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Screening programme 2015

Benzothiazoles, siloxanes, pigments & PBT compounds

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Screening Programme 2015: Benzothiazoles, siloxanes, pigments & PBT compounds
Screening program 2015: Benzotiazoler, siloksaner, pigmenter og PBT forbindelser

Summary - sammendrag

This report summarizes the findings of a screening study into the occurrence of selected benzothiazoles, siloxanes, pigments & PBT compounds in effluents, sewage sludge, surface water, sediments and biota.

Denne rapporten oppsummerer resultatene av en screeningundersøkelse i forekomsten av utvalgte benzotiazoler, siloksaner, pigmenter og PBT forbindelser i avløp, slam, overflatevann, sedimenter og biota.

4 emneord

Screening, Miljøgifter, Oslofjord, Mjøsa

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Summary

Within the 2015 screening programme the occurrence of benzothiazoles (8 compounds), siloxanes (4 compounds), pigments (10 substances), and five selected PBT compounds were measured in effluents, sewage sludge, surface water, sediments and biota in both Oslofjord and Lake Mjøsa. Several of the compounds were found in effluent, sludge, leachate and environmental samples following the targeted analysis of these compounds.

For the compound class of benzothiazoles only 2-benzothiazolol (HBT) was detected in the effluent from the Gjøvik WWTP at relatively high levels (1.3-2.5 µg/L). Additional compounds 2-(1,3-benzotiazol-2-yltio) succinsyre (BTSA), N-cyclohexyl-2-benzotiazolsulfenamid (CBTS) and 2-(Tiocyanato metyltio) benzotiazol (TCMBT) were found at low concentration in the surface waters of Lake Mjøsa. These levels pose little or no environmental risk, but care should be taken in cases where the PNEC values are close-to the lower limit of detection (LoD). However, the unexpectedly high level of HBT in the effluent from Gjøvik WWTP during the 5-day sampling period is reason for concern.

Pigments (RED-112, RED-14, RED-146 and Orange-13) were found in the majority of the WWTP samples, the sediment samples from Lake Mjøsa and the leachate from both landfill sites at concentrations above the LoD. No PNEC values were available for the compounds found in the screening samples. Pigments do not seem to bio-accumulate and were not found in any of the biological samples.

From the selected PBT compounds, only the antioxidant 4,4'-methylene bis[2,6-bis (1,1-dimethylethyl)-phenol (MB1) was found in effluent at both WWTP, and at low concentrations (just above the LoD) in the leachate samples of the Lindum landfill. This compound was also found in one of the sludge samples from the Gjøvik WTTP. The anti-oxidant was found in all biota samples at levels well above the LoD. Although MB1 does not seem to bio-accumulate, the occurrence of this compound in biological samples is of concern.

Regarding the siloxanes, decamethylcyclo pentasiloxane (D5) was found in all samples, and was present at particularly high concentrations in the sludge samples from both the HIAS and the Gjøvik WTTP. Tris (trimethyl siloxy)phenylsilane (M3T) was found in the sludge samples and in several of the sediment samples from lake Mjøsa. Levels of D5 were well below the PNEC value for sediment, but both D5 and M3T were found in biological samples and indicate bio-accumulation which is of concern. The fluorinated siloxanes 2,4,6-trimethyl-2,4,6-tris(3,3,3-trifluoropropyl)-cyclotrisiloxane (D4F) and 2,4,6,8-tetramethyl- 2,4,6,8-tetrakis(3,3,3-trifluoropropyl)-cyclotetrasiloxane (D3F) were found in the effluent (D3F) and leachate samples (D3F and D4F). D4F was also found in all sludge samples from the Gjøvik WTTP. All levels of the fluorinated siloxanes were low, but confirm their presence in effluent from WTTPs and leachate from landfills.

Sammendrag

Screeningprogrammet for 2015 målte forekomster av benzotiazoler, siloksaner, pigmenter og PBT stoffer i avløpsvann, kloakkslam, overflatevann, sedimenter og biota i Oslofjorden og Mjøsa. Basert på resultater fra tidligere screeningstudier ble 10 pigmenter, 5 PBT forbindelser, 8 benzotiazoler og 4 siloksaner og fluorerte siloksaner valgt ut for analyse. Flere av de aktuelle stofferene ble funnet i avløpsvann, slam, sigevann eller miljøprøver.

For benzotiazoler ble relativt høye nivåer (1.3 til 2.5 µg / L) av 2-benzotiazolol (HBT) påvist i avløpsvann fra Gjøvik renseanlegg. I tillegg ble 2-(1,3-benzotiazol-2-yltio) succinsyre (BTSA), N-cyclohexyl-2-benzotiazolsulfenamid (CBTS) og 2-(Tiocyanato metyltio) benzotiazol (TCMBT) funnet i lave konsentrasjoner i overflatevann fra Mjøsa. Disse nivåene medfører ingen eller lav miljørisiko. Likevel bør forsiktighet utvises i tilfeller der PNEC-verdiene er like under deteksjonsgrensen. De relativt høye nivåene av HBT i avløpsvannet fra Gjøvik renseanlegg i prøveperioden er bekymringsverdig.

Pigmenter (RED-112, RED-14, RED-146 og Orange-13) ble funnet med konsentrasjoner over deteksjonsgrensen i de fleste prøvene fra renseanleggene, sediment prøvene fra Mjøsa samt sigevann fra begge deponiene. Ingen PNEC-verdier var tilgjengelige for pigmentene som ble funnet. De undersøkte pigmentene ble ikke funnet i noen av biotaprøvene og synes ikke å bioakkumulere.

Av de utvalgte PBT-forbindelsene ble anti-oksidanten 4,4'-metylenbis [2,6-bis (1,1-dimetyletyl) -fenol (MB1) funnet i avløpsvann fra begge renseanleggene, samt i lave konsentrasjoner i sigevannprøvene fra Lindum deponi. I tillegg ble denne forbindelsen funnet i en av slamprøvene fra Gjøvik renseanlegg. Antioksidanten MB1 ble også funnet i alle biotaprøvene ved nivåer godt over deteksjonsgrensen. Selv om MB1 ikke ser ut til å bioakkumulere, er forekomsten av denne forbindelsen i biologiske prøver alarmerende.

Av siloksaner ble dekametylsyklopentasiloksan (D5) funnet i alle prøver og i relativt høye konsentrasjoner i slamprøvene fra både HIAS og Gjøvik renseanlegg. Tris(trimetylsiloksy) fenylsilan (M3T) ble funnet i slam og i flere av sedimentprøvene fra Mjøsa. Nivåene av D5 var godt under PNEC-verdiene for sedimenter. Begge forbindelsene bioakkumulerer i de biologiske prøvene (dyreplankton, lagesild, smelt og ørret), noe som er bekymringsfullt. De fluorerte siloksanene 2,4,6-trimetyl-2,4,6-tris (3,3,3-trifluorpropyl) –syklotrisiloksan (D4F) og 2,4,6,8-tetrametyl 2,4,6,8 N'-tetrakis (3,3,3-trifluorpropyl) -syklotetrasiloksan (D3F) ble funnet i avløpsvann (D3F) og av sigevannprøvene (D3F og D4F). I tillegg ble D4F funnet i alle slamprøvene fra Gjøvik renseanlegg. Alle nivåer av fluorerte siloksaner var lave, men var likevel tilstede i avløpsvann fra renseanlegg og sigevann fra deponier.

1. Background and Introduction

1.1 General

The Norwegian Environment Agency in 2015 selected several groups of compounds for target analysis for inclusion in Part 2 of its annual screening programme. These were 10 pigments and 5 selected PBT compounds, 8 benzothiazoles, and 4 selected siloxanes and fluoro-siloxanes. The objective of the project was to establish the occurrence of these chemicals in the Norwegian marine and freshwater environments, with particular focus on their potential sources. The data on the occurrence of new potential harmful chemicals in the Norwegian environment presented in the report will contribute to future national or international legislation on an EU (REACH) or global level (UNEP).

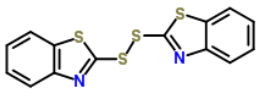
1.2 Compounds of Interest

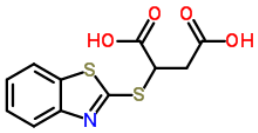
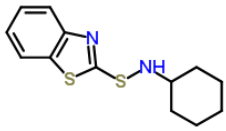
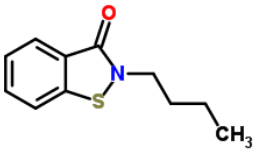
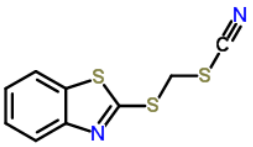
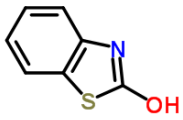
Benzothiazoles

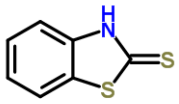
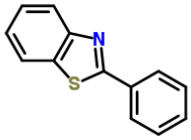
The different derivatives of benzothiazoles are widely used in many industrial applications as accelerators, stabilizers, and also as biocides and pharmaceuticals. Due to their intrinsic chemical reactivity, they are not assumed to be very persistent and bio accumulative but are soluble or slightly soluble in water.

Most data in the international literature has been published on 2-Mercapto benzothiazoles (MBT) and Di(benzothiazol-2-yl)disulphide (MBTS). Both compounds are photodegradable (Maloukia et al., 2004) and although the degradation of the compounds during biological wastewater treatment has been observed (Kloepfer et al., 2005) no complete removal in WWTPs benzothiazoles has been described. Benzothiazoles can potentially bind to sediments although very little information has been published. The individual benzothiazoles included in the study are listed in Table 1.

Table 1: Benzothiazole
Name, Acronym, CAS and Log K_{OW}

Compound	Acronym	Structure	CAS	Function	Log K_{OW}[1]
2,2'-Dithiobis (benzothiazole)	MBTS		120-78-5	Rubber accelerator, stabilizer	4.55-7.04

2-(1,3-benzothiazol-2-ylthio)succinic acid	BTSA		95154-01-1	Corrosion inhibitor	3.10^2
N-Cyclohexyl-2-benzothiazole sulfenamide	CBTS		95-33-0	Rubber accelerator	2.85-3.47
2-Butyl-1,2-benzisothiazolin-3-one	BBTO		4299-07-4	Biocide	2.32-2.76
2-(Thiocyanato methylthio) benzothiazole	TCMTB		21564-17-0	Biocide	2.64-3.30
2-Benzothiazolol	HBT		934-34-9	Biocide	1.76-2.44

2-Mercapto benzothiazole	MBT		149-30-4	Biocide	2.49-2.86
2-Phenyl benzothiazole	PBT		883-93-2	Biocide	3.49-4.26

[1] US-EPA (www.comptox.epa.gov/dashboard)

[2] Chemspider predicted (www.chemspider.com)

Suspected PBT compounds

The group of suspected PBT and other compounds are very diverse and consists of brominated flame retardants (BFR), a PCB replacement, an antioxidant, and a chlorinated chemical intermediate. Both selected BFRs have a high Kow and are expected to be persistent and bioaccumulate in the environment. N,N'-ethylenebis(3,4,5,6-tetrabromophthalimide) (EBTPI), however, has only been reported in one single environmental sample (Nyholm et al. 2013). Decabromodiphenyl ethane (DBDPE) is uses as a replacement for BDEs especially Deca BDE and has been found in both indoors as well as in waste water, sludge and sediment (Kierkegaard el al. 2004). Bioaccumulation of DBPDE has been debated due to the large size of the molecule but DBPDE has been found in Fulmar eggs from the Atlantic (Karlsson et al. 2006). Both BFRs susceptible to photo debromination bot are otherwise relatively stable in the environment.

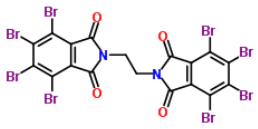
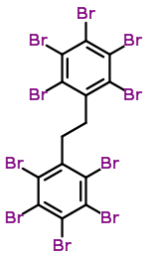
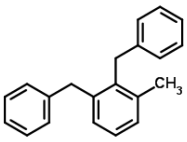
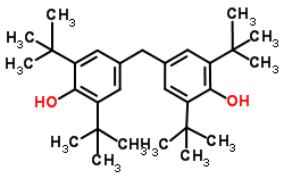
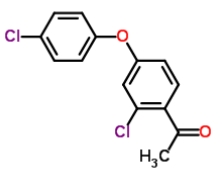
Dibenzotoluene (DBT) is used as a dielectric or heat transfer fluid, it usage and properties are very similar to PCBs. DBT has a very low water solubility and due to its high Kow it is expected to bio accumulate with a strong affinity for both lipids and sediments. DBT is not readily biodegradable, although it is difficult to conclude on persistency due to conflicting results.

4,4'-methylenebis[2,6-bis(1,1-dimethylethyl)-phenol (MB1) is used as an industrial anti-oxidant and additive to plastics. MB1 has shown a large to very large bioaccumulation potential in fish studies (Blankinship et al. 2009) due to its high K_{ow}. Primary degradation of MB1 is rapid in abiotic systems at low concentrations, but several more unidentified stable degradation products were formed. MB1 is expected to bind to sediment and suspended matter based on estimated partition coefficients.

1-[2-chloro-4-(4-chlorophenoxy)phenyl]ethanone (CCPPE) is listed as an intermediate in chemical production. Several patents are listed showing CCPPEs capacity as a fungicide. CCPPE was not found to biodegradable.

