MILJØ-
DIREKTORATET

RAPPORT

M-81/2013

Norsk institutt for vannforskning

Stockholm University

Siloxanes in freshwater food webs - a study of three lakes in Norway.

Norwegian screening program

Forord

På oppdrag av Klima og forurensningsdirektoratet (Klif) har Norsk institutt for vannforskning (NIVA) og Department of Applied Environmental Science, Stockholm Universitet (ITM) i 2012 undersøkt forekomst av flyktige sykliske metylerte siloksaner (cyclic volatile methyl siloxanes cVMS) i tre norske innsjøer (Mjøsa, Randsfjorden, Femunden). Biomagnifisering av sykliske siloksaner i det pelagiske næringsnettet med ørret som predator ble beregnet og sammenliknet med velkjente miljøgifter som PCB og DDE, og kilder av sykliske siloksaner til innsjøene ble vurdert.

Resultatene er sammenliknet med en tidligere studie om siloksaner i Mjøsa, samt data fra andre forskningsprosjekter og rapporter der dette var relevant. Dette inkluderer resultater fra NIVAs Strategiske Institutt Satsning for 2012, der bromerte flammehemmere (PBDE) ble analysert i alle prøvene fra Mjøsa.

Feltarbeidet og prøvetakning av invertebrater, fisk, sedimenter, og vann fra 2012 har vært gjort av personell fra NIVA (Jarl Eivind Løvik, Sigurd Rognerud, Eirik Fjeld, Katrine Borgå), samt feltassistent Eilif Fjeld, og lokale fiskere. Personell fra de ulike renseanleggene var behjelpelige med å prøveta renset avløpsvann.

Ved ITM har Michael McLachlan og Amelie Kierkegaard vært ansvarlig for de kjemiske analysene. Ulrika Nordlöf har hjulpet til med utvikling av metoden for siloksananalyse, og Dimitrios Panagopoulos har assistert ved siloksananalysene av prøvematerialet.

Ved NIVA har Andreas Sven Høgfeldt og Kine Bæk vært ansvarlige for de kjemiske analysene. Hovedansvarlig for rapporteringen av prosjektet har vært Katrine Borgå. Rapporten er utformet som et manuskript for internasjonal publisering som vil bli publisert parallelt med rapporten. Erik Fjeld har tatt forsidebildet.

For oppdragsgiver har ansvarlig saksbehandler vært senioringeniør Bård Nordbø.

En stor takk rettes til alle medarbeidere og involverte for et godt samarbeid.

Oslo, 21. november 2013

Katrine Borgå

Innhold

1. Extended abstract

Title: Siloxanes in freshwater food webs – a study of three lakes in Norway
Year: 2013
Authors: Katrine Borgå (NIVA), Eirik Fjeld (NIVA), Amelie Kierkegaard (ITM), Jarl Eivind Løvik (NIVA), Sigurd Rognerud (NIVA), Andreas S. Høgfeldt (NIVA), Kine Bæk (NIVA), Michael S. McLachlan (ITM)
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As part of Climate and Pollution Agency's (Klif) screening of emerging contaminants NIVA collected various members of the pelagic food web in the three Norwegian lakes Mjøsa, Randsfjorden and Femunden, from July to September 2012. The aim was to assess sources of cyclic volatile methyl siloxanes (cVMS) to these lakes, their contamination level and biomagnification in the food web leading to brown trout as top predator. In addition to fish and invertebrates from the pelagic zone, benthic fish were collected in Mjøsa, effluent water from waste water treatment plants (WWTP) from Randsfjorden and Mjøsa, water samples from Mjøsa, and surface sediments from all three lakes.

The material was analysed for the three cVMS octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) by the Department of Applied Environmental Science, Stockholm Universitet (ITM). In addition, lipid or organic carbon content in biota or sediments, respectively, and chlorinated organic contaminants (PCBs and DDT) in Mjøsa and Randsfjorden, and brominated flameretardants (PBDE) in Mjøsa, were analysed by the Norwegian Institute for Water Research (NIVA), to enable comparison of the food web biomagnification across chemicals. Dietary descriptors (stabile nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes) were analysed at the Institute for Energy Technology (IFE) for evaluation of relative trophic position in the food web and carbon source.

The previously reported high D5 levels and food web biomagnification in Mjøsa from 2010 were confirmed by the 2012 samples. The lakes that receive discharge from WWTPs had higher cVMS concentrations in both animals and sediments compared to the remote reference lake, Femunden, a lake with minor human impact in which most cVMS levels were below the limit of quantification in the analysed samples. cVMS were found in grab samples of effluent water from WWTPs, while D5 and D6 were quantified in surface sediments from Mjøsa and Randsfjorden. D4 was below the limit of quantification (LOQ) in all sediment samples, and D6 was below the LOQ in some sediment samples. Surface sediments of Randsfjorden and Mjøsa show high spatial variation in cVMS concentrations, with highest concentrations near Brandbu and Gjøvik, respectively. Due to the large difference in cVMS levels between the lakes, the presence of cVMS in effluent water, and the large spatial variation within the lakes with waste water treatment plants, local sources are the likely major input to the lakes, rather than long range transport.

Whereas D5 and D6 concentrations within the pelagic food web were significantly correlated with biomagnifying legacy contaminants (e.g. PCB-153, *p,p'*-DDE), D4 did not correlate with either D5 or D6 nor with PCB-153 or *p,p'*-DDE. Benthic feeding fish (perch, whitefish, burbot) had lower cVMS concentrations than pelagic fish at comparable trophic levels.

D5 and D6 biomagnified in the pelagic food web of Mjøsa and Randsfjorden with trophic magnification factors (TMF) for D5 of 2.9 (95% Confidence Interval (CI): 2.1-4.0) and D6 TMF 2.3 (CI: 1.8-3.0). D4 was below the LOQ in the majority of samples, and had substantially lower biomagnification than for D5 and D6. The cVMS TMFs did not differ between the lakes, whereas the legacy POP TMFs were higher in Mjøsa than Randsfjorden. Whitefish had lower cVMS bioaccumulation compared to legacy POPs, and affected the TMF significance for cVMS,

but not for POPs. TMFs of D5 and legacy contaminants in Lake Mjøsa were consistent with those previously measured in Mjøsa.

The present study has documented the potential of D5 and D6 to biomagnify in pelagic food webs, whereas D4 seems to be subject to trophic dilution.

2. Sammendrag

Tittel: Siloksaner i ferskvanns næringsnett – et studium av tre innsjøer i Norge
År: 2013
Bidragsytere: Katrine Borgå (NIVA), Eirik Fjeld (NIVA), Amelie Kierkegaard (ITM), Jarl Eivind Løvik (NIVA), Sigurd Rognerud (NIVA), Andreas S. Høgfeldt (NIVA), Kine Bæk (NIVA), Michael S. McLachlan (ITM)
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Som et ledd i Klifs screening av nye miljøgifter, har NIVA sommeren og høsten 2012 samlet inn prøvemateriale av det pelagiske næringsnettet i Mjøsa, Randsfjorden og Femunden. Formålet var å vurdere kilder av volatile (flyktige) sykliske metyl siloksaner (cVMS) i disse innsjøene, deres nivåer og biomagnifisering i næringsnettet med ørret som topp-predator. I tillegg til fisk og dyreplankton fra de frie vannmassene, ble det samlet inn bunnfisk fra Mjøsa, renset avløpsvann fra renseanlegg ved Mjøsa og Randsfjorden, vannprøver fra Mjøsa, samt overflatesediment fra alle tre innsjøer.

Materialet ble analysert for de tre sykliske siloksanene oktametylsyklotetrasiloksan (D4), dekametylsyklopentasiloksan (D5), og dodekametylsykloheksasiloksan (D6) av Department of Applied Environmental Science, Stockholms Universitet (ITM). Innhold av lipider i biota og organisk karbon i sedimenter, og konsentrasjoner av klorerte organiske miljøgifter (PCB og DDT) i prøver fra Mjøsa og Randsfjorden, samt bromerte flammehemmere (PBDE) i Mjøsa, ble analysert av NIVA for å kunne sammenlikne biomagnifisering i næringsnettet mellom kjemikalier. Stabile nitrogen ($\delta^{15}N$) og karbon ($\delta^{13}C$) isotoper ble analysert av Institutt for Energiteknikk (IFE) for bestemmelse av karbonkilde og plassering i næringsnettet.

Dette studiet fra 2012 bekreftet de høye D5 nivåene og biomagnifisering i næringsnettet funnet i Mjøsa i 2010. Innsjøene som mottar avløpsvann fra renseanlegg hadde høyere cVMS nivå i både dyr og sedimenter sammenliknet med referanseinnsjøen, Femunden, som har lav menneskelig påvirkning og hvor de fleste cVMS var under kvantifikasjonsgrensen i de analyserte prøvene. cVMS ble funnet i stikkprøver av renset avløpsvann fra renseanlegg, og D5 og D6 ble funnet i overflatesediment fra Mjøsa og Randsfjorden. D4 var under kvantifikasjonsgrensen i alle sedimentprøvene, og D6 var under i noen sedimentprøver. D5 og D6 konsentrasjoner i overflatesediment fra Randsfjorden og Mjøsa viste høy geografisk variasjon, med høyeste konsentrasjoner i nærheten av henholdsvis Brandbu og Gjøvik. Gitt den store forskjellen i cVMS mellom innsjøer med og uten menneskelig påvirkning, forekomsten av cVMS i renset avløpsvann, og den store geografiske variasjonen cVMS nivåer innen innsjøer med renseanlegg, antas lokale kilder å være hovedopphav til cVMS i innsjøene, og ikke langtransporterte prosesser.

Mens D5 og D6 nivåene i det pelagiske næringsnettet korrelerte signifikant med kjente biomagnifiserende miljøgifter (f.eks. PCB-153, *p,p'*-DDE), korrelerte D4 hverken med D5, D6, PCB-153, eller *p,p'*-DDE. Bunnfisk (abbor, sik og lake) hadde lavere cVMS konsentrasjoner enn pelagisk fisk på tilsvarende trofiske plassering i næringsnettet.

D5 og D6 biomagnifiserte i det pelagiske næringsnettet i Mjøsa og Randsfjorden med trofisk magnifikasjonsfaktor (TMF) for D5 på 2,9 (95 % konfidenseintervall KI: 2,1-4,0), og D6 TMF på 2,3 (KI: 1,8-3,0). D4 hadde flertallet av prøvene under kvantifikasjonsgrensen i både Mjøsa og Randsfjorden, og hadde lavere biomagnifisering enn D5 og D6. Trofisk magnifikasjonsfaktor for cVMS var lik mellom innsjøene, mens den for klorerte organiske miljøgifter var høyere i Mjøsa enn i Randsfjorden. Sik hadde lavere bioakkumulering av cVMS sammenliknet med klorerte og brommerte organiske miljøgifter. Dette innvirket på signifikansen av TMF for cVMS, men ikke for persistente organiske miljøgifter. TMF for D5 og POPs I Mjøsa var konsistente med de som tidligere er dokumentert fra Mjøsa.

Screeningen av siloksaner i norske innsjøer i 2012 har dokumentert at D5 og D6 kan biomagnifisere i pelagiske næringsnett, mens D4 antagelig er utsatt for trofisk fortynning.

3. Introduction

Cyclic volatile methylsiloxanes (cVMS) have been identified as emerging contaminants of concern due to their predicted persistence and bioaccumulative characteristics[1]. Siloxanes are produced in high volumes, and have several uses such as in personal care and biomedical products, consumer products such as car polish and waxes, and as additives in fuel[2]. The three cVMS octamethylcyclotetrasiloxane (D4 CAS no. 556-67-2), decamethylcyclopentasiloxane (D5 CAS no. 541-02-6), and dodecamethylcyclohexasiloxane (D6 CAS no. 540-97-6) (Figure 1) have been found to accumulate in biota[3-5] , but to a varying degree dependent on chemical, organism, and with large variation between studies. Based on the REACH criteria, D4, D5 and D6 are classified as very bioaccumulative (vB)[6-8]. Recent development and improvement of analytical quantification methods has resulted in increasing measurement in environmental matrices, including biota[9,10]. Thus, whereas previous assessment of cVMS behavior in the environment was based on model predictions[1,11] and laboratory tests[6,7], recent studies allow an interpretation of the persistence and bioaccumulation from environmental samples[3,12].

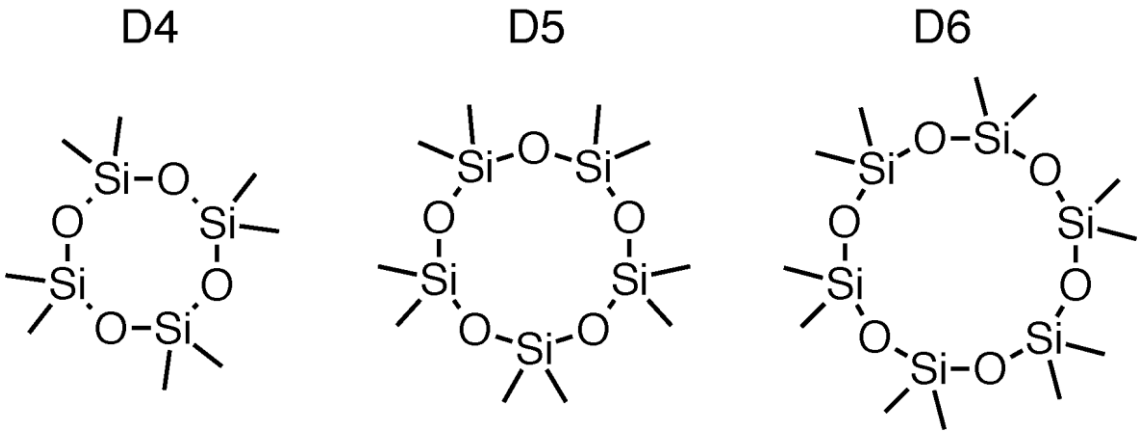


Figure 1. Molecular structure of D4, D5, and D6, the cyclic volatile methylsiloxanes included in the present study.

The bioaccumulation of chemicals in an organism from, and relative to, the diet is currently assessed by biomagnification factors (BMF) or trophic magnification factors (TMF)[13,14]. Whereas the BMF considers specific predator-prey relationships, the TMF is an estimation of the average change in contaminant concentrations, normalized for fugacity capacity,when moving one trophic level up the food web[15]. For contaminants with high octanol-water partitioning coefficient (K_{OW}), such as cVMS (log Kow 6.98 for D4, 8.07 for D5, and 8.87 for D6[16]), lipid normalization reflect the fugacity capacity normalized concentrations. TMF was suggested as the most conclusive measure of bioaccumulation of chemicals in biota that have a multitude of food choices and thus exposures to contaminants[13]. TMF is currently estimated from empirical data[15], but there is still need for improvement of the scientific understanding of TMF, how to best estimate and interpret it [15,17] [18]. The European Community Regulation on chemicals and their safe use (REACH) recently added BMF and TMF to Annex XIII as metrics that can be used in a weight of evidence assessment of bioaccumulation[19].

There are presently few studies of empirical food web magnification (TMF) of cVMS. There is currently only one study published in the peer review literature[20]. This study reported D5 TMF greater than 1 in the pelagic food web of Lake Mjøsa, Norway, indicating food web biomagnification[20]. This is in line with previous studies of D5 biomagnification[3], but in contrast with the silicon producing industry's own reports on other food webs and ecosystems, which report TMFs less than 1 for all cVMS[21,22]. The Lake Mjøsa study reported

surprisingly high cVMS levels[20], with concentrations comparable to levels reported from the inner Oslofjord of Norway[2,23], a highly populated area close to the capital of Norway. A study from Swedish lakes suggest that the sediment and fish contamination of cVMS is correlated to the wastewater treatment plant (WWTP) effluent load in the respective lakes[24]. The inner Oslofjord is the recipient for WWTP effluent from approximately 10 times greater person equivalents than Lake Mjøsa. As the Mjøsa study[20] is presently the sole study reporting significant food web biomagnification of cVMS, and because it reports surprisingly high cVMS concentrations relative to the WWTP effluent load, a closer investigation is needed to assess if this result is representative or atypical for Norwegian lake ecosystems with food webs leading to brown trout (*Salmo trutta*) as a top predator.

The present study repeated the study of the pelagic food web in Lake Mjøsa from 2010, in addition to including a comparable lake in the vicinity (Lake Randsfjorden) and a reference remote lake (Lake Femunden) far from any known sources of contaminants (Table 1. Figure 2). The aims of the present study were to increase our understanding of the food web biomagnification of cVMS, to obtain information on the potential sources of cVMS to the investigated lakes, and to identify differences in cVMS levels, biomagnification and sources between lakes. In addition to samples from the pelagic food web, samples of benthic fish, WWTP effluent, surface sediment and lake water were collected. The food web bioaccumulation behavior of cVMS was compared to that of the legacy persistent organic pollutants (POPs) dichlorodiphenyldichloroethylene (p,p'-DDE), polychlorinated biphenyls (PCB) and polybrominated diphenyl ethers (PBDE).

