

Results from the Swedish Screening 2005

Subreport 2. Biocides

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Summary <p>.As an assignment from the Swedish Environmental Protection Agency, IVL has performed a "Screening Study" of the following biocides: bronopol, 4-Chloro-3-cresol, 2-Mercaptobenzothiazole, N-didecyldimethylammoniumchloride (DDMAC), Propiconazole, Resorcinol, 2-(Tiocyanomethylthio)benzothiazole, triclosan, Methyl-, Ethyl-, Propyl-, Butyl- and Benzylparabene. The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden. In total, 132 samples of air, precipitation, effluent water, surface water, sludge, sediment, fish, foodstuff and human urine have been analyzed. The background sites were generally non-contaminated, only 2 out of 17 background samples contained detectable amounts of biocides, and in these samples only two substances were found; namely triclosan and 2-mercaptobenzothiazole. Due to regular presence in samples of wastewater effluents, sludge and urban sediments, on-going emissions are likely to occur for 2-mercaptobenzothiazole, triclosan, DDMAC and parabenes. Observed concentrations were, however, below risk levels. The atmosphere was identified as a possible transport matrix for triclosan, 2-mercaptobenzothiazole and parabenes. Bronopol, resorcinol and 2-(tiocyanomethyltio)-benzothiazole were not detected in any of the samples analyzed, and were considered to pose no environmental risks. Propiconazole may be of local concern but at present it is unlikely to cause any problems on a national level.</p>	
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Sammanfattning

IVL Svenska Miljöinstitutet har på uppdrag av Naturvårdsverket utförd en screening av biocider. Studien omfattade följande substanser: bronopol, 4-klor-3-kresol, 2-merkaptobensothiazol, N-didecyldimetylammoniumklorid (DDMAC), propikonazol, resorcinol, 2-(tiocyanometyltio)bensothiazol, triklosan, metyl-, etyl-, propyl-, butyl- samt bensylparaben. Det övergripande syftet med studien var att bestämma koncentrationer i ett antal matriser i den svenska miljön, att undersöka viktiga transportvägar samt att utvärdera om pågående emissioner kan tänkas förekomma i Sverige.

En provtagningsstrategi utarbetades. Provtagningsprogrammet baserades på användningen, identifierade möjliga utsläppskällor samt på ämnenas egenskaper. Programmet inkluderade mätningar i bakgrundsmiljöer och nära potentiella punktkällor. Även mätningar av diffusa spridningsvägar i form av avloppssystem och urbana områden inkluderades. Mätningarna omfattade totalt 132 prover av luft, nederbörd, utgående avloppsvatten, ytvatten, slam, sediment samt fisk, varav 37 prover utgjordes av bidrag från det regionala programmet, där totalt sex länsstyrelser deltog. För att belysa human exponering för/upptag av bronopol and DDMAC analyserades även 12 prover av livsmedel och urin.

Bakgrundsstationerna var generellt sett oförorenade med avseende på de inkluderade biociderna, endast 2 av 17 bakgrundsprover innehöll detekterbara mängder av biocider och i dessa prover återfanns endast två substanser: triklosan och 2-merkaptobensothiazol.

För fyra substanser är pågående emissioner sannolika, då de återfanns i ett stort antal prover av utgående avloppsvatten, reningsverksslam samt i urbana sediment: 2-merkaptobensothiazol, triklosan, DDMAC och parabener. De uppmätta koncentrationerna låg dock under risknivåer. DDMAC kunde inte detekteras i livsmedel eller i mänsklig urin.

Atmosfären identifierades som en möjlig transportmatris för triklosan, 2-merkaptobensothiazol samt parabener, då dessa påvisades i luft och depositionsprover.

Bronopol och 2-(tiocyanometyltio)-bensothiazol detekterades inte i några prover och resorcinol endast i några enstaka slamprover samt ett prov av utgående avloppsvatten. Dessa tre substanser anses inte orsaka några risker för miljön. Propikonazol kan vara av lokal betydelse, men i nuläget torde den inte innebära några problem på nationell nivå.

Summary

As an assignment from the Swedish Environmental Protection Agency, IVL has during 2005/2006 performed a "Screening Study" of biocides. Included substances were bronopol, 4-Chloro-3-cresol, 2-Mercaptobenzothiazole, N-didecyldimethylammoniumchloride (DDMAC), Propiconazole, Resorcinol, 2-(Tiocyanomethylthio)benzothiazole, triclosan, Methyl-, Ethyl-, Propyl-, Butyl- and Benzylparabene. The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden.

A sampling strategy was set up, based on known use and possible emission sources and environmental behavior of the chemicals. The programme included measurements in background areas and close to potential point sources. Measurements of diffuse pathways from the society including sewage systems and urban areas were also included. In total, 132 samples of air, precipitation, effluent water, surface water, sludge, sediment, and fish have been analyzed. To elucidate human exposure, 12 samples of foodstuffs and urine were analyzed with respect to bronopol and DDMAC. Six county administrative boards throughout Sweden contributed with samples.

The background sites were generally non-contaminated, only 2 out of 17 background samples contained detectable amounts of biocides, and in these samples only two substances were found; namely triclosan and 2-mercaptobenzothiazole.

For four of the substances on-going emissions are likely to occur as they are regularly found in wastewater effluents, in sludge and in urban sediments: 2-Mercaptobenzothiazole, triclosan, DDMAC and parabenes. Observed concentrations were, however, below risk levels. DDMAC was not detected in foodstuffs nor in human urine.

The atmosphere was identified as a possible transport matrix for triclosan, parabenes and 2-mercaptobenzothiazole.

Bronopol, and 2-(tiocyanomethylthio)-benzothiazole were not detected in any of the samples analyzed, and resorcinol only occasionally in sludge and effluents. These three substances were considered to cause no environmental problems. Propiconazole may be of local concern but at present it is unlikely to cause any problems on a national level.

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1 Introduction

As an assignment from the Swedish Environmental Protection Agency, IVL has during 2005/2006 performed a "Screening Study" of selected chemicals within the groups pharmaceuticals, biocides and perfluorinated alkylated substances (PFAS) according to Table 1. The overall objectives of the screening were to determine concentrations in a variety of media in the Swedish environment, to highlight important transport pathways, and to assess the possibility of current emissions in Sweden. A further aim was to investigate the likelihood of long-range transport and uptake in biota. The outcome of the study is aimed to serve as a basis for decision-making e.g. regarding monitoring activities of these chemicals.

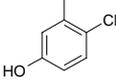
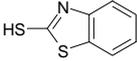
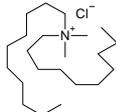
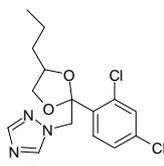
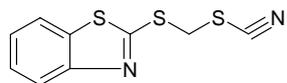
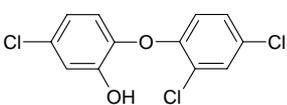
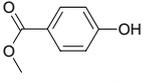
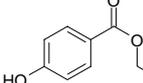
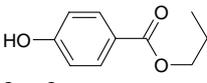
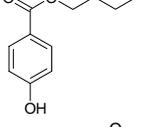
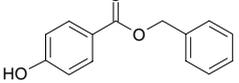
This subreport considers the screening of biocides. Results for the other chemical groups included in the screening are given in subreports 1 and 3.

Table 1. Chemicals selected for screening 2005

	Included substances	Subreport nr.
Pharmaceuticals	Antibiotics: Doxycycline, Lymecycline, Oxitetracycline, Tetracycline Antiinflammatory substances: Ibuprofen, Ketoprofen, Naproxen, Diclofenac Hormones: Ethinyl estradiol, Norethindrone, Estradiol, Estriol, Progesterone	1
Biocides	Bronopol, 4-Chloro-3-cresol, Mercaptobenzothiazole, N-didecyl-dimethylammoniumchloride, Propiconazole, Resorcinol, 2-(Tiocyanomethylthio)benzothiazole, triclosan, Methylparabene, Ethylparabene, Propylparabene, Butylparabene, Benzylparabene	2
PFAS	Perfluorobutane sulfonate (PFBS), Perfluorohexane sulfonate (PFHxS), Perfluorooctane sulfonate (PFOS), Perfluorodecane sulfonate (PFDS), Perfluorohexanoic acid (PFHxA), Perfluoroheptanoic acid (PFHpA), Perfluorooctanoic acid (PFOA), Perfluorononanoic acid (PFNA), Perfluorooctane sulfonamide (PFOSA)	3

The selected biocides together with CAS number and structures of the substances are given in Table 2. The different biocides are emitted to and spread in the environment via a variety of sources, e.g. point sources and use in products. Some of the chemicals are commonly used internationally and/or in Sweden.

Table 2. Biocides included in screening

Chemical name	CAS-no	
Bronopol	52-51-7	
4-Chloro-3-cresol	59-50-7	
2-Mercaptobenzothiazole	149-30-4	
N-didecyldimethylammonium chloride (DDMAC)	7173-51-5	
Propiconazole	60207-90-1	
Resorcinol	108-46-3	
2-(Tiocyanomethylthio)benzothiazole	21564-17-0	
Triclosan	3380-34-5	
Methylparabene	99-76-3	
Ethylparabene	120-47-8	
Propylparabene	94-13-3	
Butylparabene	94-26-8	
Benzylparabene	94-18-8	

2 Chemical properties and toxicity

The chemical properties and toxicity of the biocides included in the screening are described in the sub-chapters 2.1 - 2.9. The substances have physical and chemical properties according to Table 3.

Table 3. Physical and chemical properties of the included biocides. ^a

Substance	MW	W _{sol} (mg/l)	V _p (mm g, (25°)) ^b	H (Atm m ³ /mol) ^b	Log K _{ow}	BCF ^c	K _{oc} (L/kg) ^f
Bronopol	199.99	250000	1.26×10 ⁻⁵	1.33×10 ⁻¹¹	-0.64 ^b	3.2	5
4-Chloro-3-cresol	142.58	3830	0.05 ^a	2.45×10 ⁻⁶	3.1	49	-
2-Mercaptobenzo- thiazole	167.26	120	4.64×10 ⁻⁴	3.63×10 ⁻⁸	2.42	15	-
DDMAC	361.5 ^g	700 ^g			0 ^g	81 ^g	LogKoc = 5.9 ^g
Propiconazole	342.22	110	1.00×10 ^{-6a}	4.12×10 ⁻⁹	3.72	150	-
Resorcinol	110.11	717000	5×10 ^{-4d}	9.88×10 ⁻¹¹	0.8 ^e	3.2	65
2(Tiocyanomethyl- thio)benzothiazole	238.36	125	3.12×10 ⁻⁷	6.49×10 ⁻¹²	3.3	69	286-7896
Triclosan ^h	289.6	12	7×10 ⁻⁴		4.8		
Methylparabene	152.15	2500	2.37×10 ⁻⁴	2.16×10 ⁻⁸	1.96	6.4	
Ethylparabene	166.18	885	9.29×10 ⁻⁵	4.79×10 ⁻⁹	2.47	16	119-209
Propylparabene	180.20	500	5.55×10 ⁻⁴	6.37×10 ⁻⁹	3.04	44	
Butylparabene	194.23	207	1.86×10 ⁻⁴	8.45×10 ⁻⁹	3.57	110	
Benzylparabene	228.25	108	3.37	2.92×10 ⁻¹⁰	2.56	110	

2.1 Bronopol

Bronopol forms white to slightly yellow crystals; melting point 130 - 133° C. Due to its application in many consumer available products bronopol is released to the environment through various waste streams. The substance is highly soluble in water and not expected to bioaccumulate. Bronopol is stable to hydrolysis under normal conditions but at increased temperatures and/or higher pH rapid hydrolysis may occur (US EPA 2006a). Bronopol is easily biodegradable.

^a Experimental values retrieved from Chemfinder (2006), unless otherwise stated.

^b Estimated values retrieved from Chemfinder (2006), unless otherwise stated.

^c Estimated values retrieved from the PBT-profiler software, US EPA (2006c)

^f HSDB, 2006

^d Extrapolated value retrieved from SRC (2006a)

^e Experimental value retrieved from SRC (2006b)

^g Juergensen et al., 2000

^h Samsø-Petersen et al., 2003

Bronopol forms white to slightly yellow crystals; melting point 130 - 133° C. Due to its application in many consumer available products bronopol is released to the environment through various waste streams. The substance is highly soluble in water and not expected to bioaccumulate. Bronopol is stable to hydrolysis under normal conditions but at increased temperatures and/or higher pH (pH > 6; authors remark) rapid hydrolysis may occur (US EPA 2006a). Bronopol is easily biodegradable.

In aqueous solution bronopol is easily transformed to the carcinogenic substance formaldehyde. Other transformation products are bromonitroethane, bromoethanol, and bromonitroethanol (Wang et al., 2002).

When released to water, bronopol is not expected to adsorb to suspended solids and sediments based upon an estimated K_{OC} (Table 3). Based on the Henry's Law constant (Table 3) vaporization from the water phase is not expected to be an important transport pathway.

When released to air, bronopol will exist both in the vapor and the particulate phases and in the vapor phase it may react with hydroxyl radicals with an estimated half-life of 97 hours (HSDB, 2006).

In terms of ecotoxicity, bronopol is practically non-toxic to slightly toxic to birds; slightly to moderately toxic to freshwater fish and terrestrial invertebrates; moderately to highly toxic to estuarine/marine invertebrates and slightly toxic to estuarine/marine fish (US EPA, 2006b). Bronopol is classified as harmful if swallowed or in contact with skin; irritating to eyes, respiratory system and skin; risk of serious damage to eyes and very toxic to aquatic organisms (Xn; R21/22 - Xi; R37/38-41 - N; R50; ESIS, 2006). Most ecotoxicological data found in literature is for acute toxic effects. A predicted no effect concentration (PNEC) for water, derived from the most sensitive acute test for bronopol with a safety factor of 1000 (TGD, 2003), is 0.78 µg/l (Table 4).

2.2 4-Chloro-3-cresol

4-Chloro-3-cresol in its pure state exists in the form of needle-shaped crystals, with low water solubility. The vapor pressure indicates that 4-chloro-3 cresol mainly will exist in the gas phase of the atmosphere. The substance is degraded photo-chemically with a half-life of approximately 15 h. When released to water, it is not expected to adsorb to particles in water phase. OECD-tests have shown that 4-chlor-3-cresol was readily biodegradable and biodegradation may therefore be an important environmental pathway. The Henry's Law constant indicates that vaporization could also be an important pathway (Table 3; Forsgren et al., 2004).

The substance is classified as "harmful in contact with skin and if swallowed", "risk of serious damage to eyes", "may cause sensitization by skin contact" and "very toxic to aquatic organisms" (R 21/22, R41, R43, and R50). 4-chlor-3-chresol is classified as a priority risk reduction substance in the priority risk reduction database of the Swedish Chemical Inspectorate and as such users should evaluate the risk of its uses and consider substitution (PRIO, 2006). A PNEC for water, derived from the most sensitive acute toxicity test with a safety factor of 1000 (TGD, 2003), is 0.917 µg/, Table 4).

Log K_{OW} indicates a bioaccumulation potential but the OECD bioaccumulation tests of fish did not show any bioaccumulation potential and the chemical can therefore be assumed to have low risk for bioaccumulation (Table 3).

2.3 2-Mercaptobenzothiazole

2-Mercaptobenzothiazole occurs in its pure state as needle-shaped crystals that are insoluble in water. The substance is generally rather persistent (Haroune et al, 2004). When released to soil, it has low mobility and a half-life of 92-248 days. When released to water 2-mercaptobenzothiazole will partly dissociate but may also adsorb to sediment. In water it can be degraded photolytically with a half-life about 0.05 days in the summer and 0.21 days in the winter. When released to air, 2-mercaptobenzothiazole reacts with the hydroxyl radical with an atmospheric half-life of 8.4 hours (HSDB, 2006). Bioaccumulation tests indicate a low bioconcentration (ESIS, 2006).

The substance is classified as “very toxic to aquatic animals”, “may cause long-term effects in the aquatic environment” and “may cause sensitization by skin contact” (R 50/53, R43; ESIS; 2006). It also has shown potential mutagenic effects and has a known microbial toxicity (Haroune et al, 2004). Because of these properties, 2-mercaptobenzothiazole is classified as a risk reduction substance in the priority database of the Chemical Inspectorate in Sweden (KEMI) (PRIO, 2006). A PNEC for water, derived from the most sensitive acute toxicity test with a safety factor of 1000 (TGD, 2003), is 0.25 µg/, Table 4).

2.4 N-didecyldimethylammoniumchloride (DDMAC)

N-didecyldimethylammoniumchloride (DDMAC) is a cationic surfactant that belongs to a group of chemicals known as quaternary compounds. The mode of action in organisms is by cellular membrane disruption causing damage to exposed areas such as gills and gastrointestinal tracts (Szenasy, 1998). DDMAC is classified as “harmful when swallowed” and “can cause burns” (R22, R43, ESIS, 2006).

DDMAC has high affinity to solid materials, as soil and sediments, and its bioavailability is therefore assumed to be low. There is ambiguous data about its persistence indicating that the aquatic half-life may be between 11.2 days up to 23 years. Studies of other quaternary ammonium compounds suggest that the half-life is more likely to be in the order of days than in years (Szenasy, 1998). A PNEC for water, derived from the most sensitive acute toxicity test with a safety factor of 100 (TGD, 2003), is 0.37 µg/, Table 4).

2.5 Propiconazole

Propiconazole has a low vapor pressure and a low mobility. In soil it binds strongly to particles, which affects its spreading. Degradation by microorganisms has shown to be relatively slow and in anaerobic environments there is practically no degradation at all. Complete degradation of the most stable metabolite, 1, 2, 4-triazol, takes several years. Degradation with impact of sunlight has only been showed in water. Propiconazole has a potential for bioaccumulation (KEMI, 1997).

Toxicity for birds and insects are relatively low, probably low for earthworms as well but moderately to high for aquatic organisms. Some algae show very high sensibility to propiconazole (KEMI, 1997). A PNEC derived from the most sensitive EC50 with a safety factor of 1000 (TGD, 2003) gives 0.021 µg/l. Soil microorganisms seem unaffected by propiconazole at normal use.

Propiconazole is classified as harmful if swallowed and it may cause sensitization in contact with skin. It is very toxic to aquatic organisms and may cause long term adverse effects in the

environment (R22, R43, and R50/53). In the priority database of KEMI it is classified as risk reduction substance (PRIO, 2006).

2.6 Resorcinol

Resorcinol is a crystalline, aromatic chemical that is water-soluble and very conductive to derivatization. Due to its applications the substance may be released to air and water. No hydrolysis is expected to occur due to lack of appropriate functional groups. In the water phase adsorption to suspended solids is expected to be limited due to its low K_{OC} (Table 3).

The vapor pressure (Table 3) indicates that resorcinol in ambient air mainly will exist in the vapor phase where it may be degraded by the hydroxyl radical with an estimated half-life of 1.9 hours. Assays based on the OECD ready biodegradability and inherent biodegradability protocols indicate that resorcinol is biodegradable (INDSPEC, 2004).

Resorcinol is classified as harmful if swallowed, irritating to eyes and skin and very toxic to aquatic organisms (Xn; R22 Xi; R36/38 N; R50; ESIS, 2006). A PNEC for water, derived from the most sensitive acute toxicity test with a safety factor of 1000 (TGD, 2003), is 0.25 $\mu\text{g}/\text{Table 4}$). Resorcinol administered at high dose to rodents can also disrupt thyroid hormone synthesis. A risk assessment of exposure of resorcinol under real world conditions showed that resorcinol is not expected to cause adverse effects on thyroid function (Lynch et al., 2002).