Table 2: PBT compounds

Name, Acronym, CAS and Log K_{OW}

Compound	Acronym	Structure	CAS	Function	Log K_{OW}
N,N'-Ethylenebis (3,4,5,6-Tetra bromophthalimide)	EBTPI		32588-76-4	Flame retardant	8.44
Decabromodiphenyl ethane	DBDPE		84852-53-9	Flame retardant	11.1
Dibenzyltoluene	DBT		26898-17-9, 29589-57-9	Heat transfer fluid	6.49
4,4'-methylenebis[2,6-bis (1,1-dimethylethyl) -phenol	MB1		118-82-1	Industrial anti oxidant	8.99
1-[2-chloro-4-(4-chlorophenoxy) phenyl]ethanone	CCPPE		119851-28-4	Chemical inter mediate, Fungicide	4.63

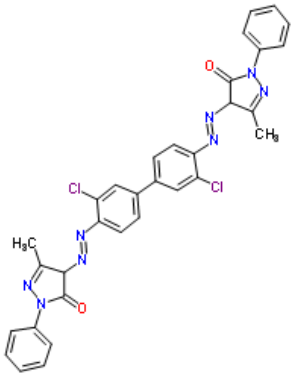
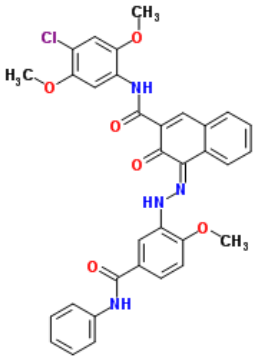
Pigments

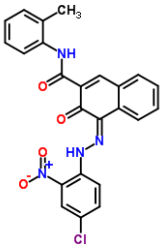
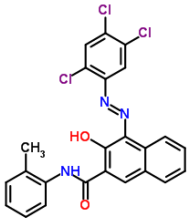
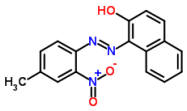
Pigments are an important class of colorants that are used in ink, painting, plastics, food, cosmetics, and other materials. An important distinction between the two colorants pigments and dyes is that pigments are insoluble in their matrix, which means that they are in suspension, whereas dyes either are liquids or are soluble in their matrix, which means they are in solution. An important consequence of this difference is that pigments are by design difficult to dissolve in organic solvents.

As the normal procedure of all trace analytical methods are based on handling of the compound of interest in organic solutions, pigments can be very difficult or even impossible to analyse with the most sensitive analytical methods available to date. To insure light stability pigments are designed to be relatively stable and persistent. In addition, they are characterized by very high log K_{ow} coefficients, which could indicate a high risk for bioaccumulation. However, their extremely low solubility restricts mobility and thus the risk of bioaccumulation.

It was the intention of this study to screen for more pigments than listed in Table 3. The study started with the development of suitable analytical methods for these pigments. Due to differences in the chemical properties including the solubility of several of the pigments, it was only possible to develop and establish suitable and sensitive methods for 5 Pigment Orange 13, Pigment Red 146, Pigment Red 14, Pigment Red 112 and Pigment Red 3 of the 11 selected pigments.

Table 3: Pigments
Name, Acronym, CAS and Log K_{OW}

Compound	Acronym	Structure	CAS	Function	Log K_{OW}
Pigment Orange 13	O13		3520-72-7	Pigment	4.65
Pigment Red 146	R146		5280-68-2	Pigment	6.97

Pigment Red 14	R14		6471-50-7	Pigment	6.54
Pigment Red 112	R112		6535-46-2	Pigment	8.35
Pigment Red 3	R3		2425-85-6	Pigment	5.24

Siloxanes and Fluorinated Siloxanes

Siloxane use is widespread throughout industry, although their dominant usage has been in the personal care product and cosmetic industry. Much focus has been placed on octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5) and dodecamethylcyclohexasiloxane (D6) due to the high concentrations in cosmetic products (Horii and Kannan, 2008) and findings of elevated concentrations within various environmental media (Kierkegaard and McLachlan, 2010; Sparham et al., 2011; Sparham et al., 2008) and have displayed potential bioaccumulative behavior (Borgå et al., 2012; Kierkegaard et al., 2011; Warner et al., 2010). However, fluorinated siloxanes have also been listed as ingredients in cosmetic products and may also be a source of other fluorinated compounds present within cosmetic products (Yukiko et al., 2013).

A comprehensive screening assessment recently performed by Howard and Muir (Howard and Muir, 2010) has provided an insight into commercial chemicals that may be persistent (P) and bioaccumulative (B). Using several chemical registry lists within Canada and the United States, the US Environmental Protection Agency EPISuite software prioritized over 610 chemicals produced in significant amounts that were meet P and B criteria (Howard and Muir, 2010). Of these chemicals, 2,4,6-trimethyl-2,4,6-tris(3,3,3-trifluoropropyl)-cyclotrisiloxane (TFP-D3) was prioritized as one of the top 10 chemicals that should be further investigated due to its atmospheric persistence, large production volumes (0.45 - 4.5 kilotons) and high log Kow (8.66 or 9.8). 2,4,6,8-tetramethyl-2,4,6,8-tetrakis(3,3,3-trifluoropropyl)-cyclotetrasiloxane (TFP-D4) was also listed as chemicals to be prioritized (Table 5).

Table 4: Siloxanes selected for screening

Name, Acronym, CAS and Log K_{OW}

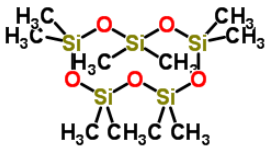
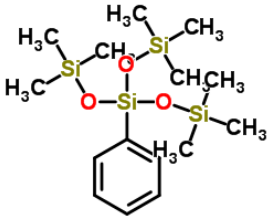
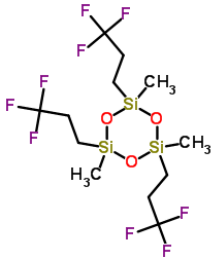
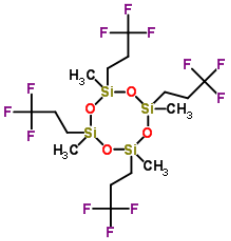
Compound	Acronym	Structure	CAS	Function	Log K_{OW}
Decamethylcyclo pentasiloxane	D5		541-02-6	Industrial, cosmetics	5.7
Tris(trimethylsiloxy) phenylsilane	M3T(Ph)		2116-84-9	Industrial, cosmetics	8.28

Table 5: Fluorinated siloxanes selected for screening

Name, Acronym, CAS and Log K_{OW}

Type	Compound	Acronym	Structure	CAS	Function	Log K_{OW}
Fluorinated siloxanes	2,4,6-trimethyl-2,4,6-tris(3,3,3-trifluoropropyl)-cyclotrisiloxane	TFP D3 (D3F)		2374-14-3	Industrial, cosmetics	9.8
	2,4,6,8-tetramethyl-2,4,6,8-tetrakis(3,3,3-trifluoropropyl)-cyclotetrasiloxane	TFP D4 (D4F)		429-67-4	Industrial, cosmetics	12.4

2. Materials and Methods

2.1 Sampling

Wastewater treatment plants

All of the wastewater treatment works (WWTW) samples were collected by staff at the respective plants. Twenty four hour composite effluent samples were collected by means of the automatic sampling equipment found at the WWTWs for routine monitoring. The effluent samples were collected in clean glass bottles and shipped to NIVA. Sludge samples were collected using a procedure based on the 'Mattilsynet' guideline for the sampling of sludge, compost and other waste-based fertilizer products. Five core samples of mixed sludge were collected from each facility. Each mixed sample was transferred to 4 glass sample jars using pre-washed stainless steel equipment provided by NIVA.

• HIAS owned and receives wastewater from approximatley 52,000 people from the municipalities of Hamar, Løten, Ringsaker, and Stange. The plant is located at Ottestad on Lake Mjøsa with the discharge point at a depth of 15 m around 250 m from the shore. Wastewater is treated mechanically, biologically (not N removal) and chemically. The sludge is treated by thermal hydrolysis (Cambiprocess at 160°C) prior to anaerobic digestion at 38°C.

• Rambekk WWTP in the municipality of Gjøvik and receives wastewater from approximately 17,900 people plus industry (11600 PE). The plant is located on Lake Mjøsa with the discharge point at a depth 7 m. The wasatewater is treated mechanically and chemically. The resulting sludge is treated by mesophilic (34-39°C) anaerobic digestion at a pH of approximately 7, followed by drying.

Landfill Leachate

Leachate sampling was performed using an ISCO 6712 automatic sampler for collecting a 24 hr composite sample from ISI landfill and Lindum Resource and Recycling AS. Flow data were obtained from the plants own water flow measurements.

• ISI landfill (Bærum Kommune) was established in 1974 and ceased being used in 2002. ISI covers an area of approximately 1.4 km^2 with a fill depth of between 12 and 21 m. Groundwater levels in the landfill can be 7.2 m above the base of the landfill. The draining water, composed of leachate and incoming groundwater, flows through a discharge tank downstream of the landfill. Leachate from ISI is sent to VEAS WWTW for treatment.

• Lindum Resource and Recycling is located in Drammen and receives solid waste from the Drammen Region. Leachate from the landfill is heavily influenced by incoming groundwater, especially in the wake of heavy rainfall events. The total annual leachate volume in the period 2000-2006 was at 366,000 to 910,000 m^3. All the leachate goes through an aerated lagoon with subsequent sedimentation before it is pumped to Solumstranda WWTW.

Lake Mjøsa

Surface water

Water samples were collected at five stations with a Ruttner water sampler, at 15 meters depth (26th May 2015). The water samples were taken at the corresponding sediment sample stations. Each water sample was transferred to two l litre PE bottles and stored cold until analysis.

Sediment

Five pooled samples of sediment were taken along a gradient from the discharge point to HIAS and south. Each pooled sample consisted of three individual subsamples taken from the upper 0-2 cm sediment layer at a water depth of 25–35 m. We used a gravity corer with a core tube and a retractable sediment stopper in stainless steel. The samples were transferred to heat-treated (500°C) glass containers sealed with heat-treated aluminium foil underneath the lids. The core tube and other sectioning equipment used were thoroughly cleaned with acetone and cyclohexane (HPLC grade) before use, and direct hand contact with the sampling matrix was avoided. They samples were stored frozen (20°C) until analysis.

Fish

From Lake Mjøsa, during 11. – 30. August 2015, the following species of pelagic fish were collected: brown trout (*Salmo trutta*), smelt (*Osmerus eperlanus*) and vendace (*Coregonus albula*). Smelt and vendace were caught with gillnets, deployed in the area around the outlet of discharge pipe of the HIAS sewage treatment plant, at a depth of about 20 – 35 m, whereas brown trout were caught north of the town of Gjøvik at a depth of 5 –20 m. The smelt were mainly small-bodied planktivorous individuals, but a few larger cannibalistic individuals were also included.

Smelt and vendace were taken out of the nets as they were hauled, instantly killed with a short blow to the head, wrapped in clean aluminium foil, kept cool and transported to a freezer (-20°C). Before freezing the aluminium foil wrapped fish were put in polyethylene bags. Brown trout were transported alive in a water filled container to the shore for biological sampling (Ref: Screening report del 1), after finishing this was the fish were wrapped in aluminium foil and frozen for later dissection of muscle samples for chemical analysis. At no time were the fish allowed to be in contact with plastics or other potentially contaminated surfaces. The time between catch and transfer to the freezer took no longer than 4 hours.

Before preparing muscle samples of the pelagic fish, they were thawed and total length and weight were registered (Table 6). They were then scraped clean of mucus with a solvent washed knife and placed on a cutting board covered with solvent rinsed aluminium foil. For each fish a solvent cleaned set of stainless steel dissection tools was used. We dissected the sagittal otoliths, and determined sex and maturity after opening of the abdomen. We dissected out samples of lateral skeleton muscles and transferred them to heat treated (500°C) glass containers sealed with heat treated aluminium foil underneath the lids. The samples were then frozen (-20°C) and sent to homogenization before analysis. Five pooled samples were prepared for smelt and vendace, respectively, to obtain sufficient material for chemical analysis, whereas 10 individual samples were prepared for brown trout.

To reduce the risk of contamination during catch and sample preparation, all personnel involved avoided use of personal care products at least 24 hours in advance. Also, dissection and preparing of samples took place outside in a non-urban area. Dissection equipment and aluminium foil that could be in direct contact with the samples were cleaned with acetone and cyclohexane (HPLC grade) before use, and direct hand contact with the sampling matrix was avoided.

2.1.4 Table 6. Mean length and weight of the studied fish. Pooled samples were made for smelt and vendace.

			Length (cm)		Weight (cm)	
Species	Sample No.	N	Mean	Std Dev	Mean	Std Dev
Smelt	1	5	21.7	0.6	55.6	9.2
	2	7	17.2	1.3	29.1	7.2
	3	8	15.8	0.2	24.3	1.5
	4	11	14.7	1.0	18.0	4.1
	5	40	11.4	0.6	10.7	1.3
Vendace	1	4	21.4	0.9	75.5	4.7
	2	3	21.7	0.4	74.3	6.5
	3	4	20.8	0.9	73.8	11.3
	4	3	21.4	1.1	75.3	10.5
	5	4	20.7	0.4	69.8	3.6
Brown trout	1:10	10	64.8	13.1	3569	2321

Mysis and zooplankton

Samples of the opossum shrimp *Mysis relicta* and zooplankton were sampled with horizontal net hauls. Epipelagic zooplankton, consisting mainly of the cladocerans *Daphnia galeata* and *Bosmina longispina* together with the copepod *Eudiaptomus gracilis,* were collected at a depth of 3–5 m, whereas Mysis were collected at a depth of 70–110 m (this is a diurnal vertical migrating Mysida, mainly feeding in the epipelagic zone during night-time). The zooplankton net used were made of nylon mesh (single strand thread, mesh size: 500 µm), equipped with a brass cup with a brass mesh, and with an opening diameter of 1 m.

Mysis were separated from copepods in the hypopelagic samples by filtering the samples through a sieve (mesh of stainless steel strands) while flushing gently with water from the lake and handpicking with tweezers. All filtering and separation of samples were done in the boat immediately after net hauling. The samples were kept on the same type of cleaned class jars as the fish, held cool on board until they could be transferred to a freezers (-20°C) no more than 8 hours after sampling. All equipment (glass or metal) and aluminium foil that could be in direct contact with the samples after they were transferred from the net were cleaned with acetone and cyclohexane (HPLC grade) before use, and direct hand contact with the samples was avoided. Five samples each of Mysis and epipelagic zooplankton were prepared.

