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REPORT

Screening of complexing agents: EDTA, DTPA,
NTA, 1,3-PDTA and ADA

11 October 2012

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REPORT

Screening of complexing agents: EDTA, DTPA, NTA, 1,3-PDTA and ADA

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Summary

The occurrence of complexing agents in the Swedish environment has been studied in a screening investigation. The studied complexing agents all belong to the group of amino-polycarboxylates:

- EDTA
- NTA
- DTPA
- 1,3-PDTA
- ADA.

These chemicals are highly polar and certain are also environmentally persistent. This investigation is part of the national environmental monitoring programme, run by the Swedish EPA, but also includes the participation of 13 county administrative boards. The investigation includes a number of subprogrammes that addresses certain key questions:

- Whether these chemicals are present in lakes and watersheds
- To what degree these chemicals are present in domestic incoming and outgoing waste waters
- If releases from municipal waste water treatment plants influence the occurrence in their recipients
- If diffuse emissions occur in urban areas
- If the chemicals are released from certain industrial point sources

In total 149 samples were analysed. The sampling medias were, listed in decreasing number of samples: effluent waste water, surface water, incoming waste water, stormwater, groundwater, drinking water, sediment, sludge and a landfill leachate. This report aims at giving a general description of the results and provides a discussion in relation to the questions shown above.

EDTA and NTA were detected in most samples of incoming and outgoing municipal waste waters from municipal waste water treatment plants (wwtp's). DTPA was relatively commonly detected in those media, whereas 1,3-PDTA and ADA were detected only in a few of these samples. The concentrations of EDTA and NTA in incoming waste waters varied significantly among the different wwtp's, with median levels of 47 µg/l (EDTA) and 93 µg/l (NTA). Concentrations of EDTA were generally similar in the incoming waste waters and effluents, indicating that EDTA is not ready biodegradable. In contrast, NTA levels in waste waters were on the average reduced ca five times in the wwtp's, suggesting that biodegradation occurs.

In order to assess whether outgoing municipal waste waters influence their recipients with regard to these chemicals, surface waters from recipients of 16 wwtp's were analysed for these complexing agents. Concentrations of EDTA and NTA were generally higher in the recipients than in local background sites. In these recipients, EDTA ranged from 1-100 µg/l and NTA from 0,1-10 µg/l. The remaining three chemicals studied were generally not detected in these wwtp recipients.

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Very high EDTA levels, up to almost 10 mg/l, were detected in the effluents to a pulp and paper mill. Also the recipient to this point source contained high EDTA levels that progressively decreased as a result of dilution further downstream. Single spot samples of surface water were also taken in several other recipients to pulp and paper mills, but there was generally no pronounced influence of these industries. DTPA was, however, detected at elevated levels downstream two paper mills.

EDTA and NTA were also detected in treated and untreated drinking water, in urban groundwater and in urban stormwaters.

Roughly equal amounts of EDTA and DTPA are used in Sweden. The large difference regarding their environmental occurrence may be attributed to the much more disparate use of EDTA as compared to that of DTPA. The occurrence of complexing agents in wwtp's and in surface waters is in agreement with the Swedish Exposure Indexes, that were calculated by the Swedish Chemicals Inspectorate and are intended to reflect the risk for diffuse emissions. The common occurrence of EDTA and NTA are matched by their high indexes; DTPA and 1,3-PDTA were detected less commonly, in agreement with their low indexes.

The detected concentrations of complexing agents are not apparently toxic to aquatic ecosystems or health. Indirect effects may nevertheless be caused by their strong ability to influence the bioavailability of both essential and toxic metals in the aquatic environments. When also considering that EDTA is environmentally persistent, the general occurrence of EDTA and NTA in surface waters may still cause some concern.

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Sammanfattning

Förekomsten i miljön av komplexbildare inom gruppen aminopolykarboxylater har undersökts genom en screeningundersökning. De studerade föröreningarna är:

- EDTA
- NTA
- DTPA
- 1,3-PDTA
- ADA.

Samtliga av dessa ämnen är starkt polära och vissa är också svårnedbrytbara i miljön. Uppdraget ingår i Naturvårdsverkets miljöövervakning men innehållar också deltagande av 13 länsstyrelser. Studien innehåller ett antal delprogram som alla är upprättade efter några huvudsakliga frågeställningar:

- Om dessa ämnen förekommer i sjöar och vattendrag
- I vilken mån ämnena sprids till och från kommunala reningsverk
- Om utsläpp från kommunala reningsverk påverkar halterna i recipienterna
- Om diffus spridning sker i urban miljö
- Om ämnena sprids till miljön från industriella punktkällor.

Totalt omfattade undersökningen 149 prov, fördelat på följande matriser i avtagande omfattning: utgående avloppsvatten, ytvatten, inkommende avloppsvatten, dagvatten, grundvatten och dricksvatten, sediment, slam och lakvatten. Denna rapport syftar till att ge en allmän beskrivning av resultaten samt att presentera övergripande tolkningar.

EDTA och NTA förekom allmänt i både inkommende och utgående avloppsvatten från kommunala reningsverk. DTPA detekterades i relativt många avloppsprov medan 1,3-PDTA och ADA endast påträffades i några få prov. Halterna av EDTA och NTA i inkommende avloppsvatten varierade påtagligt mellan reningsverken, med medianhalter på 47 µg/l (EDTA) och 93 µg/l (NTA). I utgående avloppsvatten var halterna av EDTA i stort sett opåverkade medan halterna av NTA var i medeltal ca fem gånger lägre än i inkommende vatten. Detta illustrerar att EDTA är svårnedbrytbart, medan biologisk nedbrytning av NTA sker i reningsverk.

För att studera om utgående avloppsvatten påverkar förekomsten av dessa ämnen i recipienterna provtogs ytvatten från recipienterna till sexton reningsverk. Som grupp betraktat var halterna av EDTA och NTA betydligt högre i dessa recipienter än i lokala bakgrundslokaler. För EDTA är uppmätta halter mellan ca 1-100 µg/l, och för NTA ca 0,1-10 µg/l. Övriga studerade ämnen kunde generellt inte detekteras i reningsverkens recipienter.

Förekomsten av dessa ämnen undersöktes också i utgående industriellt avloppsvatten samt i recipienter till ett pappersbruk. Mycket höga halter av EDTA uppmättes i detta industri-

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ella avloppsvatten. I den å som utgjorde recipient var halterna som mest ca 900 µg/l, för att successivt avta pga utspädning längre ned i sjösystemet. Enstaka stickprov togs också på ytvatten nedanför andra industriella punktkällor, men någon tydlig påverkan på förekomsten av komplexbildare kunde sällan påvisas. Utanför två pappersbruk påvisades dock DTPA i förhöjda halter.

EDTA och NTA påträffades också i drickvatten, i urbana grundvatten och i urbana dagvatten. Halterna var i huvudsak i storleksordningen 1 µg/l.

Mängderna av EDTA och DTPA som används i Sverige är ungefär lika stora. Att deras förekomst i miljön skiljer sig så mycket beror troligen på att DTPA i stort sett bara används inom papperindustrin, medan EDTA har en bredare användning. Förekomsten i reningsverk och i vattenmiljön stämmer relativt väl överens med det exponeringsindex som Kemikalieinspektionen presenterat, och som bygger på förutsedd risk för spridning. EDTA och NTA har höga index och förekom också i höga halter och påträffades i många prov. DTPA och 1,3-PDTA har relativt låga index och påträffades mer sporadiskt, DTPA var dock mer vanligt än 1,3-PDTA.

De uppmätta ytvattenhalterna av dessa komplexbildare förefaller inte vara direkt toxiska för ekosystem eller hälsa. Dessa ämnen kan dock ge en indirekt påverkan genom sin förmåga av komplexbilda både essentiella och toxiska metaller. Betydelsen av denna effekt har inte kunnat bedömas i föreliggande rapport. I kombination med det faktum att EDTA och NTA förekommer i nästan alla ytvattenprov som analyserats och att EDTA är svårnedbrytbar, kan det ändå finnas anledning till att uppmärksamma EDTA och NTA.

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1. Introduction

1.1. General

In environmental science and monitoring, the last ca 10 years have brought about a focus on relatively polar pollutants. Kolpin et al. (2002) demonstrated widespread pollution of rivers located downstream Urban areas in the US. Pollutants with various intended use and of various origin were found in those rivers, including e.g. pharmaceuticals, stimulants, detergents, biocides, pesticides, plasticizers, flame retardants and fragrances. Similar findings have been drawn from numerous studies since then.

The Swedish Screening program, run by the Environmental Protection Agency, has also included a large number of samples from Urban areas and waste water treatment plants (wwtp's). A review on these Swedish screening studies that encompasses Urban areas and wwtp's can be found in WSP (2010). It appears that wwtp's can be major "sources¹" of many current use polar pollutants. There is also a potential for direct diffuse emissions through e.g. Urban and industrial stormwater.

As an assignment from the Swedish Environmental Protection Agency, WSP Environmental has during 2011-2012 performed a national screening investigation of three groups of relatively polar or volatile chemicals in the Swedish environment:

1. Fragrances: OTNE, acetyl cedren and diphenylether
2. Complexing agents: EDTA, NTA, DPTA, 1,3-PDTA and ADA
3. Three polar pollutants: TPPO, TMDD and TCEP.

A number of regional screening studies of the same chemicals have also been performed by the county administrative boards, and are reported jointly with the national screening study in this and two other reports.

1.2. Brief introduction to complexing agents

EDTA and NTA are the two most well-known examples of a group of chemicals referred to as aminopolycarboxylates. This study also includes the aminopolycarboxylates DPTA, 1,3-PDTA and ADA. Characteristic is their ability to form strong soluble complexes with a range of metals, so called chelates. This has led to their use in various industrial processes, in particular in paper and pulp industry. These chemicals are also used in various household products.

The aminopolycarboxylates are highly soluble in water and some of them are fairly persistent in the environment. Thus large amounts have been released to rivers from wwtp's and industries, leading to their ubiquitous presence in anthropogenically influenced rivers and

¹ WWTP's may not be considered as primary sources, because the chemicals emitted from wwtp's are generally used upstream of the wwtp.

