# Results from the Swedish National Screening Programme 2004

Subreport 1: Adipates

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Organization  IVL Swedish Environmental Research Institute Ltd.	Report Summary
	Project title
Address P.O. Box 21060 SE-100 31 Stockholm	Uppdrag inom den nationella miljöövervakningen, screening 2004
<b>Telephone</b> +46 (0)8-598 563 00	Project sponsor  Environmental Monitoring, Swedish Environmental Protection Agency

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#### Title and subtitle of the report

Results from the Swedish National Screening Programme 2004 Subreport 1: Adipates

## Summary

Measurements were performed of 7 adipates in 128 air, water, sludge, sediment and fish samples near source areas and at background locations. Human breast milk samples were also analysed. Di(2-ethylhexyl) adipate (DEHA) was found in sludge, sediment, fish and in one water sample. Di-iso-butyl and di-decyl adipate were detected in a few samples of sediment and sludge. Two potential point sources were identified; Stenungsund industrial area and Gislaved plastics/rubber production plant, which showed elevated levels in sediments, sludge and/or fish. Current diffuse emissions via municipal sewage treatment plants are likely, but the factors governing the occurrence of DEHA in sludge are unclear. Sediment and biota data indicate DEHA presence in the dissolved phase of surface water, however, it was not detected in any surface water samples. The absence in air confirms that adipates have no potential for long-range atmospheric transport. Although adipates are not very persistent, the high usage and observed occurrence in sediments and fish indicate that they may locally reach high levels. Considering that there are no restrictions of their usage, the environmental releases of adipates are not expected to cease in the near future.

#### Keyword

Adipates, screening, Sweden, environment

#### Bibliographic data

IVL Report B1645

#### The report can be ordered via

Homepage: www.ivl.se, e-mail: publicationservice@ivl.se, fax+46 (0)8-598 563 90, or via IVL, P.O. Box 21060, SE-100 31 Stockholm Sweden

# Sammanfattning

IVL Svenska Miljöinstitutet har på uppdrag av Naturvårdsverket genomfört en sk screening av adipater. Adipater är en grupp ämnen som ofta används som alternativ till ftalater och förekommer som bl a mjukgörare och smörjmedel i flera olika produkttyper. De adipater som ingick i studien var följande: dietyl-, di-iso-butyl-, dibutyl-, di-(2-etylhexyl)- (DEHA), dioktyl-, didecyl-, di-iso-oktyl-och di-iso-decyladipat. Det finns idag inga restriktioner gällande användningen av adipater i Sverige, och trots att ingen produktion förekommer inom landet så importeras stora kvantiteter, både som rena ämnen/kemiska produkter samt sannolikt även i from av färdiga konsumentprodukter och/eller halvfabrikat. Adipater är lättnedbrytbara både i miljön och i organismer. Vissa indikationer på toxicitet finns dock och det är möjligt att omfattande användning kan leda till koncentrationer som är tillräckligt höga för att utgöra en risk för levande organismer och för miljön. Det är därför av intresse att kartlägga förekomsten av adipater i den svenska miljön.

Huvudsyftet med studien var att bestämma koncentrationer i olika matriser i miljön, att belysa viktiga transportvägar samt att bedöma sannolikheten för pågående emissioner i Sverige. Ett ytterligare syfte var att bedöma omfattningen av atmosfärisk transport samt upptag i biota. Studiens resultat skall kunna bidra med underlag för beslut om vidare miljöövervakning av dessa ämnen.

En provtagningsstrategi utarbetades utifrån ämnenas fördelningsegenskaper samt möjliga källor till utsläpp. Potentiella punktkällor, diffusa källor (reningsverk), urban miljö samt bakgrundsstationer valdes ut, och provtagning utfördes i luft, vatten, slam, sediment och fisk. För att undersöka potentialen för human exponering, analyserades även 40 prover av bröstmjölk. Elva länsstyrelser deltog i studien och bidrog med ytterligare 66 prover för analys. Det totala antalet prover som analyserats inom studien framgår av tabellen nedan.

Program	Luft	Vatten	Sediment	Slam	Biota	Bröstmjölk	Totalt
Nationellt	12	14	17	4	12	40	99
Regionalt	-	17	10	37	2	-	66
Totalt	12	31	27	41	14	40	165

DEHA var den enda adipat som kunde detekteras frekvent i proven. Detta överensstämmer väl med data över konsumtion inom Sverige, där den registrerade konsumtionen av DEHA utgör 50-90% av den totala adipatanvändningen (den registrerade användningen har under de senaste åren skiftat något mot andra adipater). DEHA hittades i slam, sediment och fisk samt i ett enstaka vattenprov (obehandlat lakvatten från deponi).

Förutom DEHA kunde di-iso-butyladipat detekteras i två regionala slamprover. Di-decyladipat hittades i fyra slamprover och ett sedimentprov från det regionala programmet.

Inga adipater kunde detekteras i luft. Inga adipater kunde detekteras i bröstmjölk.

Resultaten från mätningarna i slam, sediment och fisk tyder på att industriområdet i Stenungsund samt plast/gummiindustrin i Gislaved utgör möjliga punktkällor för adipater, framför allt DEHA. Dessa indikationer utgjordes av:

- I Stenungsund hittades förhöjda halter av DEHA framför allt i sediment utanför industriområdet, men även i prover av tånglake av hankön. Koncentrationerna i yngel och honor av tånglake var dock i nivå med uppmätta koncentrationer i sill och strömming från bakgrundsområden.
- DEHA detekterades i slam från det industriella reningsverket vid plastindustrin i Gislaved samt i fisk från samma område i koncentrationer som var upp emot 3 gånger högre än uppmätta koncentrationer i bakgrundsfisk. DEHA detekterades däremot inte i sediment, men detektionsgränserna var högre än uppmätta koncentrationer i bakgrundsprover av sediment.

Resultaten från den regionala screeningen i slam samt de förhöjda halterna i urbana sediment tyder på att användning i hushåll och konsumentprodukter är en viktig källa för förekomsten av DEHA i den svenska miljön. Inget tydligt samband mellan koncentration av DEHA och befolkningstäthet (dvs storlek på reningsverk) kunde påvisas. Vilka faktorer som styr förekomsten av DEHA i slam är därmed okänt.

Den generella frånvaron av adipater i luft bekräftar att dessa ämnen snabbt avlägsnas från atmosfären via sorption till partiklar följt av deposition eller via atmosfärisk nedbrytning. Adipater har därmed ej potential för långväga atmosfärisk transport och lokala källor dominerar sannolikt utsläppen till miljön.

DEHA kunde inte detekteras i vatten förutom i ett prov. De uppmätta koncentrationerna i fisk från bakgrundsstationer tyder dock på att DEHA kan finnas i den lösta vattenfasen i halter i samma storleksordning som detektionsgränsen, varför det är möjligt att DEHA förekommer här, om än i koncentrationer som är lägre än detektionsgränsen. DEHA har även påvisats i vatten i tidigare studier. Dessutom ger den generella förekomsten i både fisk och sediment en tydlig indikation om att DEHA faktiskt emitteras till den akvatiska miljön.

Trots att adipater inte är särskilt persistenta, kan den omfattande användningen innebära att de lokalt kan uppnå höga koncentrationer i miljön. Då det inte finns några restriktioner gällande användningen, är det inte sannolikt att utsläppen kommer att minska under den närmaste tiden.

# **Summary**

As an assignment from the Swedish Environmental Protection Agency, IVL has performed a screening study of adipates. Adipates are group of chemicals commonly used as alternatives to phthalates and occur as plasticizers, lubricants etc. The adipates included in the study were diethyl, di-iso-butyl, dibutyl, di-(2-ethylhexyl) (DEHA), dioctyl, didecyl, di-iso-octyl and di-iso-decyl adipate. There is no restriction on the use of adipates in Sweden, and although there is no production within the country, they are imported in large quantities, both as pure chemicals and most probably also within consumer products. The adipates are fairly reactive substances, which readily degrade both in the environment and in organisms. However, there are indications of toxicity of these chemicals and it is possible that extensive use may result in concentrations high enough to pose a risk to living organisms and the environment. Thus, it is of interest to map the present occurrence of adipates in the Swedish Environment.

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

A sampling strategy was set up based on the partitioning properties and possible sources of these compounds. Potential point sources, diffuse sources (sewage treatment plants), urban sites as well as background sites were selected, and sampling was performed in air, water, sludge, sediment and fish. In order to investigate human exposure, 40 breast milk samples were also analysed. Eleven county administrative boards participated in the study and provided 66 additional samples for analysis. The total number of samples analysed in the study are shown in the table below.

Programme	Air	Water	Sediment	Sludge	Biota	Breast milk	Total
National	12	14	17	4	12	40	99
Regional	-	17	10	37	2	-	66
Total	12	31	27	41	14	40	165

DEHA was the only adipate frequently detected in the samples. This is consistent with information on usage in Sweden, where the registered DEHA consumption represents 50-90% of the total use (the registered consumption has shifted towards other adipates in the last few years). It was found in sludge, sediment and fish and in one single water sample (untreated landfill leachate).

Besides DEHA, di-iso-butyl adipate was detected in two regional sludge samples, and di-decyl adipate was found in four sludge samples and one sediment sample from the regional programme.

No adipates were detected in air. Adipates were not detected in human breast milk.

The results from sludge, sediment and fish, however, point out two potential point sources; the industrial area in Stenungsund and the plastics/rubber production plant in Gislaved. The indications for these two sites as point sources were:

- In Stenungsund, elevated concentrations of DEHA were mainly found in sediments outside the industrial area, but also in male eelpout samples. The concentrations in female and young eelpout were, however, comparable to background concentrations in herring.
- DEHA was detected in sludge from the industrial STP at the plastics/rubber production plant in Gislaved and in fish from the same area in levels up to 3 times higher than background concentrations. It was not detected in sediments but detection limits were higher than the concentrations found in background sediments.

Results from the regional screening in sludge as well as the observed elevated concentrations in urban sediment samples indicate that diffuse domestic usage in household products is an important source for the environmental occurrence of DEHA. No clear correlation with population density was observed, thus the factors governing the occurrence of DEHA in sludge remain unclear.

The general lack of adipates in air confirms that these substances are quickly removed from the atmosphere both via sorption to particles followed by deposition or via atmospheric degradation. Thus, adipates have no potential for long-range atmospheric transport and local sources are likely to dominate the release into the environment.

It is surprising that DEHA was not generally found in water. The measured concentrations in fish from background stations indicate that DEHA could be present in the dissolved water phase in concentrations comparable to the detection limit. It has also been found in water samples in other studies. In addition, the general occurrence in both fish and sediments gives a clear indication that DEHA does enter the aquatic environment.

Despite that adipates are not very persistent chemicals, their high usage and their occurrence in sediments and fish indicates that they locally may reach high levels. Considering that there are no restrictions of their usage, the environmental releases of adipates are not expected to cease in the near future.

