REPORT

# Results from the Swedish National Screening Programme 2004

## Subreport 4: Siloxanes

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B1643
October 2005

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**Report Summary**

**Project title**

**Project sponsor**


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**Title and subtitle of the report**

Results from the Swedish National Screening Programme 2004. Subreport 4: Siloxanes


**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/m$^3$. 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.



**Keyword**
Screening siloxanes D4 D5 D6 MM MDM MD2M MD3M

**Bibliographic data**
IVL Report B1643

**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

# Sammanfattning

IVL Svenska Miljöinstitutet har på uppdrag av Naturvårdsverket utfört en screeningstudie av siloxaner. Studien omfattade tre cykliska polydimetyslioxaner (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 ämnenas 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 µ/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 $ng/m^3$. 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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# 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

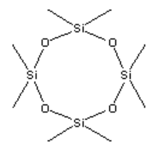
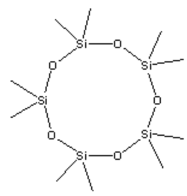
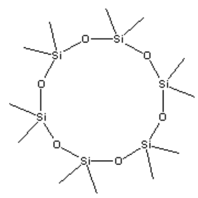
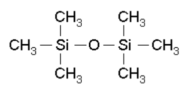
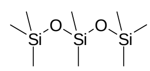
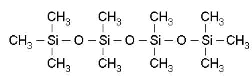
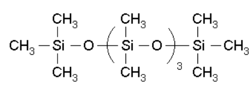
The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden. A further aim was to investigate the likelihood of atmospheric transport 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** | $W_{sol}$ (mg/l) | $V_p$ (mm Hg) | H (Atm $m^3$/mol) | Log $K_{ow}$ | **BCF** | $K_{oc}$(L/kg) |
|---|---|---|---|---|---|---|---|
| D4[a] | 296.6 | 0.9 (25°) | 1 (25°) | 0.42 | - | 12400 | $2.85\times10^4$ |
| 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] | $1.6\times10^{4}$[b] |
| D6 | 444.9 | | | | | | |
| MM[a] | 162.4 | 2 (25°) | 42 (25°) | 4.5 | 4.2 | 5000 | $4.6\times10^3$ |
| MDM | 236.5 | | | | | | |
| MD2M | 310.7 | | | | | | |
| MD3M[c] | 384.8 | $3.1\times10^{-4}$ | 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 ( Americamysis bahia) | NOEC | IMBL | 14 d | 9.1 |
| Midge (Chironomus tentans) | NOEC | GRO | 14 d | >15 |
| Midge (Chironomus tentans) | NOEC | MOR | 14 d | >15 |
| Sheepshead minnow (Cyprinodon variegatus) | NOEC | MOR | 14 d | 6.3 |
| Water flea (Daphnia magna) | NOEC | IMBL | 48 h | >15 |
| Water flea (Daphnia magna) | NOEC | REP | 21 d | 1.7 - 15 |
| Rainbow trout, donaldson trout (Oncorhynchus mykiss) | LC50 | MOR | 14 d | 10, 8.5 - 13 |
| Rainbow trout, donaldson trout (Oncorhynchus mykiss) | LOEC | MOR | 14 d | 6.9 |
| Rainbow trout, donaldson trout (Oncorhynchus mykiss) | NOEC | MOR | 14 d | <=4.4 |
| Rainbow trout, donaldson trout (Oncorhynchus mykiss) | NOEC | NOC, MULT | 93 d | 4.4 |
| Water flea (Daphnia magna) | | 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 carcinogenity 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.