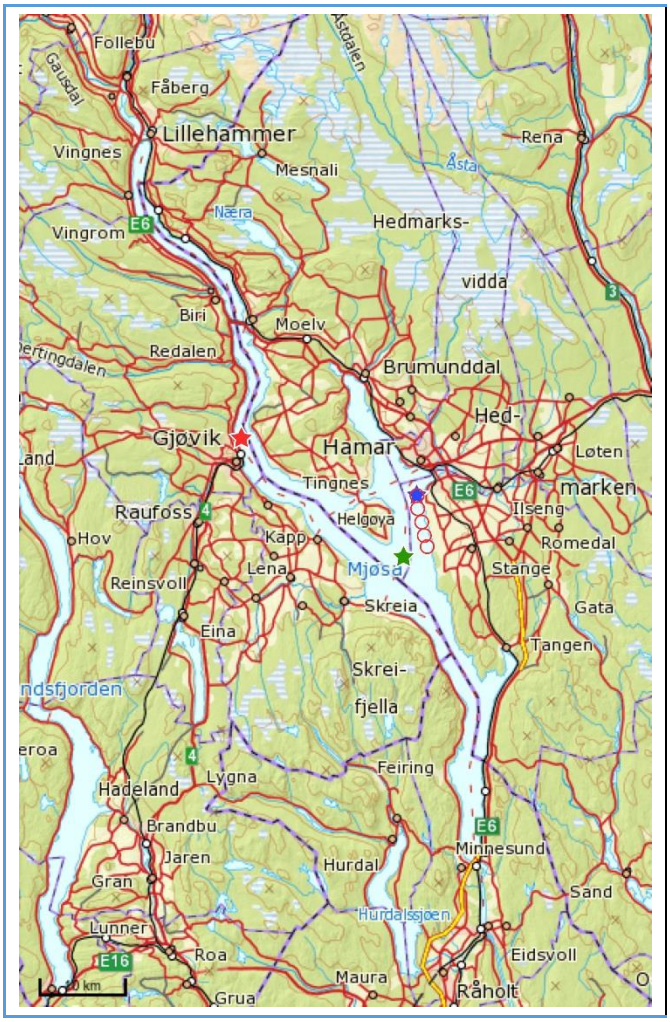


Figure 1. Map showing Lake Mjøsa, the catch sites (blue star: smelt and vendace; red star: brown trout; Mysis and zooplankton: green star) and sediment/water sampling sites (red circles). The location coordinates are given in Table 7.

Table 7. Coordinates for the Lake Mjøsa water, sediment and biota sampling stations

Station	Date	Depth (m)	UTM 33E	UTM 33 N	°E	°N
HIAS			286000	6743100	11.070	60.766
Sediments/water						
St-1	26.05.2015	sed.: 35, water: 15	285400	6743100	11.059	60.766
St-2	26.05.2015	sed.: 25, water: 15	285941	6742150	11.075	60.759
St-3	26.05.2015	sed.: 25, water: 15	285932	6740684	11.072	60.744
St-4	26.05.2015	sed.: 25, water: 15	286479	6739302	11.084	60.732
St-5	26.05.2015	sed.: 25, water: 15	287021	6737370	11.096	60.715
Fish						
St-1	11.08.15	20-35	286400	6743600	11.059	60.766
St. Gjøvik	13.-30.08.15	10-20	265100	6750000	10.680	60.816
Mysis	10.-13.08.14	70-110	284000	6735000	11.04	60.69
Zooplankton	13.09.15	3-5	284000	6735000	11.04	60.69

2.2 Analysis

Benzothiazoles

Extraction
Sulfamethoxypyridazine-d_3 was used as an internal standard for all samples matrices. Sediment and sludge (0.5 g) and biota (1 g) were extracted ultrasonically with methanol (4 ml) for 60 minutes. The extract was decanted and particulates were removed using Spin-X nylon centrifuge filters. Receiving waters and landfill leachate (200 ml) were extracted onto Oasis HLB (500 mg) solid phase extraction cartridges. Benzothiazoles were eluted with methanol/hexane (50/50). Extracts were evaporated under nitrogen and reconstituted in methanol prior to particulate removal using Spin-X nylon centrifuge filters.

LC-MS analysis
Benzothiazoles were separated on an Aquity UPLC (Waters, Manchester) using a BEH C8 column (100 x 2.1 mm, 1.7 µm) (Waters, Sweden) with an acetonitrile and water (5.2 mM ammonium acetate) mobile phase. Gradient elution was from 50% to 100% acetonitrile over a 10 minutes program. The UPLC system was connected to a mass spectrometer (Xevo G2S QToF, (Waters, Manchester)) operated in electrospray ionisation mode.

Detection limits
Detection limits (LoD) and quantification limits (LoQ) were calculated for each sample individually using the standard method of calculation of 3 x s/n ratio and 9 x s/n ratio for LoD and LoQ respectively.

Considerations
It was not possible to use labelled internal standards because these are not commercially available. Very few of the benzothiazoles are available as certified standards, where available they were purchased from Sigma-Aldrich (Germany). Where no standard was available, compounds were purchased directly from producers; however, the quality of these could not be certified. This will result in a somewhat higher uncertainty than normal between 35-50% depending on the analytes and sample matrix.

PBT compounds

Aqueous samples
Water samples (150 ml) were extracted by solid phase extraction using ChromaBond HR-X (500 mg). SPE columns were conditioned with ethyl acetate, acetonitrile and with MilliQ water, the samples were then extracted and analytes eluted with ethyl acetate. Afterwards solvent was exchanged either to toluene or methanol and samples were analyzed with GC-HRMS and LC-APPI-MS.

Solid samples
The solid phase samples were extracted with ethyl acetate by accelerated solvent extraction (ASE). Afterwards samples were further cleaned with solid phase extraction similarly to water samples.

Considerations
It was not possible to use labelled internal standards because these are not commercially available. Very few of the PBTs are available as certified standards, where available they were purchased from Sigma-Aldrich (Germany). Where no standard was available, compounds were purchased directly from producers; however, the quality of these could not be certified. This will result in a somewhat higher uncertainty than normal between 35-50% depending on the analytes and sample matrix.

Pigments

Aqueous samples
Aqueous samples were extracted by solid phase extraction using Strata X (500 mg). SPE columns were conditioned with ACN and water, the sample extracted, and analytes eluted with ethyl acetate. Afterwards solvent was exchanged to 5% dimethyformamide (DMF) in acetonitrile and analyzed with LC-ESI-TOF-MS in negative mode.

Solid samples
Sediments were extracted with ethyl acetate by accelerated solvent extraction (ASE). Afterwards solvent was exchanged to 5% dimethyformamide (DMF) in ACN and samples were filtrated with a nylon filter and analyzed with LC-ESI-TOF-MS in negative mode.

Biota samples
Biota samples were extracted with ethyl acetate by accelerated solvent extraction (ASE). Afterwards solvent was exchanged to 5% dimethyformamide (DMF) in acetonitrile. Fat was removed with hexane by liquid/liquid extraction. Prior to analysis samples were filtrated with a nylon filter and analyzed with LC-ESI-TOF-MS in negative mode.

Considerations
It was not possible to use labelled internal standards because these are not commercially available. None of the pigments is available as certified standard. This will result in a somewhat higher uncertainty than normal between 35-50% depending on the analytes and sample matrix. Of the originally selected 10 pigments only 5 were finally included in the analytical methods 3 pigment were insoluble in more than 10 organic solvents and the remaining 2 were not extracted from the samples.

Siloxanes

Extraction
Established methods based on liquid/liquid extraction (Warner et al. 2010; Warner et al. 2013) were used to extract and quantify siloxanes, in addition to headspace extraction techniques (Sparham et al. 2008) for analysing siloxanes in water and sediment samples.

Analysis
Analysis of siloxanes (D4, D5 and D6) was performed using gas chromatography with mass spectrometric detection (GC-MS).

Limits of Detection
The limit of detection (LoD) and limit of quantification (LoQ) were used to evaluate the detection of analytes. The method used to calculate the MDL has been previously reported (Warner et al. 2013). LoQ was calculated as nine times the signal/noise ratio of the GC-MS instrument.

Quality assurance
The greatest risk in the analysis is background contamination, as these chemicals (D4, D5 and D6) are applied in e.g. skin care products. NILU has previously participated in a laboratory intercalibration of siloxanes and has also worked closely with the siloxane industry. Samples were analysed in batches with at least one additive standard sample and a blank control. The data from these were used to calculate the uncertainty for each sample batch. To ensure repeatability, a random sample from each matrix was selected for duplicate analysis.

Field blanks were prepared for the sampling by packing 2 or 3 grams of XAD resin in filter bags of polypropylene/cellulose, which were then cleaned by ultrasonic treatment in hexane for 30 min. Subsequently, used hexane was removed and substituted with clean hexane and the field blanks were sonicated once more for 30 min. After ultrasonic treatment, the field blanks were dried in a clean cabinet equipped with HEPA- and a charcoal filter to prevent contamination from indoor air. After drying, the field blanks were put in sealed polypropylene containers and sent for sampling purposes. Several field-blanks were stored at NILU's laboratories and analysed to determine reference concentrations before sampling. The field blanks sent for sampling purposes were exposed and handled in the field during sampling and during preparation of samples. Reference blanks are the same as field blanks (XAD resin in filter bags of polypropylene/cellulose), but stored in cabinet at the NILU laboratory with no exposure in the field or during preparation of samples.

Fluorinated Siloxanes

There is currently no validated analytical method for these substances, thus the analysis was done according to the best state-of-the-art method. The analytical procedure included solvent extraction, purge and trap cleanup and UPLC-MS-MS analysis. Matrix effects and adjustment for recovery were achieved by analyzing spiked samples in parallel to unspiked (standard addition). With the exception of the mysis samples, two spiked samples were analyzed for each matrix.

Effluent water and leachate

The water samples were kept without headspace in fully filled 250 mL bottles (300 mL) at 8 °C until the time of analysis. The water was extracted by adding 20 mL of dichloromethane (DCM Merck, lichrosolve) into the bottle through a glass funnel. The water displaced from the bottle was discarded. The water and DCM were vigorously mixed over night with a magnetic stirrer. The phases were allowed to separate in and the DCM was collected after centrifugation. The water was reextracted with 5-10 mL of DCM. DCM extracts were combined in a solvent rinsed 250 mL Erlenmeyer flask with a magnet stir bar for purge & trap cleanup, described below.

Sewage sludge and sediment

The sediment was initially centrifuged in 50 mL PP tubes at 4000 rpm for 20 min. The water was discarded and 20-25 g of the sediment was transferred to a glass centrifuge tube. Sub-samples were taken from the sludge and from the centrifuged sediment in pre-weighed crucibles for dry weight determination (105 °C for >72 hrs).

Dewatered sludge (approximately 12 g) and sediment (20-25 g) were weighed into a 50 mL centrifuge glass tube together with 10 mL of acetone and 6 mL of DCM. The sludge samples were analyzed in 2 batches with a random selection of samples from both STP sites. The samples were rotated overnight c the following day. The organic phase was transferred to new glass tubes and the samples were re-extracted with 20 mL of DCM. After approximately 4 hrs the samples were ultra-sonicated for 5 minutes, centrifuged and the DCM extracts decanted and combined. The combined organic phases were cleaned up with the purge and trap method.

Biota samples

Fish muscle: The fish muscle was thawed and cut into small pieces using a solvent rinsed pair of scissors. 12-14 g was weighed into a 50 mL glass centrifuge tube. After the addition of 6 mL of acetone and 20 mL of DCM the tissue was homogenized with an Ultra Turrax homogenizer. The tube was centrifuged at 2600 rpm for 15 mins and the organic phase transferred to a new

tube. The tissue was re-extracted with 15 mL of DCM and ultra-sonicated for 5 minutes. The organic phases were combined, centrifuged and transferred to a 250 mL flask for purge and trap cleanup.

Zooplanktion (Mysis): The whole sample was transferred to a centrifuge tube and was centrifuged for 17 min at 2600 rpm. The water supernatant was transferred to a new tube and extracted with 10 mL DCM by vortexing. The mysis pellet was extracted with 25 mL DCM and 7 mL acetone using ultra sonication for 7 minutes. After centrifugation the DCM phases were combined in a new centrifuge tube. The mysis was re-extracted with 15 mL DCM, ultra-sonicated and centrifuged. The combined organic phase was cleaned up with the purge & trap method.

Purge and trap cleanup
All extracts were cleaned up by a purge and trap method described in Borgå *et al*. (2013). The method was slightly modified to minimize degradation of the analytes. The ENV+ sorbent was replaced by another polymer-based resin, ABN (Biotage, Uppsala, Sweden, 10-15 mg in 1 mL cartridge) for all matrices except the water where the original ENV+ was used. The cartridges were rinsed with 3 mL of DCM and 3 mL of acetonitrile (ACN) before application. The extracts were evaporated over night with a continuous flow of filtered nitrogen that via the headspace of the flask passed through the cartridge. In the morning when the extract was completely evaporated, the heating element of the stirrer was set to maximum, generating ~70-80 °C in the flasks. The purging continued for another 8 hours. The ABN cartridges were eluted with ACN in two fractions of 400 uL (1st) and 600 uL (2nd).

