaceous earth, which was exposed to the surrounding environment during the sampling time, was used as a field blank.

The four marine sampling sites (Ö Gotlandsdjupet, Ö Öland, Norrköpingsdjupet, Ö Landsortsdjupet) were chosen from areas with continuous deposition of fine-grained sediment. These sites were identified with hydroacoustic methods (shallow seismic, sub-bottom profiler and chirp side-scan sonar). Prior to sampling, the bottom at the sampling site was inspected with a submarine video camera. Furthermore, a sediment-core from the site was X-rayed with a sediment-scanner (Cato et al. 2000) in order to detect unwanted physical disturbances as strong bioturbation, anchoring, trawling, etc. Sites, which fulfilled the sedimentological demands set up, were then sampled with a Gemini corer and the cores were sliced in vertical position with a core-cutter onboard. Surface sediments (0-1 cm) from four cores taken at each site were mixed in order to neutralise sediment inhomogenities. The samples collected were stored dark and frozen in pre-cleaned and burned glass bottles.

6.1.4 Sludge

The staff at the different treatment plants collected the sludge samples from the anaerobic chambers. The sludge was transferred into pre heated (400°C) glass jars fitted with aluminium foil lined screw caps and stored at 4°C or -18°C until analysed. A glass jar filled with diatomaceous earth, which was exposed to the surrounding environment during the sampling time, was used as a field blank.

6.1.5 Fish

Fish were collected by means of fishing net, hoop net or fishing-rod, individually wrapped in aluminium foil and stored in a freezer (-20°C). Later the fish were dissected and muscle samples taken for analysis. Muscle samples from herring from background areas were directly supplied from The Environmental Specimen Bank and the Museum of Natural History (A. Bignert and Colleagues).

6.1.6 Breast milk

Human breast milk samples were provided by The University Hospital of Lund (Department of Occupational and Environmental Medicine). The sampling strategy and methodology have been described elsewhere (Appelgren 2005, in prep.).

The samples were primarily collected for analysis of phthalates. They were acidified with phosphoric acid (1 M; 125 µl/ml) immediately after collection. The samples were stored in a freezer (-80°C) and were transported to the IVL laboratory in Stockholm on dry ice. The obtained samples were marked with numbers and carried no personal information or medical history.

6.2 Analysis

6.2.1 Analysis of sludge, sediment, breast milk and water

The samples were diluted with water and purged with a gas stream passing through an adsorbent from which the analytes were thermally desorbed and analysed by GC-MS.

Approximately 2 g of wet sludge were diluted to 20 ml with MilliQ water and homogenised with a high frequency mixer (Polytron). Approximately 1 ml of the slurry was weighed in to the purge & trap apparatus and diluted to 10 ml. 0.5 ml buffer solution (2M K₂HPO₄, 0.4M HCl, 80g Na₂EDTA 2H₂O per litre) was added. Sediment was diluted in a similar way, but homogenised by shaking only. Breast milk, 2 ml, was diluted to 5 ml with Milli Q water.

The purge & trap apparatus for sludge, sediment, breast milk and waste water samples consisted of a 25 ml graduated glass test tube with an adapter with one inlet for a Pasteur pipette extending to the bottom of the tube and one side arm to which an empty adsorbent tube was connected using flexible tubing (Viton). This tube acted as a short cooler and water trap. An adsorbent tube containing 0.25 g Tenax TA was connected to the empty tube, the Pasteur pipette was connected to the purge gas (nitrogen, 50 ml/min) and the tube was immersed in a thermostatic water bath. An electric fan facilitated air cooling of the upper part of the apparatus. Samples were purged at 70°C

for 20 min (analysis of MM) and for 2h (all other siloxanes). For low contaminated water samples a gas washing bottle with glas frit gas inlet was used as purge vessel. Water volume was 60 - 80 ml, the other conditions was the same as above.

The adsorbent tube was transferred to a thermal desorber (Unity, Markes) connected to a GC-MS instrument (6890N, 5973N, Agilent). Prepurge time was 2 min, tube desorption time 5 min at 225°C, the trap was held at 3°C and heated at 32°C/s to 250°C. The desorbing flow was 30 ml/min and the split flow 10 ml/min. The flow path temperature was 150 °C and it connected directly to the GC-column, which was a CP-Sil 8CB 30m × 0.25 mm id, film thickness 0.5µm (Varian). The column temperature was 40°C for 3 min, programmed to 200°C at 12°C/min and to 260°C at 30°C/min. The carrier gas was helium held at constant pressure 10 psi measured at 40°C. The mass detector was used in single ion recording mode.

500 mg each of D3, D4, D5, MM, MDM, MD2M, MD3M (Aldrich) and D6 (Gelest) were mixed in a test tube. The chemicals were of 97% declared purity or better. 200 mg of the mixture were dissolved in methanol and diluted to 25 ml. This made a stock solution of 1 mg/ml for each component. This solution was further diluted with methanol. Different amounts of this solution were added to 10 ml MilliQ water and 0.5 ml buffer solution in the purge & trap apparatus and analysed as samples. In this way a seven-point linear calibration curve was constructed and used for quantification of the samples. The blank level and calibration was regularly checked by running water blanks and one or more of the calibration solutions together with the samples.

Table 13. Retention times, quantification and supporting ions used for analysis of siloxanes

	RT, min	Quant. Ion	Supp. ion 1	Supp. Ion 2	Supp ion 3
MM	4.28	147	148		
D3	7.07	207	208		
MDM	8.15	221	73		
D4	10.04	281	282		
MD2M	11.17	207	295	73	
D5	12.41	355	267	73	
RD3M	13.60	281	369	147	73
D6	14.71	341	429	325	

6.2.2 Analysis of air samples

The adsorbent tubes were analyzed using the thermal desorber and GC-MS system described above (6.2.1). Calibration was carried out using adsorbent tubes to which a small volume of siloxane standard solution in methanol had been injected while a stream of nitrogen was flowing through the tube.

6.2.3 Analysis of fish samples

Different methods for determination of D4 and D5 in biota are described in literature (Flassbeck et al., 2001, Flassbeck et al. 2003, Kala et al. 1998, and Varaprath et al. 2000). However, none of the described methods were using high resolution mass spectrometry for quantification. Due to the ubiquity of the cyclic siloxanes great care was given to reduce the risk for contamination of the samples with siloxanes through direct contact with the lab staff, the equipment used for sample

storage, preparation, and extraction. To avoid evaporation loss of the volatile siloxanes and to reduce the contamination risk, a very short and comprehensive sample preparation and quantification method was developed and validated.

Typically the sample material was thawed and homogenised with a household mixer. A 0.30 g aliquot of the sample was mixed thoroughly with 1 mL n-hexane on a whirl mixer for 5 min. The mixture was separated by centrifugation at 10000 rpm and the clear solution was carefully removed with a Pasteur pipette and transferred without any further treatment into a GC-MS vial. There was no significant difference in the results from sample extracts which were dried with sodium sulfate according to the procedure published by Dow Corning (Varaprath et al., 2000) and the results from undried extracts.

For method testing and calibration a solution was prepared containing D3, D4, D5, D6, MM, MDM, MD2M, and MD3M at concentrations of about 3 ng/ μ L and about 30 ng/ μ L in n-hexane.

The sample extracts were analysed on a GC-HRMS system (GC: 6890 Agilent, MS: Micromass Ultra Autospec) using the following parameters: Gas chromatograph: Splitless injection (injector temp: 200°C), Helium as carrier gas (1 mL/min; constant flow), 25m \times 0.2mm \times 0.11 μ m Agilent Ultra2 capillary; Temperature program: 35°C, 3min, 7°/min, 130°C, 0 min, 30°/min, 325° C, 5 min. Interface temperature: 250 ° C. Mass spectrometer: Ion source temperature: 200° C; Electron impact mode with accelerating voltage 8000 V, Resolution: 10000 at 5 %; Single ion monitoring mode acquiring the following masses (m/z): 147.0661 (MM), 207.0329 (D3), 221.0849 (MDM), 281.0517 (D4), 295.1037 (MD2M), 355.0705 (D5), 369.1225.

