

Contents lists available at ScienceDirect

Science of the Total Environment



journal homepage: www.elsevier.com/locate/scitotenv

Occurrence and trophic transport of organic compounds in sedimentation ponds for road runoff



Merete Grung ^{a,*}, Sondre Meland ^a, Anders Ruus ^a, Sissel Ranneklev ^a, Eirik Fjeld ^a, Alfhild Kringstad ^a, Jan Thomas Rundberget ^a, Majbrit Dela Cruz ^b, Jan H. Christensen ^b

^a Norwegian Institute for Water Research (NIVA), Gaustadalléen 21, NO-0349 Oslo, Norway

^b Analytical Chemistry Group, Department of Plant and Environmental Science, Faculty of Science, University of Copenhagen, Thorvaldsensvej 40, 1871 Frederiksberg C, Denmark

HIGHLIGHTS

GRAPHICAL ABSTRACT

- Ecosystem in sedimentation ponds for road runoff receives several organic pollutants.
- Water, sediment, plants, larvae and fish were analysed for 4 contaminant groups.
- Higher levels of pollutants in sedimentation ponds vs. reference were observed.
- Bioaccumulation observed for PACs and PBDEs, but all 4 groups detected in fish.
- Biomagnification was documented for PBDEs, alkylated PACs important in road runoff.

ARTICLE INFO

Article history: Received 3 June 2020 Received in revised form 11 August 2020 Accepted 17 August 2020 Available online 20 August 2020

Editor: Dimitra A Lambropoulou

Keywords: Sedimentation pond Organic pollutants Bioaccumulation Biomagnification Polycyclic aromatic compounds (PACs) Polybrominated diphenyl ethers (PBDEs)



ABSTRACT

Sedimentation ponds have been shown to accumulate several groups of contaminants, most importantly polycyclic aromatic compounds (PACs) and metals. But also, other urban organic pollutants have shown to be present, including polybrominated diphenyl ethers (PBDEs), organophosphate compounds (OPCs) and benzothiazoles (BTs). This investigation aimed at determining the occurrence of these four groups of contaminants in sedimentation ponds and determine their transport from water/sediment to organisms. PACs, including alkylated PACs, PBDEs; OPCs and BTs were determined in water, sediment, plants, dragonfly larvae and fish from two sedimentation ponds and one reference site. Fish were analysed for PAC metabolites.

Overall, higher concentrations of all four pollutant groups were detected in water and sediment from sedimentation ponds compared to two natural lakes in rural environments (reference sites). The concentration difference was highest in sediments, and >20 higher concentration was measured in sedimentation ponds (3.6–4.4 ng/g ww) compared to reference (0.2 ng/g ww) for sum BDE6. For PACs and PBDEs a clear transport from water/sediment to organisms were observed. Fish were the highest trophic level organism (3.5–5) in our study, and all four pollutant groups were detected in fish. For PBDEs a trophic biomagnification (TMF) was found both in sedimentation ponds. TMF was not calculated for PACs since they are metabolised by vertebrates, but a transfer from water/sediment to organisms was seen. For BTs and OPCs, no consistent transfer to plants and dragonfly larvae could be seen. One

* Corresponding author.

E-mail addresses: mgr@niva.no (M. Grung), sme@niva.no (S. Meland), aru@niva.no (A. Ruus), sra@niva.no (S. Ranneklev), efj@niva.no (E. Fjeld), akr@niva.no (A. Kringstad), tru@niva.no (J.T. Rundberget), mjha@plen.ku.dk (M. Dela Cruz), jch@plen.ku.dk (J.H. Christensen).

https://doi.org/10.1016/j.scitotenv.2020.141808

0048-9697/© 2020 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).

OPC and two BTs were detected in fish, but only in fish from sedimentation ponds. It is therefore concluded that sedimentation ponds are hotspots for urban and traffic related contaminants, of which especially PACs and PBDEs are transferred to organisms living there.

© 2020 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (http:// creativecommons.org/licenses/by/4.0/).

1. Introduction

The economy of many countries relies heavily on road transport, with e.g., goods and car passengers accounting for 46% and 73% of intra-EU transport (European Commission, 2012). In addition, road freight traffic and car passenger transport are forecasted, in the period 2010–2050, to grow by about 57% and 30%, respectively (European Environment Agency, 2016). Subsequently, roads and road transportation have a huge impact on the environment in terms of land use change causing habitat defragmentation, habitat loss and spreading of invasive organisms, noise, CO₂ emissions causing climate change, emissions of particles and a range of chemicals polluting local air, soil and water bodies (European Environment Agency, 2017).

Typical examples of pollutants from roads and traffic are particles, nutrients (nitrogen and phosphorus), metals (e.g., Cu, Pb, Ni, Zn, As, Sb), road salt (NaCl) and polycyclic aromatic compounds (PACs) (Brown and Peake, 2006; Hwang et al., 2016). These road and traffic related pollutants are readily washed out from the road surface to the surrounding environment during storm events and may finally end up in the aquatic environment being potentially detrimental for aquatic organisms (Meland et al., 2010). Hence, polluted road runoff is now acknowledged by most national road administrations (NRAs) and environmental authorities as a significant source of diffuse pollution, and mitigation measures are therefore normally part of road building schemes (Meland, 2016).

Several mitigation measures exist to protect the aquatic environment from polluted road runoff. One of the most popular is wet sedimentation ponds. These ponds, having permanent water (opposed to dry detention ponds), mimic natural processes and treat road runoff by retaining particle bound pollutants through sedimentation processes. In addition, dilution, chemical and biological degradation of pollutants are also important. Well-functioning sedimentation ponds have proved to protect nearby water bodies since they significantly reduce the concentrations of many pollutants. Sedimentation ponds are therefore recognised as a robust and cheap way of protecting nearby water bodies. Hence, thousands of sedimentation ponds have been built along Norwegian and European major roads (Meland, 2016). In Norway some sedimentation ponds receive tunnel wash water, and this is the case for one of the tunnels (Vassum) investigated in this study. Since the tunnels are washed 1-12 times/y, this represents an additional pollution pressure which is pulsed.

The high number of sedimentation ponds may underline their importance and relevance in an ecological context. Sedimentation ponds have gained interest in an ecological context due to their potential capacity to conserve and promote aquatic biodiversity (Le Viol et al., 2009; Meland et al., 2020). The biodiversity in Norwegian sedimentation ponds has been investigated by (Sun et al., 2018), and the two sedimentation ponds used in our study were among the 12 investigated by (Sun et al., 2018) who found a total of 96 taxa in the sedimentation ponds investigated. Since the function of the sedimentation ponds is retention of pollutants, the sedimentation ponds may become sinkhabitats and ecological traps (Villalobos-Jiménez et al., 2016). It is therefore important to investigate the pollutants present in sedimentation ponds, and if the organisms present are affected by the pollutant pressure.

In a previous study, we investigated the levels of PACs in the same two sedimentation ponds, and found that the concentrations of sum of PAH16 in sediments were between 1900 and 4200 ng/g dw (Grung et al., 2016a; Markiewicz et al., 2017). This was one of the possible explanations for the fact that fish in Skullerud sedimentation pond had lower condition index (CI), higher level of DNA damage and higher level of PAH-metabolites in bile than fish from the nearby river. To our knowledge, Skullerud is the only sedimentation pond in Norway where a minnow (*Phoxinus phoxinus*) population has been established.

Since a range of organic pollutants are present in road runoff (Grung et al., 2017; Markiewicz et al., 2017), we wanted to complement our previous study of PACs in sedimentation ponds with other organic pollutants that are known to be present in road runoff. In a recent investigation, we identified 50 PACs, including alkylated PACs, along with other urban markers such as OPCs and BTs by non-target methods in tunnel particles. (Grung et al., 2017). Our main objective of the present study was therefore to quantify the PACs, OPCs and BTs in different matrices from sedimentation ponds, including biota. At the same time, we wanted to investigate the potential bioaccumulation of the same compounds. A positive control was therefore needed to show that bioaccumulation occurs. Since PBDEs have been detected in low concentrations in sediments from sedimentation ponds, analyses of PBDEs were included to serve as a positive control for bioaccumulation and biomagnification of the other organic pollutants.

