

## REPORT

# Screening of organotin compounds in the Swedish environment

30 June 2006

By: John Sternbeck, Jenny Fäldt and Ann Helén Österås

Revised by: Jenny Fäldt



## REPORT

# Screening of organotin compounds in the Swedish environment

30 June 2006

### Client

Naturvårdsverket  
106 48 Stockholm  
Sweden

Contact person: Britta Hedlund, Jonas Rodhe  
Contract: 219 0503; 219 0604

### Consultant

WSP Environmental  
SE-121 88 Stockholm-Globen  
Visitors: Arenavägen 7  
Phone: +46 8 688 60 00  
Fax: +46 8 688 69 22  
WSP Sverige AB  
Corporate identity no.: 556057-4880  
Reg. office: Stockholm  
[www.wspgroup.se](http://www.wspgroup.se)

### Contacts

Project leader: John Sternbeck +46-8-688 6319

Consultant      Jenny Fäldt  
                         Ann Helén Österås  
                         Johan Persson

Representative: Marie Arnér +46-8-688 6403



## Summary

A number of organotin compounds are used in the society, but the knowledge on their environmental impact is mainly restricted to those compounds that are used in antifouling paints on ships. In this screening study, the occurrence of butyltins, octyltins, phenyltins and tricyclohexyltin in the Swedish environment was investigated. The sampling programme was setup based on the use of organotins in non-biocidal applications. There is an emphasis on the lacustrine environment in three urban regions, but the study also covers sewage treatment plants, an agricultural field, a plastics industry, landfills, foodstuffs and breastmilk. A total of 107 samples were analysed. Furthermore, the physical-chemical and toxicological properties of organotins are briefly summarised and their use in the Swedish society is described.

The major goals were to assess

- If diffuse release of organotins occurs in urban areas
- If releases of organotins from municipal sewage treatment plants (STPs) influence the levels in the aquatic environment
- If industrial use may cause a local impact of organotins
- If the use of sewage sludge as a fertiliser causes organotin accumulation in soils
- If human exposure is significant

Because this is a screening study, some of these goals had to be assessed on the basis of few samples, whereas other aspects could be more thoroughly investigated. Urban stormwater sludge from three cities shows that diffuse releases of monobutyltin, dibutyltin, tributyltin, monoctyltin and dioctyltin occur in urban areas. This diffuse release is also evident in the aquatic environment, where in particular DBT and MBT are enriched in urban sediments. The relative abundance of organotin compounds is very similar in urban stormwater sludge and sewage sludge, suggesting common sources. Furthermore, the specific load to municipal STPs ( $\mu\text{g}/\text{d}/\text{pe}$ ) is similar to earlier results from households, suggesting that diffuse releases from products in the urban environment is important.

STPs appear to influence the levels of MBT in their recipients moderately, but no other organotin compound is released in significant quantities by these STPs. The concentrations in stormwater sludge and soil close to a PVC industry suggest that emissions may occur from the current industrial use of butyltins and octyltins.

Fish from various lakes and the Baltic Sea contain butyltins and phenyltins. When considering the occurrence of these substances in sediments, and their present use, it appears that phenyltins are more strongly accumulated than butyltins in fish. Perch from Mälaren, Stockholm city and the Stockholm archipelago contains higher levels of dibutyltin, tributyltin and triphenyltin than fish from other lakes. The levels in Stockholm city is particularly high.

In conclusion, there was a good correspondence between the type of organotin compounds found in the present study and those that are used in the society. Phenyltins mainly occurred in the aquatic environment in areas where large ships travel. Octyltins were found in waste waters and sludge, in urban stormwaters and in certain urban sediments, suggesting an association to products containing octyltins. Butyltins were found in all these environments, in agreement with their use both in the marine environment and in urban areas.

## Sammanfattning

Många tennorganiska ämnen används i samhället, men kunskap om deras eventuella miljöpåverkan är huvudsakligen begränsad till de ämnen som används i båtbottnfärg. I denna screeningundersökning har butyltenn-, fenyltenn- och oktyltennföreningar samt tricyklohexyltenn undersökts med avseende på förekomst i miljön och human exponering. Ett mätprogram upprättades utifrån en översiktlig kartläggning av hur dessa tennorganiska ämnen används i samhället. Mätprogrammet har en viss tyngdpunkt mot urbana miljöer, men omfattar även kommunala reningsverk, en slambehandlad åkermark, en plastindustri, avfallsdeponier, livsmedel och bröstmjök. Totalt analyserades 107 prov. Dessutom är ämnenas fysikalisk-kemiska och ekotoxikologiska egenskaper kortfattat beskrivna.

Undersökningens primära mål var att bedöma

- Om diffus spridning sker i urban miljö
- Om utsläpp från kommunala reningsverk påverkar halterna av tennorganiska ämnen i vattenmiljön
- Om tennorganiska ämnen kan spridas från industriell användning
- Om användning av rötslam på åkermark leder till en ackumulation av tennorganiska ämnen i jorden
- I vilken grad human exponering sker

Eftersom detta är en screeningundersökning fick flera av dessa frågor bedömas preliminärt utifrån relativt få prov, medan andra aspekter kunde belysas i mer detalj.

Dagvattenslam från tre tätorter visar att monobutyltenn, dibutyltenn, tributyltenn, monooktyltenn och dioktyltenn sprids diffust i urban miljö. Dessa diffusa utsläpp syns i vissa fall även i den akvatiska miljön, där särskilt monobutyltenn och dibutyltenn var anrikade i sedimenten. De relativa halterna av dessa ämnen var mycket likartade i dagvattenslam och i slam från reningsverk, vilket indikerar ett gemensamt ursprung. Dessutom var den specifika belastningen till reningsverken i samma nivå som tidigare mätningar påvisat från hushållsavloppsvatten, vilket indikerar att diffus spridning från varor är en betydelsefull spridningsprocess i tätorter.

Reningsverk förefaller kunna orsaka en marginell haltförhöjning av monobutyltenn i recipienterna, medan halterna av övriga tennföreningar är låga i utgående avloppsvatten. I närområdet till en plastindustri förekom butyltenn och oktyltenn i höga halter i dagvattenslam, och ämnena påträffades även i jord.

Fisk från olika sjöar samt från Östersjön innehöll butyltenn- och fenyltennföreningar. I jämförelse med dessa ämnens förekomst i sediment, samt i vilken utsträckning de används i samhället, framgår det att fenyltennföreningar bioackumuleras i avsevärt högre grad än butyltennföreningar. Dibutyltenn, tributyltenn och trifenyltenn förekom i betydligt högre halter i abborre från Mälaren, centrala Stockholm och Stockholms skärgård, än i fisk från andra lokaler. Halterna i centrala Stockholm var särskilt höga, i likhet med halterna i sediment.

Sammanfattningsvis var det en god överensstämmelse mellan vilka tennorganiska ämnen som påträffas i miljön och vilka som används i samhället. Fenyltennföreningar påträffades främst i akvatisk miljö, nära farleder. Oktyltennföreningar påträffades främst i avloppsvatten, rötslam, dagvattenslam och sediment, vilket tyder på en koppling till varor innehållande oktyltenn. Butyltennföreningar påträffades i alla miljöer, i överensstämmelse med deras bredare användningsområden.



## Table of Contents

<b>1.</b>	<b>Introduction</b>	<b>6</b>
<b>2.</b>	<b>Properties of organotin compounds</b>	<b>6</b>
2.1.	Physical and chemical properties	6
2.2.	Bioaccumulation and Toxicity	8
<b>3.</b>	<b>Use and release of organotin substances</b>	<b>10</b>
<b>4.</b>	<b>Environmental occurrence</b>	<b>12</b>
4.1.	Sewage treatment plants	12
4.2.	Surface water, storm water etc	14
4.3.	Sediment, biota and foodstuffs	14
<b>5.</b>	<b>Sampling strategy and study areas</b>	<b>16</b>
5.1.	Urban areas	17
5.2.	Sewage treatment plants	20
5.3.	Potential point sources	21
<b>6.</b>	<b>Methods</b>	<b>22</b>
6.1.	Sampling	22
6.2.	Chemical analysis and quality assurance	22
<b>7.</b>	<b>Results</b>	<b>24</b>
<b>8.</b>	<b>Discussion</b>	<b>29</b>
8.1.	Background levels	29
8.2.	Urban areas	30
8.3.	Sewage treatment plants	32
8.3.1.	Occurrence in sludge and waste waters	32
8.3.2.	Sources of organotins to STPs	34
8.3.3.	Impact of STPs on surface waters	35
8.4.	Point sources	35
8.5.	Brief assessment of organotin releases in Sweden	35
8.6.	Occurrence in foods and human exposure	36
8.7.	Risks för health and the environment	37
<b>9.</b>	<b>Conclusions</b>	<b>38</b>
<b>10.</b>	<b>Acknowledgements</b>	<b>38</b>
<b>11.</b>	<b>References</b>	<b>39</b>
	<b>Appendix 1-3</b>	<b>42-53</b>

## List of abbreviations

MBT	Monobutyltin
DBT	Dibutyltin
TBT	Tributyltin
TeBT	Tetrabutyltin
MOT	Monooctyltin
DOT	Diocyltin
TCHT	Tricyclohexyltin
MPT	Monophenyltin
DPT	Diphenyltin
TPT	Triphenyltin
OTCs	Organotin compounds
STP	Sewage treatment plant
pe	Population equivalents



## 1. Introduction

Organotin compounds are characterized by a  $\text{Sn}^{4+}$  ion to which one to four organic ligands are attached. They are classified according to the type of organic ligand and the most common are butyltins, octyltins och phenyltins. A large number of organotin substances are used in the society, and some of these are well-known environmental pollutants. The use of tributyltin (TBT) in antifouling paints on ships has caused significant harm to the marine environment worldwide. Female molluscs are masculinized by TBT at levels as low as ca 1 ng/l, and this effect has severe consequences for their ability to reproduce (e.g. Gies, 2003).

Most investigations on the environmental occurrence of organotin substances have focused on TBT. However, other substances such as dibutyltin and dioctyltin are used in the society for other reasons and are found in other applications. The aim of this study is to broadly investigate the occurrence of butyltin, octyltin and phenyltin compounds in the Swedish environment, and to put this in the perspective of their current use in the society as well as their potential effects on health and environment. Tributyltin is the most well-known and well-studied of these substances, both internationally and in Sweden, why we put some focus on the other substances, and on the non-biocidal uses.

The project is an assignment from the Swedish EPA, and is part of the national environmental screening programme. This study is intended to complement a previous screening of organotin compounds, that was directed at the marine environment (Tesfalidet, 2003).

## 2. Properties of organotin compounds

### 2.1. Physical and chemical properties

The structures of the investigated organotins compounds are shown in Figure 1. Except for tetrabutyltin, all studied organotin compounds are cations. In nature, as well as in industrial chemicals, they are balanced by inorganic or organic anion ligands. Most industrial organotin chemicals are composed of an organotin cation and one or several ligands, and most of these chemicals are reconverted to the organotin cation compounds in natural waters. The cation may form dissolved complexes with e.g. chloride in seawater.

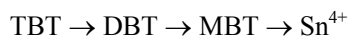
Therefore, their environmental partitioning properties such as  $K_d$  and  $K_H$  depend in part on the balancing anion in the environment. Hydrophobicity increases with increasing number of alkyl groups, and with increasing length of the alkyl chain. Organotins are moderately hydrophobic and associate strongly to particles in natural waters. In harbour sediments,  $\log K_d$  in the range 3-4.3 have been measured for various OTCs, and the particle affinity increased in the order MBT < DBT < TBT (Berg et al., 2001). In various soils, however, the reverse pattern of  $K_d$  was observed (Huang and Matzner, 2004). In organic soils,  $\log K_d$  exceeded 4.0, whereas adsorption was less strong in mineral soils. In contrast to hydrophobic pollutants such as PCBs or PAHs (that partition to lipids in organic matter), OTCs are adsorbed to the functional groups of organic matter, e.g. phenolic and carboxylic groups (Berg et al., 2001; Huang and Matzner, 2004a).

Because organotins are cations, long-range atmospheric transport has generally not been considered as important. It has though been demonstrated that TBT forms highly volatile chloride species in seawater (Mester and Sturgeon, 2002). A recent study actually demonstrated the presence of organotins in air from rural sites, showing that long-range atmospheric transport of butyltins and octyltins do occur (Huang and Klemm, 2004). MBT was the

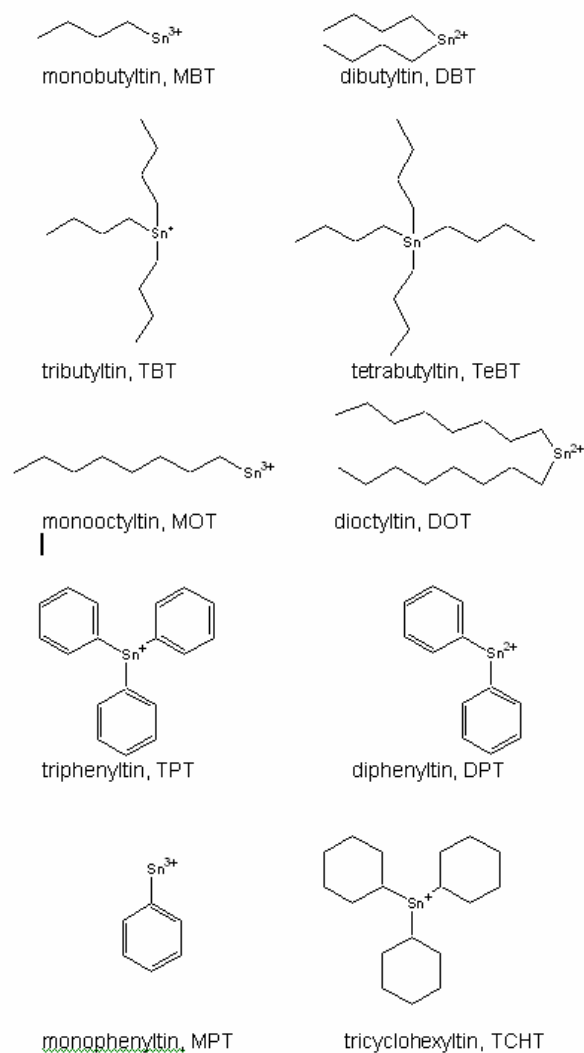


major species in precipitation and deposition. TBT mainly occurred in the gas phase and it is speculated that the source of butyltins may have been volatile TBT species. Subsequent dealkylation in the atmosphere may convert TBT to DBT and MBT.

Organotins are progressively dealkylated in nature, for instance:



Dealkylation proceeds both by photolysis and through enzymatical reactions. This is important to consider when monitoring data are evaluated, since the occurrence of, e.g., DBT may be due to direct release of DBT or to release of TBT that is subsequently dealkylated. Environmental half-lives are summarized in Huang and Matzner (2004b). Half-lives in soils and sediments are commonly one or a few years, but may be longer under reducing conditions, whereas half-lives in natural waters may range from a few days to several weeks.



**Figure 1.** Molecular structure of the investigated organotins, and the abbreviations used in this report.



## 2.2. Bioaccumulation and Toxicity

Organotin compounds have been detected in various marine organisms, from evertebrates to mammals. In fish and marine mammals, TBT and TPT bioaccumulate more strongly in liver than in muscle (e.g., Ciesielski et al., 2004; Hajjaj el Hassani et al., 2005). Bioaccumulation is often stronger in bivalves than in fish, a consequence of lower metabolic capacity in bivalves (WHO, 1999). Trisubstituted OTCs are more strongly bioaccumulated than the less lipophilic disubstituted OTCs. Because TBT is dealkylated in many organisms, DBT may be a major species in biota (e.g. Borghi and Porte, 2002) but not necessarily the organotin species that was assimilated. Most studies do not suggest that TBT is biomagnified in aquatic food-chain. However, TPT appears to be biomagnified fairly strongly in the aquatic food chain (Hu et al., 2006).

The trisubstituted substances, TPT and in particular TBT, are widely held as the most toxic organotin substances. Numerous field studies have demonstrated a direct link between TBT and imposex in certain marine organisms, mainly molluscs (reviewed in e.g. Gies, 2003). Imposex means that females are masculinized and this effect is severe because it directly influences the ability for organisms to reproduce. Imposex has been demonstrated in many coastal areas, and was recently shown to be common in gastropods on the Swedish west coast (Strand et al., 2006). These effects occur at very low levels (ca 1 ng/l) for certain organisms. It has been shown in laboratory that TBT causes masculinization also in fish (Shimasaki et al., 2003).

DBT and MBT does not cause imposex, but both TBT and DBT have negative effects on the reproductive system in mammals (e.g., Hirose et al., 2004). In line with these facts, TBT and TPT were given the highest category in a European review of endocrine disrupting chemicals (BKH, 2000): “Evidence for endocrine disruption in living organisms”. TBT was also classified as “Evidence of potential to cause endocrine disruption in humans”.

Organotins are also toxic by other mechanisms. For instance, several organotins are strongly immunosuppressive, display developmental and reproductive effects and are neurotoxic (WHO, 1999; EFSA, 2004). TPT is classified as category 3 carcinogenic in the EU, but as category 2 (probable human carcinogenic) by the USEPA (EFSA, 2004). DBT may actually be more toxic than TBT to certain enzyme systems (see Santillo et al., 2001). Immunotoxic and developmental effects in mammals may also be more sensitive to DBT than to TBT (Santillo et al., 2001). Both TBT and TPT may be classified as PBT and vPvB substances according the criteria in TGD (2002), whereas DBT and DOT may be classified as PBT (RPA, 2005). Examples of current classification for certain OTCs are given in Table 1.