Before and after a series of 10 samples including a complete method blank the calibration solution was injected in 2 parallels. Quantification was performed as external standard quantification. Analysis of biota was done by NILU, Norwegian Institute for Air Research, Kjeller, Norge.

7 Results and discussion

All individual results are given in Appendix 1, Tables A3, A4 and A5.

7.1 National programme

7.1.1 Sludge

Sludge from the three large municipal sewage treatment plants (STPs) in Stockholm, (Henriksdal), Gothenburg (Ryaverken) and Borås (Gässlösa) showed a similar pattern of siloxanes. D5 dominated with concentrations in the range 10 000 to 22 000 ng/g DW. D6 and D4 followed with concentrations close to 3000 and 300 ng/g DW, respectively. The concentrations of the linear analogues were considerably lower: MM and MDM <2 ng/g DW, MD2M 8-16 ng/g DW and MD3M 24-46 ng/g DW.

The concentrations as well as the distribution among the measured siloxanes are illustrated below (Figure 5) where also the concentrations found in sludge samples from the regional programs are given.

7.1.2 Air

The results from analysis of siloxanes in air are presented in Figure 2.

The background samples from Råö, which were all taken in November 2004, showed great variations in the concentrations; it was 6 times higher during the last occasion. The concentrations during this period were higher compared to the samples collected both at point sources and in the urban area. Back trajectories using the HYSPLIT model (NOAA, 2005), which indicate the origin of the air masses, could not explain the observed concentration differences at Råö. Contamination of the sample can not be ruled out. The samples collected close the first point source, Stenungsund show somewhat elevated concentrations compared to two of the background samples as well as to the second point source Stockvik. Concentrations in the urban area (Stockholm) were in the same order of magnitude as the concentrations at the background site and in Stockvik.

The siloxane that occurred in the highest concentration in most of the samples was D4, the most volatile of the measured cyclic siloxanes. The most volatile linear siloxane, MM, was detected in all three samples from Stenungsund and in the last sample from the background site, but not in the other samples.

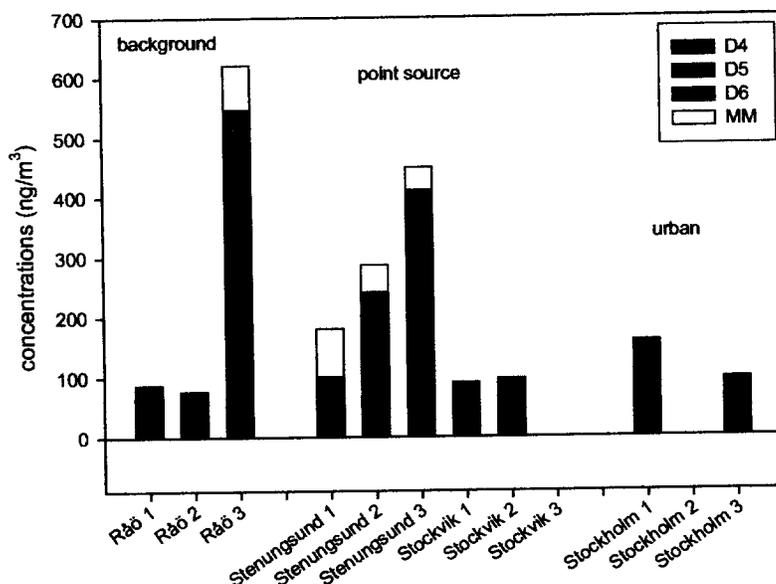


Figure 2. Measured concentrations of siloxanes in air, within the national programme.

7.1.3 Fish

No siloxanes were detected in the muscle samples from different species of fish. The obtained limits of detection (LOD) are shown in Table 14.

Table 14. LOD of siloxanes in fish muscle

Siloxane	LOD, ng/g ww
D4	5
D5	5
D6	5
MM	0.4
MDM	0.3
MD2M	0.4
MD3M	0.5

7.1.4 Water

Only two of the six water samples from the national sampling programme, which were all collected at the point sources, showed detectable concentrations of siloxanes, and in each case only one single substance was detected. D6 was found in a concentration of 0.04 µg/L in the surface water sample from the industrialised area in Stenungsund and MM was detected (0.0006 µg/L) in outgoing sewage water from the Stockvik industrial site.

7.1.5 Sediment

No cyclic siloxanes were detected in any of the sediment samples analysed within the national programme. Linear siloxanes were detected in two samples, from the bay of Sundsvall outside Stockvik industrial area. MD2M was detected in one of the deeper sediments (2-7 cm) in a concentration of 0.9 ng/g dw and MD3M was detected in both deeper sediment samples from this area in concentrations of 1.7 and 0.7 ng/g dw respectively.

7.1.6 Breast milk

One or more of the cyclic siloxanes, D4, D5 and D6, was found in 11 out of 49 human breast milk samples. The results are illustrated in Figure 3. The maximum concentration of D4 was 10 µg/l, of D5 4.5 µg/l and of D6 4.8 µg/l. Trace amounts of linear siloxanes were found in 6 of the samples, however all concentrations were below 0.04 µg/l.

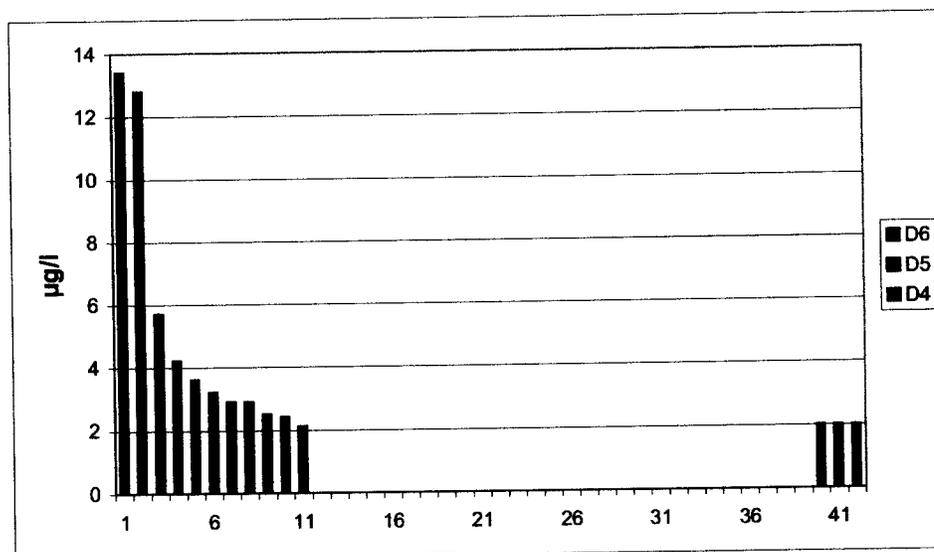


Figure 3. Concentrations of D4, D5 and D6 (µg/l) in breast milk. The samples are sorted in decreasing order of concentration. Results for sample 12-39 are below the quantification limit. The bars to the right illustrate the quantification limits.

7.2 Regional programme

7.2.1 Sludge

An overview of concentrations of siloxanes in the sludge samples from the regional screening is illustrated in "box plots" in Figure 4. The results from the three STPs in the national programme are also included in the plots. The concentrations in sludge from the different STPs are graphically illustrated in Figure 5.