2.7 2-(Tiocyanomethylthio)benzothiazole

If released to air the vapor pressure indicates that 2-(tiocyanomethylthio)benzothiazole will exist in both the vapor and particulate phases. The atmospheric half-life considering reaction with hydroxyl radical is estimated to be 4 hours. 2-(Tiocyanomethylthio)benzothiazole absorbs light in the environmental UV-spectrum and may thus undergo direct photolysis in sunlight, producing 2-mercaptobenzothiazole and benzothiazole. If released to soil, 2-(Tiocyanomethylthio)benzothiazole is expected to have moderate to slight mobility based upon K_{OC} and is expected to be non-biodegradable. If released to water it may adsorb to suspended solids and sediments. BCF- values, which range between 14 and 268, suggest that bioconcentration is possible but not extensive (Table 3).

2-(Tiocyanomethylthio)benzothiazole is classified as “harmful if swallowed”, “very toxic by inhalation”, irritating to eyes and skin”, “may cause sensitization by skin contact”, very toxic to aquatic organism, “may cause long term adverse effects in the environment” (R22, R26, R36/38, R43, R50/53; ESIS, 2006). A PNEC for water, derived from the most sensitive acute toxicity test with a safety factor of 100 (TGD, 2003), is 0.038 $\mu\text{g}/\text{Table 4}$).

2.8 Triclosan

Triclosan is a relatively lipophilic compound ($\log K_{OW}$ 4.8) and an ionizing molecule with a pK_a of 8.14 resulting in a higher lipophilicity when pH is low and the molecule is not ionized. The non-ionized molecule is stable against photochemical reduction while the ionized molecule is not (Tixier et al. 2002). Because of its antimicrobial properties triclosan does not meet the criteria set for the

standard biodegradability test (OECD 301C, 302C and 303A). Activated sludge tests contradicted these results with an almost complete removal of triclosan from waste water (Ciba, 1998).

Triclosan is classified as “irritating to skin and eyes”, “very toxic to aquatic organisms” and “may cause long term adverse effects in the environment” (R 36/38;R50/53). A PNEC water derived from the most sensitive species for triclosan with a safety factor of 10 is 0.05 µg/l (Ciba 1998, Reiss et al., 2002).

2.9 Parabenes

Environmental data have only been found for **ethylparabene** and **methylparabene** while toxicity data was only found for **methylparabene**.

When released to air **ethylparabene** will exist in both the vapor and particulate phases and it will be degraded photo-chemically by hydroxyl radical reactions, with an estimated half-life of 10 hours. If released to soil **ethylparabene** is expected to have high to moderate mobility based upon a K_{OC} ranging from 119-209. The pKa of **ethylparabene**, 8.3, indicates that this compound partially will exist as an anion in moist soil surfaces and is expected to have very high mobility in soils. The Henry law constant indicates that no volatilization from moist surfaces will take place. Aerobic biodegradation is expected to be an important environmental pathway with measured half-lives about 3.5 and 14 days. A BCF-value of 16 suggests that bioaccumulation in aquatic organisms is low (Table 3).

Methylparabene is easily biodegradable and has no potential for bioaccumulation. Toxicity values range from 10-100 mg/l with daphnia as the most sensitive species (Läkemedelsverket, 2004). Using a safety factor of 1000 (TGD, 2003) a PNEC of 10 µg/l is obtained.

Table 4. Summary for estimated PNEC-values for the selected biocides (Safety factor according to TGD, 2003)

	PNEC (µg/l)	Safety factor	Water quality guideline (Canada) (µg/l)
Bronopol	0.78	1000	
4-chloro-3 cresol	0.917	1000	
2-Mercaptobenzothiazole	0.25	1000	
DDMAC	0.37	100	1.5 (Canada; Juergensen et al., 2000)
Propiconazole (marine algae)	0.021	1000	0.1(EG; Kemi,1997)
Propiconazole	3.1	100	0.1(EG; Kemi,1997)
Resorcinol	0.25	1000	
2-(Tiocyanometyltio)benzothiazole	0.038	100	
Triclosan	0.05	10	
Parabenes	10	1000	

3 Environmental fate

In order to highlight the likely fate and partitioning behavior of the included biocides a modeling exercise was performed using the Equilibrium Criterion (EQC) model (Mackay et al., 1996). Physical-chemical properties were taken from Table 3. The degradation half-lives (in hours) used are given in Table 5 and Table 6¹.

Table 5. Estimated half-lives (hours) of biocides

Substance	T _½ (air)	T _½ (water)	T _½ (soil)	T _½ (sediment)
Bronopol	288	360	720	3360
2-Mercaptobenzothiazole	9.6	360	720	3360
4-Chloro-3-cresol	15	912	1800	8160
Propiconazole	17	1440	2880	12960
Resorcinol	2	360	720	3360
2-(Thiocyano-methylthio)benzothiazole	3.8	912	1800	8160

Table 6. Estimated half-lives (hours) of parabenes

Substance	T _½ (air)	T _½ (water)	T _½ (soil)	T _½ (sediment)
Methylparabene	36	360	720	3360
Ethylparabene	31	360	720	3360
Butylparabene	24	209	408	1872
Benzylparabene	22	360	720	3360

Emission rates were set to 1000 kg/h, only for illustrative purposes. The outcome of the modeling exercise is shown in Table 5. The numbers given in the Table should be regarded as indicative, as they are dependent on model structure as well as on chemical property data. This calculation is based on the steady state distribution of a chemical, in an environment not at equilibrium. The chemical is continuously discharged at a constant rate into the chosen environmental media, and achieves a steady-state condition at which input and output rates are equal. This involves calculating the rates of degradation and advection (from half-lives/rate constants and advective flow rates) and considering the emission. Intermedia transport processes (e.g. wet deposition, evaporation, or sedimentation) are included. The media receiving the emissions are very important and have a controlling influence on the overall fate of the chemical.

The overall residence time of the chemical in the system depends on to which compartment it is emitted and it is generally lower for the modeled biocides than the previously modeled pharmaceuticals (subreport 1).

Biocides are mostly small molecules with fairly high vapor pressures. It can thus be anticipated that the air compartment would be an environmentally important media. However, the EQC-modeling attempt indicates that except for 2-mercaptobenzothiazole, 4-chloro-3-cresol and butylparabene, air is not likely to be the most important compartment where the biocides tend to exist. Instead, these substances, due to their high water solubility, tend to primarily partition to the water compartment (Table 7, Table 8 and Table 9). Also soil will receive a substantial part. Taking the pattern of use also into consideration, such as agriculture, the soil matrix will be of importance.

¹ Values retrieved from the PBT-profiler (US EPA, 2006c)

Most of the biocides selected in this screening effort possess ionisable groups and the properties of the substances therefore depend on the pH of the environmental compartment. For instance 2-mercaptobenzothiazole has a pKa-value of 6.93, which implicates that at an environmentally relevant pH of 6.93, as in lake water and soil, 50 % of the molecules will be ionized (an-ions) and the other half will be neutral, and distribute in the environment roughly as predicted by the model. The ionic form will have an even higher propensity to be in the aqueous phase. Also the parabenes have environmentally relevant pKa-values (~ 8.4) and the previously described dissociative pattern will be valid also for them. Thus, in the environment, a large fraction of the substances will exist as anions and the predicted environmental distribution is likely to be shifted to the water compartment with persistence largely governed by the aqueous degradation kinetics.

In the case of parabenes, it is interesting to note that in the almost homologous series (methyl-, ethyl-, propyl- etc), the predicted residence time of this group shows a slight increase as a function of increasing molecular weight. But butylparabene, and to some extent also propylparabene, behave slightly different showing a shorter residence time (Figure 1). This is a result of the lower half-lives used as input data. Whether this is a true deviation in persistence or merely some artifacts in the QSAR model used for estimation of half-lives is still elusive.

Table 7. Results from EQC modeling of Bronopol (Brono) and 2-Mercaptobenzothiazole (MercBe), using emission rates of 1000 kg/h

Emission medium	% in air		% in water		% in soil		% in sediment		Persistence (h)	
	Brono	MercBe	Brono	MercBe	Brono	MercBe	Brono	MercBe	Brono	MercBe
Air	<0.001	25	26	8.6	74	67	0.05	0.05	680	47
Water	<0.001	0.01	99	99	<0.001	0.03	0.18	0.6	340	343
Soil	<0.001	0.09	22	27	78	725	0.04	0.17	717	966
All three	<0.001	0.05	39	12.	61	77	0.07	9.8	580	450

Table 8. Results from EQC modeling of 4-Chloro-3-cresol (Cresol) and Propiconazole (Propi), using emission rates of 1000 kg/h

Emission medium	% in air		% in water		% in soil		% in sediment		Persistence (h)	
	Cresol	Propi	Creso l	Propi	Creso l	Propi	Creso l	Propi	Creso l	Propi
Air	26	0.08	9.4	2.1	64	98	0.3	0.3	66	3260
Water	0.04	0.07	97	87	0.1	0.01	2.7	13	570	771
Soil	<0.01	<0.001	1.1	0.33	99	99	0.03	0.05	2471	4088
All three	0.6	0.03	19	9.3	80	89	0.54	1.4	1038	2706

Table 9. Results from EQC modeling of Resorcinol (Reso) and 2-(Tiocyno-methylthio)benzothiazole (Tiocyan), using emission rates of 1000 kg/h

Emission medium	% in air		% in water		% in soil		% in sediment		Persistence (h)	
	Reso	Tiocyan	Reso	Tiocyan	Reso	Tiocyan	Reso	Tiocyan	Reso	Tiocyan
Air	0.006	0.01	23	3.1	77	97	0.05	0.14	696	2196
Water	<0.001	<0.001	99	96	<0.001	<0.001	0.19	4.3	342	592
Soil	<0.001	<0.001	19	0.71	81	99	0.04	0.03	748	2534
All three	0.002	0.006	36	12	64	87	0.07	0.6	596	1774

Table 10. Results from EQC modeling of Methylparabene (M-Par) and Ethylparabene (E-Par), using emission rates of 1000 kg/h

Emission medium	% in air		% in water		% in soil		% in sediment		Persistence (h)	
	M-Par	E-Par	M-Par	E-Par	M-Par	E-Par	M-Par	E-Par	M-Par	E-Par
Air	0.84	0.97	10	6.2	89	93	0.04	0.04	713	720
Water	<0.001	<0.001	99	99	0.01	0.02	0.34	0.67	343	344
Soil	0.006	0.003	6.7	2.6	99	97	0.02	0.02	913	986
All three	0.3	0.34	24	20	75	79	0.08	0.14	656	683

Table 11. Results from EQC modeling of Butylparabene (B-Par) and Benzylparabene (Be-Par), using emission rates of 1000 kg/h

Emission medium	% in air		% in water		% in soil		% in sediment		Persistence (h)	
	B-Par	Be-Par	B-Par	Be-Par	B-Par	Be-Par	B-Par	Be-Par	B-Par	Be-Par
Air	13	0.4	5	3.7	81	96	0.17	0.19	148	836
Water	<0.001	<0.001	97	95	0.03	0.007	3.3	4.9	239	358
Soil	<0.001	<0.001	0.16	0.24	99	99	0.005	0.01	587	1034
All three	0.15	0.006	25	17	73	82	0.85	0.86	324	743

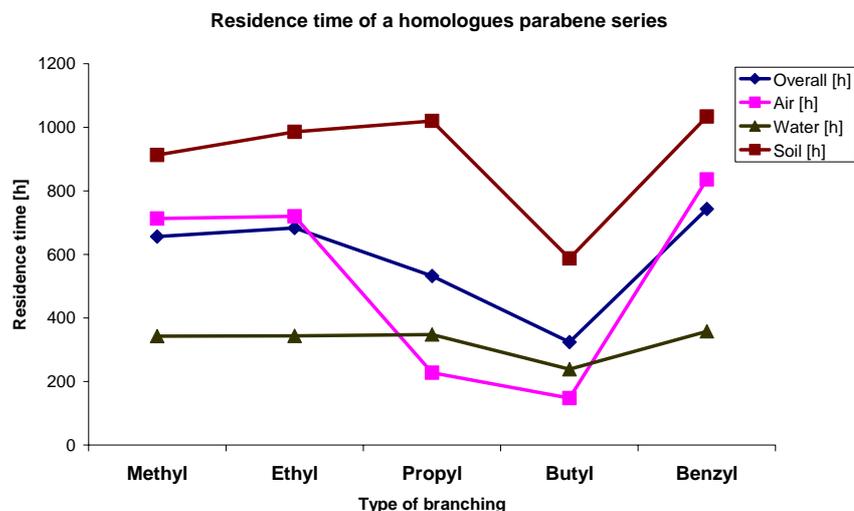


Figure 1. Predicted system residence time as a function of functional chemical composition in a homologue series of parabenes.

4 Production, consumption and regulation

Consumption of the different biocides in Sweden during recent years is shown in Figure 2 and Figure 3. Consumption data was retrieved from the SPIN-database, Substances in preparations in Nordic countries (SPIN, 2006).

Bronopol is used as a preservative in cosmetics, hygiene products and in chemical products such as glue and paint. The substance can also be used as a slimicide and bactericide in industrial processes *e.g.* paper mills, cooling water, air conditioning/humidifying systems (US EPA 2006a). Products that may contain bronopol are washing detergents, disinfectants, hygienic biocides for veterinary use, pharmaceutical products for skin application, shampoo and bathing products (Wang et al, 2002). Concentrations range between 0.01 and 0.1 % (the maximum level allowed by EC directive 76/768; Weyland et al., 1994).

As a biocide, bronopol is included in the European directive on biocides EG 2032/2003. All usage of the substance should be reported to national agencies. Bronopol is an LPV² chemical within the EU, with production registered in UK, Germany, Italy and the Netherlands. Because of the many areas of use, bronopol is expected to be imported in both industrial and consumer products

Resorcinol is an essential component of adhesive systems and an important chemical intermediate in the manufacturing of chemicals such as light screening agents for the protection of plastics from exposure to ultraviolet light. Other uses include the manufacturing of dyestuffs, pharmaceuticals, flame retardants, agriculture chemicals, fungicidal creams and lotions, explosive primers, antioxidants, chain extender for urethane elastomers and treatment to improve mechanical and

² LPV = Low production volume produced or imported in EU with a tonnage >10t/y but never more than 1000 t/y

chemical resistance of paper machine fabrics (INDSPEC, 2004). Resorcinol is an HPV³ chemical with registered production in Germany and Finland (ESIS, 2006). Because of the many areas of use resorcinol is expected to be imported with both industrial and consumer products.

2-mercaptobenzothiazole is an HPV chemical with registered production in Germany, Netherlands, Belgium, Spain, France and UK (ESIS, 2006). 2-mercaptobenzothiazole is a typical rubber additive but it also has other applications such as inhibiting biocorrosion in cooling systems or in paper manufacturing. The substance is a biocide and as such included in the European directive on biocides EG 2032/2003.

4-Chloro-3-cresol is a biocide and as such included in the European directive on biocides EG 2032/2003. All usage of the substance should be reported to national agencies. The substance is prohibited in products for contact with mucous membranes and not allowed in cosmetic products in concentrations above 0.2 % (LVFS 1993:2 4:24; Forsgren et al., 2004)

N-didecyldiammoniumchloride (DDMAC) is commonly used industrially as disinfectant and is part of many household items such as fabric softeners and anti-static agents (Szenasy, 1998). It is also used as a pesticide and is included in the European directive on biocides EG 2032/2003. DDMAC is an LPV chemical with registered production in Sweden (Stenungsund), France, Germany and Switzerland.

Propiconazole is used as a pesticide against fungus in seed, forest nurseries, in products for wood protection and impregnation (KEMI, 1997). Propiconazole is an LPV chemical with registered production in Germany and France (ESIS, 2006). Propiconazole is included in the European directive on biocides EG 2032/2003.

2-(Tiocyanomethylthio)benzothiazole (TCMTB) is a biocide used for conservation of leather and wood partly as an ingredient in wood preserving paint.

Triclosan is a chlorinated phenolic compound that has been used as a biocide in several consumer products for many years. It has been used in e.g. toothpaste to prevent loosening of teeth as well as in fiber- and plastic material because of its ability to be incorporated into the material and then slowly leak out during a long period of time. During recent years a lot of attention has been drawn to triclosan and its feared negative impacts on the environment. As a result, many producers have ceased using the compound. Triclosan is an LPV chemical in the European Union with production registered in Switzerland and Italy. (ESIS, 2006).

³ HPV = High Production Volume produced or imported in quantity of at least 1000 tonnes per year in EU by at least one industry

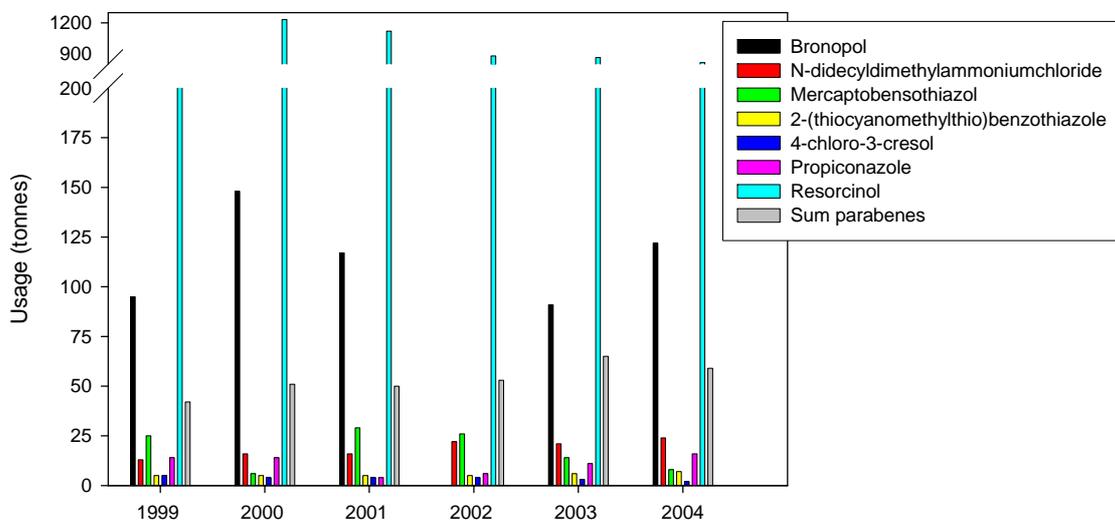


Figure 2. Consumption of biocides (tonnes) in Sweden 1999-2004 (SPIN, 2006)

Ethyl- and propylparabenes are reported as LPV chemicals within the European Union (ESIS, 2006). In Sweden, the most extensive used parabene is methylparabene. Methylparabene is used in pharmaceuticals as an "aid component". It occurs in about 170 different pharmaceutical products where 20 are for veterinary purposes. The use in pharmaceutical products answers for approximately 15 % of the total consumption of parabenes. Other uses of parabenes are as preservatives in cosmetic products.

The consumption of the selected biocides according to the SPIN-database (SPIN, 2006) is shown in Figure 2. where the different chemicals are compared. The figure shows that resorcinol is used in greatest amounts followed by bronopol. No significant trend in the consumption between the different years could be seen for any of the biocides. Figure 3 shows the consumption of different homologues of parabenes.