Some examples of ecotoxicological effect levels are presented in Table 2, which illustrates that ecotoxicity increases dramatically in the order MBT < DBT < TBT for certain endpoints. As discussed above, however, DBT is more toxic than TBT for certain modes of action. A selection of ecotoxicologically based guidevalues are presented in Table 3.

For human health, there are no epidemiological studies on chronic low level exposure available (Appel, 2004). The CSTE suggested that toxicity was equal for DBT, TBT, DOT and TPT for humans, and proposed a group TDI of  $0.1 \mu\text{g Sn (kg Bw and d)}^{-1}$  (see RPA, 2005).





**Table 1.** Examples of classification for different organotin compounds. Sources: KIFS 2005:5; PRIO; ESIS.

Substance	Health	Environment
DBT (Dibutyltinhydrogenborate)	Toxic: danger of serious damage to health by prolonged exposure if swallowed	Very toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment.
TBT (Tributyltin compounds; TBTO)	Toxic: danger of serious damage to health by prolonged exposure through inhalation, in contact with skin and if swallowed Endocrine disruption	Very toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment. PBT, vPvB Endocrine disruption
TPT (Triphenyltinhydroxide)	Limited evidence of a carcinogenic effect. Possible risk of harm to the unborn child. Toxic: danger of serious damage to health by prolonged exposure through inhalation, in contact with skin and if swallowed	Very toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment. Potential PBT, vPvB
DOT (Dichlorodioctyltin)		Potential PBT/vPvB

**Table 2.** Ecotoxicological data. Source: Prevent database.

Substance	Cas	Organotin unit	Toxicological measure	Value
Butyltintrichloride	1118-46-3	MBT	EC <sub>50</sub> (Daphnia magna)	49 mg/l/24h.
Dibutyltindichloride	683-18-1	DBT	LC <sub>50</sub> (Daphnia magna)	0,9 mg/l/24h.
Tributyltinchloride	1461-22-9	TBT	EC <sub>50</sub> (Daphnia magna)	0,0006 mg/l/48h
Tetrabutyltin	1461-25-2	TeBT	EC <sub>50</sub> (Daphnia magna)	1,3 mg/l/48h
Dichlorodioctyltin	3542-36-7	DOT	EC <sub>50</sub> (Daphnia magna)	0,005 mg/l/24h.
Dichlorodioctyltin	3542-36-7	DOT	NOEC, (Scenedesmus subspicatus)	>0,0017 mg/l/72h.
Tricyklohexyltinhydroxide	13121-70-5	TCHT	EC <sub>50</sub> (Daphnia: magna)	0,005 mg/l/48h
Triphenyltinchloride	639-58-7	TPT	LC <sub>50</sub> (Daphnia magna)	0,035 mg/l/24h.
Tetraphenyltin	595-90-4	TePT	LC <sub>50</sub> Fish (Leuciscus idus)	0,04 mg/l/48h.

**Table 3.** Ecotoxicologically based guidevalues for organotin compounds.

Substance	Media	Target	Value	Reference
TBT	freshwater, coastal marine	aquatic ecosystem	0.1 ng/l annual average	proposed EQS for WFD
TBT	freshwater, coastal marine	aquatic ecosystem	1.5 ng/l maximum transient concentration	proposed EQS for WFD
TBT	freshwater	aquatic ecosystem	6 ng Sn/l	RPA, 2005
TPT	freshwater	aquatic ecosystem	3 ng Sn/l	RPA, 2005
DBT	freshwater	aquatic ecosystem	400 ng Sn/l	RPA, 2005
TBT	marine	aquatic ecosystem	1 ng/l	CCME, 2003
TBT	freshwater	aquatic ecosystem	8 ng/l	CCME, 2003
TPT	freshwater	aquatic ecosystem	22 ng/l	CCME, 2003

L:\3655\2005\10060857\_Screening av tennororganiska ämnen\C-Genomförande\23-Skisser-Utkast\tem-rapport-rev\_060630.doc

### 3. Use of organotin substances

The following substances are included in this study: monobutyltin, dibutyltin, tributyltin, tetrabutyltin, mono-octyltin, dioctyltin, tricyclohexyltin, monophenyltin, diphenyltin och triphenyltin. The current use of organotin compounds in Sweden are summarized in this chapter. This assessment is mainly based on data from the Swedish registry of chemical products, as displayed in the SPIN-database. Data may thus be incomplete due to confidential information. Furthermore, there are no official statistics on the import of chemicals in finished goods. This aspect can only be treated qualitatively.

The basic organotin compounds are used in a variety of chemicals. The chemicals that are used in the largest quantities are shown in Table 4. The total Swedish usage varied within 36-60 ton Sn per year during 1999-2003, with no significant trend. The use of OTCs in chemical products is completely dominated by dibutyltin and dioctyltin compounds. The specific uses of the various OTCs that are introduced as chemical products on the Swedish market are summarized in appendix 1. As a general rule, trisubstituted OTCs are used as biocides and other OTCs as industrial chemicals. In Sweden, the DBT and DOT compounds that are listed in Table 4 are mostly used in the manufacturing of plastic products and water-based paint, mainly as light and heat stabilisers for PVC. DBT may also be used as catalysts for PUF production, and as vulcanising agents for silicone rubber.

**Table 4.** Total use of organotin substances as chemical products in Sweden. Only the major substances that were found in SPIN are presented. Amounts are given in tonnes Sn/year, to facilitate comparison between substances with different molar mass.

Chemical compound	OTC	CAS	1999	2000	2001	2002	2003
Monobutyltintris-(isooctylthioglycolate)	MBT	26864-37-9	0.89	0.60	0.15		
Dibutyltinbis(2-ethylhexylthioglycolate)	DBT	10584-98-2	7.8	13.4	10.6	10.2	11.9
Dibutyltindilaurat	DBT	77-58-7	7.3	4.0	6.0	7.1	1.7
Dibutyltinoxid	DBT	818-08-6	9.5	8.6	7.6	6.7	4.3
Di-n-Butylbis(methyl maleate)tin	DBT	15546-11-9	9.4	10.2	9.7	5.6	6.0
Tributyltinmetacrylate	TBT	26354-18-7	0.75	1.0	0.25		
Dioctyltin bis(thioglycolic acid) 2-ethylhexyl ester	DOT	15571-58-1	2.2	17.7	17.2	7.4	8.4
Tinbis(2-ethylhexanoate)	DOT	301-10-0		1.8	8.8	8.5	5.6
Total			36	57	60	45	38

Trisubstituted compounds such as TBT and TPT are mainly used as biocides, e.g. in antifouling paint, wood preservation, agricultural pesticides and various industrial applications. The use of TBT and TPT has decreased strongly in many countries. In Sweden, the use of TBT in antifouling paints was banned in 1989 for ships less than 25 meter. The ban was extended to also include longer ships in January 2003, in Sweden as well as in the EC. There is no registered use of phenyltin compounds in Sweden at present. One pesticide containing phenyltin was banned in 1995. In northwestern Europe during 1995, the use in marine environments



was estimated to ca 70 tonnes and 1.5 tonnes, for TBT and TPT respectively (OSPAR, 2000). TBT was previously used in wood preservation.

Imported products is probably an important source of organotins to Sweden, and for which there is no official statistics. Various common products where the presence of organotins has been demonstrated during the last years is shown in Table 5. DBT is frequently the major species.

**Table 5.** Examples of organotin substances that occur in finished goods. When their presence is considered fairly common, it is marked with X. Less common occurrence is denoted x. The major species are underlined.

Product group	Dibutyltin	Mono-butyltin	Tributyltin	Octyltin	Levels, mg/kg	Reference
Bags	X	X		X	0.07-6.6	MST, 2001
Shower drapery	X	X			0.2-2.5	MST, 2001
Earplugs	<u>X</u>	x	x	x	0.1-1300	MST, 2003
PVC flooring	<u>X</u>	X	X	X	0.1-350	MST, 2001; Allsopp et al., 2000
Vinyl wallpaper	X	X	X	X	0.03-270	MST, 2001
Glue and sealants	X		x			Sveriges Byggin-dustrier (2000)
Diaper	X	X	X		0.002-0.02	Miljö kemi (2000)
Rain clothings	x		x			Ökottest / Råd & rön
Scotch Brite	X	X			0.002-0.015	Miljö kemi (2000)
Bathing ball	x	x			1-14	Miljö kemi (2000)

The Swedish recent use of organotin compounds can be summarized as:

- Dibutyltin: mainly as stabilizer in PVC, but also in paint, sealants etc
- Octyltin: mainly stabilizer in PVC, possibly food packages etc
- Tributyltin: as a biocide in paint for ships (prohibited 1988/2003); smaller amounts are still used in unspecified applications
- Triphenyltin: biocid, as a biocide in paint for ship, no registered use
- Tricyclohexyltin: no registered use



## 4. Environmental occurrence

By far, most studies on to the environmental occurrence of organotin substances have focused on tributyltin, the biocidal use of which has caused significant environmental impact in marine harbours world-wide. However, the use of TBT is mainly restricted to marine environments and is also declining due to international regulations. As shown in chapter 3, current societal use mainly concerns mono- and dialkylated substances, which therefore may be expected to occur in freshwaters as well as in sewage sludge. It has recently been demonstrated that organotin compounds may undergo atmospheric transport (Huang and Klemm, 2004). Measurements in air and deposition are so far restricted to a region in eastern Germany, where MBT and MOT were the major species in deposition. Therefore, even remote inland regions may contain OTCs. In this chapter, we summarize some recent studies on the environmental distribution on organotin substances, mainly from Sweden.

### 4.1. Sewage treatment plants

Data on sludge from municipal STPs are summarized in Table 6. In the Västra Götaland county, sludge from 19 municipal sewage treatment plants were analysed during 2002 (Svensson, 2002). MBT and DBT were most common, occurring at roughly similar levels. They are followed by TBT, MOT and DOT. In the Loudden MSTP, much lower levels of MBT were obtained. Phenyltin compounds were not detected, in agreement with the fact that there is no reported use of these substances in Sweden (chapter 3). An earlier Swedish study found higher levels of several OTCs in sludge.

Levels in municipal and industrial waste waters are shown in Table 7. There are fairly large variations between different studies. The waste waters contain the same OTCs as the sludge samples. The levels in the industrial waste waters overlapped with those from households, but were occasionally higher.

**Table 6.** The occurrence of organotin substances in sludge from municipal STPs ( $\mu\text{g}/\text{kg dw}$ ).

Substance	V. Götaland, 19 STPs Svensson (2002)		Loudden (Tesfalidet, 2002)	5 STP Norén och Borén (1993)
	median	min-max	Average $\pm$ std. dev.	min-max
MBT	290	120-870	39 $\pm$ 14	100-770
DBT	250	37-350	690 $\pm$ 180	330-2200
TBT	44	10-96	32 $\pm$ 18	20-410
TeBT	4.4			
MPT	<1	<1		
DPT	<1	<1		<2-70
TPT	<1	<1		0-60
MOT	24	9.6-49		
DOT	14	6.5-73		<70-370

L:\3655\2005\10060857 Screening av tennorganiska ämnen\C-  
Genomförande\23-Skisser-Utkast\tem-rapport-rev\_060630.doc



**Table 7.** Organotin compounds in waste waters in Sweden (ng/l). BDT means water from bathing, showers and toilet.

	INCOMING WASTE WATERS					EFFLUENTS	
	Stockholm, household <sup>A</sup>	Stockholm, industrial areas <sup>A</sup>	Gryaab <sup>B</sup>	BDT <sup>C, D</sup>	Vibyåsen <sup>C</sup>	Bromma <sup>B</sup>	Borlänge <sup>E</sup>
MBT	36-58	22-81	35	17- 74	6	2.5 ± 1.9	11
DBT	49-73	10-120	96	5-18	10	0.06 ± 0.1	11
TBT	3-6	2-4	<1	0.8- 4.3	2		2.7
TeBT	<1	<1	<1		<1		ND
MPT			<1	<1			ND
TPT	<1	<1	<1	<1	<1	1.2 ± 0.9	2,3
MOT	5-9	5-20	8.4		<1		ND
DOT	8-12	3-28	16		<1		ND

A. Andersson, 2004; B. Tesfalidet; C. Palmquist and Hanneus, 2001 (maximum value of two points); D. Andersson and Jensen (2002); E. Junestedt et al., 2003.

**Table 8.** Organotin compounds in surface waters (ng/l). All data from Tesfalidet (2003), except Stockholm (Junestedt et.al., 2003).

	Karlsudd (Gålö)	Norrby, Umeå	Fiskebäckskilsvik	Hinsholmskil,	Stockholm city
MBT	3.9 ± 2.9	6.0 ± 3.2	3.8 - 5.9	0.05 – 0.98	5.6
DBT	1.7 ± 1.0	4.4 ± 2.4	0.40 - 0.77	0.01 – 0.77	5.8
TBT	0.9 ± 0.5	4.7 ± 2.1	0.1 – 0.6	0.1 – 0.9	2.4
MPT	ND	4.0 ± 2.3	ND - 0.7	0.01 – 0.13	ND
DPT	ND	4.7 ± 2.2	ND - 0.56	0.02 – 0.94	ND
TPT	ND	4.16 ± 2.9	ND – 0.23	0.01 – 0.82	ND

**Table 9.** Organotin substances in landfill leachates and storm waters from industrial sites (ng/l). All data from Junestedt et.al. (2003).

	Landfills, 5 Swedish	Industrial stormwaters, 5 Swedish (median, min-max)
MBT	7 - 50	100 (30-9600)
DBT	3 - 500	200 (2-18 000)
TBT	<1 - 60	300 (10-500)
TeBT	<1 - 20	< 1
MPT	<1 - 3	<1
DPT	<1 – 2	<1
TPT	<1	<1
MOT	<1 – 10	10 (1 - 1 700)
DOT	< 1 -40	20 (1 - 4 200)

L:\3655\2005\10060857 Screening av tennorganiska ämnen\C-Genomförande\23-Skisser-Utkast\tem-rappoirt-rev\_060630.doc



## 4.2. Surface water, storm water etc

Only few measurements of OTCs in surface waters have been performed in Sweden (Table 8). Butyltins are generally the major species in surface waters. Higher levels than in surface waters have been measured in landfill leachates and industrial stormwaters, where also octyltins were detected (Table 9). The levels in stormwaters from various industrial sites are particularly high.

## 4.3. Sediment, biota and foodstuffs

To date most sediment data are from the marine environment (Table 10). Butyltins are generally the major organotin group in marine sediments, although phenyltins are also found. TBT was recently analysed in sediment from Stockholm city, suburban lakes and the Svealand coast (Sternbeck et al., 2003). Almost no lake contained detectable levels, whereas the Stockholm city contained higher levels than the coast.

Butyltins and phenyltins have frequently been detected in fish, mussel and shellfish. Some recent Swedish data on aquatic biota is shown in Table 11 and the results of a large European survey on marine foodstuffs is summarized in Table 12. Shellfish contains higher levels of butyltins than of phenyltins, whereas these groups are roughly equally abundant in fish.

**Table 10.** Organotin compounds in surface sediments (ng/g dw) in Sweden.

Substance	Baltic coast and open sea (Cato, 2003)	Gothenburg harbour (Brack, 2000)	Gålö, Stockholm archipelago 0-10 cm (Tesfalidet, 2003)
MBT	<1-50	6-44	4-28
DBT	<1-210	17-98	7-88
TBT	<1-1400	17-366	3.4-70
TeBT	<1-17		
MPT	<1-7	< 0.8-5.5	1-24
DPT	<1-14	< 1 6.2	1-25
TPT	<1-72	<1.5-71	
MOT	<1		
DOT	<1		

**Table 11.** Swedish recent data on organotin compounds in fish (ng/g ww) and mussel (ng/g dw).

Substance	Salmon etc, Vänern-Vättern (Öberg, 2002).	Herring and Salmon, Umeå coast (Tesfalidet, 2003)	Blue Mussel, Fiskebäckskil (Tesfalidet 2003)
MBT		0.3-0.6	26- 600
DBT		1.0-3.6	25-370
TBT	average 1.2 (0.3-6)	10-26	89-870
MPT		1.4-25	
DPT		0.2-2.6	
TPT	average 7 (0.4-21)	18-31	



**Table 12.** European data on organotin compounds in foodstuffs (ng/g ww) (EFSA, 2004).

	Fish and fishery products			Shellfish		
	Mean	Median	95-percentile	Mean	Median	95-percentile
MBT	10	2.5	25	34	4	215
DBT	17	2.5	35	52	4	370
TBT	28	7	107	60	14	210
MPT	7.0	2.5	23	12	2.5	80
DPT	2.6	1.5	4	2.0	1.5	2.5
TPT	17	4	63	21	3	120



## 5. Sampling strategy and study areas

The study consists of a national programme, financed by the Swedish EPA, and regional programmes for the counties of Dalarna, Södermanland and Skåne, respectively. The strategy of the joint programme is outlined below, and the national and regional programmes are shown in Table 14 and Table 13. All sample details are listed in Appendix 2. The localisation of the different regions is shown in Figure 2.

- ❑ A possible urban influence, resulting from diffuse emissions, was investigated by sampling in three urban regions (Stockholm, Eskilstuna and Borås). This includes both local background, city center, and downstream.
- ❑ The role of wastewater was investigated at seven municipal sewage treatment plants and at the recipients of some of these STPs.
- ❑ To illustrate point source emissions, samples were taken close to a PVC industry and in three landfill leachates.
- ❑ An agricultural area where STP sludge is used as a fertiliser was investigated, including a local reference area.
- ❑ As indicators of human exposure, 12 different foods and 5 samples of breast milk were analysed.