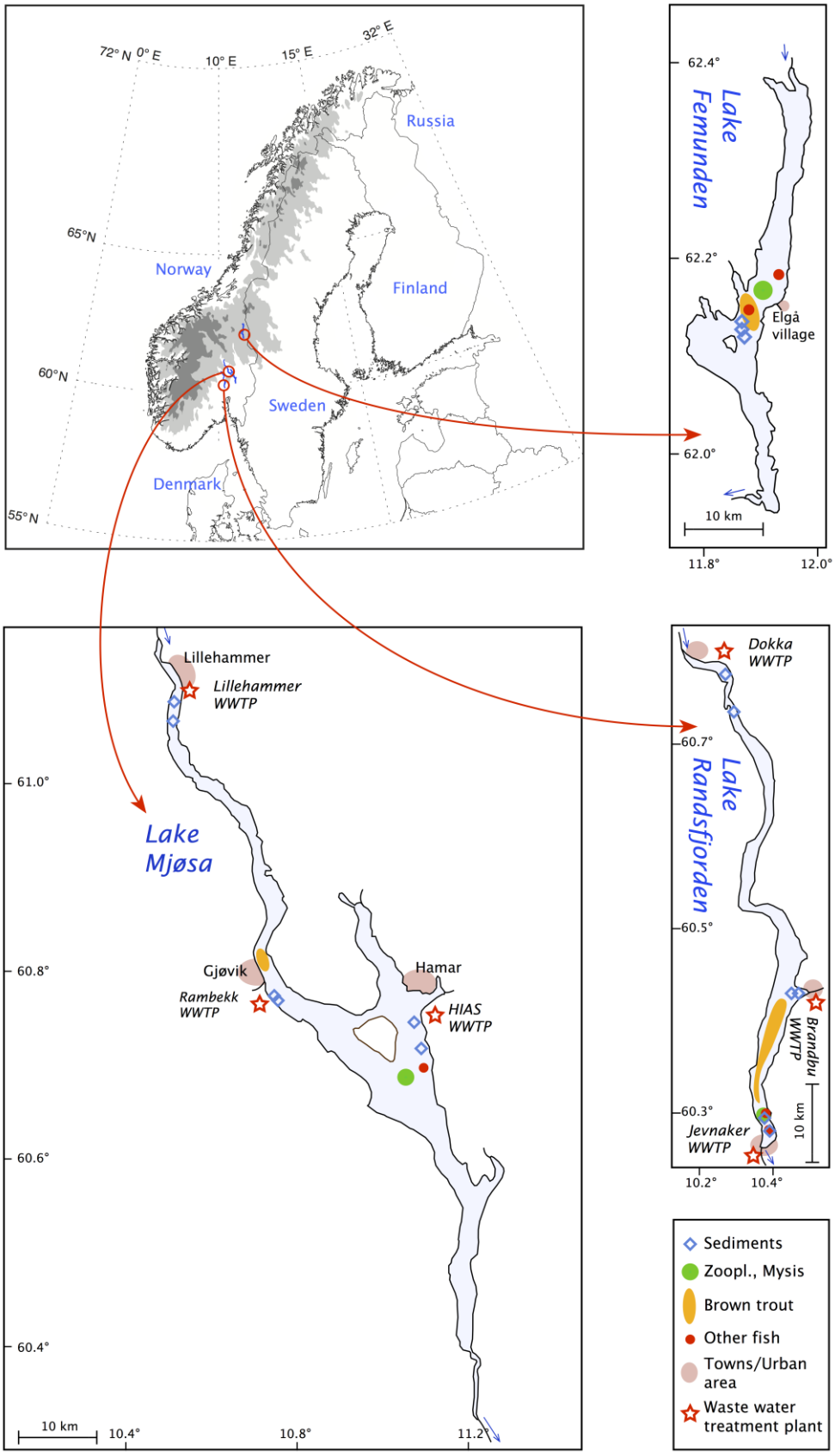


Figure 2. Map of a) Lake Mjøsa, b) Lake Randsfjorden, and c) Lake Femunden with sampling sites, major urban areas and waste water treatment plants (WWTP).

4. Materials and methods

4.1 Selected lakes and sampling description

The samples were collected in Lake Mjøsa, Lake Randsfjorden, and Lake Femunden during July-September 2012 (**Figure *2***, Table S 1). Based on person equivalents (**Table *1***), Mjøsa is subject to high to moderate human impact, Randfjorden to moderate human impact, whereas Femunden is remote from human activity. All three study lakes are deep and contain well-defined pelagic food webs including zooplankton, planktivorous fish and brown trout as top predator. The main food web difference between the lakes is that Lake Mjøsa includes *Mysis relicta* in the invertebrate community, vendace among the planktivorous fish, and exclcudes Arctic char as top predator. Whitefish is assumed to replace vendace in the pelgaic food web of Randsfjorden and Femunden, whereas it is benthic feeding in Mjøsa.

Table 1. Information on lakes included in the study

Lake	Mjøsa	Randsfjorden	Femunden
Position	60°53′N 10°41E	60°23′N 10°23′E	62°21′N 11°57′E
Length (km)	117	75	60
Volume (km^3)	65	7,3	6
Area (km^2)	362	134	203
Maximum depth (m)	453	120	153
Person equivalents[a]	206000	28500	200

a. Estimated from maps with discharge and wastewater treatment plants for the different regions

Representatives of the food webs of the respective lakes were collected according to protocols as described in Borgå et al.2012[20]. Sediment, effluent water from WWTPs, water and benthic fish (whitefish *Coregonus lavaretus*, perch *Perca fluviatilis*, burbot *Lota lota*) samples were collected as described in brief below.

In Mjøsa, zooplankton from the epilimnion (Cladocerans *Daphnia galeata, Bosmina longispina*) and hypolimnion (Copepods *Limnocalanus macrurus*), *Mysis relicta*, vendace, and smelt (*Osmerus eperlanus*) were collected mid-lake south of Helgøya, and trout close to Gjøvik. Although the trout in Mjøsa were sampled close to the Gjøvik area (Figure 2), the trout represent a larger geographic area as it uses the entire of Mjøsa in its search for food[25]. In Randsfjorden zooplankton from the epilimnion (*D. galeata,* Copepods *Eudiaptomus gracilis*) and hypolimnion (*D. galeata* and Copepods *L. macrurus, Heterocope appendiculata*), whitefish, smelt and trout were collected mid-lake, south of Brandbu. In Femunden, zooplankton from the epilimnion (Cladocerans *D. galeata, B. longispina*), whitefish, Arctic char and trout were collected in the southern basin (Figure 2). Zooplankton (epilimnic and hypolimnic) and Mysis were collected with vertical net hauls, and fish were caught using surface and bottom gill nets, traps, and angling.

Each sample of fish consisted of skinless filets from one individual fish, with the exception of small smelt from Mjøsa and Randsfjorden, where 5-6 skinless filets were pooled, and with the exception of burbot where liver was analysed in addition to filets. Brown trout from Mjøsa

was stored frozen whole until sample preparation (dissection of skinless filet) at NIVA, whereas fish from Femunden and Randsfjorden were dissected fresh. The dissected samples were stored frozen in preheated glass jars.

Precleaned field blanks (passive samplers: polyester pouches containing ~60 mg ENV+) were exposed to air and handled in the same manner as the biotic samples, as described previously[20]. After exposure the field blanks were wrapped in aluminum foil and kept frozen in sealed PE bags until analysis.

Sediment samples from the surface layer (upper 0-1 cm) were collected in stainless steel tubes (inner diameter 85 mm) using a gravity corer equipped with a core catcher. The sampling was performed according to established protocols[26]. Sediments were collected in areas with stable accumulation sediments, if possible close to the WWTP discharge area (Figure 2). Each sample consisted of three pooled cores from each station. Deeper pre-industrial sediments were collected from Mjøsa (40-42 cm) and Randsfjorden (30-32 cm Jevnaker, 40-42 cm Fluberg) to serve as reference to the surface sediments (Table 3).

In Mjøsa, high volume water samples were collected using a pre-programmed in situ water sampler at ca 15 m depth (Table 3). The in situ water sampler was custom made for NIVA, and includes a filter holder in stainless steel for collection of the particulate phase (for analysis of cVMS in the present study) and a separate chamber for polyurethan foam (PUFs) for collection of the water dissolved fraction (for analysis of PCBs in the present study). Filters for sampling were pre-heated, and the PUFs were cleaned with solvents and stored in aluminum foil prior to sampling.

Grab samples of effluent were collected directly from the outlet drain of 3 WWTPs in Mjøsa and 3 in Randsfjorden (Table 3).All of the samples were collected on the same day. As far as possible, the bottle (2.5 L) was topped to avoid air space below the cap. Aluminum foil sealed the bottle under the cap.

To reduce the risk of contamination during sampling, all sample preparation was conducted outdoors, i.e., the material was outdoors from the time of sampling until it was freezer-ready for storage until shipment to the Department of Applied Environmental Science (ITM, Stockholm University, Sweden) for analysis of cVMS in October-November 2012. All personell involved in the sampling (NIVA personnel, WWTP personell, and local fishermen) avoided personal care products at least 24 h prior to field work. All large surfaces (e.g. tubs for gill nets, gill nets after retrieval before the fish were collected, the chopping board for sample preparation and fish dissection) were covered in aluminum foil. All utensils (tweezers, knife, scalpel) were made of strnaless steel. All sampling equipment in contact with any sampling matrix was cleaned with solvents (acetone/methanol) between samples. Contact with plastics was avoided. The samples were stored in pre-heated glass jars sealed with aluminum foil under the lid. All biota samples and water samples (GFF and PUF) were stored frozen until chemical analysis. Sediment samples from Lake Femunden were stored frozen (-20°C), while sediment samples from Randsfjorden and Mjøsa were stored cooled (4ºC) until analysis. Effluent water samples were stored cooled until analysis. More details on sampling procedures can be found in the Supporting Information.

Immediately after collection, the material was divided into sub-samples for analysis of cVMS, legacy contaminants (halogenated POPs), and stable isotopes of nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$). Biota and sediment samples were analysed for cVMS, PCBs, PBDEs, stable isotopes, and lipid content or organic carbon, respectively. Effluent and suspended particulate matter (filter) samples were analysed for cVMS, whereas the dissolved water samples (PUF) were only analysed for PCBs.

4.2 Chemical analysis of cVMS

The samples were analysed for cVMS (D4, D5 and D6) at Stockholm University using a modified version of a published purge and trap method[9]. To improve the repeatability and analyte recovery of that method, the samples were extracted using an organic solvent containing the surrogate standards ^{13}C-D4, ^{13}C-D5 and ^{13}C-D6. Fish tissue, zooplankton, GFF filters, and effluent water were extracted with dichloromethan (DCM), while sediment was extracted with a mixture of DCM and acetone. The extracts were transferred to a flask containing glass beads (4 mm) and a stir bar. Purified nitrogen was led into the flask and exited via a cartridge containing 10-15 mg of Isolute ENV+ (Biotage AB, Sweden). The flask was purged with nitrogen while stirring for 2.5-3 h until all solvent had evaporated. The purging was continued for another 2 h with the heating turned on giving a temperature of ~ 72°C. Then the ENV+ cartridge was removed and eluted with 0.8 mL n-hexane. Tetrakis(trimethylsiloxy)silane, M4Q, was used as the volumetric standard. The purified extract was analysed using GC/MS as described in Kierkegaard et al.[9]. Extraction and sample preparation were performed in a clean air cabinet under a laminar flow of filtered air. A detailed description of the method is provided in the Supporting Information (text and Table S 3 - Table S 7).

In addition to procedural blanks and field blanks, an internal matrix control (homogenate of herring from the Baltic Sea for biota samples and a sediment sample from Lake Mjøsa for abiotic samples) was analyzed with each round of 8 samples. The limit of quantification (LOQ) for biota samples was set to the mean plus 10 times the standard deviation of the procedural blanks (Table S 8). For sediment the LOQs were based on the reference sediments (three times the maximum quantity measured in the reference sediments from Randsfjorden, n=2) because there were too few sediment blanks (Table S 8). The cVMS results were not blank corrected.

4.3 Chemical analysis of halogenated POPs

The biota samples from Lake Mjøsa and Lake Randsfjorden were analysed for PCBs and chlorinated pesticides. The Lake Mjøsa samples were also analysed for PBDE. The analysis were conducted at NIVA based on established methods for extraction[27] using cyclohexane and isopropanol. Extracts were analyzed on GC-EI-MS operating in single ion monitoring (SIM). Quantification of individual compounds was done using the relative response of surrogate internal standard and comparing that to a calibration curve[28]. More detailed description is found in the Supporting Information (tekst and Table S 9).

The sediment and PUF samples were spiked with the same internal standards as the biota samples, and then extracted twice by shaking with DCM and then centrifuged (sediments), or by soxhlet using 5% ether in n-hexane (PUFs).[29] The organic phase was then dried using sodium sulphate (Na_2SO_4), transferred to a new vial and solvent exchanged with isohexane. The cleanup procedure was the same procedure as for the biota samples.

The extraction of total lipids by cyclohexane and isopropanol followed the recommended method for the revised OECD 305 guideline for determination of bioconcentration factor in fish[30], with results well within the acceptable criteria for the Quasimeme ringtest for lipid determination (Table S 10).

4.4 Analysis of trophic descriptors

Stable isotopes of nitrogen (δ15N)and carbon (δ13C) were analysed to assess the relative trophic position and the dominant carbon source of the organisms, respectively[19]. All biota and sediment samples analysed for cVMS were also analysed for $\delta^{15}N$ and (δ13C) at the Institute for Energy Technology (IFE-Kjeller) according to standard protocols[31]. Lipids and carbonate were not removed or extracted from samples prior to analysis of the isotopic signature.

4.5 Data treatment

The co-occurrenceof chemicals in the various samples was investigated by pairwise correlation (in which n may vary depending on chemical). As more Mjøsa zooplankton sub-samples were analysed for cVMS than for PCB, the cVMS data were averaged to obtain a similar n to legacy POPs. As the PBDE analysis in one hypolimnion zooplankton was considered uncertain (irregular chromatogram), this sample was excluded in he correlations and further data treatment with PBDE-47 and PBDE-99.

Pelagic food web biomagnification was assessed by estimating trophic magnification factors (TMFs)[15,20]. In brief, the relative trophic level (TL) of each sample (consumer) was calculated from $\delta^{15}N$ using an enrichment factor (ΔN) of 3.4 ‰[17,18,26]. The lowest epilimnion zooplankton $\delta^{15}N$ for the respective lake was defined as the baseline primary consumer of trophic level 2 ($\delta^{15}N$ primary consumer) (Equation 1).

$$TL_{consumer}=((\delta^{15}N_{consumer}- \delta^{15}N_{primary\ consumer})/ \Delta N)+2 \quad (1)$$

TMFs were estimated as the slope (b) of the lipid normalized contaminant concentration ($[Contaminant]_{LW}$) regressed onto the TL, analysing for interaction with lake to test if the TMF differ between lakes (Equation 2 and 3). Benthic fish and abiotic samples were not included in the regression as they are not components of the pelagic food web.

$$Ln[Contaminant]_{LW} = Ln\ a + bTL + c\ Lake + d\ TLxLake \quad (2)$$

For chemicals with non-significant interaction between trophic level and lake, the term was removed from the regression, and the TMF estimated from the slope (b) (Equation 3). For chemicals with significant interaction (only PCB-180), the TMF were estimated separate for each lake.

$$TMF = e^{b} \quad (3)$$

TMFs were calculated for cVMS and selected legacy POPs. For cVMS, more than 80% of the data were quantified above the LOQthroughout the food web (Table S 12). Thus, uncensored cVMS data were included in the data analysis, using estimated values below LOQ but above LOD for cVMS. Data treatment and estimation of TMF based on original uncensored data is preferable to censored data by replacement of values below LOQ with a fixed or random value[15]. As POPs were quanfitied using LOD as the cut off, values below LOD were censored and replaced by the samples and chemical specific LOD. When data were quantified below the LOQ for cVMS, or LOD for POPs, these data were generally comparable to the data above the LOQ or LOD, respecitvely. Thus the data were included in the analysis, and if more than 50% of data were below the LOQ, or LOD, respectively, the TMFs were estimated and presented for comparison. They are labeled with an asterix for recognition (TMF*) to denote that they have a greater uncertainty. TMFs were not estimated for Femunden, as the whole

food web was not analysed for cVMS, following the observations of low levels in trout and sediments.

In Randsfjorden, one hypolimnion zooplankton sample (R8) was identified as a multivariate outlier and was excluded from the dataset for all data analysis.

5. Results and Discussion

5.1 cVMS QA/QC results

5.1.1 Control samples and repeatability of analysis

The repeatability of the method was assessed using the matrix control samples analysed during each round of extractions. The relative standard deviation (RSD) of the control samples was between 8% and 11% for D5 and D6 in both the sediment and the herring matrices (Table S 6), which is a good result, particularly in light of the low D5 and D6 concentrations in these samples. The RSD was higher for D4 in herring, which can be attributed to the very low levels in the matrix control samples (a factor of 2 above the LOQ). D4 was below the LOQ in the sediment.

5.1.2 Limit of Quantification and comparison to blank

The amount of D4, D5 and D6 was above the LOQ in 23%, 98% and 58%, respectively, of the biota samples (total biota samples n = 91), and 0%, 80% and 73%, respectively, of sediment samples (total sediment samples n = 18), depending on the lake (Table S 11). In the reference lake, Femunden, all cVMS were below LOQ in all samples except for a few trout in which D5 was above the LOQ. Low cVMS levels in Femunden had been anticipated and therefore sediments and samples of the top predators brown trout and arctic char were analysed first. Due to the low levels found, the remaining samples collected in Femunden (zooplankton, whitefish, arctic char) were not analyzed.

All of the effluent water samples contained all cVMS above the LOQ, with the exception of D6 in the sample from Lillehammer, Mjøsa (Table S 11). For the filter (GFF) samples, an error in the field unfortunately resulted in no field blank being available. Since it could therefore not be excluded that these samples were contaminated, the measured concentrations were designated “<”.

In biotia samples, the total content of D5 and D6 in the field blanks from Lake Mjøsa was in all cases low compared to the total amount extracted from the samples above LOQ (ratio >4.4 up to 3499, Table S 11). For D4 the difference between field blanks and samples was lower; still 11 of 21 samples from Mjøsa contained more than 5 times the amount in the field blank (total range 3-94). For Randsfjorden, although more samples were close to or below the LOQ for D4 and D6, the biotia sample to field blank ratio for D5 was greater than 5 for all but 6 samples (Table S 11). In Femunden only D5 was quantified above the LOQ in trout, with values 15-23 times higher than the field blank.

Table 2. Species collected in Norwegian lakes in 2012 and analysed for trophic descriptors (stable isotopes of nitrogen ($\delta^{15}N$), and carbon ($\delta^{13}C$), ‰), cyclic volatile methylsiloxanes (cVMS, ng/g lipid weight), and persistent organic pollutants (POPs, ng/g lipid weight)[a].

Species	Biometry	Length (cm)		Weight (g)		SI	$\delta^{13}C$		$\delta^{15}N$		Trophic level		Lipid	Lipid %	
	N	Mean	SE	Mean	SE	N	Mean	SE	Mean	SE	Mean	SE	N	Mean	SE
MJØSA															
Zooplankton Epilimnion						4	-31.6	0.3	7.7	0.0	2.0	0.0	4	0.72	0.04
Zooplankton Hypolimnion						5	-33.5	0.7	9.7	0.6	2.6	0.2	5	3.50	1.36
Mysis						5	-30.9	0.4	10.5	0.3	2.8	0.1	5	2.54	0.55
Vendace	7	22.0	0.2	67.9	2.4	7	-29.6	0.3	13.9	0.1	3.9	0.0	7	1.17	0.11
Smelt, small[b]	35	10.8	0.1	5.9	0.1	5	-28.9	0.1	13.5	0.2	3.8	0.1	5	1.03	0.03
Smelt, large	5	20.5	1.1	48.6	8.5	5	-27.6	0.2	15.8	0.1	4.4	0.0	5	1.27	0.23
Brown trout	5	56.4	2.3	2054	306	5	-28.3	0.5	15.6	0.1	4.4	0.0	5	2.92	0.56
Whitefish	5	32.3	1.5	241	89.6	5	-27.0	0.3	13.0	0.3	3.6	0.1	5	0.84	0.19
Perch	6	26.2	2.1	224	148	6	-26.2	0.4	14.3	0.3	4.0	0.1	6	0.69	0.04
Burbot, liver	6	31.7	1.5	206	72.1								6	40.5	2.96
Burbot, muscle	6	31.7	1.5	206	72.1	6	-25.6	0.5	15.7	0.2	4.4	0.1	6	0.69	0.02
RANDSFJORDEN															
Zooplankton Epilimnion						4	-32.3	0.1	6.3	0.0	2.0	0.0	4	0.73	0.03
Zooplankton Hypolimnion						3	-36.4	0.8	9.5	0.9	3.0	0.3	3	1.65	0.48
Whitefish	10	24.6	2.3	157	31.3	9	-28.2	1.1	10.4	0.2	3.2	0.1	9	1.24	0.22
Smelt[b]	25	12.5	0.1	10.3	0.2	5	-30.4	0.1	11.3	0.2	3.5	0.1	5	1.97	0.20
Brown trout	5	40.8	2.5	862	180	5	-28.4	0.6	12.2	0.3	3.8	0.1	5	0.68	0.09
FEMUNDEN															
Arctic char	1	32.2		321		1	-26.1		6.9				1	1.00	
Brown trout	6	38.1	2.0	550	92.2	6	-23.2	0.3	9.3	0.5			6	0.74	0.16

Table 2. cont.