2. Materials and methods

2.1. Study sites

Two highway sedimentation ponds (Skullerud and Vassum, hereafter denoted $S_{Skullerud}$ and S_{Vassum}) and two natural references (Svartoren and Lyseren, hereafter denoted $R_{Svartoren}$ and $R_{Lyseren}$) were included in the study (Fig. 1 and Table 1). The two sedimentation ponds have previously been investigated by us (Grung et al., 2016a). The ponds are situated in the greater Oslo area. $S_{Skullerud}$ and S_{Vassum} receive runoff from the four-lane highway E6. In addition, S_{Vassum} receives regularly tunnel wash water from three tunnels (Nordbytunnelen, Smiehagentunnelen and Vassumtunnelen). $S_{Skullerud}$ has a closed forebay and a main pool covering an area of ca. 880 m², while S_{Vassum} has both an open forebay and main pool covering an area of 400 m². Both sedimentation ponds have developed a dense vegetation within and along the edges of the ponds. The reference sites are two lakes that are in rural environments with little influence from traffic.

2.2. Sampling of water, sediment and biological material

The sampling took place in June–August 2016. All matrices were sampled in incinerated glassware and kept at -20 °C until analysis if not otherwise indicated. All biological samples except fish were pooled and analysed for each station. The sampling methods are presented in Table 2. The significance for interpretation of the sampling is shortly addressed here.

The water samples were not filtered, whereby a small amount of particles would be present. However, upon visual inspection the samples looked clear. At the sedimentation ponds, moss (*Fontinalis antipyretica*) was sampled upstream at the receiving stream and transferred to the sedimentation pond in June. The moss was sampled in August. At the reference site ($R_{svartoren}$), moss (*Fontinalis* sp.) was sampled at the same times, but not transplanted like in the sedimentation ponds. Fish from $S_{skullerud}$ and perch from $R_{Lyseren}$ were caught by net and length and weight were measured (see Table S1, supporting material). The sex of the fish was noted when possible. The bile was sampled by means of a capillary tube.

M. Grung et al. / Science of the Total Environment 751 (2021) 141808



Fig. 1. Map showing the various ponds included in the present study. Sedimentation ponds are marked with red circles and references are marked with blue circles. The road network is depicted by grey lines. The map was created in JMP using basemaps from © OpenStreetMap contributors (https://www.openstreetmap.org/copyright).

For several years, a small population of minnow (*Phoxinus* phoxinus) was established in S_{Skullerud}, and was investigated by us in 2014 (Grung et al., 2016a). We also wanted to investigate the minnows for this study, but instead of minnows we found pikes (Esox Lucius) in S_{Skullerud}. In 2016, the upstream river was flooded. This caused an overflow of $S_{Skullerud}$, whereby pikes were trapped in $S_{Skullerud}$ as the overflow subsided. After the pikes appeared, minnows have not been detected in S_{skullerud}, despite several attempts of both el-fishing and translocation of minnows from the nearby river. When designing this project, minnows were our target organism. However, since there were no minnows present in S_{skullerud}, the pikes were fished and analysed. Half of the pikes with content in their stomach contained minnows. We tried to get pikes from a nearby clean site, but no pikes of comparable size were found. We therefore analysed perch (Perca fluviatilis) from a nearby clean lake (R_{Lvseren}) to serve as a clean reference site for the pikes from S_{skullerud}. The perch were of roughly the same size as the pikes (Table S1, Fig. S3), and of the same trophic level (Figs. S1 and S2). The two reference locations (R_{Svartoren} and R_{Lyseren}) are not very far from each other in distance (12 km), with low levels of urbanisation and traffic.

Table 1
Coordinates of sampling locations and amount of traffic

Road runoff	Tunnel wash water	Location	Longitude (decimal grades)	Latitude (decimal grades)	Annual average daily traffic (AADT) ^a
Yes	No	Skullerud	10.83131	59.86217	70,100
	Yes	Vassum	10.73714	59.70937	66,326
No	No	Svartoren	10.98258	59.79632	n.a.
		Lyseren (fish)	11.13157	59.71041	n.a.

^a Obtained from www.vegkart.no (2019.03.22).

2.3. Chemical extraction and analyses

The chemical analyses are described below. The quality assurances for the different methods are given as text in the supplementary material and Tables S4–S7 (supplementary material).

2.3.1. Analyses of total dry matter, organic carbon and lipid

Total dry matter in the sediments and biota was analysed gravimetrically. Sediment subsamples were freeze-dried, crushed, and acidified (1 N HCl) and analysed for total organic carbon (TOC) by catalytic combustion at 1800 °C in a Carlo Erba 1106 elemental analyser (Carlo Erba SpA, Rodano, Italy). Aliquots of the homogenised biota material from each of the groups were used to determine the lipid content gravimetrically, after lipid extraction (cyclohexane and acetone).

2.3.2. PAC including alkylated PACs

Water samples (1 L) were added internal standards and extracted with dichloromethane (150 mL) twice under vigorous shaking (1 h). The aliquots were dried (Na₂SO₄), and the aliquots combined. Sediments and biological material were homogenised with equal amounts hydromatrix in an IKA 11® sample mill and extracted through pressurised liquid extraction in an ASE200 system. The extraction cell for the sediment samples consisted of 4 g of activated silica, bottom layer, 4 g of acidified copper powder (to bind elemental sulphur), 5 g of sample mix, and topped off with Ottawa sand. The extraction cell for the plant samples consisted of 4 g of activated silica (chlorophyll retainer), bottom layer, 2-5 g of sample mix, and topped off with Ottawa sand. The extraction cell for the animal samples consisted of 4 g 2% deactivated silica (as a fat retainer), bottom layer, 2-5 g of sample mix, and topped off with Ottawa sand. 200 µL (8 µg/mL) internal standard mix (Gallotta and Christensen, 2012) was added to the samples before extraction. The following extraction parameters were used: Pressure: 1500 psi, preheat time: 2 min, static time: 5 min, flush

Table 2

Sampling methods for all matrixes at all stations investigated.

Sample	Reference sites		Sedimentation ponds						
	R _{Lyseren}	R _{Svartoren}	S _{Vassum} S _{Skullerud}						
Water	No samples collected	Spot samples collected by means of large steel container. Water was thereafter transferred to larg glass flasks (5 L). Water samples were not filtered							
Sediment	No samples collected	3–5 sub-samples of the upper 5 cm layer were collected by means of a van Veen grab and combined.							
Pondweed	No samples collected	Leaves (no stems or roots) were collected from the surface.							
Moss	No samples collected	Moss were collectedMoss upstream thetwice at the samesedimentation pondstimepoints as firstwere collected andand second samplingtransplanted forin sedimentation2 months in thepondssedimentation ponds							
Dragonfly larvae	No samples collected	Dragonfly larvae were sampled using a pond no							
Fish	Perch collected by means of a net and killed by a blow to the head.	No fish samples collect R _{Svartoren} , only minnov present, and in S _{Vassum} species were living.	ted. In Pikes collected by vs were means of a net and killed by a blow to the head.						

volume: 70%, purge time: 60 s, static cycles: 2, temperature: 100 °C, solvent mixture: *n*-pentane:dichloromethane (90:10). Each cell was extracted twice into separate collection vials, and after concentrating under elevated temperature (40 °C), the two extracts were combined and evaporated to below 5 mL, filled to 5 mL with *n*-pentane:dichloromethane (90,10) and added 200 μ L 8 μ g/mL recovery standard mix (Gallotta and Christensen, 2012) (Tables S2 and S3, supporting material). For each matrix, a corresponding matrix blank was made.

GC–MS analysis: The method described in Gallotta and Christensen (2012) was utilised with a few minor differences. The extracts were analysed for PACs and alkyl PACs on an Agilent 5975C inert XL MSD with electron ionization operating in selected ion monitoring (SIM) mode. The GC parameters were as follows: 1 L sample was injected in splitless mode (inlet: 300 °C) to a 60 m HP-5 capillary column with 0.25 mm inner diameter, 0.25 μ m film thickness. The flow rate was 1.1 mL/min. The initial temperature (40 °C) was held for 2 min, ramped with 25 °C/min to 100 °C, followed by 5 °C/min to 315 °C and held for 14 min (total run time: 61.4 min). Temperatures: transfer line: 315 °C, ion source: 230 °C, quadrupole: 150 °C.

Peaks were integrated using Chemstation V2.0 (Agilent technologies, Inc.) and the data was quantified using the internal standard method and corrected for recoveries. A six-point calibration curve was used for each compound (range: 2–90 ng/mL solvent) with internal and recovery standards as given in Table S2 (supporting material) for sediment and biota and in Table S3 (supporting material) for water samples. For each EPA PAH16, the calibration curve was constructed using the compound in question. For the alkylated PACs, one selected compound representing the group was chosen for the calibration curve, except for C1-naphthalenes where the average of two calibration curves constructed from 1-methylnaphthalene and 2-methylnaphthalene was used. Please see supporting material for information on quality assurance and quality control.