There was a great variation in the concentrations in the sludges from the different STPs. D5 and D6 were detected in all 52 sludge samples except in one industrial sludge from Olofström. The sample from Fagersta STP, Borlänge, showed the highest concentrations, D5, 54 000 ng/g DW and D6, 8 400 ng/g DW which were substantially higher than in the sludges from the large STPs in Stockholm, Göteborg and Borås. The lowest concentrations (D5 54 ng/g DW and D6 37 ng/g DW) were found in a sample from Härjedalen, Björnrike STP, that also had the highest dry weight (69% DW) of all the samples. It is possible that siloxanes had been lost in the drying procedure. The sample with next lowest concentrations (Flen STP, D5 570 ng/g DW, D6 170 ng/g DW) was not exceptionally dry (28% DW)

D4 was detected in 34 of the samples. The concentration range was 130 - 2300 ng/g DW. Concentrations of D4 higher than 1000 ng/g DW was found in the samples from Kristianstad, Gnesta, and Lidköping.

The concentrations of the linear analogues were, as in the national programme, substantially lower: MM was found in only three samples with a maximum of 8 ng/g DW, MDM in 12 samples with a maximum of 22 ng/g DW. MD2M was in the range <0.5 to 37 and MD3M <0.5 to 160 ng/g DW.

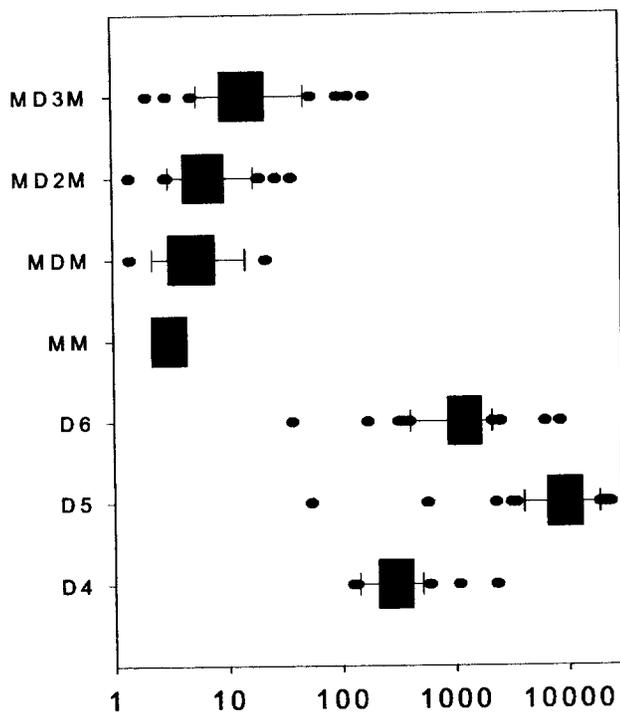


Figure 4. Concentrations of siloxanes (ng/g DW, logarithmic scale) in sludge from all municipal STPs. The lower and upper boundaries of the box represent the 25- and 75-percentiles, the line within the box is the median concentration. The whiskers represent the 10- and 90-percentiles, and the dots are individual results outside this range.

A summary of the siloxane concentrations found in the 54 sludge samples from municipal STPs (national and regional programme) is given in Table 15.

The concentration of D5 found in sludge could be compared to the concentrations of some other widespread organic contaminants without implying any similarities in their effects. In a recent study concerning sludge from 20 Swedish STPs, di-(ethylhexyl)-phthalate (DEHP) averaged to 49 000 and 4-nonylphenol to 9 000 ng/g DW (Bignert and Remberger, 2005). In another study including 23 Swedish STPs the average concentration of 4-nonylphenol was 15 000 ng/g DW (Remberger et al., 2004). Thus, the concentration level of D5 found in the present study is similar to that for 4-nonylphenol and somewhat lower than that for DEHP. Siloxanes may therefore be added to the list of organic pollutants that make it problematic to use municipal sewage sludge in a sustainable way, i.e. as a source of nutrients in agriculture.

Table 15. Summary of siloxane concentrations found in sludge from the 54 municipal STPs in the national and regional programmes.

Siloxane	Detected in number of samples	Median conc. ng/g DW	Average conc. ng/g DW	Maximum conc. ng/g DW
D4	37	310	390	2300
D5	54	9500	11000	54000
D6	54	1300	1500	8400
MM	3	5	5	8
MDM	12	5	7	37
MD2M	46	7	9	37
MD3M	44	15	24	160

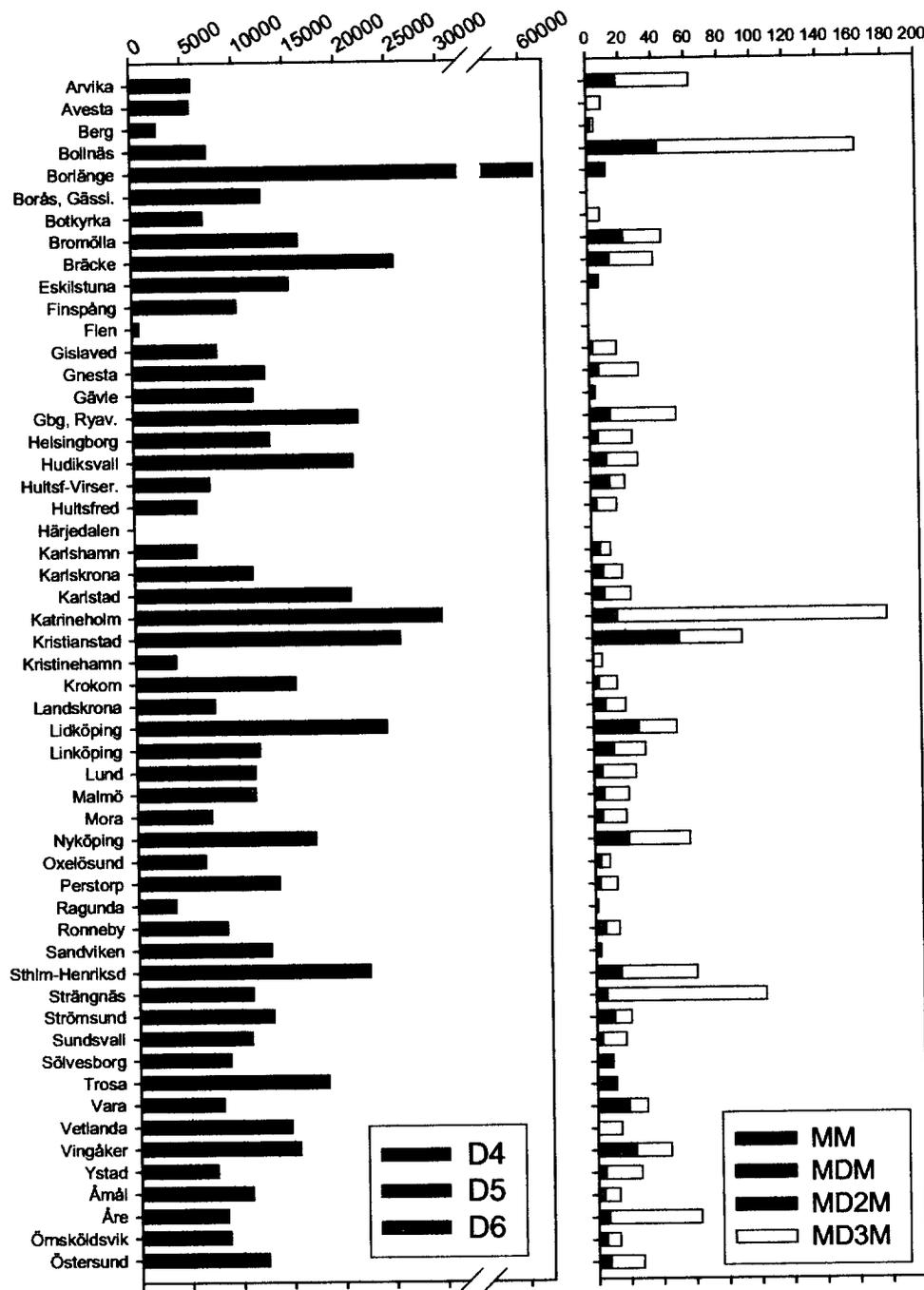


Figure 5. Concentrations of siloxanes in sludge from municipal STPs, ng/g DW. Please note the different concentration scales in the left and right parts of the figure.