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# 1 Introduction

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

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

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

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

<sup>&</sup>lt;sup>a)</sup> High Production Volume

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

b) Bioaccumulation/Persistence

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

 $<sup>^{\</sup>rm d)}\, {\sf Concerns}\,\, {\sf 3,3'-(ureylenedimethylene)-bis-(3,5,5-trimethylcyclohexyl)}\,\, {\sf diisocyanate}$ 

Due to the variety in emission sources and use as well as differences in chemical properties, the screening has been carried out in seven sub-projects. This report concerns the results for **adipates**. Results for the other chemicals are presented in subreports 2-6.

# 2 Physical-chemical properties, fate and toxicity

Adipates belong to a group of substances formed through esterification of adipic acid with various alcohols. The adipates included in this study are diethyl, di-iso-butyl-, dibutyl-, di-(2-etylhexyl)-(DEHA), dioctyl-, didecyl-, di-iso-octyl- and di-iso-decyl adipate (Table 2). Di-iso-octyl and di-iso-decyl adipate are isomeric mixtures, all others are single substances. All commercial adipates are brightly coloured, oily and odourless substances and have physical-chemical properties according to Table 3.

Table 2. Adipates included in the study

Name	CAS#	Structure
Diethyl adipate	141-28-6	
Di- <i>iso</i> -butyl adipate	141-04-8	
Dibutyl adipate	105-99-7	
Di(2-ethylhexyl) adipate (DEHA)	103-23-1	
Di-n-octyl adipate	123-79-5	
Didecyl adipate	105-97-5	
Di- <i>iso</i> -octyl adipate isomeric mixture The structure presented is one isomer only	1330-86-5	
Di-iso-decyl adipate isomeric mixture The structure presented is one isomer only	27178-16-1	

Table 3. Physical-chemical properties of adipates (HSDB 2004, Chemfinder 2004).

Ämne	MW	Wsol mg/L (°C)	<b>V</b> <sub>p</sub> (mm Hg)	H (atm m³/mol)	Log K <sub>ow</sub>	BCF L/kg	K <sub>oc</sub>
Diethyl adipate	202.25	4230 (20°)	0.058 (25°)	3. 6×10 <sup>-6</sup>		6	45
Di-iso-butyl adipate	258.36	Insoluble					
Dibutyl adipate	258.36						
Di-(2-ethylhexyl) –adipate (DEHA)	370.57	Insoluble 0.78 (22°)	8.5×10 <sup>-7</sup> (20°)	4.34×10 <sup>-7</sup>	>6.11	27	5.7×10
Di-n-octyl adipate	370.57	0.78 (22°)	8.5×10 <sup>-7</sup> (20°)			27	5×10 <sup>5</sup>
Didecyl adipate	426.68						
Di-iso-octyl adipate	370.57		2.6×10 <sup>-5</sup> (20°)	5.2×10 <sup>-5</sup>	8.1	60	
Di-iso-decyl adipate	426.68						

Explanations: Wsol is the solubility of phthalates in water.  $V_p$  is the vapour pressure. H (atm  $m^3$ /mol) is the Henry's law constant.  $K_{oc}$  is the octanol-water partition coefficient. BCF is the bioconcentration factor.  $K_{ow}$  is the organic carbon normalised solid-water partition coefficient in L/kg.

The vapour pressures (Table 3) show that DEHA, dioctyl adipate and diisooctyl adipate in the atmosphere may exist both in the gas- and particle phases. Diethyl adipate, which has the highest vapour pressure, is expected to exist mainly in the vapour phase. Adipates are removed from the air via dry and wet deposition or via degradation, which mainly takes place through reactions with hydroxyl radicals. Direct photolysis is also a possible degradation route, because of functional groups that absorb UV-light. Diethyl adipate is the most stable (Table 4) and is the only adipate that has potential to be transported long distances in the atmosphere (HSDB, 2004).

Except for the diethyl adipate, the adipates generally have high K<sub>OC</sub>-values (Table 3) and may sorb to organic carbon. When released to soil, DEHA, dioctyl adipate and diisooctyl adipate are considered to be immobile, whereas diethyl adipate is expected to have a high mobility. Some adipates may vaporise from soil, in particular diethyl adipate, which has a high vapour pressure and a high Henry's law constant. Biodegradation is expected to be an important route of removal from soil for most adipates (HSDB, 2004).

In the water environment DEHA, diisoctyl adipate and dioctyl adipate will to a large extent sorb to particles and end up in the sediment. The transport of these adipates via water is thus expected to be limited (HSDB, 2004). However, all adipates are esters which enable them to undergo hydrolysis, thus increasing their water solubility and enhancing transport in water (Table 4). Final hydrolysis products of DEHA are 2-ethylhexanol and hexanedioic acid and for dioctyl adipate, octanol och adipic acid (HSDB, 2004). Adipates are readily degraded by microorganisms (HSDB, 2004).

Table 4. Half-life  $(t_{1/2})$  of adipates when reacting with hydroxyl radicals in air and for hydrolysis in water (HSDB, 2004).

Adinata		t <sub>1/2/Water</sub>		
Adipate	t <sub>1/2/Air</sub>	pH 7	рН 8	
Diethyl adipate	2 days	1.7 years	64 days	
Di-(2-ethylhexyl) adipate	0.63 days	3 years	120 days	
Dioctyl adipate	0.67 days	5 years	170 days	
Di-iso-octyl adipate, (isomeric mixture)	16 h	75 days	2 years	

A modelling exercise was performed using the Equilibrium Criterion (EQC) model (Mackay et al., 1996) in order to highlight the likely fate and partitioning behaviour of adipates. Based on data on consumption and environmental occurrence in Sweden, DEHA was used as a model substance for the fate assessment. Physical-chemical properties were taken from Table 3. The degradation half-lives used were as follows; air: 17 h, water 170 h, soil 550 h and sediment 1700 hours. These half-lives were based on degradation (chemical and biological) data obtained from various sources (e.g. HSDB 2004; US EPA, 2005), as well as estimated data using the EPIWIN software (Meylan, 1999) and classified according to Mackay (2001). Emission rates were set to 1000 kg/h, only for illustrative purposes. The outcome of the modelling exercise is shown in Table 5.

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

Emission medium	Percentage in air	Percentage in water	Percentage in soil	Percentage in sediment	Persistence (h)
Air	6	2	83	9	224
Water	<0.001	17	< 0.01	83	811
Soil	<0.01	<0.01	100	< 0.01	794
All three	<1	8	54	38	609

The numbers in the table should be regarded as indicative, as they are dependent on model structure as well as chemical property data. However, the results indicate that the persistence of DEHA is generally low (<30 days when emitted to all media), and it is efficiently removed from the system through degradation. DEHA is predicted to partition mainly to soil and sediments. When emitted to air or water, a larger proportion is predicted to partition to these particular compartments, and DEHA may thus be available for advection out of the system.

This exercise emphasises the short residence time of DEHA in the environment and its low mobility when released to soils. As DEHA and other adipates are used in a large variety of products, emissions are likely to occur via different pathways; i.e. via air, water or even directly to soil.

There is only limited information about the toxicity of adipates available. Two adipates that have raised concern for environmental effects are DEHA and diethyl adipate. DEHA has structural similarities to other chemicals, which have proven carcinogenic properties and DEHA is therefore considered to be a potential carcinogen (HSDB, 2004). U.S. EPA has a limit for DEHA in drinking water of 0.4 mg/L and an oral reference dose (RfD) of 0.6 mg/kg/day (US EPA, 2004). Diethyl adipate is irritating at dermal exposure and has shown reproductive effects in laboratory experiments. Di-iso-octyl adipate and dioctyl adipate are considered slightly acute and chronically

toxic, but there is still limited data to support these findings. Dioctyl adipate is also irritating to skin and eye. Di-iso-butyl adipate is moderately toxic at intra-peritoneal (IP) exposure, mildly toxic when ingested with food and has experimentally shown teratogenic effects (HSDB, 2004). Ecotox data are according to Table 6 (US EPA, 2004).

Table 6. Ecotoxicity data (US EPA, 2004).

Adipate	Endpoint	Scientific name	Duration	Conc (μg/l)
Diethyl-	LC50	Pimephalis promelas	48 h	17300
Diethyl-	EC50 Behaviour	Pimephalis promelas	48 h	16900
Dibutyl-	LC50	Pimephalis promelas	96 h	3640
DEHA	LC50	Daphnia magna	48 h	660
DEHA	LC50	Pimephalis promelas	96 h	780
DEHA	LC50	Selenastrum capricornutum, (green algae)	96 h	780
DEHA	Reproduction	Daphnia magna (zooplankton)	21 d	24-52
DEHA	Growth	Daphnia magna	21 d	24-52

Bioconcentration factors (BCF) are generally low for adipates despite rather high log  $K_{\rm OW}$ -values (e.g. 8.1 for di-iso-octyl adipate; HSDB, 2004). The low bioconcentration is presumably due to degradation by organisms that absorb them. The strong sorption capacity of adipates (see above) also reduces their bioavailability. DEHA only has the potential to bioaccumulate in aquatic organisms, which lack the ability to degrade the molecule (HSDB, 2004).

# 3 Production, use, emissions and regulation

Adipates are not produced in Sweden, but they are imported and used in large amounts. (Figure 1). They are commonly used as alternatives to the well-studied phthalate esters as plasticizers. The adipates with the largest use are DEHA, di-iso-decyl adipate och di-iso-butyl adipate. DEHA is used as a plasticizer and in lubricants, glue, scotch-tape, and sealants. Di-iso-butyl adipate is mainly used in paint, laquers and fernish. Di-iso-octyl adipate and di-iso-decyl adipate are used in e.g. hydraulic liquids, lubricants and as chemical additives.

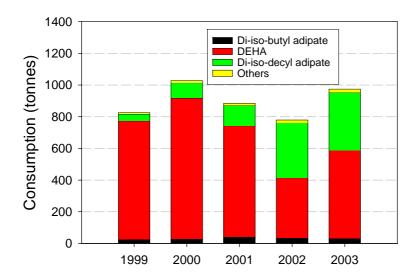


Figure 1. Use of adipates in Sweden (including export; SPIN, 2005).

The group "others" (Figure 1), does not include diethyl adipate nor didecyl adipate as their uses are confidential. The export of different chemical products containing adipates was limited in 2002; thus the majority of adipates registered in SPIN is used within Sweden (SPIN, 2005). Adipates are used in a large amount of products and application areas (Table 7).

Table 7. Number of chemical products containing adipates in Sweden (SPIN, 2005)

Tabl	e 7. Number	or chemical pro	Juucis containing a	uipates iii Sw	reden (SPIN, 20	03)	
Year	Dibutyl adipate	Di-iso-butyl adipate	Di(2-etylhexyl) adipate, DEHA	Dioctyl adipate	Di-iso-octyl adipate	Di-iso-decyl adipate	Total
1999	9 11	14	128	3	6	19	181
2000	9	13	115	4	6	20	167
2001	1 7	15	135	4	9	24	194
2002	2 7	16	141	9	10	26	209
2003	3 7	20	158	8	9	12	214

As evident from Figure 1 and Table 7, DEHA is the adipate most commonly used in Sweden, but di-iso-decyladipate is also becoming increasingly important. The equivalent of DEHA in the phthalate group is di-(ethylhexyl)-phthalate (DEHP), for which the consumption has decreased from 12000 tonnes (201 preparations) in 1999 to 1600 tonnes (149 preparations) in 2003 (SPIN,

2005). The consumption of phthalates is thus still larger than the adipate consumption, although the phthalate use is decreasing.