Species	D4		D5		D6		POPs	PCB-153		PCB-180		*p,p'*-DDE		PBDE-47		PBDE-99	
	N	Mean	SE	Mean	SE	Mean	SE	N	Mean	SE	Mean	SE	Mean	SE	Mean	SE	Mean	SE
MJØSA																		
Zooplankton Epi	3	<46		342	± 33	<48		2 (1)	10	± 0	<8		38	± 1	(11)		(1)	
Zooplankton Hypo	4	36	± 3	1664	± 296	48		3	20	± 1	5	± 1	67	± 2	35	± 5	8	± 1
Mysis	4	53	± 13	927	± 116	59	± 13	4	25	± 6	<4		82	± 17	34	± 6	10	± 2
Vendace	7	81	± 8	14160	± 2446	786	± 117	7	333	± 70	69	± 15	890	± 167	415	± 83	146	± 32
Smelt, small[b]	5	<24	± 3	3533	± 224	184	± 21	5	73	± 11	9.6	± 4	216	± 29	105	± 12	16	± 1
Smelt, large	5	<17	± 3	5256	± 737	325	± 55	5	337	± 50	50.1	± 11	853	± 120	552	± 89	16	± 4
Brown trout	5	27	± 7	5629	± 1041	285	± 45	5	322	± 81	54	± 13	837	± 208	659	± 204	68	± 18
Whitefish	5	<38		1027	± 325	<122		5	308	± 89	47	± 18	807	± 193	789	± 161	141	± 31
Perch	6	<29		403	± 47	<66		6	335	± 107	41	± 14	655	± 210	362	± 218	117	± 34
Burbot, liver	6	44	± 7	5296	± 1019	260	± 73	6	415	± 78	48	± 9	793	± 106	1406	± 267	78	± 13
Burbot, muscle	6	<61		1507	± 244	174	± 21	6	113	± 17	15	± 2	217	± 28	376	± 74	22	± 2
RANDSFJORDEN																		
Zooplankton Epi	4	<34		251	± 5	<37		4	8	± 0	<7		21	± 2				
Zooplankton Hypo	3	51	± 2	2251	± 39	48	± 10	3	26	± 7	8	± 2	51	± 13				
Whitefish	9	<19		112	± 39	<30		9	27	± 5	7	± 1	47	± 6				
Smelt[b]	5	<11		969	± 71	58	± 9	5	9	± 1	<3		23	± 2				
Brown trout	5	16	± 3	2579	± 806	132	± 31	5	59	± 4	14	± 1	113	± 8				
FEMUNDEN																		
Arctic char	1	<10		<20		<40												
Brown trout	6	<40		39	± 14	<80												

a. For cVMS or POPs with more than 50% of values below LOQ or LOD, respectively, for a given species and lake, the estimated mean include all values (also those <LOQ), and the estimate is flagged by <.

b. 5-6 small smelt filets were pooled into one sample for contaminant and stable isotope analyses. All other fish were analysed individually.

Table 3. Abiotic samples collected in Norwegian lakes in 2012; surface sediments (0-1 cm), deeper pre-industrial sediments for reference, effluent water from wastewater treatment plants, water particulate phase and water dissolved phase. All samples are reported individually, except water which is an average of 3 samples. Sediment samples were analysed for total organic carbon (TOC, %), stable isotopes of nitrogen ($\delta^{15}N$, ‰), and carbon $\delta^{13}C$, ‰), cyclic volatile methyl siloxanes (cVMS), and persistent organic pollutants (POPs)[a,b].

MATRIX / Lake	Date	Area	Depth (m)	TOC	$\delta^{13}C$	$\delta^{15}N$	D4	D5	D6	PCB-153	PCB-180	*p,p'*-DDE	PBDE-47	PBDE-99
SEDIMENTS (ng/g TOC)														
Mjøsa	Sept 3rd	Ottestad/ Gillundstranda	120	2.16	-26.1	-2.6	<125	<8	<15	<6.9	<6.9	<4.6	<9.3	<9.3
	Sept 5th	Ottestad	30	3.73	-26.7	2.9	<46	154	166	61.7	21.7	131	<5.4	<11
	Sept 5th	Lillehammer	30	3.22	-27.8	0.04	<9.3	210	95	6.2	<2.5	9.3	<6.2	<9.3
	Sept 5th	Lillehammer	80	3.56	-28.1	0.8	<14	729	264	6.5	<2.3	7.3	9.8	<11
	Sept 5th	Gjøvik	28	5.61	-27.7	2.4	<66	5086	602	32.1	13.0	14	11.9	37.4
	Sept 5th	Gjøvik	120	5.57	-26.7	2.1	<120	6022	725	104.1	44.9	29	9.3	18.0
	Sept 5th	Lillehammer-reference	80	3.31	-26.5	-0.8	<9.1	28	<30	45.3	13.3	42	<6.0	<6.0
Randsfjorden														
	Aug 30th	Jevnaker	65	2.65	-26.8	1.4	<45	37	32	23.0	9.1	67.9		
	Aug 30th	Jevnaker	70	2.51	-26.6	1.9	<155	32	40	8.0	4.0	8.8		
	Aug 30th	Brandbu	28	6.55	-27.8	4.7	<17	795	152	24.4	10.4	64.1		
	Aug 30th	Brandbu	30	5.79	-27.3	3.7	<28	2540	385	16.2	7.4	32.8		

Table 3. cont.	Date	Area	Depth (m)	TOC	$\delta^{13}C$	$\delta^{15}N$	D4	D5	D6	PCB-153	PCB-180	*p,p'*-DDE	PBDE-47	PBDE-99
Randsfjorden cont.														
	Aug 30th	Fluberg	41	4.77	-28.1	2.1	<6.3	126	93	7.5	<2.5	13.2		
	Aug 30th	Fluberg	42	5.89	-28.4	1.4	<80	193	133	<2.5	<1.7	4.4		
	Aug 30th	Jevnaker-reference	65	2.02	-26.6	1.9	17	5.6	7.6	<5.0	<5.0	<5.0		
	Aug 30th	Fluberg-reference	42	2.18	-27.2	-2.1	<64	8.6	9.6	4.6	4.6	<4.6		
Femunden														
	Aug 8-9th	Femunden	70	5.7			<16	<3.5	<11					
	Aug 8-9th	Femunden	75	5.7			<21	<8.8	<19					
	Aug 8-9th	Femunden	80	5.7			<7.0	<5.2	<12					
EFFLUENT WATER (ng/L)														
	Aug 22nd	Mjøsa Rambekk WWTP (Gjøvik)					8.9	82	12.1					
	Aug 22nd	Mjøsa Hias WWTP (Ottestad)					15	111	14.0					
	Aug 22nd	Mjøsa Lillehammer WWTP					7.7	29	<3.1					
	Aug 22nd	Randsfjorden Jevnaker WWTP					13	61	11					
	Aug 22nd	Randsfjorden Brandbu WWTP					27	351	11					
	Aug 22nd	Randsfjorden Dokka WWTP					24	368	9.3					

Table 3 cont.	Date	Area	Depth (m)	TOC	$\delta^{13}C$	$\delta^{15}N$	D4	D5	D6	PCB-153	PCB-180	*p,p'*-DDE	PBDE-47	PBDE-99
WATER PARTICULATE (ng/g TOC[c])														
Mjøsa	Sept 3rd	Ottestad/Gillundstranda					<171	<200	<143					
WATER DISSOLVED (pg/L)														
Mjøsa	Sept 3rd	Ottestad/Gillundstranda		mean						<0.9	<0.9	2	7	4
				SD								0.4	0.9	1.3

a. < = lower than limit of detection for PCBs and PBDEs.For cVMS the concentration measured in the sample is reported and designated with < if it is below the LOQ.
b. Empty cells indicates the parameter was not analysed
c. Based on 1 mg/L particles in the water column, from on turbidity measurements, and on the assumtion of 35% TOC in particles.

5.2 General trends in cVMS concentrations 2012

In both the food web and the sediments, Lake Mjøsa was more contaminated with cVMS than Lake Randsfjorden (Table 2, Table 3). In addition to a higher human population and thus higher discharge from WWTPs, Mjøsa supports more traffic, local industry and business, including garages, and agricultural activity compared to Randsfjorden. Of the cVMS, D5 dominated both biotic and abiotic samples, followed by D6 and D4 that were below LOQ in several samples.

The high levels of cVMS found in 2010 in fish from Mjøsa were confirmed in the present study. For instance, the D5 concentrations in trout ranged from 3000 ng/g lw to 9200 ng/g lw (Table 2). The D5 levels in Randsfjorden were lower, with concentrations in brown trout from 60 ng/g lw to 4900 ng/g lw. In the remote reference lake, Femunden, D5 concentrations in trout were low, ranging from <23 ng/g lw to 69 ng/g lw. The two orders of magnitude lower cVMS levels in Femunden biota compared to Mjøsa and Randsfjorden is in line with reports from Swedish lakes comparing perch in lakes receiving and not receiving discharge from WWTP.[24] The low cVMS levels in biota from the remote Femunden are also in line with a study of arctic char in remote Swedish lakes[9].

The cVMS concentrations in sediment displayed considerable variability. One sample, the deep water sediment from Ottestad/Gillundstranda (Mjøsa), contained very low D5 and D6 levels, only 7% compared to the shallower sample from the same area (Table 3). The fact that *p,p'*-DDE was not detected in this sample (despite high levels in the other sample from this area), and the low $\delta^{15}N$ level, suggested that this was disturbed sediment (e.g. from a slump or dumped material). The sediment was collected in an area with steep slope and strong currents, thus unstable sedimentation and absence of accumulation sediments is likely. Within each of the other areas there was good agreement between the two samples collected (median difference 67%). The low variability within sampling areas contrasted with high variability between sampling areas within a lake. The mean concentrations in the most contaminated sampling area of Lake Mjøsa were 36 and 4 times higher for D5 and D6, respectively, than the mean concentrations in the least contaminanted area. The corresponding values for Lake Randsfjorden were 48 and 7. Comparing the most contaminated sampling areas of the lakes with each other, the mean D5 and D6 concentrations were higher in Lake Mjøsa by a factor of 3.5 and 2.5, respectively. Smaller differences were observed when the least contaminated sampling areas of the two lakes were compared.

The dry weight normalized D5 concentrations in sediment from this work (Mjøsa: 2-400 ng/g dw; Randsfjorden: 0.1-150 ng/g dw) can be compared with other studies. The present highest sediment concentrations were 1-2 orders of magnitude lower than reported from water bodies highly impacted by wastewater such as the inner Oslofjord, Norway[32], Humber estuary[3], and other areas receiving effluent from WWTPs[3]. The highest D5 sediment concentrations (ng/g dw) were however comparable to those of other Nordic areas with moderate human impact[2], whereas the low concentration areas were comparable to those reported from the Arctic[4]. Thus, whereas the trout cVMS concentrations are comparable to those reported from the inner Oslofjord (high human impact), the cVMS concentrations in the sediments are more in line with those reported from moderate to low human impact areas.

All benthic feeding fish in Lake Mjøsa (burbot, perch and whitefish) had lower lipid normalized concentrations of cVMS in muscle compared to pelagic feeding fish at comparable trophic levels (trout, smelt and vendace, respectively) (Table 2). This may be due to lower exposure (lower fugacity) from sediment than in the water column, lower bioaccumulation at the first trophic levels of the food web, and/or lower biomagnification and enrichment from prey to predator (i.e. less efficient trophic transfer and retention) within the benthic compared to the pelagic food web.

The levels of legacy POPs were higher in fish from Mjøsa than in Randsfjorden, whereas the levels in zooplankton were comparable (Table 2). The observed levels of legacy POPs were comparable to recent studies from Mjøsa[33]. A previous comparison of Randsfjorden and Mjøsa, in 1998, also reported higher PCB levels (3 times) in Mjøsa trout compared to Randsfjorden trout[34].

5.3 Sources of cVMS

The area with highest cVMS concentration in sediments in Mjøsa was Gjøvik, and Brandbu in Randsfjorden (sediment sampling was approximately 1-5 km from the discharge area of the respective WWTPs). The higher levels in sediments close to Gjøvik compared to the other areas in Mjøsa, may be due to the its area close to a catchment area of a large industrial area including industry such as manufacturing and surface treatment of plastics for automotive industry, manufacturing of storage systems for alternative fuels, aluminum industry and junk yards. The river Hundselva passes through this industrial area and has its outlet north of Gjøvik. cVMS were found in effluent grab samples from the WWTPs of both Mjøsa and Randsfjorden. Effluent grab samples were collected from the WWTPs, rather than flow proportional samples, to avoid contamination of the sample and volatilization of cVMS. Thus, the effluent samples did not represent flow volume corrected concentrations, and could not be directly compared across WWTPs. Nevertheless, as they were collected from the WWTP outlet, they represent effluent averaged over all connected sources and at least several hours in time. The cVMS concentrations in effluent grab samples from Mjøsa WWTPs were highest and comparable at HIAS (close to Ottestad) and Rambekk (close to Gjøvik), and lowest at Lillehammer (Lillehammer). Earlier estimates of daily flow through the WWTPs, showed substantial variation during the year, with estimated daily flow of 6,000-40,000 m^3/day in Rambekk, 14,500-28,000 m^3/day in HIAS and 11,000-31,000 m^3/day at Lillehammer[35]. The highest sediment concentrations in Randsfjorden close to the Brandbu WWTP coincides with the relative denser population in this area. Although also Jevnaker has a dense population in Randsfjorden, the sediment samples were collected in the lake upstream for the WWTP discharge area, which is in the river outlet of the lake. The Jevnaker sediments therefore do not reflect the impact from the Jevnaker population.

As cVMS have high K_{OW} and high octanol air partitioning coefficient (K_{OA}), they are very hydrophobic and will tend to sorb to particles in the water column or to volatilize to the atmosphere. cVMS are therefore difficult to analyze in sirface water due to low dissolved water concentrations, risk of cross contamination, and risk of volatilization during storage of water. Nevertheless, in Lake Mjøsa the particulate phasewas collected on filters with an in situ pump. The estimated upper bounds of the OC normalized concentrations in the pelagic particulate matter (<170, <200 and <143 ng/g dw TOC for D4, D5 and D6, respectively) were similar to the concentrations measured in the near surface sediment at the same area (<125, 154 and 166 ng/g dw TOC) (Table 3). Considering that the level in the water column all represent upper limits, this suggest that the higher measured concentrations in pelagic fish compared to benthic fish at the same trophic level can not be explained by consistently higher exposure in the pelagic habitat compared to the benthic habitat. However, this should be the topic of future studies.

The majority of cVMS emissions to the environment are to the atmosphere, and they have the potential for long range atmospheric transport[12,36,37]. However, they are also released to the environment in WWTP effluent[6], as was confirmed by analysis in this project. The present study found up to two orders of magnitude higher concentrations in biota and sediments from lakes Mjøsa and Randsfjorden compared to the reference lake, Femunden. This is comparable to findings from Swedish lakes where perch D5 levels between lakes receiving WWTP effluent and lakes receiving no effluent differed by up to three orders of magnitude[24]. If long range transport was the dominant source of cVMS to the lakes, the concentrations would be expected to be more similar between the lakes. Although Femunden is remote from local sources, it is not remote from the perspective of long range atmospheric transport, and thus the deposition of cVMS from the atmosphere is not likely to differ greatly between the investigated lakes on this relatively small spatial scale. Thus, local sources such as discharged effluent water from the WWTPs are suggested to be the primary source of cVMS in the Norwegian lakes.

5.4 Dietary relationships

Analysis of the dietary descriptors $\delta^{13}C$ and $\delta^{15}N$ enabled an evaluation of the major carbon source and relative trophic position in the fish community of Lake Mjøsa (Figure 3.). The food web sampled in Mjøsa ranged across 2.4 trophic levels, from 2 to 4.4 (Table 2). The pelagic feeding fish were separated from the benthic feeding fish in the $\delta^{13}C$ signal, but overlapped in trophic position, i.e. trout and burbot occupied the highest pelagic and benthic trophic positions, followed by vendace and perch, respectively. The pelagic feeding smelt consisted of two size groups that were separated in trophic position, with the large smelt occupying a higher trophic position that overlapped with trout, while the smaller smelt occupied a lower trophic position that overlapped with vendace and perch (Figure 3., Table 2). The smelt diet shifts from being predominantly zooplankton for younger and smaller smelt, to an increasing degree of cannibalism once the fish are in their fourth year (3+) and longer than approximately 10 cm[38].

Of the Lake Mjøsa invertebrates, epilimnic zooplankton occupied the lowest trophic position, and there was little variance in the isotopic signals among the samples. Hypolimnic zooplankton had higher $\delta^{15}N$ values, and showed larger spread in data (the samples collected on August 2nd 2012 had markedly lower $\delta^{15}N$ values than the samples collected on August 21st 2012) (Figure 3.). In accordance with its main prey and predator, Mysis occupied an intermediate trophic position between the epilimnic zooplankton and the planktivorous fish, with some variation among samples due to different sampling dates (higher $\delta^{15}N$ in one sample collected in early July, compared to those sampled in early August). The $\delta^{13}C$ values did not vary greatly among the invertebrate samples.

In Randsfjorden, the food web sampled ranged 1.7 over trophic levels from 2.0 to 3.7, and was thus narrower compared to the Mjøsa food web (Table 2). Smelt had lower trophic position than large fish-feeding trout, whereas whitefish occupied the lowest trophic position. The fish overlapped in $\delta^{13}C$ values, whereby the variation was particular high for whitefish (Figure 3., Table 2). The spread in $\delta^{13}C$ suggest that there is considerable variation in diet within the whitefish, which is supported by earlier investigations of stomach contents, that identified both purely pelagic feeding fish and fish feeding on benthic and terrestrial invertebrates[39]. TMFs were thus calculated both excluding and including whitefish.

In Randsfjorden, the epilimnic zooplankton isotopic values were tightly grouped, whereas the hypolimnion samples varied substantially, particularly in $\delta^{15}N$, spanning more than one trophic level (Figure 3.). These samples were collected the same day, by the same people, and using the same methods. Thus, the most likely explanation is that the species composition differs with variable amounts of carnivorous hypolimnic zooplankton, as each sample reflects its own individual net haul.