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lakes (e.g. Schmidt et al., 2004). Actually EDTA has been identified as the major organic pollutant in wwtp effluents (Reemtsma et al., 2006) and one of the major organic pollutants in surface water (e.g. Barber et al., 1995).

It was reported that in Germany, environmental concerns led to the partial substitution of EDTA to more biodegradable complexing agents such as 1,3-PDTA and ADA (Schmidt et al., 2004). However, there are very sparse information on the environmental occurrence of 1,3-PDTA and ADA.

Previous investigations of the environmental occurrence of EDTA in Sweden is mainly restricted to paper and pulp recipients (e.g. Remberger, 2001). The latter study investigated the behavior of EDTA and DTPA in the large lake Vänern. Although EDTA is generally considered as fairly persistent in the environment, this study indicated that the behavior of EDTA was not entirely conservative within this lake.

In summary, it is known that aminopolycarboxylates are general aquatic pollutants and that they generally occur at comparably high levels.

The goals of these studies are to investigate if:

- these chemicals are found in the Swedish environment
- diffuse releases appears to occur
- they are present in background lakes
- elucidate their behavior in municipal waste water treatment plants
- providing data on the occurrence in wwtp effluents.
- industrial use may lead to a direct emission.

Based on these assessments, it is discussed whether the presence of these chemicals in the Swedish environment is a cause for concern.

2. Properties of the studied substances

2.1. Physical and chemical properties

The substances in this group are all aminopolycarboxylates, as shown in Table 1. Physical and chemical properties are summarised in Table 2. All substances have multiple carboxylate groups which explain the high solubility in water and the very low volatility. These acids will partially be deprotonated at the neutral pH values of most natural waters. None of the substances are lipophilic nor regarded as bioaccumulative.

Table 1. Structure and abbreviations of the studied compounds.

Abbreviation	CAS	Structure	Full name
EDTA	60-00-4		ethylenediaminetetraacetic acid
NTA	139-13-9		(nitrilotriacetic acid)
DTPA	67-43-6		diethylenetriamine-pentaacetic acid
1,3-PDTA	1939-36-2		propylenediaminetetraacetic acid
ADA	6245-75-6		

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Table 2. Physical and chemical properties of the studied compounds. Unless otherwise stated, data is from the compilation of COWI (2011). Data in italics are model estimates.

Property	EDTA	NTA	DTPA	PDTA	ADA
CAS	60-00-4	139-13-9	67-43-6	1939-36-2	6245-75-6
M, g mol ⁻¹	292,2	191,1	393,4		
log K _{ow}	-3	no data	-4,9	no data	no data
H, Pa m ³ mol ⁻¹	<i>10²⁰</i>	<i>2 · 10¹⁰</i>	ca <i>10⁻²⁵</i>	no data	no data
S _w , g/l	0,5-1000 ^f	640 [#]			

\$: ECHA database. E, The solubility in water is pH dependent and thus vary between the protonated form and the metal salts.# Solubility of NTA is for the sodium salt.

2.2. Degradation, bioaccumulation and toxicity

EDTA is known as a persistent chemical and is generally not degraded in municipal wwtp's (EC, 2004; COWI, 2011). Certain monitoring studies in wwtp's have suggested a degradation of less than 10%, whereas biodegradability studies generally show that EDTA is not readily biodegradable. Microbiological tests show that neither EDTA nor DTPA are biodegraded under aerobic conditions (Hinck et al., 1997). In contrast, NTA is considered as biodegradable (EC, 2008).

Photolysis is considered as the major degradation of EDTA, and this specifically concerns the Fe(III)-EDTA complex.

Due to the high water solubility, bioaccumulation is low and none of these chemicals is a PBT candidate. Neither is there is risk for secondary poisoning. The classification according to CLP appendix VI is as follows:

- EDTA: eye irritating.
- 1,3-PDTA: Acute Tox. 4 and eye damaging
- None of the other substances are classified.

Suggested PNEC values for aquatic ecosystems are presented in Table 3. The PNEC values for EDTA and NTA are from the EC risk assessments (ECB, 2004, 2005). NTA is suggested to be carcinogenic but only as a category 3, i.e. there is only weak evidence for carcinogenicity in humans.

Table 3. Bioaccumulation and toxicity of the studied compounds. Unless otherwise stated, data is from the compilation of COWI (2011). Data in italics are model estimates.

Property	EDTA	NTA	DTPA
Biodegradable	No	Ready b.d.	No
NOEC, mg/l, lowest reported value	22000	9300	1000
PNEC, µg/l, suggested value	2200	930	10
P (persistence)	Yes		

3. Use and release of aminopolycarboxylates

This chapter gives a brief presentation of how the studied chemicals are used, their function and possible emission pathways. All these compounds are used in various chemical forms, partly as the fully protonated acids but mainly as salts with e.g. sodium as the balancing cation. Thus, the actual aminocarboxylate ions are present under several CAS-numbers.

Paper- and pulpindustry is the major user of EDTA and DTPA in Sweden. Other industrial sectors using DTPA and EDTA include chemical, textile, photographic and industries. The registered use in Sweden, according to "Kemi-stat; www.kemi.se", was ca 4000-10000 tons/year of DTPA and EDTA, respectively, over the last few years. The use of NTA is roughly 2000-3000 tons per year, mainly used in detergents. The use of the sodium salts of DTPA, EDTA and NTA over the last two decades are shown in Figure 1.

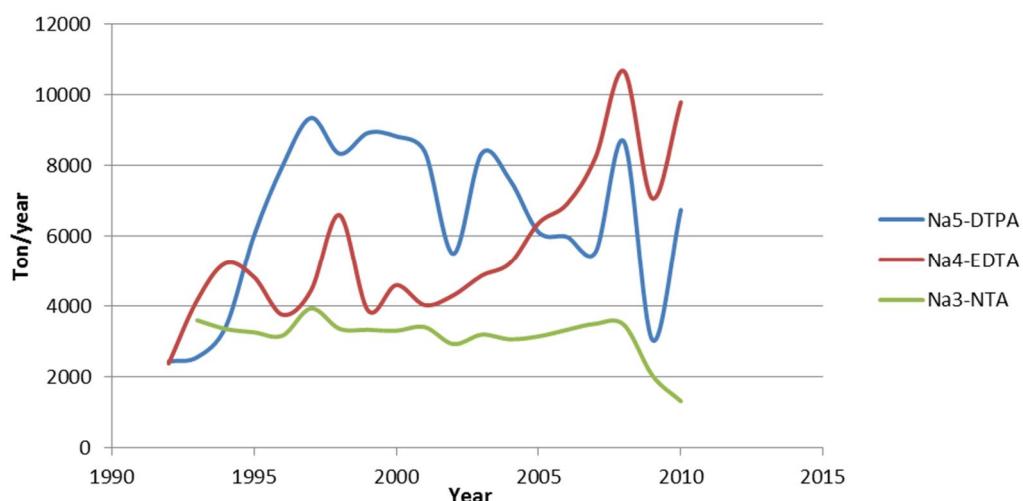


Figure 1. The use of sodium salts of DTPA, EDTA and NTA in Sweden (KemiStat; www.kemi.se).

To give a rough estimate of the potential for diffuse release of individual chemicals, the National Chemicals Inspectorate has developed an "exposure index". This index gives a value from 1 to 7, on relative terms, for the potential for release to e.g. WWTPs and for human exposure. The index considers both the amount used and the way the chemical is handled and used.

The exposure indexes for these chemicals are shown in Table 4, based on data for 2008. Of these five chemicals, EDTA and NTA display the highest exposure indexes whereas no index was estimated for ADA. It is also noteworthy that the exposure indexes for DTPA is much lower than for EDTA, although the total national use is in the same range (Figure 1). A former review of screening data from Urban areas and wwtp's showed a fairly good agreement between the exposure indexes and measured levels, although the scatter was wide (WSP, 2010).

Apart from the industrial use, these complexing agents are also used in various household products, e.g. hygiene products and detergents.

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Table 4. Exposure index (KemI, 2005; updates for 2008). The relative scale goes from 1 to 7. Trend indicates whether there has been a change in the exposure potential over the last years.

Substance	Exposure index					Trend	
	Surface water	Air	Soil	WWTP	Human	Human (-2 - +2)	Environment (-2 - +2)
EDTA 60-00-4	7	7	7	7	7	-	1
NTA 139-13-9	6	2	7	7	7		
DTPA 67-43-6	1	1	1	3	3	-	-
1,3-PDTA 1939-36-2	1	1	1	5	5	-	-
ADA 6245-75-6	-	-	-	-	-	-	-

4. Previous environmental studies

A selection of data on the occurrence in the environment and in wwtp's is presented in Table 5. The most comprehensive dataset comes from Germany, where monitoring data until 2001 was compiled by Schmidt et al. (2004). Annual average concentrations ranged from 0,7-12 µg/l (EDTA) at ca 50 sites along several major German rivers. Concentration of NTA and DTPA was slightly lower (Table 5).

1,3-PDTA and ADA have only been reported rarely in environmental studies. A few examples are given in Table 5.

Table 5. Environmental levels determined in previous studies. Sewage sludge in mg/kg dw; all other samples in µg/l.

Substance	Sewage sludge	Municipal waste water		Surface water	Study site	Reference	
		Incoming	effluent				
EDTA	0,07 (<0,02-56)	ca 30-110	ca 3-12	European wwtp's and rivers	Vättern	Reemtsma et al., 2006	
			3,3			Remberger, 2001	
			6-30	German river	Wind et al 2004		
		10-250	0,7-12	Average in several rivers	Schmidt et al., 2004		
		79-310	0,1-7,9	Norwegian Urban coast & wwtp	KLIF 2009		
NTA		1-3	0,5-6	German river	Swedish wwtp's	Svensson 2002	
		1-15	Average in several rivers			Wind et al 2004	
DTPA	1-4	0,2	Vättern	Average in several rivers	Remberger, 2001	see Schmidt et al., 2004	
		1-30	0,4-19				
1,3-PDTA		3-7				see Schmidt et al., 2004	

5. Sampling strategy and study areas

WSP developed a general strategy for the investigations, and this strategy was communicated and discussed with all participating county administrative boards as well as with the Swedish EPA. The study consists of a national programme, financed by the Swedish EPA, and regional programmes for 13 counties. In each county, the regional sampling programme was setup and implemented by the county administrative boards. The strategy of the national programme is outlined below

- A possible large-scale influence, resulting from long-range atmospheric transport
 - surface water in two national background lakes
 - groundwater from two northern sites.
- A possible diffuse urban influence was investigated by sampling in two urban regions (Stockholm, Eskilstuna): local background, city centre, and downstream.
- The role of wastewater was investigated at municipal sewage treatment plants and at the recipients of these STPs.
- To illustrate point source emissions, samples were taken in waste water from a paper industry, and in surface waters downstream.
- The influence on drinking water quality was assessed by sampling of untreated and treated drinking water from two large drinking water plants.