2.3.3. PAH-metabolites in bile of fish

Analyses of PAH-metabolites in bile of fish was based on a method described by Krahn et al. (1992) and is described in more detail in Kammann et al. (2013). Briefly, bile (20 μ L) was mixed with internal standard triphenylamine (10 μ L) and diluted with deionised water (50 μ L). Thereafter, a hydrolysation step with β -glucuronidase/arylsulphatase (10 μ L) was performed (1 h, 37 °C). Methanol (20 μ L) was added, the sample centrifuged, and the supernatant was transferred to HPLC vials. The HPLC used was a Waters 2695 Separations

Module with a 2475 fluorescence detector attached. The column was a Waters PAH C18 ($4.6 \times 250 \text{ mm}$) with 5 µm particles. The mobile phase consisted of a gradient from 40:60 acetonitrile:ammonium acetate (0.05 M, pH 4.1) to 100% acetonitrile at a flow of 1 mL/min. The column temperature was kept at 35 °C. Fluorescence was measured at (excitation/emissions wavelengths: 1-OH-phenanthrene 256/380; 1-OH-pyrene 346/384; triphenylamine 300/360; 3-OH-benzo[*a*]pyrene 380/430). 25 µL of extract was injected for each analysis. The results were calculated by use of the internal standard method. The calibration standards utilised were obtained from Chiron AS, Trondheim, Norway; and were in the range 0.2–200 ng/g.

2.3.4. PBDEs

The analytical method for PBDEs was based on a method described by Covaci et al. (2002). Water samples (1 L) were added internal standards and extracted with dichloromethane (150 mL) twice under vigorous shaking (1 h). The aliquots were dried (Na₂SO₄), and the aliquots combined. Freeze dried sediment samples (2-5 g) were extracted with dichloromethane (20 mL) in an ultrasonic bath for 120 min. Internal standards for were added during the first extraction step. This step was repeated with fresh dichloromethane (20 mL) for 60 min and the extracts combined. Biota samples were homogenised and added sodium sulphate and extracted with cyclohexane: ethyl acetate (1,1). The extracts for analyses were cleaned by partitioning with concentrated sulphuric acid, and thereafter acetonitrile, reduced in volume, and analysed by means of GC/MS. Analysis for PBDE congeners was performed with a Hewlett Packard 6890Plus GC linked to a Hewlett Packard 5973 MS detector operated in negative chemical ionization (with methane) and SIM mode. A pulsed splitless injection (4 mL, injector temperature of 280 °C and pulse pressure of 50 psi held for 2 min) was used to transfer analytes onto a DB-5MS column (Agilent Technologies; 15 m, 0.25 mm i.d., 0.1 mm film thickness). The oven temperature was set to 120 °C and held for 2 min before being increased to 345 °C at the rate of 25 °C/min and then held for 5 min. The helium flow was set to 1 mL/min for the first 13 min and increased to 1.4 mL/min. at the rate of 0.1 mL/min and held for a further 8 min. Temperatures of the ion source, quadrupole, and transfer line were 250 °C, 150 °C, and 325 °C, respectively. Ion fragments m/z 79 and 81 were used for qualifying and quantifying PBDEs. Internal standards used for PBDEs were BDE-119 and BDE-181.

2.3.5. Organophosphate compounds (OPCs)

The method for OPCs has previously been described previously (Allan et al., 2018), and therefore only an overview is described here. Deuterated OPCs were used as internal standards for all sample matrices and were added during the extraction step. Water (200 mL) was extracted by solid phase extraction (Oasis HLB (500 mg) cartridges). OPCs were eluted with methanol:hexane (50,50). Freeze dried sediment samples (0.5 g) were extracted ultrasonically with dichloromethane (4 mL, 60 min) twice. Biota samples (for fish the filet was used) were homogenised before extraction with acetonitrile:ethyl acetate (70,30, 10 mL). Extracts were concentrated and added sodium sulphate to remove water. Supercritical fluid chromatography (SFC) with MS/MS analysis was used for quantification. The following OPCs were analysed (CAS registry number in parentheses): TCEP (115-96-8), TCPP (13674-84-5), TiBP (126-71-6), TDCPP (13674-87-8), TBOEP (78-51-3), TnBP (126-73-8), TPP (115-86-6), DCP (26444-49-5), TCP (1330-78-5), EHDP (1241-94-7) and TEHP (78-42-2).

2.3.6. BT

The analytical method was based on a method described by Asheim et al. (2019). Water (200 mL) was added internal standard (sulfamethoxypyridazine-d3 which was used for all matrices) and added onto Oasis HLB (500 mg) solid phase extraction cartridges. BTs were eluted with 6 mL methanol:hexane (50,50). Freeze-dried sediment samples (0.5 g) were added internal standard and extracted ultrasonically with methanol (5 mL, 60 min) twice. Biota samples were extracted as described for OPCs. BTs were analysed by LC-MS/MS with the following conditions: They were separated on an Aguity UPLC (Waters, Manchester) using a BEH C8 column (100×2.1 mm, 1.7μ m) (Waters, Sweden) with an acetonitrile and water (5.2 mM ammonium acetate) mobile phase. Gradient elution was from 50% to 100% acetonitrile over a 10 min program. The UPLC system was connected to a mass spectrometer (Xevo G2S QToF, (Waters, Manchester)) operated in electrospray ionization mode. Detection limits (DL) was calculated for each sample individually using the standard method of calculation of 3 x s/n ratio. It was not possible to use labelled internal standards because these were not commercially available. This will result in an uncertainty of 35-50% depending on the analytes and sample matrix. Very few of the BTs were available as certified standards, where available they were purchased from Sigma-Aldrich (Germany). The following BTs were analysed: 2-hydroxybenzothiazole (2-OH-BT), 2-Phenyl-1,3-benzothiazol (phenyl-BT), 2-butyl-1,2-benzisothiazol-3 (2H)-one and N-cyclohexyl-2-benzothiazosulfenamide (the two last BTs were not detected in any matrices).

2.3.7. Stable isotopes

Stable isotope of nitrogen (δ 15N air) was analysed and used for assessment of trophic level of biota samples. δ 13C VPDB (Vienna Pee Dee Belemnite) was also included to identify the carbon sources. The Isotope analyses were performed by the Institute for Energy Technology (IFE-Kjeller, Norway) according to standard protocols (Ruus et al., 2015). The stable isotopes were calculated according to Eq. (1) as follows:

$$\partial X = \left(\frac{R_{Sample}}{R_{Standard}} - 1\right) \times 1,000 \tag{1}$$

2.4. Calculation of BAF, BSAF and trophic biomagnification

The bioaccumulation factors (BAF) were calculated according to the Eq. (2) (Borgå and Ruus, 2019):

$$BAF = \frac{C_{biota}}{C_{water}}$$
(2)

 C_{biota} is the concentration of a compound (based on the wet weight) C_{water} is the concentration of a compound in water

The biota to sediment accumulation factors (BSAF) were calculated according to Eq. (3):

$$BSAF = \frac{C_{biota \ (hw)}}{C_{sediment \ (oc)}}$$
(3)

 $C_{\text{biota}\ (lw)}$ is the lipid normalised concentration of a compound in biota

 $C_{sediment\ (oc)}$ is the organic carbon normalised concentration of a compound in sediment

The relative trophic level (TL) of each sample (consumer) was calculated from $\delta^{15}N$ (Eq. (4)) using an enrichment factor ΔN of 3.4‰ between integer trophic levels (Borgå et al., 2012). The lowest plant $\delta^{15}N$ (Potamogeton) was defined as the baseline primary consumer of trophic level 1:

$$TL_{consumer} = \frac{\partial^{15} N_{consumer} - \partial^{15} N_{plant}}{\Delta N} + 1$$
(4)

Trophic magnification factors (TMFs) were calculated as the antilogarithm of the slope (b) of the linear regression between the natural logarithm of the lipid normalised contaminant concentration and the trophic level of the sample/species in question (Fisk et al., 2001) (Eqs. (5) and (6)):

$$\ln [contaminant]_{bu} = a + b \times TL \tag{5}$$

$$TMF = e \times b \tag{6}$$

TMFs were only calculated for the brominated flame retardants BDE47, BDE99 and BDE153 as too many samples had concentrations below the DL for other compounds analysed.

2.5. Statistics

Data treatment, statistical analyses and graphical outputs were performed with JMP version 15.0.0 (SAS Institute Inc.). Regression analyses were performed by the least square's method.