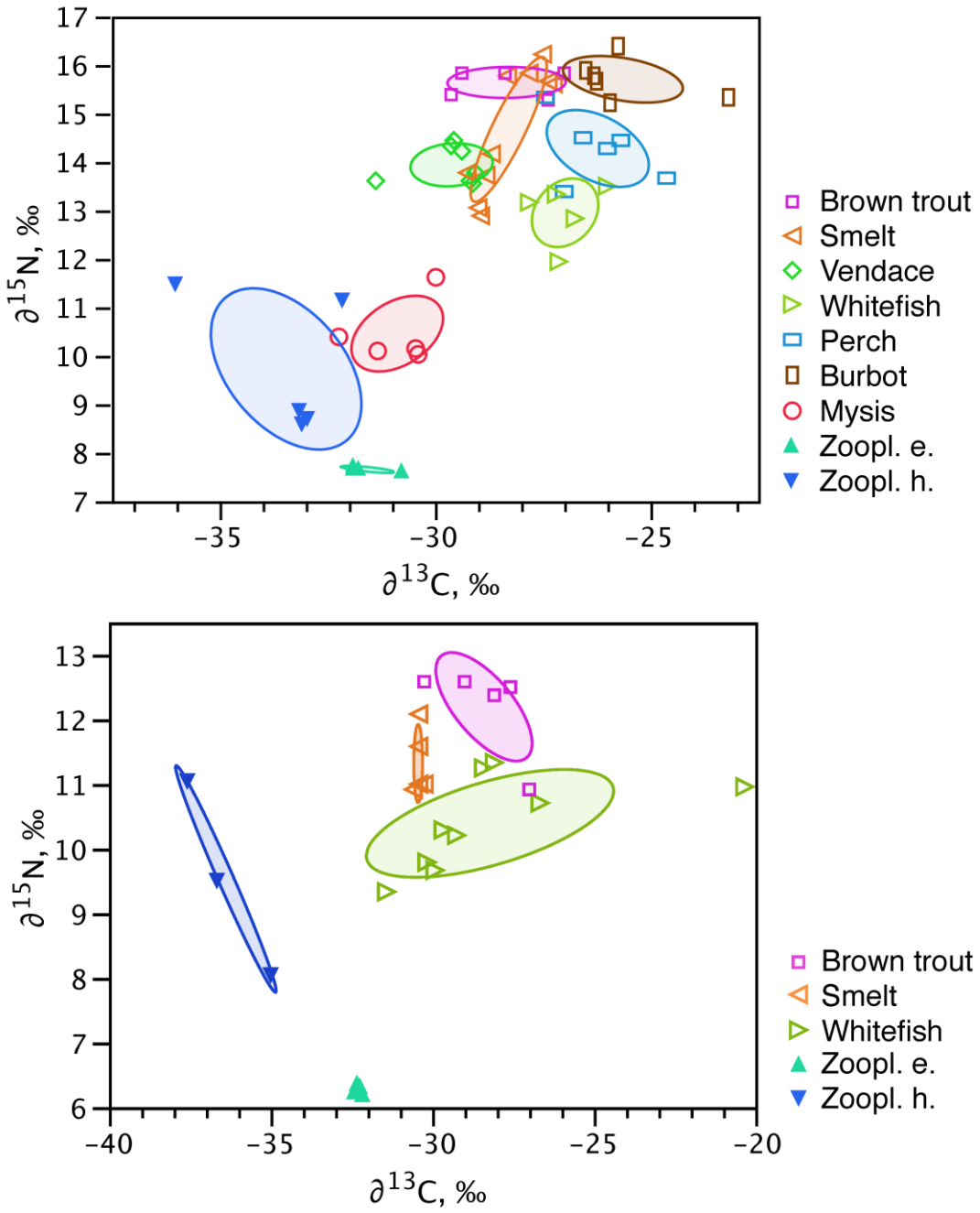
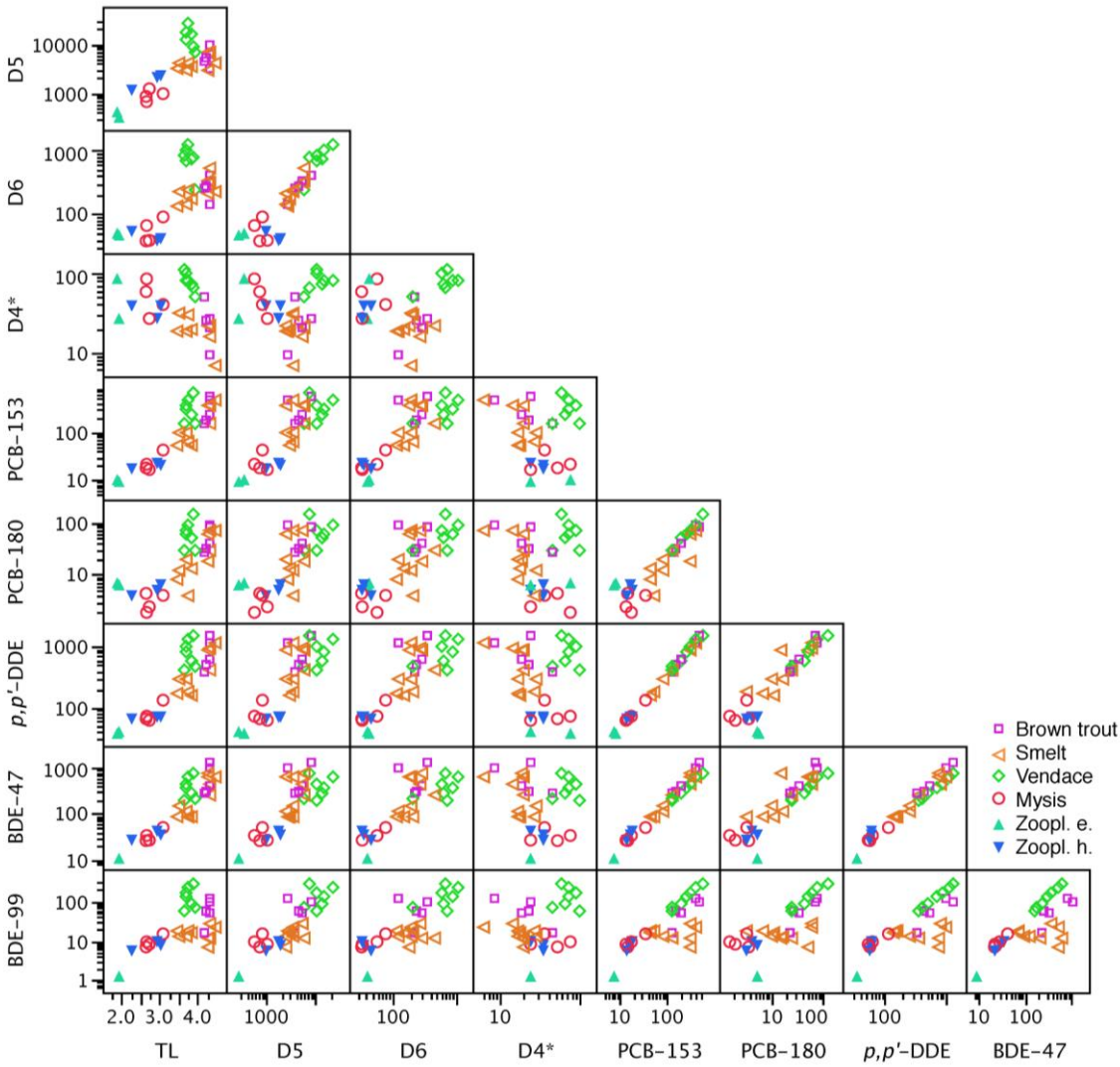


Figure 3. Relationship between the dietary descriptors δ15N and δ 13C in biota from Mjøsa (upper panel) and Randsfjorden (lower panel). Zoopl e. and h. is zooplankton epilimnion and hypolimnion, respectively.

A) MJØSA


D5
D6
D4*
PCB–153
PCB–180
p,p'–DDE
BDE–47
BDE–99
TL
Brown trout
Smelt
Vendace
Mysis
Zoopl. e.
Zoopl. h.

B) RANDSFJORDEN

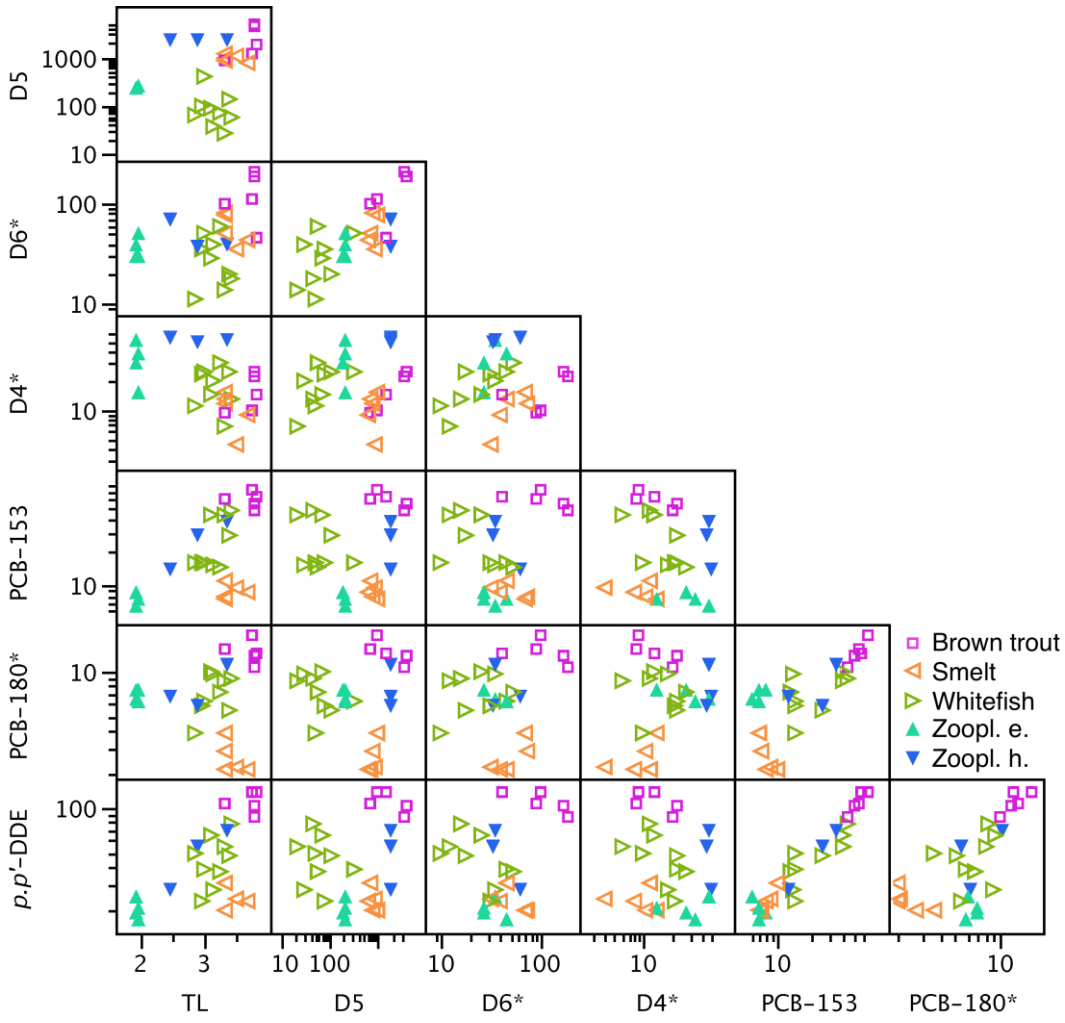


Figure 4. Scatter plot matrix for lipid normalized concentrations of cVMS and legacy POPs, and trophic level (TL) in A) Mjøsa and B) Randsfjorden. Zoopl e. and h. is zooplankton epilimnion and hypolimnion, respectively.

5.5 Food web biomagnification of cVMS

The food web biomagnification of D4 and D5 did not differ between the lakes, regardless of whitefish from Randsfjorden being included or not in the regression (i.e. the interaction *TL x Lake* was not significant $p > 0.28$, Table S13). Thus, one TMF for each of these chemicals was valid for both lakes (Table 4). Detailed results including whitefish are found in Table 4, Table S13 and Fig. S2. For D4 66% of the samples were below the LOQ for the two lakes combined, indicating a greater uncertainty for the TMF (indicated by the asterix). This uncertainty was however unbiased as the samples below the LOQ were distributed throughout the food web (Table S11). The D4 data resulted in a low TMF* of 0.7 (0.5-0.9) (Fig 5, Table 4). Due to the uncertainty associated to the D4 TMF*, it cannot firmly be concluded that D4 was subject to trophic dilution, however, the results show that D4 biomagnification, if it was occurring, would have been very low. The low D4 TMF* observed in this study is in agreement with previous findings,[21,22,40] and does not support the classification of D4 as vB.

D5 showed significant food web biomagnification with a TMF of 2.9 (2.1 - 4.0) (Table 4). The D5 TMF was within the range previously measured for the same food web.[20] The consistency with similar TMFs between lakes and between years adds to the body of evidence of significant food web biomagnification of D5 in pelagic freshwater food webs leading to brown trout. Thus, the elevated D5 concentrations in trout from Mjøsa compared to Randsfjorden were not likely due to differences in food web biomagnification, but resulted from higher D5 exposure at the base of the food web (epilimnetic zooplankton) in combination with trout occupying a higher trophic level in Mjøsa due to the presence of Mysis (Table 2, Fig. 5).

For D6, the TMF was similar between the food webs when Randsfjorden whitefish was omitted (*TL x Lake*, $p=0.0605$), resulting in a D6 TMF of 2.3 (1.8 - 3.0) (Table 4). This is the first empirical field evidence for D6 biomagnification in a food web. When whitefish from Randsfjorden was included in the regression, the D6 TMF differed between the food webs ($p=0.0360$). This resulted in a significant D6 TMF in Mjøsa of 2.7 (2.0-3.8) ($p<0.0001$), and a non-significant D6 TMF* in Randsfjorden of 1.5 (0.9-2.4) ($p=0.1173$). When omitting whitefish, D6 TMF* in Randsfjorden was significant ($p=0.0189$), and with a comparable value of 1.6 (1.1-2.3). The same shift in significance was found for D5TMF when calculated for Randsfjorden with and without whitefish (Table 4). The change in D5 TMF and D6 TMF* significance in Randsfjorden was due to the low D5 and D6 concentrations in whitefish compared to other samples at the same trophic level, which resulted in the lower confidence interval intersecting 1 when including whitefish, although the TMF value itself was not greatly affected (Table 4).

PCBs and *p,p'*-DDE TMFs were higher in Mjøsa than in Randsfjorden (i.e. significant interaction *TL x Lake*, $p < 0.005$), regardless of whether whitefish from Randsfjorden were included or not in the regression (Table 4). In Mjøsa, the TMFs of PCBs, *p,p'*-DDE and PBDEs were within or overlapped the ranges that were previously measured for the same food web.[20] For all chemicals that were compared between years, the present regression results were more precise with a narrower confidence interval and a higher R^2. The TMFs for PCBs and *p,p'*-DDE were within the range documented in Canadian lake trout food webs.[41] The TMFs for PBDEs were comparable to those reported from a Canadian lake food web.[42] The higher TMFs of PCBs and *p,p'*-DDE in Mjøsa than Randsfjorden (Table 4), suggest that the food web structure in Mjøsa differs from the structure in Randsfjorden, not just in length, but also in a manner that affected the TMF of legacy POPs, but not cVMS. This will be discussed further under *TMF sensitivity*.

Table 4. Trophic magnification factors (TMF) in freshwater food webs (Mjøsa and Randsfjorden)[a] for cyclic volatile methylsiloxanes (cVMS: D4, D5, D6) and legacy chlorinated and brominated contaminants based on lipid normalized concentrations and trophic levels estimated from stable isotopes of nitrogen[b].

Chemical	Lake	Whitefish	TMF	95% L	95% U	t Ratio	Prob>|t|	R2	N
Same TMF in the two lakes									
D5	Both	-	2.91	2.11	4.02	6.63	<0.0001	0.60	51
D5	Both	+	2.79	1.86	4.20	5.03	<0.0001	0.57	59
D6	Both	-	2.30	1.76	3.02	6.21	<0.0001	0.59	51
D4*	Both	-	0.69	0.54	0.89	-2.98	0.0045	0.29	51
D4*	Both	+	0.70	0.56	0.88	-3.10	0.0031	0.30	59
TMF for each lake separately									
cVMS									
D4*	Mjøsa	-	0.76	0.57	1.01	-1.94	0.0621	0.11	33
D4*	Randsfjorden	-	0.57	0.35	0.93	-2.46	0.0267	0.29	17
D4*	Randsfjorden	+	0.58	0.38	0.87	-2.75	0.0111	0.24	26
D5	Mjøsa	-	3.12	2.28	4.29	7.35	<0.0001	0.64	33
D5	Randsfjorden	-	2.74	1.70	4.41	4.52	0.0004	0.58	17
D5	Randsfjorden	+	2.13	0.76	5.98	1.51	0.1444	0.09	26
D6	Mjøsa	-	2.72	1.96	3.77	6.20	<0.0001	0.55	33
D6*	Randsfjorden	-	1.60	1.09	2.34	2.63	0.0189	0.32	17
D6*	Randsfjorden	+	1.46	0.90	2.36	1.62	0.1173	0.10	26
Legacy POPs									
PCB-153	Mjøsa	-	5.04	3.71	6.85	10.78	<0.0001	0.80	31
PCB-153	Randsfjorden	-	2.19	1.26	3.80	3.04	0.0083	0.38	17
PCB-153	Randsfjorden	+	2.29	1.46	3.60	3.79	0.0009	0.37	26
p,p'-DDE	Mjøsa	-	4.19	3.12	5.61	10.00	<0.0001	0.78	31
p,p'-DDE	Randsfjorden	-	1.94	1.23	3.07	3.09	0.0075	0.39	17
p,p'-DDE	Randsfjorden	+	1.96	1.36	2.84	3.76	0.0010	0.37	26
PCB-180	Mjøsa	-	4.58	2.89	7.26	6.75	<0.0001	0.61	31
PCB-180*	Randsfjorden	-	1.41	0.70	2.82	1.04	0.3137	0.07	17
PCB-180*	Randsfjorden	+	1.48	0.85	2.56	1.46	0.1564	0.08	26
BDE-47	Mjøsa	-	5.72	4.16	7.86	11.22	<0.0001	0.81	30
BDE-99	Mjøsa	-	2.95	1.62	5.35	3.71	0.0009	0.33	30

a The Mjøsa food web included: epi- and hypolimnetic zooplankton, Mysis, vendace, smelt, brown trout. The Randsfjorden food web included epi- and hypolimnetic zooplankton, whitefish (+), smelt and brown trout. The regression was also run omitting whitefish (-).
b The regression was based on natural logarithm transformed lipid normalized contaminant concentrations. Regression estimates for the intercept, slope, interactions and main effects, can be found in Table S13.
c Chemicals marked *: >50% of the data from one or both of the lakes were below LOQ (for cVMS) or LOD (for PCB and BDE). For the lakes combined, D4 and D6 had 66% and 33%, respectively, of the data below LOQ.