The national and regional programmes are summarised in Table 6 and Table 7. In total 149 samples were analysed. All sample details are listed in Appendix 1. The regional programmes were dominated by samples from wwtp's, whereas the national programme had a larger focus on the aquatic environment.

Table 6. National programme. The total number of samples is 39.

Category	Drinking water	Storm-water	Ground-water	Incoming waste water	Effluent waste water	Surface water
WWTP				5	5	
WWTP recipient						4
Background			2			2
Point source					2	5
Urban	4	4	2			4
<i>Grand Total</i>	4	4	4	5	7	15

Table 7. Regional programmes. The total number of samples is 110.

Category	Storm-water	Incoming waste water	Effluent waste water	Landfill leachate	Sediment	Sludge	Surface water
WWTP		14	50			2	4
WWTP recipient							8
Background					1		8
Point source				1	2		14
Urban	3						3
<i>Grand Total</i>	3	14	50	1	3	2	37

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6. Methods

6.1. Sampling

WSP developed general recommendations for sampling which were also communicated with the county administration boards. This protocol for sampling was sent to all personal involved in sampling, to assure similar treatment. Samples were stored dark and cold until transport to the laboratory within 1-2 days. Water samples were treated with acid before stored to stop any biological activity in the samples.

The national screening in Urban and industrial sites was performed mainly by WSP, but local contractors or personnel from the county administration boards were also involved in the sampling. Samples of surface water from background lakes were sampled by the county administration board in those counties. Water samples were generally taken as grab samples. Waste water and sewage sludge were sampled by staff at the waste water treatment plants, and was pursued in the same manner as the regulatory periodical sampling executed at each plant.

6.2. Chemical analysis

Chemical analyses were performed by ALS Scandinavia in cooperation with GBA Germany. The analyses were performed according to the methods outlined below.

Pretreatment

According to DIN EN ISO 16588-P10 (2004-4)

- Sample amount: depending on matrix 5- 20 mL
- Daily blank samples
- Internal standard: DPTA
- The sample volume is reduced to complete dryness with nitrogen
- Derivatization: tetrabutylation
- Extraction with hexane
- Concentration down to 0.2 mL

Measurement

- Analysis with GC/MS(EI), equipped with 30 m DB5ms column
- Daily 4-6 point-calibration
- Components out of linear working area: dilution

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Reporting limits

Analytical reporting limits are summarised in Table 8. These limits varied slightly between different samples due to different degrees of interfering substances. In general the lower values in the intervals given were most representative, whereas the higher values were valid for only one or a few samples.

Table 8. Reporting limits in different media.

Substance		EDTA	NTA	DTPA	1,3-PDTA	ADA
Surface water	$\mu\text{g/l}$	0,1-0,2	0,1-0,2	0,1-0,2	0,05-0,1	0,05-0,1
Sediment	mg/kg dw	0,01	0,01	0,01	0,01	0,01
wwtp incoming	$\mu\text{g/l}$	–	5	0,1-1	0,1-1	0,1-1
wwtp effluent	$\mu\text{g/l}$	–	5	0,1-1	0,1	0,1
wwtp sludge	mg/kg dw	0,02	0,04	0,08	0,04	0,04
Stormwater	$\mu\text{g/l}$	0,5	0,2	0,2	0,1	0,1
Drinking water	$\mu\text{g/l}$	–	0,2	0,2	0,1	0,1
Groundwater	$\mu\text{g/l}$	0,5	0,05	0,05-0,2	0,05-0,1	0,05-0,1

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7. Results

A general overview of the results is presented for each media in this section. Sample details are given in Appendix 1 and all data are presented in Appendix 2. Spatial trends, emission sources, environmental partitioning and possible risks to the health and environment are discussed in chapter 8.

Detection frequencies are summarised in Table 9. EDTA was the substance detected most frequently, closely followed by NTA. DTPA was commonly detected in wwtp effluents but less frequently in other media. 1,3-PDTA and ADA were only detected in a few samples of the entire study.

Table 9. The occurrence of five compounds in different media, where n is the number of samples analysed. When n < 10, the reporting frequency is given as a ratio rather than a percentage.

	EDTA	NTA	DTPA	1,3-PDTA	ADA
Surface water, n= 52	85%	63%	23%	4%	4%
Untreated drinking water, n=2	2 / 2	1 / 2	0 / 2	0 / 2	0 / 2
Treated drinking water; n=2	2 / 2	1 / 2	0 / 2	0 / 2	0 / 2
Groundwater, n=4	2 / 4	1 / 4	0 / 4	0 / 4	0 / 4
Incoming ww, n=19	100%	95%	37%	11%	11%
Effluent ww, n=55	98%	96%	73%	9%	16%
Industrial effluent, n=2	2 / 2	1 / 2	0 / 2	0 / 2	0 / 2
Stormwater n=7	5 / 7	6 / 7	0 / 7	0 / 7	0 / 7
Leachate, n=1	1 / 1	1 / 1	1 / 1	1 / 1	1 / 1
Sediment, n=3	2 / 3	2 / 3	0 / 3	0 / 3	0 / 3
Sludge, n=2	1 / 2	1 / 2	0 / 2	0 / 2	0 / 2

7.1. Surface water

Fiftytwo samples of surface water were analysed, including two national background lakes, 16 wwtp recipients, six Urban sites, 18 industrial point source recipients and 10 local background sites. EDTA was detected in most samples, followed by NTA and DTPA (Table 9). 1,3-PDTA and ADA were only detected in a few samples.

An overview of EDTA and NTA concentrations in the various site categories are shown in Figure 2. Although the scatter in each category is wide, some general observations of EDTA and NTA in surface water are:

- Waste water treatment plants are important sources of contamination
- Certain industrial sites also cause very high levels in the recipients. The importance of industrial point sources is also discussed in chapter 8.4.
- The diffuse impact on concentrations in Urban areas is moderate.

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Table 10. Summary of the results in this study. All concentrations in µg/l except sediments and sludge (mg/kg dw). Average and median values are given where detection frequency exceeds 50%. In such cases, non-detected levels are replaced by half the value of the reporting limit.

	EDTA	NTA	DTPA	1,3-PDTA	ADA
SURFACE WATER	n=52				
min	<0,1	<0,1	<0,1	<0,05	<0,05
max	930	19	12	0,1	0,13
median	1,6	0,19			
average	32	1,5			
std dev	136	3,4			
INCOMING WW	n=19				
min	7,7	<5	<0,1	<0,1	<0,1
max	350	1500	130	31	0,5
median	47	93			
average	79	212			
std dev	88	342			
EFFLUENT WW	n=55				
min	0,44	<5	<0,1	<0,1	<0,1
max	800	340	46	6	0,76
median	50	22	2,1		
average	98	44	7		
std dev	130	66	11		
INDUSTRIAL EFFLUENT	n=2				
min	8600	<5	<1	<0,5	<0,5
max	9700	26	<10	<5	<5
STORMWATER	n=7				
min	<0,5	<0,2	<0,2	<0,1	<0,1
max	310	4,5			
median	0,96	2,6			
average	45	2,2			
std dev	117	1,7			
GROUNDWATER	n=4				
min	<0,5	<0,05	<0,05	<0,05	<0,05
max	0,32	0,057			
DRINKING WATER	n=4				
min	1,1	<0,2	<0,2	<0,1	<0,1
max	1,6	2,1			
LANDFILL LEACHATE	n=1				
Value	73	0,81	22	0,6	0,13
SEDIMENT	n=3				
min	<0,01	<0,01	<0,02	<0,01	<0,01
max	0,085	0,39			
SLUDGE	n=2				
min	<0,02	<0,04	<0,07	<0,035	<0,035
max	0,072	0,19			

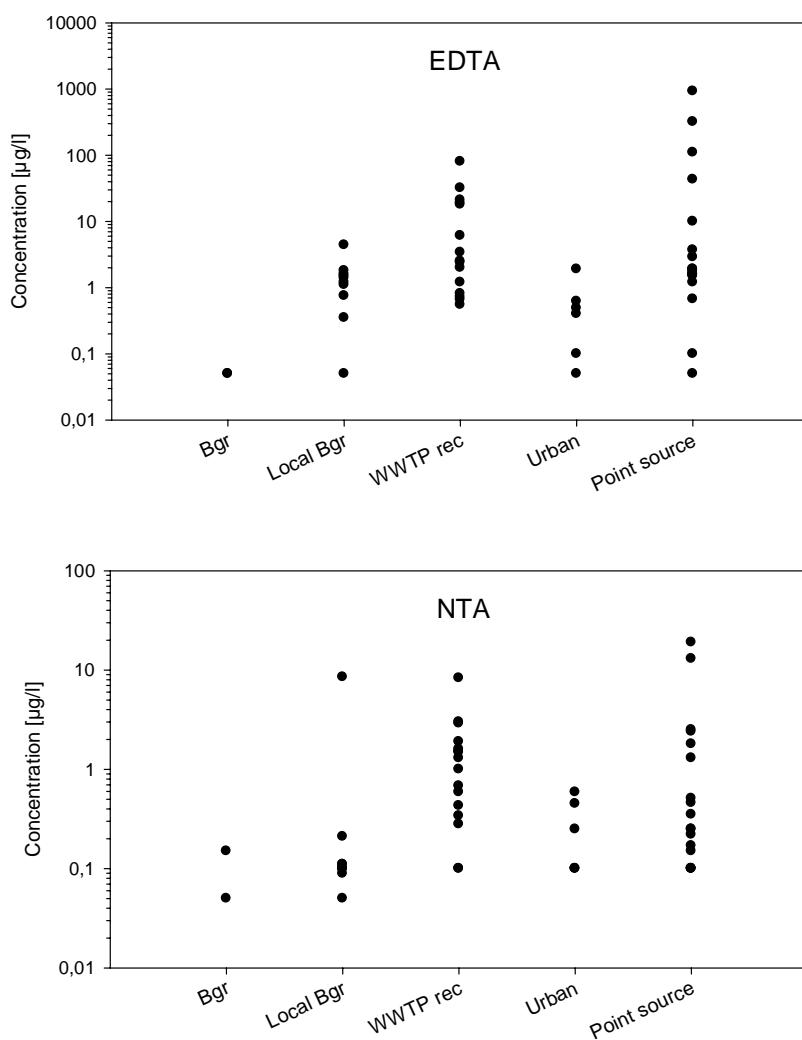


Figure 2. Concentrations of polyaminocarboxylates in surface waters, divided into the different site categories.