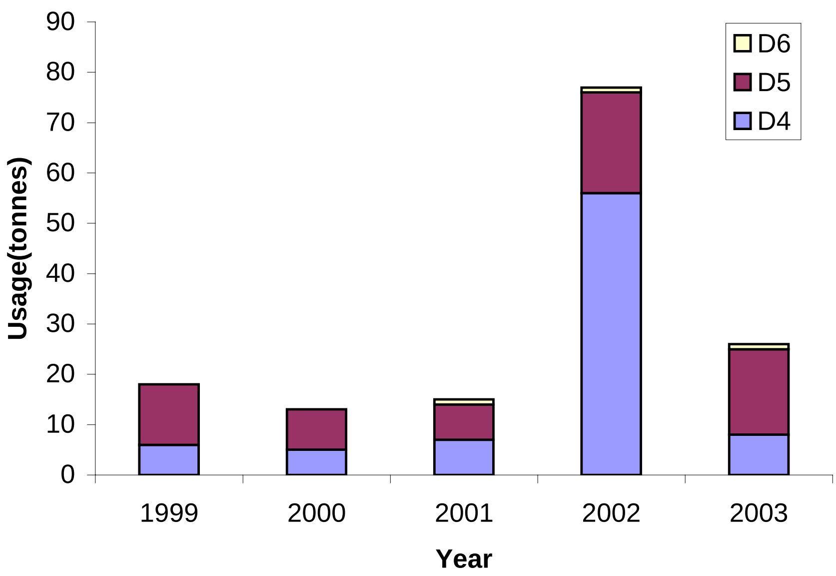



Figure 1. Swedish use of D4, D5, and D6 (SPIN, 2005)

# 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 (µg/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 µg/l. |
| Surface water near industrial area | Japan, 1997 | 2.8 –54.2 µg/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 (µg/m$^3$) | Min (µg/m$^3$) | Max (µg/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 phtalates. 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 $K_2HPO_4$, 0.4M HCl, 80g $Na_2EDTA$ $2H_2O$ 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/µL and about 30 ng/µL 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×0.2mm×0.11µm 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.

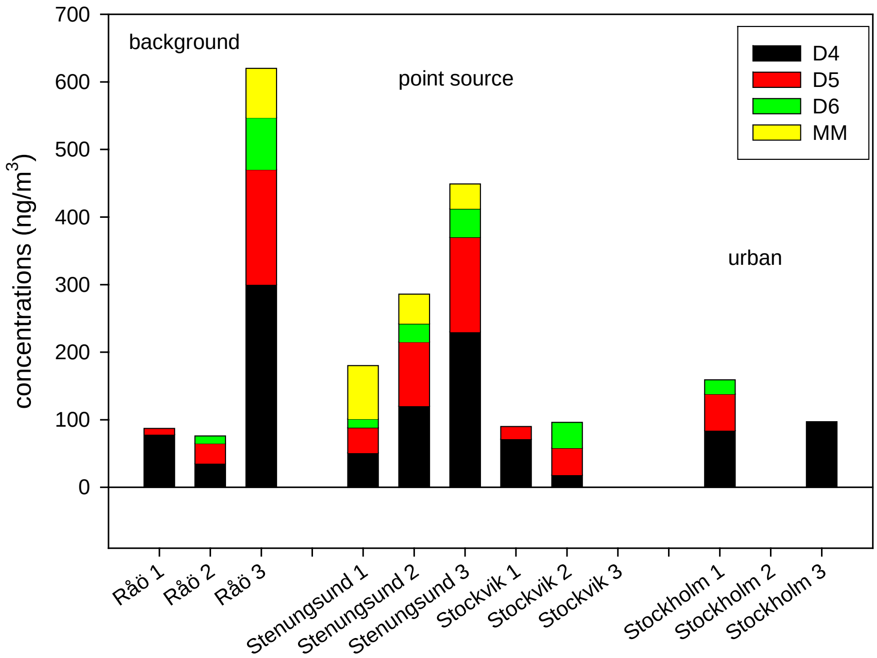



Figure 2. Measured concentrations of siloxanes in air, within the national programme.