The large number of products and application fields indicate that apart from import of adipates as pure chemicals and chemical products, they may also to a large extent be imported in consumer products. The vast occurrence in consumer products may also result in diffuse emissions from products during use as well as in the "waste stage". The amount of adipates reaching the environment is therefore presumably larger than estimates based on data from the SPIN database.

# 4 Previous measurements in the environment

Adipates have mainly been measured in water. Swedish studies of adipates are rare (Loh et al., 2003), but Paxéus (1996) found DEHA in sewage water from three Swedish STPs. Table 8 shows a summary of previous measurements of adipates in different matrices in the environment.

Table 8. Measured concentrations of adipates in various matrices in the environment

Matrix	Country	centrations of adipates Site	Bis(2- metyl- propyl) adipate	DEHA	Mono(2- ethylhexyl-) adipate	Di-octyl adipate	Reference
Indoor air	USA	Office building		2.0 ng/m³			HSDB, 2004
	Japan	Tap water, Tobata- ku, Kitakyushu		77 μg/L			HSDB, 2004
	USA	Philadelpiha treatment plant		0.002 µg/L			HSDB, 2004
Drinking water	USA	New Orleans, treatment plant		0.1 μg/L			HSDB, 2004
	USA	Evansville, pilot plant				Detected, not quantified	HSDB, 2004
	USA	Miami				20 μg/L	HSDB, 2004
	USA	Delaware river, 1976-77		0.02-0.3 μg/L			HSDB, 2004
	USA	Monatiquot river, 1973		1-30 µg/L			HSDB, 2004
Surface water	USA	Lake Itasca, Minnesota, 1984		130 ng/L			HSDB, 2004
water	USA	Mississippi river, New Orleans,1984		35 ng/L			HSDB, 2004
	USA	14 industrialised river basins				Nd – 86 ng/L (n=204)	HSDB, 2004
C	Sweden	Henriksdal STP Stockholm	3 µg/L	12 μg/L	3 μg/L		Paxéus, 1996
Sewage water	Sweden	GRYYAB STP Göteborg	-	20 μg/L	2 μg/L		d.o.
	Sweden	Sjölunda STP Malmö	-	9 μg/L	3 μg/L		d.o.
Percolate water	Sweden	Hyllstofta Landfill		3.3 µg/L			HSDB, 2004
Sediment	USA	Boston				Detected, not quantified	

DEHA has also been detected in human tissues. It was found in 1 out of 8 samples from humans at the age of 1-14, 4 out of 8 samples from humans between 15 and 44 years and in 3 out of 8 samples from people older than 45 years (HSDB, 2004).

# 5 Sampling strategy and study sites

## 5.1 National

A national sampling strategy was developed in order to determine the concentrations of adipates in different environmental matrices in Sweden. An additional aim of the sampling was to identify major emission sources as well as important transport pathways. The sampling programme was therefore based on identified possible emission sources and use of the chemicals as well as on the behaviour of the substances in the environment. The programme included both measurements in background areas and close to potential point sources. Measurements of diffuse emission pathways from the society such as sewage systems and urban areas were also included. Human breast milk samples were analysed in order to investigate the potential for human uptake. The sampling programme is summarised in table 9.

Table 9. National sampling programme

Site	Air	Water	Sediment	Sludge	Fish	Breast milk	Total
Background							
Råö	3						3
Various			5		6	40	51
Point source							
Stenungsund (various industries)	3	3	3		3		12
Gislaved (plastic and rubber plant)	3	3	3	1	3		13
Diffuse sources							
STP Henriksdal		3		1			4
STP Eslöv		3		1			4
STP Floda		2		1			3
Stockholm	3		6				9
Landfill leachate from Högbytorp, Upplandsbro		2					2
Compost leachate, Borlänge		1					1
Total	12	17	18	4	12	40	102

Two industrial sites were selected as possible point sources for adipates; one with plastics and former rubber production (Gislaved) and one with various industries such as chemical and plastics production (Stenungsund). As diffuse sources, three wastewater treatment plants were selected, which are all included in the Swedish Environmental Protection Agency's monitoring programme for environmental pollutants in sludge (Naturvårdsverkets miljöövervakningsprogram av miljögifter i slam). In addition, three samples of percolate water were taken; two from a landfill in Upplandsbro (one untreated and one treated) and one from a compost in Borlänge. Diffusive pathways were also studied in an urban area in Stockholm, where air and sediment samples were collected.

In order to determine background levels; air, sediment and biota samples were collected. The locations of the sampling sites are shown in Figure 2. The sampling station Råö is also a part of the national monitoring programme for air pollutants and included in the EMEP network. The sediment samples were collected at off shore stations in the Baltic Sea (SGU, Ingemar Cato). The biota samples were collected at background sites used within the national monitoring programme of contaminants in biota (NRM, Anders Bignert).

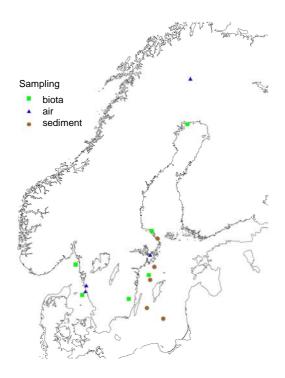


Figure 2. Geographical distribution of background and urban sampling stations

# 5.2 Regional

A regional screening programme was carried out by different Swedish county administrative boards that had the possibility to collect and send samples to IVL for analysis. Different counties have chosen different strategies for their regional sampling scheme.

The selection of the "regional" samples was meant to increase the number of samples for substances connected to their environmental programmes i.e. substances that have been regulated or included in the national priority database. Another strategy was to choose substances where environmental levels are expected to differ from national levels because of intensive use within the county i.e. local industrial areas or because of geographic proximity to European industrial areas. Furthermore, the aim of the regional sampling was to identify major emission sources as well as important transport pathways.

Eleven county administrative boards participated in the regional sampling programme with a total of 66 samples consisting of two fish samples, ten sediment samples, 37 sludge samples and 17 water samples.

# 6 Methods

# 6.1 Sampling

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

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

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

#### 6.1.1 Air

Air samples were collected by pumping air through two adsorbent tubes in series containing Tenax<sup>TM</sup>. The airflow was between 50 and 100 ml/min. Sampling proceeded for 24 hours. An additional adsorbing tube used as a field blank was sent back to the laboratory unexposed.

## 6.1.2 Water

Water samples were collected in pre-heated (400°C) glass bottles fitted with aluminium foil lined screw caps and stored in a refrigerator until analysed. 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. The samples were acidified with phosphoric acid (3 M) immediately after collection in order to avoid biodegradation of the adipates.

#### 6.1.3 Sediment

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

The four marine sampling sites (Ö Gotlandsdjupet, Ö Öland, Norrköpingsdjupet, Ö Landsortsdjupet) were chosen from areas with continuous deposition of fine-grained sediment. These sites were identified with hydroacoustic methods (shallow seismic, sub-bottom profiler and chirp side-scan sonar). Prior to sampling, the bottom at the sampling site was inspected with a submarine video camera. Furthermore, a sediment-core from the site was X-rayed with a sediment-scanner (Cato et al. 2000) in order to detect unwanted physical disturbances as strong bioturbation,

anchoring, trawling, etc. Sites, which fulfilled the sedimentological demands set up, were then sampled with a Gemini corer and the cores were sliced in vertical position with a core-cutter onboard. Surface sediments (0-1 cm) from four cores taken at each site were mixed in order to neutralise sediment inhomogenities. The samples collected were stored dark and frozen in precleaned and burned glass bottles.

# 6.1.4 Sludge

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

#### 6.1.5 Fish

The fish samples were collected by means of fishing net, a hoop net or a fishing rod. Samples of herring from background stations were supplied from The Environmental Specimen Bank at the Museum of Natural History. Fish samples were individually wrapped in aluminium foil and stored in a freezer (-20°C).

#### 6.1.6 Breast milk

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

The samples were acidified with phosphoric acid (1 M; 125 µl/ml) immediately after collection in order to avoid enzymatic hydrolysis of the adipates (David and Sandra 2001; Calafat et al., 2004). The chemicals added to the samples, phosphoric acid, heparin and EDTA, were checked for contamination prior to use by means of GC-MS. The samples were stored in a freezer (-80°C) and were transported to the IVL laboratory in Stockholm on dry ice. The obtained samples were marked with numbers and carried no personal information or medical history.

# 6.2 Analysis

#### 6.2.1 Chemicals

The solvents (HPLC-quality) acetone, hexane, metyl-tert-butylether (MTBE) were delivered from Rathburn Chemical Ltd. (Peeblesshire, Scotland). Solid phase columns containing modified silica sorbent (octadecyl and aminopropyl) were delivered from International Sorbent Technology Ltd. (UK). Sodium sulphate and silica gel was delivered from Merck and pre-heated (400°C) prior to use. All solvents, chemicals and equipment were checked by GC-MS before use. Ultra pure water was produced by a Milli-Q plus equipment (Millipore Corporation, Bedford, MA, USA). Surrogate standards used for quantification were di-n-propyl-, di-iso-propyl-, di-allyl- and di-n-nonyl adipate (delivered by TCI Europe, Belgium). The analytes diethyl-, diisobutyl- and di-n-butyl- were delivered by Aldrich. DEHA was obtained from Fluca and finally di-iso-octyl-, di-n-nonyl- and di-

n-octyl adipat from Chiron (Trondheim, Norway). The volumetric standard biphenyl was delivered from AccuStandard.

#### 6.2.2 Extraction

The Tenax<sup>TM</sup> column used to trap adipates in air was eluted with hexane:MTBE. Before GC-MS analysis, the eluate was concentrated to a final volume of 0.5 ml and spiked with a volumetric standard (biphenyl).

Sludge and sediment samples were centrifuged prior to extraction, in order to separate the water from the solid material. Surrogate standards and phosphoric acid were added and mixed carefully. The sample was extracted with (a) acetone, (b) acetone:MTBE and c) acetone:pentane:MTBE. The pooled extract was diluted with the pore water and ultra pure water to obtain 25 % acetone in water. The extract was mixed carefully. After phase separation the organic extract was safeguarded. The water phase was extracted once more with a mixture of pentane and MTBE. The combined extract was subjected to clean-up prior to GC-MS analysis (described below).

Each water sample (750 ml) was spiked with surrogate standards and extracted twice with pentane:MTBE (100 + 50 ml). The extracts were combined, dried over sodium sulphate and concentrated with a gentle stream of nitrogen. This extract was subjected to clean-up (described below).