3. Results and discussion

3.1. Occurrence in water

The water samples were not filtered prior to analyses, therefore the concentrations reported may be a result of analytes bound to particles. The concentrations for all investigated contaminants are listed in Supplementary Tables S8 and S9. Only two PBDEs (BDE47 and 99) were detected in water samples collected from all three sampling sites, while BDE153 was only detected in the sedimentation ponds ($S_{Skullerud}$ and S_{Vassum}). The concentrations were consistently lower in $R_{Svartoren}$ (reference site), and the \sum BDE6 was 0.006 ng/L in $R_{Svartoren}$, and 0.023–0.033 ng/L in S_{Vassum} and $S_{Skullerud}$ respectively. These levels are considerably lower than the maximum allowable concentration (MAC) in the water framework directive (EU European Union, 2013) 140 ng/L, based on acute toxicity studies. The WFD has not listed an annual average (AA) based on chronic toxicity data for PBDEs.

The low ring PACs (up to 4 rings) were detected in water sampled from all three sites investigated, but the higher ring sizes were only detected in the sedimentation ponds. The levels were highest in $S_{Skullerud}$ where PAH16 was 170 ng/L, while the concentration in $R_{Svartoren}$ and S_{Vassum} was 6.6 and 22 ng/L respectively. Several of the PACs (anthracene, fluoranthene, naphthalene and benzo[*a*]pyrene) have been assigned AA and MAC in freshwater systems. Only the AA of benzo[*a*] pyrene (0.017 ng/L) was exceeded in $S_{Skullerud}$, while the other results were below AA and MAC. Since the DL for benzo[*a*]pyrene was higher (0.7 ng/L) than the AA it is not known if the AA was exceeded in S_{Vassum} and $R_{Svartoren}$.

All OPCs investigated were detected in water samples from $S_{Skullerud}$ and S_{Vassum} , while only half of the investigated OPCs were detected in $R_{Svartoren}$ (Table S5, supporting material). The levels were higher in water from the two sedimentation ponds than from $R_{Svartoren}$. For TCEP, TiBP, TPP, TDCPP. TnBP and TPP the levels were more than 5 times higher. TCEP and TCPP were observed in the highest levels (up to 410 and 2000 ng/L respectively). Only TCEP among the OPCs has a national guidance for AA and MAC in water, 65 and 510 µg/L respectively. Both sedimentation ponds (140 and 410 ng/L) therefore exceeded the AA but not the MAC. In $R_{Svartoren}$ TCEP was below DL.

Previous investigation of the levels of OPC in urban areas have been done. In a study of tunnel wash water, many of the same OPCs were detected in high concentrations (Meland and Roseth, 2011). Their findings suggested that sedimentation ponds are a suitable mitigation strategy for removing OPCs, but removal percentage increased by adding peat or active carbon filter sorbents. S_{Vassum} receives tunnel wash water, while S_{Skullerud} receives only road runoff. The levels of OPCs in snow nearby airport, road and reference were measured by Marklund et al. (2005). They observed a general decrease in OPC concentrations with distance from the road (2, 100 and 250 m). In line with our findings,

TCPP was found in high concentrations (110–170 ng/kg), also at the reference site (68 ng/kg) Marklund et al. (2005). Yet, the levels of TCPP in our study were substantially higher in sedimentation ponds (1400–2000 ng/L). Also, the levels of TCEP in sedimentation ponds (140–410 ng/L) in our study were substantially higher than what was measured in snow nearby a road in Umeå, Sweden (12 ng/kg) Marklund et al. (2005). A study of water from rivers in South Korea was investigated by Yoon et al. (2010). Both clean sites and sites influenced by effluents from sewage treatment plants (STPs) were investigated. Again, the levels of TCPP in sedimentation ponds were higher than in creeks strongly influenced by STP effluent. In a study of rivers in Spain (Cristale et al., 2013) levels were in general low, but near urban influence (Barcelona) and STPs, levels up to 1800 ng/L (TCPP) and 330 ng/L (TCEP) were observed.

In a non-target study of tunnel particles, three BTs were identified in tunnel particles, BT, 2-OH-BT and phenyl-BT (Grung et al., 2017). BTs in traffic environment are strongly linked to emissions of tire wear (Asheim et al., 2019). Regardless of that, BTs have gained little attention in previous pollution studies of road runoff compared to other organic pollutants (e.g. PACs). Only two of the BTs investigated were detected in any of the matrices investigated, 2-OH-BT and phenyl-BT (Table S8 and S9, supporting material). For the water phase, only phenyl-BT was detected in low concentrations in water from S_{Skullerud} and S_{Vassum} (6.6–19 ng/L), but not in the water from R_{Svartoren}. BTs are not listed in the WFD.

Water is not the preferred analytical matrix for many of the compounds reported here, especially PACs and PBDEs with a high logKow (range 3.34–6.22 and 5.6–8.27 respectively) are mainly particle bound. In addition, the concentrations in water will be highly variable since a dilution will take place after each rainfall. The analysis of the water was done mainly to account for compounds with a low logKow (OPCs (1.44–9.49) and BTs (1.8–4.26)), and to calculate BAFs of compounds that were detected both in water and in biota. However, because of the highly variable concentrations and since we have only done one measurement of water, the results reported here must be interpreted with caution. Yet, we believe that the general pattern that all the compound classes investigated have higher concentrations in water from sedimentation ponds than at the reference site are valid.

3.2. Occurrence in sediment

Elevated levels of PACs were detected in sediment from sedimentation ponds S_{Skullerud} and S_{Vassum} compared to the reference site (R_{Svartoren}) (Table S8, supporting material). PACs are the most abundant pollutant in the sedimentation ponds of the target analytes included in this study as can be clearly seen from the sediment results. Some PACs are regulated in the WFD. In addition to the environmental quality standards (EQS) for the EU prioritised compounds (EU European Union, 2013), Norway has EQS for the sum of PAH16 since PACs are considered an environmental risk in Norway (Direktoratsgruppen vanndirektivet, 2018). The PAH16 concentrations in sediments are listed in Table 3 and are classified according to the national guidelines. In short, a yellow, orange or red colour indicate exceedance of the EQS, and a higher level of toxicity from yellow to red, while blue (background), and green (no toxic effects) are concentrations not exceeding EQS. In addition, Norway's National EQS for sum of PAH16 has been used for its usefulness for an overall risk (Direktoratsgruppen vanndirektivet, 2018). The concentrations of PAH16 in sedimentation ponds were similar to the levels that was previously reported for $S_{Skullerud}\xspace$ and $S_{Vassum}\xspace$ in 2016 (Grung et al., 2016a). The concentration in S_{Skullerud} then was higher (4200 vs. 3000 ng/g dw now) while concentration in S_{Vassum} was similar (1900 vs. 2200 ng/g dw now).

Contrary to our previous analyses of PACs in sediment, this time the alkylated homologues of selected PACs (naphthalene, fluorene, phenanthrene/anthracene, pyrene/fluoranthene and chrysene) were also analysed in sediment (Table). As can be clearly seen, the alkylated PACs were very high in sediment from $S_{Skullerud}$ and S_{Vassum} , while substantially lower/non-existing at $R_{Svartoren}$ (Fig. S4, supporting material). High occurrence of three-four ring alkylated PACs is an indication of petrogenic origin of the PACs and may stem from the bitumen of the asphalt, spilled lubricating oils or soot. Commercial extraction of bitumen has led to increased environmental levels of alkylated PACs in a lake in Alberta, Canada (Korosi et al., 2013). In Norway, asphalt wear is particularly high in the winter season when studded winter tires are commonly used. The alkylated PAC pattern was typically bell-shaped (Stogiannidis and Laane, 2015), with the C3 alkylated homologues as the highest, indicating a petrogenic origin (Fig. S4, supporting material). The alkylated chrysenes were of the highest concentrations among the alkylated PACs, and the histogram of PACs of the sedimentation ponds resembled the road asphalt shown in Stout et al. (2004).

By assuming that the alkylated homologues exert the same toxicity as the non-alkylated PACs (Richter-Brockmann and Achten, 2018; Wayland et al., 2008), an estimation of the associated risk can be made. It is evident that inclusion of alkylated homologues of PAH16 changes the risk of hazardous effects to sediment living organisms (Table 3). Notably, the change in the environmental risk of chrysene has changed dramatically for the two sedimentation ponds. Also, the change in the classification for PAH16 has changed from exceeding the AA (yellow) to exceeding the MAC (orange) for the two sedimentation ponds, while R_{Svartoren} has changed classification from background (blue) to good (green). Since the toxicity of all alkylated PACs have not been investigated, this is for the present only an approximation. However, we believe that until the toxicity has been estimated, this approximation is a valid precautionary principle.