**Table 13.** National programme. The total number of samples is 83.

Description	STP influent	STP effluent	STP sludge	Storm water sludge	Sediment	Surface water	Soil	Foods	Breast milk	Fish
STP Henriksdal	2	2	3			3				
STP Gässlösa, Borås	2	2	3							
PVC-compounding industry				2			2			
Sludge end-use						2	4			
Urban regions: Stockholm				2	7	6				2
Urban region: Eskilstuna				2	2	3				
Urban region: Borås, river Viskan					3	3				
Urban region: Helsingborg				2						
Foods								12		
Breast milk									5	
Background areas										2
Marine coast										5
<b>National total</b>	<b>4</b>	<b>4</b>	<b>6</b>	<b>8</b>	<b>12</b>	<b>17</b>	<b>6</b>	<b>12</b>	<b>5</b>	<b>9</b>

L:\3655\2005\10060857 Screening av tennororganiska ämnen\C-Genomförande\23-Skisser-Utkast\tem-rapport-rev\_060630.doc





**Table 14.** Regional programmes. The total number of samples is 25.

AREA	Leachate	STP influent	STP effluent	STP Sludge	Sediments	Surface water	Biota
<b>Dalarna</b>							
STP Fagersta, Borlänge		1	1	1			
Landfill Borlänge	1						
STP Krylbo, Avesta			1	1			
River Bäringen					1	1	
<b>Södermanland</b>							
STP Eskilstuna		1	1	1		1	
Landfill Eskilstuna	1					1	
STP Nyköping			1	1	2		
Landfill Nyköping	1						
STP Flen			1	1			
<b>Skåne</b>							
Mussel, Öresund							4
<b>Total regional</b>	<b>3</b>	<b>2</b>	<b>5</b>	<b>5</b>	<b>3</b>	<b>3</b>	<b>4</b>

## 5.1. Urban areas

Three urban areas of varying sizes were investigated: Stockholm, Eskilstuna and Borås. Sediment and surface waters were sampled in Stockholm, Eskilstuna and river Viskan close to Borås, and in upstream sites representing urban background (Figure 3, Figure 4). Pooled samples of mature perch (*Perca fluviatilis*) in a transect from lake Mälaren, through Stockholm city and in the Stockholm archipelago were also analysed. In order to investigate whether organotin compounds are released by diffuse emissions in urban areas, six samples of urban stormwater sludge were also taken in Stockholm, Eskilstuna and Helsingborg.

In Stockholm, water flows from the large lake Mälaren in the west and mixes with brackish water in the Stockholm city. The sediments in Stockholm are highly polluted with respect to metals as well as many organic contaminants (e.g., Sternbeck et al., 2003). With respect to organotin compounds, large ships frequently travel through Stockholm on their way into other cities around the large Lake Mälaren.

The eastern part of Lake Hjälmaren flows into the river Eskilstunån, which runs through Eskilstuna city and finally reaches lake Mälaren. A number of smaller industries are located in Eskilstuna.

In the river Viskan, the two lakes downstream of Borås city (Guttasjön and Djupasjön) are severely polluted with e.g. PCDD/Fs due to earlier emissions from the textile industry. A municipal sewage treatment plant (Gässlösa) has its effluent located between Druvefors and Djupasjön (Figure 4). The local background site Öresjön serves as the water supply for Borås.

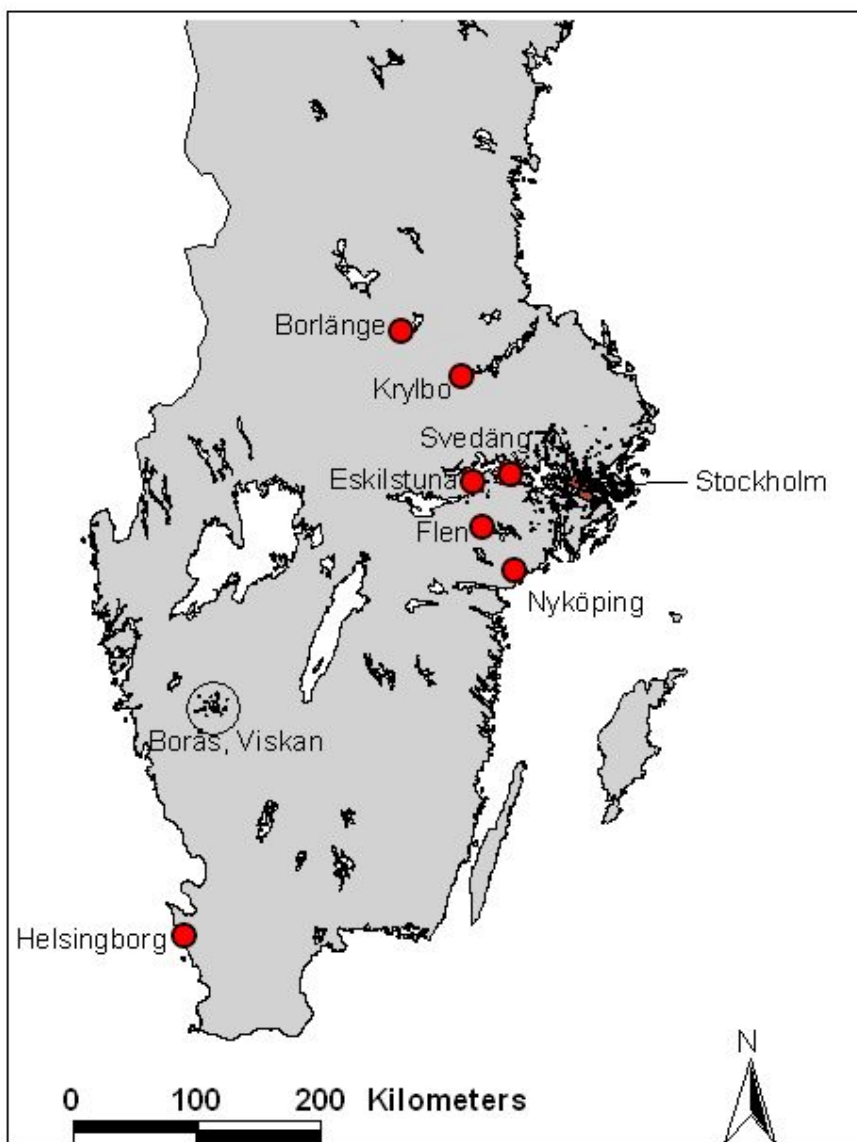
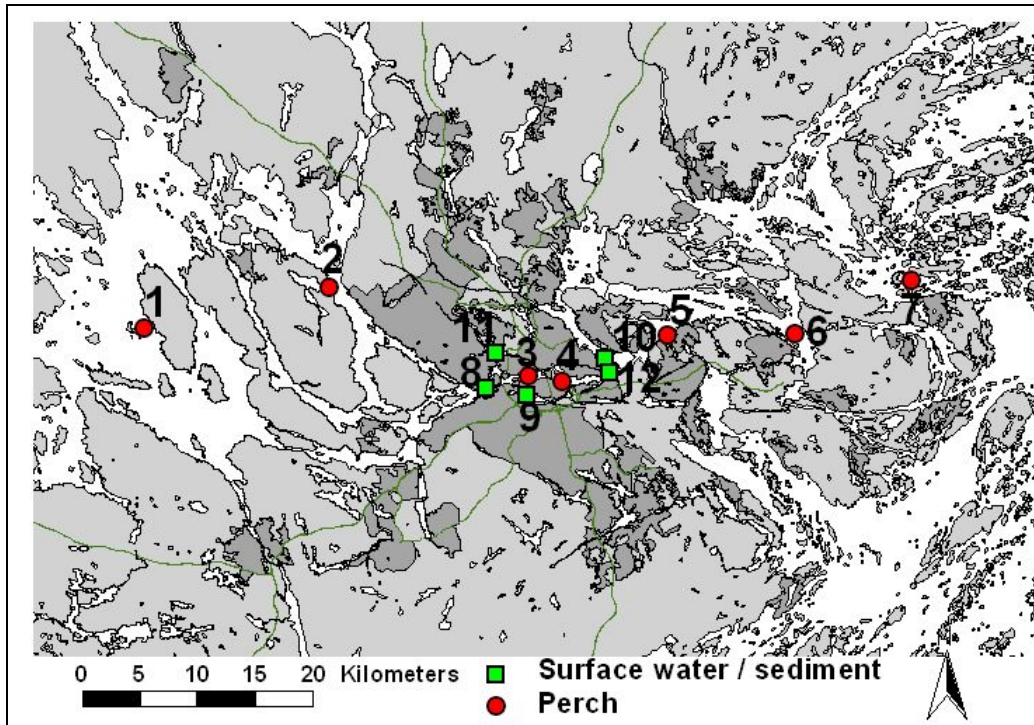
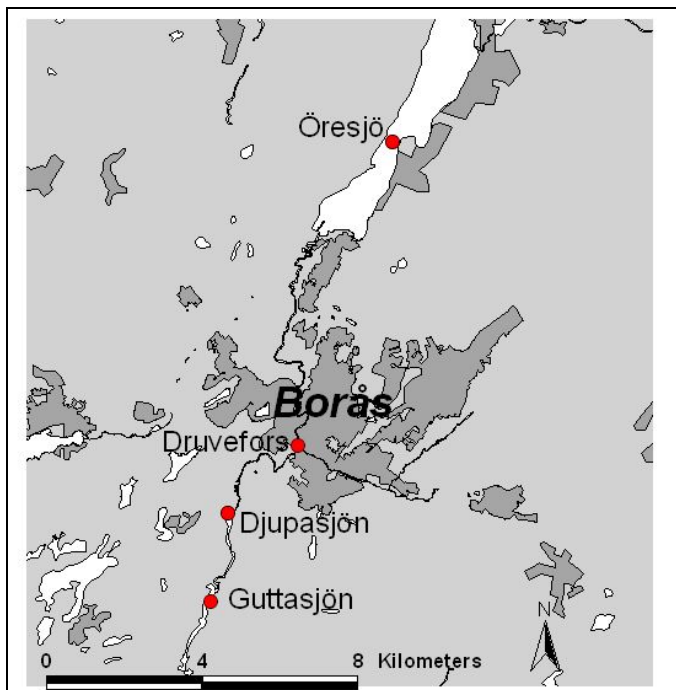


Figure 2. Overview of the investigated regions.

L:\3655\2005\10060857 Screening av tenororganiska ämnen\C-  
Genomförande\23-Skisser-Utkast\tem-rapport-rev\_060630.doc



**Figure 3.** Sampling stations in the Stockholm area. 1. Adelsö; 2. Färingsö; 3. Riddarfjärden; 4. Slussen; 5. Kummelnäs; 6. Torsbyfjärden; 7. Vindö; 8. Mälaren Rotholmen; 9. Årstaviken; 10. Lilla Värtan; 11. Ulvsundasjön; 12. Fjäderholmarna.



**Figure 4.** Sampling stations in Viskan, Borås.

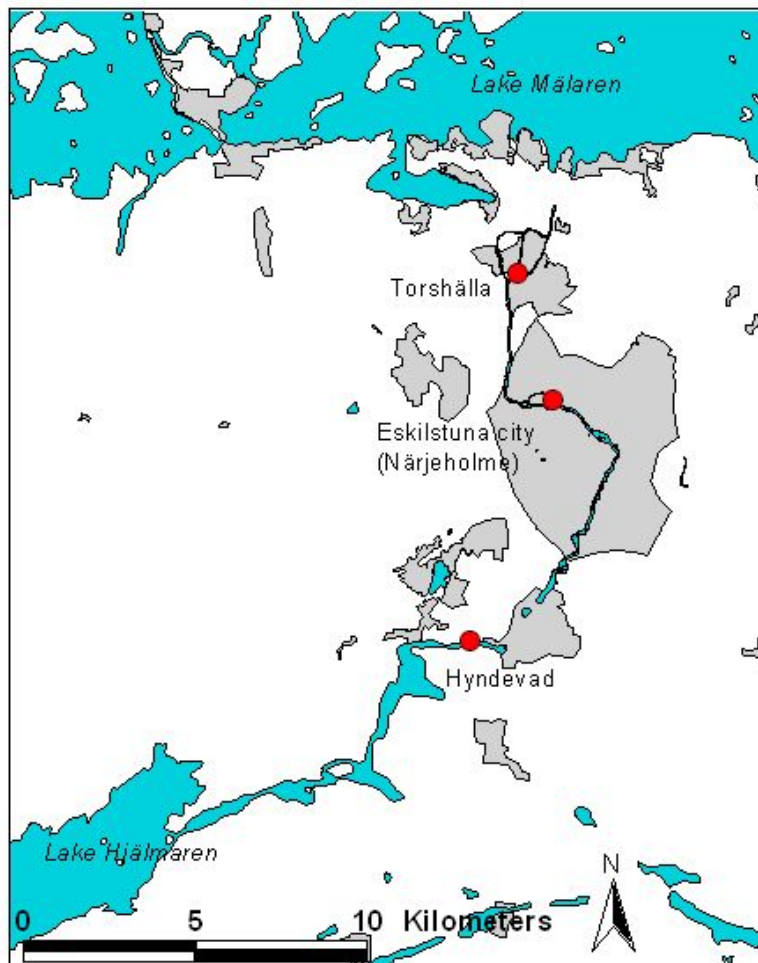


Figure 5. Sampling stations in Eskilstuna.

## 5.2. Sewage treatment plants

Seven municipal sewage treatment plants (STP) were sampled for incoming waste waters, sludge and effluents. Also surface waters and sediments were analysed in the recipients of some of these STPs, in order to investigate whether effluents influenced the environmental levels of organotin compounds. Details of the STPs are summarized in Table 15. At Henriksdal and Gässlösa, sludge was sampled in September and November 2005 and in February 2006; influents and effluents were sampled in September 2005 and February 2006. All other samples were taken in September 2005. Gässlösa has a fairly high industrial load from textile industries.



**Table 15.** Details of the sewage treatment plants that were sampled. The size of the STPs are given as population equivalents, pe.

Sewage treatment plant	Digestion	Total PE	Industrial load, in pe	Household load, in pe	Stormwater (% of water flow)
Henriksdal, Stockholm	+	750 000	70 000	680 000	30
Gässlösa, Borås	+	101 200	21 200	80 000	47
Eskilstuna	+	64 000	9800		40
Fagersta, Borlänge	+				8
Flen	-	15 000	6000	9000	no data
Brandholmen, Nyköping	+				no data
Krylbo, Avesta	+	24 000	7500	17 000	44

### 5.3. Potential point sources

To illustrate whether organotin compounds may be released to the environment during industrial processing, soil and stormwater sludge were collected close to a PVC processing industry (Hydro Polymers in Helsingborg). Upper soil was sampled in two directions from the industry, within ca 250 meters. Sludge from the gutter in the immediate surroundings was also sampled at two directions from the industry.

Leachates from three landfills were sampled in the regional programmes.

To illustrate whether the use of sewage sludge as a fertiliser may pollute agricultural soils, samples of upper soil were taken in an agricultural field where sewage sludge were used in the production biofuel (*Salix* plants). A control field in the surroundings was also sampled. Surface waters were also sampled from two points (upstream and downstream) in a small river at this site.



## 6. Methods

### 6.1. Sampling

Surface waters were collected with a Ruttner sampler and were stored in dark 1 liter glass bottles. The bottles were first rinsed with the sample. Sediments were taken with a Willner sampler or similar, and sliced into 2 cm slices in the field. Upper soil samples were taken as pooled samples within a smaller area, and litter was removed. Samples at the STPs were taken by their staff, following common procedures. A protocol for sampling was sent to all personal involved in sampling, to assure similar treatment. Waste waters were sampled for five consecutive days and pooled at the fifth day. Individual samples were stored cold and dark. Sludge was taken as a pooled sample on one day. Perch from the Stockholm region were caught during 2000/2001 by ITM, Stockholm University, who kindly supplied us with these samples.

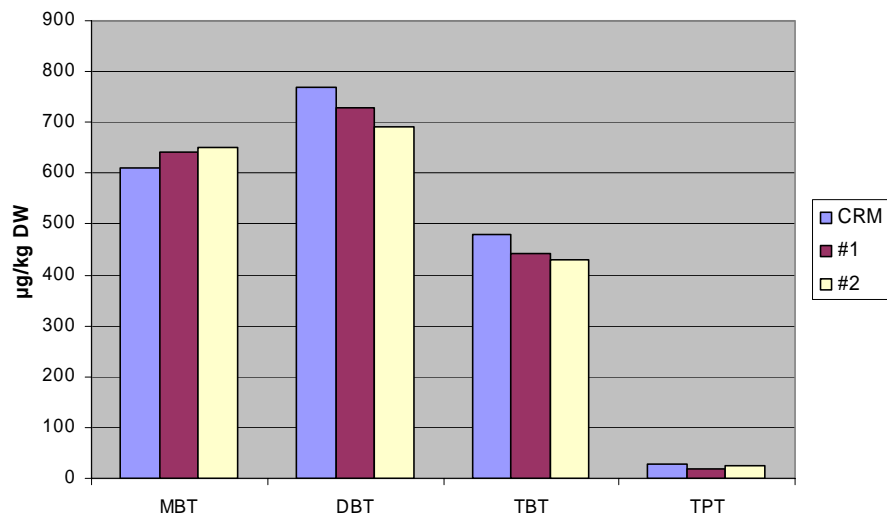
Foodstuffs and breast milk were kindly supplied by the Swedish National Food Administration. For the foodstuffs, pooled samples of fish muscle were caught 2001-2002. The meat sample was collected in a so called food-basket survey, bought in Sundvall in spring 2005. The canned mussel sample was bought in Malmö 2005, and the field collected mussels were collected on the Swedish west coast during 2001. Breast milk were collected from primiparous women and all milk were gathered during the third week after birth. During the sampling week, breast milk was stored in acetone rinsed bottles in their freezers.