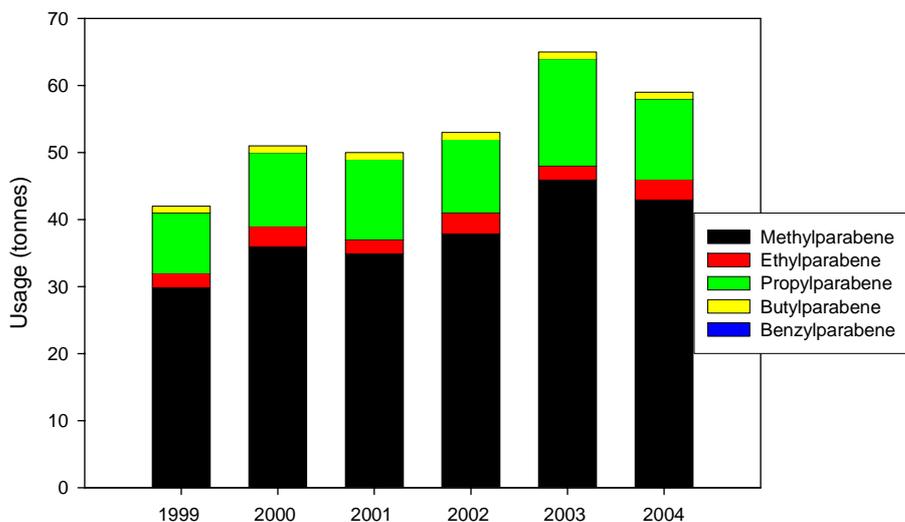


Figure 3. Consumption of parabenes (tonnes) in Sweden 1999-2004 (SPIN, 2006)

5 Previous measurements in the environment

Data from previous measurements of the selected biocides are scarce. The data found in the literature are summarized in Table 12. Possible sources of various benzothiazoles (benzothiazole, BT, 2-hydroxybenzothiazole, HOBT and 2-(4-morpholino)benzothiazole, 24MoBT) to the environment have previously been discussed by Reddy & Quinn (1997). They analyzed recycled car tires, antifreeze, urban runoff, sediments, road dust and urban particulate matter and estimated fluxes of benzothiazoles to the environment. One of the analyzed compounds was found in leachate from a rubber landfill but fluxes could not be estimated. They also detected various benzothiazoles in urban runoff, in road dust and urban particulate matter, and hypothesized leaching from car tires and/or use of antifreezers. Fluxes from tire wear were estimated, and antifreeze was identified as a possible source to the environment. Reddy & Quinn concluded that benzothiazoles may enter the environment via leaching of rubber products, tire wear and antifreeze.

Triclosan has previously been measured at several sites in the environment (Table 12). Triclosan has also been detected in human breast milk, where three out of five randomly selected samples showed levels that were considered high (60-300 µg/kg lipid weight; Adolfsson-Erici et al., 2003). Triclosan was also detected in different foodstuffs such as meat(fat), egg, milk and fish in concentrations between 1.4-8.4 while it could not be detected in vegetables nor in cereals in a Swedish screening study (Remberger et al., 2002).

Table 12. Previous measurements of selected biocides.

Substance	Matrix	Comment	Country	Concentration	Reference
4-Chloro-3-cresol	Surface water		Germany	0.1-0.4 µg/l (mean-max)	ESIS, 2006
	Effluent water (public WTP)		Germany	0.5-1.5 µg/l	ESIS, 2006
	Effluent (paper mill)		Germany	1.8 µg/l	ESIS, 2006
	Sludge		Germany	0.12-0.4 µg/l	ESIS, 2006
	Sludge		Great Britain	220 µg/kg dw	ESIS, 2006
	Effluent		Great Britain	0.073 µg/l	ESIS, 2006
Propiconazole	Surface water		Sweden	≤ 1.2 µg/l	KEMI,1997
Resorcinol	Sewage water			7-22 mg/l	ESIS, 2006
DDMAC	Sediment		Fraser river, Canada	0.52-1.26 µg/g dw	Szenasy, 1998
Triclosan	River water	Downstream population centers	USA	Ave= 140 ng/l Max 2300 ng/L	Kolpin et al.,2002
	Sea water	Influenced by STPs	Schweiz	1.4-74 ng/l	Lindström et al., 2002
	Sea & river water	Influence from chemical industries	USA	600-40000ng/l	Lopez-Avila and Hites, 1980
	Sea sediment	Influence from chemical industries	USA	<d.l-100mg/kg	Lopez-Avila and Hites, 1980
	Sewage water	STP	Switzerland	600-1300 ng/l	Lindström et al., 2002
	Sludge	STP	Sweden	2.8-4.4 µg/g d.w	Remberger et al., 2002
	Influent water	STP	Sweden	0.1-1.5 µg/L	Cited in Samsö-Petersen et al., 2003
	Effluent water	STP	Sweden	0.2 µg/L	Cited in Samsö-Petersen et al., 2003
	Influent water	STP	Denmark	2.5-1.7 µg/L	Cited in Samsö-Petersen et al., 2003
	Effluent water	STP	Denmark	<1	Cited in Samsö-Petersen et al., 2003
	Biota	STP	Switzerland	35 ng/g ww	Balmer et al. 2004
	Effluent water	STP		42-213 ng/l	Singer et al.,2002
	River water	STP recipient	Switzerland	11-98 ng/l	Singer et al.,2002
	Surface water	Industrial influence	Sweden	<2-160	Remberger et al. 2002
	Sediment	Industrial influence	Sweden	2-25 µg/kg DW	Remberger et al., 2002
	Soil	Industrial influence	Sweden	3-15 µg/kg DW	Remberger et al., 2002
	Biota	Muscle	Sweden	0.8-13 µg/kg ww	Remberger et al., 2002
Air	Urban	Sweden	0.02- 0.17ng/m3	Remberger et al., 2002	
Air	Ambient	Sweden	0.003-0.005 m3	Remberger et al., 2002	
Deposition	Urban	Sweden	9.7-20 ng/m2/d	Remberger et al., 2002	
Deposition	Ambient	Sweden	0.2-0.41 ng/m2/d	Remberger et al., 2002	

6 Sampling program and study sites

A national sampling program was developed in order to determine the environmental concentrations of selected biocides in different environmental matrices in Sweden. An additional aim of the sampling program was to identify major emission sources as well as important transport pathways. The sampling program was based on identified possible emission sources and the behavior of the substances in the environment.

Emissions from diffuse sources were identified as important pathways of biocides to the environment due to the variable use of these substances. Based on modeling of the distribution pattern of the biocides to different environmental compartments, water, sediment and sludge were the prioritized matrices for sampling. Municipal sewage treatment plants (STPs) were also considered as important sources for the occurrence of the biocides in the aquatic environment. STP samples e.g. sludge are frequently used as indicators of diffuse spreading of chemicals in the environment.

One of the depicted routes of biocides into the environment is from consumer products being added to the domestic garbage disposal system. Therefore, landfills and landfill leachate water were important sampling matrices, which were considered as point sources when the sampling scheme was set up.

Environmental background levels in water, sediment and biota were determined in samples from three background lakes where the influence from human activities was considered minor. Finally, to investigate human exposure some foodstuff samples and human urine samples were also included in the sampling program.

Air samples were collected at a background site at the Swedish West Coast, Råö as well as at a remote site, Pallas in the north of Finnish Lapland. Råö and Pallas are sampling stations used within the national monitoring program for air pollutants. The air monitoring site hosted by the City of Stockholm Environment and Health Administration (Miljöförvaltningen) at rooftop level in central Stockholm (Södermalm) was used as urban station for diffuse sources.

During the winter and spring 2005 three measurement campaigns were carried out at Råö, at Pallas and in central Stockholm. At the sampling station in Stockholm also rain/deposition samples were taken during the early spring (pooled monthly). Sediment, fish and water samples were also collected in the Stockholm area.

A harbor area and a wood impregnation industry were selected as point sources. In addition to this, effluent water from industries with known use of resorcinol (sample provided within the regional program) and parabenes were added.

The general sampling program is summarized in Table 13. Site information and sample characteristics of the samples collected within the program are given Appendix 3 & 4.

Table 13. General sampling strategy of the biocide screening

		Air	Precipitation	Water	Sediment	Biota	Sludge	Provisions	Urine
Back-ground	Pallas	x							
	Råö	x	x						
	Lilla Öresjön			x	X	x			
	Stensjön			x	X	x			
	Tärnan			x	X	x			
Diffuse	Stockholm	x	x						
	Riddarfjärden			x	X	x			
	Stora Essingen			x	X	x			
	Årstaviken			x	X	x			
	Mälaren				X				
	Municipal STPs			x			x		
	Landfills			x					
Point source	Industrial plants Harbor	x		x	X				
Human exposure								x	x

6.1 Sampling

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

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

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

6.1.1 Water

The water samples were collected using a Ruttner sampler and transferred to cleaned plastic bottles and stored and stored in a freezer (-18°C) until analyzed. A bottle with ultra pure water (Milli-Q), which was exposed to the surrounding environment during the sampling time, was used as a field blank.

6.1.2 Sediment

Sediment samples from lakes were collected by means of a Kajak sampler. The sediment core was sliced and transferred into plastic jars and stored in a freezer (-18°C) until analyzed. A plastic jar filled with water saturated diatomaceous earth that was exposed to the surrounding environment during the sampling time was used as field blank.

6.1.3 Sludge

The staff at the different treatment plants collected the sludge samples from the anaerobic chambers. After the process in the anaerobic chamber the sludge was concentrated by means of added polymers and centrifugation. The de-watered sludge was transferred into pre cleaned plastic jars (polypropylene) and stored in a freezer (-18°C) until analyzed. A plastic jar filled with water saturated diatomaceous earth, which was exposed to the surrounding environment during the sampling time, was used as a field blank.

6.1.4 Air and deposition

Air samples were collected using a low volume air sampler (LVS) with a flow of approximately 1 m³/h. The air was passed through a glass fiber filter (MG160, Munktell) where particles were collected and then through a glass column packed with the adsorbents XAD-2 (Amberlite) and polyurethane foam (PUF). Sampling duration was four weeks. Filter and adsorbents were changed every week. Prior to sampling, glass fiber filters were heated to 400°C, and the adsorbent columns were cleaned by Soxhlet extraction with acetone. After sampling, the filter and columns were wrapped in aluminum foil and sent to the laboratory, where they were stored in a freezer (-18°C) until analysis. Additional filters and columns used as field blanks were sent back to the laboratory unexposed.

The deposition was collected using an open sampler (bulk sampler). This sampler consists of a 1 m² Teflon coated surface with 10 cm-high edges. The bottom declines slightly to a central opening where a cassette with an adsorbent (PUF) is attached. The deposition sample includes both compounds in the precipitation and compounds deposited to the collection surface of the sampler. Both the precipitation and the deposited particles are included in the analysis. This type of sampler is used for deposition measurements of organic contaminants within the Swedish national monitoring of air pollutants (Brorström-Lundén, 1996).

6.1.5 Fish

The fish (perch and roach) samples from Stockholm were collected by means of fishing net. The fish samples were individually wrapped in aluminum foil and stored in freezer (-20°C) until analyzed. Muscle samples of perch from background lakes was supplied from The Environmental Specimen Bank and the Swedish Museum of Natural History (A. Bignert and colleagues).

7 Analytical methods

7.1 Chemicals

The solvents, HPLC-quality, methanol, acetonitrile, hexane, pentane, methyl-tert-butylether (MTBE), dichloromethane (DCM) and ethylacetate were delivered from Rathburn (Chemical Ltd., Peeblesshire, Scotland).

Merck (Darmstadt, Germany) delivered diethyl ether, (ethanol-stabilized) phosphoric acid, ammonium acetate, sodium chloride, di-potassium carbonate, ascorbic acid, sodium sulphate and silica gel.

Sodium sulphate, silica gel and sodium chloride were thermally cleaned (400°C) prior use. The silica gel was deactivated (5 % water) before used.

Solid phase columns containing a hydroxylated steredivenylbensene copolymer sorbent (ENV+; 500 mg) were delivered from IST, (International sorbent Technology, Mid Glamorgan, U.K).

Ultra-pure water was produced by a Milli-Q plus (Millipore Corporation, Bedford, MA, USA).

The 3-fluoro-methyl-, ethyl- and propylparabene used as recovery standard (for parabene, resorcinol and bronopol) were synthesized in the laboratory and used as surrogate standards. The purity of three products was > 98 % (GC-MS).

The analytical methods developed to quantify the selected biocides in various matrices are briefly described below.

7.2 Propiconazole and 2-mercaptobenzothiazole

7.2.1 Water

The sample (200 ml) was spiked with recovery standard following ammonium acetate and ascorbic acid addition. The sample was acidified (pH 2-6) with phosphoric acid and extracted twice with pentane:diethyl ether. The combined extract was. Polar and acidic compounds were washed out by shaking the extract with buffer (pH 7). The extract was finally dried over sodium sulphate. The extract was derivatised as described below.

7.2.2 Sludge/Sediment

The sample (10 g) was spiked with recovery standards. Ammonium acetate and ascorbic acid were added and the sample was carefully mixed. Phosphoric acid was added to a final pH of 2-6. The sample was extracted twice with methanol. The extracts were combined and diluted with water and the mixture was extracted in two cycles with pentane: diethyl ether. The extract was concentrated and the co-extracted polar and acidic compound was washed out in the same manner as for the water samples. The extract was dried and finally derivatized as below.

7.2.3 Air/Aerosols

The filters, the XAD/PUF and the glass cartridges used were all subjected to Soxhlet extraction using acetone as solvent for 24 h. The sample extract was concentrated first on a RotoVap and further under a nitrogen gas stream. The extract was dried and finally derivatized as below.

7.2.4 Derivatization

The 2-mercaptobenzothiazole, dissolved in ethylacetate/diethyl/methanol, was derivatized with ethereal diazomethane. The reaction was performed at room temperature in darkness.

The reagent excess was evaporated with the aid of a nitrogen jet and the methanol was washed away by shaking the extract with water after addition of hexane. The extract was dried over sodium sulphate and concentrated before clean up on a silica gel column (see below).

7.2.5 Clean-up by silica gel chromatography

The deactivated silica gel column was prepared in a Pasteur pipette. Two fractions were collected: (a) fraction F-3, containing the S-methylated 2-mercaptobenzothiazol, was obtained by elute the column with hexane: MTBE and (b) fraction F-5, containing propiconazole, was obtained by elute with MTBE.

Both fractions F-3 and F-5 where concentrated with nitrogen and finally spiked with injection standard prior GC-MS-analysis.

The extraction and work-up procedure is summarized schematically in Figure 4.

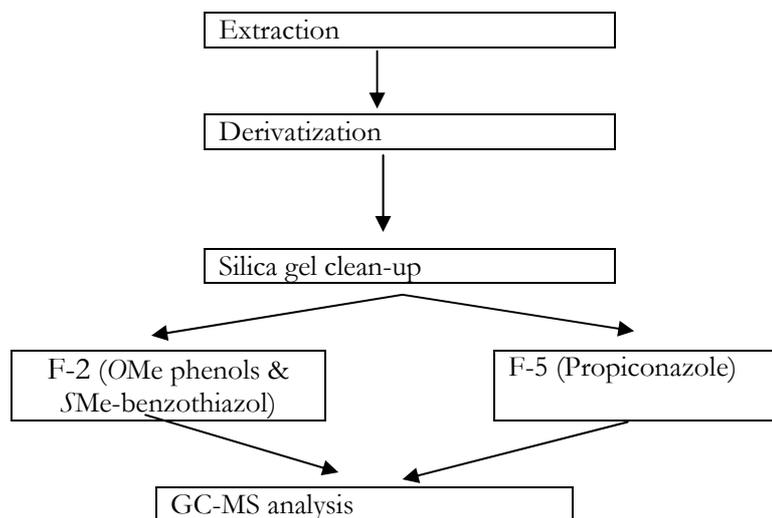


Figure 4. Scheme of the analytical sample work-up

7.2.6 GC-MS analysis

The extracts were analyzed on a 6890N gas chromatograph with a 5973N mass selective detector (Agilent). The injection was done at 250°C in pulsed splitless mode. The fused silica capillary column (VF-5MS 30 m × 0.25 mm i.d. × 0.25 µm film thickness, Varian) was held at 45°C for 1 min, ramped 15°C/min to 200°C and 5°C/min to 300°C, and then held at 300°C for 2 min. Helium was used as carrier gas. The mass spectrometer transfer line temperature was 280°C. The detector was used in selected ion monitoring mode with electron ionization at energy of 70 eV. Their characteristic retention time, one target ion (Tgt-ion) used for quantification of the analytes and one qualifier ion used (Q-ion) to increase specificity was recorded (table 3 and 4). Quantification was based on comparison of peak abundance to the known response of the internal standard (diphenyl). The reported analyte concentrations were corrected for the losses according to the determined recovery standard as described above. The analytes, injection standard (IS), and recovery standard (RS) used in this study are summarized in Table 14.

Propiconazole is not a single compound but two isomers. The analytical results in present report are reported as the sum of these two isomers.

Table 14. Molecular weight (MW), target ion (Tgt-ion) and qualifier ion (Q-ion) used for the GC-MS quantification.

Compound	MW	Tgt-ion	Q-ion
Diphenyl IS	154	154	153
2-Mercaptobenzothiazole S-Me	181	181	148
Atrazin RS	216	215	200
Chlorfenvinfos RS	351	323	267
Propiconazole	342	259	173
Tetrachlorbisphenol-A-(OMe) ₂ US	394	379	377

Molecular weight (MW); The target ion (Tgt-ion) were used for quantification; The qualifier ion (Q-ion) and retention time (data not shown) was used to tentatively confirm the identity of the compounds. Injection standard (IS); Recovery standard (RS); S-methylated (SMe). Compounds marked with bold text are the studied compounds in present investigation.

7.3 Bronopol and resorcinol

7.3.1 Water

When thawing the sample, acid was added in order to stabilize the analytes. An aliquot of the water sample (200 ml) was spiked with surrogate standard, acidified (pH 2) and fortified with sodium chloride (20 g/l) before concentration on a SPE-column. The analytes were eluted with ethyl acetate and MTBE. The extract was concentrated azeotropic evaporation and subsequently derivatized (see below).

7.3.2 Sludge/Sediment

The sample was fortified with surrogate standards and extracted with acetonitrile. The sample was centrifuged and the organic extract was transferred to a test tube and diluted with sodium chloride solution and subsequently MTBE-extracted. The combined extract was concentrated and dried in the same manner as for the water samples (see above) and was thereafter derivatized (see below).

7.3.3 Biota (fish)

The sample was fortified with recovery standards and homogenized in acetonitrile. The extraction was performed by gentle agitation on a shaking board. The sample was centrifuged and the organic extract was transferred to a test tube and diluted with sodium chloride solution and subsequently MTBE-extracted. The combined extract was concentrated and dried in the same manner as the water samples (see above) and was thereafter derivatized (see below).

7.3.4 Derivatization

The extract dissolved in hexane/MTBE was reacted with acetic acid anhydride in the presence of sodium acetate. The reaction was accomplished on a heating block. The excess reagents were removed by shaking with di-potassium carbonate. The derivatized extract was dried over sodium sulphate and finally cleaned up on a silica gel column (see below).

7.3.5 Clean-up by silica gel chromatography

A deactivated silica gel column was prepared in a Pasteur pipette. The derivatized analytes were transferred to the column and eluted by a mobile phase containing hexane:MTBE. The eluate was concentrated and internal standard (diphenyl) was added prior to analysis by GC-MS.

7.3.6 GC-MS analysis

The extracts were analyzed on a 6890 N gas chromatograph coupled to a 5973N mass selective detector (Agilent). The injection, 1 µl, was done in splitless mode at 250°C. The fused silica capillary column (VF-5MS 30 m x 0.25 mm i.d. x 0.25 µm film thickness, Varian) was held at 50°C for 3 min., ramped 12°C/min to 280°C and held at 280°C for 10 min. Helium was used as carrier gas. The detector was used in selected ion monitoring mode (SIM). Resorcinol was analyzed in electron ionization mode at energy of 70 eV. Bronopol was analyzed in negative chemical ionization mode with methane as reaction gas. The analytes were identified by their characteristic retention time and one target ion (Tgt-ion, used for quantification) and in most cases one or two qualifier ions (Q-ion) used to increase specificity was recorded. Quantification was based on comparison of peak abundance to the known response of the internal standard (biphenyl). The reported analyte concentrations were corrected according to the determined surrogate standard losses.

Table 15. Target ion (Tgt-ion) and qualifier ions (Q-ion) used for the GC-MS quantification.