There are several reports linking exposure of alkylated PACs to toxicity (Andersson and Achten, 2015). Raine et al. (2017) found that lower embryo survival corresponded to higher total and alkylated PAC content in a 31 day exposure of fish to oil sands tailings pond sediments in the laboratory. In addition, delay in development and increased percentages of larvae with heart and yolk sac oedema, cranial and spinal malformations were observed. Lee et al. (2017) showed that PACs (naphthalene, fluorene, dibenzothiphene, phenanthrene and chrysene) and their alkylated analogues disrupt endocrine functions. Especially 1methylchrysene, followed by phenanthrene and its alkylated analogues possessed endocrine mediating potencies. In a binding study with aryl hydrocarbon receptor (AhR), 1-methylchrysene and 1,2,6,9tetramethylphenanthrene were shown to possess high bindingpotencies (145% and 83% of TCDDmax respectively) (Lee et al., 2015). Vignet et al. (2014) ascribed a large part of the toxicity to alkylated PACs for the toxicity observed after exposure to PAC fractions representative of either pyrolytic or petrogenic composition.

The petrogenic composition are characterised with high occurrence of alkylated PACs, especially phenanthrenes, naphthalenes and anthracenes. In addition to the toxic concerns of alkylated PACs, several investigations have shown that alkylated PACs are more persistent in the environment than the parent PACs (Barron and Holder, 2003). We therefore believe that environmental management need to take into consideration the increased risk associated with alkylated PACs in the sedimentation ponds. We have previously shown that alkylated PACs are found in tunnel particles (Grung et al., 2017), and the same is true for sedimentation ponds receiving road runoff.

The levels of PBDEs in sediments were quite low both in sedimentation ponds and reference site. The \sum BDE6 were 4 ng/g dw in both sedimentation ponds and 0.20 ng/g dw in R_{Svartoren}. These concentrations are substantially lower than the EQS for sediment (310 ng/g dw). There are few investigations of PBDEs in sedimentation ponds, probably since PBDEs are not assumed to be one of the major organic pollutants in such ponds. The results reported for PBDEs are somewhat lower than previously reported concentration in S_{Vassum} of 21 ng/g dw of penta-BDEs (Meland, 2012). In a study of stormwater ponds in South Carolina (Crawford et al., 2010), no PBDEs were detected above the detection limit of 6–10 ng/g dw.

Table 3

PAH16 and alkylated PACs in sediment (µg/kg dw). Sum alkylated PACs are the sum of C1–C4 naphthalenes, C1–C3 fluorenes, C1–C4 phenanthrenes/anthracenes, C1–2 pyrenes/ fluoranthenes and C1–C3 chrysenes. Percent alkylated are the percentage of alkylated divided by the sum of PAC and alkylated PACs. Letters in red are above the detection limit (DL), but below the LOQ (limit of quantification). The colour of each cell indicate classification according to Direktoratsgruppen vanndirektivet (2018). PACs marked with an asterisk are prioritised pollutants according to EU, while the others are prioritised by Norway, including the sum of PAH16. Two significant digits are used.

		R _{Svartoren}			S _{Skullerud}		S _{Vassum}			
Sample name	PAC	Sum alk. PAC	% alk	PAC	Sum alk. PAC	% alk	PAC	Sum alk. PAC	% alk	
Naphthalene*	45	76	63	79	470	86	56	310	85	
Acenaphthylene	9			32			18			
Acenaphthene	4			9			5			
Fluorene	4	nd		28	1,200	98	61	1,400	96	
Phenanthrene	22	20	42	190	2 400	04	200	3,200	93	
Anthracene*	4	20	45	30	5,400	94	25			
Fluoranthene*	32	han		360	1 000	64	310	1,700	66	
Pyrene	29	na		730	1,900		570			
Benzo(a)-anthracene	11			80			50			
Chrysene	18	18	50	400	8,000	95	270	6,100	96	
Benzo[b]fluoranthene*	23			240			130			
Benzo[k]fluoranthene*	18			110			66			
Benzo[a]pyrene*	11			130			69			
Indeno(1,2,3-	15			150			82			
c,d)pyrene*										
Dibenzo[ah]anthracene	nd			58			37			
Benzo[ghi]perylene*	14			330			220			
∑РАН16	260	110	30	3,000	15,000	83	2,200	13,000	86	

In sediment samples, fewer OPCs were detected than in water, and TCEP, TiBP, and TnBP were not detected in any sediment samples. Overall, levels were higher in sediment from $S_{Skullerud}$ and S_{Vassum} than from $R_{Svartoren}$. The highest levels were observed for TEHP (530 ng/g dw) and TCP (70 ng/g dw) which are among the OPCs investigated with highest logKow. Only for the OPCs with logKow >5 there was a clear tendency that the concentrations in sediments from sedimentation ponds were higher than from the reference site.

BTs were detected in sediment samples from both sedimentation ponds, and phenyl-BT (90–140 ng/g dw) and 2-OH-BT (520–1000 ng/ g dw) were detected. No BTs were detected in the sediment from $R_{Svartoren}$. In a study of BTs in road dust samples from Trondheim in Norway (Asheim et al., 2019), 2-OH-BT was found in concentrations of 100–1100 ng/g dw depending on the season. 2-OH-BT constituted roughly 70–90% of total BTs investigated, and 2-OH-BT is a probable degradation product of several other BTs. In a study of BTs in road dust and roadside soil, 2-OH-BT was also the major BT in both matrices alongside benzothiazole (Zhang et al., 2018).

3.3. Occurrence in biota

The data for concentrations in plants and dragonfly larvae are shown in Table S8 (supporting material), while the concentrations in individual fish are shown in Table S9 (supporting material). The transplantation of moss showed little accumulation of contaminants. This was probably due to contamination from traffic reaching the locations upstream the sedimentation ponds where the mosses were sampled. In Table S8 (supporting material), the mosses have therefore been treated as two individual samples of moss (Table S8).

The PAC concentrations were in general twice as high in biota from sedimentation ponds than from the two reference sites. The concentrations in plants were lower (on a dry weight basis) than the sediments, and in general few (2–4) PACs were measured in plants and dragonfly larvae. In our previous investigation (Grung et al., 2016a) much higher levels in pondweed were observed than in this study, whereas the levels in dragonfly larvae were similar. In our previous study, the root of the pondweed was included, but this time, only leaves of the plants were analysed. According to Gao and Zhu (2004), the uptake of PACs in pondweed is mainly by the roots.

PACs are not accumulated in fish, but metabolised and excreted via bile. However, the PAH-metabolites in bile are exposure markers for approximately the last week prior to sampling (Jonsson et al., 2004). Therefore, bile samples were analysed for PAH-metabolites, and results for 1-OH-pyrene and 1-OH-phenanthrene are shown in Fig. 2. The ICES guidance limits for background assessment criteria (BAC) and environmental assessment criteria (EAC) for marine fish species (Hylland et al., 2012) are indicated. 1-OH-pyrene and 1-OH-phenanthrene were not detected in bile of fish from R_{Lyseren}. In S_{Skullerud}, 1-OH-pyrene and 1-OH-phenantrene were detected in all fish. The minnows from S_{Skullerud} in our previous study had significantly higher levels of 1-OH-pyrene (median 3600 (min-max, 480–12,000 ng/g bile)) than the pikes. One explanation may be the difference in diet. Minnows were probably a substantial part of the pikes' diet. Minnows have already metabolised the PACs, thereby facilitating a rapid excretion in the pikes, and lowering their exposure to PACs.

There are some limitations to interpretation of data from fish. The species are not the same but are similar in size trophic level and eating habits. More importantly the location of reference for fish ($R_{Lyseren}$) are different from reference site for water and sediment ($R_{Svartoren}$). The plan was to investigate minnow which had been living in $S_{Skullerud}$ for years and was investigated in our previous study (Grung et al., 2016a). However with this drawback of fish species and location in mind some important conclusions can be drawn from the comparison of perch from $R_{Lyseren}$ and pike from $S_{Skullerud}$

In biota, the detection frequency of OPC were much less than for water and sediments. Only for EHDP and TEHP a consistent detection in plants were observed. Only TPP was detected above the DL, and only in fish from $S_{Skullerud}$. The detection frequency was 45%, and the mean level was 1.5 ng/g ww. For BTs there were very few detections in plants. Both 2-OH-BT and phenyl-BT were detected in fish from $S_{Skullerud}$, but not in fish from $R_{Lyseren}$. 2-OH-BT was detected in many of the fish, 82% had concentrations above the DL. The mean level was 11 ng/g ww (range <1–16). 2-OH-BT is the probable transformation product of several BTs, and an increase in concentration after a period of storage of 2-OH-BT has been observed in other studies at NIVA. Phenyl-BT had a lower detection frequency (36%), and the concentrations were also lower (mean 0.40 ng/g ww, range <0.30–0.42).