### 6.2. Chemical analysis and quality assurance

All samples were stored cold and dark until analysis, and were sent to the laboratory within two days after sampling. Samples were analysed by Analytica AB within ca ten days after sampling. The samples are homogenized and extracted by a mixture of methanol:hexane (1:1). The organic phase (methanol and hexane) is thereafter purified by alumina oxide and derivatized with NaBEt<sub>4</sub> prior to chemical analysis. The chemical analysis is performed by GC-AED (Agilent Technologies). The yield is justified with recovery tests using internal standard. The compounds are identified by comparison of retention times with standards.

Reporting limits are 1 ng/l for aqueous samples, 1 ng/g dw for soil, sludge and sediments, and 0.3-0.4 ng/g ww for food and biota. Analytical precision is 10-20 %. Results from analyses of a certified reference sediment (CRM 646) are shown in Figure 6.

The total uncertainty in the results is also influenced by natural heterogeneity and possible effects from sampling. As an indicative measure of the total uncertainty in the results, sediments and surface waters were sampled in duplicate from two sites in Stockholm, and pooled samples of perch from one site were analysed in triplicate. The uncertainty is presented as the relative standard deviation, which typically is high due to the low number of replicates. For perch, the uncertainty was in the range 2-13 % for individual substances. The uncertainty was frequently higher for surface waters and sediments. At one site, the uncertainty varied between 16 % and 108 % (average 58%) for individual substances. This is likely due to strongly heterogeneous levels in sediments. For the other sediments and surface waters, the average uncertainty was 10 % to 28 %, i.e. only slightly higher than the analytical precision.



**Figure 6.** Results from analysis of a certified reference materials. CRM represent the certified concentrations and #1 and #2 two replicate measurements.





## 7. Results

A general overview of the levels of the studied organotin compounds is presented for each media in this section. Sample details are given in Appendix 2 and all data are presented in Appendix 3. A discussion on spatial trends, emission sources, environmental partitioning and possible risks to the health and environment is given in chapter 8.

All analysed organotin compounds except TCHT were detected (Table 16, Appendix 3). The most commonly found substances were MBT, DBT and TBT. The number of media that each substance was detected in decreased in the following order: MBT, DBT > TBT > MOT, DOT, DPT > TPT > TeBT = MPT. TeBT was only detected in storm water sludge and in sediment, whereas MPT was only found in sediment and fish. Butyltins were detected in all media, octyltins mainly in wastewaters, sediments and urban runoff, and phenyltins mainly in the aquatic environment and in foods.

All concentrations are presented as the amount of the cation. The detection frequency varied widely for different substances. Substances with lower detection frequency will thus display average levels that are biased toward higher levels. Therefore we also show the detection frequency in separate graphs.

**Table 16.** The occurrence of different organotin compounds in different media, where n-tot is the number of samples analysed. The last row gives the overall detection frequency. Substances that only rarely occur in a certain media are shown with a small x.

Media	n-tot	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT
Soil	6	x	x			x	x				
Waste water	16	X	X	X		X	X				
Sewage sludge	11	X	X	X		X	X				
Storm water sludge	8	X	X	X	x	X	X			x	x
Landfill leachate	3	X	X								
Sediment	15	X	X	X	X	X	X		X	X	X
Surface water	18	X	X	X							
Aquatic biota	13	X	X	X					x	X	X
Foods	12	X	X	X						X	X
Breast milk	5	X									
Detection frequency	n <sub>tot</sub> =107	84%	86%	60%	7%	31%	30%	0%	10%	25%	28%

### Sediments

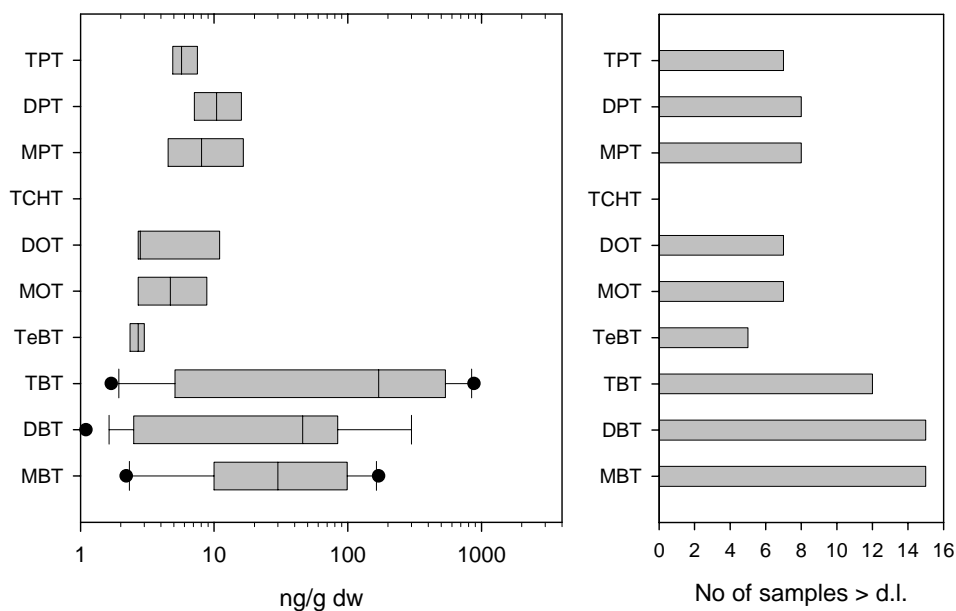
All analysed substances except TCHT were detected in sediments (Figure 7). TBT, DBT and MBT were most abundant. The levels of TBT, DBT and MBT were particularly high in Stockholm, and for DBT and MBT also in the lakes downstream of Borås. TBT was the major butyltin in Stockholm, but not at the other sites. Phenyltins were mainly detected in samples from Stockholm and Viskan, and at relatively low levels. Octyltins occurred at similar levels and sites as phenyltins.



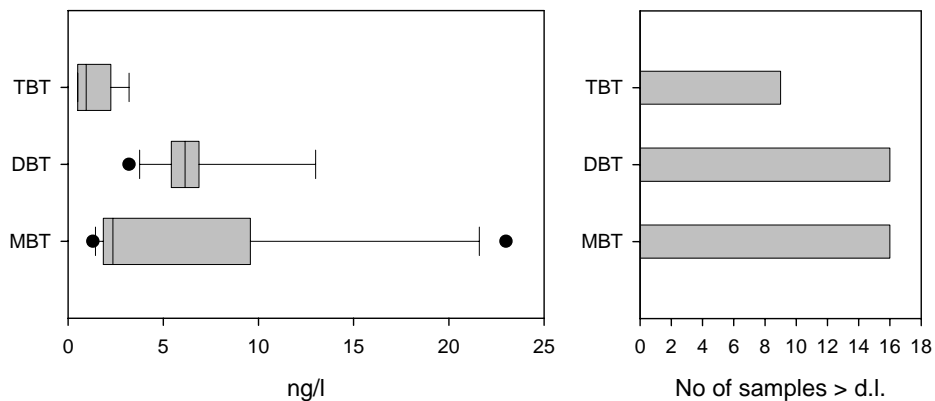


## Surface water

Only butyltin compounds were above the analytical reporting limit in surface waters (Figure 8). This is generally the case in other studies too, although Tesfalidet (2004) detected low levels of phenyltins in marine surface waters (Table 8). DBT and MBT were more abundant than TBT in this study, both in terms of concentrations and detection frequency. TBT was only detected in surface water from Stockholm. Eskilstuna displayed much higher levels of MBT than the other sites, whereas DBT occurred at similar levels in all three urban areas.



**Figure 7.** Levels of organotin compounds in surface sediments (n=15). Note the logarithmic concentration axis. The right graph shows the detection frequency. The detection limit was 1 ng/g dw.



**Figure 8.** Levels of organotin compounds in surface waters (n=18). The right graph shows the detection frequency. The detection limit was 1 ng/l.



## Sludge and waste water

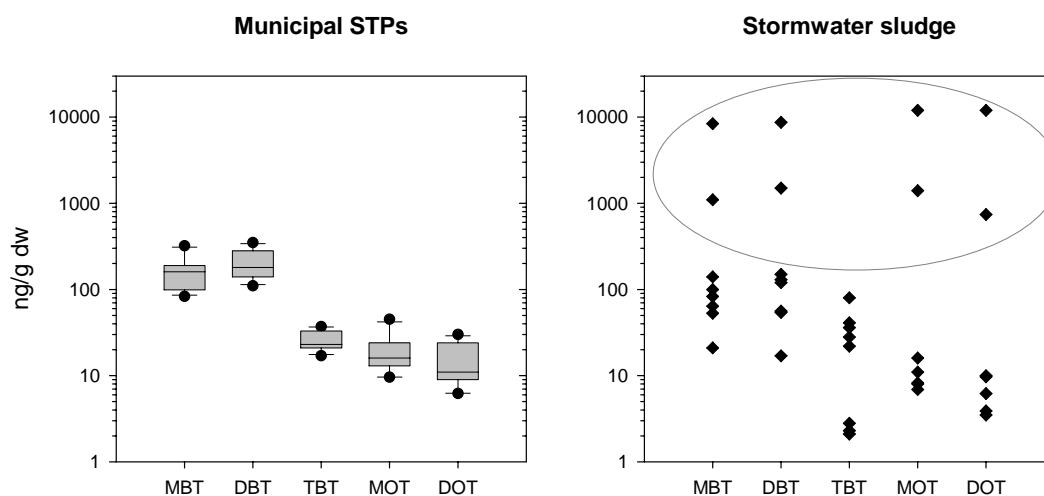
Municipal wastewater and sewage sludge from municipal sewage treatment plants (STPs) contained butyltins and octyltins, but no phenyltins (Figure 9 and Figure 10). The concentrations in sewage sludge and waste waters were in the same range as in other recent studies (Table 6, Table 7), with MBT and DBT as the major species. The five OTCs detected in sewage sludge were present in all sludge samples and in similar relative proportions. MBT and DBT were present in all samples of incoming waste water, MOT and DOT in all but one, and TBT in only two out of six (Figure 10). The effluents only contained MBT and DBT at detectable levels. Octyltins were present in one effluent sampled after a wetland (used as a final removal stage), but not in the sample before the wetland. No chemicals are used in this stage and neither is stormwater introduced between the sampling points, why we have no explanation for this anomaly.

Urban stormwater sludge contained the same substances as in sewage sludge, and in similar or slightly lower concentrations (Figure 9). Additionally, samples from Helsingborg and the PVC industry also contained TeBT and in a single case TPT and DPT. Stormwater sludge from the PVC industry contained orders of magnitude higher concentrations than the urban samples (Figure 9).

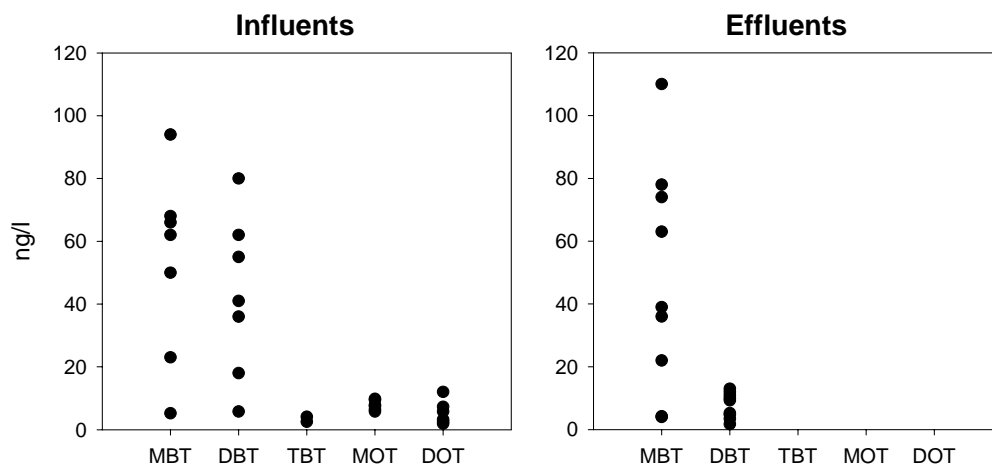
## Soil and landfill leachates

In the few soil samples that we studied, no OTCs were found in the agricultural soils. MBT, DBT, MOT and DOT were, however, found in soil close to the PVC industry at levels in the range 5-60 ng/g dw, with DBT as the major species. We are not aware of any other Swedish measurements of organotin substances in soil.

Leachates from three landfills were analysed and contained MBT and DBT in levels similar to those in STP effluents. Higher levels have occasionally been measured in landfill leachates earlier (Junestedt et al., 2003; see Table 9).



**Figure 9.** Levels of organotin compounds in sludge from municipal waste water treatment plants (n=11) and in stormwaters (n=6+2). In the right graph, samples from an industry are encircled. Note that the concentration axes are logarithmic. Detection frequency was 100% for these substances in sewage sludge. TeBT, DPT and TPT were also found in a few stormwater sludge samples.



**Figure 10.** Levels of organotin compounds in waste waters from municipal wastewater treatment plants.

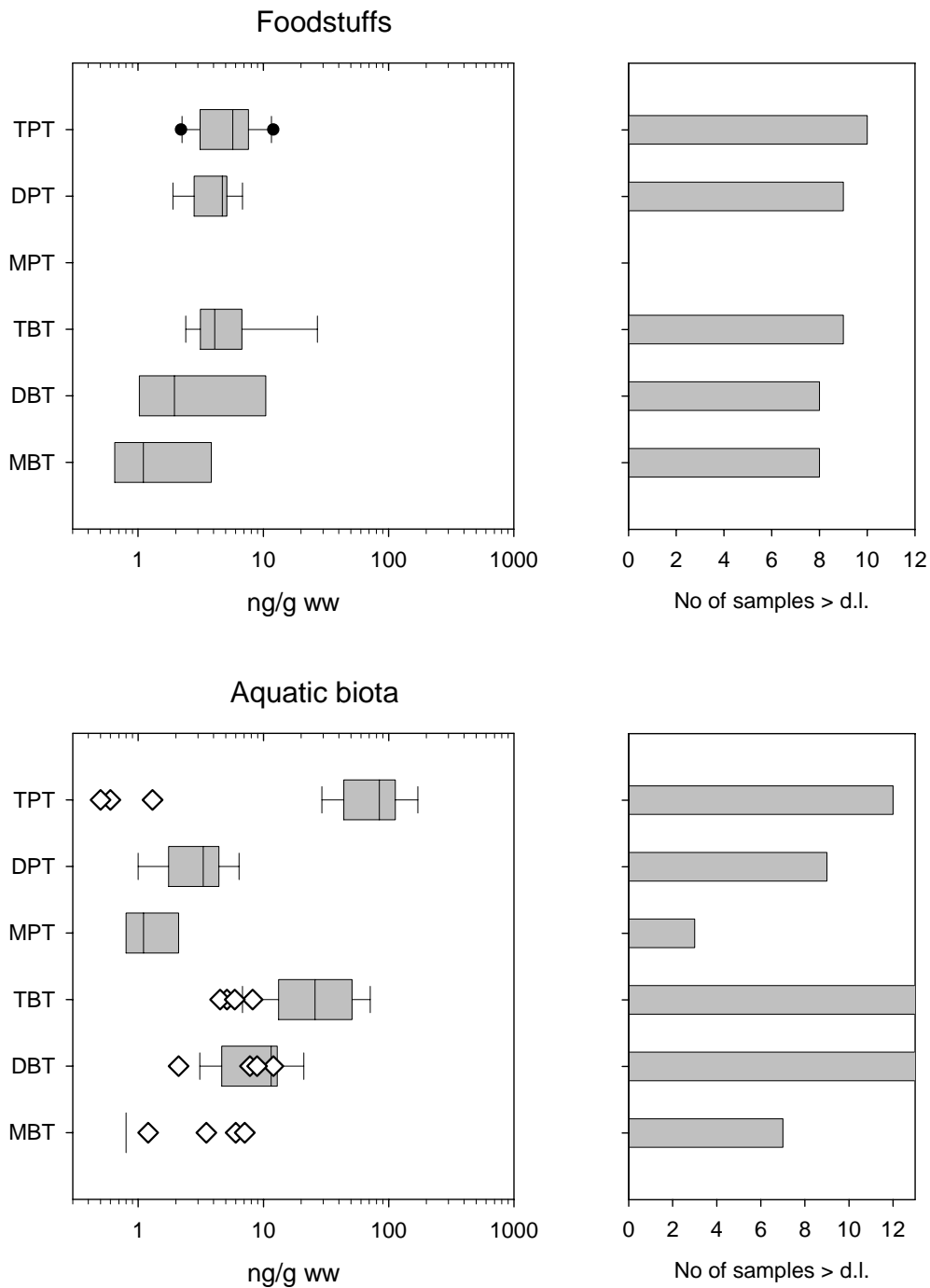
### Biota and foodstuffs

Aquatic organisms, including fish from Stockholm, mussels from Öresund, and samples of foodstuffs from various regions, are presented in Figure 11. Fish samples were analysed on muscle. Only butyltin and phenyltin compounds were found. TBT and TPT were detected in almost every sample. Perch from the Stockholm region consistently contained much higher levels of DBT, TBT and TPT than did fish in the food group. Even perch from the urban background sites in lake Mälaren contained higher levels than fish from other areas, in particular for TPT. Phenyltins are frequently more abundant than butyltins in fish, whereas the opposite was true in mussels. In addition, one pooled sample of meat contained DBT and DPT at concentrations of 1.1 and 1.9 ng/g ww, respectively. More detailed description of these data are given in chapters 8.2 and 8.6.