Substance	Tgt-ion	Q-ion 1
Diphenyl (EI-mode)	154	-
3-Fluor ethyl parabene(EI-mode)	139	184
Resorcinol(EI-mode)	110	152
Bronopol (NCI-mode)	79	81

7.4 Parabenes

7.4.1 Water

The water samples were extracted using a solid phase extraction approach using ENV+ as solid phase. The column was activated using MTBE, acetonitrile and acidified water. To sample was the acidified, spiked sample. Sodium chloride was added in order to drastically increase the ionic strength of the sample solution and thus salt out the polar analytes in the solid phase extraction. After applying the sample onto the SPE-column, the column was rinsed with acidified water, left to dry and the sequentially eluted with of acetonitrile followed by an elution of hexane:MTBE (1:1). Acidified water was the added to the retrieved extracts and the analytes in the supernatant organic phase was isolated. The extract was further extracted using hexane:MTBE (1:1). The extract was concentrated and dried prior to derivatization (se below).

7.4.2 Sludge/Sediment samples

5-10 grams of sample was first spiked with internal standards and acidified. The samples were then extracted with acetone using an ultrasonic bath. The procedure was then repeated using acetone:MTBE. The extract was pooled and concentrated. Acetone was washed of bay shaking the extract with acidified water. The organic extract was then dried with sodium sulphate and resolved in hexane. The hexane-base samples where then back-extracted using a borax buffer based procedure. The extract was dried with sodium sulphate prior to derivatization.

7.4.3 Biota (fish)

Sample of fish muscle (10 g) was homogenized in acetone and acidified with phosphoric acid. Magnesium sulfate was added and the sample was extraction on a shaking board. The acetonitrile extract was withdrawn, diluted with acidified water and extracted with two aliquot of a solvent mixture of hexane and MTBE. This extract was dried and the solvent was exchanged to hexane prior the back extraction procedure using a borax buffer. The extract was finally derivatized, and chromatographed on a silica column prior to GC-MS analysis (se below).

7.4.4 Derivatisation

The extract, dissolved was reacted with acetic acid anhydride in the present of the base sodium acetate. The reaction was accomplished on a heating block. The excess reagents were removed by shaking the extract with di-potassium carbonate. The derivatized extract was dried over sodium sulfate and finally subjected to clean up on a silica gel column (see below).

7.4.5 Clean-up by silica gel chromatography

A deactivated silica gel column was prepared in a Pasteur pipette. The derivatized analytes were transferred to the column and eluted with a aliquot of hexane:MTBE. The eluate was concentrated and internal standard (diphenyl) was added prior to analysis by GC-MS.

7.4.6 GC-MS analysis

The extracts were analyzed on a 6890 N gas chromatograph coupled to a 5973N mass selective detector (Agilent). The injection, 1 µl, was done in splitless mode at 275°C. The fused silica capillary column (VF-5MS 30 m x 0.25 mm i.d. x 0.25 µm film thickness, Varian) was held at 45°C for 1 min., ramped 15°C/min to 200°C, 5°C/min until 300°C and held at 300°C for 5 min. Helium was used as carrier gas. The detector was used in selected ion monitoring mode (SIM). The parabenes were analyzed in electron ionization mode at energy of 70 eV. The analytes were identified by their characteristic retention time and one target ion (Tgt-ion) and in most cases one or two qualifier ions (Q-ion) used to increase specificity was recorded (see Table 16). Quantification was based on comparison of peak abundance to the known response of the internal standard. The reported analytes concentrations were corrected according to the determined surrogate standard losses.

Table 16. Target ion (Tgt-ion) and qualifier ion (Q-ion) used for the GC-MS quantification.

Substance	Tgt-ion	Q-ion 1	Q-ion 2
3-Fluoromethyl parabene	139	170	-
Methyl parabene	121	152	-
3-Flouroethyl parabene	139	184	-
Ethyl parabene	121	166	-
3-Fluoro propyl parabene	156	139	-
Propyl parabene	138	121	180
Buthyl parabene	138	121	194
Benzyl parabene	121	228	270

7.5 N-didecyldimethylammonium chloride (DDMAC)

7.5.1 Water

Analysis of DDMAC in water was performed according to (Ding, et al. 2003; Hind, et al. 1997).

The water sample was spiked with recovery standard, acidified with HI and fortified with KI. The sample was left on a shaking board for 20 min. Potassium hydroxide was added until a final pH of 10-11. The iodide salt of the analyte was extracted with three aliquot of dichloromethane. The polled extract was concentrated, spiked with injection standard prior analysis on GC-MS.

7.5.2 Sludge/sediment

Quaternary ammonium compounds have a high affinity to particles and are therefore expected to be found in sediments and sludge. The strength of the binding to these matrices is powerful and it is therefore important to use a powerful extraction method capable to release the analytes. The method used was developed by Fernández et al. (1996) and Picó et al. (2000) to meet these needs and was used with minor modifications in this study. Briefly, the sample was spiked with recovery

standard and extracted with 1 M HCl in methanol. The extraction was first performed in an ultrasonic bath and then on a heating block (85°C). This procedure was repeated twice and the extracts were pooled. The methanol in the extract was blown off with the aid of a nitrogen jet at elevated temperature.

The residue was extracted twice with hexane:MTBE. This extraction was a clean up step and was discarded.

The cleaned up sample extract was evaporated to dryness and reconstituted in water. In order to exchange the counter ion from chlorine in DDMAC to iodide (DDMAI), hydrogen iodide (HI) and potassium iodide (KI) were added in excess followed by addition of potassium hydroxide until the pH reached 10-11. The DDMAI was extracted with three aliquots of dichloromethane. The combined DCM-extract was concentrated and analyzed with GC-MS after the addition of injection standard.

7.5.3 Biota samples (fish)

Ground fish meat was spiked with recovery standard and mixed with methanol and acidified with HCl. The extraction was first performed in an ultrasonic bath. After this step, the sample was transferred to a SPE-column and the extract was forced down into a test tube by vacuum. The sample was then further extracted with two aliquots of methanol. The methanol extract was centrifuged and the supernatant was withdrawn and evaporated to dryness. After this stage the counter ion was exchanged using the same procedure as for sediment/sludge samples (see above).

7.5.4 GC-MS analysis

The extracts were analyzed on a 6890 N gas chromatograph coupled to a 5973N mass selective detector (Agilent). The injection, 1 µl, was done in splitless mode at 300°C. The fused silica capillary column (VF-5MS 30 m x 0.25 mm i.d. x 0.25 µm film thickness, Varian) was held at 45°C for 1 min., ramped 15°C/min to 200°C, 5°C/min until 300°C and held at 300°C for 5 min. Helium was used as carrier gas. The detector was used in selected ion monitoring mode (SIM). DDMAC was analyzed in electron ionization mode at energy of 70 eV. The analytes were identified by their characteristic retention time and one Target ion (Tgt-ion) and in most cases one or two qualifier ion (Q-ion) used to increase specificity were recorded (see Table 17). Quantification was based on comparison of peak abundance to the known response of the internal standard. The reported analyte concentrations were corrected according to the determined surrogate standard losses.

Table 17 Target ion (Tgt-ion) and qualifier ion (Q-ion) used for the GC-MS analysis of DDMAC.

Compound	Tgt-ion	Q-ion
N-Didecyldimethylammonium iodide	184	185
N,N-dimethylundecylamine	58	199
Biphenyl (injection standard)	154	-

7.6 Quality control

The recoveries of the different analytes were tested in separate experiments. Acceptable recovery of added surrogate standards and analytes was generally $\geq 75\%$. Analytes were quantified using the

internal standard approach: all reported values were recovery corrected. Method blanks and field blanks were run for each sample batch, to assess background interference and contamination.

The limit of detection (LOD) was defined as 3 times background blank level noise but with consideration of the actual sample noise. For some of the analytes background levels were detected. Background levels were subtracted from measured sample values.

All glass equipment, glass fiber filters, sodium sulphate and sodium chloride were cleaned by heating to 400°C for 4 h prior to use. All other equipment was washed with organic solvent before use.

The following quality criteria were used to ensure correct identification and quantification of the target compound : (a) the retention time matched those of the standard compounds within 0.1 min, S/N was >3 (b) the ratio of the selected ions (Tgt- and Q-ions) was within $\pm 15\%$ of expected / theoretical value. The recovery of all internal standards in the analyzed samples was within acceptable limits, 40-90 %.

8 Results

The concentrations of the biocides found in the different samples from the national and regional screening are shown in Appendix 4 and Appendix 5 and are presented below per site category. An overview of the results and a concluding discussion is given in chapter 9.

8.1 Bronopol

The concentrations of bronopol were below the detection limit in all the analyzed samples. Detection limits are given in Table 18.

Table 18. LOD bronopol

Matrix	Limit of Detection
Air	0.05-0.07 ng/m ³
Precipitation	0.16 µg/L
Water	0.05 µg/l
Sediment	10-17 µg/kg dw
Fish	10 µg/kg fw
Sludge	12-24 µg/kg dw
Food stuffs	10 µg/kg fw

8.2 4-Chloro-3-cresol

8.2.1 Background

The concentrations of 4-chloro-3-cresol were below limit of detection in all samples from background locations (biota LOD 0.5µg/kg fw; sediment LOD 5 - 20 µg/kg dw; water LOD 0.01 µg/l, air LOD 0.03 ng/m³).

8.2.2 STPs and landfills

4-chloro-3-cresol was found in 16 out of 26 **sludge** samples from municipal STPs in concentrations ranging from 7 to 67 µg/kg dw with the highest concentration found in Borås STP (Figure 5). The concentrations were lower than what has been detected in Great Britain (220 µg/kg dw; Table 12). In the remaining 10 samples, the concentrations were below the detection limit (<3 - <7 µg/kg dw)

In **effluent water**, detectable amounts were only found in the sample from the STP in Piteå in a concentration of 0.019 µg/l (LOD 0.01 µg/l). This is similar to, or lower than what has been measured in Germany (effluent water 0.5-1.5 µg/l; Table 12).

An apparently elevated water concentration of 0.11 µg/l was found in a sample representing a **storm drain overflow** event in Göteborg.

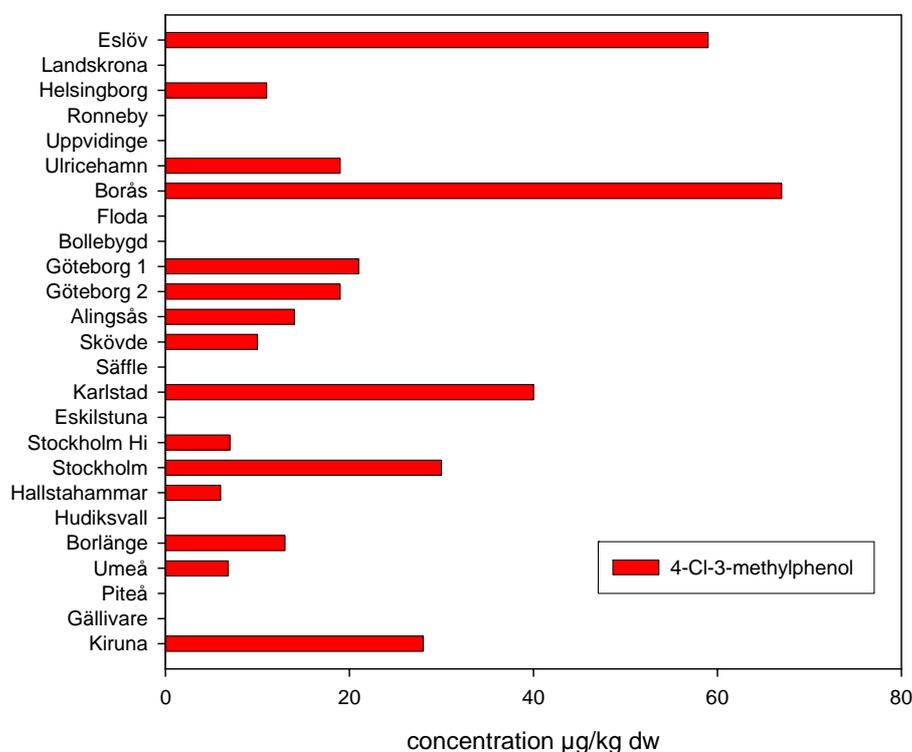


Figure 5. Concentrations of 4-Cl-3-cresol in sludge from municipal STPs

One **leachate water sample** from Högbytorp landfill in Bro contained 4-chloro-3-cresol in a concentration of 0.7 µg/L. This was the only sample from landfills with concentrations above LOD (0.01-0.03 µg/l). This sample was taken at Högbytorp landfill before the treatment process. The concentration in the sample taken after treatment was below the LOD.

8.2.3 Urban and industrial areas

4-chloro-3-cresol was found in **atmospheric deposition** in the urban area in two out of three samples. The deposition fluxes during the measuring period in March and April were 4.2 and 1.8 ng/m² day respectively. The concentration of 4-chloro-3-cresol in urban air, Stockholm, was below the detection limit (0.04 ng/m³).

No **sediment** or **biota** samples adjacent to urban or industrial areas contained any detectable amounts of 4-chloro-3-cresol (**sediment LOD**: 2 - 9 µg/kg dw; **biota LOD**: 5 µg/kg fw).

8.3 2-Mercaptobenzothiazole

8.3.1 Background

2-Mercaptobenzothiazole was detected in one **sediment** sample from Lilla Öresjön at a concentration of 7.2 µg/kg dw. The concentrations in all the other background samples were below the limit of detection (**water LOD** 0.05-0.1 µg/l; **sediment LOD** 20 -25 µg/kg dw; **biota LOD** 10 -20 µg/kg fw).

8.3.2 STPs and landfills

Mercaptobenzothiazole was found in 16 out of 24 sludge samples from municipal STPs. The variation between different STPs was great and the concentrations ranged between 22 and 950 µg/kg dw (Figure 6). The two samples from Göteborg STP were collected with one week in between and showed great variations (<20 µg/kg dw and 200 µg/kg dw).

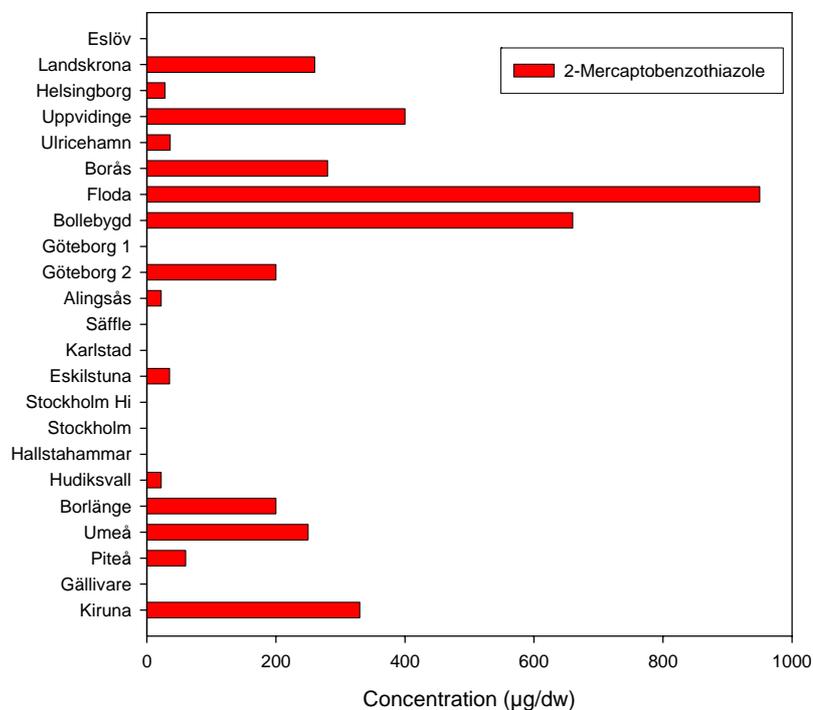


Figure 6. Concentration of 2-Mercaptobenzothiazole in sludge from municipal STPs

2-Mercaptobenzothiazole was found in seven of the 14 analyzed **effluent water** samples from municipal STPs in concentrations between 0.13 and 0.63 µg/l (Figure 7). LOD was 0.05 µg/l.

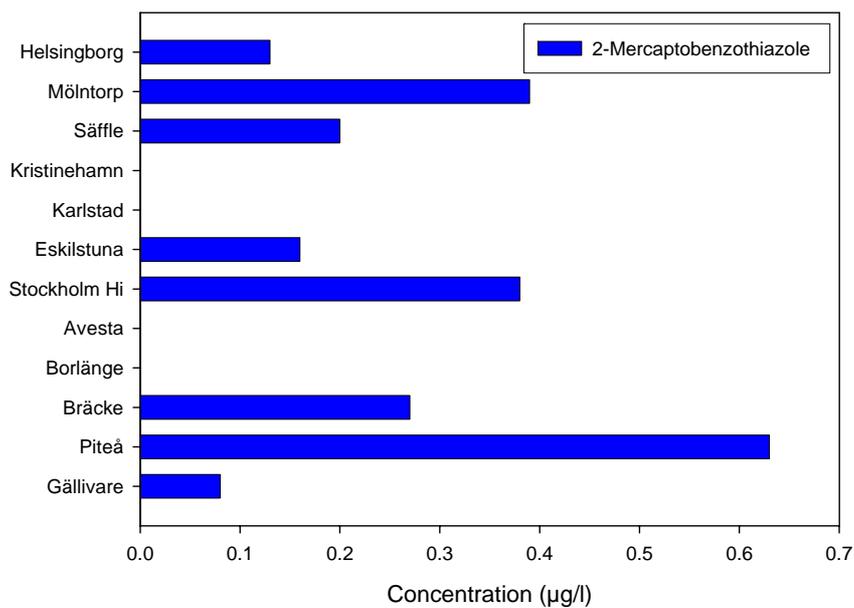


Figure 7. Concentrations of 2-Mercaptobenzothiazole in effluent water from municipal STPs

2-Mercaptobenzothiazole was found in three out of six analyzed **leachate water** samples in concentrations ranging between 0.11 and 18 µg/l. (LOD 0.05-0.5 µg/l). The highest concentration was found in the landfill Djupadalen in Karlstad.

8.3.3 Urban and industrial areas

2-Mercaptobenzothiazole was found in **atmospheric deposition** and in **sediment** samples from Stockholm. The **atmospheric deposition** fluxes were 35-37 ng/m² day and **sediment** concentrations ranged between 33 and 70 µg/kg dw, which is somewhat higher than what was found in the background sample from Lilla Öresjön (Figure 14).

The concentrations of 2-mercaptobenzothiazole were below LOD in all samples of **surface water**, **biota** and **air** (water LOD 0.01 µg/l; biota LOD 10 µg/kg fw; air LOD 0.2 - 0.05 ng/m³).

8.4 N-didecyldimethylammoniumchloride (DDMAC)

8.4.1 Background

DDMAC was not found in any of the analyzed background samples (**water** LOD 0.05 µg/l; **sediment** LOD 50 µg/kg dw).

8.4.2 Municipal STPs, landfills and point sources

DDMAC was detected in all analyzed **sludge** samples from municipal STPs in concentrations ranging between 280 - 61 000 µg/kg dw, with the highest detection found in Gässlösa STP, Borås (Figure 8).

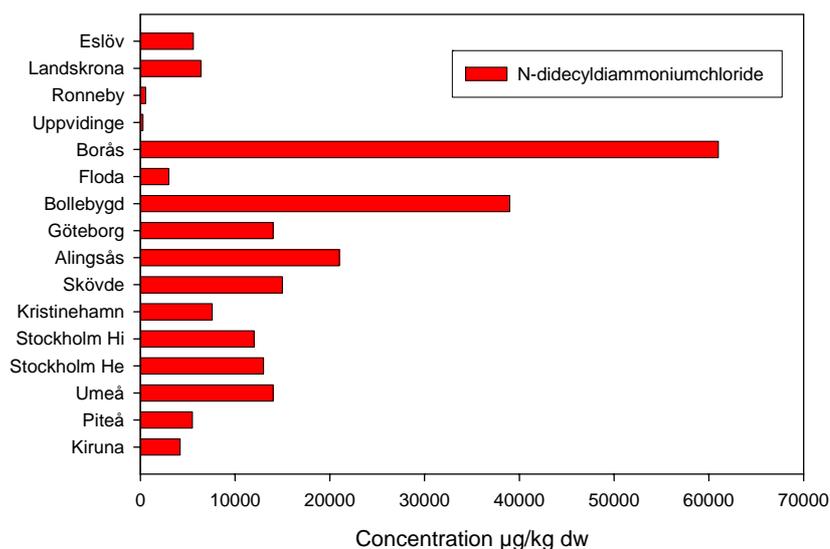


Figure 8. Concentrations of DDMAC in sludge from municipal STPs

DDMAC was detected in one **effluent water** sample from Himmerfjärden STP in a concentration of 0.17 µg/l. DDMAC was also found in sludge from the STP at Casco Adhesive (220 µg/kg dw). The concentrations in all **leachate** water samples were below LOD (< 0.04 - < 1 µg/l).