UHPLC-MS-MS analysis
Multiple reaction monitoring (MRM) was applied in ES negative mode on a XEVO UPLC/MS/MS (Waters, Sweden). Five MRM transitions of the quasi-molecular ions $[M+OH]^-$ were recorded for each of the substances. The column was a 2.1x 50 mm Acquity UPLC BEH C18, 1.7 um (Waters, Sweden), held at 50 °C. The hydrolysis of the analytes in the column was minimized by reducing the length of the ACN/water gradient and increasing its steepness. Hydrolysis is however required in the ion source in order to ionize the analytes, thus to enable ionization of D4F that elutes when the gradient is at 100% ACN, water was introduced into the ion source post column by a built in syringe pump via a mixing valve. ACN/H_2O (1:1) was added with a flow rate of 100 uL min^{-1}. The solvent gradient started with 50 % ACN/H_2O (A) and 50 % ACN (B) at 0.5 mL min^{-1} followed by a linear increase up to 100% B after 1.2 min. Both of the fractions collected after the cleanup were analyzed on UPLC/MS-MS but for the majority of the samples only fraction 1 contained DF. Injection volume was 5 uL.

External standards and quantification
The original spiking solution of D3F and D4F in ACN was freshly made from the pure chemicals purchased from Fluorochem Ltd (Derbyshire, UK), in 2012. When the first samples were analyzed, the new spiking solution D3F turned out to be partly degraded. After making a new standard it was concluded that the degradation reactions had occurred in the original jar. The chemical was at the time of this study no longer sold by Fluorochem Ltd and was therefore purchased from AK Scientific, Inc. (CA, USA). However, the D3F from AK Sci. did not fully correspond to the D3F originating from Fluorochem Ltd. The new D3F gave a broad split peak of equal intensity in the UPLC-MS-MS analysis, most likely reflecting the 2 possible stereoisomers of D3F, whereas the Fluorochem D3F did not. The response factor for D3F was calculated from the summed area of the 2 isomers and the hydrolyzed product.

The spiking of the samples (generally 5 ng of each DF in 25 uL ACN) was performed after the addition of the extraction solvent. With the exception of zooplankton a minimum of two spiked samples for each matrix or species was analyzed. No blank correction was made.

An estimation of the absolute recovery of D4F calculated versus a matrix free external standard showed acceptable recoveries for all matrices, 49-88%, except for sludge 34% (Table 1). For D3F the sediment had a recovery of 80% whereas for the other matrices it was 1-8%.

The standard addition to the water samples was performed using the initial degraded D3F solution. The amount of D3F remaining in the degraded spiking solution used for the water samples was estimated against external standards from other extraction rounds, and the response factors were adjusted accordingly. The concentrations of D3F in the water samples should therefore be regarded as semi-quantitative.

QA/QC

Extraction and clean up were carried out in a clean air cabinet under a laminar flow of charcoal-filtered air. The glassware used in the analysis was rinsed with acetone before use. With the exception of the water samples, procedural blanks were analyzed with every extraction round. Between 50-80 mg of corn oil was added to the solvent blank as a matrix surrogate/keeper.

Field blanks were collected for HIAS and Rambekk effluent. The leachate water samples lacked field blanks, so tap water was used as a surrogate blank sample. Control samples containing background levels of the analytes were analyzed to set the limit of quantification (LOQ). No such material was available from the present sampling campaign. The control samples used were lake sediment and Brown trout (*Salmo trutta*) from other studies. The control sediment was analyzed together with sediment from Lake Mjøsa and sludge from Rambekk and HIAS. The trout samples, 4 individuals 2-3 replicates of each were analyzed in parallel with the biota samples.

The LOQ, defined as the mean amount plus 10 times its standard deviation, was similar for D4F in sediment and trout (0.26 and 0.33 ng, respectively, Table 2). The LOQ for the water samples, based on 2 field blanks and one tap water blank, was somewhat higher, 0.59 ng. Only the sediment control samples had detectable D3F concentrations so that a corresponding LOQ could be derived. The D3F LOQs for the other matrices were calculated individually from the estimated area of an imaginary peak distinguished from the noise in each sample. For water the calculated amounts were subsequently multiplied with a factor of 2 to take account of the additional uncertainty in this matrix. For samples with concentrations over the LOQ, the identity of the peak was checked using the relative ratios of the intensities of the 5 transition products.

The precision (CV) of the repeated analysis of D4F in the control samples was 15% for the sediment (n=4) and 78% for the trout samples (n=10). Excluding one outlier trout the CV was reduced to 27% (n=9). The larger variation of the biota samples may partly be explained by differences between individuals (the trout control matrix consisted of intact tissue, not homogenate). None of the control materials contained any DF3.

Table 8. Recovery of standard additon samples			
Matrix	n	D4F Recovery (%)	D3F+D3OH Recovery (%)
Effluent	2	84	Not determined
Sludge	4	34	8
Sediment	2	86	80
Vendace	2	49	7
Trout	2	65	5
Smelt	2	73	1

Table 9. Recovery of standard additon samples			
Matrix	n	D4F (ng)	D3F+D3OH (ng)
Water	3	0.59	Estim. Area*2[b]
Sediment/sludge	4	0.26	0.67[a]
Biota	10	0.33	Estim. Area*1[b]

*a mean of n control samples + 10*SD*

*b estimated peak area of minimal quantifiable peak*1 or2*

Supporting Parameter analysis

Particle Size Analysis

Wet sediment was shaken by mechanical fractionation with < 63 µm sieves. Dry weight measurements were used for the particle size calculations.

Sediment TOC

Freeze dried sediment sample aliquots (0.5-10 mg) were heated in a furnace at 1,800 ºC in the presence of oxygen free helium. The carbon dioxide gas produced was passed through a chromatography column and the total organic carbon was measured.

Water DOC

Samples (4 ml) were injected into an inorganic carbon chamber and 0.5 ml 21% phosphoric acid was added. The inorganic bound carbon from carbonates, bicarbonates and dissolved CO_2 is released to an NDIR detector for CO_2 quantification.

Lipid content

Lipids were calculated gravimetrically after solvent extraction; this was done in duplicate for each sample. A column packed with a sample aliquot (approximately 0.5-1 g liver or 5 g filet) and sodium sulphate was extracted with 100 ml dichloromethane. The solvent extract was evaporated to dryness and the remaining lipids were dried at 110 ºC until constant weight.

δ 13C/δ 13N ratio analysis

Samples were dried at 60 ºC for 24 hours before grinding to fine powder. Approx 1 mg of sample was combusted in the presence of O_2 and Cr_2O_3 at 1700 ºC in a Eurovector element analyser. Reduction of NO_x to N_2 was done in a Cu oven at 650 ºC. H_2O was removed in a chemical trap of $Mg(ClO_4)_2$ before separation of N_2 and CO_2 on a 2 m Porapolt Q GC column. The C/N ratio was quantified on the basis of the m/z 44/28 ratio. N_2 and CO_2 were directly injected online to an isotope ratio mass spectrometer (Nu Instruments Horizon) for the determination of $\delta^{13}C$ and $\delta^{13}N$. The mean stable N-isotope ratios, $\delta^{15}N$, reflects the relative trophic position of the organisms. Likewise, the stable C-isotope ratio, $\delta^{13}C$, reflects the carbon sources of the organism. A low $\delta^{13}C/\delta^{13}N$ ratio indicates influence from a pelagic food chain whereas a higher ratio indicates a more littoral food chain.

3. Results and Discussion

3.1 Wastewater treatment plants

Benzothiazoles

HIAS WWTW
At the HIAS WWTW none of the selected benzothiazole derivatives could be detected in effluent samples above the LoD. None of the measured benzothiazoles could be detected above the LoD in sludge samples from HIAS.

Rambekk WWTW
At Rambekk WWTW the only benzothiazole derivative detected in effluent was HBT with a median concentration of 2.5 µg/L corresponding to a daily load of 19 000 g/day. This is somewhat higher than has been previously reported in effluent from a WWTT in Berlin (0.14-0.50 ug/L) and in the same range as effluent from a WWTT in Bejing (1.54 ug/L) (Koepler et al. 2005) In the sludge samples from Rambekk none of the measured benzothiazoles were detected above LoD.

PBT compounds

HIAS WWTW
At HIAS the only PBT compound detected in effluent was the antioxidant MB1 with a concentration of 120 ng/L corresponding to a daily load of 2 600 g/day. This are similar levels (100 ng/L, max 2000 ng/L) as have been reported for surface water collected from WTTPs in Sweden, but higher than the WTTP effluent where all levels were below the LoD (< 100 ng/L) (Arner et al. 2004) In sludge samples from HIAS none of the measured PBT compounds could be detected above LoD.

Rambekk WWTW
At Rambekk the only PBT compound detected in effluent was the antioxidant MB1 with a concentration of 77 ng/L corresponding to a daily load of 900 g/day. In sludge samples from Rambekk none of the measured PBT compounds could be detected above LoD.

Pigments

HIAS WWTW
At HIAS the only pigment detected in effluent was Red-14 in one single sample at a concentration of 0.7 ng/L just above the LoD. In sludge four pigments were detected with a median sludge concentration 4.2 ng/g dw (Red-112), 13 ng/g dw (Red-14), 6.0 ng/g dw (Red-146), and 191 ng/g dw (Orange-13) as shown in Figure 2.

Rambekk WWTW
Three pigments were detected in effluent samples from Rambekk WWTW with a median concentration of 0.6 ng/L (Red-112), 2.6 ng/L (Red-14), and 1.5. ng/L (Red-146) as shown in Figure 3.

Four pigments were detected in sludge with a median sludge concentration 30 ng/g dw (Red-112), 186 ng/g dw (Red-14), 29 ng/g dw (Red-146), and 50 ng/g dw (Orange-13) as shown in Figure 4.

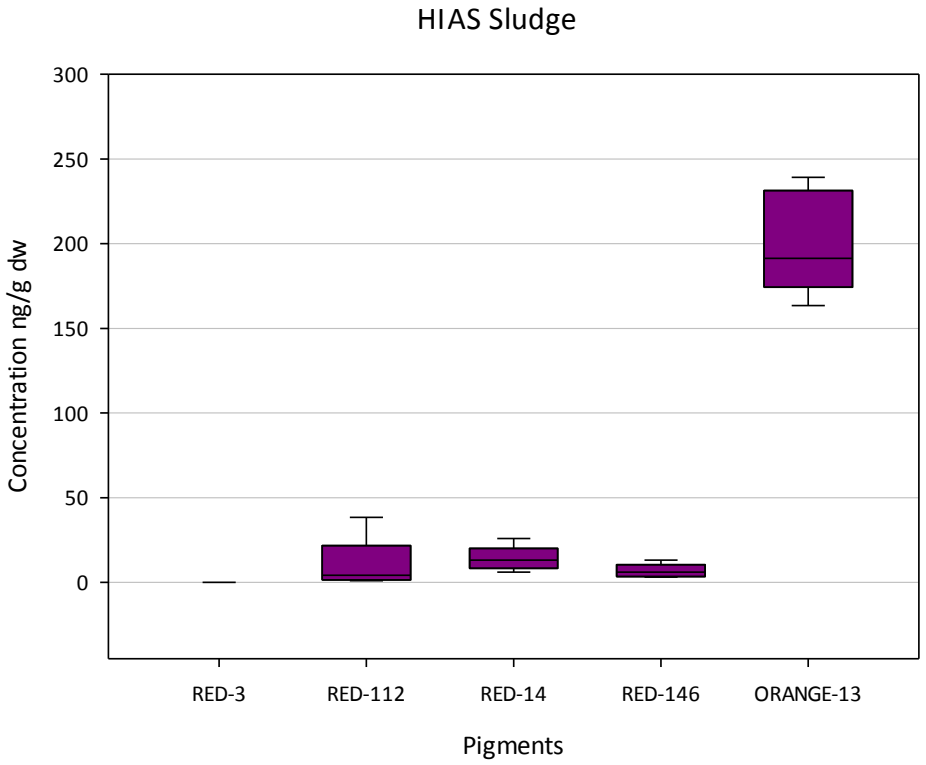


Figure 2. Pigment concentration in five sewage sludge from HIAS WWTW given in ng/g dw.

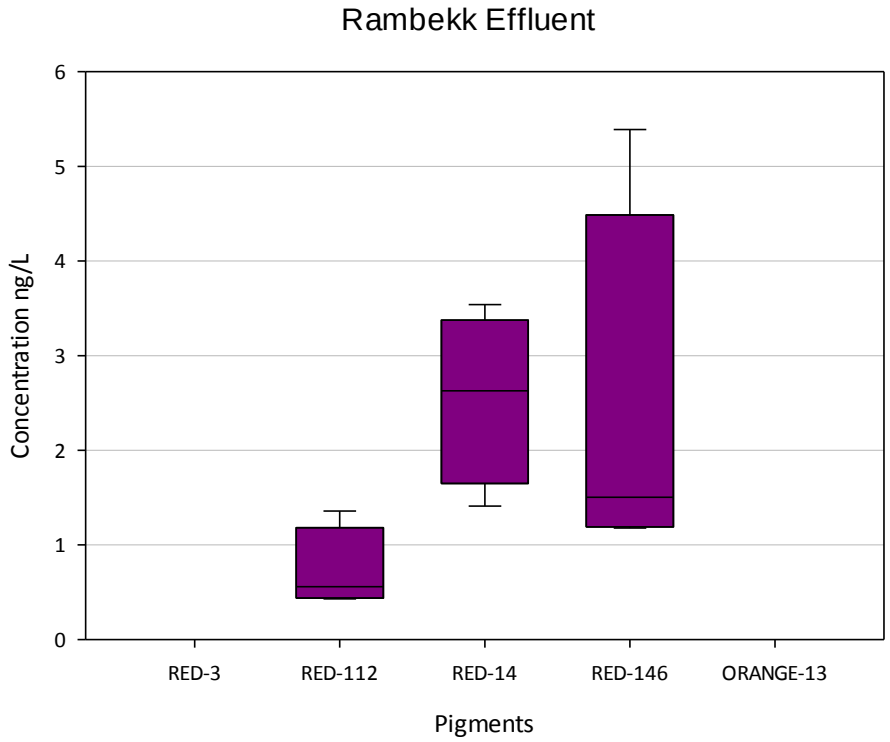


Figure 3. Pigment concentration in five effluent samples from HIAS WWTW given in ng/L.