Fish samples (muscle, 10 g) were homogenised in acetone and agitated for 30 min. The acetone was safeguarded and the samples were extracted twice with hexane:MTBE. The extracts where combined and the acetone was removed by shaking with water for 5 min. The extracts were dried, concentrated and the solvent changed to hexane prior to clean-up.

Breast milk samples (10 ml) were thawed and mixed with acetonitrile (20 ml). Sodium chloride (10 g) was added and the samples were agitated for 5 min. The acetonitrile was safeguarded after centrifugation. Ultra pure water (60 ml) was added to the acetonitrile extracts and the mixture was extracted twice with hexane:MTBE. Blanks (ultra pure water) were included for each batch of samples extracted, following the same analytical procedure as the samples. The extracts were subjected to clean-up prior to GC-MS analysis.

### 6.2.3 Clean-up of sample extracts

Large amounts of natural organic compounds are co-extracted with the analytes. Therefore, an effective clean-up method is needed to obtain a low detection limit. Clean-up was performed on three different columns: (a) silica gel, (b) aminopropyl and (c) aluminium oxide containing AgNO<sub>3</sub>. Elution was carried out using the same solvents for the three columns: fraction F-1 (hexane) and fraction F-2 (hexane: MTBE). The adipates were eluted in fraction F-2. The various types of samples analysed in this investigation (sediment, sludge, fish and breast milk) were cleaned up with different combinations of columns. The combinations used for the different matrices are presented below. All fractions used for analysis by GC-MS were spiked with a volumetric standard (biphenyl) prior to analysis.

Sludge and sediment extracts were first fractionated on a silica column. The F-2 fraction was further fractionated on an aminopropyl column. F-2 from the latter column was analysed using GC-MS.

Fish extracts were fractionated on an aluminium oxide/Ag column. Fraction F-2 was further fractionated on an aminopropyl column. F-2 from the latter column was analysed using GC-MS.

Breast milk extracts were cleaned up on an aminopropyl column. Fraction F-2, containing the adipates, was concentrated and analysed using GC-MS.

# 6.2.4 GC-MS analysis

The extracts were analysed on a 6890N gas chromatograph with a 5973N mass selective detector (Agilent). The injection was done in pulsed splitless mode at 275°C. The fused silica capillary column (VF-5MS 30 m  $\times$  0.25 mm i.d.  $\times$  0.25 µm film thickness, Varian) was held at 45°C for 1 min, ramped 15°C/min to 200°C and further 5°C/min to 300°C where it was held for 5 minutes. 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 ionisation at an energy of 70 eV. The analytes were identified by their characteristic retention times in combination with one quantification ion and one supporting ion, which was used to increase specificity.

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.

# 7 Results and discussion

## 7.1 National

All results of the samples analysed in the national programme are shown in Appendix , Table A 3 to Table A 6.

The only adipate detected in the national screening was DEHA. This is also the adipate with the highest consumption in Sweden (Figure 1). It was only found in sediment, sludge and fish and in one water sample. The detection limits for adipates in different matrices are shown in Table 10.

Table 10. Detection limits of adipates in different matrices.

Dw = dry weight, FW = fresh weight

	Air (ng/m³)	Water (µg/L)	Sludge (µg/kg DW)	Sediment (µg/kg DW)	Fish (ng/g FW)	Breast milk (µg/L)
Diethyl adipate	1	0.01	5	5	10	2
Di-iso-butyl adipate	2	0.05	10	10	10	1
Dibutyl adipate	1	0.05	10	10	10	1
(DEHA)	23	0.2	24	0.5, 24*	10	2
Dioctyl adipate	2	0.05	5	5	10	1
Di-decyl adipate	1	0.05	10	10	10	3
Di-iso-octyl adipate	20	1	50	50	100	20
Di-iso-decyl adipate	20	1	50	50	100	20

<sup>\*</sup>Due to the nature of the sample, the detection limit for DEHA in sediment at Gislaved was higher than in other samples.

## 7.1.1 Air and water

As mentioned above, adipates were not detected in any of the air samples. This general lack of adipates in air confirms that these substances are quickly removed from the atmosphere via sorption to particles and deposition, or via atmospheric degradation. Thus, adipates have limited potential for long-range atmospheric transport, and local sources are likely to dominate the release of adipates into the environment.

DEHA was detected in a concentration of 370  $\mu$ g/L in untreated landfill leachate from Högbytorp but it was not found in the treated leachate from the same site. Nor was it found in any other water sample. This is in disagreement with the findings by Paxéus (1996) where DEHA was detected in Swedish sewage effluents in concentrations of 9-20  $\mu$ g/L.

# 7.1.2 Sludge and sediment

Four sludge samples were included in the national screening programme, three from municipal sewage treatment plants and one from the potential point source in Gislaved. DEHA was only detected in the sludge sample from Gislaved, with a concentration of  $300 \,\mu\text{g/kg}$  dry weight (Table A 5).

DEHA was found in most of the sediment samples and the concentrations are shown in Figure 3.

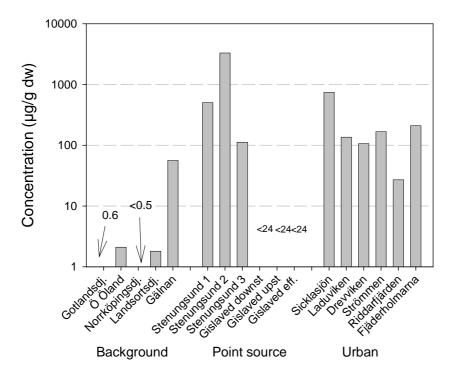


Figure 3. Concentrations of DEHA in sediments within the national sampling programme

There was a large variation in the concentrations in sediment from the different locations, with highest concentrations in sediments collected close to the industrial area in Stenungsund and lowest in sediments from offshore stations in the Baltic Sea. The background station Gälnan is a coastal site, which is closer to land than the other Baltic Sea stations. A marina with a petrol station is also situated in the area. These factors may possibly explain the higher concentrations found here.

DEHA was not detected in the sediments from the plastics/rubber industrial site in Gislaved. However, the detection limits in these samples ( $24 \,\mu g/kg$  dry weight) were higher than the detected levels at background stations. Increased concentrations of DEHA were found in Stenungsund compared to the background stations. The sediment samples in Stenungsund were taken in a gradient from the industrial area, but no decreasing concentrations with increasing distance from the potential source were found. Currents in the bay where the samples were taken may explain the higher concentration in sample number two compared to sample number one (Figure 3). Despite the lack of replicates and thus lack of possibility to use statistical methods, the industrial area in Stenungsund is suggested to be a possible point source for DEHA. DEHA also occurred in the

sediment samples collected in the urban area, Stockholm, indicating that diffuse sources may be important for the occurrence of DEHA in the sediments.

#### 7.1.3 Fish and human breast milk

DEHA was found in 8 out of 12 analysed fish samples. The results are illustrated in Figure 4.

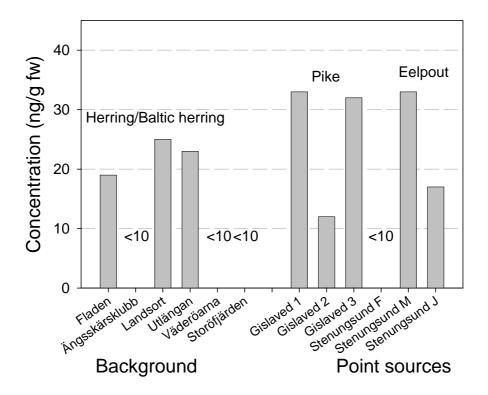


Figure 4. Measured concentration of DEHA in fish. F=females, M= males, J=juveniles

The average concentration of DEHA in herring and Baltic herring from background stations was 14 ng/g fresh weight (using half the detection limit when the substance was not detected) with the observed range <10 – 25 ng/g fresh weight. The concentrations of DEHA in fish from the point sources were generally higher compared to the average concentration in herring collected at background sites. The fish samples collected close to the plastics/rubber industrial site in Gislaved showed elevated concentrations relative to background sites in two out of three samples (32 and 33 ng/g fresh weight). Even though the species analysed here, pike, represents a higher trophic level than herring, the ready degradation of DEHA in organisms implies that concentrations in pike are not expected to be higher than in herring, possibly even lower due to trophic dilution. Thus, the higher concentrations observed in fish from the plastics/rubber industrial site may be a sign of local releases from the area.

The average background concentration in fish of 14 ng/g fresh weight corresponds to a concentration of 0.5  $\mu$ g/L (BCF = 27 – see Table 3) in the dissolved phase of the water (not considering any kind of uptake via the food chain). This is in the same order of magnitude as the detection limit for water. Thus, although DEHA was only detected in one of the water samples, the

biota data indicate that it may be present in the water, but in levels lower than the present limit of detection.

The general occurrence in fish from background sites also indicates that transport of DEHA may take place in water, alternatively that diffuse emissions of adipates occur generally all along the Swedish coastline, resulting in releases to the Baltic and the west coast.

The concentration of the adipates in the milk samples were all below the detection limits. The detection limits were for diethyl- and DEHA 2  $\mu g/l$ , for di-iso-butyl-, dibutyl- and di-n-octyl adipate 1  $\mu g/l$  and for didecyl adipate 3  $\mu g/l$ . The detection limit for the isomeric adipates (di-iso-octyl-, di-iso-decyl adipate) was 50  $\mu g/l$ .

# 7.2 Regional

The results for the regional programme are shown in Appendix (Table A 8). As in the national programme, DEHA was the adipate most frequently detected, and occurred only in sludge, sediments and fish. Other adipates were occasionally identified; they were di-iso-butyl and didecyl adipate.

# **7.2.1 Sludge**

In the regional sampling programme, adipates were frequently occurring in sludge, but there was a great variation in the concentrations in samples from the different STPs.

Di-iso-butyl adipate was found in Malmö and Finspång in concentrations of 11 and 13  $\mu$ g/kg dry weight, respectively. Didecyl adipate was detected in four samples from Krokom, Kristianstad, Landskrona and Vara in concentrations of 61, 190, 93 and 54  $\mu$ g/kg dry weight respectively.

DEHA was detected in all the analysed sludge samples but two (from Borlänge and Finspång). The DEHA concentrations from the regional sludge measurements are shown in Figure 5. The phtalate DEHP is a chemical spread in the society in the same manner as adipates and for comparison, concentrations of DEHP are also included in the figure where applicable. DEHP has been analysed in the same sample extract as the adipates.