8.4.3 Urban areas

DDMAC was found in 3 **sediment samples** from Stockholm in concentrations ranging between 59 and 140 µg/kg dw. This is about 10 times lower than observed in the Fraser river sediments in Canada (520 -1260 µg/kg; Table 12). All other urban samples showed concentrations below LOD (**water** LOD 0.05 µg/l; **fish** LOD 20 µg/kg fw).

8.4.4 Human exposure

DDMAC was not detected neither in **food stuff** (LOD 20 µg/kg fw) nor in **urine** (LOD 1 µg/l)

8.5 Propiconazole

8.5.1 Background

Propiconazole was not found in any of the background samples (**water** LOD 0.01 µg/l; **sediment** LOD 5 µg/dw; **biota** LOD 10 µg/fw).

8.5.2 Municipal STPs and landfills

Propiconazole was found in 2 out of 24 **sludge** samples from municipal STPs, 54 µg/kg dw in the **sludge** sample from the Kavaheden STP in Gällivare and 18 µg/kg dw in the **sludge** sample from the Öresundsverket STP in Helsingborg. The LOD was 5 µg/kg dw).

Propiconazole was also found in 2 out of 14 **effluent water** samples from municipal STPs. These were Ekeby STP in Eskilstuna and Kavaheden STP in Gällivare and the concentrations were 0.1 and 0.02 µg/l respectively (LOD 0.01 µg/l).

Propiconazole was found in 1 out of 6 **leachate water** samples, in Djupadalen landfill in Karlstad (0.03 µg/l). LOD was 0.01 µg/l.

8.5.3 Urban areas

Propiconazole was not found in samples collected in urban areas (**surface water** LOD 0.01µg/l; **sediment** LOD 2-5 µg/kg dw; **biota** LOD 10 µg/kg ww; **air** LOD 0.002-0.005 ng/m³).

8.5.4 Point sources

Propiconazole was found in **air** samples in concentrations between 0.25 and 0.39 ng/m³ (three monthly samples) taken in the door opening to a roofed outdoor storage area for impregnated wood. It was also detected in a **storm water well** at the premises of the same wood impregnation

factory at a concentration of 0.13 µg/l. The water concentration in the well was 1×10^6 times lower than the concentrations in the liquor used for a closed recirculation stain bath for wood impregnation. In **sediment** from the same storm water well, propiconazole was found in a concentration of 20 µg/kg dw. Notable is that all storm water wells and stain baths from the factory are plugged and all waste water from the factory is processed by SAKAB. The concentrations of propiconazole in water samples taken outside the factory area were below the detection limit (0.01 µg/l).

Propiconazole was not found in the water sample taken in a harbor (<0.1 µg/l).

8.6 Resorcinol

8.6.1 Background

Resorcinol could not be found in the background samples (air LOD 0.002 ng/m³; water LOD 0.05 µg/l; sediment LOD 5-20 µg/kg dw; biota LOD 5 µg/kg fw).

8.6.2 STPs and landfills

16 **sludge** samples have been analyzed with respect to resorcinol. In three of these samples considerable amounts of resorcinol was detected; 380 µg/kg dw in a sludge sample from Kiruna, 160 µg/kg dw in sludge from Borås (Gässlösa STP) and 22 µg/kg dw in Ulricehamn STP. The LODs for the remaining 13 samples were 10-50 µg/kg dw.

Resorcinol was only detected in one **effluent water** sample from Stockholm (Himmerfjärden STP) in a concentration of 0.078 µg/l. The detection limit for other samples varied between 0.01 and 0.05 µg/l.

The **leachate water** samples from landfills did not contain any detectable amounts of resorcinol (LOD 0.01 - 0.05 µg/l).

8.6.3 Urban and industrial areas

Resorcinol could not be found in samples from urban or industrial areas (LOD: **sediment** 5-20 µg/kg dw; **air** 0.05 ng/m³; **water** 0.01-0.05 µg/l; **biota** 5 µg/kg fw).

8.7 2-(Tiocyanomethylthio)benzothiazole

2-(tiocyanomethylthio)benzothiazole was not found in any of the samples analyzed (LOD: **air** 0.01 ng/m³; **water** 0.05 µg/l; **sediment** 50 µg/kg dw; **sludge** 50 µg/kg dw; **biota** 10 µg/kg fw).

8.8 Triclosan

8.8.1 Background

Triclosan was found in **precipitation** in a concentration of 0.012 µg/l and in one **sediment** sample in a concentration of 9 µg/kg dw (LOD < 2 µg/kg dw). In all other samples from background sites, the concentrations were below the LODs; **air** 0.002 ng/m³, **biota** 0.1 µg/kg fw **surface water**, 0.003-0.005 µg/l.

8.8.2 STPs and landfills

Triclosan was found in five **effluent water** samples from municipal STPs in concentrations ranging between 0.02 and 0.09 µg/l (LOD < 0.005 µg/l). Triclosan was also found in **sludge** in concentrations ranging between 2000 – 5600 µg/kg dw with one sample deviating with a concentration of 43 000 µg/kg dw (Borlänge). Triclosan was previously measured in a screening study performed by IVL in concentrations of 2800-4400 µg/kg dw in sludge (Remberger et al, 2002) and in effluent water by Adolfsson-Erici et al. (2003) in concentrations of 84-330 ng/l. Compared to the previous studies, the concentrations in the current study were in the same range in sludge but somewhat lower in effluent water.

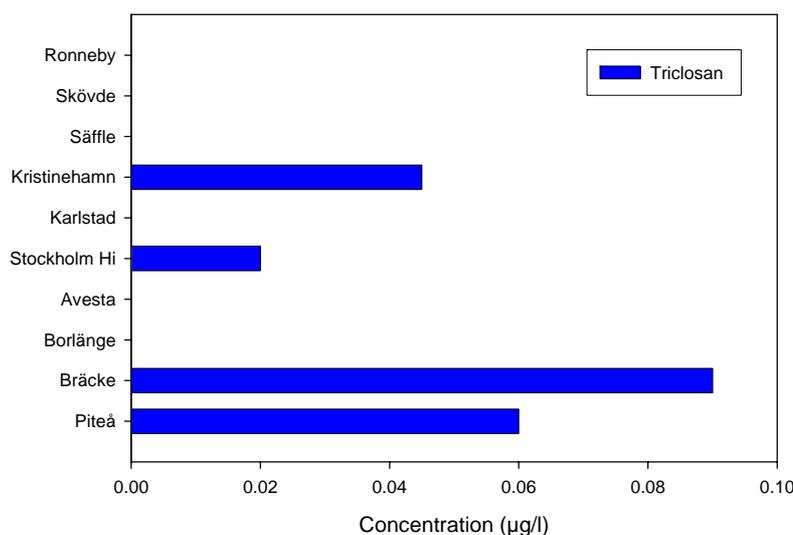


Figure 9. Concentration of triclosan in effluent water from municipal STPs

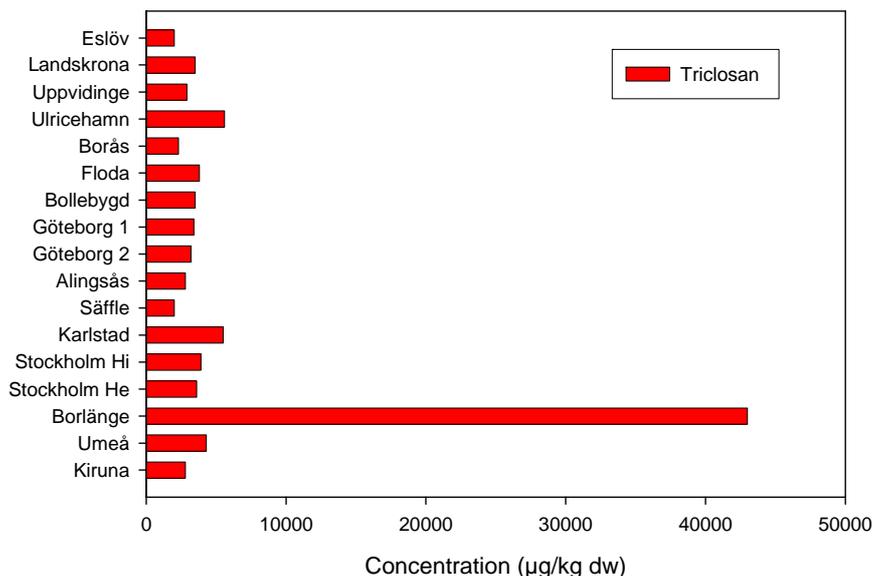


Figure 10. Concentration of triclosan in sludge from municipal STPs

8.8.3 Urban areas

Triclosan was detected in one urban **air** sample at a concentration of 0.01 ng/m³. LOD for the two others was 0.003 ng/m³. Triclosan was also found in two out of three atmospheric **deposition** samples. The deposition fluxes were 1.6-3.5 ng/m² day (LOD: 0.5 ng/m² day), which is lower than the fluxes measured in a previous study by IVL (Remberger et al., 2002; 9.7-20 ng/m² day).

Triclosan was found in one out of four analyzed **biota** (fish) samples at a concentration of 0.56 µg/kg fw (LOD 0.1 µg/kg fw). Triclosan was also found in **sediment** in concentrations ranging between 3.6 and 29 µg/kg dw with the highest concentration in Himmerfjärden. The concentrations in sediment and air are in the same level as previous measurements performed by IVL with concentrations ranging between 2-15 µg/kg dw for sediment and 0.05- 0.17ng/m³ for air (Remberger et al, 2002).

8.9 Parabenes

8.9.1 Background

Parabenes were not found in the background samples (LOD: water 0.01 - 0.02 µg/l; sediment 5 - 7; µg/kg biota 0.5-1 µg/kg fw).

8.9.2 Municipal STPs and landfills

Methylparabene was detected in all **sludge** samples from municipal STPs except for the sample taken in Kiruna (Figure 1). Highest concentrations were found in Borås, Uppvidinge and Göteborg at concentrations of 59 and 52 µg/kg dw respectively. The two samples from Göteborg were taken

with one week in between and showed substantial differences in concentrations. Propylparabene was found in four samples in concentrations ranging between 3.6 and 10 µg/kg dw. Ethylparabene and butylparabene were only detected in the sample from Hallstahammar STP in concentrations of 10 and 63 µg/kg dw respectively.

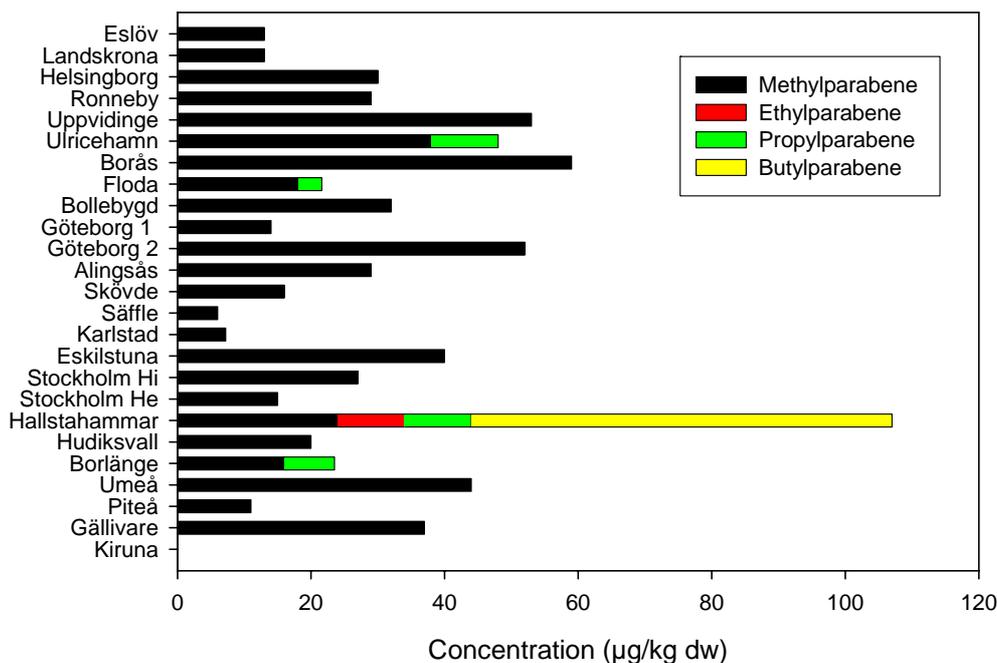


Figure 11. Concentrations of parabenes in sludge from municipal STPs

Two **influent water** samples to municipal STPs were analyzed. Propylparabene was detected in both of them in concentrations of 0.72 µg/l and 0.75 µg/l and butylparabene was detected in one of them in a concentration of 0.045 µg/l.

A cosmetics factory with known use of parabenes releases its effluent water to Borlänge STP. A **water effluent** sample from the factory contained ethylparabene (190 µg/l) and propylparabene (560 µg/l), but Borlänge STP did not show any elevated concentrations of the parabenes compared to other STPs, neither in sludge, nor in effluent water.

Propylparabene was the most frequently detected parabene in the **effluent water** samples from municipal STPs. Concentration ranged between 0.011 and 0.066 µg/l with the highest concentrations found in the sample from Piteå STP. The LOD was 0.01-0.02 µg/l. Methylparabene occurred in four samples in concentrations between 0.010 and 0.048 µg/l with the highest concentration in Helsingborg STP. Ethylparabene and butylparabene was found in one sample each in concentrations of 0.10 µg/l.

In a sample representing **storm drain water overflow** concentrations of parabenes were as follows: methylparabene 5.9 µg/l, ethylparabene 0.68 µg/l, propylparabene 0.28 µg/l and butylparabene <LOD. This was higher than concentrations in influent water to STPs regarding methyl- and ethylparabene but in the same order of magnitude for propylparabene.

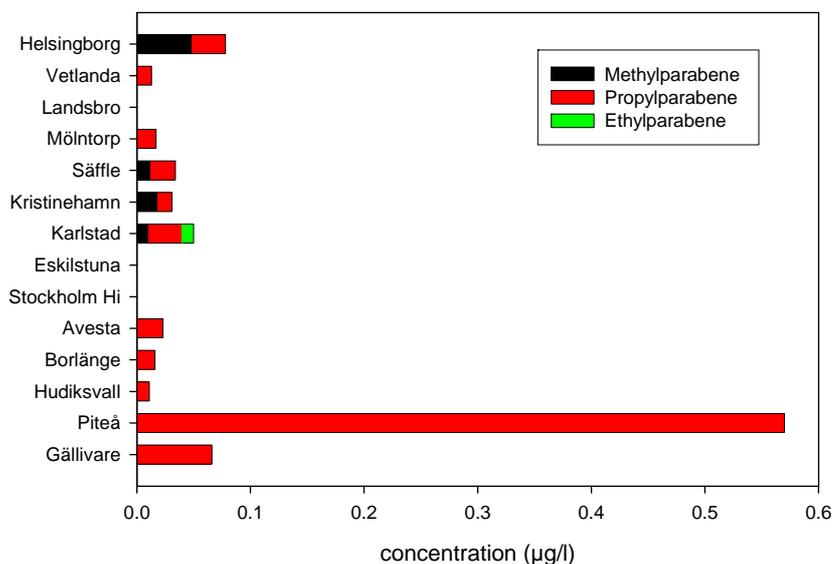


Figure 12. Concentrations of parabenes in effluent water from municipal STPs

8.9.3 Urban and industrial areas

Methylparabene was detected in the **biota** samples from Riddarfjärden and Årstaviken in Stockholm in concentrations of 1 and 2.5 µg/kg fw respectively.

Propylparabene was detected in the water from a **storm water well** at a wood impregnation factory area in a concentration of 0.011 µg/l, propylparabene as well as methylparabene was detected in **coast water** outside the factory in concentrations of 0.017 and 0.021 µg/l respectively. The detected concentrations are close to the detection limit of the background water samples and contribution from the factory can be disputed as they have no known use of the compounds.

An **atmospheric deposition** of methyl-, ethyl- and propylparabene occurred in the urban area Stockholm. The deposition fluxes were 1- 3.4, 2.6 -50 and <0.7 -1.7 ng/m² day respectively.

Parabenes were not found in **sediment** samples from urban or industrial areas (LOD: methyl-, ethyl-, propyl-, butylparabene 2 -6 µg/kg dw, benzylparabene 3 -9 µg/kg dw).

9 Summary and discussion

This chapter contains a summary of the previous section and a discussion regarding some of the results.

9.1 Substance specific discussion

Despite a high consumption of **bronopol** in Sweden as well as in the EU, it was not found in any of the samples analyzed. Bronopol undergoes rapid hydrolyzation as well as biodegradation, which may explain its absence in environmental samples.

4-chloro-3-cresol was detected in 60% of the sludge samples and in atmospheric deposition from Stockholm. Concentrations were lower compared to the other biocides and lower than measured concentrations in sludge in Great Britain. The consumption of this compound is limited and this together with the biodegradability may explain its low occurrence in the environment.

2-Mercaptobenzothiazole was found in substantial quantities in sludge and effluent water from municipal STPs. These findings are in good agreement with previous results (Paxéus 1996). Bacterial methylation of the thiol group to the methylthiobenzothiazole may occur in the aerated lagoons of the waste water treatment plants (Kloepfer et al., 2004). Unfortunately, it was not possible to analytically distinguish between these compounds since the sample was methylated during the analytical procedure.

2-Mercaptobenzothiazole is considered to be a rather recalcitrant in the environment (Haroune 2004) but the compound may oscillate between two forms depending on the redoxpotential: the reduced form, 2-mercaptobenzothiazole, and the oxidized form, the disulphide (Haroune et al. 2004; Svenson 1977; Manta et al. 2000).

In addition to sludge and sediment, 2-Mercaptobenzothiazole was also detected in the atmospheric deposition and sediment from the urban area of Stockholm as well as in one background sediment sample. The results thus indicate that there is an ongoing release of 2-mercaptobenzothiazole to the Swedish environment that derives from the diffuse usage of the substance. Therefore, it may be motivated to control the use, emission and environmental concentrations of this substance, and possibly also of its oxidized form, the disulphide. The 2-mercaptobenzothiazole is a high production volume chemical within the EU and its occurrence in deposition samples indicates that the atmosphere is a possible transport matrix for this chemical.

DDMAC was found in substantial quantities in sludge but not in effluent water from municipal STPs. Highest levels were found in Gässlösa STP in Borås, which receives input from the textile industry. DDMAC is commonly used as a fabric softener, which may explain its high occurrence in Gässlösa. The substance was also found in urban sediment but not in surface water. The results indicate an ongoing release of the substance to the environment but its high affinity to solid materials may decrease its bioavailability and hence its environmental effects. There is ambiguous data about the biodegradability of the compound and with consideration of the extensive use of DDMAC, accumulation of the compound in the environment can not be ruled out. DDMAC was not found in foodstuffs nor in human urine, thus human exposure is likely to be low.