### 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.

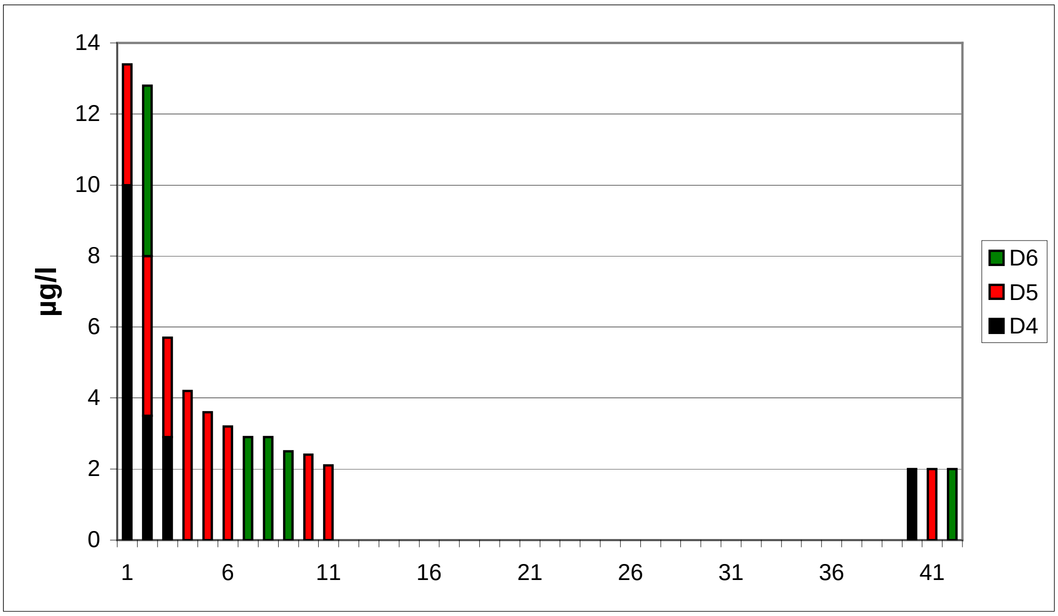



Figure 3. Concentrations of D4, D5 and D6 (µg/l) in breast milk. The samples are sorted in decreasing order of concentration. Results for sample 12-39 are below the quantification limit. The bars to the right illustrate the quantification limits.

## 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.

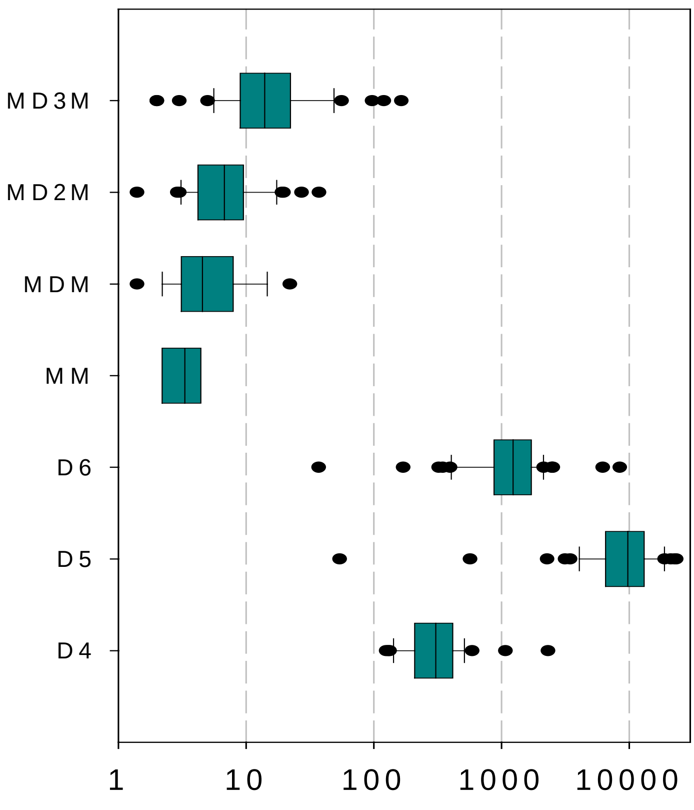



Figure 4. Concentrations of siloxanes (ng/g DW, logaritmic scale) in sludge from all municipal STPs. The lower and upper boundaries of the box represent the 25- and 75-percentiles, the line within the box is the median concentration. The whiskers represent the 10- and 90-percentiles, and the dots are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