7.2.2 Water

Four samples of influent water to municipal STPs were included in the regional program. D5 was found in three of the samples in concentrations from 1.1 µg/L (Gislaved) to 0.1 µg/L. The limit of detection was 0.04 µg/L. D6 was like D5 highest in Gislaved (0.27 µg/L) and 0.06-0.08 µg/L in the other influents. In the 12 effluent water samples from municipal STPs D5 was found in one (Gislaved, 0.051 µg/L) and D6 in five. The concentration of D6 was highest in Gislaved (0.23 µg/L) and 0.05-0.08 µg/L in the remaining three effluents. The concentrations of D5 and D6 in the industrial effluent sample from a pulp and paper production plant were 0.059 and 0.15 µg/L respectively. D4 was not found in any of the water samples.

In percolate waters from landfills detectable concentrations of MM (0.058 and 0.0015 µg/L) were found, while no cyclic siloxanes were detected. MM was also found in one effluent from a municipal STP (Arvika STP 0,0098 µg/L. MM was the only linear siloxane detected. in the water samples.

7.2.3 Sediment

The siloxanes D5, D6, MD2M and MD3M were found in sediments from Himmerfjärden, Botkyrka in the concentrations 190, 51, 1.2 and 0.44 ng/g DW respectively.

In lake Vänern, D5, D6 and MD3M occurred in sediment from Åsjorden (36, 200, 0.45 ng/g DW respectively) and MD2M (0,3 ng/g DW) in sediment from Kattfjorden.

7.2.4 Biota

No siloxanes were detected in biotic samples, which were all muscle samples from different species of fish. The obtained limits of detection (LOD) are shown in Table 14 above.

8 Conclusions

D4, which is classified as a phase out substance was found in 37 out of 54 municipal sludge samples in concentrations from 130 to 2 300 ng/g DW and in various air samples in concentrations up to 300 ng/m³. D4 was not found in any of the water, sediment or fish samples.

D5 was the dominating siloxane in most samples. It was found in all sludge samples from municipal treatment plants. The average concentration was 11 000 ng/g DW.

MM, which is on the OSPAR candidate list for dangerous substances, was found in leachate water from landfills and in low concentrations in air in the Stenungsund chemical industrial area.

Siloxanes were not found in biota (fish muscle).

9 Acknowledgements

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Appendix 1. Sample description and results

Table A 1 Sample list for national screening of siloxanes. DW = dry weight (sediment, sludge), LW = lipid weight (biota)

Category	Sample ID	City	Site	Matrix Notes	Sampling date	X RT90	Y RT90	DW/LW(%)
Back-ground		Rådö		Air	2004-11-16			-
		Rådö		Air	2004-11-23			-
		Rådö		Air	2004-11-30			-
	MR-3750		Ö Gotlandsdjupet	Sed 0-2 cm, 121 m depth, SGU 04-0058	6216805	1697299	9.5	
	MR-3751		Ö Öland	Sed 0-2 cm, 77m depth SGU 04-0368	6277308	1607374	8.3	
	MR-3752		Norrköpingsdjupet	Sed 0-2 cm, 179m depth SGU 071-001	6435399	1625504	6.6	
	MR-3639		V. Fladen	biota herring	57° 04' 47"	15 °55' 18"	1.7	
	MR-3640		Ångskärsklubb	biota baltic herring	65*43'	23**03'	2.3	
	MR-3641		Landsort	biota baltic herring	57° 12' 27"	11°39' 53"	4.5	
	Potential point sources		Stenungsund		Air	2004-11-08		
		Stenungsund		Air	2004-11-09			-
		Stenungsund		Air	2004-12-08			-
MR-3872			Stenungsund	Sed A1	6446247	1264535	27.2	
MR-3874			Stenungsund	Sed D7	6446760	1264051	31.1	
MR-3876			Stenungsund	Sed E1	6446077	1253472	27.2	
MR-3878			Stenungsund	water A1	6446247	1264535	-	
MR-3880			Stenungsund	water D7	6446760	1264051	-	
MR-3882			Stenungsund	water E1	6446077	1253472	-	
MR-4053			Stenungsund	biota Eelpout, females			-	
MR-4054			Stenungsund	biota Eelpout, males			-	
MR-4055			Stenungsund	biota Eelpout, juveniles			-	
MR-4050			Sundsvall	Sundsvall bay			-	
MR-4051			Sundsvall	Sundsvall bay			-	
MR-4052			Sundsvall	Sundsvall bay			-	

Table A 1. Cont.

Category	Sample ID	City	Site	Matrix Notes	Sampling date	X RT90	Y RT90	DW/LW(%)
Potential point sources			Stockvik	Air	2004-11-04	-	-	-
			Stockvik	Air	2005-01-26	-	-	-
			Stockvik	Air				
	MR-3600	Sundsvall	Bay outside Stockvik 1	Sed 0-2 cm	2004-10-21	6918274	1581483	41.3
	MR-3601	Sundsvall	Bay outside Stockvik 1	Sed 2-7 cm	2004-10-21	6918274	1581483	43.6
	MR-3603	Sundsvall	Bay outside Stockvik 2	Sed 0-2 cm	2004-10-21	6915504	1581340	44.0
	MR-3604	Sundsvall	Bay outside Stockvik 2	Sed 2-7 cm	2004-10-21	6915504	1581340	30.4
	MR-3612	Sundsvall	Akzo-Nobel Stockv. Expanceel Tp	water effluent	2004-10-22			
	MR-3613	Sundsvall	Akzo-Nobel Stockvik	water	2004-10-22			
	MR-3610	Sundsvall	Bay outside Stockvik	water recipient	2004-10-21	6914691	1581178	-
					2004-11-18			
	Diffuse sources		Stockholm	Hudiksvallsgatan 2	Air	2004-12-02	-	-
		Stockholm	Hudiksvallsgatan 2	Air	2004-12-03	-	-	-
		Stockholm	Hudiksvallsgatan 2	Air				
MR-3760		Stockholm	Henriksdal STP	sludge	2004-11-16			28.6
				700.000 pe, mech, chem, bio cleaning. No major industrial impact				
				p.e 98250, mec, chem and bio cleaning, textile-, graphic- industries, slaughterhouse, laundry finishing activities, landfill leachate, incineration				
MR-3790		Borås	Gässlösa STP	sludge	2004-11-24			22.4
MR-3867		Gothenburg	Ryaverket STP	sludge	2004-12-15			28.8
				p.e 620 683, mech, chem, bio cleaning, incineration, industrial and municipal				

Table A 2 Sample list for regional screening of siloxanes. DW = dry weight (sediment, sludge), LW = lipid weight (biota)