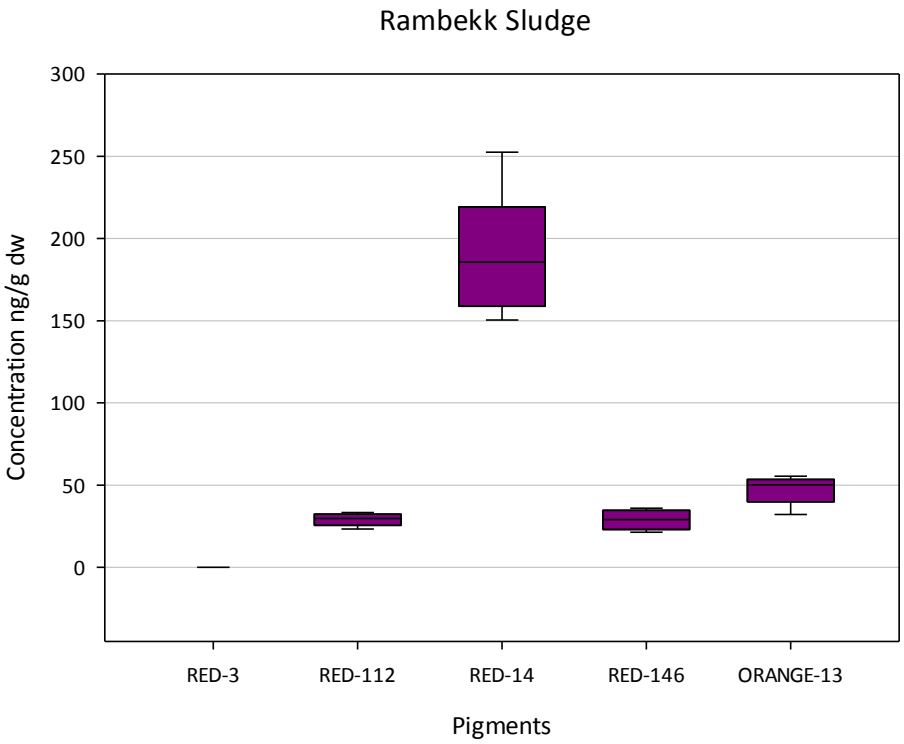


Figure 4: Pigment concentration in five sewage sludge from Rambekk WWTW given in ng/g dw.

On average the pigment concentration in sludge from Rambekk WWTW were slightly higher compared to what was detected in the HIAS samples. In addition there is also a different pigment pattern with Red-14 dominating in Rambekk and Orange-13 dominating in HIAS. As we have no data for the influent it is not possible to decide, if these differences are caused by the influent pattern or be differences in treatment technologies. In addition no comparable studies were found in the international literature on the pigments found in the Norwegian environment.

Siloxanes and fluorinated siloxanes

HIAS WWTW

D5 and D3F were detected in the effluent samples from HIAS at a median concentration of 31 and 0.16 ng/L respectively corresponding to daily loads of 360 and 1.8 g/day. In sludge samples from HIAS D5 and M3T were found with a median concentration of 7.9 and 94 µg/g dw respectively.

Rambekk WWTW

In the effluent samples form Rambekk D5 and D3F were detected at a median concentration of 93 and 0.09 ng/L respectively corresponding to daily loads of 150 and 2.1 g/day. D5 was detected at a median concentration of 5.8 µg/g dw in sludge samples from Rambekk WWTW. M3T and D4F were detected at a much lower median concentration of 57 and 0.18 ng/g, which is lower compared to measurements made in Stockholm with ~500 and 0.6 ng/g dw, respectively (McLachlan et al., 2014).

3.2 Landfill Leachate

Benzothiazoles

None of the measured benzothiazoles could be detected above LoD in the landfill leachate from Lindum and ISI.

PBT compounds

The antioxidant MB1 was detected in three samples with a concentration of 0.01 to 0.11 ng/L in the landfill leachate from Lindum and ISI. DBT was detected at a concentration of 6 ng/L in a single sample from Lindum.

Pigments

It was possible to measure three pigments at a median concentration of 0.5 ng/L (Red-112), 0.8 ng/L (Red-14), and 1.7 ng/L (Red-146) in leachate samples from the ISI landfill (Figure 5).

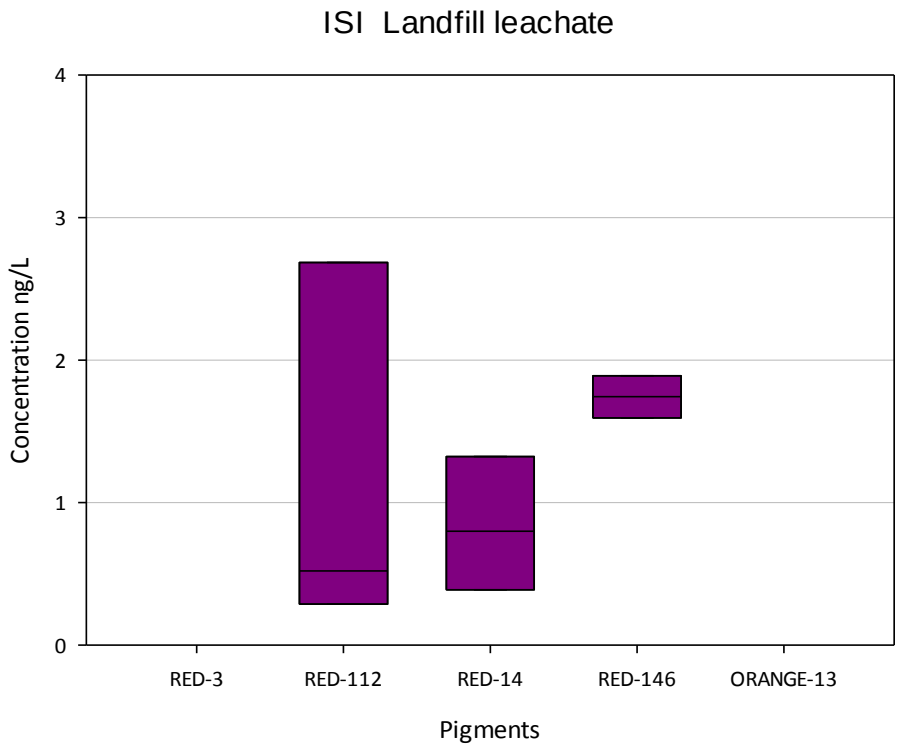


Figure 5. Pigment concentration in three leachate samples from ISI landfill

Siloxanes and fluorinated siloxanes

D5 was detected in all samples at a median concentration of 82 ng/L and D3F in two of three samples at a median concentration of 0.12 ng/L in leachate from the Lindum landfill.

In leachate from the ISI landfill D5 was detected in all samples at a median concentration of 307 ng/L. D4F and D3F were found with a median concentration of 2.8 and 0.27 ng/L, respectively.

3.3 Lake Mjøsa

Benzothiazoles

Benzothiazoles were not detected >LoD in any of the sediment or biota samples from Lake Mjøsa. BTSA was found at a level just above the LOD (25.1 ng/L) in one of the surface water samples (station 4) and CBTS was found in two samples (station 3 and 4) at a concentration of 10.2 and 76.2 ng/L. In addition TCMTB was found at concentration sof 8.8 and 15.4 at the same sampling stations at Lake Mjøsa. The first sampling point is located 600 m to the west of the HIAS WWTP (Table 7) and sampling points 3 and 5 are located at about 2 and 5 km from the WWTP.

PBT compounds

Sediments

None of the PBT compounds were detected in sediments at concentrations >LoD.

Biota

The antioxidant MB1 was detected at concentrations ranging from 30 to 1,500 ng/g ww in all of the biota samples (Figure 6). As shown in figure 6, the concentration ranges are somewhat higher for MB1 in the different organisms compared to a typical bioaccumulating compound, such as PCB-153 (Figure 7), which was also measured in all biota samples. The bio accumulation potential of MB 1 has been reported both in laboratory experiments and as well as calculations based on chemical properties (Arnot et al. 2006, Inoue et al. 2012). The screening data from Lake Mjøsa however does not show strong bioaccumulation or bioconcentration of MB1, but it confirms the environmental occurrence of this antioxidant in the studied foodchain.

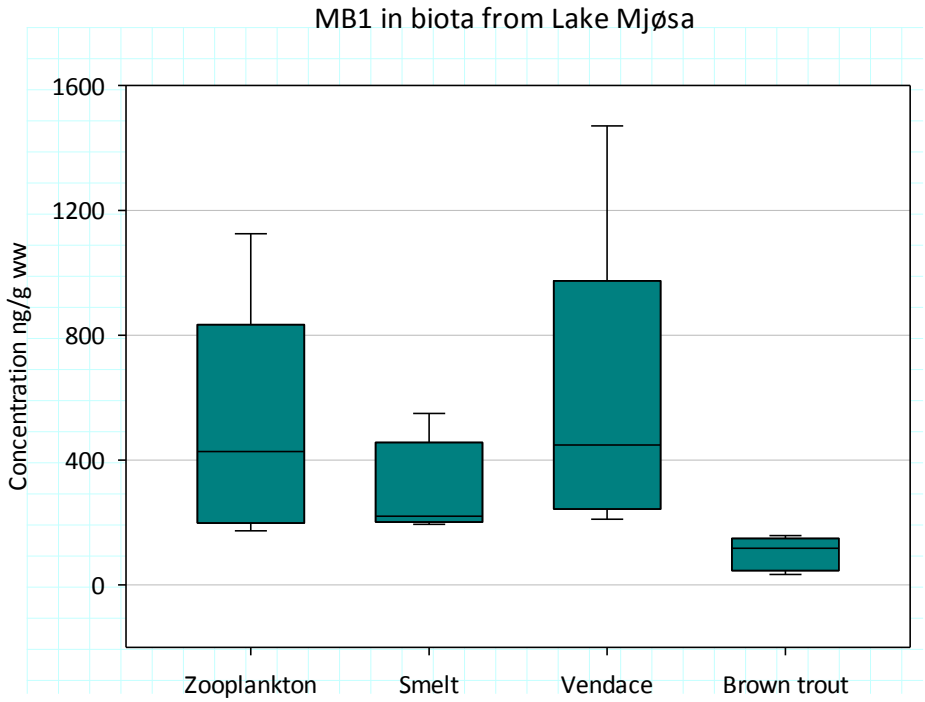


Figure 6. Concentration ranges of the antioxidant MB1 in four different freshwater organisms from Lake Mjøsa showing no indication for bioaccumulation.

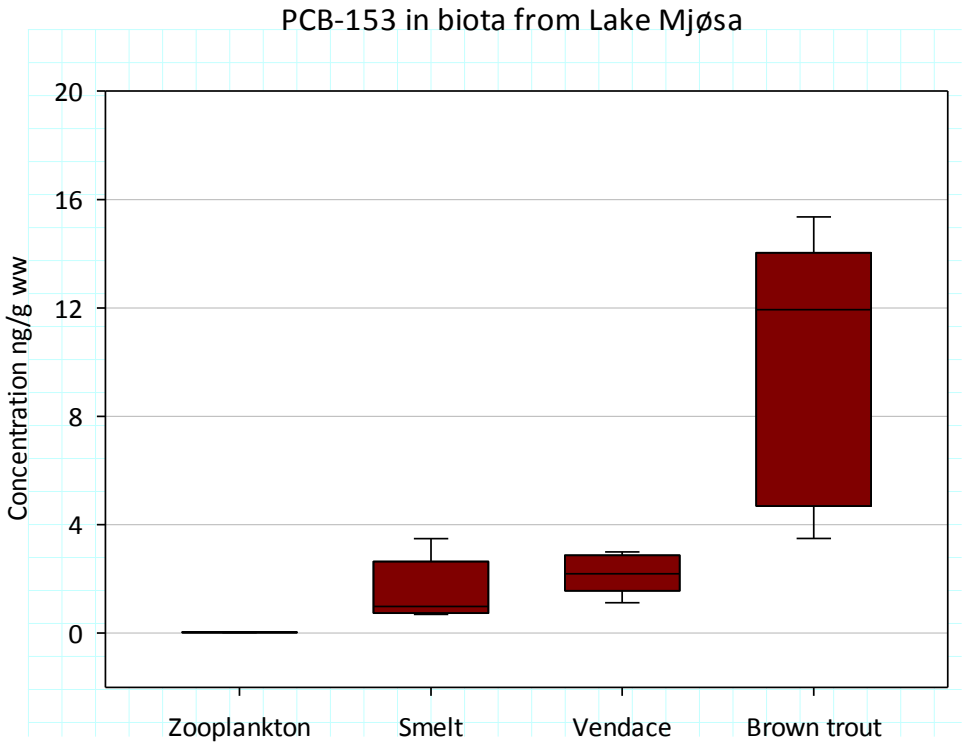


Figure 7. Concentration ranges of PCB-153 in four different freshwater organisms from Lake Mjøsa (same samples as used for MB1 analysis) showing bioaccumulation.

From the other measured PBT compounds DBT was detected in one single sample of vendace at a concentration (1200 ng/g ww) above the LoD (60-730 ng/g ww).

Pigments

Sediments

Two pigments were detected in sediments from Lake Mjøsa at a median concentration of 0.6 ng/g dw (Red-112) and 1.2 ng/g dw (Red-146). Red-146 was only detected in one of the five samples at a concentration just above the LoD.

In a very recent study the occurrence of a long range of pigment particles in sediments from Lake Garda has been shown (Imhof et al., 2016). However, this group used a completely different chemical detection mode, which give the number of microparticles per gram sample and not the concentration (ng/g) and is thus not direct comparable.

Biota

No pigments were detected in the biota samples from Lake Mjøsa (brown trout, smelt, vendace and zooplankton) above LoD.