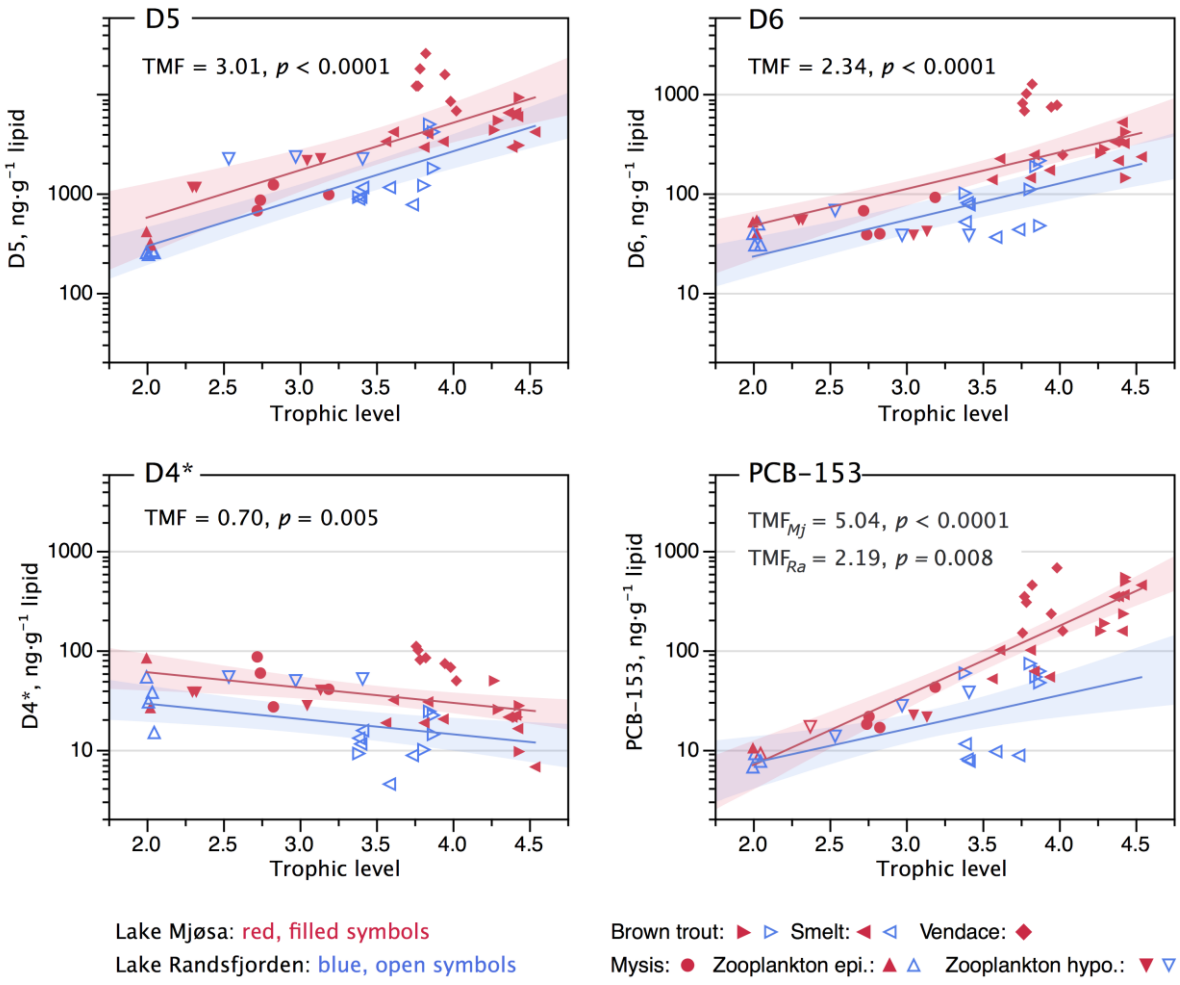


Figure 5. Relationship between lipid normalized concentrations of cVMS (D4, D5 D6) and PCB-153, and trophic level (TL) from Lake Mjøsa and Randsfjorden pelagic food webs. Chemicals marked with asterix (*) have >50% of data below LOQ. Zooplankton epi and hypo are epi- and hypolimnetic zooplankton, respectively. The trophic magnification factor (TMF) was estimated separately for Mjøsa (Mj) and Randsfjorden (Ra) when the interaction *TLxLake* was significant.

TMF sensitivity

Due to indications of semi-pelagic feeding of whitefish based on previous diet analysis, the wide spread in $\delta^{13}C$ in whitefish, and low levels of cVMS in whitefish compared to other species with same trophic level, the sensitivity of the TMF to the inclusion of Randsfjorden whitefish was investigated more closely for cVMS and legacy POPs. In Randsfjorden, the D5 TMF and D6 TMF* estimates were not greatly affected by the inclusion or omission of whitefish in the food web (Table 4). However, it resulted in non-significant versus significant TMF, respectively, as including whitefish reduced the lower confidence interval so that it intersected 1 (Table 4). The PCB and *p,p'*-DDE TMF regressions were not affected, as whitefish had concentrations of legacy contaminants that were comparable to concentrations in other species at the same trophic level (Table 2, Table 4, Fig. 4). The difference in food web magnification between the chemicals in lake Randsfjorden is also illustrated by the poor correlation among D5, D6 and legacy POPs, a correlation that was only significant when

whitefish was omitted, despite the decrease in sample size (Table S14). The sensitivity of the cVMS TMF to the low cVMS concentrations in whitefish suggest that its cVMS source and uptake differ from the other species in the purely pelagic food web, e.g., as a result of feeding more in the littoral zone on terrestrial and benthic prey. It also suggests that the environmental distribution of cVMS and legacy POPs differs so that littoral and benthic prey are less contaminated with cVMS compared to pelagic prey, whereas legacy POPs do not differ as much in prey concentrations depending on habitat. This hypothesis remains to be addressed, but one can imagine several explanations for differences in cVMS concentrations in littoral and benthic prey. One is that cVMS have a much lower fugacity in the terrestrial environment due to their comparatively rapid elimination in air via phototransformation, which would explain lower concentrations in terrestrial prey such as surface insects. Another would be that cVMS have important ongoing point source discharges to lakes from WWTP effluent,[24] whereas the legacy contaminants have had a historic diffuse presence in Mjøsa since the 1970s with several identified minor point sources.[43] This consideration illustrates that although PCBs are good benchmarks from a bioaccumulation point of view, they do not reflect variability in TMF that is caused by other factors such as differences in environmental distribution between chemicals. The present results suggest that TMF in pelagic food webs leading to trout is robust with respect to spatial variation in contaminant exposure between and within lakes for point source present-use chemicals such as cVMS, and that their TMF is more affected by habitat related differences in contaminant concentrations and food web structure.

The present study confirms and provides new documentation of significant D5 and D6 food web biomagnification with TMF > 1 for the freshwater pelagic food web in two Norwegian lakes. This is in contrast to the two other available food web studies, which report trophic dilution and TMFs < 1 for cVMS in the benthic freshwater food web from Lake Pepin, Mississippi, USA,[21] and in the marine benthopelagic food web of Oslofjorden, Norway.[22] The silicone industry also reported TMF < 1 for D4 and D5 based on preliminary data from pelagic marine fish in Tokyo Bay, Japan.[40,44] As suggested in the present study, as well as the previous Lake Mjøsa study,[20] the cVMS TMF is sensitive to food web composition, and an explanation for differences in TMFs between studies may be ecosystem characteristics that affect both the trophic transfer and retention of contaminants, and thus the degree of biomagnification. Some obvious differences among the existing cVMS TMF studies, in addition to the habitat (pelagic versus benthic/benthopelagic), are water temperature, water residence time, water depth, species composition, and salinity. The influence of differences in these characteristics on cVMS biomagnification should be the subject of future investigations.

One explanation for the different biomagnification behavior of D4 compared to D5 and D6 could be a more rapid metabolism of D4 than the other cVMS. This is suggested by biotransformation rates in fish derived from inverse modeling of bioconcentration studies.[45] There are, however, few empirical studies of metabolism and elimination of cVMS.[40,44] Although mammals have been shown to rapidly metabolise and eliminate D4 and D5, fish seem to have a slower metabolism of D4 (2% of recovered dose was present as metabolites) than of D5 (14% of recovered dose was present as metabolites).[40,44] In many environmental biota studies, the D4 concentrations are close to or below the LOQ,[3,20] which add an uncertainty to the evaluation of the biomagnification of D4.

Although the TMFs vary within and between studies and ecosystems for a given chemical (Table 4), the present study documents a consistent pattern regarding the TMF being > or < 1, i.e. biomagnification or not, as long as the majority of data are above the quality threshold, and the organisms included in the estimation reflect a clearly defined food web. The present results support a consistency in significant D5 TMF above 1 between lakes and years, and present novel documentation of D6 food web biomagnification above 1 in a pelagic food web leading to brown trout.

6. Conclusions

- The present study from 2012 has confirmed the high cVMS concentrations, and food web biomagnification of D5, in Mjøsa that was reported from 2010. The levels in predatory fish are comparable to those reported for the Inner Oslofjord.
- The present study has documented the potential of D5 and D6 to biomagnify in pelagic freshwater food webs (TMF > 1), whereas D4 seems to be subject to trophic dilution (TMF* < 1).
- Whereas D5 and D6 concentrations within the pelagic food web were significantly correlated with biomagnifying legacy contaminants (e.g. PCB-153, *p,p'*-DDE), D4 correlated with neither D5 or D6 nor PCB-153 or *p,p'*-DDE.
- The cVMS levels were highest in Mjøsa, intermediate in Randsfjorden, and below limit of quantification in most samples in Femunden.
- cVMS were quantified in grab samples of WWTP effluent, while D5 and D6 were quantified in surface sediments from Mjøsa and Randsfjorden. The surface sediments from Randsfjorden and Mjøsa showed high spatial variation in cVMS concentrations, with highest concentrations near Brandbu and Gjøvik, respectively.
- Due to the large difference in cVMS levels between the lakes, the presence of cVMS in effluent water, and the large spatial variation in cVMS concentrations within the lakes with WWTPs, local sources rather than long range atmospheric transport, are the likely major sources of cVMS to the lakes,.

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8. Supporting information

CHAPTERS

TABLES

Lake description

The primary research lake, Lake Mjøsa is Norway's largest lake (Table 1. , Figure 2), with a species rich fish community and a well defined pelagic food web leading to brown trout (*Salma trutta*) as the top predator, smelt (*Osmerus eperlanus*) and vendace (*Coregonus albula*) as primary plantivorous prey, and an invertebrate community consisting of cladocerans, copepods and *Mysis relicta*.
There are no other lakes in Norway with an identical food web and human impact from agriculture, industry and the general population. Ecologically, Lake Randsfjorden wich is Norway's 4th largest lake, is the lake with most similarities to Mjøsa. Randsfjorden is a deep fjordlake with a well-defined pelagic food web with brown trout and arctic char (*Salvelinus alpinus*) as top predators, and whitefish (*Coregonus lavaretus*) and smelt as planktivorous prey. In contrast to Lake Mjøsa, the invertebrate community lacks *Mysis relicta*.

As a reference site, the remote Lake Femunden, Norway's third largest lake was selected as it is in the wilderness. Femunden's main basin is deep with a pelagic fish community of brown trout, arctic char and whitefish. *Mysis relicta* is not found in the invertebrate community.

Table S 1. Sampling dates, Mjøsa, Randsfjorden and Femunden

Vial ID	Species	Tissue/Matrix	Date sampled	Notes
MJØSA				
S-1	Mysis	whole body, pooled ind.	2 August	Gillundstranda
S-2	Mysis	whole body, pooled ind.	2 August	Gillundstranda
S-3	Mysis	whole body, pooled ind.	2 August	Gillundstranda
S-4	Zooplankton hypo	whole body, pooled ind.	2 August	Gillundstranda
S-5	Zooplankton hypo	whole body, pooled ind.	2 August	Gillundstranda
S-6	Zooplankton hypo	whole body, pooled ind.	2 August	Gillundstranda
S-7	Zooplankton hypo	whole body, pooled ind.	2 August	Gillundstranda
S-8	Zooplankton hypo	whole body, pooled ind.	2 August	Gillundstranda
S-9	Zooplankton epi	whole body, pooled ind.	3 August	Gillundstranda
S-10	Zooplankton epi	whole body, pooled ind.	3 August	Gillundstranda
S-11	Zooplankton epi	whole body,	3 August	Gillundstranda

		pooled ind.		
S-12	Zooplankton epi	whole body, pooled ind.	3 August	Gillundstranda
S-13	Zooplankton epi	whole body, pooled ind.	3 August	Gillundstranda
B-1	Mysis	whole body, pooled ind.	21 August	Gillundstranda
B-2	Zooplankton hypo	whole body, pooled ind.	21 August	Gillundstranda
B-3	Mysis	whole body, pooled ind.	21 August	Gillundstranda
B-4	Zooplankton hypo	whole body, pooled ind.	21 August	Gillundstranda
B-5	Zooplankton epi	whole body, pooled ind.	21 August	Gillundstranda
M-14	Mysis-1	whole body, pooled ind.	6 July	Gillundstranda
L-1	Vendace	skin free filet	3 July	Gillundstranda
L-2	Vendace	skin free filet	3 July	Gillundstranda
L-3	Vendace	skin free filet	3 July	Gillundstranda
L-4	Vendace	skin free filet	3 July	Gillundstranda
L-6	Vendace	skin free filet	3 July	Gillundstranda
L-7	Vendace	skin free filet	3 July	Gillundstranda
L-8	Vendace	skin free filet	3 July	Gillundstranda
K-1	Smelt	skin free filet, homogenate 6 ind.	6 July	Gillundstranda
K-2	Smelt	skin free filet, homogenate 6 ind.	6 July	Gillundstranda
K-3	Smelt	skin free filet, homogenate 6 ind.	6 July	Gillundstranda
K-4	Smelt	skin free filet, homogenate 6 ind.	6 July	Gillundstranda
K-5	Smelt	skin free filet, homogenate 6 ind.	6 July	Gillundstranda

K-7	Smelt	skin free filet	7 September	Ottestad
K-8	Smelt	skin free filet	7 September	Ottestad
K-9	Smelt	skin free filet	13 September	Ottestad
K-10	Smelt	skin free filet	13 September	Ottestad
K-11	Smelt	skin free filet	13 September	Ottestad
MS-1	Whitefish	skin free filet	3 July	Gillundstranda
MS-2	Whitefish	skin free filet	3 July	Gillundstranda
MS-3	Whitefish	skin free filet	3 July	Gillundstranda
MS-4	Whitefish	skin free filet	3 July	Gillundstranda
MS-6	Whitefish	skin free filet	3 July	Gillundstranda
MT-1	Brown trout	skin free filet	29 August	Gjøvik
MT-2	Brown trout	skin free filet	29 August	Gjøvik
MT-3	Brown trout	skin free filet	29 August	Gjøvik
MT-4	Brown trout	skin free filet	29 August	Gjøvik
MT-5	Brown trout	skin free filet	29 August	Gjøvik
A-1	Perch	skin free filet	3 July	Gillundstranda
A-2	Perch	skin free filet	3 July	Gillundstranda
A-3	Perch	skin free filet	3 July	Gillundstranda
A-4	Perch	skin free filet	3 July	Gillundstranda
A-5	Perch	skin free filet	3 July	Gillundstranda
A-6	Perch	skin free filet	3 July	Gillundstranda
Lake-1	Burbot	Liver and filet	5 July	Gillundstranda
Lake-2	Burbot	Liver and filet	5 July	Gillundstranda
Lake-3	Burbot	Liver and filet	5 July	Gillundstranda
Lake-4	Burbot	Liver and filet	5 July	Gillundstranda
Lake-5	Burbot	Liver and filet	5 July	Gillundstranda
Lake-6	Burbot	Liver and filet	5 July	Gillundstranda
MJ-1	PUF	Water dissolved	03 September	Gillundstranda
MJ-2	PUF	Water dissolved	03 September	Gillundstrand

				a
MJ-3	PUF	Water dissolved	03 September	Gillundstrand a
MF-1	Filter	Water particle	3 September	Gillundstranda
MF-2	Filter	Water particle	3 September	Gillundstranda
MF-3	Filter	Water particle	3 September	Gillundstranda
Msed-1	Sediment	Sediment	3 September	Gillundstranda
Msed-2	Sediment	Sediment	5 September	Ottestad
Msed-3	Sediment	Sediment	5 September	Lillehammer
Msed-4	Sediment	Sediment	5 September	Lillehammer
Msed-5	Sediment	Sediment	5 September	Lillehammer
Msed-6	Sediment	Sediment	5 September	Gjøvik
Msed-7	Sediment	Sediment	5 September	Gjøvik
WTP-1	Effluent water grab sample	Effluent water	21 August	Hias (Ottestad)
WTP-2	Effluent water grab sample	Effluent water	21 August	Rambekk (Gjøvik)
WTP-3	Effluent water grab sample	Effluent water	21 August	Lillehammer
RANDSFJORDEN				
R1	zooplankton epilimnion	whole body, pooled ind.	27-29 August	
R3	zooplankton epilimnion	whole body, pooled ind.	27-29 August	
R4	zooplankton epilimnion	whole body, pooled ind.	27-29 August	
R5	zooplankton epilimnion	whole body, pooled ind.	27-29 August	
R6	zooplankton hypolimnion	whole body, pooled ind.	27-29 August	
R7	zooplankton hypolimnion	whole body, pooled ind.	27-29 August	
R8	zooplankton hypolimnion	whole body, pooled ind.	27-29 August	

R9	zooplankton hypolimnion	whole body, pooled ind.	27-29 August	
R11	Smelt	skin free filet	27-29 August	
R12	Smelt	skin free filet	27-29 August	
R14	Smelt	skin free filet	27-29 August	
R15	Smelt	skin free filet	27-29 August	
R16	Smelt	skin free filet	27-29 August	
R17	Whitefish	skin free filet	27-29 August	
R18	Whitefish	skin free filet	27-29 August	
R19	Whitefish	skin free filet	27-29 August	
R20	Whitefish	skin free filet	27-29 August	
R21	Whitefish	skin free filet	27-29 August	
R22	Whitefish	skin free filet	27-29 August	
R23	Whitefish	skin free filet	27-29 August	
R24	Whitefish	skin free filet	27-29 August	
R25	Whitefish	skin free filet	27-29 August	
R36	Brown trout	skin free filet	27-29 August	
R37	Brown trout	skin free filet	27-29 August	
R38	Brown trout	skin free filet	27-29 August	
R39	Brown trout	skin free filet	27-29 August	
R40	Brown trout	skin free filet	27-29 August	
RS-26	Sediment	Sediment	30 August	Jevnaker
RS-27	Sediment	Sediment-reference	30 August	Jevnaker
RS-28	Sediment	Sediment	30 August	Jevnaker
RS-29	Sediment	Sediment	30 August	Brandbu
RS-30	Sediment	Sediment	30 August	Brandbu
RS-31	Sediment	Sediment	30 August	Fluberg
RS-32	Sediment	Sediment	30 August	Fluberg
RS-33	Sediment	Sediment-reference	30 August	Fluberg
WTP-4	Effluent water	Effluent water	21 August	Brandbu -1

WTP-5	Effluent water	Effluent water	21 August	Jevnaker-1
WTP-6	Effluent water	Effluent water	21 August	Dokka - 2
FEMUNDEN				
F2	Zooplankton epi	whole body, pooled ind.	8-9 August	
F3	Zooplankton epi	whole body, pooled ind.	8-9 August	
F4	Zooplankton epi	whole body, pooled ind.	8-9 August	
F5	Zooplankton epi	whole body, pooled ind.	8-9 August	
F6	Zooplankton epi	whole body, pooled ind.	8-9 August	
F7	Sediments	Sediment	8-9 August	
F8	Sediments	Sediment	8-9 August	
F10	Sediments	Sediment	8-9 August	
F12	Whitefish	skin free filet	8-9 August	
F13	Whitefish	skin free filet	8-9 August	
F14	Whitefish	skin free filet	8-9 August	
F15	Whitefish	skin free filet	8-9 August	
F16	Whitefish	skin free filet	8-9 August	
F17	Whitefish	skin free filet	8-9 August	
F19	Arctic char	skin free filet	8-9 August	
F20	Arctic char	skin free filet	8-9 August	
F21	Arctic char	skin free filet	8-9 August	
F22	Arctic char	skin free filet	8-9 August	
F23	Arctic char	skin free filet	8-9 August	
F24	Arctic char	skin free filet	8-9 August	
F25	Arctic char	skin free filet	8-9 August	
F26	Brown trout	skin free filet	8-9 August	

F27	Brown trout	skin free filet	8-9 August
F28	Brown trout	skin free filet	8-9 August
F29	Brown trout	skin free filet	8-9 August
F30	Brown trout	skin free filet	8-9 August
F31	Brown trout	skin free filet	8-9 August

Sampling description

Zooplankton from the epilimnion and from the hypolimnion were collected by horizontal trawling at separate depths above and below the thermocline (zooplankton net 250 µm Nylon single strand, custom made at the Norwegian Institute for Water Research (NIVA), with brass cup and brass mesh). In Mjøsa, *Mysis relicta* was picked with tweezers from the hypolimnion trawls. Mysis and zooplankton contaminant analysis were kept in preheated glass jars, and material for stable isotopes was wrapped in aluminum foil. Some of the zooplankton material was difficult to concentrate (i.e. filter off all water), thus some samples contained more water, leading to a higher estimate of water content (Table S 2).