The only detected substance in breast milk was MBT (Table 17). The levels varied from 1 to 10 ng/l. There are to our knowledge no earlier measurements made of organotin compounds in breast milk from Sweden or elsewhere.

**Table 17.** Levels of MBT in breast milk. Reporting limits were 0.3-0.4 ng/l for individual OTCs.

City	Year	MBT (ng/l)
Uppsala	2002	1.2
Uppsala	2004	1.9
Göteborg	2001	9.7
Lund	2003	ND
Lycksele	2004	5.8



**Figure 11.** Levels of organotin compounds in various foodstuffs (upper graph) and aquatic biota (lower graph). In the lower graph, perch from the Stockholm region is shown as boxes and mussel from Öresund is shown as diamonds. The concentration axes are logarithmic. The right graphs show the number of samples with levels > d.l. Total number of samples are 12 and 13, respectively, and the detection limit was 0.3-0.4 ng/g w.w.



## 8. Discussion

There is a good correspondence between the type of organotin compounds found in the present study and those that are used in the society. Phenyltins mainly occur in the aquatic environment in areas where large ships travel. Octyltins are found in waste waters and sludge, in urban stormwaters and in certain urban sediments, suggesting an association to products containing octyltins. Butyltins are found in all these environments, in agreement with their use both in the marine environment and in urban areas. The high abundance of MBT in spite of its fairly low societal use is most likely explained by its formation in the environment from dealkylation of DBT.

### 8.1. Background levels

A background site may in this case be defined as a site where there is no input of organotin compounds from point sources or diffuse sources. Due to the release of organotin compounds from ships in the marine environment, marine areas are not expected as background areas. The broad range of non-marine applications of OTCs suggests that also inland areas may be impacted by their release. The occurrence of OTCs in rural and remote terrestrial regions has rarely been investigated and OTCs are generally not considered for long-range transport. A recent study demonstrated the presence of butyltin and octyltin compounds in air and precipitation in rural forest sites in Eastern Germany (Huang and Klemm, 2004). The levels in precipitation were ca 15 ng/l for MBT, 4 ng/l for DBT and 6 ng/l for MOT. The partitioning to particles and the washout factors in rain increased with decreasing alkylation number; i.e. deposition to the terrestrial ecosystem was dominated by MBT and MOT. This agreed with the occurrence of organotin substances in background soils, where MBT and MOT ranged from 0-30 and 0-9 ng Sn/g dw, respectively (Huang et al., 2004). Other OTCs occurred at less than 1 ng/g dw.

In this study only one general background (Svedäng) was included where soil and surface water were sampled in an agricultural area. There were no signs of the presence of OTCs. Although there may be an atmospheric deposition of in particular MBT and MOT, levels in agricultural topsoils was below the detection limit, possibly due to the mixing effect of plowing. In a German study, agricultural soils that were not treated with sludge contained sum-OTC levels up to ca 3 ng/g dw (LFU, 2003).

Urban background samples of surface water, sediment and fish were also studied in Mälaren, Viskan and Eskilstuna. These areas are not remote background areas. All these local background samples contained detectable levels of OTCs. The background sites at Eskilstuna (Hyndevad) and Viskan (Öresjö) contained MBT, DBT and TBT, with MBT as the major species. On the basis of this investigation, it is not possible to conclude whether these observations are due to local releases in these background sites, or due to atmospheric deposition. It is, however, interesting to note that the levels in surface water were in the same range as found in precipitation in Germany (see above).

In the background sites to Stockholm (Mälaren Rotholmen), the sediments also contained octyltins and phenyltins, and the butyltins were detected at much higher concentrations than in Öresjö and Hyndevad (Table 18). Samples of perch from lake Mälaren contained DBT, TBT, DPT and TPT. The dominating compound in perch was TPT (40 ng/g ww) followed by TBT (13 ng/g ww), DBT (5 ng/g ww), and DPT (1-2 ng/g ww). These sites were selected as local background to Stockholm, but are clearly influenced by local or regional OTC releases, probably from ships but also other sources since octyltins were found in the sediments.



## 8.2. Urban areas

Stormwater sludge from three cities all showed the presence of MBT, DBT, TBT, MOT and DOT (Figure 9), demonstrating that diffuse emissions of OTCs occur in urban areas. We are not aware of any previous measurements of organotin compounds in urban runoff, although high levels have been detected in runoff from industrial sites (Junestedt et al., 2003). The organotins that were found in these sludge samples are those that are commonly used in various building products, why their presence in urban runoff is expected. The influence of urban runoff on the local aquatic environment depends partly on whether these stormwaters are treated in STPs or are directly emitted to the recipients.

A summary of the organotin levels in urban sediments is given in Table 18, showing that elevated concentrations are found in all three cities. Stockholm is more strongly contaminated than the other cities, and all the three substance groups were found. Tributyltin was consistently higher than DBT and MBT in Stockholm, whereas the opposite was true for Eskilstuna and Viskan. Phenyltins were also found in Stockholm, although there is no registered use of phenyltins in Sweden since 1995 (chapter 3). These facts suggest that release from antifouling paints on ships is a major source of butyltins and phenyltins in Stockholm. The butyltins peaked in the city center and were slightly lower upstream (Mälaren Rotholmen) and downstream (Fjäderholmarna and Lilla Värtan).

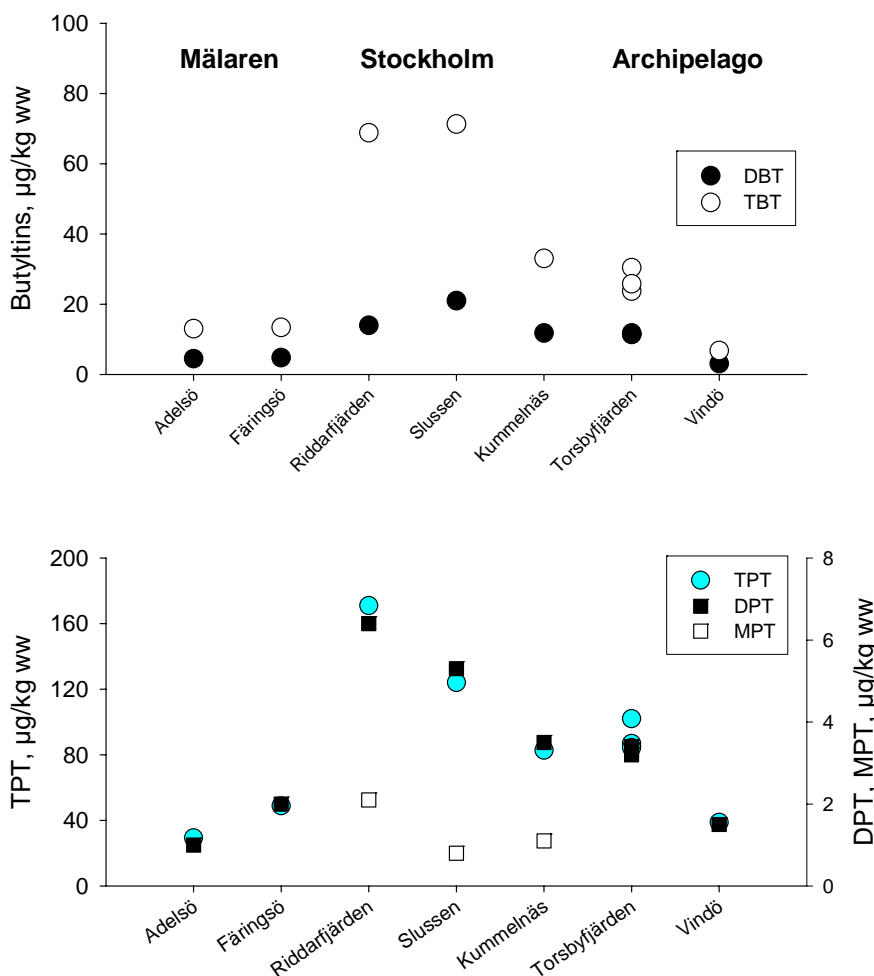
**Table 18.** Average levels of organotin compounds in surface sediments from three cities and their background sites. Because concentrations are given as a sum, the unit is ng Sn/g dw.

Urban site	no of samples	sum Butyltins	sum Octyltins	sum Phenyltins
Stockholm	5	386	2	13
Stockholm background	2	187	3	35
Eskilstuna	1	23	< 1	< 1
Eskilstuna background	1	3	< 1	< 1
Viskan	2	95	10	1
Viskan background	1	11	< 1	< 1

In order to further investigate the large-scale spatial trends in Stockholm, and to evaluate whether these pollutants were bioavailable, we analysed pooled samples of perch (*Perca fluviatilis*). Perch was collected in a gradient from Mälaren, through Stockholm and into the archipelago (Figure 3). Perch contained butyltins and phenyltins in all samples, and showed a pronounced enrichment in the city center (Figure 12). The concentrations were much higher than previous measurements in Sweden (Table 11), and also much higher than in other samples of fish that were part of the foodstuffs. Furthermore, organotin compounds were more abundant than ΣPCB (Hansson et al., 2006) and ΣDDT (Linderoth et al., 2006) in the samples. In contrast to sediments, where TPT was a relatively minor OTC compound, TPT was the major compound in perch at all sites. This suggests much stronger bioaccumulation of TPT than of TBT. The samples also contain relatively high levels of TBT metabolites but low levels of TPT metabolites, which indicate that fish metabolises TPT more slowly than TBT. Similar conclusions were recently presented from a Chinese coastal bay, where it was also shown that TPT but not TBT biomagnifies fairly strongly in the marine environment (Hu et al., 2006).



In Viskan and Eskilstunaån, DBT and MBT were more abundant than TBT. Because ships do not traffic these rivers, antifouling paint is not a likely source of OTCs at these sites. Viskan also contains octyltins at higher levels than in Stockholm. Actually, of the three urban areas in this study, only Viskan (downstream Borås city) displays octyltin levels that are elevated above the local background level. In Eskilstuna the stormwater is allowed to settle before reaching the recipient, and this may possibly explain the absence of octyltins in sediments. This pattern of organotin substances are similar to those in stormwater sludge and sewage sludge, and most probably reflects the diffuse release from products, e.g. building materials such as roof paintings. The presence of, e.g., DBT and DOT in stormwater sludge from Stockholm show that these diffuse releases also occur in Stockholm, but their impact on the aquatic environment is less obvious due to the strong influence of OTC release from ships. Compared to sediment and fish, surface water generally showed negligible spatial trends in the three urban areas.



**Figure 12.** Butyltins and phenyltins in perch from the Stockholm region. Please note the two concentration axes on the lower graph.



## 8.3. Sewage treatment plants

### 8.3.1. Occurrence in sludge and waste waters

The levels of organotin compounds in sewage sludge at Henriksdal and Gässlösa STPs displayed relatively low temporal variations (Figure 13). Furthermore, the same substances were detected at all times, i.e., MBT, DBT, TBT, MOT and DOT, with MBT and DBT as the most abundant substances. Sludge from all seven STPs contained these substances and at similar relative abundance (Table 19). This suggests a common major source of OTC's.

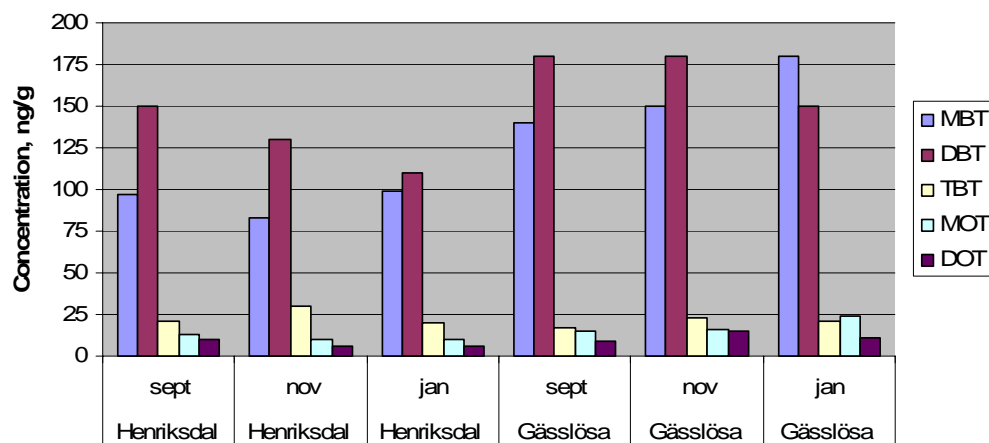
The measured concentrations of the organotin compounds in sewage sludge are in the same order of magnitude as in earlier Swedish studies (Svensson, 2002). The concentration of MBT and DBT in sewage sludge of different sewage treatment plants ranged from 100 to 350 ng/g dw (Table 19), and the total OTC levels was on the average 400-500 ng/g dw. This can be compared to the national average concentration of mercury, PAH-16 and  $\Sigma$ 7-PCB, which were 1000, 1200 and 100 ng/g respectively, in year 2000 (MI, 2002). The average level of  $\Sigma$ 5-PBDE (tetra to hexa) were 130 ng/g dw (Naturvårdsverket, 2002).

The concentrations of individual organotin compounds in sludge vary by a factor of three between different plants (Table 19). Because temporal variation appears much lower, this difference is probably significant, with highest concentrations in sludge from Fagersta and Krylbo.

The incoming wastewaters contained the same organotin compounds as the sewage sludge did (Figure 14). TBT was also detected at low concentrations in two incoming waters. The composition of the incoming wastewater differed both between months and between sewage treatment plants. MBT and DBT displayed large variations at Henriksdals sewage treatment plant during the two sampling occasions, and were anomalously low during the first sampling (Figure 14). These variations are not reflected in the sludge (Figure 13). DBT, TBT, MOT and DOT were consistently lower in effluents than in influents. In contrast, MBT was occasionally higher in the effluent, suggesting that DBT is dealkylated to MBT in the STP. On the average, 17% of DBT in influents were found in the effluents whereas there was no difference for MBT.

Consistently for all sewage treatment plants, only MBT and DBT were detectable in the effluent wastewaters, with MBT dominating in most of the cases. The absence of TBT, MOT and DOT in the effluents indicates a more efficient removal of these compounds from the incoming wastewaters, but do in part also reflect the lower levels in incoming waste waters. The removal is at least partly due to the accumulation in the sewage sludge (Table 19).





**Figure 13.** The temporal variation of organotin concentrations in sewage sludge at Henriksdal and Gässslösa sewage treatment plants.

**Table 19.** The concentration (ng/g dw) of organotin compounds detected in sewage sludge of different sewage treatment plants (STP) in september 2005.

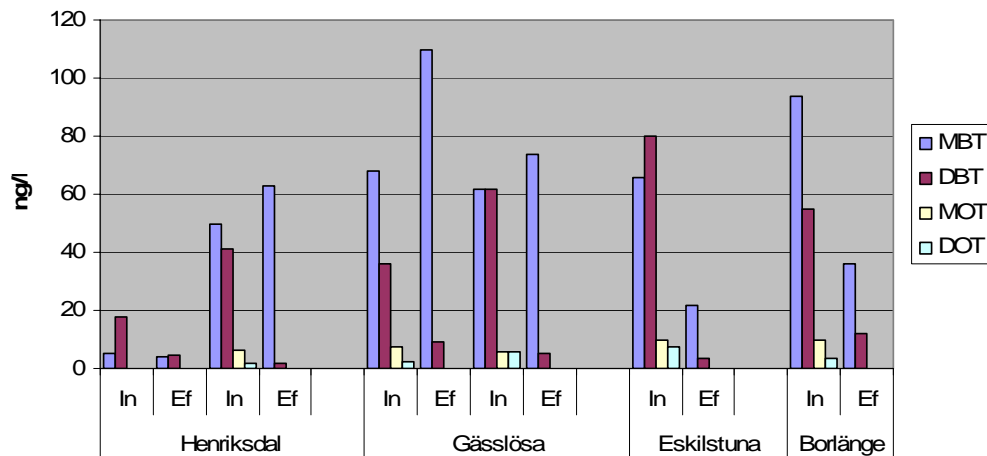
STP	Digestion	MBT	DBT	TBT	MOT	DOT
Henriksdal	+	97	150	21	13	9.9
Gässslösa	+	140	180	17	15	9
Eskilstuna	+	190	280	37	22	24
Fagersta, Borlänge	+	320	350	27	45	25
Flen	-	170	140	33	23	30
Brandholmen	+	160	200	22	15	9,6
Krylbo, Avesta	+	270	300	35	30	20
Average ± Standard dev.		190 ± 77	230 ± 81	27 ± 8	23 ± 11	20 ± 9

A tentative mass-balance was performed for the STPs in Gässslösa, Henriksdal, Fagersta and Eskilstuna. It was not always possible to achieve data on water flow and sludge production during the relevant period, why annual averages occasionally were used. Furthermore, the study period was fairly short with influents and effluents sampled during a week. The results should thus be viewed as rough estimates. Mass-balance is performed for sum of butyltins (recalculated to tin-units) and shows that the combined outflows (effluent plus sludge production) frequently were slightly higher than the inflows. This result is difficult to explain unless butyltins were added via the chemicals used in the treatment, but may be due to too short study periods or to inherent uncertainties in the sampling protocol. For instance, a large share of organotin compounds in influents is present in particulates (Baggenstoss, 2004) and it is thus difficult to get representative samples (Andersson, 2004).

As stated above, DBT is probably dealkylated to MBT in some cases, but MBT appears not to be further degraded. This is in agreement with WHO (1999) who state that DBT is more



rapidly degraded than MBT. In summary, this tentative mass-balance does not demonstrate that butyltins as a group are efficiently degraded in these STPs, but that a transformation from TBT and DBT to MBT may occur. A recent more detailed mass-balance for a Swiss STP suggested that degradation of TBT and DBT did occur during digestion, but the fate of MBT was not investigated (Baggenstoss, 2004).