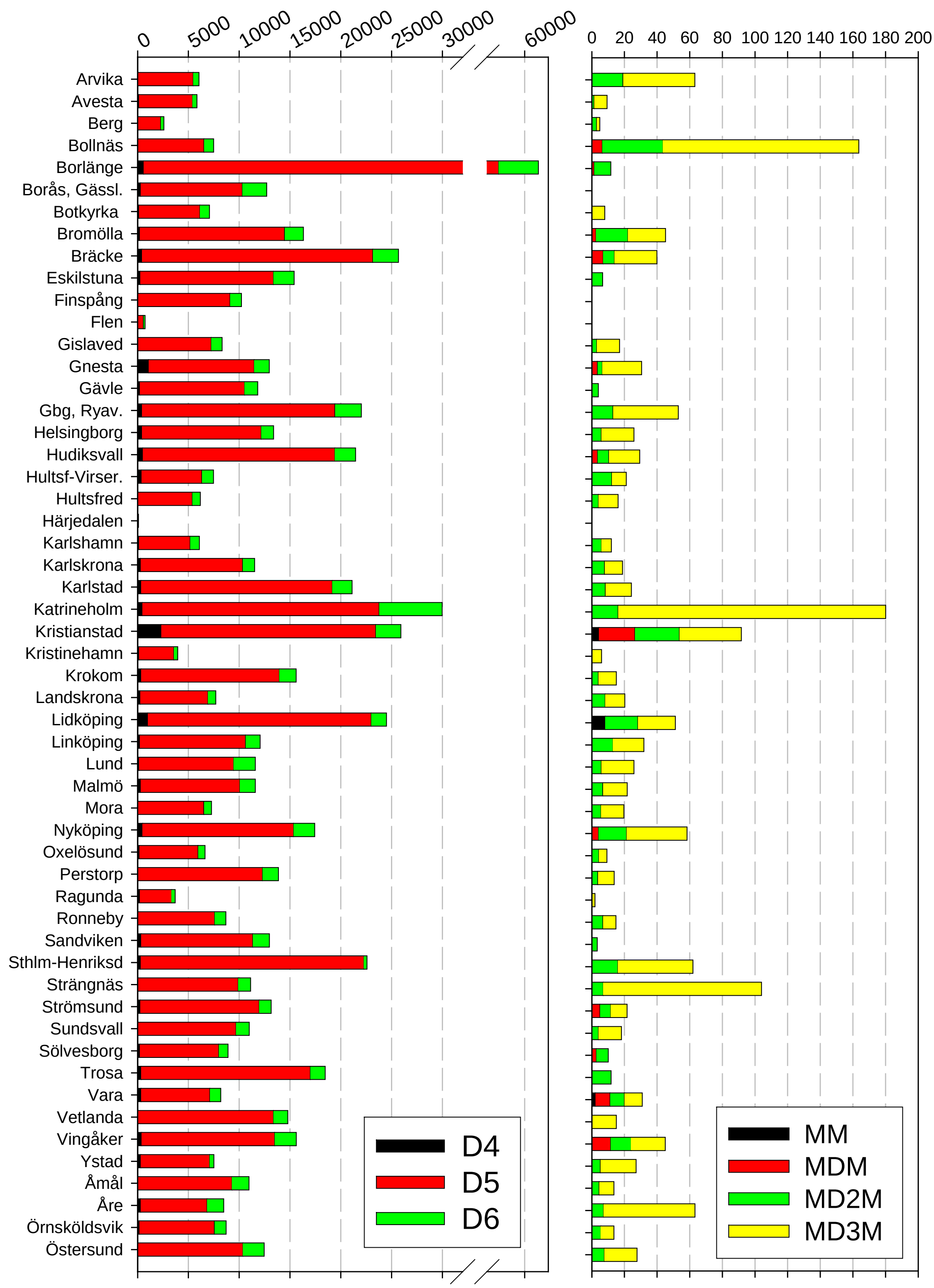



Figure 5. Concentrations of siloxanes in sludge from municipal STPs, ng/g DW. Please note the different concentration scales in the left and right parts of the figure.

### 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 occured 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/$m^3$. 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

Thanks to all staff at the county administrative boards and different municipalities that have contributed to the sampling. Especially thanks to Anders Bignert, NRM, and staff at potential point source industry for contributing samples to the national programme. The study was financed by Environmental Monitoring at Swedish Environmental Protection Agency.

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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** | | | Råö | Air | | 2004-11-16 | | | - |
| | | | Råö | Air | | 2004-11-23 | | | - |
| | | | Råö | 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 07i-001 | | 6435399 | 1625504 | 6.6 |
| | MR-3639 | | V.Fladen | biota | herring | 2003-09-16 | 57° 04' 47'' | 15 °55' 18'' | 1.7 |
| | MR-3640 | | Ängsskärsklubb | biota | baltic herring | 2003-10-01 | 65*43' | 23°*03' | 2.3 |
| | MR-3641 | | Landsort | biota | baltic herring | 2003-11-12 | 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 | biota | baltic herring | | | | - |
| | MR-4051 | Sundsvall | Sundsvall bay | biota | herring | | | | - |
| | MR-4052 | Sundsvall | Sundsvall bay | biota | salmon | | | | - |