Siloxanes and fluorinated siloxanes

Sediments

Both D5 and M3T(Ph) were detected in freshwater sediments from Lake Mjøsa. The concentrations of D5 were in the range of 2.7 to 10 ng/g dw. The concentrations found for M3T(Ph) were much lower ranging from LoD (<0.04 ng/g dw) up to 0.2 ng/g dw. This is one order of magnitude lower than what was measured earlier in Lake Mjøsa (1.7 ng/g dw) (McLachlan et al., 2014).

The fluorinated siloxanes were not found above LoD (0.004 – 0.2 ng/g dw), which is in contrast to the levels measured earlier (1.6 and 1.8 ng/g dw) (McLachlan et al., 2014).

Biota

Both D5 and M3T(Ph) were detected in freshwater organisms from Lake Mjøsa. The concentrations of D5 were in the range of 0.7 ng/g ww for zooplankton up to 100 ng/g ww for brown trout. The concentrations found for M3T(Ph) were much lower ranging from LoD (<0.1 ng/g ww) for zooplankton up to 2 ng/g ww for brown trout. Both siloxanes show a typical bioaccumulation pattern, as shown in Figures 8 and 9. The fluorinated siloxanes were only detected occasionally in one sample of brown trout (D4F: 0.024 ng/g ww) and zooplankton (D4F: 0.01 ng/g ww and D3F: 0.012 ng/g ww) and only slightly above LoD.

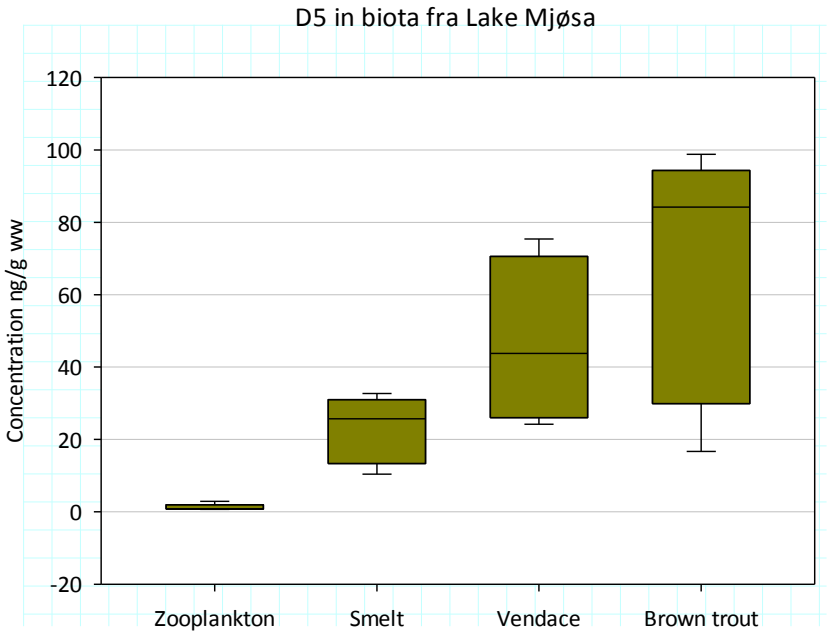


Figure 8. Concentrations of D5 in four different freshwater organisms from Lake Mjøsa showing bioaccumulation

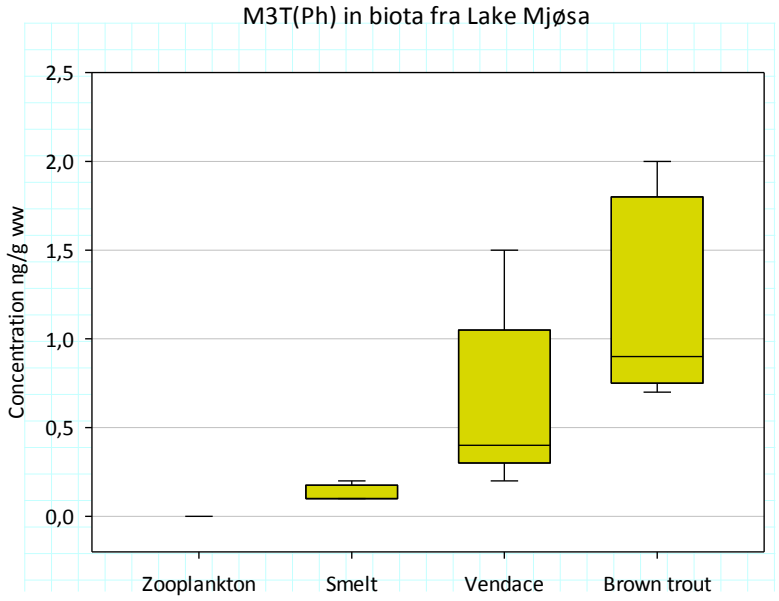


Figure 9. Concentrations of M3T(Ph) in four different freshwater organisms from Lake Mjøsa showing bioaccumulation.

3.4 Supporting parameters

The following support parameters were measured on the samples when appropriate; Particle Size Analysis, Sediment TOC, Water DOC, Lipid content δ13C/δ13N ratio analysis. The results are summarized in appendix E. The results of the isotope analysis are given in Figure 10.

The stable isotope analysis from the food chain (zooplankton, vendace, smelt and trout) in lake Mjösa revealed some special circumstances for 2015. Only very small differences were observed in the δ13N especially between smelt and brown trout. This might reflect a shortage of prey and cannibalism of larger fish of the smaller fish. This makes the isotope data less useful in the interpretation of the screening data presented in this report. The isotope distribution of lake Mjøsa is discussed in detail in 'Miljøgifter i store norske innsjøer, 2015Forekomst og biomagnifisering i fisk og zooplankton' Miljødirektorat 2016.

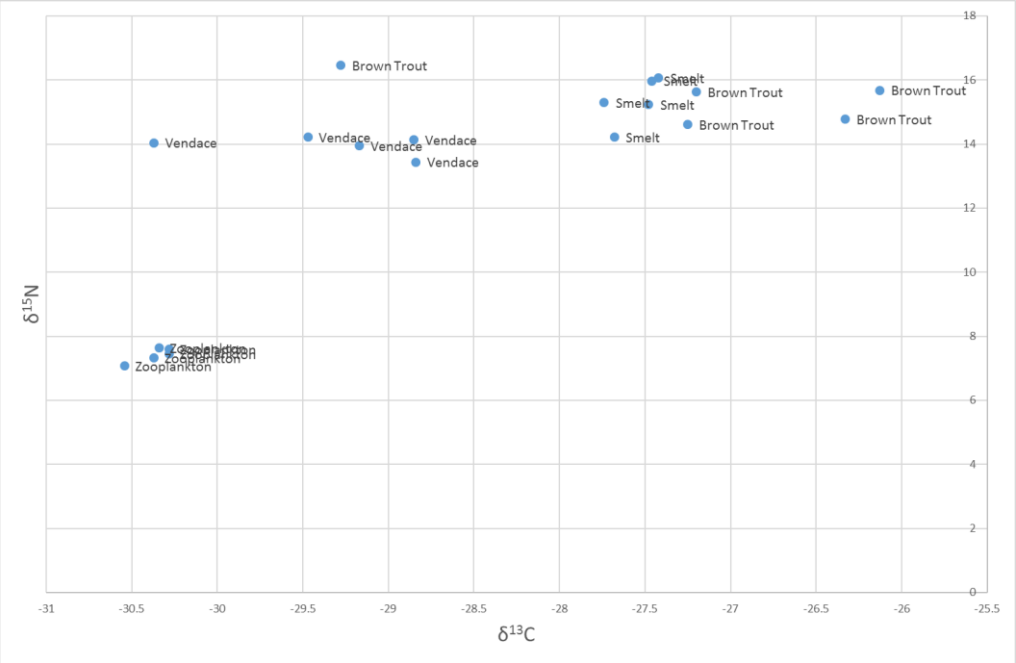


Figure 10. Stable δ13C and δ15N isotopes in the Lake Mjøsa biota.

4. Environmental Risk

4.1 Benzothiazoles

The environmental risk was assessed by comparing the highest measured concentrations (MEC) of the compounds in water and sediment with available PNEC values (Table 10) by calculating MEC/PNEC ratios. MEC/PNEC ratios above 1 generally indicate an environmental risk. In cases where the measured concentrations were below the LOD, a worst-case scenario was used in the MEC/PNEC calculations in order to assess whether the LOD is sufficiently low. As PNEC values were only available for a limited number of compounds, it was not possible to perform a complete environmental risk assessment. A comparison of the LOD values and PNEC for the four compounds in Table 11 showed that the LOD was higher than the PNEC sediment for 2-Benzothiazolol and 2-Mercaptobenzothiazole. Therefore, a potential environmental risk for sediment dwelling organisms of these compounds cannot be excluded even though the measured concentrations were below LOD.

No environmental risk was identified at the highest measured concentrations of 2,2'-dithiobis(benzothiazole) in water and sediment. A potential environmental risk was observed for N-cyclohexyl-2-benzothiazolesulfenamide and 2-benzothiazolol using PNEC for marine waters. However, the compounds were only detected in concentrations above LOD in freshwater, indicating a low environmental risk of these compounds.

Generally, low environmental risk was identified, based on the measured concentrations of the four compounds in Table 10. However, the LOD for 2-Benzothiazolol and 2-Mercaptobenzothiazole in sediment samples were above PNEC sediment values, meaning that concentrations below LOD might still pose an environmental risk. The concentrations measured in fresh water posed low environmental risk. However, similar concentrations in marine water would indicate an environmental risk, highlighting the importance of screening outlets and water bodies related to both fresh and marine waters. The highest concentrations of 2-benzothiazolol was found in the outlet form a water treatment plant to a fresh water lake. Thus, waste water treatment plants might be a source for this compound also to marine waters.

Table 100. PNEC values and calculated MEC/PNEC ratios for freshwater (f), marine water (m) and sediment (s)
Ratios above 1 which is indicative of environmental risk are shown in bold

Compound	$PNEC_f$ (ng/L)	$PNEC_m$ (ng/L)	PNECs (ng/g wwt)	MEC/ $PNEC_f$	MEC/ $PNEC_m$	MEC/ $PNEC_{sed}$
2,2'-Dithiobis(benzothiazole) (MBTS)	600	60	59	0.008*	0.08*	0.08*
N-Cyclohexyl-2-benzothiazole sulfenamide (CBTS)	320	32	86.6	0.24	2.38	0.03
2-Benzothiazolol (HBT)	16100	1600	36,7	0.16	1.58	1.36*
2-Mercaptobenzothiazole (MBT)	820	82	22,8	0.02*	0.24*	2.19*

***LOD used as MEC**
All PNEC values were obtained from European commission, 2008. European Union Risk Assessment Report N-Cyclohexylbenzothiazol-2-sulpenamide. R035_HH_ENV_0805.DOC, 272p. (ECHA 1)

4.2 Pigments

No PNECs for pigments were found in the international literature. Low concentrations of pigments in the low ng/L level were found in effluent. Five of the pigments were present in all sludge samples of both WWTPs. The pigments do not seem to bio accumulate and were not found in concentration above the LoD in the all biota samples from Mjøsa.

4.3 PBT compounds

PNECs were only available for DBT (ECHA 2) 110 ug/kg dw in sediment and DBDPE in both fresh water and marine sediment (100 mg/kg dw and 10 mg/kg dw respectively). DBT was not detected in any of the samples except for one vendace at a level of 1 µg/g ww (ECHA 2). DBDPE was not detected in any of the samples including sediment at a LoD of 10 ng/g dw (10 ug/kg) (ECHA 3) which is long under the PNEC for this compound. No PNEC was available for MB1.

4.4 Siloxanes and fluorinated siloxanes

For the siloxanes, including the fluorinated siloxanes, only a PNEC was available for D5 (ECHA 4) in fresh water and in marine sediment (11 and 1.1 mg/kg dw respectively). Levels of D5 in sediment from lake Mjøsa varied from 3-10 µg /kg dw which is below the PNEC. M3T(Ph) was detected in lake Mjøsa sediment at even lower concentration < 0.04- 0.2 µg /kg. Levels in sludge however are in the range from 7.6-8.1 mg/kg and thus close to or over the sediment PNEC. The levels in sludge are however not as relevant as sediment levels for risk assessment. Of concern is that both D5 and M3T(Ph) were shown to bio accumulate in biota.

5. Conclusion

Several of the target compounds were found in effluent, sludge, leachate and environmental samples of the analysis of 5 pigments and 5 selected PBT compounds, 8 benzothiazoles, and 4 selected siloxanes and fluoro-siloxanes based on earlier screening studies.

- 2-benzothiazolol was detected in effluent from the Gjøvik WWTP at high levels (1.3-2.5 µg/L). BTSA, CBTS and TCMBT were found at low concentrations in the surface water of lake Mjøsa. These levels indicate little environmental risk when these levels are compared with their PNEC values.

- Pigments (RED-112, RED-14, RED-146 and Orange-13) were found in the majority of the WWTP samples, sediment samples from Lake Mjøsa and the leachate from both landfill sites. Pigments were not found in any of the biota samples.

- The anti-oxidant 4,4'-methylenebis[2,6-bis(1,1-dimethylethyl)-phenol (MB1) was found in effluent of both WWTP, and leachate samples. Although 4,4'-methylenebis[2,6-bis (1,1-dimethylethyl)-phenol does not seem to bio accumulate, the occurrence of this compound in biological samples is of concern.

- Decamethylcyclo pentasiloxane (D5) was present in all samples and at relatively high concentrations in sludge samples from both the Hias and the Gjøvik WTTP. Tris (trimethyl siloxy)phenylsilane (M3T) was present in sludge and several of the sediment samples from lake Mjøsa. Levels of decamethylcyclo pentasiloxane (D5) were well below the PNEC value for sediment. Both compounds bio accumulate in the biological samples analysed (zooplankton, vendace, smelt and brown trout).