County	Sample ID	City	Site	Site information	Matrix	Notes	pa (STP)	Treatment (STP)	Sampling date	DW/LW (%)
Blekinge	MR-3462	Karlskrona	Mörum landfill		water	leachate			2004-09-23	-
	MR-3468	Karlskrona	Mörum STP		sludge		3900	active sludge, chem, mech	2004-09-28	26.1
	MR-3537	Karlskrona	Koholmen STP		sludge		40000	bio, chem post susp. mech.	2004-10-07	14.7
	MR-3529	Olofström	Volvo personvagnar		sludge				2004-10-06	28.3
	MR-3533	Ronneby	Angelskog landfill		water	leachate			2004-10-06	-
	MR-3538	Ronneby	Angelskog landfill		water	background				-
	MR-3531	Ronneby	Ronneby STP		sludge		15000	bio, chem, mech.	2004-10-06	15.4
	MR-3487	Sölvesborg	Sölvesborg STP		sludge		10300	active sludge with N-elimination	2004-09-29	18.8
	MR-3498	Avesta	Båsingan		sediment	lake			2004-10-04	21.4
	MR-3501	Avesta	Båsingan		fish				2004-09-29	-
Dalarna	MR-3478	Avesta	Krylbo		sludge	digested	17000	suspension, active sludge	2004-09-29	24
	MR-3540	Borlänge	Fagersta		sludge	digested	44000	suspension, active sludge	2004-10-12	29
	MR-3535	Mora	Venjan STP		sludge	digested	500	mech, chem	2004-10-06	0.7
	MR-3496	Mora	Venjansjön		sediment	lake			2004-10-03	5.19
	MR-3500	Mora	Venjansjön		fish				2004-10-01	-
	MR-3584	Bollnäs	Bollnäs STP		sludge				2004-10-20	14.6
	MR-3549	Gävle	Duvbacken STP		sludge				2004-10-12	19.9
	MR-3846	Hudiksvall	Resselvans STP		sludge				2004-12-07	26.6
	MR-3555	Sandviken	Sandviken STP		sludge				2004-10-13	22.0
	MR-3687	Berg	Myrviken STP	Mun.	sludge	Primary		mech, dehydration, compost	2004-11-01	19.2
Gävleborg	MR-3619	Bräcke	Bräcke STP	Mun., Ind.	sludge	Primary	1800	mech, bio	2004-10-19	10.5
	MR-3470	Härjedalen	Björnrike STP	Mun. (sample taken during tourist season not according to sampling instructions)	sludge	Primary	2325	mech, chem	2004-06-21	76.7
	MR-3574	Krokom	Hissmöfors STP	Mun., Ind.	sludge	Primary	3600	mech, chem, bio	2004-10-19	13.4
	MR-3586	Ragunda	Överammer STP	Mun.	sludge	Primary		Dehydratation	2004-10-19	28.8
	MR-3625	Strömsund	Strömsund STP	Mun., Ind.	sludge	Primary	7000	mech, chem, bio	2004-10-27	17.6
	MR-3469	Åre	Åre STP	Mun. (sample taken during tourist season not according to sampling instructions)	sludge	Primary	16000	mech, chem, bio	2004-04-13	18
	MR-3472	Östersund	Gövikens STP	Mun., Ind. (Dairy)	sludge	digested	55900	mech, chem, bio	2004-09-29	27.3

Table A 2. Cont

County	Sample ID	City	Site	Site information	Matrix	Notes	pe (STP)	Treatment (STP)	Sampling date	DW/LW (%)
	MR- 3733	Gislaved	Gislaveds STP		sludge					16.5
	MR- 3735	Gislaved	Gislaveds STP		water	effluent				-
	MR-3731	Gislaved	Gislaveds STP		water	influent				-
Jönköping	MR- 3803	Vetlanda	Gröpplebäcken		sediment				2004-11-24	22,8
	MR- 3802	Vetlanda	Landsbro STP		sludge				2004-11-24	3,87
	MR- 3807	Vetlanda	Landsbro STP		water	influent			2004-11-24	-
	MR-3808	Vetlanda	Landsbro STP		water	effluent			2004-11-24	-
	MR-3800	Hultsfred	Hullingen		sediment				2004-11-24	34,4
	MR-3799	Hultsfred	Hultsfred STP		sludge				2004-11-24	18,8
Kalmar	MR-3809	Hultsfred	Hultsfred STP		water	Effluent			2004-11-24	-
	MR-3810	Hultsfred	Hultsfred STP		water	Influent			2004-11-24	-
	MR-3798	Hultsfred	Virserum		sediment	Recipient			2004-11-24	58,5
	MR- 3805	Hultsfred	Virserum STP		water	Effluent			2004-11-24	-
	MR-3797	Hultsfred	Virserum STP		sludge				2004-11-24	14,5
	MR-3804	Hultsfred	Virserum STP		water	Influent			2004-11-24	-
	MR-3627	Oskarshamn	Mouth of Emån		sediment	Recipient			2004-11-24	22,7
	MR-3466	Bromölla	Bromölla STP		sludge				2004-09-28	25,7
	MR-3679	Bromölla	Ivösjön		sediment				2004-10-19	6,68
	MR-3682	Bromölla	Ivösjön		fish	Perch			2004-10-19	1,1
Skåne	MR-3459	Helsingborg	Helsingborg STP		sludge				2004-09-22	25
	MR-3706	Helsingborg	Coast	55 59 110-12 44 265	sediment				2004-11-03	43,4
	MR-3708	Helsingborg	Coast	55 58 915-12 44 313	fish	Flounder			2004-10-15	1,1
	MR-3677	Kristianstad	Hammarsjön		sediment				2004-10-19	14,2
	MR-3681	Kristianstad	Hammarsjön		fish	Flounder			2004-10-19	1
	MR-3514	Kristianstad	Kristianstad STP	Possible siloxane contamination from STP	sludge				2004-10-05	18,2
	MR-3489	Landskrona	Lundåkraverket	d.o	sludge				2004-09-29	22
	MR-3464	Lund	Källby STP		sludge	Thermophilic Incineration			2004-09-27	26,1
	MR-3456	Malmö	Sjölunda STP		sludge				2004-09-22	23,8
	MR-3623	Perstorp	Perstorp STP		sludge				2004-10-26	18,7

Table A 2. Cont

County	Sample ID	City	Site	Site information	Matrix	Notes	pa (STP)	Treatment (STP)	Sampling date	DW/LW (%)
Skåne	MR-3670	Perstorp	Storarydsdammen		sediment				2004-10-22	9.66
	MR-3672	Perstorp	Storarydsdammen		fish	Perch			2004-10-22	1.1
	MR-3453	Ystad	Ystad STP		slam				2004-09-22	15.8
Stockholm	MR-3480	Botkyrka	Himmerfjärdsverket		water	Effluent				-
	MR-3482	Botkyrka	Himmerfjärdsverket		sludge	Mixed sample (3 days)			2004-09-28 till 2004-100930	23.3
	MR-3449	Botkyrka	Himmerfjärden	X: 654978 Y:161021	sediment				2004-09-01	24.4
	MR-3450	Botkyrka	Himmerfjärden		fish	Perch			2004-09-01	-
	MR-3448	Södertälje	St Envättern	X:655613 Y:158793	sediment				2004-09-01	3.75
	MR-3451	Södertälje	St Envättern		fish	Perch			2004-07-22	-
	MR-3551	Flen	Flen STP	GB	sludge	Primary	10200	mech, chem,biol	2004-10-11	27.8
Södermanland	MR-3576	Gnesta	Gnesta STP	Chem., technical ind.	sludge	Digested	5200	mech, chem	2004-10-19	18
	MR-3568	Eskilstuna	Eskilstuna STP	Stormwater (possibly containing siloxanes), car repair shops	sludge	Digested	54763	mech,chem,biol, Wetland	2004-10-18	20.5
	MR-3578	Katrineholm	Katrineholm STP	printing ind., concrete ind., chicken slaughterhouse	sludge	Primary	32000	mech, chem, bio	2004-10-20	17.2
	MR-3570	Nyköping	Nyköping STP		sludge	Mesophilic Incineration	44500	mech, chem, bio		24.4
	MR-3637	Oxelösund	Oxelösund STP	SSAB, harbour, mechanical ind. (no process water)	sludge	Digested	12000	mech, wetland	2004-10-27	23
	MR-3572	Strängnäs	Strängnäs STP	Mun.	sludge	Primary	25000	mech, chem, bio		15.8
	MR-3553	Trosa	Vagnhärad STP	Mun., chemical, technical ind.	sludge	Digested	4100	mech, chem, wetland	2004-10-13	27.3
	MR-3557	Vingåker	Vingåker STP		sludge	Digested	7000	mech, chem, bio, wetland	2004-10-13	19.5
	MR-3709	Arvika	Vik STP	Mun., varying ind.	sludge	Digested	19500	mech, chem, bio	2004-11-02	14.5
	MR-3711	Arvika	Vik STP	d.o	water	Effluent	19500	mech, chem, bio	2004-11-02	-
Värmland	MR-3502	Grums	Vänern, Åsforden	Pulp and paperind., viscous manufacturing	sediment				2004-09-30	18.9
	MR-3506	Grums	Vänern, Åsforden	d.o	fish	Perch			2004-09-30	-
	MR-3507	Hammarö	Vänern, Kattfjorden	pulp and paper ind., chlorine/alkali and pesticide ind.	fish	Perch			2004-09-30	-
	MR-3504	Hammarö	Vänern, Kattfjorden	d.o	sediment				2004-09-30	13.1
	MR-3580	Karlstad	Skåre STP	Mun., varying ind.	water	Effluent	4005	mech, chem, bio	2004-10-19	-
	MR-3582	Karlstad	Skåre STP	d.o	sludge	Primary	4005	mech, chem, bio	2004-10-19	14.5
	MR-3629	Kristinehamn	Fiskartorpet STP	Mun.	sludge	Digested	16109	mech, chem, bio	2004-10-26/28	29.9
	MR-3631	Kristinehamn	Fiskartorpet STP	d.o	water	Effluent	16109	mech, chem, bio		-