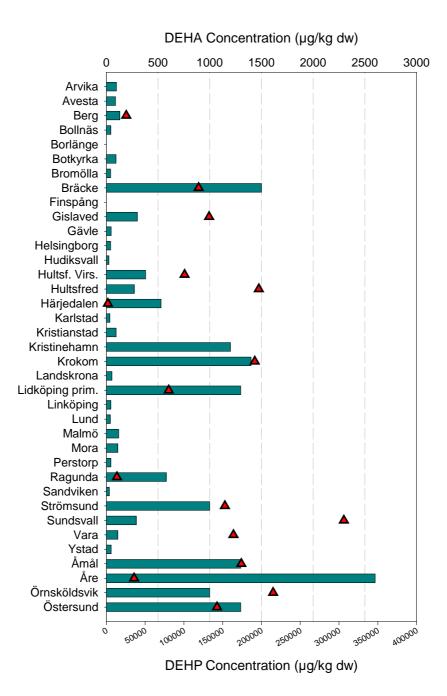


Figure 5. Concentration of DEHA (bars) and DEHP (triangles) in sludge from the regional sampling programme. Note the different scales on the x-axes.

As evident from Figure 5, there was a great variation in DEHA concentrations in sludge samples from the different regional sewage treatment plants (STPs). The concentrations varied between <10 and 2600 µg/kg dry weight. Only one sample was analysed per site and this was collected at one sampling occasion, thus differences in concentration should be interpreted with caution. The sources to the STPs with the highest concentrations of DEHA in the sludge (Åre, Östersund, Krokom, Bräcke, Åmål, Lidköping, and Kristinehamn) were mainly municipal. They all have

different treatment systems and are of varying size - from 1800 to 56 000 pe (person equivalents). The STPs with sludge with lower DEHA concentrations, have either not specified their sources, or are affected by a combination of municipal and industrial sources. Consequently, source type and treatment systems do not seem to influence the observed concentrations substantially. Figure 6 shows the correlation between the DEHA concentrations in sludge and the size of the STPs.

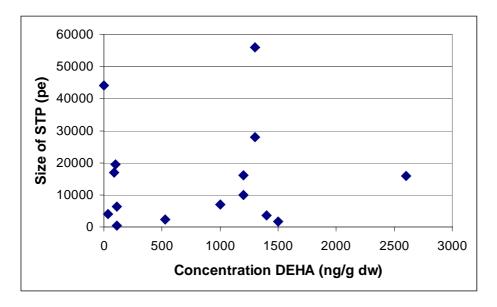


Figure 6. Correlation between DEHA concentration in sludge and STP size (pe).

It is evident from the figure that there is no clear relationship between the size and the DEHA concentrations. Although a positive correlation between concentration and size (i.e. population density) might be expected, Figure 6 indicates that the higher concentrations are randomly distributed between STPs of different size.

## 7.2.2 Water, sediment and fish

No adipates were detected in any of the regional water samples, while DEHA were detected in one sediment sample from St Envättern, (11 ng/g dry weight). The concentration of DEHA was lower than in the urban sediment samples in the national programme but higher than in the background samples from the Baltic Sea.

Two fish samples were analysed within the regional programme, showing DEHA concentrations similar or slightly higher than those measured at Swedish background sites (17 and 15 ng/g fresh weight) in perch from Himmerfjärden and St Envättern respectively). No other adipates were detected in the regional fish samples.

# 8 Conclusions

DEHA was the only adipate frequently detected in the Swedish environment. This is consistent with information on usage in Sweden, where the registered DEHA consumption represents 50-90% of the total adipate use (the registered consumption has shifted towards other adipates in the last few years). DEHA was detected in sludge, sediment and fish and in one single water sample (untreated landfill leachate).

Two possible point sources have been identified; the industrial area in Stenungsund, and the rubber/plastics production site in Gislaved. In Stenungsund, elevated concentrations were measured in sediments outside the industrial area and also in male eelpout samples. The concentrations in female and young eelpout were, however, comparable to background concentrations in herring.

DEHA was detected in sludge from the STP, which treats wastewater from the rubber/plastics industrial site in Gislaved and in fish from the same area, in concentrations that were generally higher than the background concentrations in fish. The results of the analyses in sludge as well as the observed elevated concentrations in urban sediment samples indicate that diffuse domestic usage in household products is an important source for the environmental occurrence of DEHA.

None of the adipates were found in air, neither close to sources nor at background sites. Atmospheric transport thus seems to be of minor importance for the occurrence of adipates in the Swedish environment. Adipates were not generally found in water. However, the general occurrence of DEHA in both fish and sediments, indicates that DEHA does enter the aquatic environment. No adipates were detected in breast milk.

Although adipates are not very persistent chemicals, their high usage and their occurrence in sediments and fish indicates that they locally may reach high levels. Considering that there are no restrictions of their usage, the environmental releases of adipates are not expected to cease in the near future.

# 9 Acknowledgements

Thanks to all staff at the county administrative boards and different municipalities that have contributed to the sampling. Especially thanks to Anders Bignert, NRM and personnel at point source industry for contributing with samples for the national programme. We also want to thank Staffan Skerfving and Inger Bensryd at the University Hospital of Lund (Department of Occupational and Environmental Medicine) for providing the milk samples. The study was funded by Environmental Monitoring at the Swedish Environmental Protection Agency.

# 10 References

- Andersson, J. 2004. Teoretisk förstudie till screening av miljögifter i Skåne 2004. M Sc. Thesis. Department of Ecology, University of Lund. *In Swedish*
- Hanberg, A., Högberg, J., Berglund, M., Bensryd, I., Skerfving, S., Remberger, M., Calafat, A., Appelgren, M., Falk Filipsson, A., Bo Jansson, Håkansson, H.2005. Phthalates and their metabolites in human breast milk, blood and urine as measures for monitoring exposure in human risk groups. (In prep).
- Calafat, M. A., Slakman, R. A., Silva, J. M., Herbert, R. A., Needham, L. K. 2004. Automated solid phase extraction and quantitative analysis of human milk for 13 phthalate metabolites. J. Chromatogr., B 805: 49-56.
- Cato, I., Rindby, A., Rudolfsson, J., 2000: Unik sedimentscanner utvecklad. Geologiskt forum 25, 13-15. *In Swedish*
- Chemfinder, 2004 http://chemfinder.cambridgesoft.com/chemicals/. CambridgeSoft Corporation.
- David, F., Sandra, P.2001. Phthalate esters in the environment: Monitoring program for the determination of phthalates in air, vegetation, cattle feed, milk and fish in The Netherlands (1999-2001). Korttijk, Belgium, Research Institute of Chromatography, RIC. 45.
- HSDB.2004. Hazardous Substance Data Base. U.S. National Library of Medicine Available at: <a href="http://www.toxnet.nlm.nih.gov/cgi-bin/sis/htmlgen?HSDB">http://www.toxnet.nlm.nih.gov/cgi-bin/sis/htmlgen?HSDB</a>, Aug 2005
- Loh C., Johansson V., Ivarsson, P. 2003. Utredning av ämnen inför screening 2004. EnviroPlanning Rapport 1003-01/10/01/rap001. *In Swedish*
- Mackay, D. 2001. Multimedia Environmental Models: The Fugacity Approach, CRC Boca Raton, FL, USA.
- Mackay, D., Di Guardo A., Paterson S., Cowan C.E. 1996. Evaluating the environmental fate of a variety of types of chemicals using the EQC model. Environ. Toxicol. Chem., 15, 1627-1637
- Meylan W. 1999. EPIWIN x3.04 [computer program: US EPA Version for Windows], Syracuse Research Corporation, Syracuse, NY, US. http://www.epa.gov/oppt/exposure/docs/episuitedl.htm (June 13, 2005)
- OSPAR, 2005. OSPAR List of Chemicals for Priority Action Replaced OSPAR Annex 2 from June 2002, www.ospar.org, March, 2005.
- Paxéus, N. 1996. Organic Pollutants in the effluents of large wastewater treatment plants in Sweden. Wat. Res., 30:1115-1122.
- SPIN, 2005. Substances for Preparation in Nordic Countries. Chemical Substance Database. Available at: <a href="http://www.spin2000.net/spin.html">http://www.spin2000.net/spin.html</a> Aug, 2005.

UNEP. 2005. Stockholm Convention on Persistent Organic Pollutants (POPs) http://www.pops.int/

US EPA, 2004. Ecotox, 2004 http://www.epa.gov/ecotox/

US EPA, 2005. Technical Factsheet on: DI (2-ETHYLHEXYL) ADIPATE. Available at: <a href="http://www.epa.gov/OGWDW/dwh/t-soc/di-adipa.html">http://www.epa.gov/OGWDW/dwh/t-soc/di-adipa.html</a>, Aug, 2005

Appendix Information on samples and lists of results

 Table A 1. List of samples for adipate analysis, national sampling programme.

 Mech.= Mechanical, Bio.=biological, Chem.= chemical, Dehydr.= dehydration. DW = dry weight (sediment, sludge), LW = lipid weight (fish)

Background	Sample ID MR-3995	Site	Matrix	Notes	Sampling date	X(RT90)	Y(RT90)	(%)
	MR-3995	¢0		:				
		אמט	Air	Sampling time 24 hours	2004-11-16			-
	MR-4058	Råö	Air	Sampling time 24 hours	2004-10-30			,
	MR-4059	Råö	Air	Sampling time 24 hours	2005-02-07			1
-	MR-3750	Ö Gotlandsdjupet, SGU 04-0058	Sediment	121m depth, 0-2 cm	Autumn 2004	6216805	1697299	9.5
	MR-3751	Ö Öland, SGU 04-0368	Sediment	77m depth, 0-2 cm	Autumn 2004	6277308	1607374	8.3
	MR-3752	Norrköpingsdjupet, SGU 07i-001	Sediment	179m depth, 0-2 cm	Autumn 2004	6435399	1625504	9.9
<u>                                     </u>	MR-3753	Ö Landsortsdjupet, SGU 09i-0841	Sediment	403m depth, 0-2cm	Autumn 2004	9668059	1648002	3.8
<u>.                                      </u>	MR-3559	Stockholm, Gälnan,	Sediment	24 m depth, 0-2 cm	2004-10-16	6664669	1666854	18.8
<u>                                     </u>	MR-3639	V. Fladen	Fish	Herring, females, 10	2003-09-16			4.5
<u> </u>	MR-3640	Ängsskärsklubb	Fish	Baltic herring, females, 10	2003-10-01			4.6
	MR-3641	Landsort	Fish	Baltic herring, females, 10	2003-11-12			4.4
	MR-3642	Utlängan	Fish	Herring, females, 10	2003-10-10			1.7
	MR-3643	Väderöarna	Fish	Herring, females, 10	2003-09-04			6.1
	MR-3644	Storöfjärden, Kalix skärgård	Fish	Baltic herring, females, 10	2003-09-29			2.3
Diffuse sources	MR-3762	Henriksdal STP, Sickla	Water	Influent, 700 000 pe, mech, chem,bio cleaning. No major industrial impact	2004-11-16			1
	MR-3764	Henriksdal STP	Water	Effluent, see above	2004-11-16			-
<u>                                     </u>	MR-3760	Henriksdal STP	Sludge	Digested, see above	2004-11-16			26.7
	MR-3848	Eslöv STP	Sludge	Digested, ca 100 000 pe, mech., chem., bio., treatment. Food industry (75%)	2004-12-07			18.4
	MR-3850	Eslöv STP	Water	Influent, municipal, see above	2004-12-10			
	MR-3851	Eslöv STP	Water	Influent, industrial, see above	2004-12-10			•
	MR-3852	Eslöv STP	Water	Effluent, see above	2004-12-10			-
	MR-3703	Floda STP	Water	Influent	2004-11-08			-
	MR-3704	Floda STP	Water	Effluent	2004-11-08			-
	MR-3701	Floda STP	Sludge	Digested	2004-11-08			27.7
	MR-3928	Upplandsbro landfill, Högbytorp	Water	Untreated leachate	2005-02-16			-
	MR-3926	Upplandsbro landfill, Högbytorp	Water	Treated leachate	2005-02-16			1
	MR-3758	Borlänge landfill	Water	Compost leachate	2004-11-16			1