Table S 2. Water content and lipid content of zooplankton samples

Species	Vial ID	Sample weight (g)	Dry weight %	Lipid %
MJØSA				
Zooplankton epilimnion	S-13	5.2	6.2	0.76
Zooplankton epilimnion	B-5	54.7	4.9	0.59
Zooplankton hypolimnion	B-2	16.8	13	6.2
Zooplankton hypolimnion	S-8	5.8	5.7	1.3
Zooplankton hypolimnion	B-4	24.4	14	7.4
Mysis relicta	S-3	2.4	11	2.1
Mysis relicta	B-1	18.2	13	3.4
Mysis relicta	B-3	21.0	13	4.1
Mysis relicta	M-14	7.3	6.6	0.99
RANDSFJORDEN				
Zooplankton epilimnion	R1	27.0	6.0	0.76
Zooplankton epilimnion	R3	44.0	4.8	0.67
Zooplankton epilimnion	R4	19.9	6.75	0.80
Zooplankton epilimnion	R5	33.1	6.0	0.67
Zooplankton hypolimnion	R6	13.3	2.4	0.73
Zooplankton hypolimnion	R7	16.0	3.6	1.85
Zooplankton hypolimnion	R8	27.2	1.28	0.23
Zooplankton hypolimnion	R9	29.7	4.33	2.37

In Lake Mjøsa, vendace and small smelt were collected with gill nets in the surface waters, whereas perch, whitefish, burbot and larger smelt were collected in gill nets deployed at deeper waters. In Randsfjorden, smelt were collected with gill nets in the surface waters, and whitefish was collected from large traps used for commercial fishing. Brown trout from Mjøsa and Randsfjorden, and arctic char from Femunden, was fished by angling by local fishermen according to specific protocol and instruction by NIVA. In Femunden, whitefish and brown trout were collected with pelagic gill nets, by local fishers according to specific protocol and instruction by NIVA. All Femunden fish were immediately wrapped in aluminium foil and sealed polyethylene bags, and stored cooled until dissected back on land. From Lake Mjøsa, some benthopelagic species (perch (*Perca fluviatilis*), burbot (*Lota lota*), whitefish (not primarily a member of the pelagic food web as it is in Lake Randsfjorden and Lake Femunden) were sampled to allow comparison between fish feeding from different carbon sources (benthic versus pelagic). Only brown trout larger than 30 cm were included, to ensure fish-feeding specimens.

In Mjøsa, high volume water samples (n=3; 191 L, 237.6 L, 237.8 L) were collected using a pre-programmed in situ water sampler at ca 15 m depth (**Figure *2***). The in situ water sampler was custom made for NIVA, and includes a filter holder in stainless steel for collection of the particulate phase (for analysis of cVMS in the present study) and a chamber for polyurethan foam (PUFs) for collection of the water dissolved fraction (for analysis of PCBs in the present study). Glassfibre filters (GFF, 29.9 cm, 1 µm pore size) were purchased from Chongqing Zaisheng Technology Development Co., Ltd, and PUFs were purchased as polyurethan foam from Gumotex and cleaned with solvents at the Research Center for Toxic Compounds in the Environment (RECETOX).

Grab samples of effluent water were sampled during the same day (from 8 am to 3 pm Tuesday August 21st 2012) direclty from the outlet drain of WWTPs in Lake Mjøsa and Lake Randsfjorden (Table 3). It was heavy rainfall on Monday August 20th, whereas August 21st was sunny with little/no rainfall. Effluent water was sampled directly from the outlet drain by trained personnel at the WWTPs instructed by NIVA personnel regarding sampling precautions. In Randsfjorden, the effluent was collected directly onto the clean glass bottles (2.5 L), and in Mjøsa the water was transferred to the glass bottle using a clean beaker of stainless steel. Aluminum foil sealed the bottle under the cap. The samples were stored cooled until analysis at ITM. Two bottles were collected from each plant, and were pooled into one sample (5 L) in the laboratory.

Chemical analysis

cVMS analysis - Method Description

Fish. About 10 g of tissue (1 g for three of the fish liver samples) was weighed into 50 mL centrifuge tubes. After addition of 20 mL of dichloromethane (DCM) (Lichrosolve, Merck, Germany) and 60 uL of the surrogate standard solution (containing ^{13}C labeled D4, D5, and D6), the tubes were closed with aluminum foil under the lid and left to stand overnight in the clean air cabinet. The tissue was homogenized with an ultra turrax and centrifuged for 10 min at 2200 rpm. This resulted in 3 phases, with DCM at the bottom, fish homogenate in the middle, and water on the top. The water phase was decanted and discarded. The homogenate was punctured and the DCM extract was transferred to a 250 mL Erlenmeyer flask containing 16-21 g of glass beads (diameter 4 mm, Marienfeld, Germany) and a magnetic stir bar. For the procedural blanks, 75-200 mg of corn oil was also added to simulate the sample matrix. A gas washing bottle stopper was placed on the flask. The inlet port of the stopper was connected to a nitrogen gas supply, which was equipped with purification cartridges containing ENV+ to remove any traces of cVMS. The outlet port of the stopper was connected to a sorbent cartridge. The 1mL plastic cartridges were manually filled with 10-15 mg of Isolute ENV+ packed between 2 PE frits (all from Biotage AB, Sweden). After the first 4 extractions the PE frits were identified as a source of D6 contamination. The frits were from that point stored in DCM and repeatedly ultrasonicated and rinsed with DCM prior to use and the lower frit was

replaced with glass wool.. The cleanup of the extract was started by turning on the magnetic stirrer and purging the flask with N_2 at a flow rate of 200-300 mL/min until the solvent was fully evaporated (2.5-3 h). Then the heating element of the magnetic stirrer (5 positions, IKAMAG, Germany) was set to maximum, giving a flask wall temperature of ~72 °C, and purging was continued for a further 2 h. The sorbent cartridge was removed and eluted with 0.8 mL hexane. Tetrakis(trimethylsiloxy)silane, M4Q, was added as a volumetric standard, and the cVMS were analysed by GC/MS as described in Kierkegaard et al. (2010).

Zooplankton and Mysis. Sub-samples were transferred from the sample jar to two 50 mL centrifuge tubes using a spoon, stirring the sample jar between each spoonful, and alternating between centrifuge tubes. Surrogate standard solution and 20 mL of DCM were added and the tubes were ultrasonicated for 2*15 min, mixing the tubes between the sonications. The tubes were centrifuged, the water discarded, and the DCM transferred to Erlenmeyer flasks. The extraction was then repeated with another 15 mL of DCM. The extracts were cleaned up and analysed in the same manner as the fish samples.

Sediment. The sediment was weighed after centrifugation and the water discarded. An aliquot (2-3 g) of the centrifuged wet weight was taken for dry weight determination. The remaining sediment was weighed and 15 mL acetone, 3 mL DCM and the surrogate standard solution were added. The tube was ultrasonicated for 2*15 min with thorough mixing between the sonications, centrifuged, and the organic phase was transferred to a new tube. The procedure was repeated with 10 mL acetone and 5 mL DCM. The organic phases were combined and mixed with 20 mL of MilliQ water containing 1% NaCl. After centrifugation the DCM phase was transferred to a 250 mL Erlenmeyer flask and the acetone/water phase was reextracted with another 5 mL DCM. The DCM extract was cleaned up and analysed in the same manner as the fish samples.

As part of the quality assurance program, 9 sediment samples were solvent extracted and analysed without cleanup. About 10 g dewatered sediment was ultrasonicated (2*15 min) in a centrifuge tube with 15 ml acetone, 2 mL n-hexane and the surrogate standards. The organic phase was transferred to a new centrifuge tube and the extraction repeated with 6.5 mL acetone plus 2 ml n-hexane and 15 min ultrasonification. 15 mL of 2% NaCl in MilliQ water was added to the combined extract. The hexane phase was transferred to a GC vial and analyzed by GC/MS.

Suspended particulate material. The damp filters were extracted with 80 mL DCM in a 250 mL Erlenmeyer flask employing ultrasonification for 3*15 min. The extraction was repeated with 50 mL DCM (2*15 min). The combined DCM extract was cleaned up and analysed in the same manner as the fish samples with the only exception that the extract was evaporated overnight.

STP effluent. The effluent samples were stored at 4 °C in fully filled 2.5 L brown glass bottles until analysis. Before analysis 250 mL of the effluent water was replaced by 250 mL DCM containing the surrogate standards dissolved in ethyl acetate. The bottle was vigorously stirred with a magnetic stir bar overnight, after which it was allowed to stand for 1 h. 25 g of NaCl was added and the bottle was slowly stirred for 30 min. Most of the water was decanted and discarded. The bottle was shaken with the remaining water/DCM fraction, which was then transferred to a 250 mL flask. Following phase separation (emulsions were centrifuged), the DCM phase was transferred to a 500 mL Erlenmeyer flask. The DCM extract was cleaned up and analysed in the same manner as the fish samples, but with a higher N_2 flow (~ 350 mL/min) and a sorbent cartridge with more ENV+ (25 mg). The procedural blank consisted of 2.5 L of MilliQ water and 36 mg of corn oil.

Field blanks. The pouches were transferred to a glass tube. 1.5 mL of n-hexane and the surrogate standard solution were added. The tube was mixed with a vortex mixer for about 15 s. The n-hexane was transferred to a GC vial and analyzed.

Method Evaluation and QA/QC

cVMS formation. It has previously been shown that D5 can be transformed into D4 and D3 during sampling out of the gas phase onto ENV+ (Krogseth et al., 2013). To test whether this was occurring, two blank samples with 100 mg of corn oil were analysed in which the surrogate standard solution of ^{13}C labeled D4, D5 and D6 was replaced with a single ^{13}C labeled cVMS: ^{13}C-D5 for one of the blank samples and ^{13}C-D6 for the other. After these standards were added to the extraction solvent and submitted to the sample cleanup, they were quantified against the volumetric standard M4Q. The results showed that there was a high recovery of the labeled D5 and D6 and no evidence for the formation of ^{13}C labeled D4, D5, or D6 during the sample cleanup procedure (Table S 3).

Table S 3. Concentrations* of ^{13}C labeled cVMS in standards of ^{13}C labeled D5 and D6 before and after having been submitted to the cleanup procedure.

	^{13}CD4	^{13}CD5	^{13}CD6
^{13}CD5 before cleanup	0.002	1.025	0.000
^{13}CD5 after cleanup	0.001	0.916	0.000
^{13}CD6 before cleanup	0.003	0.002	0.330
^{13}CD6 after cleanup	0.003	0.002	0.310

*Concentration approximated as the peak area of the analyte normalized to that of the volumetric standard (M4Q).

Extraction efficiency. Extraction efficiency was assessed in two manners. First the effect of extending the second (heated) phase of the purge and trap cleanup was studied. Two smelt samples were extracted and subjected to the cleanup. However, instead of using one ENV+ cartridge on the outlet of the Erlenmeyer flask, the cartridge was exchanged, first after the end of the solvent evaporation phase, and then at intervals of 30 min, 40 min, 60 min and 30 min during the heating phase. The recovery of the surrogate standards was quantified in each of the samples. The results showed that no further cVMS were transferred from the extract to the cartridge after the solvent evaporation phase plus 70 min of the heating phase (see Figure S 1). On the basis of these results we chose a 2 h duration for the heating phase as more than sufficient to transfer all of the cVMS from the extract.

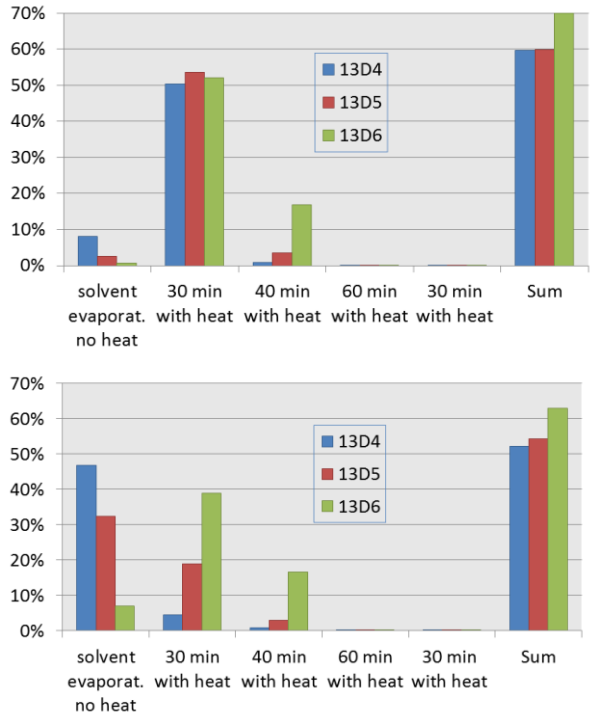

Figure S 1. Recovery of the surrogate standards from 2 smelt samples for which 5 sorbent cartridges were deployed sequentially during the purge and trap cleanup.

The second test of the extraction efficiency was to re-extract 8 biota samples. The same extraction method was applied, and a second batch of surrogate standard was added to the solvent used for re-extraction. The results showed that the second extract contained of the order of 10-20 % of quantity of D5 present in the initial extract (see Table S 4, the results for D4 and D6 are not shown due to the low levels present in the second extract). A burbot muscle sample and a zooplankton sample showed higher values (28% and 37% respectively). The higher value for the zooplankton was attributed to the high water content of the sample, and it was thus decided to extract all zooplankton and mysis samples twice. The extraction efficiency of 80-90% for the other samples was judged sufficient. Note that the percent underestimation of the concentrations due to incomplete extraction is likely to be lower than suggested by the extraction efficiencies estimated here because the extraction efficiency of the surrogate standard was also incomplete, e.g. due to residual solvent in the extracted matrix. If the extraction efficiency of the surrogate standard and the native compound were the same, then there would be no error in the measured concentration.

Table S 4. Quotient of D5 in the second and first extracts of biota samples

Sample	Extract 2/Extract 1 (D5, in %)
Zooplankton epilimnion	37
Mysis	18
Smelt	10
Burbot 1	16
Burbot 3	18
Burbot 4a	21
Burbot 4b	10
Burbot 5	28

Recovery. The recovery of the ^{13}C labeled D4, D5, and D6 surrogate standards was determined for each sample. High and consistent recoveries were observed for all analytes in almost all matrices (see Table S 5). The recoveries were higher in the zooplankton and Mysis samples than in the fish samples, which could be due to the fact that the former were extracted twice while the fish samples were extracted once. Very variable recoveries were observed for D4 in sediment. In some samples the recovery approached 100% while in one it was <10%. The reason for this is unknown. In the 8 samples for which the D4 recovery was <25% the D4 concentration was designated "<" (the matrix control samples showed that when recovery was low the concentrations were overestimates, see below).

Table S 5. Recovery of cVMS surrogate standards from the analysed samples (mean ± std dev in %).

Matrix	*N*	^{13}CD4	^{13}CD5	^{13}CD6
Zooplankton/Mysis	19	81±8	81±7	86±14
Fish	85	72±13	71±13	74±13
Sediment	22	50±32	70±11	80±14
Suspended particulate matter	3	87±1	86±1	87±8
Effluent	7	83±11	86±12	90±14

Repeatability. The repeatability of the method was assessed using the matrix control samples analysed during each round of extractions. The relative standard deviation (RSD) was between 8% and 11% for D5 and D6 in both the sediment and the herring matrices (seeTable S 6). This is a good result, particularly in light of the low D5 and D6 concentrations in these samples. The RSD was higher for D4, which can be attributed to the very low levels in the matrix control samples (a factor of 2 above the LOQ for herring). D4 was below the LOQ in the sediment.