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. Expancel TP | water | effluent | 2004-10-22 | | | - |
| | MR-3613 | Sundsvall | Akzo-Nobel Stockvik | water | water from well on factory area | 2004-10-22 | | | - |
| | MR-3610 | Sundsvall | Bay outside Stockvik | water | recipient | 2004-10-21 | 6914691 | 1581178 | - |
| **Diffuse sources** | | Stockholm | Hudiksvallsgatan 2 | Air | | 2004-11-18 | | | - |
| | | Stockholm | Hudiksvallsgatan 2 | Air | | 2004-12-02 | | | - |
| | | Stockholm | Hudiksvallsgatan 2 | Air | | 2004-12-03 | | | - |
| | MR-3760 | Stockholm | Henriksdal STP | sludge | 700.000 pe, mech, chem,bio cleaning. No major industrial impact | 2004-11-16 | | | 28.6 |
| | MR-3790 | Borås | Gässlösa STP | sludge | p.e 98250, mec, chem and bio cleaning, textile-, graphic- industries, slaughterhouse, laundry, finsihing activities, landfill leachate, incineration | 2004-11-24 | | | 22.4 |
| | MR-3867 | Gothenburg | Ryaverket STP | sludge | p.e 620 683, mech, chem, bio cleaning, incineration, indstrial and municipal | 2004-12-15 | | | 28.8 |

**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 | pe (STP) | Treatment (STP) | Sampling date | DW/LW (%) |
|---|---|---|---|---|---|---|---|---|---|---|
| Blekinge | MR-3462 | Karlshamn | Mörrum landfill | | water | leachate | | | 2004-09-23 | - |
| | MR-3468 | Karlshamn | Mörrum 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 |
| Dalarna | MR-3498 | Avesta | Bäsingen | | sediment | lake | | | 2004-10-04 | 21.4 |
| | MR-3501 | Avesta | Bäsingen | | fish | | | | 2004-09-29 | - |
| | 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 | - |
| Gävleborg | 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 |
| Jämtland | MR-3687 | Berg | Myrviken STP | Mun. | sludge | Primary | | mech, dehydration, compost | 2004-11-01 | 19.2 |
| | 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 | Hissmofors STP | Mun., ind. | sludge | Primary | 3600 | mech, chem, bio | 2004-10-19 | 13.4 |
| | MR-3586 | Ragunda | Överammer STP | Mun. | sludge | Primary | | Dehydration | 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 | Göviken 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 (%) |
|---|---|---|---|---|---|---|---|---|---|---|
| Jönköping | MR- 3733 | Gislaved | Gislaveds STP | | sludge | | | | | 16.5 |
| | MR- 3735 | Gislaved | Gislaveds STP | | water | effluent | | | | - |
| | MR-3731 | Gislaved | Gislaveds STP | | water | influent | | | | - |
| | 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 | - |
| Kalmar | MR-3800 | Hultsfred | Hulingen | | sediment | | | | 2004-11-24 | 34.4 |
| | MR-3799 | Hultsfred | Hultsfred STP | | sludge | | | | 2004-11-24 | 18.8 |
| | 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 | | | 2004-10-27 | 22.7 |
| Skåne | 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 |
| | 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 | pe (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 |
| Stock-holm | 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 | - |
| Söder-manland | MR-3551 | Flen | Flen STP | GB | sludge | Primary | 10200 | mech, chem,biol | 2004-10-11 | 27.8 |
| | 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,bio. 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 |
| Värmland | 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 | - |
| | MR-3502 | Grums | Vänern, Åsfjorden | Pulp and paperind., viscous manufacturing | sediment | | | | 2004-09-30 | 18.9 |
| | MR-3506 | Grums | Vänern, Åsfjorden | 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 | | | | | - |
| | MR-3726 | Örnsköldsvik | Bodum STP | | sludge | | | | 2004-11-10 | 31 |
| Västra Götaland | 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-götland | MR-3633 | Finspång | Finspångs STP | | sludge | | | | 2004-10-26 | 5.4 |
| | MR-3635 | Finspång | Finspångs STP | | water | | | | 2004-10-27 | - |
| | MR-3683 | Finspång | 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* |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Råö | Air | ng/m$^3$ | 78 | 9 | <12 | <20 | <5 | <5 | <5 | ***87*** | - |
| | | Råö | Air | ng/m$^3$ | 35 | 30 | 11 | <26 | <5 | <5 | <5 | ***76*** | - |
| | | Råö | Air | ng/m$^3$ | 300 | 170 | 77 | 73 | <6 | <6 | <6 | ***547*** | ***73*** |
| | MR-3750 | Ö Gotlandsdjupet | Sediment | ng/g dw | <22 | <6 | <3 | <1 | <1 | <1 | <1 | - | - |
| **Background** | MR-3751 | Ö Öland | Sediment | ng/g dw | <44 | <11 | <6 | <1 | <1 | <1 | <1 | - | - |
| | MR-3752 | Norrköpingsdjupet | Sediment | ng/g dw | <14 | <4 | <2 | <1 | <1 | <1 | <1 | - | - |
| | MR-3639 | V.Fladen | Biota | ng/g ww | <5 | <5 | <5 | <0.4 | <0.3 | <0.4 | <0.5 | - | - |
| | MR-3640 | Ängsskärsklubb | Biota | ng/g ww | <5 | <5 | <5 | <0.4 | <0.3 | <0.4 | <0.5 | - | - |
| | MR-3641 | Landsort | Biota | ng/g ww | <5 | <5 | <5 | <0.4 | <0.3 | <0.4 | <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 | - | - |