Propiconazole was occasionally detected in sludge samples but not found in any environmental samples. It was, however, found in air at a roofed outdoor storage area for wood impregnation which indicates a possible release of the compound to the environment. The magnitude of this release is difficult to assess but samples of other matrices taken just outside the area did not contain any detectable amounts. The persistence of the compound could possibly indicate a potential for accumulation of the substance in the environment if the use increases. At present, the environmental impact regarding this substance is likely to be low.

Despite an extensive use of **resorcinol** both in Sweden and within the EU, the substance was only occasionally detected in sludge samples and not detected at all in any environmental samples. Similar to bronopol, this substance is readily biodegradable, which may explain the low occurrence in the environment.

2-(Thiocyanomethylthio)benzothiazole is expected to be non-biodegradable but may undergo chemical degradation to 2-(methylthio)benzothiazole (Agüera et al, 2000; Kloepfer et al, 2005) and

thereby contribute to the detected amount of 2-mercaptobenzothiazole since the latter compound was analyzed as the methylthioderivative (see 7.2.4). The compound was not found in any of the samples analyzed in this study, which may be a result of its chemical instability and limited use in Sweden and the EU.

Triclosan was found in high concentrations in STP sludge, particularly in the STP in Borlänge (43000 µg/kg dw). This may be a reflection of the use of triclosan in health care products as Borlänge receives wastewater input from a cosmetics plant. Triclosan was occasionally detected in STP effluent water, with highest levels in Piteå and Bräcke STP. Triclosan is also found in precipitation, urban atmospheric deposition, in urban air and in urban sediment. The results confirm a previous study (Remberger et al, 2002), which indicated an ongoing release of the substance to the environment and spreading of the compound through both STP pathways and via the atmosphere. The present study shows that, despite a stated decreased usage of the compound within the industry, levels in the environment as well as in STP samples are not declining.

Metylparabene and to a lower extent **propylparabene** were found in sludge samples. Hallstahammar STP deviated from the others, showing detectable concentrations of all parabenes, particularly butyl parabene. Parabenes, especially propylparabene, were also found in effluent water from STPs. Elevated concentrations were measured in the effluent water from one point source (cosmetic plant). This effluent is lead to a municipal STP (Borlänge) where concentrations in effluent water were in the same range as other municipal STPs. This illustrates the efficient removal of parabenes in sewage treatment plants. One exception of this is Piteå STP where levels of parabenes were about 10 times higher compared to other STPs. Piteå STP also stands out with high concentrations of triclosan (see above) and pharmaceuticals (Andersson et al., 2006). Methyl-, ethyl- and propylparabenes were also detected in deposition samples from the urban area in Stockholm and methylparabene was found in biota. The results thus indicate that there is an ongoing release of the substances to the environment and that the atmosphere is a possible transport matrix. For propylparabene, STP effluents may also play a certain role, even if efficient removal seems to take place within the STPs. Environmental data about the parabenes are limited in literature but the modeling exercise indicates a similar distribution pattern but a difference in overall residence time with propyl as the most persistent parabene.

9.2 Risk assessment MEC/PNEC

A way to relate the measured concentrations to known environmental effects is to perform a risk assessment and derive risk characterization quotients based on the measured concentrations (MEC) and the predicted no effect concentrations (PNEC). When the $MEC/PNEC \leq 1$ no negative effects of the substance are expected but when $MEC/PNEC \geq 1$ the substance is considered to be problematic in the environment and further investigations are needed (TGD, 2003). PNECs for the risk assessment are according to Table 4.

In order to use the effluent data from the STPs where no recipient water samples were provided a dilution factor of 10 was applied to all effluent concentrations (TGD, 2003). Since the PNEC calculation is associated with certain uncertainties depending on availability and quality of data, applied safety factors and considering that the screening results are not statistically reliable but rather “a snap shot” of the situation the results should be interpreted with care and regarded as indications to where there is a need for further investigations. This is especially important regarding the point source samples that are limited in number.

Within this preliminary risk assessment, the MEC/PNEC ratios for all substances were <1.

9.3 Variation between sampling site categories

The background sampling sites were generally non-contaminated, i.e. concentrations were generally below detection limits. Only 2 out of 17 background samples showed detectable concentrations of biocides. Of these, one was a sediment sample from Lilla Öresjön (2-mercaptobenzothiazole and triclosan; Figure 14). In the sampling procedure at this site, the sampling staff observed cottages, which were presumably not affiliated with STPs, which could contribute to the detected levels, in particular triclosan, which is a common substance within household products. The other background sample which showed detectable concentrations of biocides was a precipitation sample from Råö, which contained detectable amounts of triclosan.

The urban area showed signs of contamination in particular of 2-mercaptobenzothiazole, triclosan, DDMAC and parabenes. The two former occurred in urban sediments as well as in atmospheric deposition, whereas DDMAC was found only in sediments (Figure 13 and Figure 14). Parabenes were detected in deposition along with 4-Cl-3-cresol (Figure 13), and also in urban biota.

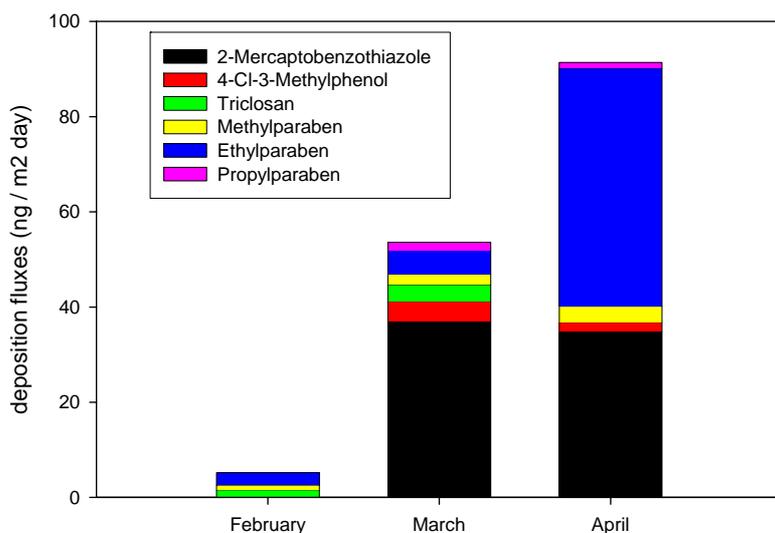


Figure 13. Deposition fluxes of biocides in Stockholm

Urban surface water was similar to background stations non-contaminated, i.e. concentrations were below detection limits.

As 2-mercaptobenzothiazole, triclosan and DDMAC were found in sediments, they may be expected to occur also in water and biota, especially given that the two former occurred also in deposition. The concentration expected to be found in water can be estimated assuming equilibrium partitioning between sediment and water. Doing this for 2-mercaptobenzothiazole as an example, and adopting an organic carbon content in the sediments (f_{oc}) of 0.07 kg/kg (Gustafsson, 2004), the water concentration corresponding to observed sediment concentrations can be estimated according to:

$$C_s = f_{OC} \times K_{OC} \times C_w + f_{BC} \times K_{BC} \times C_w^{0.7} \text{ (Schwarzenbach et al., 2003),}$$

where C_s is the concentration in sediments (mol/kg), K_{OC} is the organic carbon-water partition coefficient; $= 0.41 \times K_{OW}$, C_w the concentration in water (mol/m³), and K_{BC} is related to the black carbon content in the sediments. The impact of black carbon is presumably low for hydrophilic compounds such as 2-mercaptobenzothiazole and was thus assumed to be negligible. Using an average sediment concentration of 40 µg/kg dw (dw = 23 %) gives a water concentration of 0.0012 µg/l, which is nearly 100 times lower than the detection limits for water and below the PNEC value of 0.25 µg/l. The corresponding biota concentrations would be 0.02 µg/kg fw, which is 500 times lower than the detection limits. Thus, even if the compound is present in the water column, the concentrations are likely to be too low for detection, and also below the risk levels.

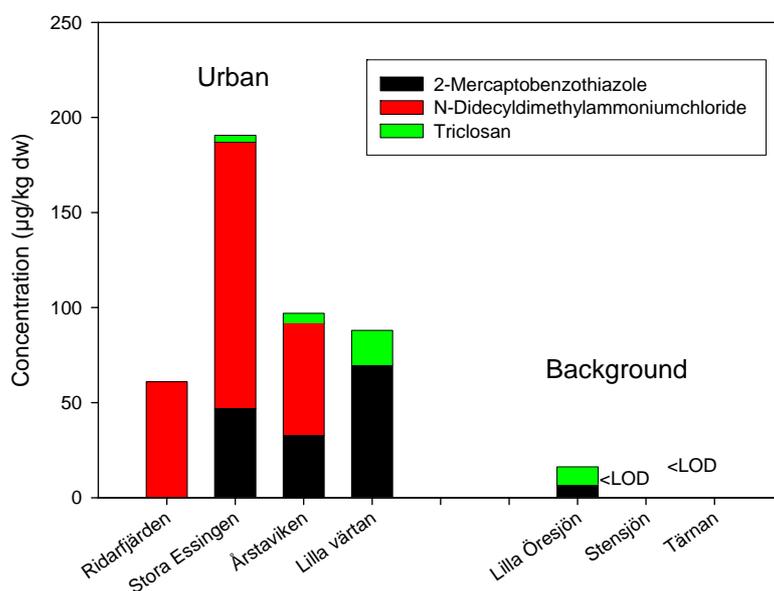


Figure 14. Concentrations of biocides in urban sediments, as compared to background stations

Among the pre-identified point sources; the harbor area; the wood impregnation industry and industrial effluents from known users, the determined chemicals did not occur in such extent as might have been expected; they were e.g. not found in surface water or sediment in the harbor area. **DDMAC** was found in sludge from the glue factory in Värmland but the concentration was lower than what was found at municipal STPs. Resorcinol and bronopol were analyzed but not detected in the effluent water from the same factory.

Propiconazole was found in outgoing air from the wood impregnation plant and in water and sediment from a plugged well on the premises from the same factory, but it appeared absent in the samples taken outside the factory. Regarding the limited number of analyzed samples, and considering the stability of this compound further measurements in the area may be motivated.

Parabenes were detected in the outgoing water from a cosmetics factory, but the receiving STP showed no elevated concentration of these compounds, indicating efficient removal from the wastewater.

9.4 Samples from the national surveillance program of sludge

All substances except bronopol were analyzed in the STPs, included in the national surveillance program for sludge (Haglund and Olofsson, 2006). The distribution of the different substances between the different STPs are illustrated in Figure 15. DDMAC, triclosan and methylparabene were the most frequently found substances with detectable concentrations in all STPs, followed by 2-mercaptobenzothiazole and 4-chloro-3-cresol, which were found in four of the STPs. Highest concentrations were detected for DDMAC and triclosan followed by 2-mercaptobenzothiazole, 4-chloro-3-cresol and methylparabene with average concentrations according to the following order: 21 325 > 3 187 > 393 > 33 > 32 µg/kg dw. The concentrations of other substances were below LOD.

The variability of concentrations between the different STPs was most pronounced for DDMAC where concentrations varied from 3 000 µg/kg dw in Floda to 61 000 µg/kg dw in Gässlösa STP in Borås. Gässlösa STP showed a total biocide concentration of up to 6.5 times higher than the other STPs, which illustrates the use of biocides within the textile industry. Resorcinol was only found in Gässlösa STP (160 µg/kg dw) and propylparabene was only detected in Floda STP (3.6 µg/kg dw; to little to be shown in Figure 15).

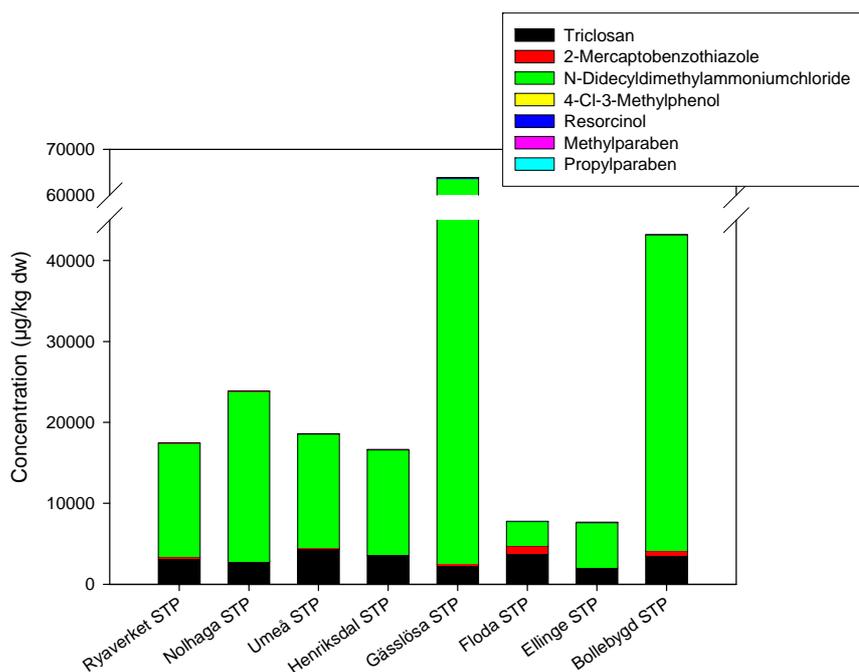


Figure 15. Concentrations of biocides in sludge from STPs.

10 Conclusions

Swedish background sites are generally non-contaminated with respect to biocides.

For four of the substances on-going emissions are likely to occur as they are regularly found in wastewater effluents, in STP sludge and in urban sediments: 2-Mercaptobenzothiazole, triclosan, DDMAC and parabenes. Observed concentrations are, however, below risk levels.

Three biocides were not at all detected in environmental samples and with a few exceptions not in samples from diffuse or point sources either. Thus, they are considered to cause no environmental problems: Bronopol, Resorcinol and 2-(thiocyanomethylthio)-benzothiazole.

Propiconazole may be of local concern but at present it is unlikely to cause any problems on a national level.

Four municipal STPs were identified as containing high levels of certain biocides: Piteå (2-mercaptobenzothiazole, triclosan, parabenes in effluent water), Borlänge (triclosan in sludge), Gässlösa (DDMAC in sludge) and Hallstahammar (parabenes in sludge).

All samples had MEC/PNEC ratios <1, thus at current levels, biocides are not expected to give rise to any environmental effects.

The atmosphere is identified as a possible transport matrix for triclosan, 2-mercaptobenzothiazole and parabenes.

11 Acknowledgements

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Appendix 1. Aquatic toxicity (IMBL: Immobilisation, GRO: growth, MOR: mortality, REP: Reproduction, ITX: intoxication, NOC; MATC; Maximum acceptable toxic concentration)

	Scientific name, Common name	Endpoint	Effect Measure	Duration	Conc (mg/L)
Bronopol¹	Americamysis bahia	LC50	MORT	96 h	5.9
	Opossum shrimp				
	Crassostrea gigas	EC50	IMBL	48 h	0.78
	Pacific oyster				
Mercapto-benzothiazol²	Cyprinodon variegatus	LC50	MORT	96 h	59
	Sheepshead minnow				
	Daphnia Magna	LC50	IMBL	48h	1.8
	Water Flea				
	Daphnia Magna	NOEC	REP	21 d	0.34
	Water Flea				
N-didecyldimethyl ammoniumchloride¹	Selenastrum capricornutum	LC50	GGRO	96h	0.25
	Green algae				
	Onchorhynchus Mykiss	Embryo survival;		89 d	0.041-0.078
	Rainbow trout, Donaldsson trout	MATC			
N-didecyldimethyl ammoniumchloride¹	Chlorella pyrenoidosa	EC50	GGRO	24 h	0.059-0.280 (F)
	Green algae				
	Daphnia magna	EC50	MORT	48 h	0.037
	Water flea				
	Hyalella azteca	LC50	MORT	48 h	0.110
	Scud				
	Lepomis macrochirus	LC50	MORT	96 h	0.270
	Bluegill				
	Oncorhynchus kisutch	LC50	MORT	96 h	0.390
	Coho salmon, silver salmon				
2-(Thiocyanomethylthio) benzothiazole¹	Daphnia Magna ⁵	LC50	MOrt	96	0,093 (F)
	Water Flea				
4-Chloro-3-cresol	Daphnia Magna ⁵	NOEC	Fecundity	7 days	0,037(F)
	Water Flea				
	Pimephales promelas	LC50	MORT	96 h	0.330
2-(Thiocyanomethylthio) benzothiazole¹	Fathead minnow				
	Oncorhynchus mykiss	LC50	MORT	96 h	0.420 (F)
	Rainbow trout, donaldson trout				
	Tetrahymena pyriformis	EC50	GGRO	24 h	10 (F)
	Ciliate				
	Carassius auratus		MORT	48 h	2 (F)
4-Chloro-3-cresol	Goldfish				
	Daphnia Magna ⁴	NOEC	MORT	21 d	0,0038
4-Chloro-3-cresol	Water Flea				
	Daphnia magna ¹	NOEC	GREP	21 d	1.3
	Water flea				
	Oncorhynchus mykiss ²	LC50	MORT	96 h	0.917
	Rainbow trout, donaldson trout				
	Scenedesmus subspicatus ²	EC50	BMAS	96 h	>10 (F)
Propiconazole³	Green algae				
	Scenedesmus subspicatus ²	EC50	BMAS	72 h	1.85 (F)
Propiconazole³	Green algae				
	Leiostomus xanthurus	LC50	MORT	96h	2.6-6.2
	Fish				
	Daphnia Magna	EC50	IMBL	48h	2.2-11.5
	Water Flea				
	Daphnia Magna	NOEC	REP	21d	0,31
Propiconazole³	Water Flea				
	Skeletonema Costatum diatom(marine algae)	EC50		11d	0.021

	Scientific name, Common name	Endpoint	Effect Measure	Duration	Conc (mg/L)
	Anabena flos-aquae Green algae	EC50		11d	6.78
Resorcinol²	Leuciscus Idus (fish)	LC50		96 h	34.7
	Brachydanio rerio (fresh water fish)	LC50		7d (early life stages)	262 (embryo)
	Brachydanio rerio (fresh water fish)	EC50		7d (early life stages)	54.8 (malformations)
	Brachydanio rerio (fresh water fish)	NOEC		7d (early life stages)	25
	Daphnia Magna (crustacea)	LC50		48 h	0.25
	Daphnia magna (crustacea)	LC50		24 h	107.6
	Palaemonetes pugio (Grasgarnelle, aquatic artropod)	LC50		96 h	42.2
	Elodea Canadensis (canadian pondweed)	LC50		9 days	143.1
Triclosan	Chlorella Pyrenoidea (algae)	LC50		72 h	1.1
	Scenedesmus (algae)	LC50		4 days	60
	Zebra fish (Brachidanio rerio)	LC50		96h	0.54mg/l
	Rainbow Trout (Oncorhynchus mykiss)	LC50		96h	0.35 mg/l
	Golden Orfe (fish; Leuciscus idus)	LC50		96h	0.56 mg/l
	Fathead Minnow (fish; Pimephales promelas)	LC50		96h	0.26 mg/l
	Daphnia Magna	EC50		48h	0.39 mg/l
	Ceriodaphnia dubia	EC50		48 h	0.13 mg/l
	Rainbow trout (fish)	NOEC		(Early life stage toxicity (ELS))	24 µg/l
	Daphnia magna (crustacea)	NOEC		21 days (OECD 202)	40 µg/l
	Ceriodaphnia dubia (crustacea)	NOEC		7days	6 µg/l
	Scenedesmus subspicatus (algae)	EC50		96h	1.4 µg/l
	Scenedesmus subspicatus (algae)	NOEC		96h	0.5 µg/l

¹ US EPA, 2006b

² ESIS, 2006

³ Kemi, 1997

⁴ Forsgren et al., 2004

⁵ Juergensen et al, 2000

(F) Not measured concentrations

Appendix 2. Sample information regional samples.