Table S 6. Results of the analyses of the matrix control samples

	D4	D5	D6
Herring (ng/g ww)			
N	13	14	14
Mean	0.9	6.8	2.3
standard deviation	0.3	0.7	0.2
RSD	32%	10%	11%
Sediment (ng/g dw)			
N	6	6	6
Mean	<LOQ	6.9	2.9
standard deviation		0.6	0.2
RSD		8%	8%

Accuracy. Due to the absence of a certified standard reference material for trace analysis of cVMS, the accuracy was evaluated by comparing the method with existing methods for which accuracy information is available. The method of Kierkegaard et al. (2010) for analyzing cVMS in biota has been shown to perform successfully in an interlaboratory comparison (McGoldrick et al., 2011). This method had also been used to analyze the herring homogenate matrix control sample used in this study. The means concentrations for D5 (6.0 ng/g ww, n=18) and D6 (1.7 ng/g ww, n=18) obtained with this method during the year prior to the development of the new method are in reasonable agreement with the values of 6.8 ng/g ww and 2.3 ng/g ww obtained with the new method (Table S 7). The somewhat higher mean concentrations measured with the new method as well as the better repeatability (10 versus 23% and 11 versus 31% for D5 and D6, respectively) may be a reflection of better and more repeatable extraction with the new method.
For sediment, the method in this study was compared with the direct injection of raw extracts, a method which we have previously evaluated in an interlaboratory comparison (Kierkegaard et al., 2013). Seven of the sediments in this study were analysed with both methods. The results for D5 and D6 are shown in Table S 7; D4 was below the LOQ. No consistent difference between the methods was observed and the difference between the results was <25% for 9 of the 14 data pairs.

Table S 7. Comparison of the method with ENV cleanup used in this paper with a reference method involving direct injection of the raw extract; duplicate analyses of 7 different sediments.

Sediment	Method	D5	D6
Ottestad 30 m	ENV cleanup	5.8	6.2
	Raw extract	4.7	4.6
Gjövik 28m	ENV cleanup	285	34
	Raw extract	395	42
Gjövik 120m	ENV cleanup	335	40
	Raw extract	358	40
Ottestad 120 m	ENV cleanup	0.2	<0.3
	Raw extract	2.0	<0.4
Lillehammer, reference	ENV cleanup	0.9	< 1.0

	Raw extract	1.7	<1.0
Lillehammer 80m	ENV cleanup	26	9.4
	Raw extract	25	8.7
Jevnaker, 65 m	ENV cleanup	0.99	0.85
	Raw extract	1.3	<0.9

Table S 8. Limit of quantification (LOQ) for a) biota samples, based on mean procedural blanks + 10 x standard deviation (SD), and b) sediment samples, based on maximum concentration measured in reference sediments from Randsfjorden x 3. The reference sediment was sampled 40-42 cm deep in the sediment core.

a) LOQ for biota samples

	D4 mean ng	SD	LOQ, ng	**D5** mean ng	SD	LOQ, ng	**D6**[a] mean ng	SD	LOQ, ng
Procedural blanks 1-4							4.9	0.8	**13**
Procedural blanks 5-20							1.0	0.4	**5.2**
Procedural blanks 1-20	0.50	0.41	**4.6**	0.42	0.22	**2.6**			

[a]*For D6 two LOQ were applied (the 4 first extraction rounds were contaminated from a source that was later identified)*

b) LOQ for sediment samples

		ng D4	ng D4/g dw	LOQ ng/g dw	ng D5	ng D5/g dw	LOQ ng/g dw	ng D6	ng D6/g dw	LOQ ng/g dw
sediment, Fluberg, reference	RS33	< 13	< 1.4	**4.2**	1.8	0.2	**0.6**	2.0	0.2	**0.6**
sediment, Jevnaker, reference	RS27	<2.4	<0.3		0.8	0.1		1.1	0.2	

[a]For D6 two LOQ were applied (the 4 first extraction rounds were contaminated from a source that was later identified)

POP chemical analysis

Extraction Biological samples

Samples were homogenized and an aliquot was taken for extraction and added internal standards PCB 30, 53, 204 (Ultra Scientific) and BDE 30, 119, 181 (Cambridge isotope laboratories). Samples were then extracted twice using a 50/50 mixture of isopropanol/cyclohexane followed removal of isopropanol by addition of water. All cyclohexane was evaporated off and the fat was dried until stable weight was achieved for total lipid determination. The fat were then dissolved in isohexane and then repeatedly treated with concentrated sulphuric acid. Extract was evaporated to about 100µl followed by PCB analysis. After PCB analysis the extract was solvent extracted twice using acetonitrile saturated with isohexane followed by evaporation and analysis of PBDEs.

Extraction Sediment samples

Sediments samples were added the same internal standards as the biota samples and extracted twice using dichloromethane. The organic phase was then dried using sodium sulphate and then solvent exchanged into isohexane. Cleanup then followed the same procedure as biological samples.

Extraction Polyuretanefoam (PUF) samples.

Each PUF was added the same internal standard as biota and then extracted by soxhlet using 5% ether in n-hexane (EPA method TO-10A, www.epa.org). The organic phase was dried by Na2SO4, transferred off and solvent exchanged into isohexane. Cleanup then followed the same procedure as biological samples.

PCB Quantification

Extracts was analyzed on an Agilent 7890A gas chromatograph coupled to an Agilent 5975C masspektrometer Agilent JW schientific, Santa Clara, USA. The instrument was operated in single ion monitoring (SIM) mode using electron impact ionization (69,9EV). The gas chromatograph was equipped with a 30m Agilent DB-5 column (0,25mm i.d. and 0,25µm film thickness) and kept at constant flow of 1,2ml/min of He. The GC-oven was kept at 60°C for 2 min and then raised to 250°C at a rate of 7°C/min and then finally raised to 310°C at a rate of 15°C/min, the oven was then held at 310°C for 2min. Samples injection was 1µl pulsed splitless injection at 20psi for 1,2min and the injector temperature was set to 300°C. Transfer line, ion source and quadrupole were kept at 280, 230 and 150°C, respectively. Quantification of individual compounds was done using the relative response of surrogate internal standard and comparing that to a calibration curve.

PBDE analysis

Determination of PBDEs was performed with a Hewlett Packard 6890Plus GC linked to a Hewlett Packard 5973 MS detector operated in negative chemical ionisation (with methane) and SIM mode. A 4 µL pulsed splitless injection (injector temperature of 280 °C and a pulse pressure of 50 psi held for 2 min) allowed transfer of analytes onto a DB-5MS column (Agilent Technologies Inc., 15 m, 0.25 mm i.d., 0.1 µm film thickness). The oven temperature was set to 120 °C. It was held for 2 min before being increased to 345 °C at the rate of 25 °C min^{-1} (then held for 5 min). The carrier gas (helium) flow was set to 1 mL min^{-1} for the first 13 min and increased to 1.4 mL min^{-1} at the rate of 0.1 mL min^{-1}. Ion source, quadrupole and transfer line temperatures were 250, 150 and 325 °C, respectively. Ion fragments m/z 79 and 81 were used for qualifying and quantifying PBDEs.

Table S 9. Analytical uncertainty for a) sediment, with SRM 1944 as reference material and b) fish muscle with HSD8 as reference material.

a) SEDIMENT	Average %	This study
HCB	30	<30
PCB 52	20	22
PCB 101	30	<30
p,p-DDE	30	<30
PCB 118	30	<30
p,p-DDD	40	<40
PCB 153	30	<30

PCB 105	26	<26
p,p-DDT	60	<60
PCB 138	23	<23
PCB 156	32	<32
PCB 180	25	36
PCB 209	36	<36

b) FISH	Average %	This study
HCB	40	<40
PCB 52	30	<30
PCB 101	26	<26
p,p-DDE	26	<26
PCB 118	26	<26
PCB 153	26	<26
PCB 105	26	<26
PCB 138	26	<26
PCB 156	26	<26
PCB 180	26	30
PCB 209	40	<40

Table S 10. Lipid quantification results in the the Quasimeme test programme.Z-score ≤ |2| is acceptable.

Year	Sample	Assigned value % lipid	NIVA % lipid	Lipid Z-score
2007	R50: 92	14.044	17	0
	93	2.643	3	0.9
2008	R52: 94	57.49	58.1	0.1
	95	2.629	3.17	1.4
2010	R62:104	17.36	20	1.2
	105	2.705	3.1	1
2011	R64:106	11.82	10	-1.2
	107	3.22	3.2	0
2011	R66:108	57	49.7	-1
	109	4.079	3.9	-0.3
2012	R68:110	2.391	2.4	0
	111	3.19	3.2	0
2012	R70:112	2.08	2.14	0.2
	113	3.938	3.89	-0.1

Results

Table S 11. cVMS measured in A) Lake Mjøsa B) Lake Randsfjorden and C) Lake Femunden. Biota (ng/g ww), sediments (ng/g dw), effluent water (ng/L) and filtered particles from the water (ng/L), and their respective field blanks or reference material.

A) MJØSA

FIELD BLANKS (FB)	**Identification**	**D4**	**D5**	**D6**
		ng D4	**ng D5**	**ng D6**
Zooplankton FB-23	M-8a	0.4	0.6	0.1
Mysis FB-25	M-4a	2.7	1.8	0.8
Fish FB-24	MS-7. Gill net bottom	2.8	3.0	1.5
Fish FB-18	MS-5. Gill net surface	3.2	2.5	1.2
Unexposed FBs (*mean of 3*)	FB28. 29. 5	1.6	1.1	1.2

Species	Sample	ng D4	D4 ng/g ww	ng D5	D5 ng/g ww	ng D6	D6 ng/g ww
Zooplankton epi	S9 + S10	< 1.8	< 0.2	17	2.3	< 2.5	< 0.3
Zooplankton epi	S11 + S12	< 1.9	< 0.2	20	2.4	< 3.0	< 0.4
Mysis	S1 + S2	6.1	1.8	49	14	< 4.9	< 1.4
Mysis	M-14	< 2.1	< 0.4	52	9.6	< 5.1	< 0.9
Zooplankton hypo	S4 + S5	< 3.2	< 0.5	89	15	< 4.0	< 0.7
Zooplankton hypo	S6 + S7	< 3.0	< 0.5	85	15	< 3.8	< 0.7
Zooplankton epi	B-5	12	0.5	55	2.4	7.9	0.3
Mysis	B-1	31	2.0	441	29	20	1.3
Mysis	B-3	10	1.1	491	50	16	1.6
Zooplankton hypo	B-2	35	2.4	2019	139	38	2.6
Zooplankton hypo	B-4	29	2	1953	156	35	2.8
Perch	A-1	< 1.4	< 0.1	29	2.8	< 4.1	< 0.4
Perch	A-2	< 2.5	< 0.2	47	4.4	< 5.2	< 0.5
Perch	A-3	< 1.7	< 0.2	30	2.7	< 4.0	< 0.4
Perch	A-4	< 1.7	< 0.2	13	1.3	< 5.0	< 0.5
Perch	A-5	< 1.4	< 0.2	25	2.9	< 4.4	< 0.5
Perch	A-6	< 3.3	< 0.3	32	2.8	< 5.1	< 0.4
Whitefish	MS-3	< 1.0	< 0.2	32	5.2	< 5.0	< 0.8

Whitefish	MS-4	< 2.1	< 0.3	93	15	9	1.5
Whitefish	MS-6	< 2.7	< 0.4	39	6.0	< 5.2	< 0.8
Whitefish	MS-1	< 2.2	< 0.2	57	6	6	0.7
Whitefish	MS-2	< 1.7	< 0.2	28	3.0	< 3.3	< 0.4
Vendace	L-2	9.8	0.9	2083	196	119	11
Vendace	L-1	8.7	1.1	906	120	62	8.1
Vendace	L-3	7.5	1.0	2451	311	118	15
Vendace	L-4	6.8	1.1	830	134	46	7.4
Vendace	L-6	6.3	0.9	832	120	30	4.4
Vendace	L-7	6.1	0.8	1301	176	60	8.1
Vendace	L-8	< 4.5	< 0.6	593	76	55	7.0
Smelt. homogenate 6 ind.	K-1	< 2.1	< 0.2	394	36	16	1.5
Smelt. homogenate 6 ind.	K-2	< 2.3	< 0.2	373	34	19	1.7
Smelt. homogenate 6 ind.	K-3	< 2.1	< 0.2	331	32	16	1.6
Smelt. homogenate 6 ind.	K-4	< 3.6	< 0.3	457	39	24	2.1
Smelt. homogenate 6 ind.	K-5	< 3.1	< 0.3	374	39	23	2.4
Smelt	K-7	< 0.7	< 0.1	383	62	22	3.5
Smelt	K-8	< 0.4	< 0.1	243	38	13	2.0
Smelt	K-9	< 1.6	< 0.4	528	126	27	6.4
Smelt	K-10	< 1.1	< 0.2	313	59	25	4.7
Smelt	K-11	< 1.5	< 0.3	238	41	17	3.0
Brown trout	MT-1	5.3	0.8	1146	166	60	8.6
Brown trout	MT-2	6.1	0.8	1716	235	93	12.7
Brown trout	MT-3	< 4.3	< 0.6	1548	203	69	9.1
Brown trout	MT-4	< 2.7	< 0.4	804	131	38	6.1
Brown trout	MT-5	4.9	0.6	421	52	25	3.1
Burbot M	L-1 muscle	< 3.5	< 0.4	45	4.6	7.5	0.8
Burbot L	L-1 liver	< 3.9	< 8.2	314	667	30	64
Burbot M	L-2 muscle	< 4.1	< 0.4	96	9.7	13	1.3
Burbot L	L-2 liver	10	22.7	675	1489	39	86
Burbot M	L-3 muscle	< 4.2	< 0.4	91	8.7	11	1.0
Burbot L	L-3 liver	< 3.7	< 27	425	3080	35	252
Burbot M	L-4 muscle	4.8	0.3	237	15	26	1.7
Burbot L	L-4 liver	38	21.4	3855	2192	242	138
Burbot M	L-5 muscle	< 4.4	< 0.4	92	9.0	9.8	1.0
Burbot L	L-5 liver	13	12.6	2011	1888	39	37

Burbot M	L-6 muscle	6.5	0.6	151	14	14	1.3
Burbot L	L-6 liver	11	11.7	2767	2937	58	62

		ng D4	D4 ng /g dw	ng D5	D5 ng/g dw	ng D6	D6 ng/g dw
Sediment. Lillehammer. reference	Msed-5	<1.8	<0.3	5.7	0.9	< 6.0	< 1.0
Sediment Ottestad 30 m	Msed-2	<2.9	<1.7	10	5.8	10.3	6.2
Sediment Ottestad 120 m	Msed-1	<12.5	<2.7	<0.8	<0.2	<1.5	0.3
Sediment Gjövik 28m	Msed-6	<9.9	<3.7	761	285	90	34
Sediment Gjövik 120m	Msed-7	<15	<6.7	751	335	90	40
Sediment. Lillehammer 30m	Msed-3	< 1.6	< 0.3	30	6.8	14	3.1
Sediment. Lillehammer 80m	Msed-4	<1.5	<0.5	74	26	27	9.4

		ng D4	D4 ng/L	ng D5	D5 ng/L	ng D6	D6 ng/L
Solvent blank		*1.2*		*1.1*		*1.6*	
Filter blank	*1 filter*	*< 5.9*		*46*		*48*	
Filter particulate phase	MF-1	< 16	< 0.08	< 16	< 0.09	< 11	< 0.06
Filter particulate phase	MF-2	< 12	< 0.05	< 14	< 0.06	< 11	< 0.04
Filter particulate phase	MF-3	< 12	< 0.06	< 12	< 0.05	< 8.6	< 0.04

		ng D4	D4 ng/L	ng D5	D5 ng/L	ng D6	D6 ng/L
Blank	*MilliQ (2.5L)*	*2.2*	*0.9*	*2.3*	*0.9*	*5.7*	*2.3*
Effluent water	Hias	38	15	274	111	35	14.0
Effluent water	Gjövik	22	8.9	201	82	30	12.1
Effluent water	Lillehammer	19	7.7	72	29	< 7.7	< 3.1

B) LAKE RANDSFJORDEN

FIELD BLANKS (FBs)	SAMPLE	ng D4	ng D5	ng D6
Zooplankton epi FB-20	R2	1.5	0.9	0.9
Fish whole procedure FB-21	R10	1.3	3.4	0.6

Smelt- sample preparation. FB-26	R13	2.1		3.9		1.9	
Unexposed FBs (*mean of 3*)	FB28. 29. 5	1.6		1.1		1.2	

SPECIES	**SAMPLE**	**ng D4**	**D4 ng/g ww**	**ng D5**	**D5ng/g ww**	**ng D6**	**D6 ng/g ww**
zooplankton epilimnion	R1	9.5	0.4	48	1.9	8.3	0.3
zooplankton epilimnion	R3	< 2.5	< 0.1	36	1.7	< 4.2	< 0.2
zooplankton epilimnion	R4	< 2.6	< 0.3	21	2.1	< 4.0	< 0.4
zooplankton epilimnion	R5	< 3.7	< 0.2	34	1.6	< 4.8	< 0.2
zooplankton hypolimnion	R6	< 2.3	< 0.4	83	16	< 2.7	< 0.5
zooplankton hypolimnion	R7	12	0.9	566	43	9.4	0.7
zooplankton hypolimnion	R8	< 1.4	< 0.1	40	3.0	< 1.9	< 0.1
zooplankton hypolimnion	R9	16	1.2	727	53	13	0.9
Whitefish	R17	< 2.4	< 0.2	16	1.2	< 3.2	< 0.2
Whitefish	R18	< 1.4	< 0.2	4.0	0.5	< 3.1	< 0.4
Whitefish	R19	< 2.3	< 0.2	37	3.3	< 4.5	< 0.4
Whitefish	R20	< 1.3	< 0.1	4.7	0.4	< 2.3	< 0.2
Whitefish	R21	< 0.8	< 0.1	2.8	0.2	< 1.9	< 0.2
Whitefish	R22	< 2.6	< 0.2	11	0.9	< 3.1	< 0.3
Whitefish	R23	< 1.3	< 0.1	6.5	0.6	< 2.4	< 0.2
Whitefish	R24	7.7	0.5	41	2.8	5.4	0.4
Whitefish	R25	< 2.8	< 0.3	13	1.3	< 3.6	< 0.4
Smelt	R11	< 1.8	< 0.2	131	15	8.7	1.0
Smelt	R12	< 2.4	< 0.3	169	20	10	1.2
Smelt	R14	< 1.8	< 0.2	129	16	11	1.4
Smelt	R15	< 1.4	< 0.1	324	25	10	0.8
Smelt	R16	< 1.9	< 0.2	201	18	11	1.0
Brown trout	R36	< 3.0	< 0.3	492	56	25	2.9
Brown trout	R37	< 0.7	< 0.1	81	9.9	< 8.5	< 1.1
Brown trout	R38	< 0.7	< 0.1	99	12	< 8.8	< 1.1
Brown trout	R39	9.6	0.9	1161	115	31	3.0
Brown trout	R40	< 2.0	< 0.2	422	41	17	1.6

	ng D4	**D4 ng /g dw**	**ng D5**	**D5 ng/g dw**	**ng D6**	**D6 ng/g**

							dw
sediment. Fluberg. reference	RS33	< 13	< 1.4	1.8	0.2	2.0	0.2
sediment. Jevnaker. reference	RS27	2.4	0.3	0.8	0.1	1.1	0.2
sediment. Jevnaker	RS26	<7.9	<1.2	6.6	1.0	5.7	0.8
sediment. Jevnaker	RS28	< 29	< 3.9	6.0	0.81	7.5	1.00
sediment. Brandbu	RS29	7.8	1.1	356	52	68	10
sediment. Brandbu	RS30	9.4	1.6	858	147	130	22
sediment. Fluberg	RS31	< 1.9	< 0.3	38	6.0	28	4.5
sediment. Fluberg	RS32	< 24	< 4.7	57	11	40	7.8
		ng D4	**D4 ng/L**	**ng D5**	**D5ng/L**	**ng D6**	**D6ng/L**
Blank	*MilliQ (2.5L)*	*2.2*	*0.9*	*2.3*	*0.9*	*5.7*	*2.3*
Effluent water	Brandbu	66	27	861	351	27	11
Effluent water	Jevnaker	33	13	151	61	28	11
Effluent water	Otta	60	24	912	368	23	9.3