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/m³ | 71 | 19 | <12 | <19 | <3 | <3 | <3 | ***90*** | - |
| | | Stockvik | Air | ng/m³ | 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 | - | ***0.00063*** |
| | MR-3613 | Akzo-Nobel Stockvik | Water | µg/L | <0.06 | <0.03 | <0.06 | <0.0005 | <0.0005 | <0.0005 | <0.0005 | - | - |
| | 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/m³ | 84 | 54 | 21 | <19 | <3 | <3 | <3 | ***160*** | - |
| | | Hudiksvallsgatan 2 | Air | ng/m³ | <23 | <13 | <12 | <26 | <3 | <3 | <3 | - | - |
| | | Hudiksvallsgatan 2 | Air | ng/m³ | 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 landfill | 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 | Angelskogs landfill | Water | µg/L | <0.07 | <0.04 | <0.04 | 0.058 | 0.00085 | <0.0005 | <0.0005 | ***-*** | ***0.059*** |
| | MR-3538 | Angelskogs landfill, 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*** |
| Dalarna | 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 | | | | | | ***-*** |
| | MR-3478 | Krylbo STP | Sludge | ng/g DW | 130 | 5300 | 430 | <1 | <1 | 1.4 | 8 | ***5800*** | ***10*** |
| | MR-3540 | Fagersta STP | Sludge | ng/g DW | 590 | 54000 | 8400 | <0.5 | 1.4 | 10 | <3 | ***63000*** | ***12*** |
| | MR-3535 | Venjan STP | Sludge | ng/g DW | <470 | 6500 | 740 | <0.5 | <0.5 | 5.7 | 14 | ***7300*** | ***20*** |
| | 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 | | ***-*** |
| Gävleborg | 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*** |
| Jämtland | MR-3687 | Myrviken STP | Sludge | ng/g DW | <150 | 2300 | 320 | <0.5 | <0.5 | 2.9 | 2 | ***2600*** | ***5*** |
| | MR-3619 | Bräcke STP | Sludge | ng/g DW | 430 | 23000 | 2500 | <0.5 | 6.9 | 7.0 | 26 | ***26000*** | ***40*** |
| | MR-3470 | Björnrike 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*** |
| | 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*** |

Table A 4. Cont

| County | Sample ID | Site | Matrix | Unit | D4 | D5 | D6 | MM | MDM | MD2M | MD3M | Sum D | Sum M |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 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*** | - |
| Jönköping | MR- 3803 | Gröpplebäcken | 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 | Hulingen | Sediment | ng/g DW | <44 | <22 | <17 | <0.5 | <0.5 | <0.5 | <0.5 | - | - |
| | 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 | - | - |
| Kalmar | 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 | - | - |
| | 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 | - | - |
| Skåne | 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 | Sjölunda 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 | - | - |
| Söder-manland | MR-3551 | Flen 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*** |
| | 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*** |
| Värmland | 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, Åsfjorden | Sediment | ng/g DW | <28 | 37 | 196 | <0.3 | <0.3 | <0.3 | 0.5 | ***230*** | ***0.5*** |
| | MR-3506 | Vänern, Åsfjorden | 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 | - | - |
| | 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 | - | - |
| Västernorrland | 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*** |

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 | 0.008 | 0.008 | <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 |