County	City	Site	Matrix	Dimension (STP)	Affiliated (STP) pe	STP-treatment	Site information	Coordinates	Sampling date	DW, %	Sample ID
Dalarna	Avesta	Krylbo STP	effluent	30000	17000	Pre-sedimentation/ active sludge	Steal, brewery, slaughterhouse, industry landfill, metal, wood, small industries, car wash, waste incineration		2005-10-05 kl.07.30		4162
Dalarna	Avesta	Krylbo STP	sludge	30000	17000	Pre-sedimentation /active sludge	Steal, brewery, slaughterhouse, industry landfill, metal, wood, small industries, car wash, waste incineration		2005-10-05 kl.09.00	23	4163
Dalarna	Avesta	Lake Bäringen	sediment						2005-10-06	39	4175
Dalarna	Avesta	Lake Bäringen	Biota, perch						2005-10-06		4177
Dalarna	Avesta	Lake Bäringen	Surface water						2005-10-06		4178
Dalarna	Borlänge	Fagersta By STP	effluent	60000	44000	Pre-sedimentation, simultaneous sedimentation and active sludge	Household, smaller industries, car wash, landfill		2005-10-05		4158
Dalarna	Borlänge	Fagersta By STP	sludge	60000	44000	Pre-sedimentation, simultaneous sedimentation and active sludge	Household, smaller industries, car wash, landfill		2005-10-05	29	4160
Dalarna	Borlänge	Fågelmyra Landfill	leachate						2005-10-05		4157
Jönköping	Landsbro	Landsbro STP	influent						2005-11-02		4330
Jönköping	Landsbro	Landsbro STP	effluent						2005-11-02		4332
Jönköping	Vetlanda	Vetlanda STP	influent						2005-11-02		4334
Jönköping	Vetlanda	Vetlanda STP	effluent						2005-11-02		4336
Stockholm	Stockholm	Himmerfjärden STP	effluent						2005-10-06		4171
Stockholm	Stockholm	Himmerfjärden STP	sludge					x=49,67 y=80,302	2005-10-06	21	4173
Stockholm		Himmerfjärden	sediment				0-3 cm, 22 meters depth	6549185; 1609285		33	4360
Stockholm		Lilla Värtan	sediment				0-3 cm, 25 meters depth	6581100; 1634095		19	4361
Stockholm		Norrtäljeviken	sediment				0-3 cm, 22 meters depth	6634360; 1680790		42	4359
Stockholm		St Envättern	sediment				0-3 cm, 12 meters depth	6556175; 1587935		11	4362
Värmland	Karlstad	Landfill Djupdalen	leachate after aeration pond						2005-10-26		4279

County	City	Site	Matrix	Dimension (STP)	Affiliated (STP) pe	STP-treatment	Site information	Coordinates	Sampling date	DW, %	Sample ID
Värmland	Karlstad	Sjöstad STP	effluent	97000	55800	mech/chem/biol and nitrogen reduction	Regional hospital, food industry (dairy, charcutery, bakery), dental clinics, chemical industry, smaller metal industry, waste incineration plant		2005-10-11/12		4200
Värmland	Karlstad	Sjöstad STP	sludge	97000	55800	mech/chem/biol and nitrogen reduction	Regional hospital, food industry (dairy, charcutery, bakery), dental clinics, chemical industry, smaller metal industry, waste incineration plant			28	4201
Värmland	Karlstad	Tjärnsjön	sediment				Previous textile industries		2005-11-07	23	4352
Värmland	Kristinehamn	Bergsjön	surface water					6586800;1408540	2005-10-17		4246
Värmland	Kristinehamn	Casco Adhesives AB	effluent				Producers of glue and registered use of resorcinol, bronopol och N-didecyldimetylammoniumklorid		2005-10-24		4286
Värmland	Kristinehamn	Casco Adhesives AB	sludge				Producers of glue and registered use of resorcinol, bronopol och N-didecyldimetylammoniumklorid.		2005-10-24		4287
Värmland	Kristinehamn	Fiskartorpet STP	effluent	18000	16300	mech/chem/biol and nitrogen reduction	Hospital, industries(mainly metal industries)	6578124; 1401080	2005-10-18		4248
Värmland	Kristinehamn	Fiskartorpet STP	sludge	18000	16300	mech/chem/biol and nitrogen reduction	Hospital, industries(mainly metal industries)	6578124; 1401080	2005-10-17	23	4250
Värmland	Kristinehamn	Landfill Strandmossen	leachate						2005-11-01		4323
Värmland		Landfill Strandmossen	sediment						2005-11-01	22	4325
Värmland	Kristinehamn	Varnumsviken	surface water						2005-10-12		4238
Värmland	Säffle	Säffle STP	effluent	15000	7750	mech/chem/biol and nitrogen reduction	Hospital		2005-10-18		4262
Värmland	Säffle	Säffle STP	sludge	15000	7750	mech/chem/biol and nitrogen reduction	Hospital		2005-10-18	31	4263
Västerbotten	Lycksele	STP	sludge		9000		industrial , hospital		2005-11-02	25	4344
Västerbotten	Skellefteå	Tuvan STP	sludge		43000			175009	2005-10-24	23	4305
Västra Götaland	Göteborg	Kodammarnas pumping station	water						2005-12-20		4472
Västra Götaland	Rya	GRYAAB	sludge						2005-10-12		4202
Västra Götaland	Ulricehamn	Ulricehamn STP	sludge						2005-10-10		4226

Appendix 3. Sample information national samples

Sample type	City	Site	Matrix	Dimension (STP)	Remarks	Coordinates	Sampling date	DW, %	Sample ID
National, Background		Pallas A	air				2005-12-19 till 2006-01-02		4594
National, Background		Pallas B	air				2006-02-22/03-08		4706
National, Background		Pallas C	air				2006-04-13/24		4791
National, Background		Råö A	air				2005-12-19/27		4495
National, Background		Råö B	air				2006-02-08/14		4622
National, Background		Råö C	air				2006-05-03/11		4758
National, Background		Råö B	precipitation				2006-01-31 - 2006-03-01		4677
National, Background		Råö C	precipitation				060503-060601		4800
National, Background		Lilla Öresjön	surface water				2006-01-12		4483
National, Background		Lilla Öresjön	sediment				2004-08-31	12	4504
National, Background		Lilla Öresjön	fish						4686
National, Background		Stora Envättern	surface water				2005-11-13		4367
National, Background		Stensjön	sediment				2004-08-12	15	4506
National, Background		Stensjön	fish						4688
National, Background		Tärnan	surface water			X=6608828; Y=1645102			4368
National, Background		Tärnan	sediment				2004-10-06	25	4505
National, Background		Tärnan	fish						4687
National, Diffuse	Stockholm	Stockholm A	air				2006-02-05/22		4632
National, Diffuse	Stockholm	Stockholm B	air				2006-03-15/22		4692
National, Diffuse	Stockholm	Stockholm C	air				2006-04-12/19		4726
National, Diffuse		Stockholm A	Deposition				2006-02-15/03-16		4685
National, Diffuse		Stockholm B	Deposition				2006-03-16/04-13		4721
National, Diffuse		Stockholm C	Deposition				2006-04-13/05-17		4764
National, Diffuse		Stockholm A	precipitation				2006-02-15 - 2006-03-15		4634-82
National, Diffuse		Stockholm B	precipitation				2006-03-15 - 2006-4-12		4693-4719
National, Diffuse		Stockholm C	precipitation				2006-04-12/19		4727
National, Diffuse	Stockholm	Riddarfjärden	surface water			6580118; 1627286	2005-11-23		4388
National, Diffuse	Stockholm	Riddarfjärden	sediment			6580118; 1627286	2005-11-23	28	4484
National, Diffuse	Stockholm	Riddarfjärden	perch		3-7 years; 65-266 g	6580118; 1627286	2005-11-23		4488
National, Diffuse	Stockholm	Stora Essingen	surface water			6579225; 1623638	2005-11-23		4386

Sample type	City	Site	Matrix	Dimension (STP)	Remarks	Coordinates	Sampling date	DW, %	Sample ID
National, Diffuse	Stockholm	Stora Essingen	sediment			6579225; 1623638	2005-11-23	21	4485
National, Diffuse	Stockholm	Stora Essingen	perch		2-4 years; 40-61 g	6579225; 1623638	2005-11-23		4489
National, Diffuse	Stockholm	Årstaviken	surface water			6578147; 1628330	2005-11-23		4387
National, Diffuse	Stockholm	Årstaviken	sediment			6578147; 1628330	2005-11-23	24	4486
National, Diffuse	Stockholm	Årstaviken	perch		2-4 years; 48-76 g	6578147; 1628330	2005-11-23		4491
National, Diffuse	Stockholm	Årstaviken	roach		118 g	6578147; 1628330	2005-11-23		4492
National, Diffuse	Stockholm	Mälaren, Broviken	sediment				2006-03-29	44	4709
National, Diffuse	Stockholm	Mälaren, Broviken	sediment				2006-03-29	44	4710
National, diffuse	Bräcke	Bräcke STP	effluent						4185
National, diffuse	Eskilstuna	Ekeby STP	effluent				2005-10-04		4165
National, Diffuse	Borås	Gässlösa STP	effluent				2006-01-24		4573
National, Diffuse	Stockholm	Henriksdal STP	effluent						4585
National, diffuse	Gällivare	Kavaheden STP	effluent	20 000	Hospital, health centers	7287700;1795760	2005-10-12		4233
National, diffuse	Hallstahammar	Mölnortorp STP	effluent	25000	1300 pe from industri	6602672	2005-10-12		4207
National, diffuse	Hudiksvall	Reffelmansverket	effluent			6844618;1568169	2005-10-26		4302
National, diffuse	Ronneby	Rustorp STP	effluent				2005-10-11		4195
National, diffuse	Piteå	Sandholmen STP	effluent	35000	Hospital, health centers	7458000;1714000	2005-10-19		4271
National, diffuse	Skövde	Stadskvarn STP	effluent				2005-10-12/13		4215
National, diffuse	Helsingborg	Öresundsverket	effluent				2005-10-25		4288
National, Diffuse		Bollebygd	sludge				2005-10-17	12	4404
National, diffuse	Eskilstuna	Ekeby STP	sludge				2005-10-04	28	4168
National, Diffuse	Eslöv	Ellinge	sludge				2005-10-20	16	4398
National, Diffuse		Floda	sludge				2005-10-20	31	4403
National, Diffuse	Borås	Gässlösa STP	sludge				2006-01-24	5,2	4574
National, Diffuse	Stockholm	Henriksdal STP	sludge				2005-10-18	31	4397
National, diffuse	Gällivare	Kavaheden STP	sludge	20 000	Hospital, health centers	7287700;1795760	2005-10-12	20	4234
National, Diffuse	Kiruna	Kiruna STP	sludge					19	4572
National, diffuse	Hallstahammar	Mölnortorp STP	sludge	25000	1300 pe from industri	6602672	2005-10-12	27	4208
National, Diffuse	Alingsås	Nolhaga	sludge				2005-10-18	22	4399
National, diffuse	Hudiksvall	Reffelmansverket	sludge			6844618;1568169	2005-10-26	28	4303

Sample type	City	Site	Matrix	Dimension (STP)	Remarks	Coordinates	Sampling date	DW, %	Sample ID
National, diffuse	Ronneby	Rustorp STP	sludge				2005-10-11	18	4196
National, Diffuse	Göteborg	Ryaverket	sludge				2005-10-19	27	4402
National, diffuse	Piteå	Sandholmen STP	sludge	35000	Hospital, health centers	7458000;1714000	2005-10-19	25	4273
National, diffuse	Skövde	Stadskvarn STP	sludge				2005-10-12	17	4217
National, Diffuse	Landskrona	STP	sludge					22	4414
National, Diffuse	Umeå	Umeå	sludge				2005-10-19	28	4401
National, Diffuse	Uppvidinge	Åseda STP	sludge					18	4481
National, diffuse	Helsingborg	Öresundsverket	sludge				2005-10-25	23	4290
National, Point source		Wood impreg. plant Smp A	air				2006-01-25 - 02-01		4591
National, Point source		Wood impreg. plant Smp B	air						4474
National, Point source		Wood impreg. plant Smp C	air				2006-02-22 - 03-01		4645
National, point source		Wood impreg. plant	Stain bath				2005-12-15		4450
National, point source		Wood impreg. plant	Stain bath				2005-12-15		4451
National, Point source		Wood impreg. plant	water		storm water well		2005-12-15		4441
National, Point source		Wood impreg. plant	surface water			6726965; 1588288	2005-12-15		4445-4446
National, Point source		Wood impreg. plants	surface water		sea water		2005-12-15		4447-4448
National, Point source		Wood impreg. plant	sediment		storm water well		2005-12-15	69	4443
National, Point source	Fiskebäckskil	Harbour	shellfish						4778
National, Point source	Fiskebäckskil	Harbour	sediment					31	4779
National, Point source	Fiskebäckskil	Harbour	surface water						4780
National, Point source		Cosmetics plant	effluent				2005-12-15		4453
National, point source	Bro	Högbytorp	leachate before treatment						4381
National, point source	Bro	Högbytorp	leachate after treatment						4382
National, point source		Deponi	leachate						4609
National, human exp		food 1	meat						4097
National, human exp		food 2	dairy products						4102
National, human exp		food 3	vegetables						4113
National, human exp		food 4	cultivated salmon						4588

Sample type	City	Site	Matrix	Dimension (STP)	Remarks	Coordinates	Sampling date	DW, %	Sample ID
National, human exp		food 5	wild salmon						4589
National, human exp		food 6	wild salmon						4590
National, human exp		human urine 1	urine						4751
National, human exp		human urine 2	urine						4962
National, human exp		human urine 3	urine						4963
National, human exp		human urine 4	urine						4964
National, human exp		human urine 5	urine						4965
National, human exp		human urine 6	urine						4966

Appendix 4. Results regional samples. n.c = not commissioned, n.a. = not analysed.

Sample ID	County	City	Matrix	Unit	2-Mercapto-benzothiazole	2-(Tiocyanmet)benzotiazol	Bronopol	4-Chloro-3-methylphenol	N-Didecylimethylammonium chloride	Propiconazole	Triclosan	Resorcinol	Methylparaben	Ethylparaben	Propylparaben	Butylparaben	Benzylparaben
4162	Dalarna	Avesta	effluent	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	<0.01	<0.02	<0.01	<0.01	<0.02	0.023	<0.01	<0.01
4158	Dalarna	Borlänge	effluent	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	<0.01	<0.02	<0.01	<0.02	<0.01	0.016	<0.01	<0.01
4332	Jönköping	Landsbro	effluent	µg/l	n. c.	n. c.	n. c.	<0.01	n. c.	n. c.	n. c.	n. c.	<0.01	<0.01	<0.01	<0.01	<0.01
4336	Jönköping	Vetlanda	effluent	µg/l	n. c.	n. c.	n. c.	<0.01	n. c.	n. c.	n. c.	n. c.	<0.01	<0.01	0.013	<0.01	<0.01
4171	Stockholm	Himmerfj. STP	effluent	µg/l	0.38	<0.05	<0.5	<0.01	0.17	<0.01	0.020	0.078	<0.02	<0.01	<0.02	<0.01	<0.01
4200	Värmland	Karlstad	effluent	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	<0.01	<0.02	n.a.	0.010	0.010	0.030	<0.01	<0.01
4248	Värmland	Kristinehamn	effluent	µg/l	<0.05	<0.05	<0.05	<0.01	n. c.	<0.01	0.045	<0.02	0.018	<0.01	0.013	<0.01	<0.01
4286	Värmland	Kristinehamn Casco adhesive	effluent	µg/l	n. c.	n. c.	<0.05	n. c.	n. c.	n. c.	n. c.	<0.04	n. c.	n. c.	n. c.	n. c.	n. c.
4262	Värmland	Säffle	effluent	µg/l	0.2	<0.05	n. c.	<0.01	n. c.	<0.01	<0.02	n.a.	0.012	<0.01	0.022	<0.01	<0.01
4330	Jönköping	Landsbro	influent	µg/l	n. c.	n. c.	n. c.	0.023	n. c.	n. c.	n. c.	n. c.	<0.02	<0.04	0.75	0.045	<0.02
4334	Jönköping	Vetlanda	influent	µg/l	n. c.	n. c.	n. c.	0.055	n. c.	n. c.	n. c.	n. c.	<0.03	<0.04	0.72	<0.05	<0.04
4157	Dalarna	Borlänge	leachate	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	<0.01	<0.02	<0.03	0.031	<0.02	<0.03	<0.01	<0.2
4323	Värmland	Kristinehamn	leachate	µg/l	<0.05	<0.05	0.05	<0.01	n. c.	<0.01	<0.005	<0.05	<0.01	<0.02	<0.01	<0.01	<0.01
4279	Värmland	Karlstad	leachate after aeration pond	µg/l	18	<0.05	n. c.	<0.01	<1	0.03	<0.2	<0.01	<0.01	0.057	<0.01	<0.01	<0.01
4177	Dalarna	Avesta	perch	µg/kg FW	<10	<10	n. c.	<0.5	n. c.	<10	<0.1	<5	2.5	<0.5	0.4	<0.5	<1
4175	Dalarna	Avesta	sediment	µg/kg DW	<20	<50	n. c.	<3	n. c.	<5	<1	<10	<3	<3	<3	<3	<4
4360	Stockholm	Himmerfjärden	sediment	µg/kg DW	<25	<50	<6	<2	<50	<2	29	<5	<2	<2	<2	<2	<4
4361	Stockholm	Lilla värtan	sediment	µg/kg DW	70	<50	<10	<9	<50	<5	18	<5	<9	<9	<9	<9	<41
4359	Stockholm	Norräljeviken	sediment	µg/kg DW	<20	<50	<10	<2	<50	<5	<1	<5	<2	<2	<2	<2	<4
4362	Stockholm	St. Envättern	sediment	µg/kg DW	<20	<50	<10	<20	<50	<5	<1	<10	<20	<20	<20	<20	<32
4352	Värmland	Karlstad	sediment	µg/kg DW	n. c.	n. c.	n. c.	<5	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4325	Värmland	Kristinehamn	sediment	µg/kg DW	<20	<50	n. c.	<5	<50	<5	<1	<10	<5	<5	<5	<5	<7
4163	Dalarna	Avesta	sludge	µg/kg DW	190	<50	n. c.	18.0	n. c.	<5	4400	<10	13.0	<5	<5	<5	<8
4160	Dalarna	Borlänge	sludge	µg/kg DW	200	<50	n. c.	13.0	n. c.	<5	43 000	<10	16.0	<4	7.5	<4	<6
4173	Stockholm	Himmerfj. STP	sludge	µg/kg DW	<20	<0.05	<24	7.0	12 000	<5	3900	<10	27	<5	<5	<5	<8
4201	Värmland	Karlstad	sludge	µg/kg DW	<20	<0.05	n. c.	40.0	n. c.	<5	5 500	<10	7.2	<4	<4	<4	<6

Sample ID	County	City	Matrix	Unit	2-Mercapto-benzothiazole	2-(Tiocyanomet hyltio) bensotiazol	Bronopol	4-Chloro-3-methylphenol	N-Didecyl dimethyl ammonium chloride	Propiconazole	Triclosan	Resorcinol	Methylparaben	Ethylparaben	Propylparaben	Butylparaben	Benzylparaben
4250	Värmland	Kristinehamn	sludge	µg/kg DW	n. c.	n. c.	n. c.	n. c.	7 600	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4287	Värmland	Kristinehamn Casco adhesive	sludge	µg/kg DW	n. c.	n. c.	n. c.	n. c.	220	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4263	Värmland	Säffle	sludge	µg/kg DW	<20	<50	n. c.	<4	n. c.	<5	2000	<10	6.0	<4	<4	<4	<6
4344	Västerbotten	Lycksele	sludge	µg/kg DW	n. c.	n. c.	<16	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4305	Västerbotten	Skellefteå	sludge	µg/kg DW	n. c.	n. c.	<15	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4202	Västra Götaland	Rya	sludge	µg/kg DW	<20	<50	<12	21	n. c.	<5	3400	<10	14	<4	<4	<4	<6
4226	Västra Götaland	Ulricehamn	sludge	µg/kg DW	36	<50	<17	19	n. c.	<5	5600	22	38	<10	10.0	<5	<9
4178	Dalarna	Avesta	Surface water	µg/l	<0.05	<50	n. c.	<0.01	n. c.	<0.01	<0.005	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
4238	Värmland	Kristinehamn	surface water	µg/l	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	<0.01	n. c.	n. c.	n. c.	n. c.	n. c.
4246	Värmland	Kristinehamn	surface water	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	<0.01	<0.005	<0.01	<0.02	<0.01	<0.01	<0.01	<0.01
4472	Västra Götaland	Göteborg, Kodammarna	Drain overflow sewage water	µg/l	0.08	<0.05	<0.05	0.11	n. c.	<0.01	0.46	<0.03	5.9	0.68	3.0	0.28	<0.01

Appendix 5. Results national samples n.c = not commissioned, n.a. = not analysed.