DTPA was mainly detected in lake Vättern, where six samples were taken close to a paper mill and showed concentrations of DTPA in the range 0,6-3,9 µg/l. Background concentration in Vättern was in the range 0,43-0,66 µg/l. In 1998, the average DTPA concentration in lake Vättern was ca 0,2 µg/l, with a maximum of 0,6 µg/l close to the paper mill (Remberger, 2001).

In contrast to the spatial gradients of DTPA in lake Vättern, EDTA occurred at similar concentrations (ca 1,5 µg/l) in both the "paper mill site" and the other sites in lake Vättern. Measurements of EDTA in Vättern in 1998 also showed homogenous concentrations but with an average of ca 3,3 µg/l (Remberger, 2001).

7.2. Sediment

Sediments were sampled from the recipients to two paper mills, and from an upstream reference site to one of these paper mills. Both EDTA and NTA were detected in the recipients, but not at the reference site. This is a clear indication of a point source influence. Concentrations at the recipients were 72-85 µg/kg for EDTA and 48-390 µg/kg for NTA. Previous measurements of EDTA in sediments from Edeboviken, also a paper mill recipient, showed similar concentrations (<7-153 µg/kg ts, Remberger, 2001).

7.3. Waste water and sewage sludge

Municipal waste water treatment plants were a major study category in this study, comprising 19 incoming waste waters, 55 effluents and 2 sludge samples. EDTA and NTA were detected in most samples of incoming waste water and effluents, whereas DTPA was less frequently detected. 1,3-PDTA and ADA were only detected in ca one tenth of the samples. All data for EDTA and NTA are shown statistically in Figure 3. The concentrations range over more than two orders of magnitude for each dataset. All the incoming and effluent waste waters are not from the same wwtp's. It nevertheless appears that EDTA levels are similar in incoming waste waters and effluents whereas for NTA, there is a tendency for lower levels in the effluents.

There are pairwise data for incoming waters and effluents from 19 wwtp's. According to a pairwise statistical test (Wilcoxon signed rank test), there is no difference between incoming waters and effluents for EDTA. For NTA, however, concentrations are on the average ca five times higher in the incoming water waters than in the effluents. German studies also showed that EDTA passes the wwtp essentially unretarded, whereas NTA is strongly reduced due to biodegradation (Schmidt et al., 2004).

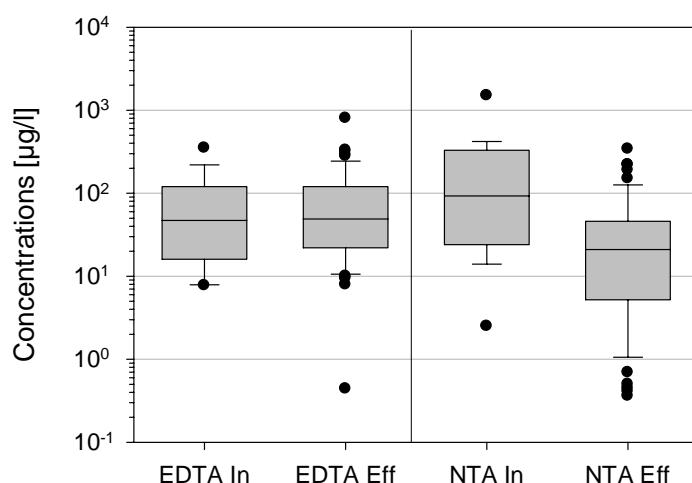


Figure 3. Comparison of concentrations of EDTA and NTA in incoming waste waters and effluents. Data consists of 19 samples of incoming waste water and 55 effluents.

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7.4. Drinking water and groundwater

Untreated and treated drinking water was collected from two drinking water plants. Both plants use surface water from Lake Mälaren to produce drinking water. EDTA and NTA were detected in the untreated water. In the treated drinking water, concentrations of EDTA was 20-30 % lower than in the untreated water. In one of the plants, NTA was reduced ca 50% following the treatment. In the other plant NTA was below reporting limit.

Groundwater was sampled at two background sites in northern Sweden, and in two Urban sites close to Stockholm. None of these complexing agents were found in the background sites, except for one hit of NTA, very close to the analytical reporting limit. EDTA was the only complexant detected in the Urban groundwaters, and the concentrations (0,32 µg/l) were lower than e.g. in the treated drinking water (ca 1 µg/l).

7.5. Stormwater, landfill leachate and industrial waste water

Seven samples of Urban stormwater were analysed. NTA was detected in six of these and at a few µg/l. EDTA was detected in five samples with one high outliers (320 µg/l). Neither DTPA, 1,3-PDTA nor ADA were detected in the stormwater samples.

Samples of industrial effluents were taken in the two ponds close to the outlet from a paper mill. EDTA was detected at almost 10 000 µg/l. None of the other substances were detected, except for NTA at moderate levels (26 µg/l). Samples of surface water were also taken upstream and downstream this industry, as will be discussed in chapter 8.4.

One leachate sample was taken from a landfill. Relatively high concentrations were found for EDTA (73 µg/l) and DTPA (22 µg/l). Also NTA, 1,3-PDTA and ADA were detected at sub-µg/l concentrations.

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8. Discussion

8.1. Background areas

Surface water from two national background lakes was sampled. Low levels (<0,1 µg/l) of 1,3-PDTA and ADA were detected in one of these lakes. The levels were very close to analytical reporting limit. Otherwise none of the complexants were detected in these background lakes.

As examples of local background sites, five lakes and a coastal site were also sampled. None of these sites can be regarded as remote sites. EDTA was detected in most of these samples and NTA was also common. DTPA was only detected in the samples from lake Vättern and neither 1,3-PDTA nor ADA were detected in the local background sites.

8.2. Urban areas

Many current use chemicals are released by diffuse processes. This may result in elevated levels in the Urban aquatic environment, which has been recognized in many studies of the Swedish national screening programme (see a review in WSP, 2010). In this report, we try to distinguish between this direct result of diffuse emissions and the impact that is caused by releases from wwtp's.

Several samples of stormwater (n=7) and surface water (n=6) were sampled in Urban areas in order to investigate whether there was a general diffuse influence on their environmental occurrence.

EDTA and NTA were detected in most stormwater samples but at relatively low levels; the remaining substances were not detected in Urban stormwater. EDTA and NTA were also detected in a few of the Urban surface waters and at low levels (compare Figure 2). Conclusively, diffuse Urban releases of polyaminocarboxylates do not appear as a major issue.

8.3. Waste water treatment plants

8.3.1. Variation between different wwtp's

The degree to which concentrations vary in waste waters is indicative of the chemicals sources to waste water. High variability or the existences of anomalously high values are indicative of e.g. point sources; low variability is indicative of a diffuse input.

Table 11 shows CV and skewness as two measures of variability for EDTA and NTA. CV is calculated as standard deviation divided by arithmetic average. A CV-value larger than ca 50% indicates non-normal distribution. A skewness close to zero would indicate normal distribution. Clearly both EDTA and NTA display significant variability between the samples, which is also indicated by the statistical summary in Table 10.

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Concentration variability was not apparently related to wwtp parameters such as size, domestic vs mixed load, stormwater etc. Certain wwtp's were sampled twice, with a few months in between. Concentrations of EDTA and NTA within each of these wwtp's varied widely at the two occasions. The differences were generally within a factor five but occasionally much larger. The variation was particularly large for NTA, with some indications of a seasonal pattern: the degradation appears to be more efficient during the warmer season.

Because of this temporal variation in concentration within individual wwtp's, it is unlikely that the results from single spot samples are representative for the long term load in each wwtp. Thus, the difference in long term load of EDTA and NTA to many of the wwtp's studied here may be lower than indicated by the concentrations in the spot samples. A general diffuse input of EDTA and NTA to the wwtp's is likely, although clearly point sources are important EDTA/NTA sources for at least some of the wwtp's in this study.

Table 11. Statistical description on variability of waste waters.

Chemical	n	CV	Skewness
EDTA incoming	19	112%	1,9
EDTA effluent	55	133%	3,4
NTA incoming	19	161%	3,3
NTA effluent	55	150%	2,8

The concentrations found in effluents and sludge from municipal wwtp's are compared to those reported in other studies in Table 12. Although the scatter is wide, a majority of the levels measured in this study are comparable to earlier data. A few high outliers are also present in the dataset.

It is also apparent that concentrations of EDTA and NTA are much higher than any of the large number of organic pollutants measured in Swedish effluents recently (e.g, Lilja et al., 2010; Österås et al, 2012; Sternbeck et al., 2012).

Table 12. Levels of EDTA, NTA and DTPA in sewage sludge (SS) and effluents (EFF) compared to previous studies. Concentrations are given as average and min-max. Average values are not shown where the detection frequency was low.

Substance	Present study		Previous studies		Reference
	Average	min-max	Average	min-max	
EDTA					
EFF ($\mu\text{g/l}$)	98	0,44-800		30-110	Reemstma et al., 2006
SS (mg/kg)		<0,02-0,072	0,07	<0,02-56	Svensson, 2002
NTA					
EFF ($\mu\text{g/l}$)	44	<5-340		1-15	Schmidt et al. 2004
SS (mg/kg)		<0,04-0,19			
DTPA					
EFF ($\mu\text{g/l}$)	7	<0,1-46		1-30	Schmidt et al., 2004
SS (mg/kg)		<0,07			

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8.3.2. Influence on recipients

As shown in the preceding section, most the EDTA and substantial fractions of NTA and DTPA appear to pass wwtp's unretained. Waste water treatment plants will thus be potentially important sources of these pollutants to recipients.