C) LAKE FEMUNDEN

FIELD BLANK (FB)	**SAMPLE**	**ng D4**	**ng D5**	**ng D6**
Fish FB-10	F18	1.7	1.3	1.0
Sediments FB-4	F11	1.4	0.8	0.5
Zooplankton FB-8	F1	2.0	1.5	0.9
Unexposed FBs (*mean of 3*)	FB28. 29. 5	1.6	1.1	1.2

SPECIES	**SAMPLE**	**ng D4**	**D4 ng/g ww**	**ng D5**	**D5 ng/g ww**	**ng D6**	**D6 ng/g ww**
Char	F19	< 0.9	< 0.1	< 2.3	< 0.2	< 4.7	< 0.4
Brown trout	F26	< 4.4	< 0.5	3.3	0.4	< 8.0	< 0.9
Brown trout	F27	< 3.4	< 0.3	2.9	0.3	< 4.2	< 0.4
Brown trout	F28	< 3.3	< 0.3	2.9	0.3	< 4.1	< 0.4
Brown trout	F29	< 1.7	< 0.1	< 1.4	< 0.1	< 5.9	< 0.5
Brown trout	F30	< 1.6	< 0.2	4.3	0.4	< 3.6	< 0.4
Brown trout	F31	< 2.7	< 0.2	4.0	0.4	< 4.0	< 0.4

		ng D4	**D4 ng/g dw**	**ng D5**	**D5 ng/g dw**	**ng D6**	**D6 ng/g dw**
Sediment	F7	< 3.7	< 0.9	< 0.9	< 0.2	< 2.4	< 0.6
Sediment	F8	< 5.9	< 1.2	< 2.6	< 0.5	< 5.5	< 1.1
Sediment	F10	< 1.9	< 0.4	< 1.3	< 0.3	< 3.3	< 0.7

Table S 12. Percentage (%) samples in the pelagic food web of Mjøsa and Randsfjorden, that were quantified below the limit of quantification (LOQ) for cyclic volatile methylsiloxanes (D4, D5, D6), or limit of detection (LOD) for PCBs, ppDDE and PBDEs.

	Mjøsa	Randsfjorden (excluding/including whitefish)
D4	52	82 / 81
D5	0	0 /0
D6	13	61 / 54
PCB-153	0	0 /0
PCB-180	19	53 / 50
p,p'-DDE	0	0 /0
PBDE-47	0	N/A
PBDE-99	3	N/A

Table S 13. Trophic magnification factor (TMF) statistics for cyclic volatile methylsiloxanes (D4, D5, D6) and selected legacy chlorinated and brominated contaminants in the Lakes Mjøsa [M] and Randsfjorden [R]. TMFs are based on the regression of lipid normalised concentrations onto trophic level (TL) estimated from stable isotopes of nitrogen.

Chemical	Whitefish [R]	Model term	Estimate	SE	t Ratio	p>|t|	Estimate CI Lower 95%	Estimate CI Upper 95%	TMF	TMF CI Lower 95%	TMF CI Upper 95%	R^2	N	Interactions (TLxLake) t-test	Interactions (TLxLake) p(t)	Comments
D5	Included	Intercept	3.60	0.70	5.12	0.0000	2.19	5.01								
D5	Included	Lake[M]	0.80	0.15	5.31	0.0000	0.50	1.11								
D5	Included	Lake[R]	-0.80	0.15	-5.31	0.0000	-1.11	-0.50								
D5	Included	TL	1.03	0.20	5.03	0.0000	0.62	1.43	2.79	1.86	4.20	0.57	59	0.86	0.4	
D5	Excluded	Intercept	3.80	0.44	8.72	0.0000	2.93	4.68								
D5	Excluded	Lake[M]	0.33	0.11	3.15	0.0029	0.12	0.54								
D5	Excluded	Lake[R]	-0.33	0.11	-3.15	0.0029	-0.54	-0.12								
D5	Excluded	TL	1.10	0.13	8.73	0.0000	0.85	1.36	3.01	2.33	3.88	0.66	50	0.47	0.64	
D4*	Included	Intercept	4.43	0.40	11.15	0.0000	3.63	5.22								
D4*	Included	Lake[M]	0.38	0.09	4.47	0.0000	0.21	0.55								
D4*	Included	Lake[R]	-0.38	0.09	-4.47	0.0000	-0.55	-0.21								
D4*	Included	TL	-0.36	0.12	-3.10	0.0031	-0.59	-0.13	0.70	0.56	0.88	0.16	59	1.08	0.28	
D4*	Excluded	Intercept	4.44	0.42	10.64	0.0000	3.60	5.28								

D4*	Excluded	Lake[M]	0.37	0.10	3.62	0.0007	0.16	0.57							
D4*	Excluded	Lake[R]	-0.37	0.10	-3.62	0.0007	-0.57	-0.16							
D4*	Excluded	TL	-0.36	0.12	-2.95	0.0050	-0.60	-0.11	0.70	0.55	0.89	0.12	50	1.05	0.3	
D6	Included	Intercept[M]	1.62	0.59	2.73	0.0103	0.41	2.82								
D6	Included	TL[M]	1.00	0.16	6.20	0.0000	0.67	1.33	2.72	1.96	3.77	0.55	33	2.15	0.04	Separate regression after significant test for interaction
D6*	Included	Intercept[R]	2.61	0.75	3.47	0.0020	1.06	4.16								
D6*	Included	TL[R]	0.38	0.23	1.62	0.1173	-0.10	0.86	1.46	0.90	2.36	0.10	26	2.15	0.04	Separate regression after significant test for interaction
D6*	Excluded	Intercept	1.80	0.44	4.06	0.0002	0.91	2.69								
D6*	Excluded	Lake[M]	0.36	0.11	3.37	0.0015	0.15	0.58								
D6*	Excluded	Lake[R]	-0.36	0.11	-3.37	0.0015	-0.58	-0.15								
D6*	Excluded	TL	0.85	0.13	6.63	0.0000	0.59	1.11	2.34	1.81	3.02	0.61	50	1.92	0.06	
PCB-153	Included	Intercept[M]	-1.30	0.56	-2.32	0.0279	-2.45	-0.15								
PCB-153	Included	TL[M]	1.62	0.15	10.78	0.0000	1.31	1.92	5.04	3.71	6.85	0.80	31	2.98	0.005	Separate regression after significant test for interaction
PCB-153	Included	Intercept[R]	0.35	0.70	0.50	0.6196	-1.10	1.81								

PCB-153	Included	TL[R]	0.83	0.22	3.79	0.0009	0.38	1.28	2.29	1.46	3.60	0.37	26	2.98	0.005	Separate regression after significant test for interaction
PCB-153	Excluded	Intercept[R]	0.42	0.83	0.51	0.6200	-1.35	2.19								
PCB-153	Excluded	TL[R]	0.78	0.26	3.04	0.0083	0.23	1.33	2.19	1.26	3.80	0.38	17	3.01	0.004	Separate regression after significant test for interaction
PCB-180	Included	Intercept[M]	-2.78	0.85	-3.29	0.0027	-4.50	-1.05								
PCB-180	Included	TL[M]	1.52	0.23	6.75	0.0000	1.06	1.98	4.58	2.89	7.26	0.61	31	3.91	0.003	Separate regression after significant test for interaction
PCB-180*	Included	Intercept[R]	1.63	0.65	2.50	0.0198	0.28	2.98								
PCB-180*	Included	TL[R]	0.08	0.20	0.41	0.6838	-0.34	0.50	1.09	0.72	1.65	0.00	26	3.91	0.0003	Separate regression after significant test for interaction
PCB-180*	Excluded	Intercept[R]	0.43	1.05	0.41	0.6849	-1.80	2.67								
PCB-180*	Excluded	TL[R]	0.34	0.33	1.04	0.3137	-0.36	1.04	1.41	0.70	2.82	0.07	19	3.01	0.004	Separate regression after significant test for interaction
ppDDE	Included	Intercept[M]	0.45	0.54	0.84	0.4068	-0.65	1.55								
ppDDE	Included	TL[M]	1.43	0.14	10.00	0.0000	1.14	1.73	4.19	3.12	5.61			3.24	0.002	Separate regression after significant test for

															interaction
ppDDE	Included	Intercept[R]	1.59	0.58	2.75	0.0110	0.40	2.79							
ppDDE	Included	TL[R]	0.67	0.18	3.76	0.0010	0.30	1.04	1.96	1.36	2.84	0.76	31	3.24	0.002	Separate regression after significant test for interaction
ppDDE	Excluded	Intercept[R]	1.62	0.69	2.35	0.0330	0.15	3.09							
ppDDE	Excluded	TL[R]	0.66	0.21	3.09	0.0075	0.21	1.12	1.94	1.23	3.07	0.37	17	3.05	0.004	Separate regression after significant test for interaction
PBDE-47	Not relevant	Intercept[M]	-1.37	0.59	-2.33	0.0275	-2.58	-0.16								PBDE only measured in [M]
PBDE-47	Not relevant	TL[M]	1.74	0.16	11.22	0.0000	1.42	2.06	5.72	4.16	7.86	0.81	30			PBDE only measured in [M]
PBDE-99	Not relevant	Intercept[M]	-0.83	1.10	-0.75	0.4601	-3.09	1.43								PBDE only measured in [M]
PBDE-99	Not relevant	TL[M]	1.08	0.29	3.71	0.0009	0.48	1.68	2.95	1.62	5.35	0.33	30			PBDE only measured in [M]

*more than 50% of data quantified below quality threshold (LOQ for cVMS, LOD for legacy POPs).

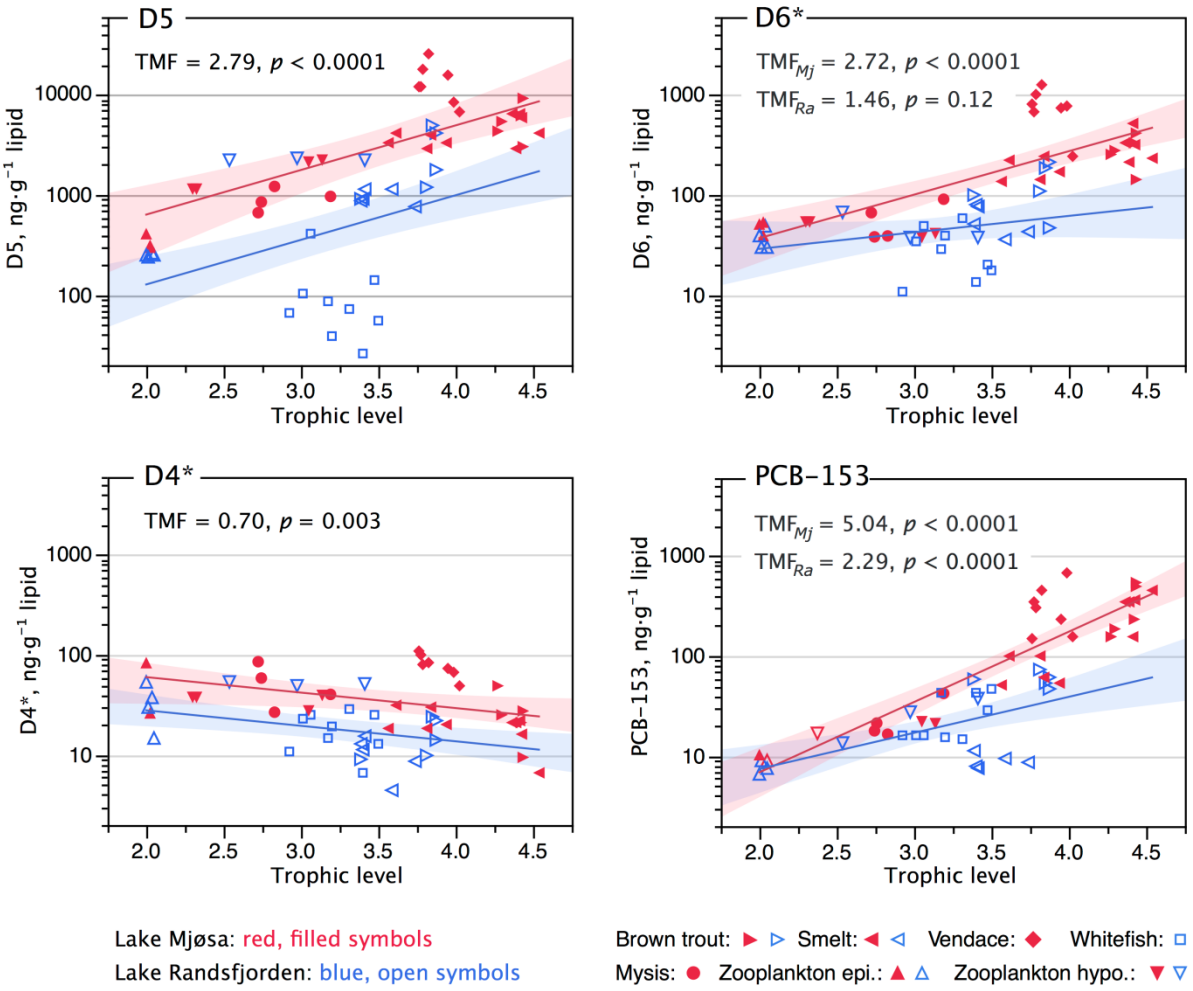

Figure S2. Relationship between lipid normalized concentrations of cVMS (D4, D5 D6) and PCB-153, and trophic level (TL) from Lake Mjøsa and Randsfjorden pelagic food webs, including whitefish from Randsfjorden. Chemicals marked with asterix (*) have >50% of data below LOQ in one or both of the lakes. Zooplankton epi and hypo are epi- and hypolimnetic zooplankton, respectively. Trophic magnification factor (TMF) estimated separately for Mjøsa (Mj) and Randsfjorden (Ra) when the interaction TLxLake was significant.

Table S 14. Product-moment correlation coefficients (r: left triangular matrix) between trophic position (TL), and log-transformed concentrations of cVMS and selected legacy contaminants in the pelagic food web in a) Mjøsa and b) Randsfjorden.

a) MJØSA[a] n = 31 (30 hvor PBDE inngår)

Variable	TL	D5	D6	D4	PCB-153	PCB-180	ppDDE	PBDE-47	PBDE-99
TL	**1.00**								
D4[c]	0.76	**1.00**							
D5	0.71	0.91	**1.00**						
D6	-0.39	0.13	0.21	**1.00**					
PCB-153	0.89	0.84	0.85	-0.15	**1.00**				
PCB-180	0.76	0.77	0.80	-0.09	0.92	**1.00**			
ppDDE	0.88	0.84	0.86	-0.14	1.00	0.92	**1.00**		
PBDE-47	0.90	0.79	0.80	-0.14	0.99	0.90	0.98	**1.00**	
PBDE-99	0.57	0.82	0.78	0.31	0.78	0.75	0.78	0.74	**1.00**

b) RANDSFJORDEN[b] Upper right diagonal without whitefish (n=17). Lower left diagonal with whitefish (n=26).

Variable	TL	D5	D6	D4	PCB-153	PCB-180	ppDDE
TL	**1.00**	0.55	0.51	-0.53	0.60	0.27	0.61
D5	0.41	**1.00**	0.78	0.14	0.53	0.39	0.53
D6[c]	0.41	0.81	**1.00**	-0.18	0.55	0.47	0.53
D4[c]	-0.51	0.23	-0.02	**1.00**	-0.18	0.01	-0.19
PCB-153	0.59	0.40	0.40	-0.23	**1.00**	0.91	0.99
PCB-180[c]	0.27	0.34	0.43	0.00	0.86	**1.00**	0.90
ppDDE	0.59	0.47	0.46	-0.20	0.97	0.85	**1.00**

a Mjøsa correlation zooplankton epilimnion and hypolimnion, Mysis relicta, vendace, smelt and trout.
b Randsfjorden correlation included zooplankton epilimnion and hypolimnion, whitefish, smelt and trout.
c correlation included >50 % values below the established limit of quantification (LOQ) for cVMS and LOD for PCB-180.

Utførende institusjon

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Oppdragstakers prosjektansvarlig

Katrine Borgå

Kontaktperson

Bård Nordbø

M-nummer

M-81

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36 + vedlegg

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Katrine Borgå (NIVA), Eirik Fjeld (NIVA), Amelie Kierkegaard (ITM), Jarl Eivind Jarl Eivind Løvik (NIVA), Sigurd Rognerud (NIVA), Andreas S. Høgfeldt (NIVA), Kine Bæk (NIVA), Michael S. McLachlan (ITM)

Tittel – norsk og engelsk

Siloksaner i ferskvanns næringsnett – et studie av tre innsjøer i Norge.

Siloxanes in freshwater food webs – a study of three lakes in Norway

Sammendrag – summary

Presence, sources and food web biomagnification of cyclic volatile methylsiloxanes (cVMS) was investigated in three Norwegian lakes (Mjøsa, Randsfjorden, Femunden). The high levels and food web biomagnification of the cVMS D5 in Mjøsa reported in 2010 was confirmed.
The cVMS levels in lakes with discharge from waste water treatment plants were higher than in the reference lake with minor human impact (Femunden), suggesting that local sources are the major input of cVMS, rather than long range transport. The cVMS analysed for (D4, D5, D6) were found in effluent water from the investigated waste water treatment plants in Mjøsa and Randsfjorden.
D5 and D6 biomagnified in the pelagic food web with trophic magnification factors above 1 in both Mjøsa and Randsfjorden. Results indicated that D4 is not subject to biomagnification, with decreasing concentrations with increasing trophic level.

4 emneord

Sykliske volatile metylerte siloksaner
Biomangifisering i næringsnett
Nivåer og kilder
Trofiske magnifikasjonsfaktorer

4 subject words

Cyclic volatile methylsiloxanes
Biomagnification in food webs
Levels and sources
Trophic magnification factors

Miljødirektoratet
Telefon: 03400/73 58 05 00 | Faks: 73 58 05 01
E-post: post@miljodir.no
Nett: www.miljødirektoratet.no
Post: Postboks 5672 Sluppen, 7485 Trondheim
Besøksadresse Trondheim: Brattørkaia 15, 7010 Trondheim
Besøksadresse Oslo: Strømsveien 96, 0602 Oslo

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