Fig. 2. 1-OH-pyrene (left) and 2-OH-phenanthrene (right) in fish bile, both in ng/g bile. The highest and lowest BAC and EAC for different marine fish species are indicated for both metabolites. The most frequently DL is indicated for each metabolite as dashed lines. The concentrations below DL have been replaced by half DL

The levels of \sum BDE6 in plants and dragonfly larvae were low in matrices from both reference site (R_{Svartoren}) and sedimentation ponds. However, the levels in matrices from sedimentation ponds were higher than matrices from the reference site, and more congeners were detected in matrices from sedimentation ponds. The levels in perch from R_{Lyseren} were comparable to brown trout from two non-polluted lakes in Norway (lake Femunden 0.49 ng/ g ww, and lake Eikedalsvannet 0.18 ng/g ww) (Jartun et al., 2018). The brown trout in lake Mjøsa in Norway used to be heavily contaminated with PBDEs due to releases from a nearby factory, but the levels have been significantly reduced from >300 to 8.0 ng/g ww in 2017 (Jartun et al., 2018). The current levels in pike from Lake Mjøsa were comparable to the levels observed in pike from R_{Lvseren} (9.3 ng/g ww). The levels in pikes from S_{Skullerud} were significantly higher than in perch from R_{Lvseren}. The levels of \sum BDE6 from both locations were higher than the EQS. (0.0085 ng/g ww). The EQS of \sum BDE6 is often exceeded in fish species, also in pristine rivers in Norway (Moe et al., 2019). Analysing the liver with higher fat content than filet must however be considered a worst-case scenario, and levels in filet were probably lower.

3.4. Transfer of contaminants to plants and biota

The concentrations in water, plants and organisms are shown in Fig. 3. Since few compounds were detected in all matrices, bioaccumulation factors were calculated using the sum of the contaminant groups. This is an approximation of the reality but illustrate the difference in potential of bioaccumulation for the four different contaminant groups. The bioaccumulation potential of \sum BDE6 can clearly be seen, while PACs also were shown to accumulate in plants. However, PACs are metabolised by most vertebrates, and therefore further bioaccumulation in higher trophic levels is discontinued. For these two groups, there was an obvious pattern that levels in sedimentation ponds were higher than in reference for all matrices. For the OPCs there was no indication of bioaccumulation, while for BTs there seemed to be higher levels in plants and organisms than in water. However, especially for BTs with exception for sediments and fish, there was no consistent pattern of higher levels in sedimentation ponds than reference site. The analyses of BTs in water and organisms were challenging.

BAFs, BSAFs and TMFs were estimated for the compounds/sites where concentrations were above DLs. The results are shown in



Fig. 3. Figures showing transfer of contaminants. The concentrations are shown in ng/g ww for all compounds, and the range of the vertical axis (concentration range) are the same for all compounds. The figures depict the sum of compounds within the same chemical group. When a compound has not been detected in a matrix it is omitted from the figure. PACs were only analysed as PAH-metabolites in fish which are not shown in the figure.

BAFs and TMFs calculated for different compounds and sites. The compounds have been ordered according to their logKow values (from pubchem.ncbi.nlm.nih.gov). Where no numbers for BAF/TMF are presented, one or both required concentrations were below DL/LOQ. Numbers for BAFs and TMFs are given with two significant digits. The BAF means have been calculated by taking the mean of only fish with positive detections of the compound in question. The lowest BAF has been calculated based on the DL

		TMF	Fish									4.0		5.6		2.4	
			Fish average min - max	20	<15-130	110	<30-180					520,000	370,000-700,000	240,000	160,000-430,000	34,000 21,000-50.000	
			Dragonfly					200	520			600				670	
			Potamogeton					65	300			260		92		100	
	Skullerud	BAF	Moss					340	1400		61	8900		5000		2500	
n ponds			Dragonfly					940	069			340		670		1200	
			Potamogeton					780	1100			340		240		290	
Sedimentat	Svassum	BAF	Moss					420	660		62	10,000		3600		0066	
		TMF	Fish									3.4		3.2		1.9	
	$R_{Lyseren}$	BAF	Fish average min - max									150,000	47,000-420,000	110,000	33,000-250,000		
			Dragonfly					4700	1800	43		1000		1500			
			Potamogeton					2400	1500		120	530					
Reference	R _{Svartoren}	BAF	Moss					3100	2700	43	52	5100		6400			
			Log Kow	4.26		4.59		4.88	5.16	5.48	5.73	6.81		7.32		7.90	
			Compound	Phenyl-BT		TPP		pyrene	fluoranthene	TCPP	EHDP	BDE47		BDE99		BDE153	

Table (BAFs and TMFs) and (BSAFs). The results for BAFs must be treated with caution since water samples were taken as point samples, thereby violating the criterion of long-term average conditions (Burkhard, 2003).

From Tables 4 and 5, BAFs and BSAFs could be calculated for some PACs and PBDEs, and only in a few cases for BTs and OPCs. This implies that most BTs and OPC are not prone to transfer from water/sediment to biota in substantial concentrations. Fish was the only species with consistent findings of BTs and OPCs in biota, however the detection frequency was between 36 and 82% indicating that the uptake is variable. For phenyl-BT, a low mean BCF of 20 for the fish with detection of the compound was found. No 2-OH-BT was detected in water so BAF could not be calculated. We did not analyse the lipid content of the fish filets, so BSAF could not be analysed. The 2-OH-BT occurred in high concentrations in sediment but did not occur above DL in water. One explanation is that the fish were exposed to the compound in sediment. More probable is that the occurrence of 2-OH-BT is a result of metabolism of other BTs that transform/degrade to 2-OH-BT. The BCF of TPP in different species of fish has previously been shown to be between 100 and 500 in different studies according to hazardous substances data bank (HSDB) (US National Library of Medicine, n.d.), and our results are therefore in accordance with this. Also, BAF for TCPP and EHDP were calculated for moss, but were not very high.

The PACs and PBDEs had significantly higher BAFs than the BTs and OPCs. For PACs, the BAFs were higher in reference than in sedimentation ponds, while the BDEs were in the same order in sedimentation ponds and reference. Since water samples were not filtered prior to analysis, it is possible that the high BAFs in sedimentation ponds can be ascribed to high measured concentrations in the water phase that are not available for bioaccumulation in biota. The runoff likely contains fine particles with a large surface for adsorption of hydrophobic contaminants. Furthermore, they likely contain soot/black carbon from combustion with a high adsorptive capacity (Burgess et al., 2004; Rust et al., 2004). The BAF for BDE47 and BDE99 were higher than pyrene and fluoranthene, especially in the sedimentation pond. One explanation can be that PACs are prone to biotransformation, and in the algae Chara we have demonstrated a biotransformation of pyrene (Grung et al., 2016b) suggesting that biotransformation may be an option. We have also demonstrated low concentrations of PAH-metabolites in dragonfly larvae, demonstrating a low rate of metabolism is a possibility (Girardin et al., 2020).

Based on occurrence in potamogeton, dragonfly larvae and fish, the TMFs for BDE47, BDE-99 and BDE-153 could be calculated for sedimentation ponds and reference sites including all relevant concentrations from $R_{Svartoren}$ and $R_{Lyseren}$, $S_{Skullerud}$ and S_{Vassum} (Fig. S5). The assumptions are that the two sedimentation ponds represent a different environment than the two reference sites. The mean of the trophic levels for the perch from $R_{Lyseren}$ was higher than the mean for $S_{Skullerud}$ (see Fig. S5), and thereby if anything higher levels of BDEs would be expected.

The levels of PBDEs in plants vs. fish showed a trophic biomagnification as expected for BDE47, BDE99 and BDE153 with positive slopes (TMF > 1) at both reference sites and $S_{Skullerud}$ (Table 4). The slopes tended to be higher in $S_{Skullerud}$ than in reference sites, but the uncertainties are too high to draw any conclusions if this a general trend. One striking feature from the figures is that the levels of BDE in dragonfly larvae were very low based on the high trophic level of the organism. This may be explained by the fact that whole organisms were homogenised, including the exoskeleton. Girardin et al. (2020) has shown a substantially lower lipid content of the exoskeleton than tissue in these organisms (7 vs. 2% ww). PBDEs could therefore have been analysed only in the tissues of dragonfly larvae to better account for the internal accumulation of PBDEs.