Table A 1. Cont	1							
Category	Sample ID	Site	Matrix	Notes	Sampling date	X(RT90)	Y(RT90)	DW/LW (%)
Urban	MR-4000	Stockholm, Hudiksvallsgatan 2	Air	Sampling time 24 hours	2004-11-25			,
	MR-4002	Stockholm, Hudiksvallsgatan 2	Air	Sampling time 24 hours	2004-11-30			
	MR-4056	Stockholm, Hudiksvallsgatan 2	Air	Sampling time 24 hours	2005-05-29			-
	MR-2535	Stockholm Sicklasjön	Sediment	0-2 cm, mixed samples, 4.5 m depth	Spring 2002	98821299	1632207	4.8
	MR-2539	Stockholm Laduviken	Sediment	0-2 cm, mixed samples, 3 m depth	Spring 2002	6584432	1629075	3.2
	MR-2541	Stockholm Drevviken	Sediment	0-2 cm, mixed samples, 12 m depth	Spring 2002	620299	1635190	5.8
	MR-2530	Stockholm, Strömmen	Sediment	0-2 cm, mixed samples, 27 m depth	Spring 2002	6581049	1634496	5.5
	MR-2547	Stockholm, Riddarfjärden	Sediment	0-2 cm, mixed samples, 19 m depth	Spring 2002	6580116	1627280	27.8
	MR-2549	Stockholm, Fjäderholmarna	Sediment	0-2 cm, mixed samples, 28 m depth	Spring 2002	6580073	1629454	6.4
Various	MR-3996	Stenungsund	Air	Sampling time 24 hours	2004-11-09			
chemical and plastics	MR-3997	Stenungsund	Air	Sampling time 24 hours	2004-12-08			1
industries	MR-3998	Stenungsund	Air	Sampling time 24 hours	2004-11-08			1
	MR-3878	Stenungsund, A1	Water		2004-11-30	6446247	1264535	1
	MR-3872	Stenungsund, A1	Sediment		2004-11-30	6446247	1264535	27.2
	MR-3874	Stenungsund, D7	Sediment		2004-11-30	6446760	1264051	31.1
	MR-3876	Stenungsund, E1	Sediment		2004-11-30	6446077	1253472	27.2
	MR-4053	Stenungsund	Fish	Eelpout, 5 females	2004-11-30			0.8
	MR-4054	Stenungsund	Fish	Eelpout, 5 males	2004-11-30			9.0
	MR-4055	Stenungsund	Fish	Eelpout, 5 juveniles	2004-11-30			0.7

Category	Sample ID	Site	Matrix	Notes	Sampling date	X(RT90) Y(RT90)	Y(RT90)	(%) DW/LW
Plastics-	MR-3989	Gislaved	Air	Sampling time 24 hours	2004-11-23	23		-
rubber	MR-3991	Gislaved	Air	Sampling time 24 hours	2004-11-17	17		
production	MR-3993	Gislaved	Air	Sampling time 24 hours	2004-11-18	81		
	MR-3766	Nissan Gislaved	Water	Downstream Storm water effluent	2004-11-17	17 6352791	1 1363001	
	MR-3768	Nissan Gislaved	Water	Upstream Storm water effluent	2004-11-16	16 6356021	1364021	
	MR-3770	Gislaved	Water	Storm water effluent	2004-11-16	16 6353937	7 1364076	٠
	MR-3776	Nissan Gislaved	Sediment	Downstream storm water effluent	2004-11-17	17 6352791	1363001	86.1
	MR-3772	Nissan Gislaved	Sediment	Upstream Storm water effluent	2004-11-16	16 6356021	1364021	80.3
	MR-3774	Gislaved	Sediment	At storm water effluent	2004-11-16	16 6353937	7 1364076	76.8
	MR-3733	Gislaved STP	Sludge		2004-11-17	17		16.9
	MR-3892	Nissan Skeppshult, Gislaved	Fish	Pike, 870g/51cm	2004-04-07	7(		
	MR-3893	Nissan Skeppshult, Gislaved	Fish	Pike, 2350g/70cm	2004-04-07	7(		
	MR-3894	Nissan Rydöbruk Gislaved	Fish	Pike, 1400g/59cm	2004-04-07	70		

Table A 2. Samples for regional screening of adipates.

Mun. = Municipal, Ind. = Industrial, Mech. = Mechanical, Bio. = biological, Chem. = chemical, Dehydr. = dehydration DW = dry weight (sediment, sludge), LW = lipid weight (fish)

County	Sample ID	Municipality	Site	Matrix	Notes	Site	Pe (STP)	Treatmen	Sampling date	X (RT90)	Y (RT90)	DW/LW
Dalarna	MR-3498	Avesta	Lake Bäsingen	Sediment					2004-10-04		,	21
	MR-3478	Avesta	Krylbo STP	Sludge	Digested		17000	Suspension, Active sludge	2004-09-29			24
	MR-3540	Borlänge	Fagersta STP	Sludge	Digested		44000	Suspension, Active sludge	2004-10-12			29
	MR-3535	Mora	Venjan STP	Sludge	Digested		500	Suspension, Active sludge	2004-10-06			0.7
	MR-3496	Mora	Lake Venjan	Sediment					2004-10-03			5.2
Gävleborg	MR-3584	Bollnäs	Bollnäs STP	Sludge					2004-10-20			15
	MR-3549	Gävle	Duvbacken STP	Sludge					2004-10-12			20
	MR-3846	Hudiksvall	Resselvans STP	Sludge					2004-12-07			27
	MR-3555	Sandviken	Sandviken STP	Sludge					2004-10-13			22
Jämtland	MR-3687	Berg	Myrviken sludge lagoon	Sludge	Primary	Mun.		Mech., Dehydr., compost	2004-11-01			19
	MR-3619	Bräcke	Bräcke STP	Sludge	Primary	Mun., Ind., workshops	1800	Mech. Bio.	2004-10-19			11
	MR-3470	Härjedalen	Björnrike STP	Sludge	Primary, Sample taken during season	Mun.	2325	Mech., Chem.	2004-06-21			77
	MR-3574	Krokom	Hissmofors STP	Sludge	Primary	Mun., Ind., workshops	3600		2004-10-19			13
	MR-3586	Ragunda	Överammaer sludge lagoon	Sludge	Primary	Mun.,		Dehydr.	2004-10-19			29
	MR-3625	Strömsund	Strömsund STP	Sludge	Primary	Mun., Ind., workshops	7000	Mech., chem., bio.	2004-10-27			18
	MR-3469	Åre	Åre STP	Sludge	Primary, Sample taken during season	Mun.	16000	Mech., chem., bio.	2004-04-13			18
	MR-3472	Östersund	Göviken STP	Sludge	Digested	Mun., Ind. (dairy)	55900	Mech., chem., bio.	2004-09-29			27
Jönköping	MR-3731	Gislaved	Gislaveds STP	Water	Influent				2004-11-15			-
	MR-3735	Gislaved	Gislaveds STP	Water	Effluent				2004-11-15			,
	MR-3733	Gislaved	Gislaved STP	Sludge					2004-11-15			17

Table A 2. Cont	1:					i				;		
County	Sample ID	Municipality	Site	Matrix	Notes	Site information	Pe (STP)	Treatmen	Sampling date	X (RT90)	Y (RT90)	DW/LW (%)
Kalmar	MR-3800	Hultsfred	Hulingen	Sediment					2004-11-24			34
	MR-3809	Hultsfred	Hultsfred STP	Water	Effluent				2004-11-24			-
	MR-3810	Hultsfred	Hultsfred STP	Water	Influent				2004-11-24			-
	MR-3799	Hultsfred	Hultsfred STP	Sludge					2004-11-24			19
	MR-3804	Hultsfred	Virserum STP	Water	Influent				2004-11-24			
	MR-3805	Hultsfred	Virserum STP	Water	Effluent				2004-11-24			
	MR-3797	Hultsfred	Virserum STP	Sludge					2004-11-24			15
	MR-3798	Hultsfred	Virserum	Sediment	Recipient				2004-11-24			5.5
	MR-3627	Oskarshamn	Mouth of river Emån	Sediment					2004-10-27			23
Skåne	MR-3466	Bromölla	Bromölla STP	Sludge					2004-09-28			26
	MR-3459, MR-3460	Helsingborg	Helsingborgs STP	Sludge					2004-09-22			25
	MR-3514	Kristianstad	Kristianstad STP	Sludge					2004-10-05			18
	MR-3489	Landskrona	Lundåkraverket STP	Sludge					2004-09-29			22
	MR-3464	Lund	Lund STP	Sludge	Thermophilic Digested + centrifuged				2004-09-27			26
	MR-3456, MR-3457	Malmö	Malmö STP	Sludge					2004-09-22			24
	MR-3623	Perstorp	Perstorp STP	Sludge					2004-10-26			19
	MR-3453, MR-3454	Ystad	Ystad STP	Sludge					2004-09-22			16
Stockholm	MR-3480	Botkyrka	Himmerfjärdsverket STP	Water	Effluent				2004-09-30			,
	MR- 3482,84,86	Botkyrka	Himmerfjärdsverket STP	Sludge	Average of three days sampling				2004-09-28 –30			23
	MR-3449	Botkyrka	Himmerfjärden bay	Sediment					2004-09-01	654978	161021	24
	MR-3450	Botkyrka	Himmerfjärden bay	Fish	Perch				2004-09-01	654978	161021	0.7
	MR-3448	Södertälje	Lake St Envättern	Sediment					2004-09-01	655613	158793	3.8
	MR-3451	Södertälje	Lake St Envättern	Fish	Perch				2004-07-22	655613	158793	1.0
		Ī							•			1