In this study surface water from 16 recipients were analysed for these complexing agents. It was demonstrated in chapter 7.1 that in general, concentrations of EDTA and NTA were much higher in wwtp recipients than in sites not influenced by wwtp effluents. Only in a few cases, however, did the sampling programmes of the wwtp recipients also include a sample from a local background or upstream site. These pair-wise samples confirmed the general conclusion that wwtp may influence the occurrence of EDTA and NTA in their recipients. The actual degree of contamination in each recipient is, of course, related the degree of waste water dilution in that recipient. The high potential of municipal wwtp's to influence recipients with EDTA was previously demonstrated by Reemstma et al (2006).

8.4. Point sources

A paper mill was selected in order to illustrate the influence of an industrial point source on surface water. The paper mill, that uses EDTA in the industrial process, was located along a smaller river. The industrial effluent passed two artificial ponds before reaching the river. A municipal waste water treatment plant was also located along this river, downstream the industrial outlet. The industrial effluent contained almost 10 mg/l of EDTA, low levels of NTA and non-detectable levels of the other complexants. The influence of the industrial effluent on EDTA in the surface water was significant (Figure 4). Although river water was diluted in the lakes, EDTA was present at 110 µg/l in the downstream lake, a concentration that by far exceeded most other surface water data in this study.

Surface waters were also sampled at ca 10 other sites where the industrial use of EDTA or other complexants was suspected or confirmed. As shown in Figure 2, most of these samples display EDTA and NTA levels in the same range as local background sites or wwtp recipients.

DTPA was also investigated in several surface water samples from lake Vättern. As presented in chapter 7.1, elevated concentrations adjacent to a paper and pulp mill clearly illustrated the influence of this point source. Actually the DTPA concentrations were slightly higher than in an earlier study in this area (Remberger, 2011). DTPA was also detected in this screening at 12 µg/l downstream a paper mill in Dalarna county.

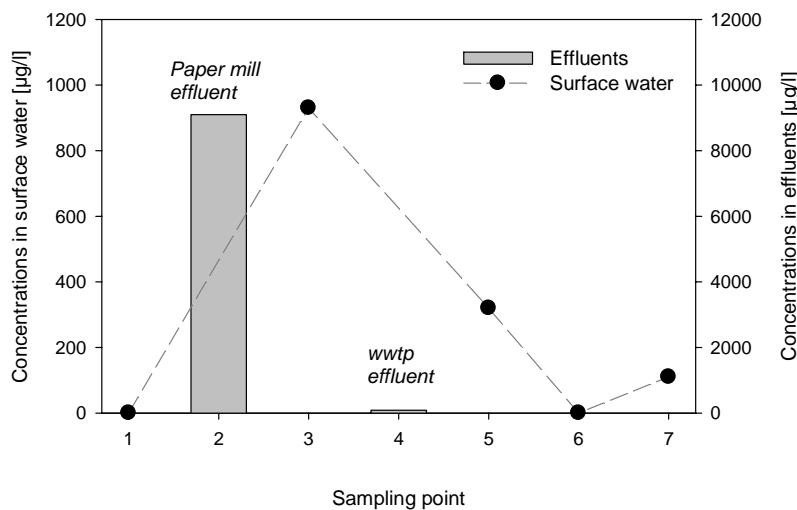


Figure 4. Concentrations of EDTA in surface waters in relation to concentrations in industrial and wwtp effluents. Sampling points of surface water are located as follows: 1. river upstream the point sources; 3. river, downstream the industry; 5. river downstream the wwtp; 6. a small lake below the river; 7. a larger lake downstream point 6.

8.5 Environmental significance of the observations

Based on results of this study, EDTA and NTA are those polyaminocarboxylates that are most widespread in the Swedish environment. The results agree well with the exposure indexes (Table 4): EDTA and NTA are widespread, DTPA occurs sporadically, and 1,3-PDTA and ADA occurs only rarely.

Possible hazards include

- direct toxicological effects on aquatic organisms or on the microbiological processes in wwtp's,
- human health effects due to exposure via drinking water
- indirect effects through the impact on trace metals in the environment

The safety margin between concentrations measured in the 52 samples of surface water and the corresponding PNEC values (Table 3) is generally high. Even the very high EDTA concentrations of 930 µg/l measured in a river just downstream a paper mill is lower than the PNEC value (2200 µg/l). However, considering the probable temporal variability in this river, a cause for concern cannot be excluded.

The PNEC values for microorganisms are used to assess the risk for negative impact on the microbial processes in municipal wwtp's. For EDTA and NTA these PNEC values are 50 and 540 mg/l, respectively (ECB, 2004; 2005). Compared to measured concentrations in incoming waste waters, there is clearly no risk at present.

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Human health effects that would be caused by other exposure pathways than the environment are not treated in this report. As stated before (ECB (2004), drinking water is the only significant human exposure pathway from the environment. EDTA and NTA were found in treated drinking water at ca 1-2 µg/l. This results in a human exposure many orders of magnitude lower than the corresponding TDI values (WHO, 2011).

The most pronounced consequence of the general presence of EDTA and NTA in the environment may be the influence of metal speciation, leaching, transport and bioavailability. To assess this question further requires detailed analysis in each case. A general conclusion cannot be drawn since the actual effect depends on the concentrations of metals at each site.

In summary, there are several factors that taken together call for a certain concern of EDTA and NTA:

- The ability of EDTA and NTA to influence metal cycling in natural waters
- Their general occurrence at relatively high concentrations in wwtp's and surface waters
- The persistence of EDTA in natural waters.

9. Conclusions

- Five complexing agents in the group of aminopolycarboxylates were studied in this report.
 - EDTA and NTA were common pollutants in both surface waters and waste waters.
 - DTPA was detected only in a few lakes, but more commonly in waste waters.
 - 1,3-PDTA and ADA were only detected in a few samples.
- Concentrations of EDTA in both surface waters and waste waters are higher than for most other known organic pollutants.
- EDTA and NTA appears to be emitted both by point sources and by diffuse inputs.
- Municipal waste water treatment plants are important general sources of contamination.
- Concentrations of EDTA are similar in incoming and effluent waste waters.
- NTA is partially degraded in municipal waste water treatment plants.
- Certain pulp and paper industries cause substantial contamination of EDTA in their recipients.
- Based on risk assessment, neither EDTA nor NTA appears to pose a direct eco-toxicological risk.

				Not covered in this study	Not covered in this study
	Long range transport	Diffuse emissions	Point sources	Bioaccumulation	Human exposure
EDTA	No	Yes	Yes		
NTA	No	Yes	?		
DTPA	No	?	Yes		
1,3-PDTA	No	No	No		
ADA	No	No	No		

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10. Acknowledgments

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WSP Environmental

11 October 2012

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Appendix 1. Sample details

Appendix 1.1. Drinking water, groundwater and stormwater. Note that positions are given in different geographical systems.

Sample	Programme	County	Municipality	Media	Site name	Position X	Position Y	Category	Sampling date
WSP_439	National	Stockholm		Untreated drinking water	Norrvatten			Urban	111011
WSP_441	National	Stockholm		Untreated drinking water	Norsborg			Urban	sept-11
WSP_440	National	Stockholm		Drinking water	Norrvatten			Urban	111011
WSP_442	National	Stockholm		Drinking water	Norsborg			Urban	sept-11
WSP_397	National	Norrbotten	Gällivare	Groundwater	Ripats	7436641	754080	Background	2011-08-15
WSP_398	National	Norrbotten	Arjeplog	Groundwater	Kallkällmyran	7317761	638561	Background	2011-08-16
WSP_437	National	Stockholm	Stockholm-Vällingby	Groundwater	Råcksta	143150	6581898	Urban	2011-09-28
WSP_438	National	Stockholm	Stockholm-Älvsjö	Groundwater	Älvsjömässan	150791	6573018	Urban	2011-09-28
C_305	Regional	Uppsala	Uppsala	Stormwater	Librobäck	6640771	1600349	Urban	2011-10-04
C_306	Regional	Uppsala	Uppsala	Stormwater	Bärby	6641071	1600882	Urban	2011-10-04
C_307	Regional	Uppsala	Uppsala	Stormwater	Industristaden	6638043	1603366	Urban	2011-10-04
WSP_411	National	Stockholm	Stockholm	Stormwater	Stockholm, Mälarhöjden,			Urban	2012-06-01
WSP_412	National	Stockholm	Stockholm	Stormwater	Stockholm, Liljeholmskajen,			Urban	2012-06-01
WSP_431	National	Södermanland	Eskilstuna	Stormwater	Eskilstuna	6583283	585485	Urban	110921
WSP_432	National	Stockholm	Stockholm	Stormwater	Stockholm innerstad, Södermalm			Urban	2012-06-01

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Status: Final report



Appendix 1.2. Surface water. Note that positions are given in different geographical systems.

Provnummer	Programme	County	Municipality	Media	Site name	Position X	Position Y	Category	Sampling date
W_08	Regional	Dalarna	Falun	Surface water	Runn	6707902	1490933	Local back-ground	2011-09-29
W_11	Regional	Dalarna	Avesta	Surface water	Bäsingen	6671468	1532945	Point source	2011-09-25
W_13	Regional	Dalarna	Borlänge	Surface water	Dalälven, downstream paper mill	6711589	1479245	Point source	2011-10-06
W_14	Regional	Dalarna	Falun	Surface water	Grycken	6727689	1484482	Point source	2011-09-28
F_39	Regional	Jönköping	Jönköping	Surface water	Munksjön	6403642	450352	WWTP REC	2011-09-22
F_41	Regional	Jönköping	Gislaved	Surface water	Anderstorpsån	6346442	415225	Urban	2011-10-05
F_42	Regional	Jönköping	Vadstena	Surface water	Vättern Jungfrun	6484467	481083	Local back-ground	2011-08-30
F_43	Regional	Jönköping	Jönköping	Surface water	Vättern Edeskvarna	6418631	454151	Local back-ground	2011-08-30
F_44	Regional	Jönköping	Vadstena	Surface water	Vättern Jungfrun	6484467	481083	Local back-ground	2011-08-30
F_45	Regional	Jönköping	Jönköping	Surface water	Vättern Edeskvarna	6418631	454151	Local back-ground	2011-08-30
F_46	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6509573	488568	Point source	2011-08-30
F_47	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6509374	489910	Point source	2011-08-30
F_48	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6509965	491677	Point source	2011-08-30
F_49	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6511089	492898	Point source	2011-08-30
F_50	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6512558	493271	Point source	2011-08-30
F_51	Regional	Jönköping	Askersund	Surface water	Vättern paper mill	6513792	493221	Point source	2011-08-30
F_52	Regional	Jönköping	Eksjö	Surface water	Torsjöän	6387132	499682	Point source	2011-11-21
F_57	Regional	Jönköping	Vetlanda	Surface water	Vetlandabäcken	6363872	506233	Urban	2011-11-23

Appendix 1.2. Continued.