- The fluorinated siloxane 2,4,6-trimethyl-2,4,6-tris(3,3,3-trifluoropropyl)-cyclotrisiloxane (D3F) was found in the effluent samples from both WWTPs and in the leachate four of the leachate samples. 2,4,6,8-tetramethyl- 2,4,6,8-tetrakis(3,3,3-trifluoropropyl)-cyclotetrasiloxane (D4F) was found in one of the leachate samples from the Lindum landfill and all sludge samples from the Gjøvik WTTP (D4F). All levels of the fluorinated siloxanes were relatively low.

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ECHA 2 *http://echa.europa.eu/registration-dossier/-/registered-dossier/2137/6/1*

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Attachments

Attachment A: Pigments

Pigments (Effluent, Leachate)							
		CAS	2425-85-6	6535-46-2	5280-68-2	5280-68-2	3520-72-7
		Compound	RED-3	RED-112	RED-14	RED-146	ORANGE-13
Sample	Location	ID.nr.	ng/l	ng/l	ng/l	ng/l	ng/l
Effluent	Hias WWTP	Day 1	< 7	< 0.1	< 0.4	< 0.6	< 7
Effluent	Hias WWTP	Day 2	< 4	< 0.1	0.7	< 0.2	< 6
Effluent	Hias WWTP	Day 3	< 5	< 0.1	< 0.3	< 0.5	< 11
Effluent	Hias WWTP	Day 4	< 5	< 0.1	< 0.4	< 0.7	< 8
Effluent	Hias WWTP	Day 5	< 6	< 0.1	< 0.5	< 0.4	< 6
Effluent	Rambekk WWTP	Day 1	< 14	0.5	1.4	1.2	< 15
Effluent	Rambekk WWTP	Day 2	< 12	0.4	2.4	1.2	< 17
Effluent	Rambekk WWTP	Day 3	< 13	1.4	2.9	5.4	< 42
Effluent	Rambekk WWTP	Day 4	< 19	0.7	3.5	1.8	< 24
Effluent	Rambekk WWTP	Day 5	< 10	< 0.0	< 0.1	< 0.1	< 16
Leachate	ISI Bærum	Day 1	< 6	0.5	0.8	1.6	< 12
Leachate	ISI Bærum	Day 2	< 4	0.3	0.4	< 2.7	< 32
Leachate	ISI Bærum	Day 3	< 18	2.7	1.3	1.9	< 13
Leachate	Lindum	Day 1	< 10	< 0.2	< 0.7	< 1.3	< 16
Leachate	Lindum	Day 2	< 11	< 0.1	< 0.5	< 0.7	< 13

Pigments (Sludge and Sediment)							
		CAS	2425-85-6	6535-46-2	5280-68-2	5280-68-2	3520-72-7
		Compound	RED-3	RED-112	RED-14	RED-146	ORANGE-13
Sample	Location	ID.nr.	ng/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw
Sludge	Hias WWTP	Day 1	< 16	1.0	25.9	7.8	239
Sludge	Hias WWTP	Day 2	< 31	1.9	10.5	13.2	185
Sludge	Hias WWTP	Day 3	< 31	4.2	6.1	6.0	164
Sludge	Hias WWTP	Day 4	< 26	38.4	14.3	3.1	191
Sludge	Hias WWTP	Day 5	< 29	5.0	13.1	3.8	224
Sludge	Rambekk WWTP	Day 1	< 6	31	150	21	55
Sludge	Rambekk WWTP	Day 2	< 8	28	167	36	48
Sludge	Rambekk WWTP	Day 3	< 11	30	253	25	50
Sludge	Rambekk WWTP	Day 4	< 10	33	186	29	52
Sludge	Rambekk WWTP	Day 5	< 7	23	186	33	32
Sediment	Mjøsa	Station 1	< 17	1.2	< 0.5	< 0.4	< 3
Sediment	Mjøsa	Station 2	< 7	1.9	< 0.6	1.2	< 5
Sediment	Mjøsa	Station 3	< 17	1.7	< 1.6	< 1.7	< 10
Sediment	Mjøsa	Station 4	< 7	0.6	< 0.5	< 0.4	< 3
Sediment	Mjøsa	Station 5	< 6	1.3	< 1.1	< 0.8	< 4

Pigments (Biological samples)							
		CAS	2425-85-6	6535-46-2	5280-68-2	5280-68-2	3520-72-7
		Compound	RED-3	RED-112	RED-14	RED-146	ORANGE-13
Sample	Location	ID.nr.	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww
Brown Trout (Muscle)	Mjøsa	MØ-2-15	< 13	< 0.1	< 1.1	< 1.6	< 5
Brown Trout (Muscle)	Mjøsa	MØ-5-15	< 7	< 0.1	< 1.4	< 2.0	< 4
Brown Trout (Muscle)	Mjøsa	MØ-6-15	< 5	< 0.1	< 0.4	< 0.5	< 2
Brown Trout (Muscle)	Mjøsa	MØ-7-15	< 12	< 0.1	< 1.0	< 2.3	< 5
Brown Trout (Muscle)	Mjøsa	MØ-8-15	< 13	< 0.1	< 0.8	< 1.1	< 3
Smelt (Muscle)	Mjøsa	12081	< 6	< 0.1	< 2.1	< 1.7	< 10
Smelt (Muscle)	Mjøsa	12082	< 24	< 0.1	< 2.3	< 1.4	< 9
Smelt (Muscle)	Mjøsa	12083	< 6	< 0.1	< 2.7	< 2.7	< 15
Smelt (Muscle)	Mjøsa	12084	< 5	< 0.1	< 1.2	< 1.5	< 9
Smelt (Muscle)	Mjøsa	12085	< 5	< 0.1	< 3.8	< 1.2	< 9
Vendace (Muscle)	Mjøsa	ML-1	< 11	< 0.1	< 1.6	< 3.2	< 7
Vendace (Muscle)	Mjøsa	ML-2	< 13	< 0.0	< 2.3	< 3.0	< 7
Vendace (Muscle)	Mjøsa	ML-3	< 10	< 0.1	< 1.0	< 1.6	< 5
Vendace (Muscle)	Mjøsa	ML-4	< 22	< 0.1	< 3.9	< 10.1	< 12
Vendace (Muscle)	Mjøsa	ML-5	< 21	< 0.1	< 2.6	< 5.3	< 10
Zooplankton	Mjøsa	12096	< 10	< 0.4	< 2.8	< 1.7	< 13
Zooplankton	Mjøsa	12097	< 16	< 0.4	< 8.4	< 3.0	< 24
Zooplankton	Mjøsa	12098	< 10	< 0.3	< 3.2	< 2.0	< 13
Zooplankton	Mjøsa	12099	< 19	< 0.7	< 12.1	< 3.0	< 21
Zooplankton	Mjøsa	12100	< 9	< 0.3	< 3.7	< 2.0	< 16

Attachment B: PBT Compounds

PBT (Effluent, Leachate)							
		CAS	119851-28-4	26898-17-9 29589-57-9	118-82-1	84852-53-9	32588-76-4
		Compd.	CCPPE	DBT	MB1	DBDPE	EBTPI
Sample	Location	ID.nr.	ng/L	ng/L	ng/L	ng/L	ng/L
Effluent	Hias WWTP	Day 1	< 67	< 67	121	< 50	< 50
Effluent	Hias WWTP	Day 2	< 184	< 22	72	< 50	< 50
Effluent	Hias WWTP	Day 3	< 249	< 33	40	< 50	< 50
Effluent	Hias WWTP	Day 4	< 357	< 42	129	< 50	< 50
Effluent	Hias WWTP	Day 5	< 66	< 15	127	< 50	< 50
Effluent	Rambekk WWTP	Day 1	< 224	< 51	77	< 50	< 50
Effluent	Rambekk WWTP	Day 2	< 268	< 87	58	< 50	< 50
Effluent	Rambekk WWTP	Day 3	< 50	< 110	133	< 50	< 50
Effluent	Rambekk WWTP	Day 4	< 59	< 84	25	< 50	< 50
Effluent	Rambekk WWTP	Day 5	< 242	< 35	107	< 50	< 50
Leachate	ISI Bærum	Day 1	<2	<0.2	0.028	< 50	< 50
Leachate	ISI Bærum	Day 2	<1	<0.1	<0.007	< 50	< 50
Leachate	ISI Bærum	Day 3	<0.5	<0.1	<0.005	< 50	< 50
Leachate	Lindum	Day 1	<0.3	6.0	0.01	< 50	< 50
Leachate	Lindum	Day 2	<0.2	<0.4	0.1	< 50	< 50

PBT (Sludge and Sediment)							
		CAS	119851-28-4	26898-17-9 29589-57-9	118-82-1	84852-53-9	32588-76-4
		Compd.	CCPPE	DBT	MB1	DBDPE	EBTPI
Sample	Location	ID.nr.	ng/L	ng/L	ng/L	ng/L	ng/L
Sludge	Hias WWTP	Day 1	< 1392	< 3053	< 203	< 50	< 50
Sludge	Hias WWTP	Day 2	< 1142	< 3199	< 222	< 50	< 50
Sludge	Hias WWTP	Day 3	< 489	< 1661	< 107	< 50	< 50
Sludge	Hias WWTP	Day 4	< 866	< 3130	< 185	< 50	< 50
Sludge	Hias WWTP	Day 5	< 684	< 2848	< 138	< 50	< 50
Sludge	Rambekk WWTP	Day 1	< 1820	< 3947	< 345	< 50	< 50
Sludge	Rambekk WWTP	Day 2	< 2625	< 3663	< 371	< 50	< 50
Sludge	Rambekk WWTP	Day 3	< 2169	< 4170	1655	< 50	< 50
Sludge	Rambekk WWTP	Day 4	< 1113	< 1090	< 46	< 50	< 50
Sludge	Rambekk WWTP	Day 5	< 2299	< 4420	< 1323	< 50	< 50
Sediment	Mjøsa	Station 1	< 10	< 10	< 10	< 10	< 10
Sediment	Mjøsa	Station 2	< 10	< 10	< 10	< 10	< 10
Sediment	Mjøsa	Station 3	< 10	< 10	< 10	< 10	< 10
Sediment	Mjøsa	Station 4	< 10	< 10	< 10	< 10	< 10
Sediment	Mjøsa	Station 5	< 10	< 10	< 10	< 10	< 10

PBT (Biological Samples)							
		CAS	119851-28-4	26898-17-9	118-82-1	84852-53-9	32588-76-4
				29589-57-9			
			CCPPE	DBT	MB1	DBDPE	EBTPI
Sample	Location	ID.nr.	ng/L	ng/L	ng/L	ng/L	ng/L
Brown Trout (Muscle)	Mjøsa	MØ-2-15	< 124	< 10	58	< 10	< 10
Brown Trout (Muscle)	Mjøsa	MØ-5-15	< 195	< 20	34	< 10	< 10
Brown Trout (Muscle)	Mjøsa	MØ-6-15	< 414	< 105	158	< 10	< 10
Brown Trout (Muscle)	Mjøsa	MØ-7-15	< 334	< 71	117	< 10	< 10
Brown Trout (Muscle)	Mjøsa	MØ-8-15	< 430	< 59	139	< 10	< 10
Smelt (Muscle)	Mjøsa	12081	< 373	< 62	220	< 10	< 10
Smelt (Muscle)	Mjøsa	12082	< 335	< 86	194	< 10	< 10
Smelt (Muscle)	Mjøsa	12083	< 1366	< 97	363	< 10	< 10
Smelt (Muscle)	Mjøsa	12084	< 999	< 94	549	< 10	< 10
Smelt (Muscle)	Mjøsa	12085	< 994	< 146	208	< 10	< 10
Vendace (Muscle)	Mjøsa	ML-1	< 214	< 62	478	< 10	< 10
Vendace (Muscle)	Mjøsa	ML-2	< 153	1174	210	< 10	< 10
Vendace (Muscle)	Mjøsa	ML-3	< 543	< 69	275	< 10	< 10
Vendace (Muscle)	Mjøsa	ML-4	< 5432	< 732	1471	< 10	< 10
Vendace (Muscle)	Mjøsa	ML-5	< 531	< 68	448	< 10	< 10
Zooplankton	Mjøsa	12096	< 786	< 93	427	< 10	< 10
Zooplankton	Mjøsa	12097	< 914	< 333	1125	< 10	< 10
Zooplankton	Mjøsa	12098	< 594	< 85	173	< 10	< 10
Zooplankton	Mjøsa	12099	< 408	< 122	223	< 10	< 10
Zooplankton	Mjøsa	12100	< 464	< 85	543	< 10	< 10

Attachment C: Siloxanes

Siloxanes (Effluent, Leachate, Sludge and Sediment)						
		CAS	541-02-6	2116-84-9	429-67-4	2374-14-3
		Compd.	D5	M3T(Ph)	D4F	D3F +D3FOH
Sample	Location	ID.nr.	ng/L	ng/L	ng/L	ng/L
Effluent	Hias WWTP	Day 1	83	< 4	<0.28	<0.035
Effluent	Hias WWTP	Day 2	93	< 4	<0.20	<0.047
Effluent	Hias WWTP	Day 3	95	< 4	<0.27	<0.074
Effluent	Hias WWTP	Day 4	76	< 4	<0.19	0.08
Effluent	Hias WWTP	Day 5	147	< 4	<0.20	0.11
Effluent	Rambekk WWTP	Day 1	46	<0.7	<0.36	0.50
Effluent	Rambekk WWTP	Day 2	35	<0.7	<0.15	<0.054
Effluent	Rambekk WWTP	Day 3	21	<0.7	<0.48	0.16
Effluent	Rambekk WWTP	Day 4	31	<0.7	<0.59	<0.063
Effluent	Rambekk WWTP	Day 5	24	<0.7	<0.34	0.13
Leachate	ISI Bærum	Day 1	84	< 1	<0.12	<0.089
Leachate	ISI Bærum	Day 2	82	< 1	<0.094	0.12
Leachate	ISI Bærum	Day 3	74	< 1	<0.3	0.11
Leachate	Lindum	Day 1	243	< 1	< 2	0.24
Leachate	Lindum	Day 2	371	< 1	2.80	0.30
			ng/g dw	ng/g dw	ng/g dw	ng/g dw
Sludge	Hias WWTP	Day 1	8077	98	<0.036	<0.049
Sludge	Hias WWTP	Day 2	7830	95	<0.026	<0.051
Sludge	Hias WWTP	Day 3	7595	89	<0.08	<0.068
Sludge	Hias WWTP	Day 4	7922	87	<0.036	<0.033
Sludge	Hias WWTP	Day 5	8077	98	<0.036	<0.049
Sludge	Rambekk WWTP	Day 1	5852	87	0.56	<0.024
Sludge	Rambekk WWTP	Day 2	5852	56	0.12	<0.028
Sludge	Rambekk WWTP	Day 3	6431	57	0.19	<0.069
Sludge	Rambekk WWTP	Day 4	6344	60	0.18	<0.024
Sludge	Rambekk WWTP	Day 5	5815	52	0.16	<0.03
Sediment	Mjøsa	Station 1	10	0.2	<0.0056	<0.008
Sediment	Mjøsa	Station 2	5.4	0.1	<0.01	<0.0033
Sediment	Mjøsa	Station 3	3.5	<0.04	<0.018	<0.011
Sediment	Mjøsa	Station 4	3.3	0.1	<0.011	<0.005
Sediment	Mjøsa	Station 5	2.7	<0.08	<0.013	<0.0038