Table A 2. Cont.												Ī
County	Sample ID	Municipality	Site	Matrix	Notes	Site information	Pe (STP)	Treatmen	Sampling date	Х (RT90)	Y I (RT90)	DW/LW (%)
Värmland	MR-3711	Arvika	Vik STP	Water	Effluent	Mun., Ind.	19500	Mech. Chem. bio.	2004-11-02			,
	MR-3709	Arvika	Vik STP	Sludge	Digested	Mun., Ind.	19500	Mech., chem., bio.	2004-11-02			15
	MR-3580	Karlstad	Skåre STP	Water	Effluent	Mun., Ind.	4005	Mech., chem., bio.	2004-10-19			
	MR-3582	Karlstad	Skåre STP	Sludge	Primary	Mun., Ind.,	4005	Mech., chem., bio.	2004-10-19			30
	MR-3631	Kristinehamn	Fiskartorpet STP	Water	Effluent	Mun.	16109	Mech., chem., bio.	2004-10-29			
	MR-3629	Kristinehamn	Fiskartorpet STP	Sludge	Digested, Average of three days sampling	Mun.	16109	Mech., chem., bio.	2004-10-26/28			1.5
	MR-3719	Säffle	Östby landfill	Water	Leachate	Mainly mun.			2004-11-08			-
	MR-3721	Säffle	Östby landfill	Sediment		Mainly mun.			2004-11-08			26
Väster- norrland	MR-3754	Sundsvall	Tivoliverket	Sludge					2004-11-16			19
	MR-3726	Örnsköldsvik	Bodum STP	Sludge					2004-11-10			31
Västra Götaland	MR-3508	Lidköping	Lidköping STP	Water	Effluent	Mun.	28000	Mech., chem.	2004-10-05			
	MR-3510	Lidköping	Lidköping STP	Sludge	Primary	Mun.	28000	Mech., chem.	2004-10-05			20
	MR-3512	Vara	Vara STP	Sludge	Digested	Mun., laundry	0989	Mech., chem., bio.	2004-10-04			19
	MR-3474	Åmål	Âmâl STP	Water	Effluent	Mun., workshops.	10000	Mech., chem.	2004-09-29			1
	MR-3476	Åmål	Åmål STP	Sludge	Digested	Mun., workshops.	10000	Mech., chem.	2004-09-29			20
Östergötland	MR-3635	Finspång	Finspångs STP	Water					2004-10-27			-
	MR-3633	Finspång	Finspångs STP	Sludge					2004-10-26			4.1
	MR-3683	Finspång	Skuten	Sediment					2004-10-29			7.7
	MR-3728	Linköping	Linköping STP	Water					2004-11-09			-
	MR-3729	Linköping	Linköping STP	Sludge					2004-11-09			28
	MR-3685	Linköping	Roxen	Sediment					2004-10-21			13
	MR-3788	Norrköping	Häradsudden landfill	Water	Leachate				2004-11-22			1
	MR-3789	Norrköping	Häradsudden landfill	Water	Leachate				2004-11-22			,

Table A 3. Concentration of adipates in air, national programme

Sample ID	Site	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso-octyl adipate	Di-iso-decyl adipate
MR-3995	Råö	ng/m³	1>	<2	<b>1</b> >	<23	<2	\ 	<50	<50
MR-4058	Råö	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-4059	Råö	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-3996	Stenungsund	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-3997	Stenungsund ng/m <sup>3</sup>	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-3998		ng/m³	^	< 2	<u>^</u>	<23	<2	<u>^</u>	<50	<50
MR-3989	Gislaved	ng/m³	^	< 2	^	<23	<2	<u>~</u>	<50	<50
MR-3991	Gislaved	ng/m³	^	< 2	^	<23	<2	<u>~</u>	<50	<50
MR-3993	Gislaved	ng/m³	^	< 2	<u>^</u>	<23	<2	<u>^</u>	<50	<50
MR-4000	Stockholm	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-4002	Stockholm	ng/m³	^	< 2	^	<23	<2	^	<50	<50
MR-4056	Stockholm	ng/m³	<u>^</u>	<2	^	<23	<2	^	<50	<50

Table A 4. Concentration of adipates in water, national programme

Sample I D	Matrix	Site	Unit	Dietyl adipate	Di-iso- butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso- octyladipate	Di-iso-decyl adipate
MR-3878	Surface water	Stenungsund, A1	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<b>!</b> >	
MR-3766	MR-3766 Surface water	Nissan, downstream Gislaved	hg/L	<0.01	<0.05	<0.05	< 0.2	< 0.05	<0.05	^	^
MR-3768	MR-3768 Surface water	Nissan, upstream Gislaved	hg/L	<0.01	<0.05	<0.05	< 0.2	< 0.05	<0.05	^	^ _
MR-3770	Storm water	Gislaved	hg/L	<0.01	<0.05	<0.05	< 0.2	<0.05	<0.05	^	^
MR-3762 Influent	Influent	Henriksdal STP, Sickla	hg/L	<0.01	<0.05	<0.05	< 0.2	<0.05	<0.05	^	\ 
MR-3764 Effluent	Effluent	Henriksdal STP	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	^	\ 
MR-3850	Influent, municipal	Eslöv STP	hg/L	<0.01	<0.05	<0.05	<0.2	< 0.05	<0.05	^	^
MR-3851	Influent, industrial	Eslöv STP	hg/L	<0.01	<0.05	<0.05	<0.2	< 0.05	<0.05	^	^
MR-3852	Effluent	Eslöv STP	hg/L	<0.01	<0.05	<0.05	< 0.2	<0.05	<0.05	^	\ 
MR-3703 Influent	Influent	Floda STP	hg/L	<0.01	<0.05	<0.05	< 0.2	<0.05	<0.05	^	^
MR-3704 Effluent	Effluent	Floda STP	hg/L	<0.01	<0.05	<0.05	< 0.2	<0.05	<0.05	^	^
MR-3928	Untreated leachate	Upplandsbro, Högbytorp landfill	hg/L	<0.01	<0.05	<0.05	370	< 0.05	<0.05	^	^
MR-3926	Treated leachate	MR-3926 Treated leachate Upplandsbro, Högbytorp landfill	hg/L	<0.01	<0.05	<0.05	< 0.2	< 0.05	<0.05	^	^
MR-3758	MR-3758 Leachate	Borlänge, Högbytorp, compost	hg/L	<0.01	<0.05	<0.05	<0.2	< 0.05	<0.05	\ \ -	\ 

Table A 5. C	Table A 5. Concentration of adipates in sediment and	sediment		, national <sub>I</sub>	sludge, national programme						
Sample ID	Site	Matrix	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso- octyladipate	Di-iso-decyl adipate
MR-3750	Ö Gotlandsdjupet,	Sediment µg/	hg/kg DW	× 2	<10	< 10	9.0	< 5	<10	<50	<50
MR-3751	Ö Öland	Sediment	Sediment µg/kg DW	V 2	<10	<10	2.1	< 2	<10	<50	< 50
MR-3752	Norrköpingsdjupet	Sediment	µg/kg DW	V 2	<10	<10	<0.5	V 2	<10	<50	< 50
MR-3753	Ö Landsortsdjupet	Sediment	µg∕kg DW	× 5	<10	<10	1.8	< 2	<10	<50	< 50
MR-3559	Stockholm, Gälnan, 0-2 cm	Sediment	µg/kg DW	V 2	<10	<10	26	× 2	<10	<50	< 50
MR-3872	Stenungsund, A1	Sediment	µg∕kg DW	V 2	<10	<10	200	< 2	<10	<50	< 50
MR-3874	Stenungsund, D7	Sediment	µg∕kg DW	V 2	<10	<10	3300	< 2	<10	<50	< 50
MR-3876	Stenungsund, E1	Sediment	µg/kg DW	V 2	<10	<10	110	V V	<10	<50	< 50
MR-3776	Gislaved, downstream	Sediment	µg∕kg DW	V 2	<10	<10	<24	< 2	<10	<50	< 50
MR-3772	Gislaved, upstream	Sediment µg/kg	µg∕kg DW	V 2	<10	<10	<24	V V	<10	<50	< 50
MR-3774	Gislaved	Sediment	µg∕kg DW	V 2	<10	<10	<24	V V	<10	<50	< 50
MR-2535	Stockholm, Sicklasjön	Sediment	µg∕kg DW	V 2	<10	<10	740	< 2	<10	<50	< 50
MR-2539	Stockholm, Laduviken	Sediment µg/kg	hg/kg DW	V 2	<10	< 10	140	× 2	<10	<50	< 50
MR-2541	Stockholm, Drevviken	Sediment	hg/kg DW	V 21	<10	< 10	110	× 2	< 10	<50	< 50
MR-2530	Stockholm, Strömmen	Sediment µg/	µg∕kg DW	V 21	<10	< 10	170	× 2	< 10	<50	< 50
MR-2547	Stockholm, Riddarfjärden	Sediment	hg/kg DW	V 2	<10	< 10	27	× 2	<10	<50	< 50
MR-2549	Stockholm, Fjäderholmarna	Sediment	µg∕kg DW	V 2	<10	<10	210	< 2	<10	<50	< 50
MR-3760	Henriksdal STP	Sludge	µg∕kg DW	V 2	<10	<10	100	< 2	<10	<50	< 50
MR-3848	Eslöv STP	Sludge	µg/kg DW	× 5	<10	<10	110	× 2	<10	<50	< 50
MR-3701	Floda STP	Sludge	µg/kg DW	V 2	<10	<10	40	V V	<10	<50	< 50
MR-3733	Gislaved STP	Sludge	µg/kg DW	< 5	<10	< 10	310	× 2	<10	<50	< 50

Table A 6. Concentration of adipates in fish, national programme

Sample I D	Site	Species	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso-octyl adipate	Di-iso-decyl adipate
MR-3639 Fladen	Fladen	Herring	ng/g FW	<10	<10	<10	19	<10	<10	<100	<100
MR-3640	MR-3640 Ångsskärsklubb	Baltic herring	ng/g FW	<10	^ 10	<10	<10	<10	<10	<100	< 100
MR-3641	MR-3641 Landsort	Baltic herring	ng/g FW	<10	^ 10	<10	25	<10	<10	<100	<100
MR-3642	MR-3642 Utlängan	Herring	ng/g FW	<10	<10	<10	23	<10	<10	< 100	<100
MR-3643	MR-3643 Väderöarna	Herring	ng/g FW	<10	<10	<10	<10	<10	<10	<100	< 100
MR-3644	MR-3644 Storöfjärden	Baltic herring	ng/g FW	<10	^ 10	<10	<10	<10	<10	<100	< 100
MR-4053	MR-4053 Stenungsund	Eelpout	ng/g FW	<10	<10	<10	<10	<10	<10	<100	< 100
MR-4054	MR-4054 Stenungsund	Eelpout	ng/g FW	<10	<10	<10	33	<10	<10	<100	<100
MR-4055	MR-4055 Stenungsund	Eelpout	ng/g FW	<10	<10	<10	17	<10	<10	<100	<100
MR-3892	MR-3892 Gislaved	Pike	ng/g FW	<10	<10	<10	33	<10	<10	<100	<100
MR-3893	Gislaved	Pike	ng/g FW	<10	<10	<10	12	<10	<10	<100	<100
MR-3894 Gislaved	Gislaved	Pike	ng/g FW	<10	<10	<10	32	<10	<10	<100	<100