Sample	Programme	County	Municipality	Media	Site name	Position X	Position Y	Category	Sampling date
F_59	Regional	Jönköping	Vetlanda	Surface water	Emån ned Vetlanda	6364546	507434	WWTP REC	2011-11-22
I_61	Regional	Gotland	Gotland	Surface water	Åminne	6391826	724271	Local back-ground	2011-09-26
D_155	Regional	Södermanland	Katrineholm	Surface water	Djulösjön	6537300	570920	WWTP REC	2011-09-28
D_160	Regional	Södermanland	Flen	Surface water	Gårdsjön	6546818	590520	WWTP REC	2011-09-28
H_241	Regional	Kalmar	Mönsterås	Surface water	Lervik	594111	6326224	Point source	25-okt-11
H_242	Regional	Kalmar	Mönsterås	Surface water	Outside the harbour	595106	6329397	Local back-ground	25-okt-11
H_243	Regional	Kalmar	Mönsterås	Surface water	Outside Ödängla	596670	6324945	Local back-ground	25-okt-11
H_244	Regional	Kalmar	Mönsterås	Surface water	Emån	592031	6332863	Point source	25-okt-11
C_301	Regional	Uppsala	Uppsala	Surface water	Vindbron, Fyrisån	6636137	1604114	WWTP REC	2011-09-28
C_308	Regional	Uppsala	Östhammar	Surface water	Krutuddens wwtp	6685572	1643415	WWTP REC	2011-10-04
C_312	Regional	Uppsala	Enköping	Surface water	Enköpings wwtp	6612069	1571592	WWTP REC	2011-09-28
S_331	Regional	Värmland	Ärjäng	Surface water	Kyrkbruds wwtp	6584925	335558	WWTP	2011-12-13
S_335	Regional	Värmland	Storfors	Surface water	Storforsälven	6601914	1412306	WWTP	jan-12
S_339	Regional	Värmland	Kristinehamn	Surface water	Kristinehamn wwtp	6577984	1400628	WWTP	2011-12-14
S_343	Regional	Värmland	Sunne	Surface water	Kolsnäs	6634409	129770	WWTP	120104
E_375	Regional	Östergötland	Linköping	Surface water	Stångån downstream Nykvarns wwtp			WWTP REC	111025
E_376	Regional	Östergötland	Linköping	Surface water	Stångån upstream Nykvarns wwtp			Urban	111025

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Sample	Programme	County	Municipality	Media	Site name	Position X	Position Y	Category	Sampling date
WSP_391	National	Västernorrland	Örnsköldsvik	Surface water	Remmarsjön			Background	2011-07-12
WSP_394	National	Örebro	Hällefors	Surface water	Limmingssjön			Background	2011-08-17
WSP_413	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån	6584822	583205	WWTP REC	110921
WSP_414	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån, Torshälla	6587467	583397	WWTP REC	110921
WSP_415	National	Stockholm	Stockholm	Surface water	Blockhusudden, 0,5 m			WWTP REC	september
WSP_416	National	Stockholm	Stockholm	Surface water	blockhusudden, 10 m			WWTP REC	september
WSP_425	National	Södermanland		Surface water	Eskilstunaån, Rosenfors	6577910	584015	Urban	110921
WSP_426	National	Södermanland	Eskilstuna	Surface water	Eskilstunaån	6583857	583152	Urban	110921
WSP_427	National	Stockholm	Stockholm	Surface water	Klubbensborg			Urban	sept-11
WSP_428	National	Stockholm	Stockholm	Surface water	Årstaviken			Urban	sept-11
WSP_460	National	Kronoberg	Lessebo	Surface water	Lesseboån inlet	517502	6289606	Point source	
WSP_463	National	Kronoberg	Lessebo	Surface water	Lesseboån, downstream paper mill	515910	6289246	Point source	
WSP_464	National	Kronoberg	Lessebo	Surface water	Lesseboåns outlet	515452	6289040	Point source	
WSP_465	National	Kronoberg	Lessebo	Surface water	Osets outlet	514826	6288736	Point source	
WSP_466	National	Kronoberg	Lessebo	Surface water	Sörsjöns outlet	514442	6287749	Point source	
F_491	Regional	Jönköping	Vetlanda	Surface water	Paper mill	6368906	531285	Point source	2011-11-22
F_492	Regional	(Jönköping)Kalmar	Hultsfred	Surface water	Storgölen, landfill	6364673	549172	Point source	2011-11-22

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Appendix 1.3. Industrial effluent, leachate, sediments and sludge.

Sample	Programme	County	Municipality	Media	Site name	Position X	Position Y	Category	Sampling date
WSP_461	National	Kronoberg	Lessebo	Industrial effluent	Paper mill, treatment pond			Point source	May -12
WSP_462	National	Kronoberg	Lessebo	Industrial effluent	Paper mill, treatment pond			Point source	May-12
D_154	Regional	Södermanland	Eskilstuna	Landfill leachate	Lilla Nyby	6579430	587524	Point source	110906
W_10	Regional	Dalarna	Falun	Sediment	Runn	6707902	1490933	Local back-ground	2011-09-29
W_12	Regional	Dalarna	Avesta	Sediment	Bäsingen	6671468	1532945	Point source	2011-09-25
W_15	Regional	Dalarna	Falun	Sediment	Grycken	6727689	1484482	Point source	2011-09-28
F_32	Regional	Jönköping	Jönköping	Sludge	Simsholmen wwtp	6403325	450565	WWTP	2011-10-10
F_34	Regional	Jönköping	Jönköping	Sludge	Huskvarna wwtp	6405841	456163	WWTP	2011-10-11
S_344	Regional	Värmland	Sunne	Sludge	Sunne wwtp	6636693	131770	WWTP	120104

Appendix 1.4. Incoming waste waters.

Sample	Pro-gramme	County	Municipali-ty	Site name	Category	Sampling date	Size, pe	Load	Stormwater load	Active sludge	Chem. Prec.
W_02	Regional	Dalarna	Borlänge	Borlänge	WWTP	111006	34 000 ,	mix	yes	yes	yes
W_05	Regional	Dalarna	Falun	Främby	WWTP	111011	45000	mix	yes	yes	yes
T_121	Regional	Örebro	Ljusnars-berg	Bångbro	WWTP	110629	33000	ind	yes	yes	yes
T_124	Regional	Örebro	Nora	Nora	WWTP	110629	8500	dom	yes	yes	yes
T_127	Regional	Örebro	Ljusnars-berg	Bångbro	WWTP	111027	33000	ind		yes	yes
T_130	Regional	Örebro	Nora	Nora	WWTP	111027	8500	dom		yes	yes
M_271	Regional	Skåne	Helsingborg	Öresundsverket	WWTP	111213	200000	dom	yes	yes (bio-P)	no
C_302	Regional	Uppsala	Uppsala	Kungsängsverket	WWTP	110928	200 000	dom	no	yes	yes
C_309	Regional	Uppsala	Östhammar	Krutudden	WWTP	111004	4700	dom	yes	yes	yes
C_313	Regional	Uppsala	Enköping	Enköping	WWTP	110928	30 000	mix	yes	yes	yes
S_332	Regional	Värmland	Årjäng	Kyrkbrud	WWTP	111213	5000	dom	no	yes	yes
S_336	Regional	Värmland	Storfors	Storfors	WWTP	jan-12	4500	dom	yes	no	yes (pax)
S_340	Regional	Värmland	Kristine-hamn	Kristinehamn	WWTP	111214	12081	mix	yes	yes	yes
WSP_399	National	Söderman-land	Eskilstuna	Ekeby	WWTP	110628	94000	dom	yes	yes	yes
WSP_402	National	Stockholm	Stockholm	Henriksdal	WWTP	110726	ca 700 000	dom		yes	yes
WSP_405	National	Söderman-land	Eskilstuna	Ekeby	WWTP	sep-11	94000	dom	yes	yes	yes
WSP_408	National	Stockholm	Stockholm	Henriksdal	WWTP	110919	ca 700 000	dom		yes	yes
WSP_467	National	Kronoberg	Lessebo	Lessebo	WWTP						

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Appendix 1.5. Effluents from municipal waste water treatment plants.