Siloxanes (Biological Samples)						
		CAS	541-02-6	2116-84-9	429-67-4	2374-14-3
		Compound	D5	M3T(Ph)	D4F	D3F+D3FOH
Sample	Location	ID.nr.	ng/g ww	ng/g ww	ng/g ww	ng/g ww
Brown Trout (Muscle)	Mjøsa	MØ-2-15	43	0.8	<0.024	<0.012
Brown Trout (Muscle)	Mjøsa	MØ-5-15	99	2.0	0.024	<0.0095
Brown Trout (Muscle)	Mjøsa	MØ-6-15	84	0.9	<0.0093	<0.0098
Brown Trout (Muscle)	Mjøsa	MØ-7-15	90	1.6	<0.019	<0.0083
Brown Trout (Muscle)	Mjøsa	MØ-8-15	17	0.7	<0.016	<0.0076
Smelt (Muscle)	Mjøsa	12081	33	0.2	<0.0081	<0.0078
Smelt (Muscle)	Mjøsa	12082	10	<0.05	<0.0037	<0.0075
Smelt (Muscle)	Mjøsa	12083	29	0.1	<0.0071	<0.0067
Smelt (Muscle)	Mjøsa	12084	26	0.1	<0.01	<0.0077
Smelt (Muscle)	Mjøsa	12085	16	0.1	<0.0029	<0.0044
Vendace (Muscle)	Mjøsa	ML-1	24	0.2	<0.0065	<0.0094
Vendace (Muscle)	Mjøsa	ML-2	75	0.6	<0.0076	<0.011
Vendace (Muscle)	Mjøsa	ML-3	56	0.4	<0.021	<0.0099
Vendace (Muscle)	Mjøsa	ML-4	306	1.5	<0.012	<0.01
Vendace (Muscle)	Mjøsa	ML-5	32	0.4	<0.016	<0.01
Zooplankton	Mjøsa	12096	0.7	<0.05	0.010	0.012
Zooplankton	Mjøsa	12097	0.8	<0.05	<0.0014	<0.0025
Zooplankton	Mjøsa	12098	2.9	<0.05	<0.0013	<0.0022
Zooplankton	Mjøsa	12099	1.0	<0.05	<0.0012	<0.0031
Zooplankton	Mjøsa	12100	0.8	<0.05	<0.0032	<0.0027

Attachment D: Benzothiazoles

Benzothiazoles (Effluent, Surface Water and Leachate)										
		CAS	120-78-5	95154-01-1	95-33-0	4299-07-4	21564-17-0	934-34-9	149-30-4	883-93-2
		Compound	MBTS	BTSA	CBTS	BBTO	TCMTB	HBT	MBT	PBT
Sample	Location	ID.nr.	ng/L	ng/L	ng/L	ng/L	ng/L	ng/L	ng/L	ng/L
Effluent	Hias WWTP	Day 1	< 5	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Effluent	Hias WWTP	Day 2	< 5	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Effluent	Hias WWTP	Day 3	< 5	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Effluent	Hias WWTP	Day 4	< 5	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Effluent	Hias WWTP	Day 5	< 5	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Effluent	Rambekk WWTP	Day 1	< 5	< 10	< 1	< 10	< 5	1985	< 20	< 20*
Effluent	Rambekk WWTP	Day 2	< 5	< 10	< 1	< 10	< 5	2520	< 20	< 20*
Effluent	Rambekk WWTP	Day 3	< 5	< 10	< 1	< 10	< 5	1608	< 20	< 20*
Effluent	Rambekk WWTP	Day 4	< 5	< 10	< 1	< 10	< 5	1305	< 20	< 20*
Effluent	Rambekk WWTP	Day 5	< 5	< 10	< 1	< 10	< 5	1657	< 20	< 20*
Surface Water	Mjøsa	Station 1	<5	<10	<1	<10	<5	<20	<20	< 20*
Surface Water	Mjøsa	Station 2	<5	<10	<1	<10	<5	<20	<20	< 20*
Surface Water	Mjøsa	Station 3	<5	<10	10.2	<10	8.8	<20	<20	< 20*
Surface Water	Mjøsa	Station 4	<5	25.1	<1	<10	<5	<20	<20	< 20*
Surface Water	Mjøsa	Station 5	<5	<10	76.2	<10	15.4	<20	<20	< 20*
Leachate	ISI Bærum	Day 1	< 5*	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Leachate	ISI Bærum	Day 2	< 5*	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Leachate	ISI Bærum	Day 3	< 5*	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Leachate	Lindum	Day 1	< 5*	< 10	< 1	< 10	< 5	< 20	< 20	< 20*
Leachate	Lindum	Day 2	< 5*	< 10	< 1	< 10	< 5	< 20	< 20	< 20*

Benzothiazoles (Sludge and Sediment)										
		CAS	120-78-5	95154-01-1	95-33-0	4299-07-4	21564-17-0	934-34-9	149-30-4	883-93-2
		Compound	MBTS	BTSA	CBTS	BBTO	TCMTB	HBT	MBT	PBT
Sample	Location	ID.nr.	ng/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw
Sludge	Hias WWTP	Day 1	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Hias WWTP	Day 2	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Hias WWTP	Day 3	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Hias WWTP	Day 4	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Hias WWTP	Day 5	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Rambekk WWTP	Day 1	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Rambekk WWTP	Day 2	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Rambekk WWTP	Day 3	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Rambekk WWTP	Day 4	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sludge	Rambekk WWTP	Day 5	< 1*	< 1*	< 1	< 10	< 5	< 50	< 50	< 50*
Sediment	Mjøsa	Day 1	< 10*	< 10*	< 1	< 10	< 5	< 50	< 50	< 20*
Sediment	Mjøsa	Day 2	< 10*	< 10*	< 1	< 10	< 5	< 50	< 50	< 20*
Sediment	Mjøsa	Day 3	< 10*	< 10*	< 1	< 10	< 5	< 50	< 50	< 20*
Sediment	Mjøsa	Day 4	< 10*	< 10*	< 1	< 10	< 5	< 50	< 50	< 20*
Sediment	Mjøsa	Day 5	< 10*	< 10*	< 1	< 10	< 5	< 50	< 50	< 20*

*LOD based on low recovery of spike experiments

Benzothiazoles (Biological Samples)										
		CAS	120-78-5	95154-01-1	95-33-0	4299-07-4	21564-17-0	934-34-9	149-30-4	883-93-2
		Compound	MBTS	BTSA	CBTS	BBTO	TCMTB	HBT	MBT	PBT
Sample	Location	ID.nr.	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww
Brown Trout (Muscle)	Mjøsa	MØ-2-15	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Brown Trout (Muscle)	Mjøsa	MØ-5-15	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Brown Trout (Muscle)	Mjøsa	MØ-6-15	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Brown Trout (Muscle)	Mjøsa	MØ-7-15	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Brown Trout (Muscle)	Mjøsa	MØ-8-15	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Smelt (Muscle)	Mjøsa	12081	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Smelt (Muscle)	Mjøsa	12082	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Smelt (Muscle)	Mjøsa	12083	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Smelt (Muscle)	Mjøsa	12084	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Smelt (Muscle)	Mjøsa	12085	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Vendace (Muscle)	Mjøsa	ML-1	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Vendace (Muscle)	Mjøsa	ML-2	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Vendace (Muscle)	Mjøsa	ML-3	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Vendace (Muscle)	Mjøsa	ML-4	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Vendace (Muscle)	Mjøsa	ML-5	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Zooplankton	Mjøsa	12096	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Zooplankton	Mjøsa	12097	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Zooplankton	Mjøsa	12098	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Zooplankton	Mjøsa	12099	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*
Zooplankton	Mjøsa	12100	< 10*	< 10	< 1	< 10	< 5	< 50	< 50	< 50*

*LOD based on low recovery of spike experiments

Attachment E. Support parameters

Support parameters for effluent samples			
			DOC
Sample	Location	ID.nr.	mg C/L
Effluent	Hias WWTP	Day 1	19.2
Effluent	Hias WWTP	Day 2	20.4
Effluent	Hias WWTP	Day 3	18.9
Effluent	Hias WWTP	Day 4	22.2
Effluent	Hias WWTP	Day 5	18.7
Effluent	Rambekk WWTP	Day 1	16.9
Effluent	Rambekk WWTP	Day 2	25.2
Effluent	Rambekk WWTP	Day 3	13.9
Effluent	Rambekk WWTP	Day 4	15.3
Effluent	Rambekk WWTP	Day 5	15.4
Leachate	ISI Bærum	Day 1	35.3
Leachate	ISI Bærum	Day 2	33.4
Leachate	ISI Bærum	Day 3	34.5
Leachate	Lindum	Day 1	473
Leachate	Lindum	Day 2	204

Support parameters for sludge and sediment samples				
			DOC	<63 µm
Sample	Location	ID.nr.	mg C/L	
Sludge	Hias WWTP	Day 1	256	-
Sludge	Hias WWTP	Day 2	291	-
Sludge	Hias WWTP	Day 3	287	-
Sludge	Hias WWTP	Day 4	294	-
Sludge	Hias WWTP	Day 5	293	-
Sludge	Rambekk WWTP	Day 1	293	-
Sludge	Rambekk WWTP	Day 2	257	-
Sludge	Rambekk WWTP	Day 3	264	-
Sludge	Rambekk WWTP	Day 4	266	-
Sludge	Rambekk WWTP	Day 5	263	-
Sediment	Mjøsa	Station 1	36.9	76
Sediment	Mjøsa	Station 2	50.3	81
Sediment	Mjøsa	Station 3	41.8	75
Sediment	Mjøsa	Station 4	44.1	73
Sediment	Mjøsa	Station 5	43.5	70

Support parameters for biological samples							
			d13CVPDB	d15NAIR	W% C	W% N	C/N
Sample	Location	ID.nr.	ng/g ww	ng/g ww	ng/g ww	ng/g ww	ng/g ww
Brown Trout (Muscle)	Mjøsa	MØ-2-15	-27.25	14.61	46.49	13.77	3.38
Brown Trout (Muscle)	Mjøsa	MØ-5-15	-27.20	15.63	46.28	13.09	3.54
Brown Trout (Muscle)	Mjøsa	MØ-6-15	-26.13	15.68	44.68	14.07	3.18
Brown Trout (Muscle)	Mjøsa	MØ-7-15	-29.28	16.46	49.45	10.82	4.57
Brown Trout (Muscle)	Mjøsa	MØ-8-15	-26.33	14.78	44.08	14.02	3.14
Smelt (Muscle)	Mjøsa	12081	-27.42	16.07	44.13	13.80	3.20
Smelt (Muscle)	Mjøsa	12082	-27.74	15.30	45.29	13.73	3.30
Smelt (Muscle)	Mjøsa	12083	-27.46	15.97	46.05	12.89	3.57
Smelt (Muscle)	Mjøsa	12084	-27.48	15.24	44.81	13.78	3.25
Smelt (Muscle)	Mjøsa	12085	-27.68	14.21	44.15	14.04	3.14
Vendace (Muscle)	Mjøsa	ML-1	-29.17	13.94	45.74	13.06	3.50
Vendace (Muscle)	Mjøsa	ML-2	-28.84	13.44	45.75	13.25	3.45
Vendace (Muscle)	Mjøsa	ML-3	-30.37	14.04	46.93	11.11	4.22
Vendace (Muscle)	Mjøsa	ML-4	-29.47	14.22	43.89	12.67	3.46
Vendace (Muscle)	Mjøsa	ML-5	-28.85	14.14	45.07	12.94	3.48
Zooplankton	Mjøsa	12096	-30.37	7.32	38.69	6.81	5.69
Zooplankton	Mjøsa	12097	-30.28	7.45	39.21	6.99	5.61
Zooplankton	Mjøsa	12098	-30.34	7.63	41.08	6.19	6.64
Zooplankton	Mjøsa	12099	-30.28	7.60	39.52	7.18	5.51
Zooplankton	Mjøsa	12100	-30.54	7.08	39.27	6.63	5.92

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The Norwegian Environment Agency is working for a clean and diverse environment. Our primary tasks are to reduce greenhouse gas emissions, manage Norwegian nature, and prevent pollution.

We are a government agency under the Ministry of Climate and Environment and have 700 employees at our two offices in Trondheim and Oslo and at the Norwegian Nature Inspectorate's more than sixty local offices.

We implement and give advice on the development of climate and environmental policy. We are professionally independent. This means that we act independently in the individual cases that we decide and when we communicate knowledge and information or give advice.

Our principal functions include collating and communicating environmental information, exercising regulatory authority, supervising and guiding regional and local government level, giving professional and technical advice, and participating in international environmental activities.