**Figure 14.** The organotin compounds of incoming and effluent wastewater at four different sewage treatment plants. For Henriksdal and Gässlösa the organotin content is given for two different months. In: influent; Ef: effluent.

### 8.3.2. Sources of organotins to STPs

The levels in sludge and in influents are not necessarily reflecting how the load of OTCs vary between different STPs, because levels are influenced by dilution from stormwater, industrial waste water, the amounts of solids, mass loss during digestion etc. The daily load of organotins has therefore been calculated, based on levels in influents, and is shown in Table 20. Slightly higher values are obtained if the calculations are based on effluents and sludge. The values are quite similar to data on Swedish greywater (i.e. household waste water except the toilet fractions; Palmquist, 2004). Because the specific flows agree well, it is suggested that waste water from households is an important source of organotins to STPs. This is in line with the observations that sludge from all STPs showed similar levels and patterns of organotin compounds. Interestingly, this implies that the highly toxic TBT is present in our homes. The relative abundance of organotins in the influents and in the sludge is also similar to that in in-door dust (Santillo et al., 2001), further supporting the hypothesis that households are an important source of organotins to STPs. Furthermore, the relative abundance of OTCs in sewage sludge is very similar to that in urban stormwater sludge. This also points to a common diffuse origin.

**Table 20.** Daily loads of organotins, expressed in µg per day and population equivalent. Values are given as average and 95 % confidence interval of the mean.

	MBT	DBT	TBT	MOT	DOT
Average	28 ± 13	17 ± 13	1.6	3.1 ± 1.7	1.8 ± 1.4



### 8.3.3. Impact of STPs on surface waters

Only MBT and DBT were detectable in effluents from the seven STPs studied. MBT were on the average ca 5 times more abundant than DBT and the maximum level of MBT was 110 ng/l. MBT was generally much higher in effluents than in surface waters. Whether STP effluents may impact the levels of MBT in their recipient surface waters depends on the local dilution in the recipient. We sampled surface waters upstream and downstream of three STPs (Gässlösa, Henriksdal, Eskilstuna). A moderate increase in the downstream MBT levels could be observed at two of these sites: Viskan (Gässlösa) and Waldemarsudde (Henriksdal).

### 8.4. Point sources

The levels of MBT, DBT, MOT and DOT in storm water sludge were several orders of magnitude higher from the PVC-industry than from urban areas (Table 21). The same substances were also found in park soil within a few 100 meters from industrial site. Both the sludge and the soil samples contain butyltins and octyltins in roughly similar levels, whereas butyltins were ca 10 times more abundant than octyltins in urban stormwater sludge. This clearly shows that the occurrence of OTCs in soil was due to emissions from the industry. These observations illustrate that organotin may be released from industrial point sources, probably as dust via the ventilation. Previous measurements in stormwater from various industrial sites have also found high levels of OTCs (Junestedt et al., 2003; Table 9).

**Table 21.** Concentrations of organotin compounds (ng/g dw) in stormwater sludge from the PVC industry, and the three cities.

Study areas	MBT	DBT	TBT	MOT	DOT
PVC industry	8 400	8 700	80	12 000	12 000
PVC industry	1 100	1 500	28	1 400	740
Three cities (average ± std dev.; n=6)	77 ± 41	88 ± 53	18 ± 18	10 ± 4	7 ± 3

The three landfill leachates contained MBT and DBT in the same concentration range as found in earlier studies (Table 9). The levels are similar to those in the STP effluents. Whether landfill leachates results in an environmental impact on the surroundings has not been studied.

All the analysed organotin compounds from the sludge treated agricultural soils were under the detection level. We cannot therefore not exclude that sludge affects the concentration of OTCs in soil, but the levels are apparently low. A German study has shown an accumulation of OTCs in soil treated with sludge (LFU, 2003).

### 8.5. Brief assessment of organotin releases in Sweden

Based on the preceding chapters 8.2-8.4 and the information on how organotin compounds are used in Sweden (chapter 3), a brief assessment on the release pathways of OTCs to the Swedish environment is presented. Except for the releases from antifouling paint on ships, there is very limited knowledge on the release of organotin compounds to the environment.



The Swedish Pollutant release and transfer register (<http://www.naturvardsverket.se/kur/>) contains no data on reported emissions from industries that use these chemicals.

Apparently, TBT and TPT are still released to the aquatic environment, both marine but also to freshwaters that are connected to the sea and where ships travel. The current use of organotins in Sweden appears to result in the following principle releases:

- Industrial points sources: DBT, MBT, DOT, MOT
- Diffuse urban emissions through stormwater: DBT, MBT, DOT, MOT
- Households and industries through STPs: DBT, MBT, DOT, MOT
- Due to impure technical products, the release of DBT may include fractions of TBT.

## 8.6. Occurrence in foods and human exposure

In the food samples analysed and in the aquatic biota (perch and mussels), butyltin and phenyltin compounds were detected (Figure 15). The highest levels in the foodstuffs occurred in mussels, where MBT and DBT dominated. TPT, DPT and TBT generally dominated in fish, and phenyltins were frequently more abundant than butyltins in fish. The two salmon samples from lake Vänern contained phenyltins but no butyltins, in agreement with an earlier study (Öberg, 2002). This is somewhat unexpected for a lake, but may be due to the presence of large international ships in this lake. Six samples of salmon were analysed. The farmed Norwegian salmon was slightly lower in total OTCs than the others. Salmon from three different regions of the Baltic Sea was almost identical, and were slightly higher than those from Vänern and the farmed one.

Perch from lake Mälaren, Stockholm city and the Stockholm archipelago (Figure 12) consistently had much higher levels of OTCs than the fish foodstuffs that originated from Vänern, Vättern and the Baltic Sea. Perch from Stockholm city contained up to 70 ng/g ww TBT and 170 ng/g ww TPT. These samples of perch have previously been analysed for ΣPCB (Hansson et al., 2006) and ΣDDT (Linderoth et al., 2006). OTCs show similar spatial trends as PCB and DDT, but the levels of OTCs are even higher.

There was no relation between organotin levels and fat content. The levels in fish and mussels are close to the mean or median levels of European data (Table 12; EFSA, 2004). The meat sample contained low levels of DBT and DPT. There were no traces of octyltins, which are used in certain food packaging materials.

Pooled samples of blue mussel (*Mytilus edulis*) from four sites in Öresund were analysed. Butyltins were more abundant than TPT. There were no major differences in levels between the four sites, and the levels were lower than those mussels from the foodstuff group.

Phenyltins were frequently more abundant than butyltins in fish, particularly in perch, whereas the mussels contained more butyltins than phenyltins. This pattern is most likely due to the fact that TPT but not TBT biomagnifies in the aquatic food chain (chapter 2.2).

The only species found in human breast milk was MBT. This does not match the pattern of organotins in foods, where MBT generally is a minor species. This discrepancy may either be explained by human metabolism of other organotins, or that other than dietary sources of organotin exposure exist. Earlier studies on human blood found both MBT, DBT and TBT, with MBT as the major species (Kannan et al., 1999). The levels in breast milk ranged from 1-10 ng/l, which is about 0.03-0.3 ng/g fat. Compared to other recent measurements on Swedish human breast milk, the concentration of MBT is lower than those of e.g. PCB, DDT and PBDE (Aune et al., 2002).

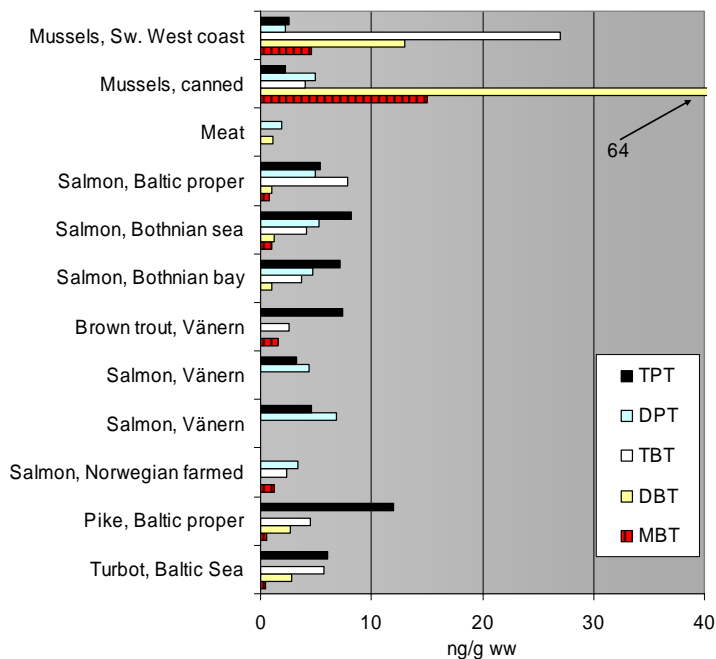


Figure 15. Organotins in foodstuffs.

### 8.7. Risks för health and the environment

It is difficult to assess the risk to health or the environment from this limited dataset. Furthermore, environmental quality standards are only available for a few of the OTCs that we investigated, and mainly for water. Therefore, only a very small part of the data presented can be assessed for risks. One should also note that several of the compounds have been proposed as PBT or vPvB compounds. Releases of such substances should always be kept to a minimum, regardless of the present levels.

However, we can note that TBT was present in all surface waters from Stockholm at levels above the proposed European EQS value for transient conditions (1.5 ng/l). There is also an EQS for annual average proposed (0.1 ng/l), and this value is lower than the analytical reporting limit in this study, why we cannot exclude that TBT presents a risk also in other freshwaters. The single guidevalue for DBT that we found were higher than the concentrations measured in surface waters. Considering that DBT is more toxic than TBT for certain endpoints and that DBT is the most abundant OTC in this study, it still seems that the omnipresence of DBT deserves more attention.

For the health assessment, a group TDI of 0.1 µg Sn/kg bw/d for the sum of DBT, DOT, TBT and TPT was proposed, based on immunotoxic effects (see RPA, 2005). This sum was in the range 1-94 ng Sn/g ww for all foodstuffs and fish that were investigated. An adult person of 50 kg can thus eat at least 50 gram fresh fish per day. The estimate is valid for the most contaminated perch from Stockholm city. This assessment is limited in the sense that no long-term epidemiological studies are available for assessing health risks of organotins, that TPT is classified as having limited evidence for carcinogenic effects and possible risk of harm to the unborn child, and that the current dataset is limited. Neither are synergistic effects with other pollutants considered. It thus seems that the presence of organotins in fish from other navigable channels, and in other fish species, deserves more attention.

L:\3655\2005\10060857 Screening av tennorganiska ämnen\C-Genomförande\23-Skisser-Utkast\tem-rapport-rev\_060630.doc



## 9. Conclusions

This study verifies that organotin compounds are present in Swedish inland waters. The following general conclusions can be drawn.

- ❑ Organotins are present in urban background sites.
- ❑ The current use of butyltin and octyltin in plastics and other applications causes diffuse release in urban areas.
- ❑ STPs appears mainly to be influenced by diffuse releases, and their effluents may influence the levels of MBT in their recipients.
- ❑ TBT and DBT are dealkylated to MBT in STPs.
- ❑ Butyltin and octyltins may be released from industries where these substances are handled.
- ❑ Butyltins were found in all areas and all medias that were investigated, and DBT and MBT were the most abundant substances.
- ❑ Octyltins were mainly found in waste water, sewage sludge, stormwater sludge and sediments, but never in biological specimen.
- ❑ Phenyltins were found in both the lacustrine and marine environment, mainly in areas where international ships travel, and accumulated strongly in fish.
- ❑ Stockholm was the most contaminated region in this study.
- ❑ Organotin compounds were more abundant than PCB and DDT in perch from Stockholm.
- ❑ Fish and other foodstuffs contained butyltins and phenyltins.
- ❑ Fairly low levels of MBT were found in human breast milk.
- ❑ A tentative risk assessment shows that the levels of organotins are close to or above acceptable levels for health and the environment in certain areas.

## 10. Acknowledgements

We wish to thank the following persons who assisted in various ways:

Morten Christensen and Maria Bigner (Analytica AB); Kierstin Pettersson-Grawe, Per Ola Darnerud and Marie Aune (Livsmedelsverket); Anita Lundmark (Dalarna county administration board); Sofi Nordfeldt (Södermanland county administration board); Fredrik Andreasson (Skåne county administration board); Kjell Westermarck (Eskilstuna municipality); Lenart Balk and Marsha Hanson (ITM Stockholm University); Thomas Terne (WSP), Rolf Wolfe, Anette Ohlsson, Pernilla Norwald, Lotta Edström, Britt Louise Hultman, Barbro Andersson nad Agneta Bergman at the sewage treatment plants.

The study was financed by the Swedish Environmental Protection Agency.



## 11. References

- Allsopp M., Santillo D. and Johnston P. (2000) Hazardous Chemicals in PVC Flooring. Greenpeace Research Laboratories Technical Note, N: 14/00, November 29th 2000.
- Appel K.E. (2004) Organotin Compounds: Toxicokinetic Aspects. *Drug metabolism reviews*, 36, 763–786.
- Andersson Å. (2004) Bromstens industriområde - Inventering av industriella verksamheter samt mätning av spillvattenkvalitet år 2004. Stockholm Vatten, rapport.
- Aune M, Barregård L, Claesson A. and Darnerud P.O. (2002) Organiska miljögifter i bröstmjolk från Göteborg 2001. Livsmedelsverket, rapport till Naturvårdsverket.
- Baggenstoss J. (2004) The fate of organotin compounds in a waste water treatment plant. Diploma Thesis, Ecole Polytechnique Federale de Lausanne.
- Berg M., Arnold C.G., Muller S.R., Muhlemann J. and Schwarzenbach R.P. (2001) Sorption and desorption behavior of organotin compounds in sediment-pore water systems. *Environ. Sci. Technol.* 35, 3151-3157.
- BKH (2000) Towards the establishment of a priority list of substances for further evaluation of their role in endocrine disruption. Report for the European Commission.
- Borghi V. and Porte C (2002) Organotin pollution in deep-sea fish from the northwestern Mediterranean. *Environ. Sci. Technol.* 36, 4224-4228.
- Cato I. (2003) Organotin compounds in Swedish sediments – an overlooked environmental problem. Swedish Geological Survey, report 2003:4, 6-8.
- Ciesielski, T.; Wasik, A.; Kuklik, I.; Skora, K.; Namiesnik, J.; Szefer, P (2004) Organotin Compounds in the Liver Tissue of Marine Mammals from the Polish Coast of the Baltic Sea. *Environ. Sci. Technol.* 38, 1415-1420.
- EFSA (2004) Opinion of the Scientific Panel on Contaminants in the Food Chain on a request from the Commission to assess the health risks to consumers associated with exposure to organotins in foodstuffs. *The EFSA Journal* (2004) 102, 1-119.
- Gies A. (2003) Government view of endocrine disruption in wildlife. *Pure Appl. Chem.* 75, 2563–2574.
- Hajjaj el Hassani L. et al. (2005) Study of the accumulation of tributyltin and triphenyltin compounds and their main metabolites in the sea bass, *Dicentrarchus labrax*, under laboratory conditions. *Sci. Tot. Environ.* 348, 191– 198.
- Hansson T., Schiedek D., Lehtonen K.K., Vuorinen P.J., Liewenborg B., Noaksson E., Tjärnlund U., Hanson M. and Balk L. (2006) Biochemical biomarkers in adult female perch (*Perca fluviatilis*) in a chronically polluted gradient in the Stockholm recipient (Sweden). *Mar. Poll. Bull.* In press.
- Hirose A., Takagi A., Nishimura T., Kanno J. and Ema M. (2004) Review of reproductive and developmental toxicity induced by organotins in aquatic organisms and experimental animals. *Organohalogen compounds*, 66, 3042-3047.
- Hu J et al., (2006) Trophic magnification of triphenyltin in a marine food web of Bohai bay, north China: comparison to tributyltin. *Environ. Sci. Tech.* 40, 3142-3147.
- Huang J.H. and Metzner E. (2004a) Adsorption and desorption of organotin compounds in organic and mineral soils. *Eur. J. Soil Sci.* 55, 693–698.