Sample	Pro-gramme	County	Municipality	Site name	Sampling date	Size,	Load	Stormwater load	Active sludge	Chem prec.
W_04	Regional	Dalarna	Borlänge	Borlänge	2011-10-06	34 000 ,	mix	yes	yes	yes
W_07	Regional	Dalarna	Falun	Främby	111011	45 000	mix	yes	yes	yes
F_31	Regional	Jönköping	Jönköping	Simsholmen	2011-10-10	61700	dom	yes	yes	yes
F_33	Regional	Jönköping	Jönköping	Huskvarna	2011-10-11	27543	dom	yes	yes	yes
F_35	Regional	Jönköping	Jönköping	Bankeryd	2011-10-10	4143	dom	yes	yes	yes
F_37	Regional	Jönköping	Jönköping	Gränna	2011-10-10	4083	dom	yes	yes	yes
F_55	Regional	Jönköping	Vetlanda	Vetlanda	2011-11-23	19300	mix	yes	yes	yes
I_64	Regional	Gotland	Gotland	Visby	2011-09-27	60000	mix		yes	yes
G_92	Regional	Kronoberg	Alvesta	Alvesta	111212	12000	dom	no	yes	yes
G_96	Regional	Kronoberg	Lessebo	Lessebo	111201	9000	mix	no	yes	yes
G_100	Regional	Kronoberg	Ljungby	Ljungby	111122	33000	mix	no	yes	yes
G_104	Regional	Kronoberg	Markaryd	Ribersdals	120305	10000	dom	no	yes	yes
G_108	Regional	Kronoberg	Tingsryd	Tingsryds	111201	42000	dom	no	yes	yes
G_111	Regional	Kronoberg	Uppvidinge	Åseda	111206	6000	mix	no	yes	yes
G_114	Regional	Kronoberg	Växjö	Sundet	111201	80000	mix	no	yes	yes
G_118	Regional	Kronoberg	Älmhult	Älmhult	111123	22700	dom	no	yes	yes
T_122	Regional	Örebro	Ljusnarsberg	Bångbro	110629	33000	ind	yes	yes	yes
T_125	Regional	Örebro	Nora	Nora	110629	8500	dom	yes	yes	yes
T_128	Regional	Örebro	Ljusnarsberg	Bångbro	111027	33000	ind		yes	yes
T_131	Regional	Örebro	Nora	Nora	111027	8500	dom		yes	yes
D_151	Regional	Södermanland	Eskilstuna	Ekeby	110906	< 94000	dom/mix	yes	yes	yes

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Sample	Pro-gramme	County	Municipality	Site name	Sampling date	Size,	Load	Stormwater load	Active sludge	Chem prec.
D_157	Regional	Södermanland	Katrineholm	Rosenholm	111011	< 53000	mix	yes	no	yes
D_162	Regional	Södermanland	Flen	Flens	111011	< 19900	mix	yes	yes	yes
Y_181	Regional	Västernorrland	Sundsvall	Fillanverket	2011-10-19	21600	mix		yes	yes
Y_182	Regional	Västernorrland	Sundsvall	Tivoliverket	2011-10-19	53000	mix		yes	yes
Y_183	Regional	Västernorrland	Örnsköldsvik	Knorthems	2011-10-04	12500	dom		yes	yes,
Y_184	Regional	Västernorrland	Sollefteå	Hågesta	2011-09-01	13150	mix	yes	no	yes,
BD_211	Regional	Norrbotten	Luleå	Uddebo	120209	Ca 60 000	dom		no	yes
BD_213	Regional	Norrbotten	Piteå	Sandholmen	111028	Ca 30 500	dom		no	yes
BD_215	Regional	Norrbotten	Gällivare	Kavahedens		18 000-20 000	mix		yes	yes
M_272	Regional	Skåne	Helsingborg	Öresundsverket	111213	200000	dom	yes	yes	no
C_303	Regional	Uppsala	Uppsala	Kungsängsverket	40814	200 000	dom	no	yes	yes
C_310	Regional	Uppsala	Östhammar	Krutuddens	40820	4700	dom	yes		
C_314	Regional	Uppsala	Enköping	Enköpings	2011-09-28	30 000	mix	yes	yes	yes
S_333	Regional	Värmland	Årjäng	Kyrkbruds	2011-12-13	5000	dom	no	yes	yes
S_337	Regional	Värmland	Storfors	Storfors	jan-12	4500	dom	yes	no	yes
S_341	Regional	Värmland	Kristinehamn	Kristinehamn	2011-12-14	12081	mix	yes	yes	yes
S_345	Regional	Värmland	Sunne	Sunne rv	120104	7500	dom	no	no	yes
E_361	Regional	Östergötland	Åtvidaberg	Håckla	110926	7700	dom	yes	no	yes
E_363	Regional	Östergötland	Mjölby	Gudhem	111019	6000	dom	yes	yes	yes
E_365	Regional	Östergötland	Mjölby	Mjölkulla	111019	55000	dom	yes	yes	yes
E_367	Regional	Östergötland	Motala	Karshult	10-16 okt 2011	40000	dom	yes	yes	yes
E_369	Regional	Östergötland	Vadstena	Vadstena	10-16 okt 2011	9500	dom	yes	no	yes
E_371	Regional	Östergötland	Norrköping	Slottshagen	111012	200000	dom	yes	yes	yes

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Sample	Pro-gramme	County	Municipality	Site name	Sampling date	Size,	Load	Stormwater load	Active sludge	Chem prec.
E_373	Regional	Östergötland	Linköping	Nykvarns	111025	235000	dom	yes	yes	yes
WSP_400	National	Södermanland	Eskilstuna	Ekeby	2011-06-28	94000	dom	yes	yes	yes
WSP_403	National	Stockholm	Stockholm	Henriksdal	2011-07-26	ca 700 000	dom			yes
WSP_406	National	Södermanland	Eskilstuna	Ekeby	september	94000	dom	yes	yes	yes
WSP_409	National	Stockholm	Stockholm	Henriksdal	110919	ca 700 000	dom			yes
WSP_468	National	Kronoberg	Lessebo	Lessebo						
F_495	Regional	Jönköping	Gislaved	Gislaved	2011-10-11	17573	dom	yes	no	yes
F_496	Regional	(Jönköping)Kalmar	Hultsfred	Hultsfred	2011-09-27	8500	mix	yes	yes	yes
F_497	Regional	Jönköping	Eksjö	Eksjö	2011-09-20	16200	mix	yes	yes	yes
F_498	Regional	Jönköping	Nässjö	Nässjö	2011-09-27	17079	dom	yes	no *	yes
F_499	Regional	Jönköping	Tranås	Tranås	2011-10-17	18000	mix	yes	yes	yes

Appendix 2. Analytical results

Sample no	Unit	Media	Category	EDTA	NTA	DTPA	1,3-PDTA	ADA
WSP_439	µg/l	Untreated drinking water	Urban	1,6	2,1	<0.20	<0.10	<0.10
WSP_441	µg/l	Untreated drinking water	Urban	1,2	<0.20	<0.20	<0.10	<0.10
WSP_440	µg/l	Drinking water	Urban	1,3	1,1	<0.22	<0.12	<0.10
WSP_442	µg/l	Drinking water	Urban	1,1	<0.20	<0.20	<0.10	<0.10
C_305	µg/l	Stormwater	Urban	2,4	2,8	<0.20	<0.10	<0.10
C_306	µg/l	Stormwater	Urban	0,3	<0.20	<0.20	<0.10	<0.10
C_307	µg/l	Stormwater	Urban	310	0,5	<0.20	<0.10	<0.10
WSP_411	µg/l	Stormwater	Urban	0,96	3,9	<1.0	<0.50	<0.50
WSP_412	µg/l	Stormwater	Urban	<0.50	4,5	<1.0	<0.50	<0.50
WSP_431	µg/l	Stormwater	Urban	1,3	0,87	<0.20	<0.10	<0.10
WSP_432	µg/l	Stormwater	Urban	<0.50	2,6	<1.0	<0.50	<0.50
WSP_397	µg/l	Groundwater	Background	<0.50	<0.050	<0.050	<0.050	<0.050
WSP_398	µg/l	Groundwater	Background	<0.50	0,057	<0.050	<0.050	<0.050
WSP_437	µg/l	Groundwater	Urban	0,32	<0.10	<0.20	<0.10	<0.10
WSP_438	µg/l	Groundwater	Urban	0,32	<0.10	<0.20	<0.10	<0.10
WSP_461	µg/l	Ind effluent	point source	9700	26	<10	<5.0	<5.0
WSP_462	µg/l	Ind effluent	point source	8600	<5.0	<1.0	<0.50	<0.50
D_154	µg/l	landfill leachate	point source	73	0,81	22	0,6	0,13
W_10	mg/kg	SED	Lokal Back-ground	<0.010	<0.010	<0.020	<0.010	<0.010
W_12	mg/kg	SED	point source	0,085	0,048	<0.034	<0.017	<0.017
W_15	mg/kg	SED	point source	0,072	0,39	<0.12	<0.056	<0.056
F_32	mg/kg	sludge	wwtp	0,072	0,19	<0.070	<0.035	<0.035
F_34	mg/kg	sludge	wwtp	<0.020	<0.040	<0.080	<0.040	<0.040
S_344	mg/kg	sludge	wwtp					
W_08	µg/l	surface water	Lokal back-ground	0,75	8,5	<0.20	<0.10	<0.10
W_11	µg/l	surface water	point source	10	19	<0.20	<0.10	<0.10
W_13	µg/l	surface water	point source	43	1,3	12	<0.10	<0.10
W_14	µg/l	surface water	point source	0,67	13	<0.20	<0.10	<0.10
F_39	µg/l	surface water	wwtp rec	2,4	1,5	<0.20	<0.10	<0.10
F_41	µg/l	surface water	Urban	0,4	0,45	<0.20	<0.10	<0.10
F_42	µg/l	surface water	Lokal back-ground	1,5	0,089	0,43	<0.050	<0.050
F_43	µg/l	surface water	Lokal back-ground	1,8	0,11	0,48	<0.050	<0.050
F_44	µg/l	surface water	Lokal back-	1,6	0,11	0,62	<0.050	<0.050