4. Conclusions

Sedimentation ponds are the most common mitigation strategy for road runoff, and therefore contain traffic related organic compounds

Table 5

BSAFs calculated for different compounds and sites. Where no numbers are presented, one or both of the required concentrations were below LOQ. Numbers are given with two significant digits.

Reference						Sedimentation ponds							
Site R _{Svartoren}				R _{Lyseren}	S _{Vassum}			S _{Skullerud}					
Matrix	Moss	Moss Potamogeton Dragonfly		Fish average min - max	Moss	Potamogeton	Dragonfly	Moss	Potamogeton	Dragonfly	Fish average min - max		
Pyrene Fluoranthene	0.33 0.44	0.084	0.016 0.019		0.47 0.46	0.061 0.11	0.021 0.02	0.46 0.42	0.053 0.03	0.017 0.02			
BDE47	1.2	1.7	0.12	7.5 2.8–21	0.23	0.059	0.016	0.28	0.026	0.0069	2 1.2–2.9		
BDE99	0.32	1.2	0.07	2.3 0.80–4.9	0.059	0.031	0.023	0.24	0.014	0.0037	1.4 0.81–1.8		

such as PACs, OPCs, BTs and BDEs. PACs are the most prominent organic contaminant in sedimentation ponds, and levels exceed levels that are expected to affect sediment living organisms. Concentrations of alkylated PACs were high in sedimentation ponds, and the percentage was higher in sedimentation ponds compared to reference. Alkylated PACs are expected to be of the same toxicological concern as parent PACs and should therefore be included in future analyses of sediments in traffic related matrices. BDEs were shown to bioaccumulate and biomagnify both in sedimentation ponds and in reference, and concentrations were higher in sedimentation ponds than reference. The bioaccumulation potential of OPCs and BTs were low. However, concentrations of compounds from both these groups were only detected in fish from sedimentation ponds, not in fish from reference sites. PACs have low potential to bioaccumulate in higher organisms due to metabolic conversion and excretion but accumulate in plants and non-metabolising organisms. Furthermore, PACs also bind strongly to particulate phases such as soot and therefore may be less bioavailable for uptake.

CRediT authorship contribution statement

Merete Grung: Conceptualization, Methodology, Formal analysis, Writing - original draft, Writing - review & editing, Visualization. Sondre Meland: Conceptualization, Methodology, Writing - original draft, Writing - review & editing. Anders Ruus: Writing - original draft, Writing - review & editing. Sissel Ranneklev: Conceptualization, Methodology, Writing - review & editing. Eirik Fjeld: Conceptualization, Writing - original draft. Alfhild Kringstad: Formal analysis, Writing - review & editing. Majbrit Dela Cruz: Formal analysis, Resources, Writing - review & editing. Jan H. Christensen: Resources, Writing - original draft, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We want to thank Espen Lund (NIVA) for collection of biological materials.

Funding

This work was supported and partly funded by the Danish, Norwegian and Swedish Road Administration through their NordFoU R&D programme Reducing Highway Runoff Pollution (REHIRUP, Grant number 604133, www.nordfou.org). In addition, the work was partly funded by NIVA's basic funding from the Norwegian Research Council (Grant number 160016).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2020.141808.

References

- Allan, I.J., Garmo, Ø.A., Rundberget, J.T., Terentjev, P., Christensen, G., Kashulin, N.A., 2018. Detection of tris(2,3-dibromopropyl) phosphate and other organophosphorous compounds in Arctic rivers. Environ. Sci. Pollut. Res. 25, 28730–28737. https://doi.org/ 10.1007/s11356-018-2947-5.
- Andersson, J.T., Achten, C., 2015. Time to say goodbye to the 16 EPA PAHs? Toward an upto-date use of PACs for environmental purposes. Polycycl. Aromat. Compd. 35, 330–354. https://doi.org/10.1080/10406638.2014.991042.
- Asheim, J., Vike-Jonas, K., Gonzalez, S.V., Lierhagen, S., Venkatraman, V., Veivåg, I.-L.S., Snilsberg, B., Flaten, T.P., Asimakopoulos, A.G., 2019. Benzotriazoles, benzothiazoles and trace elements in an urban road setting in Trondheim, Norway: re-visiting the chemical markers of traffic pollution. Sci. Total Environ. 649, 703–711. https://doi. org/10.1016/j.scitotenv.2018.08.299.
- Barron, M.G., Holder, E., 2003. Are exposure and ecological risks of PAHs underestimated at petroleum contaminated sites? Hum. Ecol. Risk. Assess. 9, 1533–1545.
- Borgå, K., Ruus, A., 2019. Quantifying Bioaccumulation in the Aquatic Environment. pp. 1–18 https://doi.org/10.1007/7653_2019_36.
- Borgå, K., Fjeld, E., Kierkegaard, A., McLachlan, M.S., 2012. Food web accumulation of cyclic siloxanes in Lake Mjøsa, Norway. Environ. Sci. Technol. 46, 6347–6354. https:// doi.org/10.1021/es300875d.
- Brown, J.N., Peake, B.M., 2006. Sources of heavy metals and polycyclic aromatic hydrocarbons in urban stormwater runoff. Sci. Total Environ. 359, 145–155. https://doi.org/ 10.1016/j.scitotenv.2005.05.016.
- Burgess, R.M., Ryba, S.A., Perron, M.M., Tien, R., Thibodeau, L.M., Cantwell, M.G., 2004. Sorption of 2,4-dichlorobiphenyl and fluoranthene to a marine sediment amended with different types of black carbon. Environ. Toxicol. Chem. 23, 2534–2544.
- Burkhard, L.P., 2003. Factors influencing the design of bioaccumulation factor and biotasediment accumulation factor field studies. Environ. Toxicol. Chem. 22, 351–360. https://doi.org/10.1002/etc.5620220216.
- Covaci, A., de Boer, J., Ryan, J.J., Voorspoels, S., Schepens, P., 2002. Determination of polybrominated diphenyl ethers and polychlorinated biphenyls in human adipose tissue by large- volume injection - narrow-bore capillary gas chromatography/electron impact low-resolution mass spectrometry. Anal. Chem. 74, 790–798.
- Crawford, K.D., Weinstein, J.E., Hemingway, R.E., Garner, T.R., Globensky, G., 2010. A survey of metal and pesticide levels in stormwater retention pond sediments in coastal South Carolina. Arch. Environ. Contam. Toxicol. 58, 9–23. https://doi.org/10.1007/ s00244-009-9347-2.
- Cristale, J., García Vázquez, A., Barata, C., Lacorte, S., 2013. Priority and emerging flame retardants in rivers: occurrence in water and sediment, Daphnia magna toxicity and risk assessment. Environ. Int. 59, 232–243. https://doi.org/10.1016/j. envint.2013.06.011.
- Direktoratsgruppen vanndirektivet, 2018. Veileder 02:2018 Klassifisering. Direktoratsgruppen for gjennomføringen av vannforskriften http://www. vannportalen.no/globalassets/nasjonalt/dokumenter/veiledere-direktoratsgruppa/ Klassifisering-av-miljotilstand-i-vann-02-2018.pdf.
- EU European Union, 2013. Directive 2013/39/EU of the European Parliament and of the Council of 12 August 2013 Amending Directives 2000/60/EC and 2008/105/EC as Regards Priority Substances in the Field of Water Policy.
- European Commission, 2012. Road Transport A Change of Gear. No. ISBN 978-92-79-22827-8. https://doi.org/10.2832/65952 (Luxembourg).
- European Environment Agency, 2016. Transitions Towards a More Sustainable Mobility System: TERM 2016: Transport Indicators Tracking Progress Towards Environmental Targets in Europe.
- European Environment Agency, 2017. Monitoring Progress of Europe's Transport Sector Towards its Environment, Health and Climate Objectives. Publication No. ISBN 978-92-9213-930-8. https://doi.org/10.2800/620161.
- Fisk, A.T., Hobson, K.A., Norstrom, R.J., 2001. Influence of chemical and biological factors on trophic transfer of persistent organic pollutants in the Northwater Polynya marine food web. Environ. Sci. Technol. 35, 732–738. https://doi.org/10.1021/es001459w.