- Huang J.H. and Metzner E. (2004b) Degradation of organotin compounds in organic and mineral forest soils. *J. Plant Nutr. Soil Sci.* 167, 33-38.
- Huang J.H. and Klemm O. (2004) Atmospheric speciation of ionic organotin, organolead and organomercury compounds in NE Bavaria. *Atmos. Environ.* 38, 5013–5023.
- Huang J.H., Schwesig D., Matzner E. (2004) Organotin compounds in precipitation, fog and soils of a forested ecosystem in Germany. *Environmental Pollution* 130, 177-186.
- Junestedt C., Solyom P., Ek M., Palm A., Öman C. (2003) Karakterisering av utsläpp - jämförelse av olika utsläpp till vatten. IVL B1544.
- Kannan, K., Senthilkumar, K., Giesy, J. P. (1999). Occurrence of butyl compounds in human blood. *Environ. Sci. Technol.* 33:1776–1779.
- Linderoth M., Hansson T., Liewenborg B., Sundberg H., Noaksson E., Hanson M., Zebuhr Y. and Balk L (2006) Basic physiological biomarkers in adult female perch (*Perca fluviatilis*) in a chronically polluted gradient in the Stockholm recipient (Sweden). *Mar. Poll. Bull.*, in press.
- LFU (2003) Contaminants in arable soils in Baden-Württemberg fertilised with sewage sludge - Concise Report. Landesanstalt für Umweltschutz, Baden-Württemberg, Bodenschutz 16.
- Mester Z. and Sturgeon R. E (2002) Detection of volatile organometal chloride species in model atmosphere above seawater and sediment. *Environ. Sci. Technol.* 36, 1198-1201.
- MI (2002) Utsläpp till vatten och slamproduktion 2000. MI 22 SM 0101. Naturvårdsverket och SCB.
- MST (2001) Phthalates and organic tin compounds in PVC products. Survey No 1. Miljöstyrelsen i Danmark.
- MST (2003) Survey no. 28- 2003. Mapping of chemical substances in earplugs Phase 2: Analysis of substances
- Miljö-Kemi (2000) Analyse af forbrugerprodukter. Miljöstyrelsen i Danmark. Uppdrag 101655-72-67.
- Naturvårdsverket (2002) Bromerade flamskyddsmedel i avloppsslam. Rapport 5188 (in Swedish).
- Palmquist H. (2004) Hazardous substances in wastewater management. Doctoral Thesis, Luleå University.
- RPA (2005) Risk assessment studies on targeted consumer applications of certain organotin compounds. Report for the European Commission. Risk & Policy Analysts Limited, (RPA), UK.
- Santillo D., Johnston P. and Brigden K. (2001) The presence of brominated flame retardants and organotin compounds in dusts collected from Parliament buildings from eight countries. Greenpeace Research Laboratories. Technical Note 03/2001.
- Shimasaki Y. et al. (2003) Tributyltin causes masculinization in fish. *Environ. Tox. Chem.* 22, 141-144.
- Sternbeck J. Brorström-Lundén E., Remberger M., Kaj L., Palm A., Junedahl E. and Ingemar Cato. WFD Priority substances in sediments from Stockholm and the Svealand coastal region. IVL report B1538.





Strand J., Larsen M.M., Naes K., Cato I. and Dahhlöf I. (2006) Tributyltin, forekomst og effekter i Skagerrak. Forum Skagerrak II ([www.forumskagerrak.com](http://www.forumskagerrak.com)).

Svensson A. (2002) Miljögifter i avloppsslam. Länsstyrelsen Västra Götaland, Rapport 2002:39.

Sveriges Byggindustrier (2000) Hållbara val – färg, fog, lim.

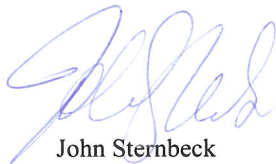
Tesfalidet S. (2003) Screening of organotin compounds in the Swedish marine environment. Analytical Chemistry, Umeå University. April 2003.

WHO (1999) Tributyltin oxide. Concise International Chemical Assessment Document 14.

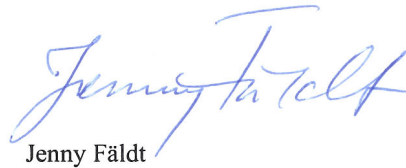
Öberg T. (2003) Miljögifter i fisk 2001/2002. Vänerns Vattenvårdsförbund, rapport nr 25

Stockholm 2006-06-30

WSP Environmental



John Sternbeck



Jenny Fäldt



**Appendix 1.** The Swedish use of organotin compounds in different applications, according to data in SPIN. The amounts are tonnes of Sn. Because of confidentiality, the amounts of DBT are lower here than in table 4.

<b>Application</b>	<b>1999</b>	<b>2000</b>	<b>2001</b>	<b>2002</b>	<b>2003</b>
<b>Monobutyltin compounds</b>					
Stabilizers	0.89	0.60	0.15		
<i>Total</i>	<i>0.89</i>	<i>0.60</i>	<i>0.15</i>		
<b>Dibutyltin compounds</b>					
Adhesives, glues	0.19	0.19	0.19		
Catalysts	4.9	0.38	0.38	2.8	0.38
Caulking compounds	0.19	0.19	0.19		
Intermediates (plastics manufacture)		0.19			
Other stabilizers				14.6	16.8
Padding (filling) materials				1.1	0.66
Sealing compounds	0.38	0.19	0.19		
Stabilizers	5.4	4.1	1.7		
<i>Total</i>	<i>11</i>	<i>5.2</i>	<i>2.6</i>	<i>19</i>	<i>18</i>
<b>Tributyltin compounds</b>					
Antifoulant paints	0.75	1.0	0.25		
<i>Total</i>	<i>0.75</i>	<i>1.0</i>	<i>0.25</i>		
<b>Ethyltin compounds</b>					
Catalysts	2.6	1.2	3.5	7.3	5.6
<i>Total</i>	<i>2.6</i>	<i>1.2</i>	<i>3.5</i>	<i>7.3</i>	<i>5.6</i>
<b>Octyltin compounds</b>					
Stabilizers		17.7	17.2	14.3	16.6
<i>Total</i>		<i>17.7</i>	<i>17.2</i>	<i>14.3</i>	<i>16.6</i>



**Appendix 2. Sample details.** This appendix shows sample details for all samples. The corresponding analytical results are shown in appendix 3.

Sample nr	Programme	Study site / sample	Media	Category	Sampling details	Sampling date	Comments	X RT90	Y RT90
WSP_60857_1	National	Henriksdal STP	STP, incoming waste water	STP	pooled sample	3-7 sept. 2005			
WSP_60857_2	National	Henriksdal STP	STP, incoming waste water	STP	pooled sample	3-7 sept. 2005			
WSP_60857_3	National	Henriksdal STP	STP effluent	STP	pooled sample	3-7 sept. 2005			
WSP_60857_4	National	Henriksdal STP	STP effluent	STP	pooled sample	30 jan-3 feb, 2006			
WSP_60857_5	National	Henriksdal STP	sewage sludge	STP	pooled sample	3-7 sept. 2005			
WSP_60857_6	National	Henriksdal STP	sewage sludge	STP	pooled sample	november, 2005			
WSP_60857_7	National	Henriksdal STP	sewage sludge	STP	pooled sample	30 jan-3 feb, 2006			
WSP_60857_8	National	Mälaren Rotholmen	surface water	Urban	duplicate sample	3/9 2005	0-4 cm, ca 28 m depth	6579255	1623652
WSP_60857_9	National	Mälaren Rotholmen	surface water	Urban	duplicate sample	3/9 2005	0-4 cm, ca 28 m depth	6579255	1623652
WSP_60857_10	National	Mälaren Rotholmen	sediment	Urban	duplicate sample	3/9 2005	0-4 cm, ca 28 m depth	6579255	1623652
WSP_60857_11	National	Mälaren Rotholmen	sediment	Urban	duplicate sample	3/9 2005	0-4 cm, ca 28 m depth	6579255	1623652
WSP_60857_12	National	Årstaviken, 1	surface water	Urban	duplicate sample	3/9 2005	0-4 cm, ca 7m depth	6578637	1627117
WSP_60857_13	National	Årstaviken, 2	surface water	Urban	duplicate sample	3/9 2005	0-4 cm, ca 7m depth	6578637	1627117
WSP_60857_14	National	Årstaviken, 1	sediment	Urban	duplicate sample	3/9 2005	0-4 cm, ca 7m depth	6578637	1627117
WSP_60857_15	National	Årstaviken, 2	sediment	Urban	duplicate sample	3/9 2005	0-4 cm, ca 7m depth	6578637	1627117
WSP_60857_17	National	Lilla Värtan	sediment	Urban		4/9 2005	0-4 cm, 24 m depth	6581780	1633924
WSP_60857_18	National	Ulvsundasjön, Sthlm	sediment	Urban		3/9 2005	0-4 cm, 14 m depth	6582278	1624438
WSP_60857_19	National	Fjäderholmarna	sediment	Urban		4/9 2005	0-4 cm, 28 m depth	6580622	1634298
WSP_60857_20	National	Ulvsundasjön, Sthlm	surface water	Urban		3/9 2005	0-4 cm, 14 m depth	6582278	1624438
WSP_60857_21	National	Fjäderholmarna	surface water	Urban		4/9 2005	0-4 cm, 28 m depth	6580622	1634298
WSP_60857_23	National	Stockholm, stormwater sludge	Stormwater sludge	Urban		27/10 05	Storage site for stormwater sludge from Stockholm city (Fruängen)		
WSP_60857_24	National	Stockholm, traffic storm-water retention pond	Stormwater sludge	Urban		25/11-05	Ryska Smällen, Johanneshovsbron		



## Appendix 2. continued..

Sample nr	Programme	Study site / sample	Media	Category	Sampling details	Sampling date	Comments	X RT90	Y RT90
WSP_60857_25	National	Eskilstuna stormwater gutter	Stormwater sludge	Urban		15-nov-05	Hamngatan, city center		
WSP_60857_26	National	Eskilstuna stormwater gutter	Stormwater sludge	Urban		15-nov-05	Hamngatan, city center		
WSP_60857_27	National	Eskilstuna upstream, Hyndevad	sediment	Urban background		15-nov-05	0-2 cm, 2 m depth	6578038	1536361
WSP_60857_28	National	Eskilstuna, central, Närjeholmsv.	sediment	Urban		15-nov-05	0-2 cm, 7 m depth	6585040	1538769
WSP_60857_31	National	Eskilstuna, central, Närjeholmsv.	surface water	Urban		15-nov-05		6585040	1538769
WSP_60857_32	National	Eskilstuna downstream, Torshälla	surface water	Urban, STP recipient		15-nov-05	downstream the STP	6588735	1537751
WSP_60857_34	National	Eskilstuna upstream, Hyndevad	surface water	Urban background		15-nov-05		6578038	1536361
WSP_60857_35	National	Gässlösa STP	STP, incoming waste water	STP	pooled sample	3-7 sept. 2005			
WSP_60857_36	National	Gässlösa STP	STP, incoming waste water	STP	pooled sample	060130-060203			
WSP_60857_37	National	Gässlösa STP	STP effluent	STP	pooled sample	3-7 sept. 2005			
WSP_60857_38	National	Gässlösa STP	STP effluent	STP	pooled sample	060130-060203			
WSP_60857_39	National	Gässlösa STP	sewage sludge	STP	pooled sample	3-7 sept. 2005			
WSP_60857_40	National	Gässlösa STP	sewage sludge	STP	pooled sample	november 2005			
WSP_60857_41	National	Gässlösa STP	sewage sludge	Urban	pooled sample	03-feb-06			
WSP_60857_42	National	Viskan (Öresjö)	surface water	Urban background		03-okt-05		640995	133150
WSP_60857_43	National	Viskan (Druvefors)	surface water	Urban		03-okt-05		640217	132909
WSP_60857_44	National	Viskan (Djupasjön)	surface water	Urban, STP recipient		03-okt-05		640045	132730
WSP_60857_45	National	Viskan (Öresjö)	sediment	Urban	pooled sample	03-okt-05	0-2 cm; 29 m depth	640995	133150
WSP_60857_46	National	Viskan (Djupasjön)	sediment	Urban	pooled sample	03-okt-05	0-2 cm; 5 m depth	640045	132730
WSP_60857_47	National	Viskan (Guttasjön)	sediment	Urban	pooled sample	03-okt-05	0-2 cm; 4 m depth	639818	132685



## Appendix 2. continued..

Sample nr	Programme	Study site / sample	Media	Category	Sampling details	Sampling date	Comments	X RT90	Y RT90
WSP_60857_48	Regional, Nyköping	Brandholmen STP	STP effluent	STP	pooled sample	3-7 sept. 2005			
WSP_60857_49	Regional, Nyköping	Brandholmen STP	sewage sludge	STP	pooled sample	3-7 sept. 2005			
WSP_60857_50	Regional, Nyköping	Östra mellanfjärden	Sediment			september 2005	Recipient to Brandholmen STP	6513908	1573804
WSP_60857_51	Regional, Nyköping	Örsbaken	Sediment			september 2005	Background site to Brandholmen	6511581	1576742
WSP_60857_52	Regional, Nyköping	Eskilstuna STP	STP, incoming waste water		pooled sample	10-14 sept 2005			
WSP_60857_53	Regional, Nyköping	Eskilstuna STP	STP effluent		pooled sample	10-14 sept 2005			
WSP_60857_54	Regional, Nyköping	Eskilstuna STP	sewage sludge		pooled sample	10-14 sept 2005			
WSP_60857_55	Regional, Nyköping	Eskilstuna STP	STP effluent, downstream a wetland		pooled sample	10-14 sept 2005			
WSP_60857_56	Regional, Nyköping	Flen STP	STP effluent		pooled sample	3-7 sept. 2005			
WSP_60857_57	Regional, Nyköping	Flen STP	sewage sludge		pooled sample	3-7 sept. 2005			
WSP_60857_58	Regional, Dalarna	Fagersta STP, Borlänge	STP, incoming waste water		pooled sample	3-7 sept. 2005			
WSP_60857_59	Regional, Dalarna	Fagersta STP, Borlänge	STP effluent		pooled sample	3-7 sept. 2005			
WSP_60857_60	Regional, Dalarna	Fagersta STP, Borlänge	sewage sludge		pooled sample	3-7 sept. 2005			
WSP_60857_61	Regional, Dalarna	Krylbo STP, Avesta	STP effluent		pooled sample	3-7 sept. 2005			
WSP_60857_62	Regional, Dalarna	Krylbo STP, Avesta	sewage sludge		pooled sample	05-okt 2005			
WSP_60857_63	Regional, Dalarna	River Bäringen, Dalälven	surface water			september, 2005	Recipient to Krylbo STP		
WSP_60857_64	Regional, Dalarna	River Bäringen, Dalälven	sediment		pooled sample	september, 2005	Recipient to Krylbo STP 0-2 cm, 11-12m depth, sandy sediment		
WSP_60857_65	Regional, Dalarna	Fågelmäyra landfill	Landfill leachate		pooled sample	september, 2005			
WSP_60857_66	Regional, Nyköping	Eskilstuna landfill	Landfill leachate		pooled sample	september 2005			
WSP_60857_68	Regional, Nyköping	Björshult landfill	Landfill leachate		pooled sample	september, 2005			
WSP_60857_72	National	Svedäng, agricultural land-use, sludge treated	soil		pooled sample	november 2005	Sludge treated agricultural soil (0-2 cm)		
WSP_60857_73	National	Svedäng, agricultural land-use, sludge treated	soil		pooled sample	november 2005	Sludge treated agricultural soil (0-2 cm)		



## Appendix 2. continued..

Sample nr	Programme	Study site / sample	Media	Sampling details	Sampling date	Comments	X RT90	Y RT90
WSP_60857_74	National	Svedäng, agricultural land-use	soil	pooled sample	nov-05	Reference agricultural soil (0-2 cm)		
WSP_60857_75	National	Svedäng, agricultural land-use	soil	pooled sample	nov-05	Reference agricultural soil (0-2 cm)		
WSP_60857_76	National	Svedäng stream	surface water		nov-05	Svedäng sludge treated		
WSP_60857_77	National	Svedäng stream	surface water		nov-05	Svedäng control		
WSP_60857_78	Regional, Skåne	Öresund 1	Mussel ( <i>Mytilus edulis</i> )	pooled sample	2005	ÖVF 1:5 Domsten mussels		
WSP_60857_79	Regional, Skåne	Öresund 2	Mussel ( <i>Mytilus edulis</i> )	pooled sample	2005	ÖVF 3:5 Landskrona mussels		
WSP_60857_80	Regional, Skåne	Öresund 3	Mussel ( <i>Mytilus edulis</i> )	pooled sample	2005	ÖVF 4:12 Spillepeng mussels		
WSP_60857_81	Regional, Skåne	Öresund 4	Mussel ( <i>Mytilus edulis</i> )	pooled sample	2005	ÖVF 5:5 Klagshamn mussels		
WSP_60857_82	National	Foods 1	Turbot Baltic Sea	pooled sample	2002	Muscle, FF20020509		
WSP_60857_83	National	Foods 2	Pike, Baltic Sea	pooled sample	2002	Muscle, FISK2 C414		
WSP_60857_84	National	Foods 3	Salmon, farmed in Norway	pooled sample	2001	Muscle, FF20010030		
WSP_60857_85	National	Foods 4	Salmon Vänern	pooled sample	2003	Muscle, FF20030077		
WSP_60857_86	National	Foods 5	Salmon Vänern	pooled sample	2003	Muscle, FF20030011		
WSP_60857_87	National	Foods 6	Brown trout Vänern	pooled sample	2001	Muscle, FF20010193		
WSP_60857_88	National	Foods 7	Salmon Bothnian bay	pooled sample	2002	Muscle, FF20020335		
WSP_60857_89	National	Foods 8	Salmon Bothnian sea	pooled sample	2002	Muscle, FF20020436		
WSP_60857_90	National	Foods 9	Salmon, Baltic proper	pooled sample	2002	Muscle, FF20020407		
WSP_60857_91	National	Foods 10	Meat, pooled of various animals	pooled sample	2005	Slurry 3 S:I 2005		
WSP_60857_92	National	Foods 11	Mussels, canned	pooled sample	2005	Abba från M:II 2005		
WSP_60857_93	National	Foods 12	Mussels Swed. west coast	pooled sample	2001	X226 dnr2925/01		
WSP_60857_94	National	Uppsala 2002	Human breast milk	pooled sample	2002	HF20030032		
WSP_60857_95	National	Uppsala 2004	Human breast milk	pooled sample	2004	H0500111		
WSP_60857_96	National	Göteborg 2001	Human breast milk	pooled sample	2001	HF20030031		
WSP_60857_97	National	Lund 2003	Human breast milk	pooled sample	2003	HF20040042		
WSP_60857_98	National	Lycksele 2004	Human breast milk	pooled sample	2004	HF20040054		