Sample ID	County/ National	City	Site	Matrix	Unit	2-Mercapto benzothiazole	2-(Thio- cyano- methyl- tio)- benso- tiazol	Brono- pol	4-Cl-3- Methyl phenol	N- Didecyldi methyl- ammo- nium- chloride	Propicon azole	Tri- closan	Reso- rcinol	Methyl para- ben	Ethyl para ben	Propyl- paraben	Butyl- para- ben	Benzyl para- ben
4594	Background		Pallas A	air	ng/m ³	n. c.	n. c.	<0.07	n. c.	n. c.	n. c.	<0.00 2	<0.05	n. c.	n. c.	n. c.	n. c.	n. c.
4706	Background		Pallas B	air	ng/m ³	n. c.	n. c.	<0.07	n. c.	n. c.	n. c.	<0.00 2	<0.05	n. c.	n. c.	n. c.	n. c.	n. c.
4791	Background		Pallas C	air	ng/m ³	n. c.	n. c.	<0.07	n. c.	n. c.	n. c.	<0.00 2	<0.05	n. c.	n. c.	n. c.	n. c.	n. c.
4495	Background		Råö A	air	ng/m ³	n. c.	n. c.	<0.05	<0.03	n. c.	n. c.	<0.00 2	<0.05	<0.6	<0.0 3	<0.3	<0.03	<0.03
4622	Background		Råö B	air	ng/m ³	n. c.	n. c.	<0.05	<0,03	n. c.	n. c.	<0.00 2	<0.05	<0.1	<0.0 3	<0.1	<0.03	<0.03
4758	Background		Råö C	air	ng/m ³	n. c.	n. c.	<0.05	<0.03	n. c.	n. c.	<0.00 2	<0.05	<0.2	<0.0 3	<0.1	<0.03	<0.03
4677	Background		Råö B	precipitation	µg/l	n. c.	n. c.	<0.05	n. c.	n. c.	n. c.	0.012	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4483	Background		Lilla Öresjön	surface water	µg/l	<0.05	<0.05	<0.05	<0.01	<0.05	<0.01	<0.00 3	<0.05	<0.02	<0.0 2	<0.01	<0.01	<0.01
4367	Background		Stora Envättern	surface water	µg/l	<0.05	<0.05	<0.05	<0.01	<0.05	<0.01	<0.00 3	<0.05	<0.01	<0.0 1	<0.01	<0.01	<0.01
4368	Background		Tärnan	surface water	µg/l	<0.1	<0.05	<0.05	<0.01	<0.05	<0.01	<0.00 5	<0.05	<0.01	<0.0 1	<0.01	<0.01	<0.01
4504	Background		Lilla Öresjön	sediment	µg/kg DW	7.2	<50	<17	<7	<50	<5	9.0	<20	<7	<7	<7	<7	<12
4506	Background		Stensjön	sediment	µg/kg DW	<20	<50	<11	<5	<50	<5	<2	<10	<5	<5	<5	<5	<7
4505	Background		Tärnan	sediment	µg/kg DW	<25	<50	<10	<6	<50	<2	<2	<5	<6	<6	<6	<6	<10
4686	Background		Lilla Öresjön	fish	µg/kg ww	<10	<10	<10	<0.5	n. c.	<10	<0.1	<5	<0.5	<0.5	<0.5	<0.5	<1
4688	Background		Stensjön	fish	µg/kg ww	<30	<10	<10	<0.5	n. c.	<10	<0.1	<5	<0.5	<0.5	<0.5	<0.5	<1
4687	Background		Tärnan	fish	µg/kg ww	<10	<10	<10	<0.5	n. c.	<10	<0.1	<5	<0.5	<0.5	<0.5	<0.5	<1
4632+	Diffuse		Stockholm A	air	Ng/m ³	<0.05	<0.01	<0.05	<0.04	n. c.	<0.002	<0.00 3	<0.05	0.06	<0.0 5	<0.03	<0.03	<0.03
4692+	Diffuse		Stockholm B	air	Ng/m ³	<0.2	<0.01	<0.05	<0.04	n. c.	<0.004	0.010	<0.05	0.05	<0.0 5	<0.03	<0.03	<0.03
4726+	Diffuse		Stockholm C	air	Ng/m ³	<0.2	<0.01	<0.05	<0.04	n. c.	<0.005	<0.00 3	<0.05	0.07	<0.0 5	<0.03	<0.03	<0.03
4685	Diffuse		Stockholm A	deposition	ng/m ² . d	<5	<	n. c.	<1	n. c.	<0.5	1.6	n. c.	1.0	2.6	<0.7	<0.7	<0.7

Sample ID	County/ National	City	Site	Matrix	Unit	2-Mercapto benzothia zole	2-(Tio- cyano- methyl- tio)- benso- tiazol	Brono- pol	4-Cl-3- Methyl phenol	N- Didecyldi methyl- ammo- nium- chloride	Propicon azole	Tri- closan	Reso- rcinol	Methyl para- ben	Ethyl para- ben	Propyl- paraben	Butyl- para- ben	Benzyl para- ben
4721	Diffuse		Stockholm B	deposition	ng/m ² . d	37	<	n. c.	4.2	n. c.	<0.5	3.5	n. c.	2.2	5.0	1.7	<0.7	<0.7
4764	Diffuse		Stockholm C	deposition	ng/m ² . d	35	<	n. c.	1.8	n. c.	<0.5	<0.5	n. c.	3.4	50	1.2	<0.7	<0.7
4634- 82	Diffuse		Stockholm A	precipitatio n	µg/l	n. c.	n. c.	<0.16	n. c.	n. c.	n. c.	<0.00 3	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4693- 4719	Diffuse		Stockholm B	precipitatio n	µg/l	n. c.	n. c.	<0.16	n. c.	n. c.	n. c.	<0.00 3	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4388	Diffuse		Riddarfjärden	surface water	µg/l	<0.1	<0.05	n.c.	<0.01	<0.05	<0.01	<0.00 2	<0.05	<0.05	<0.0 1	<0.01	<0.01	<0.01
4386	Diffuse		Stora Essingen	surface water	µg/l	<0.1	<0.05	n.c.	<0.01	<0.05	<0.01	<0.00 2	<0.05	<0.05	<0.0 1	<0.01	<0.01	<0.01
4387	Diffuse		Årstaviken	surface water	µg/l	<0.1	<0.05	n.c.	<0.01	<0.05	<0.01	<0.00 2	<0.05	<0.05	<0.0 1	<0.01	<0.01	<0.01
4484	Diffuse		Riddarfjärden	sediment	µg/kg DW	<25	<50	<10	<6	61	<2	<1	<5	<6	<6	<6	<6	<9
4485	Diffuse		Stora Essingen	sediment	µg/kg DW	47	<50	<10	<6	140	<5	3.6	<5	<6	<6	<6	<6	<9
4486	Diffuse		Årstaviken	sediment	µg/kg DW	33	<50	<10	<4	59	<5	5.0	<5	<4	<4	<4	<4	<6
4488	Diffuse		Riddarfjärden	perch	µg/kg FW	<10	<10	<10	<0.5	<20	<10	<0.1	<5	1.0	<0.5	<0.5	<0.5	<1
4489	Diffuse		Stora Essingen	perch	µg/kg FW	<10	<10	<10	<0.5	<20	<10	<0.1	<5	<0.5	<0.5	<0.5	<0.5	<1
4491	Diffuse		Årstaviken	perch	µg/kg FW	<10	<10	n. c.	<0.5	n. c.	<10	<0.1	n. c.	2.5	<0.5	0.4	<0.5	<1
4492	Diffuse		Årstaviken	roach	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	0.56	<5	n. c.5	n. c.	n. c.	n. c.	n. c.
4185	Diffuse	Bräcke	Bräcke STP	effluent	µg/l	0.27	<0.05	n. c.	n.c.	n. c.	<0.01	0.09	n.c.	n.c.	n.c.	n.c.	n.c.	n.c.
4165	Diffuse	Eskilstuna	Ekeby STP	effluent	µg/l	0.16	<0.05	N.c.	<0.01	n. c.	0.1	n.c.	N.c.	<0.01	<0.0 1	<0.01	<0.01	<0.01
4573	Diffuse	Borås	Gässlösa STP	effluent	µg/l	n. c.	n. c.	n. c.	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4585	Diffuse	Stockholm	Henriksdal STP	effluent	µg/l	n. c.	n. c.	n. c.	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4233	Diffuse	Gällivare	Kavaheden STP	effluent	µg/l	0.08	<0.05	N.c.	<0.01	n. c.	0.02	N.c.	n.c.	<0.01	<0.0 1	0.066	<0.01	<0.01
4207	Diffuse	Mölntorp	Mölntorp STP	effluent	µg/l	0.39	<0.05	N.c.	<0.01	n. c.	<0.01	N.c.	N.c.	<0.02	<0.0 1	0.017	<0.01	<0.01
4302	Diffuse	Hudiksvall	Reffelmans- verket	effluent	µg/l	n. c.	n. c.	n. c.	<0.01	n. c.	n. c.	n. c.	n. c.	<0.01	<0.0 1	0.011	<0.01	<0.01
4195	Diffuse	Ronneby	Rustorp STP	effluent	µg/l	n. c.	n. c.	<0.05	n. c.	n. c.	n. c.	<0.00 5	<0.05	n. c.	n. c.	n. c.	n. c.	n. c.
4271	Diffuse	Piteå	Sandholmen	effluent	µg/l	0.63	<0.05	n. c.	0.019	n. c.	<0.01	0.06	n.c.	<0.02	<0.0	0.570	0.1	<0.03

Sample ID	County/ National	City	Site	Matrix	Unit	2-Mercapto benzothia zole	2-(Tio- cyano- methyl- tio)- benso- tiazol	Brono- pol	4-Cl-3- Methyl phenol	N- Didecyldi methyl- ammo- nium- chloride	Propicon azole	Tri- closan	Reso- cinol	Methyl para- ben	Ethyl para ben	Propyl- paraben	Butyl- para- ben	Benzyl para- ben
			STP												2			
4215	Diffuse	Skövde	Stadskvarn STP	effluent	µg/l	n. c.	n. c.	<0.05	n. c.	n. c.	n. c.	<0.00 5	<0.05	n. c.	n. c.	n. c.	n. c.	n. c.
4288	Diffuse	Helsingborg	Öresunds- verket	effluent	µg/l	0.13	<0.05	N.c.	<0.01	n. c.	<0.01	N.c.	N.c.	0.05	<0.0 2	0.030	<0.01	<0.01
4709	Diffuse		Mälaren, Broviken	sediment	µg/kg DW	<20	<50	<5	<2	<50	<5	<0.5	<5	<2	<2	<2	<2	<3
4710	Diffuse		Mälaren, Broviken	sediment	µg/kg DW	<20	<50	<5	<2	<50	<5	<0.5	<5	<2	<2	<2	<2	<3
4404	Diffuse	Bollebygd	Bollebygd	sludge	µg/kg DW	660	<50	n. c.	<7	39 000	<5	3500	<50	32.0	<7	<7	<7	<11
4168	Diffuse	Eskilstuna	Ekeby STP	sludge	µg/kg DW	35	<50	N.c.	<5	n. c.	<5	N.c.	N.c.	40.0	<5	<5	<5	<8
4398	Diffuse	Eslöv	Ellinge	sludge	µg/kg DW	<20	<50	n. c.	59	5 600	<5	2000	<50	13	<7	<7	<7	<12
4403	Diffuse	Floda	Floda	sludge	µg/kg DW	950	<50	n. c.	<3	3 000	<5	3800	<50	18.0	<3	3.6	<3	<5
4574	Diffuse	Borås	Gässlösa STP	sludge	µg/kg DW	280	<50	<	67	61 000	<5	2300	160	59	<30	<23	<23	<36
4397	Diffuse	Stockholm	Henriksdal STP	sludge	µg/kg DW	<25	<50	<15	30	13 000	<2	3600	<10	15	<10	<4	<4	<6
4234	Diffuse	Gällivare	Kavaheden STP	sludge	µg/kg DW	<20	<50	n.c.	<5	n. c.	54	N.c.	N.c.	37.0	<5	<5	<5	<8
4572	Diffuse	Kiruna	Kiruna STP	sludge	µg/kg DW	330	<50	<17	28	4 200	<5	2800	380	<20	<6	<7	<6	<10
4208	Diffuse	Hallstaham mar	Mölntorp STP	sludge	µg/kg DW	<40	<50	n.c.	6.0	n. c.	<10	N.c.	N.c.	24	10.0	10.0	63.0	<8
4399	Diffuse	Alingsås	Nolhaga	sludge	µg/kg DW	22	<50	n. c.	14	21 000	<5	2800	<20	29	<5	<6	<5	<8
4303	Diffuse	Hudiksvall	Reffelmans- verket	sludge	µg/kg DW	22	<50	n.c.	<5	n. c.	<5	N.c.	N.c.	20.0	<5	<5	<5	<8
4196	Diffuse	Ronneby	Rustorp STP	sludge	µg/kg DW	n. c.	n. c.	n.c.	<5	570	n. c.	n. c.	n. c.	29.0	<5	<5	<5	<8
4402	Diffuse	Göteborg	Ryaverket	sludge	µg/kg DW	200	<50	n. c.	19	14 000	<5	3200	<25	52	<4	<4	<4	<7
4273	Diffuse	Piteå	Sandholmen STP	sludge	µg/kg DW	60	<50	n.c.	<5	5 500	<5	N.c.	N.c.	11.0	<5	<5	<5	<8
4217	Diffuse	Skövde	Stadskvarn STP	sludge	µg/kg DW	n. c.	n. c.	n.c.	10	15 000	n. c.	N.c.	N.c.	16	<5	<5	<5	<8
4414	Diffuse	Landskrona	Landskrona STP	sludge	µg/kg DW	260	<50	n. c.	<5	6 400	<5	3500	<30	13.0	<5	<5	<5	<8
4401	Diffuse	Umeå	Umeå	sludge	µg/kg DW	250	<50	n. c.	6.8	14 000	<0.01	4300	<10	44	<4	<4	<4	<6

Sample ID	County/ National	City	Site	Matrix	Unit	2-Mercapto benzothia zole	2-(Tio- cyano- methyl- tio)- benso- tiazol	Brono- pol	4-Cl-3- Methyl phenol	N- Didecyldi methyl- ammo- nium- chloride	Propicon azole	Tri- closan	Reso- cinol	Methyl para- ben	Ethyl para ben	Propyl- paraben	Butyl- para- ben	Benzyl para- ben
4481	Diffuse	Uppvidinge	Åseda STP	sludge	µg/kg DW	400	<50	n.c.	<5	280	<5	2900	<10	53.0	<5	<5	<5	<8
4290	Diffuse		Öresundsverket	sludge	µg/kg DW	28	57	n. c.	11	n. c.	18	n.c.	n.c.	30	<5	<5	<5	<8
4591	Point source		Wood impreg. plant Smp A	air	ng/m3	<1	<0.5	n. c.	n. c.	n. c.	0.39	<0.00 5	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4474	Point source		Wood impreg. plant Smp B	air	ng/m3	<1	<0.5	n. c.	n. c.	n. c.	0.26	<0.00 5	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4645	Point source		Wood impreg. plant Smp C	air	ng/m3	<1	<0.5	n. c.	n. c.	n. c.	0.25	<0.00 5	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4450	Point source		Wood impreg. plant	Stain bath	µg/l	n. c.	n. c.	n. c.	n. c.	n. c.	33000	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4451	Point source		Wood impreg. plant	Stain bath	µg/l	n. c.	n. c.	n. c.	n. c.	n. c.	33000	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4441	Point source		Wood impreg. plant	water	µg/l	<0.05	<0.05	n. c.	<0.01	n. c.	0.13	<0.00 5	n. c.	<0.01	<0.0 1	0.011	<0.01	<0.01
4445- 4446	Point source		Wood impreg. plant	surface water	µg/l	<0.05	<0.05	<0.05	<0.01	n. c.	<0.01	<0.00 5	<0.05	<0.01	<0.0 1	<0.01	<0.01	<0.01
4447- 4448	Point source		Wood impreg. plant	surface water	µg/l	<0,05	<0.05	n. c.	<0.01	n. c.	<0.01	2.0	<0.05	0.02	<0.0 1	0.017	<0.01	<0.01
4443	Point source		Wood impreg. plant	sediment	µg/kg DW	<20	<50	n. c.	<1	n. c.	20	<2	<10	<1	<1	<1	<1	<2
4780	Point source		Harbour	surface water	µg/l	<0.05	<0.05	n. c.	n. c.	n. c.	<0.1	<0.00 5	<0.01	n. c.	n. c.	n. c.	n. c.	n. c.
4779	Point source		Harbour	sediment	µg/kg DW	<20	<50	n. c.	n. c.	n. c.	<5	<2	<10	n. c.	n. c.	n. c.	n. c.	n. c.
4778	Point source		Harbour	biota, shellfish	µg/kg FW	n.c.	n.c.	<10	n. c.	n. c.	n.c.	<0.1	<5	n. c.	n. c.	n. c.	n. c.	n. c.
4453	Point source		Cosmetics plant	effluent	µg/l	<0.05	<0.05	n. c.	<2	n. c.	<0.01	n.c.	n.c.	<10	190	560	<10	<10
4381	Point source		Högbytorp	leachate before treatment	µg/l	<0.5	<0.05	n.c.	0.7	<1	<0.1	<0.02	n.c.	<0.07	<0.2	<0.1	<0.06	<0.01
4382	Point source		Högbytorp	leachate after treatment	µg/l	11	<0.05	n.c.	<0.03	<0.05	<0.1	<0.02	<0.4	<0.05	<0.1	<0.05	<0.05	<0.05
4609	Point source		Deponi	leachate	µg/l	0.11	<0.05	n.c.	<0.01	<0.04	<=0.3	<0.02	<0.02	<0.03	<0.0 5	<0.02	<0.01	<0.1
4097	human exp		food 1	meat	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4102	human exp		food 2	Dairy products	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4113	human exp		food 3	vegetables	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.

Sample ID	County/ National	City	Site	Matrix	Unit	2-Mercapto benzothia zole	2-(Tio- cyano- methyl- tio)- benso- tiazol	Brono- pol	4-Cl-3- Methyl phenol	N- Didecyldi methyl- ammo- nium- chloride	Propicon azole	Tri- closan	Reso- rcinol	Methyl para- ben	Ethyl para ben	Propyl- paraben	Butyl- para- ben	BenzyI para- ben
4588	human exp		food 4	cultivated salmon	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4589	human exp		food 5	wild salmon	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4590	human exp		food 6	wild salmon	µg/kg FW	n. c.	n. c.	<10	n. c.	<20	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4751	human exp		human urine 1	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4962	human exp		human urine 2	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4963	human exp		human urine 3	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4964	human exp		human urine 4	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4965	human exp		human urine 5	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.
4966	human exp		human urine 6	Urine	µg/l	n. c.	n. c.	<0.5	n. c.	<1	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.	n. c.