Table A 2. Cont

County	Sample ID	City	Site	Site information	Matrix	Notes	pe (STP)	Treatment (STP)	Sampling date	DW/LW (%)
Väster-norrland	MR-3754	Sundsvall	Tivoliverket		sludge					19
	MR-3615	Timrå	SCA Östrand		sediment					24.0
	MR-3756	Timrå	SCA Östrand		water					-
Västra Götaland	MR-3726	Örnsköldsvik	Bodum STP		sludge				2004-11-10	31
	MR-3508	Lidköping	Lidköping STP	Mun.	water	Effluent	28000		?	-
	MR-3510	Lidköping	Lidköping STP	d.o	sludge	Primary	28000		?	19.5
	MR-3512	Vara	Vara STP	Mun., laundry	sludge	Digested	6360	mech,chem,biol	2004-10-04	19
	MR-3474	Åmål	Åmål STP	municipal, car treatment plants, work shops	water	Effluent	10000	mech, chem	2004-09-29	-
	MR-3476	Åmål	Åmål STP	d.o	sludge		10000	mech, chem	2004-09-29	20.2
Öster-götaland	MR-3633	Finnsång	Finnsångs STP		sludge				2004-10-26	5.4
	MR-3635	Finnsång	Finnsångs STP		water				2004-10-27	-
	MR-3683	Finnsång	Skuten		sediment				2004-10-29	7.65
	MR-3728	Linköping	Linköping STP		water				2004-11-09	-
	MR-3729	Linköping	Linköping STP		sludge				2004-11-09	27.6
	MR-3685	Linköping	Roxen		sediment				2004-10-21	12.7

Table A.3. Results from the national programme. Sum D = D4+D5+D6. Sum M = MM+MDM+ MD2M+MD3M. Values below the detection limit are not included in the sum.

Category	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M	
Background		Råb	Air	ng/m ³	78	9	<12	<20	<5	<5	<5	87	-	
		Råb	Air	ng/m ³	35	30	11	<26	<5	<5	<5	76	-	
		Råb	Air	ng/m ³	300	170	77	73	<6	<6	<6	547	73	
		Ö Gotlandsdjupet	Sediment	ng/g dw	<22	<6	<3	<1	<1	<1	<1	<1	-	-
		Ö Öland	Sediment	ng/g dw	<44	<11	<6	<1	<1	<1	<1	<1	-	-
		MR-3752 Norrköpingsdjupet	Sediment	ng/g dw	<14	<4	<2	<1	<1	<1	<1	<1	-	-
		MR-3639 V. Floden	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	<0.5	-	-
		MR-3640 Ängsskärsklubb	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	<0.5	-	-
		MR-3641 Landsort	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	<0.5	-	-
Potential point sources		Stenungsund	Air	ng/m ³	51	37	13	79	<5	<5	<5	101	79	
		Stenungsund	Air	ng/m ³	120	95	27	44	<5	<5	<5	242	44	
		Stenungsund	Air	ng/m ³	230	140	42	37	<6	<6	<6	412	37	
		MR-3872 Stenungsund	Sediment	ng/g dw	<9	<7	<6	<0.1	<0.1	<0.1	<0.1	-	-	
		MR-3874 Stenungsund	Sediment	ng/g dw	<14	<7	<6	<0.2	<0.2	<0.2	<0.2	-	-	
		MR-3876 Stenungsund	Sediment	ng/g dw	<18	<9	<7	<0.2	<0.2	<0.2	<0.2	-	-	
		MR-3878 Stenungsund	Water	µg/L	<0.06	<0.03	0.038	<0.0005	<0.0005	<0.0005	<0.0005	0.038	-	
		MR-3880 Stenungsund	Water	µg/L	<0.06	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.0005	-	-	
		MR-3882 Stenungsund	Water	µg/L	<0.06	<0.03	<0.03	<0.0005	<0.0005	<0.0005	<0.0005	-	-	
		MR-4053 Stenungsund	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
		MR-4054 Stenungsund	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
		MR-4055 Stenungsund	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
		MR-4050 Sundsvallsbukten	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
		MR-4051 Sundsvallsbukten	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
	MR-4052 Sundsvallsbukten	Biota	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-		

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Table A 3. Cont

Category	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M
Potential point sources		Stockvik	Air	ng/m ³	71	19	<12	<19	<3	<3	<3	90	-
		Stockvik	Air	ng/m ³	18	40	38	<19	<3	<3	<3	96	-
	MR-3600	Bay outside Stockvik 1	Sediment	ng/g dw	<11	<6	<5	<0.2	<0.2	<0.2	<0.2	-	-
	MR-3601	Bay outside Stockvik 1	Sediment	ng/g dw	<22	<11	<10	<0.3	<0.3	0.9	1.7	-	2.6
	MR-3603	Bay outside Stockvik 2	Sediment	ng/g dw	<11	<6	<5	<0.2	<0.2	<0.2	<0.3	-	-
	MR-3604	Bay outside Stockvik 2	Sediment	ng/g dw	<12	<6	<6	<0.2	<0.2	<0.2	0.7	-	-
	MR-3612	Akzo-Nobel Stockv. STP	Water	µg/L	<0.06	<0.03	<0.06	0.00063	<0.0005	<0.0005	<0.0005	-	0.0006
	MR-3613	Akzo-Nobel Stockvik	Water	µg/L	<0.06	<0.03	<0.06	<0.0005	<0.0005	<0.0005	<0.0005	-	3
	MR-3610	Bay outside Stockvik	Water	µg/L	<0.06	<0.03	<0.06	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	Diffuse sources		Hudiksvallsgatan 2	Air	ng/m ³	84	54	21	<19	<3	<3	<3	160
		Hudiksvallsgatan 2	Air	ng/m ³	<23	<13	<12	<26	<3	<3	<3	-	-
		Hudiksvallsgatan 2	Air	ng/m ³	97	<13	<12	<26	<3	<3	<3	97	-
MR-3760		Henriksdal STP	Sludge	ng/g DW	280	22000	3100	<2	<2	16	46	25000	62
MR-3790		Gässlösa STP	Sludge	ng/g DW	310	10000	2400	<2	<2	8	24	13000	32
MR-3867		Ryaverket STP	Sludge	ng/g DW	430	19000	2600	<2	<2	13	40	22000	53

Table A 4 Results from regional screening of siloxanes. Sum D = D4+D5+D6. Sum M = MM+MDM+MD2M+MD3M. Values below the detection limit are not included in the sum.