## Appendix 2. continued..

Sample nr	Programme	Study site / sample	Media	Category	Sampling details	Sampling date	Comments	X RT90	Y RT90
WSP_60857_99	National	Waldemarsudde	surface water, 1 m depth	STP, urban		04-sep-05	local reference	6579784	1631142
WSP_60857_100	National	Waldemarsudde	surface water 10.-16 m depth	STP, urban	pooled sample	04-sep-05	intermediate depth, where STP effluents are intermixed	6579784	1631142
WSP_60857_101	National	Waldemarsudde	surface water ca 21 m depth	STP, urban		04-sep-05	bottom water, at the depth where STP effluents are released	6579784	1631142
WSP_60857_102	National	Hydropolymer, PVC-prod. site East of industrial building	soil	Point source	Pooled from 4 individual samples	17-mar-06			
WSP_60857_103	National	Hydropolymer PVC-prod. site. North of industrial building	soil	Point source	Pooled from 4 individual samples	17-mar-06			
WSP_60857_104	National	Hydropolymer PVC-prod. site. stormwater gutter, south	Stormwater sludge	Point source		17-mar-06			
WSP_60857_105	National	Hydropolymer PVC-prod. site. stormwater gutter, north	Stormwater sludge	Point source		17-mar-06			
WSP_60857_106	National	Helsingborg, stormwater retention pond, Stenbrogården	Stormwater sludge	Urban		17-mar-06			
WSP_60857_107	National	Helsingborg, stormwater retention pond, Regnbågsdamm	Stormwater sludge	Urban		17-mar-06			
WSP_60857_108	National	Adelsö, Mälaren	Perch	Lake	pooled sample	2000 / 2001	Muscle	6584399	1594118
WSP_60857_109	National	Färingsö, Mälaren	Perch	Lake	pooled sample	2000 / 2001	Muscle	6587920	1610058
WSP_60857_110	National	Riddarfjärden, Stockholm	Perch	Urban	pooled sample	2000 / 2001	Muscle	6580200	1627260
WSP_60857_111	National	Slussen, Stockholm	Perch	Urban	pooled sample	2000 / 2001	Muscle	6579800	1630200
WSP_60857_112	National	Kummelnäs	Perch	Coastal	pooled sample	2000 / 2001	Muscle	6583790	1639290
WSP_60857_113	National	Torsbyfjärden	Perch	Coastal	pooled sample	2000 / 2001	Muscle	6583923	1650300
WSP_60857_114	National	Torsbyfjärden	Perch	Coastal	pooled sample	2000 / 2001	Muscle	6583923	1650300
WSP_60857_115	National	Torsbyfjärden	Perch	Coastal	pooled sample	2000 / 2001	Muscle	6583923	1650300
WSP_60857_116	National	Vindö	Perch	Coastal	pooled sample	2000 / 2001	Muscle	6588500	1660380

### Appendix 3. Analytical data

Table A1. Concentrations in soil, stormwater sludge and sewage sludge.

Site	Category	Media	Sample nr.	DW, %	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT
Hydropolymer East	Industry	Soil	WSP_60857_102		30	58	<1,0	<1,0	25	27	<1,0	<1,0	<1,0	<1,0
Hydropolymer north	Industry	Soil	WSP_60857_103		6	15	<1,0	<1,0	5.4	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng sludge treated salix field		Soil	WSP_60857_72	74.9	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng sludge treated salix field		Soil	WSP_60857_73	72.5	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng untreated field (control)		Soil	WSP_60857_74	76.7	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng untreated field (control)		Soil	WSP_60857_75	75.6	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna	Urban	Stormwater sludge	WSP_60857_25	33.2	53	54	2.3	<1,0	8.2	3.5	<1,0	<1,0	<1,0	<1,0
Eskilstuna	Urban	Stormwater sludge	WSP_60857_26	46	64	56	2.8	<1,0	8	3.9	<1,0	<1,0	<1,0	<1,0
Helsingborg, Stenbrogården	Urban	Stormwater sludge	WSP_60857_106		140	130	41	2	16	10	<1,0	<1,0	<1,0	<1,0
Helsingborg, Regnbågsdamm	Urban	Stormwater sludge	WSP_60857_107		83	150	36	3.2	6.9	6.2	<1,0	<1,0	<1,0	25
Hydropolymer, Dagvattenbrunn södra	Industry	Stormwater sludge	WSP_60857_104		8400	8700	80	<1,0	12000	12000	<1,0	<1,0	29	<1,0
Hydropolymer, Dagvattenbrunn norra	Industry	Stormwater sludge	WSP_60857_105		1100	1500	28	8.4	1400	740	<1,0	<1,0	<1,0	<1,0
Stockholm, Johanneshovsbron, Ryska smällen	Urban	Stormwater sludge	WSP_60857_24	50.5	100	120	22	<1,0	11	9.7	<1,0	<1,0	<1,0	<1,0
Stockholm, pooled from many sites	Urban	Stormwater sludge	WSP_60857_23	41.9	21	17	2.1	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Fagersta-STP (Borlänge)	STP	Sewage sludge	WSP_60857_60	26	320	350	27	<1,0	45	25	<1,0	<1,0	<1,0	<1,0
Brandholmen STP	STP	Sewage sludge	WSP_60857_49	26.1	160	200	22	<1,0	15	9.6	<1,0	<1,0	<1,0	<1,0
Eskilstuna STP	STP	Sewage sludge	WSP_60857_54	31.3	190	280	37	<1,0	22	24	<1,0	<1,0	<1,0	<1,0
Flen STP	STP	Sewage sludge	WSP_60857_57	11.9	170	140	33	<1,0	23	30	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Sewage sludge	WSP_60857_39	24.8	140	180	17	<1,0	15	9	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Sewage sludge	WSP_60857_40	19.6	150	180	23	<1,0	16	15	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Sewage sludge	WSP_60857_41	20.4	180	150	21	<1,0	24	11	<1,0	<1,0	<1,0	<1,0
Henriksdals STP	STP	Sewage sludge	WSP_60857_5	26.2	97	150	21	<1,0	13	9.9	<1,0	<1,0	<1,0	<1,0
Henriksdals STP	STP	Sewage sludge	WSP_60857_6	26.6	83	130	30	<1,0	9.7	6.5	<1,0	<1,0	<1,0	<1,0
Henriksdals STP	STP	Sewage sludge	WSP_60857_7	26.1	99	110	20	<1,0	9.6	6.2	<1,0	<1,0	<1,0	<1,0
Krylbo STP	STP	Sewage sludge	WSP_60857_62	25.6	270	300	35	<1,0	30	20	<1,0	<1,0	<1,0	<1,0





Table A2. Concentrations in municipal waste waters and landfill leachates (all data in ng/l).

Site	Category	Media	Sample nr	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT
Fagersta STP	STP	Incoming waste water	WSP_60857_58	94	55	<1,0	<1,0	9.5	3.2	<1,0	<1,0	<1,0	<1,0
Eskilstuna STP	STP	Incoming waste water	WSP_60857_52	66	80	4.1	<1,0	9.8	7.3	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Incoming waste water	WSP_60857_35	68	36	<1,0	<1,0	7.5	2.5	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Incoming waste water	WSP_60857_36	62	62	2.5	<1,0	5.8	5.8	<1,0	<1,0	<1,0	<1,0
Henriksdal STP	STP	Incoming waste water	WSP_60857_1	5.2	18	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Henriksdal STP	STP	Incoming waste water	WSP_60857_2	50	41	<1,0	<1,0	6.4	1.9	<1,0	<1,0	<1,0	<1,0
Fagersta STP	STP	Effluent	WSP_60857_59	36	12	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Brandholmen STP	STP	Effluent	WSP_60857_48	4.2	10	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna STP	STP	Effluent	WSP_60857_53	22	3.4	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna STP	STP	Effluent, after wetland	WSP_60857_55	23	5.8	<1,0	<1,0	7.8	12	<1,0	<1,0	<1,0	<1,0
Flen STP	STP	Effluent	WSP_60857_56	39	13	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Effluent	WSP_60857_37	110	9.3	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Gässlösa STP	STP	Effluent	WSP_60857_38	74	5.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Henriksdal STP	STP	Effluent	WSP_60857_3	4	4.7	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Henriksdal STP	STP	Effluent	WSP_60857_4	63	1.7	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Krylbo STP, Avesta	STP	Effluent	WSP_60857_61	78	11	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Björshult landfill	Landfill	Leachate	WSP_60857_68	56	16	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Borlänge landfill	Landfill	Leachate	WSP_60857_65	11	19	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna landfill	Landfill	Leachate	WSP_60857_66	6.7	6.4	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0



Table A3. Concentrations in surface sediments (all data in ng/g dw).

Site	Category	Sample nr	TS, %	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT	TOC, %
Bäsingen, Dalälven	Recipient to STP	WSP_60857_64	21.6	10	2.50	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	4.00
Eskilstuna upstream, Hyndevad	Urban background	WSP_60857_27	33	2.8	1.1	1.7	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	
Eskilstuna, central, Närjeholmsv.	Urban	WSP_60857_28	23.3	21	17	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	
Rotholmen 1, Mälaren, Stockholm	Urban background	WSP_60857_10	14.9	74	84	380	2.70	5.50	3.70	<1,0	45	66	12	7.10
Rotholmen 2, Mälaren, Stockholm	Urban background	WSP_60857_11	14.5	59	46	150	<1,0	2.70	2.80	<1,0	6.90	8.90	4.50	7.30
Årstaviken 1, Stockholm	Urban	WSP_60857_14	21.4	160	250	590	2.10	3.10	2.40	<1,0	9.20	12	5	7.10
Årstaviken 2, Stockholm	Urban	WSP_60857_15	22	140	300	880	3	2.60	2.80	<1,0	12	13	7.50	6.80
Ulvsundasjön, Stockholm	Urban	WSP_60857_18	19.1	170	300	760	3	4.70	2.70	<1,0	18	17	4.90	7.10
Lilla värtan, Stockholm	Urban	WSP_60857_17	8.9	21	30	190	<1,0	<1,0	<1,0	<1,0	5.20	6.70	5.70	7.70
Fjäderholmarna, Stockholm	Urban	WSP_60857_19	12.8	30	50	260	2.60	<1,0	<1,0	<1,0	4.30	8.40	6.10	7.60
Öresjö, Viskan (Borås)	Urban background	WSP_60857_45	9.7	11	4	2.50	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	15
Djupasjön, Viskan, (Borås)	Urban	WSP_60857_46	13.7	99	78	19	<1,0	14	14	<1,0	2.40	1.10	<1,0	16
Guttasjön, Viskan, (Borås)	Urban	WSP_60857_47	20.4	69	48	12	<1,0	8.80	11	<1,0	<1,0	<1,0	<1,0	11
Örsbaken, Nyköping	Recipient to STP	WSP_60857_51	52.9	2.4	2.1	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	1.30
Östra mellanfjärden, Nyköping	Background to Örsbaken	WSP_60857_50	33.9	2.2	2	2.8	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	2.10



Table A4. Concentrations in surface waters (ng/l).

Site	Category	sample nr	unit	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT
Bäsingen, Dalälven	STP-recipient	WSP_60857_63	ng/l	1.6	4.6	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna upstream, Hyndevad	Urban background	WSP_60857_34	ng/l	14	13	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna, central, Närjeholmsv.	Urban	WSP_60857_31	ng/l	21	5.5	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Eskilstuna, Torshälla	Urban	WSP_60857_32	ng/l	23	6.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Mälaren Rotholmen	Urban background	WSP_60857_8	ng/l	2	11	1.8	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Mälaren Rotholmen	Urban background	WSP_60857_9	ng/l	2	4	1.4	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Ulvsundasjön, Sthlm	Urban	WSP_60857_20	ng/l	2.2	6.2	2.9	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Årstaviken, 1	Urban	WSP_60857_12	ng/l	2.9	6.1	2.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Årstaviken, 2	Urban	WSP_60857_13	ng/l	2.1	6.9	2.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Waldemarsudde, 10- 16 m depth	Urban	WSP_60857_100	ng/l	1.3	3.2	3.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Waldemarsudde 1 m depth	Urban	WSP_60857_99	ng/l	1.5	5.7	2.4	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Waldemarsudde 21 m depth	Urban	WSP_60857_101	ng/l	2.8	6.7	3.2	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Fjäderholmarna	Urban coast	WSP_60857_21	ng/l	1.8	5.4	1.7	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Viskan (Öresjö)	Urban background	WSP_60857_42	ng/l	5.3	13	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Viskan (Druvefors)	Urban	WSP_60857_43	ng/l	2.5	6.8	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Viskan (Djupasjön)	Urban	WSP_60857_44	ng/l	11	5.8	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng , small river	sludge treated field	WSP_60857_76	ng/l	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0
Svedäng, small river	control field	WSP_60857_77	ng/l	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0



Table A5. Concentrations in foodstuff and aquatic biota (alla data in ng/g wet weight).

Site	Category	Sample details	Sample nr	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT	fat, %
	Foodstuff 1	Piggvar Östersjön	WSP_60857_82	0.5	2.8	5.7	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	6	
	Foodstuff 2	Pike, Baltic Sea	WSP_60857_83	0.6	2.7	4.5	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	12	
	Foodstuff 3	Salmon, farmed in Norway	WSP_60857_84	1.2	<0,30	2.4	<0,30	<0,40	<0,40	<0,30	<0,30	3.4	<0,30	
	Foodstuff 4	Salmon Vänern	WSP_60857_85	<0,30	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	6.8	4.6	
	Foodstuff 5	Salmon Vänern	WSP_60857_86	<0,30	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	4.4	3.3	
	Foodstuff 6	Öring Vänern	WSP_60857_87	1.6	<0,30	2.6	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	7.4	
	Foodstuff 7	Salmon Bothnian bay	WSP_60857_88	<0,30	1	3.7	<0,30	<0,40	<0,40	<0,30	<0,30	4.7	7.2	
	Foodstuff 8	Salmon Bothnian sea	WSP_60857_89	1	1.2	4.1	<0,30	<0,40	<0,40	<0,30	<0,30	5.3	8.2	
	Foodstuff 9	Salmon, Baltic proper	WSP_60857_90	0.8	1	7.8	<0,30	<0,40	<0,40	<0,30	<0,30	4.9	5.4	
	Foodstuff 10	Meat, pooled of var. animals	WSP_60857_91	<0,30	1.1	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	1.9	<0,30	
	Foodstuff 11	Mussels, canned	WSP_60857_92	15	64	4	<0,30	<0,40	<0,40	<0,30	<0,30	4.9	2.2	
	Foodstuff 12	Mussels, Swed. west coast	WSP_60857_93	4.6	13	27	<0,30	<0,40	<0,40	<0,30	<0,30	2.2	2.6	
Adelsö	Urban background	<i>Perch (perca fluviialis)</i>	WSP_60857_108	<0,300	4.5	13	<0,300	<0,400	<0,400	<0,300	<0,300	1	29.3	0.8
Färingsö	Urban background	<i>Perch (perca fluviialis)</i>	WSP_60857_109	<0,300	4.8	13.4	<0,300	<0,400	<0,400	<0,300	<0,300	2	48.9	1.4
Riddarfjärden	Urban	<i>Perch (perca fluviialis)</i>	WSP_60857_110	0.8	14	68.8	<0,300	<0,400	<0,400	<0,300	2.1	6.4	171	2.2
Slussen	Urban	<i>Perch (perca fluviialis)</i>	WSP_60857_111	0.8	21	71.3	<0,300	<0,400	<0,400	<0,300	0.8	5.3	124	0.7
Kummelnäs	Marine coast	<i>Perch (perca fluviialis)</i>	WSP_60857_112	0.8	11.8	33	<0,300	<0,400	<0,400	<0,300	1.1	3.5	82.8	0.5
Torsbyfjärden	Marine coast	<i>Perch (perca fluviialis)</i>	WSP_60857_113	<0,300	11.5	30.4	<0,300	<0,400	<0,400	<0,300	<0,300	3.4	102	0.7
Torsbyfjärden	Marine coast	<i>Perch (perca fluviialis)</i>	WSP_60857_114	<0,300	11.8	23.7	<0,300	<0,400	<0,400	<0,300	<0,300	3.3	86.9	0.7
Torsbyfjärden	Marine coast	<i>Perch (perca fluviialis)</i>	WSP_60857_115	<0,300	11.4	25.8	<0,300	<0,400	<0,400	<0,300	<0,300	3.2	84.3	0.7
Vindö	Marine coast	<i>Perch (perca fluviialis)</i>	WSP_60857_116	<0,300	3.1	6.8	<0,300	<0,400	<0,400	<0,300	<0,300	1.5	38.8	1.1
Öresund 1	Marine	ÖVF 1:5 Domsten mussels	WSP_60857_78	3.5	7.8	5.1	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	0.6	0.8
Öresund 2	Marine	ÖVF 3:5 Landskrona mussels	WSP_60857_79	1.2	2.1	4.5	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30	0.9
Öresund 3	Marine	ÖVF 4:12 Spillepeng mussels	WSP_60857_80	6	12	8.2	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	1.3	1
Öresund 4	Marine	ÖVF 5:5 Klagshamn mussels	WSP_60857_81	7.1	8.9	5.9	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	0.5	0.9



Table A6. Concentrations in breast milk (ng/l)

Provtyp	Study sites	Provrnr	MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	MPT	DPT	TPT
Breast milk 1	Uppsala 2002	WSP_60857_94	1.2	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30
Breast milk 2	Uppsala 2004	WSP_60857_95	1.9	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30
Breast milk 3	Göteborg 2001	WSP_60857_96	9.7	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30
Breast milk 4	Lund 2003	WSP_60857_97	<0,30	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30
Breast milk 5	Lycksele 2004	WSP_60857_98	5.8	<0,30	<0,30	<0,30	<0,40	<0,40	<0,30	<0,30	<0,30	<0,30