Di-iso-decyl adipate < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 < 20 Di-iso-octyl adipate <20 <20 <20 < 20 <20 <20 < 20 <20 <20 <20 <20 <20 <20 <20 <20 <20 <20 <20 < 20 Didecyl adipate 8 Dioctyl adipate Table A 7. Concentration of adipates in breast milk, national programme Dibutyl adipate Di-iso-butyl adipate adipate Dietyl hg/L Lund Site Lund MR-3007 MR-3008 MR-3009 MR-3010 MR-3011 MR-3012 MR-3013 MR-3014 MR-3015 MR-3016 MR-3017 MR-3018 MR-3019 MR-3020 MR-3021 MR-3022 MR-3023 MR-3024 MR-3025 MR-3027 MR-3028 MR-3029 MR-3030 MR-3031 MR-3032 MR-3181 MR-3280 Sample

Sample I D	Site	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso-octyl adipate	Di-iso-decyl adipate
MR-3282	Lund	hg/L	< 2	^	^	<2	\ \ -	× 3	<20	<20
MR-3283	Lund	hg/L	< 2	^	^	<2	\ \ \	× 3	<20	<20
MR-3284	Lund	hg/L	< 2	^	^	<2	^	× 3	<20	<20
MR-3285	Lund	hg/L	< 2	^	^ _	<2	\ \ \	× 3	<20	<20
MR-3286	Lund	hg/L	< 2	^	^	<2	\ \ \	× 3	<20	<20
MR-3287	Lund	hg/L	< 2	^	^	<2	\ \ \	× 3	<20	<20
MR-3288	Lund	hg/L	<2	<u>^</u>	^	<2	\ \ \	< 3	<20	<20
MR-3289	Lund	hg/L	< 2	^	^	<2	\ \ -	× 3	<20	<20
MR-3290	Lund	hg/L	< 2	^	^	<2	\ \ -	× 3	<20	<20
MR-3291	Lund	hg/L	< 2	^	^	<2	\ \ -	× 3	<20	<20
MR-3292	Lund	hg/L	< 2	^	^	<2	\ \ \	× 3	<20	<20
MR-3293 Lund	Lund	hg/L	<2	> 1	> 1	<2	> 1	<3	<20	<20

County	Sample ID	Municipality	Site	Matrix	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	Di-iso- octyl adipate	Di-iso- decyl adipate
Dalarna	MR-3498	Avesta	Bäsingen	Sediment	µg/kg DW	× 2	<10	<10	< 10	× 5	< 10	<50	<50
	MR-3478	Avesta	Krylbo STP	Sludge	µg/kg DW	× 2	<10	<10	86	< 5	<10	<50	<50
	MR-3540	Borlänge	Fagersta STP	Sludge	µg/kg DW	× 2	<10	<10	<10	<5	<10	<50	<50
	MR-3535	Mora	Venjan STP	Sludge	µg/kg DW	× 2	<10	<10	110	< 5	<10	<50	<50
	MR-3496	Mora	Lake Venjan	Sediment	µg/kg DW	× 2	<10	<10	<10	<5	< 10	<50	<50
Gävleborg	MR-3584	Bollnäs	Bollnäs STP	Sludge	µg/kg DW	< 5	<10	<10	43	<5	<10	<50	<50
	MR-3549	Gävle	Duvbacken STP	Sludge	µg/kg DW	× 2	<10	<10	47	× 2	<10	<50	<50
	MR-3846	Hudiksvall	Resselvans STP	Sludge	µg/kg DW	× 2	<10	<10	25	× 2	<10	<50	< 50
	MR-3555	Sandviken	Sandviken STP	Sludge	µg/kg DW	V 21	<10	<10	28	× 2	<10	<50	<50
Jämtland	MR-3687	Berg	Berg STP	Sludge	µg/kg DW	< 5	<10	<10	130	<5	<10	<50	<50
	MR-3619	Bräcke	Bräcke STP	Sludge	µg/kg DW	× 2	<10	<10	1500	< 5	<10	<50	<50
	MR-3470	Härjedalen	Härjedalen STP	Sludge	µg/kg DW	< 5	<10	<10	530	< 5	<10	<50	<50
	MR-3574	Krokom	Krokom STP	Sludge	µg/kg DW	× 2	<10	<10	1400	< 5	19	<50	< 50
	MR-3586	Ragunda	Ragunda STP	Sludge	µg/kg DW	< 2	<10	<10	280	< 2	<10	<50	< 50
	MR-3625	Strömsund	Strömsund STP	Sludge	µg/kg DW	× 2	<10	<10	1000	< 2	<10	<50	<50
	MR-3469	Åre	Åre STP	Sludge	µg/kg DW	< 2	<10	<10	2600	<5	<10	<50	<50
	MR-3472	Östersund	Östersund STP	Sludge	µg/kg DW	× 2	<10	<10	1300	< 5	<10	<50	<50
Jönköping	MR-3731	Gislaved	Gislaveds STP, influent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<1	<1
	MR-3735	Gislaved	Gislaveds STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
Kalmar	MR-3804	Hultsfred	Virserum STP, influent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3805	Hultsfred	Virserum STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>\</u>
	MR-3797	Hultsfred	Virserum STP	Sludge	µg/kg DW	× 2	<10	<10	380	< 2	<10	<50	<50
	MR-3798	Hultsfred	Virserum, recipient	Sediment	µg/kg DW	× 2	<10	<10	<10	< 2	<10	<50	< 50
	MR-3809	Hultsfred	Hultsfred STP, effluent	Water	µg/L	<0.01	<0.05	<0.05	<0.2	< 0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3810	Hultsfred	Hultsfred STP, influent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3799	Hultsfred	Hultsfred STP	Sludge	µg/kg DW	× 2	<10	<10	270	< 2	<10	<50	< 50
	MR-3800	Hultsfred	Hulingen	Sediment	µg/kg DW	< 5	<10	<10	<10	< 5	<10	<50	< 50
	MR-3627	Oskarshamn	Mouth of river Eman	Sediment	MQ by	\ Г.	<10	<10	7	ν.	7	Ļ	,

I able A 6. Coll.	COLIF.											Di-iso-	Di-iso-
County	Sample ID	Municipality	Site	Matrix	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DEHA	Dioctyl adipate	Didecyl adipate	octyl adipate	ø
Skåne	MR-3466	Bromölla	Bromölla STP	Sludge	µg/kg DW	× 2	<10	<10	42	× 2	<10	<50	<50
	MR-3459, MR- 3460	Helsingborg	Helsingborg STP	Sludge	µg/kg DW	V 21	<10	<10	42	٧ 5	<10	<50	0 20 20
	MR-3514	Kristianstad	Kristianstad STP	Sludge	hg/kg DW	× 21	<10	<10	98	\ \ \ \	190	<50	<50
	MR-3489	Landskrona	Lundåkraverket STP	Sludge	µg/kg DW	V 21	<10	<10	22	× 2	93	<50	<50
	MR-3464	Lund	Lund STP	Sludge	µg/kg DW	v 2	<10	<10	40	× 2	<10	<50	<50
	MR-3456, MR- 3457	Malmö	Malmö STP	Sludge	µg/kg DW	V 2	11	< 10	120	\ \ \	< 10	<50	<50
	MR-3623	Perstorp	Perstorp STP	Sludge	µg/kg DW	V 21	<10	<10	45	× 2	<10	<50	<50
	MR-3453, MR- 3454	Ystad	Ystad STP	Sludge	µg/kg DW	V 2	< 10	< 10	47	V 21	< 10	<50	<50
Stock- holm	MR-3480	Botkyrka	Himmerfjärdsverket STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	^	\ -
	MR-3482,84,86	Botkyrka	Himmerfjärdsverket STP	Sludge	µg/kg DW	<b>~</b>	< 5	<10	<10	94	<5	<50	<50
	MR-3449	Botkyrka	Himmerfjärden bay	Sediment	µg/kg DW	V 2	<10	<10	<10	< 5	<10	<50	<50
	MR-3450	Botkyrka	Himmerfjärden bay	Fish	ng/g FW	<10	<10	<10	17	< 10	<10	<100	<100
	MR-3448	Södertälje	Lake St Envättern	Sediment	µg/kg DW	× 2	<10	<10	11	× 2	,	<50	<50
	MR-3451	Södertälje	Lake St Envättern	Fish	ng/g FW	< 10	<10	<10	15	<10	<10	<100	<100
Värmland	MR-3711	Arvika	Arvika STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	^	\ 
	MR-3709	Arvika	Arvika STP	Sludge	µg/kg DW	× 2	<10	<10	26	< 5	<10	<50	<50
	MR-3580	Karlstad	Skåre STP effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	< 0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3582	Karlstad	Skåre STP	Sludge	µg/kg DW	× 2	<10	<10	35	< 2	<10	<50	<50
	MR-3631	Kristinehamn	Kristinehamn STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3629	Kristinehamn	Kristinehamn STP	Sludge	µg/kg DW	× 2	<10	<10	1200	<5	<10	<50	<50
	MR-3719	Säffle	Östby landfill, leachate	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3721	Säffle	Östby landfill	Sediment	µg/kg DW	<5	<10	<10	<10	<5	<10	<50	<50
Väster- norrland	MR-3754	Sundsvall	Tivoliverket	Sludge	µg/kg DW	<5	<10	<10	300	< 5	<10	<50	<50
	MR-3726	Örnsköldsvik	Bodum STP	Sludge	µg/kg DW	<5	<10	<10	1000	<5	<10	<50	<50
Västra Götaland	MR-3508	Lidköping	Lidköping STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	\ 	<u>^</u>
	MR-3510	Lidköping	Lidköping STP, primary	Sludge	µg/kg DW	× 2	<10	<10	1300	< 5	<10	<50	<50
	MR-3512	Vara	Vara STP	Sludge	µg/kg DW	× 2	<10	<10	110	< 5	54	<50	<50
	MR-3474	Åmål	Åmål STP, effluent	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	<u>^</u>
	MR-3476	Åmål	Åmål STP	Sludge	µg/kg DW	< 5	<10	<10	1200	< 5	<10	<50	<50

Table A 8. Cont.	Cont.												
County	Sample ID	Municipality	Site	Matrix	Unit	Dietyl adipate	Di-iso-butyl adipate	Dibutyl adipate	DЕНА	Dioctyl   adipate	Didecyl adipate	Di-iso- octyl adipate	Di-iso- decyl adipate
Öster- götland	MR-3635	Finspång	Finspångs STP	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	\ \ \	\ \ \
)	MR-3633	Finspång	Finspångs STP	Sludge	µg/kg DW	× 2	13	<10	<10	× 2	<10	<50	<50
	MR-3683	Finspång	Skuten	Sediment	µg/kg DW	< 5	<10	<10	<10	<5	<10	<50	<50
	MR-3728	Linköping	Linköping STP	Water	µg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	^
	MR-3729	Linköping	Linköping STP	Sludge	µg/kg DW		<10	<10	44	< 2	<10	<50	<50
	MR-3685	Linköping	Roxen	Sediment	µg/kg DW	< <sub>2</sub>	<10	<10	<10	<5	<10	<50	<50
	MR-3788	Norrköping	Häradsudden landfill	Water	hg/L	<0.01	<0.05	<0.05	<0.2	<0.05	<0.05	<u>^</u>	^