County	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M
Blekinge	MR-3468	Karlshamn STP	Sludge	ng/g DW	150	5000	900	<0.5	<0.5	6.0	6	6100	12
	MR-3462	Mörrum landfyll	Water	µg/L	<0.07	<0.04	<0.04	0.0015	<0.0005	<0.0005	<0.0005	-	0.0015
	MR-3537	Karlskrona STP	Sludge	ng/g DW	300	10000	1200	<0.5	<0.5	7.8	11	12000	19
	MR-3529	Volvo cars STP	Sludge	ng/g DW	<120	<28	<16	<0.5	<0.5	<0.5	3	-	3
	MR-3533	Angeliskogs landfyll	Water	µg/L	<0.07	<0.04	<0.04	0.058	0.00085	<0.0005	<0.0005	-	0.059
Dalarna	MR-3538	Angeliskogs landfyll, Water Angleån	Water	µg/L	<0.07	<0.04	0.076	<0.0005	<0.0005	<0.0005	<0.0005	0.08	-
	MR-3531	Ronneby STP	Sludge	ng/g DW	<250	7600	1100	<0.5	<0.5	6.8	8	8700	15
	MR-3487	Sölvesborg STP	Sludge	ng/g DW	230	7800	930	<1	2.8	7.3	<4	8900	10
	MR-3498	Lake Bäsingen	Sediment	ng/g dw	<19	<9.3	<7.2	<0.2	<0.2	<0.2	<0.2	-	-
	MR-3501	Lake Bäsingen	Fish	ng/g WW	<5	<5	<5	<1	<1	1.4	8	5800	10
Gävleborg	MR-3478	Krylbo STP	Sludge	ng/g DW	130	5300	430	<0.5	1.4	10	<3	63000	12
	MR-3540	Fagersta STP	Sludge	ng/g DW	590	54000	8400	<0.5	<0.5	5.7	14	7300	20
	MR-3535	Venjan STP	Sludge	ng/g DW	<470	6500	740	<0.5	<0.5	<0.48	<0.48	-	-
	MR-3496	Lake Venjan	Sediment	ng/g dw	<47	<23	<18	<0.48	<0.48	<0.48	<0.48	-	-
	MR-3500	Lake Venjan	Fish	ng/g WW	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
Jämtland	MR-3584	Bollnäs STP	Sludge	ng/g DW	<280	6500	950	<0.5	6.4	37	120	7500	163
	MR-3549	Duvbacken STP	Sludge	ng/g DW	200	10000	1300	<0.5	<0.5	4.0	<4	12000	4
	MR-3846	Resselvans STP	Sludge	ng/g DW	490	19000	2000	<0.5	3.51	6.9	19	21000	29
	MR-3555	Sandviken STP	Sludge	ng/g DW	350	11000	1600	<2	<0.5	3.3	<4	13000	3
	MR-3687	Myrviken STP	Sludge	ng/g DW	<150	2300	320	<0.5	<0.5	2.9	2	2600	5
Västmanland	MR-3619	Bräcke STP	Sludge	ng/g DW	430	23000	2500	<0.5	6.9	7.0	26	26000	40
	MR-3470	Björnsrike STP	Sludge	ng/g DW	<78	54	37	<0.5	<0.5	<0.5	<0.5	91	<2
	MR-3574	Hissmofors STP	Sludge	ng/g DW	370	14000	1700	<0.5	<0.5	4.0	11	16000	15
	MR-3586	Ragunda STP	Sludge	ng/g DW	210	3100	350	<0.5	<0.5	<0.5	2	3700	2
	MR-3625	Strömsund STP	Sludge	ng/g DW	260	12000	1200	<0.5	5.0	6.6	10	13000	21
Västergötland	MR-3469	Åre STP	Sludge	ng/g DW	310	6500	1600	<0.5	<0.5	7.2	56	8500	63
	MR-3472	Östersund STP	Sludge	ng/g DW	<140	10000	2100	<0.5	<0.5	7.7	20	12000	27

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Table A 4. Cont

County	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M
Jönköping	MR-3733	Gislaveds STP	Sludge	ng/g DW	<270	7200	1100	<0.5	<0.5	3.0	14	8300	17
	MR-3735	Gislaveds STP	Water	µg/L	<0.07	1.1	0.27	<0.0005	<0.0005	<0.0005	<0.0005	1.37	-
	MR-3731	Gislaveds STP	Water	µg/L	<0.07	0.051	0.23	<0.0005	<0.0005	<0.0005	<0.0005	0.28	-
	MR-3803	Gröpplebacken	Sediment	ng/g DW	<16	17.2	<6.1	<0.16	<0.16	<0.16	<0.16	17.2	-
	MR-3802	Landsbro STP	Sludge	ng/g DW	<680	13000	1400	<0.5	<0.5	<0.6	15	15000	15
	MR-3807	Landsbro STP	Water	µg/L	<0.06	<0.03	0.055	<0.0005	<0.0005	<0.0005	<0.0005	0.055	-
	MR-3808	Landsbro STP	Water	µg/L	<0.06	<0.03	0.046	<0.0005	<0.0005	<0.0005	<0.0005	0.046	-
	MR-3800	Hullingen	Sediment	ng/g DW	<44	<22	<17	<0.5	<0.5	<0.5	<0.5	-	-
Kalmar	MR-3799	Hultsfred STP	Sludge	ng/g DW	<210	5400	790	<0.5	<0.5	4.1	12	6200	16
	MR-3809	Hultsfred STP	Water	µg/L	<0.07	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3810	Hultsfred STP	Water	µg/L	<0.08	0.21	0.069	<0.0005	<0.0005	<0.0005	<0.0005	0.28	-
	MR-3798	Virserum	Sediment	ng/g dw	<6.9	<3.4	<2.6	<0.1	<0.1	<0.1	<0.1	-	-
	MR-3805	Virserum STP	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3797	Virserum STP	Sludge	ng/g dw	380	5900	1200	<0.5	<0.5	12	9	7500	21
	MR-3804	Virserum STP	Water	µg/L	<0.08	0.099	0.076	<0.0005	<0.0005	<0.0005	<0.0005	0.18	-
	MR-3627	Mouth of Emån	Sediment	ng/g dw	<12	<6	<5	<0.1	<0.1	<0.1	<0.1	-	-
Skåne	MR-3466	Bromölla STP	Sludge	ng/g dw	190	14000	1900	<1	2.6	20	23	16000	45
	MR-3679	Ivösjön	Sediment	ng/g dw	<60	<29	<23	<0.6	<0.6	<0.6	<0.6	-	-
	MR-3682	Ivösjön	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3459/60	Öresunds STP	Sludge	ng/g dw	450	12000	1200	<1	<1	5.8	20	13000	26
	MR-3706	Helsingborg	Sediment	ng/g dw	<16	<8	<6	<0.2	<0.2	<0.2	<0.2	-	-
	MR-3708	Helsingborg	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3677	Hammarsjön	Sediment	ng/g dw	<20	<10	<8	<0.2	<0.2	<0.2	<0.2	-	-
	MR-3681	Hammarsjön	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3514	Kristianstad STP	Sludge	ng/g dw	2300	21000	2500	5	22	27	38	26000	92
	MR-3489	Lundåkrav. STP	Sludge	ng/g dw	270	6700	770	<1	<1	8.2	12	7700	20
	MR-3464	Källby STP	Sludge	ng/g dw	130	9300	2100	<1	<1	5.8	20	12000	26
	MR-3456/57	Sjölanda STP	Sludge	ng/g dw	300	9800	1500	<1	<1	6.7	15	12000	21
MR-3623	Perstorp STP	Sludge	ng/g dw	<220	12000	1600	<0.5	<0.02	3.7	10	14000	14	
MR-3670	Storarydsdammen	Sediment	ng/g dw	<28	<14	<10	<0.3	<0.3	<0.3	<0.3	-	-	
MR-3672	Storarydsdammen	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-	
MR-3453/54	Ystad STP	Sludge	ng/g dw	330	6800	420	<2	<3	5.2	22	7500	27	