Sample no	Unit	Media	Category	EDTA	NTA	DTPA	1,3-PDTA	ADA
			ground					
F_45	µg/l	surface water	Lokal Back-ground	1,4	0,1	0,66	<0.050	<0.050
F_46	µg/l	surface water	point source	1,9	0,51	3,9	<0.050	<0.050
F_47	µg/l	surface water	point source	1,9	0,35	1,5	<0.050	<0.050
F_48	µg/l	surface water	point source	1,6	0,15	0,58	<0.050	<0.050
F_49	µg/l	surface water	point source	1,8	0,46	3,3	<0.050	<0.050
F_50	µg/l	surface water	point source	1,5	0,17	0,64	<0.050	<0.050
F_51	µg/l	surface water	point source	1,6	0,22	0,74	<0.050	<0.050
F_52	µg/l	surface water	point source	1,2	<0.50	<0.20	<0.10	<0.10
F_57	µg/l	surface water	Urban	<0.20	<0.50	<0.20	<0.10	<0.10
F_59	µg/l	surface water	wwtp REC	<0.20	<0.20	<0.20	<0.10	<0.10
I_61	µg/l	surface water	Lokal Back-ground	0,35	<0.10	<0.20	<0.10	<0.10
D_155	µg/l	surface water	wwtp REC	18	3	<0.20	<0.1	<0.1
D_160	µg/l	surface water	wwtp REC	1,2	1,9	<0.20	<0.1	<0.1
H_241	µg/l	surface water	point source	3,7	<0.20	<0.20	<0.10	<0.10
H_242	µg/l	surface water	lokal Back-ground	4,4	<0.20	<0.20	<0.10	<0.10
H_243	µg/l	surface water	lokal Back-ground	1,1	<0.20	<0.20	<0.10	<0.10
H_244	µg/l	surface water	point source	1,6	<0.20	<0.20	<0.10	<0.10
C_301	µg/l	surface water	wwtp REC	80	2,9	0,37	<0.10	0,13
C_308	µg/l	surface water	wwtp REC	2,5	0,43	<0.20	<0.10	<0.10
C_312	µg/l	surface water	wwtp REC	19	1,6	<0.20	<0.10	<0.10
S_331	µg/l	surface water	wwtp	0,55	0,34	<0.20	<0.10	<0.10
S_335	µg/l	surface water	wwtp	0,72	1,5	<0.20	<0.10	<0.10
S_339	µg/l	surface water	wwtp	2	1,3	<0.20	<0.10	<0.10
S_343	µg/l	surface water	wwtp	21	<0.20	<0.20	<0.10	<0.10
E_375	µg/l	surface water	wwtp REC	32	8,3	<0.20	<0.10	<0.10
E_376	µg/l	surface water	Urban	<0.10	<0.20	<0.20	<0.10	<0.10
WSP_391	µg/l	surface water	Background	<0.1	<0.1	<0.1	<0.1	<0.1
WSP_394	µg/l	surface water	Background	<0.5	0,15	<0.050	0,084	0,056
WSP_413	µg/l	surface water	wwtp REC	0,66	<0.57	<0.20	<0.10	<0.10
WSP_414	µg/l	surface water	wwtp REC	0,81	0,59	<0.20	<0.10	<0.10
WSP_415	µg/l	surface water	wwtp REC	3,4	0,68	<0.20	<0.10	<0.10
WSP_416	µg/l	surface water	wwtp REC	6,1	1	<0.10	0,1	<0.10
WSP_425	µg/l	surface water	Urban	0,62	<0.20	<0.20	<0.10	<0.10
WSP_426	µg/l	surface water	Urban	0,49	<0.20	<0.20	<0.10	<0.10
WSP_427	µg/l	surface water	Urban	1,2	0,21	<0.20	<0.10	<0.10
WSP_428	µg/l	surface water	Urban	1,9	0,59	<0.20	<0.10	<0.10
WSP_460	µg/l	surface water	point source	<0.10	<0.20	<0.20	<0.10	<0.10
WSP_463	µg/l	surface water	point source	930	<5.0	<1.0	<0.50	<0.50

Sample no	Unit	Media	Category	EDTA	NTA	DTPA	1,3-PDTA	ADA
WSP_464	µg/l	surface water	point source	320	2,4	<1.0	<0.50	<0.50
WSP_465	µg/l	surface water	point source	<0.10	<0.20	<0.20	<0.10	<0.10
WSP_466	µg/l	surface water	point source	110	1,8	<1.0	<0.50	<0.50
F_491	µg/l	surface water	point source	<0.20	<0.50	<0.20	<0.10	<0.10
F_492	µg/l	surface water	point source	2,9	<0.20	<0.20	<0.10	<0.10
W_02	µg/l	Incoming waste water	wwtp	80	420	<0.20	<0.10	<0.10
W_05	µg/l	Incoming ww	wwtp	18	330	<0.20	<0.10	<0.10
T_121	µg/l	Incoming ww	wwtp	9,3	14	0,41	<0.1	<0.1
T_124	µg/l	Incoming ww	wwtp	150	15	0,94	<0.1	<0.1
T_127	µg/l	Incoming ww	wwtp	47	24	<1.0	<0.50	<0.50
T_130	µg/l	Incoming ww	wwtp	7,7	14	<1.0	<0.50	<0.50
M_271	µg/l	Incoming ww	wwtp	57	340	4,1	<0.50	<0.50
C_302	µg/l	Incoming ww	wwtp	33	97	<0.20	<0.10	<0.10
C_309	µg/l	Incoming ww	wwtp	67	220	2,1	<0.10	0,5
S_332	µg/l	Incoming ww	wwtp	31	24	130	<0.10	<0.10
S_336	µg/l	Incoming ww	wwtp	13	38	<1.0	<0.25	<0.25
S_340	µg/l	Incoming ww	wwtp	16	160	4,4	<0.10	<0.10
WSP_399	µg/l	Incoming ww	wwtp	350	93	<0.1	<0.1	<0.1
WSP_402	µg/l	Incoming ww	wwtp	120	260	<0.2	31	<0.1
WSP_405	µg/l	Incoming ww	wwtp	90	55	<0.20	<0.10	<0.10
WSP_408	µg/l	Incoming ww	wwtp	220	1500	<1.0	5,2	<0.50
WSP_467	µg/l	Incoming ww	wwtp	28	<5.0	<2.0	<1.0	<1.0
C_313	µg/l	Incoming ww	wwtp	150	370	<2.0	<1.0	<1.0
W_04	µg/l	Effluent	wwtp	140	0,45	12	<0.10	<0.10
W_07	µg/l	Effluent	wwtp	49	26	27	<0.10	<0.10
F_31	µg/l	Effluent	wwtp	28	5,2	15	<0.10	<0.10
F_33	µg/l	Effluent	wwtp	37	3,2	14	<0.10	<0.10
F_35	µg/l	Effluent	wwtp	14	24	<0.20	<0.10	<0.10
F_37	µg/l	Effluent	wwtp	20	1,3	<0.20	<0.10	<0.10
F_55	µg/l	Effluent	wwtp	35	15	1,8	<0.10	0,42
I_64	µg/l	Effluent	wwtp	49	1,8	2	<0,1	,22
G_92	µg/l	Effluent	wwtp	53	69	8,7	3,9	<0.10
G_96	µg/l	Effluent	wwtp	120	40	<0.20	<0.10	<0.10
G_100	µg/l	Effluent	wwtp	87	31	2,8	0,83	0,35
G_104	µg/l	Effluent	wwtp	15	20	<1.0	<0.25	<0.25
G_108	µg/l	Effluent	wwtp	150	6,6	<0.20	<0.10	<0.10
G_111	µg/l	Effluent	wwtp	21	150	4,4	<0.50	0,76
G_114	µg/l	Effluent	wwtp	120	66	6,1	<0.10	<0.10
G_118	µg/l	Effluent	wwtp	67	110	<0.20	<0.10	<0.10
T_122	µg/l	Effluent	wwtp	11	0,69	0,45	<0.1	<0.1

Sample no	Unit	Media	Category	EDTA	NTA	DTPA	1,3-PDTA	ADA
T_125	µg/l	Effluent	wwtp	160	0,41	5,5	<0.1	<0.1
T_128	µg/l	Effluent	wwtp	200	1,9	17	<0.10	<0.10
T_131	µg/l	Effluent	wwtp	21	41	0,78	<0.10	<0.10
D_151	µg/l	Effluent	wwtp	10	5,1	0,13	<0.050	0,067
D_157	µg/l	Effluent	wwtp	320	21	14	<0.10	<0.10
D_162	µg/l	Effluent	wwtp	91	46	<0.20	<0.10	<0.10
Y_181	µg/l	Effluent	wwtp	39	84	8,7	<0.10	<0.10
Y_182	µg/l	Effluent	wwtp	44	28	24	6	<0.10
Y_183	µg/l	Effluent	wwtp	16	190	0,51	<0.10	<0.10
Y_184	µg/l	Effluent	wwtp	0,44	0,36	3,1	<0.050	<0.050
BD_211	µg/l	Effluent	wwtp	23	35	2,2	<0.10	0,17
BD_213	µg/l	Effluent	wwtp	52	340	12	<0.10	<0.10
BD_215	µg/l	Effluent	wwtp	24	43	<0.20	<0.10	<0.10
M_272	µg/l	Effluent	wwtp	200	4,6	34	<0.10	0,14
C_303	µg/l	Effluent	wwtp	220	4,4	1,2	<0.10	0,28
C_310	µg/l	Effluent	wwtp	31	31	43	<0.10	<0.10
S_333	µg/l	Effluent	wwtp	120	59	46	<0.10	<0.10
S_337	µg/l	Effluent	wwtp	7,9	16	<1.0	<0.25	<0.25
S_341	µg/l	Effluent	wwtp	22	6,4	2,6	<0.10	0,15
S_345	µg/l	Effluent	wwtp	15	31	3,8	<0.10	<0.10
E_361	µg/l	Effluent	wwtp	26	58	1,7	<0.1	<0.1
E_363	µg/l	Effluent	wwtp	330	8	19	<0.10	<0.10
E_365	µg/l	Effluent	wwtp	140	6,8	<1.0	<0.50	<0.50
E_367	µg/l	Effluent	wwtp	54	31	12	<0.10	<0.10
E_369	µg/l	Effluent	wwtp	800	82	<0.20	<0.10	<0.10
E_371	µg/l	Effluent	wwtp	94	18	13	<0.10	<0.10
E_373	µg/l	Effluent	wwtp	290	46	5,3	<0.10	<0.10
WSP_400	µg/l	Effluent	wwtp	120	14	0,41	<0.1	<0.1
WSP_403	µg/l	Effluent	wwtp	57	0,5	0,78	3,2	<0.1
WSP_406	µg/l	Effluent	wwtp	110	7,7	12	<0.10	<0.10
WSP_409	µg/l	Effluent	wwtp	210	220	2,1	2,3	<0.10
WSP_468	µg/l	Effluent	wwtp	52	<40	<10	<5.0	<5.0
F_495	µg/l	Effluent	wwtp	9,4	63	<0.20	<0.10	<0.10
F_496	µg/l	Effluent	wwtp	34	9,4	1,1	<0.10	<0.10
F_497	µg/l	Effluent	wwtp	9,7	11	<0.20	<0.10	<0.10
F_498	µg/l	Effluent	wwtp	27	220	0,32	<0.10	0,17
F_499	µg/l	Effluent	wwtp	44	3,3	7,7	<0.10	<0.10
C_314	µg/l	Effluent?	wwtp	280	24	1,3	<0.20	<0.20