Table A 4. Cont

County	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M
Stockholm	MR-3480	Himmerfjärdsverket	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3482/84/86	Himmerfjärdsverket	Sludge	ng/g dw	140	6000	940	<1	<1	<1	8	7100	8
	MR-3449	Himmerfjärden	Sediment	ng/g dw	<23	190	51	<0.2	<0.2	1.2	0.4	240	1.6
	MR-3450	Himmerfjärden	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3448	St Envättern	Sediment	ng/g dw	<115	<57	<44	<1.2	<1.2	<1.2	<1.2	-	-
	MR-3451	St Envättern	Fish	ng/g ww	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3551	Fien STP	Sludge	ng/g dw	<130	570	170	<2	<0.5	<1	<3	740	0
	MR-3576	Gnesta STP	Sludge	ng/g dw	1100	10000	1500	<0.5	3.4	3.1	24	13000	30
	MR-3568	Eskilstuna STP	Sludge	ng/g dw	280	13000	2000	<0.5	<0.5	6.6	<4	15000	7
	MR-3578	Katrineholm STP	Sludge	ng/g dw	460	23000	6200	<0.5	<0.5	16	160	30000	181
Södermanland	MR-3570	Nyköping STP	Sludge	ng/g dw	490	15000	2100	<0.5	4	17	37	17000	58
	MR-3637	Oxelösund STP	Sludge	ng/g dw	180	5800	680	<0.5	<0.5	4.3	5	6600	9
	MR-3572	Strängnäs STP	Sludge	ng/g dw	<260	9900	1200	<2	0.00	7.0	97	11000	104
	MR-3553	Vagnhärad STP	Sludge	ng/g dw	370	17000	1500	<2	<0.5	12	<3	18000	12
	MR-3557	Vingåker STP	Sludge	ng/g dw	410	13000	2100	<0.5	12	13	21	16000	45
	MR-3709	Vik STP	Sludge	ng/g DW	<300	5500	570	<0.5	<0.5	19	44	6000	63
	MR-3711	Vik STP	Water	µg/L	<0.06	<0.04	<0.04	0.0098	<0.0005	<0.0005	<0.0005	0.0098	-
	MR-3502	Vänern, Åsffjorden	Sediment	ng/g DW	<28	37	196	<0.3	<0.3	<0.3	0.5	230	0.5
	MR-3506	Vänern, Åsffjorden	Fish	ng/g WW	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
	MR-3507	Vänern, Kattfjorden	Fish	ng/g WW	<5	<5	<5	<0.4	<0.3	<0.4	<0.5	-	-
Värmland	MR-3504	Vänern, Kattfjorden	Sediment	ng/g DW	<23	<11	<8.8	<0.2	<0.2	<0.2	1.3	-	1.3
	MR-3580	Skåre STP	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3582	Skåre STP	Sludge	ng/g DW	340	19000	2000	<0.5	<0.5	8.3	16	21000	25
	MR-3629	Fiskartorpet STP	Sludge	ng/g DW	130	3400	400	<2	<3	<4	6	4000	6
	MR-3631	Fiskartorpet STP	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3754	Tivoliverket	Sludge	ng/g DW	<270	9700	1300	<0.5	<0.5	4.1	14	11000	18
	MR-3615	SCA Östrand	Sediment	ng/g DW	<16	<7.8	<6.0	<0.2	<0.2	<0.2	<0.2	-	-
	MR-3756	SCA Östrand	Water	µg/L	<0.07	0.059	0.15	<0.0005	<0.0005	<0.0005	<0.0005	0.21	-
	MR-3726	Bodum STP	Sludge	ng/g DW	180	7400	1100	<0.5	<0.5	5.5	8	8700	13

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Table A 4. Cont

County	Sample ID	Site	Matrix	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M	Sum D	Sum M
Västra Götaland	MR-3508	Lidköping STP	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3510	Lidköping STP	Sludge	ng/g DW	1000	22000	1500	8.2	<1	20	23	25000	52
	MR-3512	Vara STP	Sludge	ng/g DW	350	6800	1100	2.2	9.0	8.8	11	8200	31
	MR-3474	Åmål STP	Water	µg/L	<0.06	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3476	Åmål STP	Sludge	ng/g DW	<190	9300	1700	<0.5	<0.5	4.5	9	11000	13
Östergötland	MR-3633	Finspångs STP	Sludge	ng/g DW	<840	9100	1100	<0.5	<0.5	<0.5	<0.5	10000	-
	MR-3635	Finspångs STP	Water	µg/L	<0.07	<0.04	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3683	Skuten	Sediment	ng/g DW	<28	26	<11	<0.3	<0.3	<0.3	<0.3	26	-
	MR-3728	Linköping STP	Water	µg/L	<0.06	<0.03	<0.04	<0.0005	<0.0005	<0.0005	<0.0005	-	-
	MR-3729	Linköping STP	Sludge	ng/g DW	220	10000	1400	<0.5	<0.5	13	19	12000	32
MR-3685	Roxen	Sediment	ng/g DW	<45	<22	<17	<0.5	<0.5	<0.5	<0.5	-	-	

Table A5 Concentration of siloxanes in breast milk

Sample nr	Unit	D4	D5	D6	MM	MDM	MD2M	MD3M
MR3007	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3008	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3009	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3010	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3011	µg/L	<2	3.6	<2	<0.004	0.007	0.013	<0.04
MR3012	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3013	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3015	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3016	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3017	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3018	µg/L	<2	<2	2.9	<0.004	0.003	<0.005	<0.04
MR3019	µg/L	<2	<2	2.9	<0.004	0.003	<0.005	<0.04
MR3020	µg/L	<2	<2	<2	<0.004	0.005	<0.005	<0.04
MR3021	µg/L	<2	2.1	<2	<0.004	<0.002	<0.005	<0.04
MR3023	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3024	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3025	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3026	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3027	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3028	µg/L	<2	<2	2.5	0.005	<0.002	<0.005	<0.04
MR3029	µg/L	<2	3.2	<2	<0.004	<0.002	<0.005	<0.04
MR3030	µg/L	<2	<2	<2	<0.004	0.006	0.006	<0.04
MR3031	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3032	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3181	µg/L	3.5	4.5	4.8	<0.004	<0.002	<0.005	<0.04
MR3280	µg/L	<2	4.2	<2	<0.004	<0.002	<0.005	<0.04
MR3281	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3282	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3283	µg/L	<2	<2	<2	0.006	<0.002	<0.005	<0.04
MR3284	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3285	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3286	µg/L	10	3.4	<2	<0.004	<0.002	<0.005	<0.04
MR3287	µg/L	<2	<2	<2	<0.004	0.003	<0.005	<0.04
MR3288	µg/L	<2	2.4	<2	<0.004	<0.002	<0.005	<0.04
MR3289	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3290	µg/L	2.9	2.8	<2	<0.004	<0.002	<0.005	<0.04
MR3291	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3292	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04
MR3293	µg/L	<2	<2	<2	<0.004	<0.002	<0.005	<0.04