- Gallotta, F.D.C., Christensen, J.H., 2012. Source identification of petroleum hydrocarbons in soil and sediments from Iguaçu River Watershed, Paraná, Brazil using the CHEMSIC method (CHEMometric analysis of Selected Ion Chromatograms). J. Chromatogr. A 1235, 149–158. https://doi.org/10.1016/j.chroma.2012.02.041.
- Gao, Y., Zhu, L., 2004. Plant uptake, accumulation and translocation of phenanthrene and pyrene in soils. Chemosphere 55, 1169–1178. https://doi.org/10.1016/j. chemosphere.2004.01.037.
- Girardin, V., Grung, M., Meland, S., 2020. Polycyclic aromatic hydrocarbons: bioaccumulation in dragonfly nymphs (Anisoptera), and determination of alkylated forms in sediment for an improved environmental assessment. Sci. Rep. 10, 10958. https://doi. org/10.1038/s41598-020-67355-1.
- Grung, M., Petersen, K., Fjeld, E., Allan, I., Christensen, J.H., Malmqvist, L.M.V., Meland, S., Ranneklev, S., 2016a. PAH related effects on fish in sedimentation ponds for road runoff and potential transfer of PAHs from sediment to biota. Sci. Total Environ. 566–567, 1309–1317. https://doi.org/10.1016/j.scitotenv.2016.05.191.
- Grung, M., Ruus, A., Schneider, S.C., Hjermann, D.Ø., Borgå, K., 2016b. Toxicokinetics of pyrene in the freshwater alga Chara rudis. Chemosphere 157, 49–56. https://doi. org/10.1016/j.chemosphere.2016.04.128.
- Grung, M., Kringstad, A., Bæk, K., Allan, I.J., Thomas, K.V., Meland, S., Ranneklev, S.B., 2017. Identification of non-regulated polycyclic aromatic compounds and other markers of urban pollution in road tunnel particulate matter. J. Hazard. Mater. 323 (Part A), 36–44. https://doi.org/10.1016/j.jhazmat.2016.05.036.
- Hwang, H.-M., Fiala, M.J., Park, D., Wade, T.L., 2016. Review of pollutants in urban road dust and stormwater runoff: part 1. Heavy metals released from vehicles. Int. J. Urban Sci. 20, 334–360. https://doi.org/10.1080/12265934.2016.1193041.
- Hylland, Ketil, Vethaak, D., Davies, I.M., 2012. Background document: polycyclic aromatic hydrocarbon metabolites in fish bile. In: Davies, I.M., Vethaak, D. (Eds.), Integrated Marine Environmental Monitoring of Chemicals and Their Effects, ICES Cooperative Research Report. ICES/CIEM, Copenhagen, pp. 18–25.
- Jartun, M., Fjeld, E., Bæk, K., Løken, K., Rundberget, J.T., Grung, M., Schlabach, M., Warner, N.A., Johansen, I., Lyche, J.L., Berg, V., Nøstbakken, O.J., 2018. Monitoring of environmental contaminants in freshwater ecosystems. Occurrence and Biomagnification (No. M–1106). Norwegian Environment Agency. https://www.miljodirektoratet.no/ globalassets/publikasjoner/M1106/M1106.pdf.
- Jonsson, G., Bechmann, R.K., Bamber, S.D., Baussant, T., 2004. Bioconcentration, biotransformation, and elimination of polycyclic aromatic hydrocarbons in sheepshead minnows (Cyprinodon variegatus) exposed to contaminated seawater. Environ. Toxicol. Chem. 23, 1538–1548. https://doi.org/10.1897/03-173.
- Kammann, U., Askem, C., Dabrowska, H., Grung, M., Kirby, M.F., Koivisto, P., Lucas, C., McKenzie, M., Meier, S., Robinson, C., Tairova, Z.M., Tuvikene, A., Vuorinen, P.J., Strand, J., 2013. Interlaboratory proficiency testing for measurement of the polycyclic aromatic hydrocarbon metabolite 1-hydroxypyrene in fish bile for marine environmental monitoring. J. AOAC Int. 96, 635–641. https://doi.org/10.5740/jaoacint.12-080.
- Korosi, J.B., Irvine, G., Skierszkan, E.K., Doyle, J.R., Kimpe, L.E., Janvier, J., Blais, J.M., 2013. Localized enrichment of polycyclic aromatic hydrocarbons in soil, spruce needles, and lake sediments linked to in-situ bitumen extraction near Cold Lake, Alberta. Environ. Pollut. 182, 307–315. https://doi.org/10.1016/j.envpol.2013.07.012.
- Krahn, M.M., Burrows, D.G., Ylitalo, C.M., Brown, D.W., Wigren, C.A., Collier, T.K., Chan, S.L., Varanasi, U., 1992. Mass spectrometric analysis for aromatic compounds in bile of fish sampled after the Exxon Valdez oil spill. Environ. Sci. Technol. 26, 116–126. https:// doi.org/10.1021/es00025a012.
- Le Viol, I., Mocq, J., Julliard, R., Kerbiriou, C., 2009. The contribution of motorway stormwater retention ponds to the biodiversity of aquatic macroinvertebrates. Biol. Conserv. 142, 3163–3171. https://doi.org/10.1016/j.biocon.2009.08.018.
- Lee, S., Shin, W.-H., Hong, S., Kang, H., Jung, D., Yim, U.H., Shim, W.J., Khim, J.S., Seok, C., Giesy, J.P., Choi, K., 2015. Measured and predicted affinities of binding and relative potencies to activate the AhR of PAHs and their alkylated analogues. Chemosphere 139, 23–29. https://doi.org/10.1016/j.chemosphere.2015.05.033.
- Lee, S., Hong, S., Liu, X., Kim, C., Jung, D., Yim, U.H., Shim, W.J., Khim, J.S., Giesy, J.P., Choi, K., 2017. Endocrine disrupting potential of PAHs and their alkylated analogues associated with oil spills. Environ. Sci. Processes Impacts 19, 1117–1125. https://doi.org/ 10.1039/C7EM00125H.
- Markiewicz, A., Björklund, K., Eriksson, E., Kalmykova, Y., Strömvall, A.-M., Siopi, A., 2017. Emissions of organic pollutants from traffic and roads: priority pollutants selection and substance flow analysis. Sci. Total Environ. 580, 1162–1174. https://doi.org/ 10.1016/j.scitotenv.2016.12.074.
- Marklund, A., Andersson, B., Haglund, P., 2005. Traffic as a source of organophosphorus flame retardants and plasticizers in snow. Environ. Sci. Technol. 39, 3555–3562. https://doi.org/10.1021/es0482177.

Meland, S., 2012. Chemical Characterisation of Sediment From Vassum Sedimentation Basin (No. 94).

- Meland, S., 2016. Management of Contaminated Runoff Water: Current Practice and Future Research Needs.
- Meland, S., Roseth, R., 2011. Organophosphorus compounds in road runoff. Sedimentation and filtration as a mitigation strategy. Presented at the World Congress on Engineering and Technology, Institute of Electrical and Electronics Engineers Inc., Shanghai, pp. 653–656.
- Meland, S., Borgstrøm, R., Heier, L.S., Rosseland, B.O., Lindholm, O., Salbu, B., 2010. Chemical and ecological effects of contaminated tunnel wash water runoff to a small Norwegian stream. Sci. Total Environ. 408, 4107–4117. https://doi.org/10.1016/j. scitotenv.2010.05.034.
- Meland, S., Sun, Z., Sokolova, E., Rauch, S., Brittain, J.E., 2020. A comparative study of macroinvertebrate biodiversity in highway stormwater ponds and natural ponds. Sci. Total Environ. 740, 140029. https://doi.org/10.1016/j.scitotenv.2020.140029.
- Moe, T.F., Persson, J., Bækkelie, K.A.E., Myrvold, K.M., Garmo, Ø.A., Grung, M., Hindar, A., Guerrero Calidonio, J.-L., de Wit, H., 2019. Overvåking av referanseelver 2018. Basisovervåking i henhold til vannforskriften.
- Raine, J.C., Turcotte, D., Tumber, V., Peru, K.M., Wang, Z., Yang, C., Headley, J.V., Parrott, J.L., 2017. The effect of oil sands tailings pond sediments on embryo-larval walleye (Sander vitreus). Environ. Pollut. 229, 798–809. https://doi.org/10.1016/j. envpol.2017.06.038.
- Richter-Brockmann, S., Achten, C., 2018. Analysis and toxicity of 59 PAH in petrogenic and pyrogenic environmental samples including dibenzopyrenes, 7H-benzo[c]fluorene, 5-methylchrysene and 1-methylpyrene. Chemosphere 200, 495–503. https://doi. org/10.1016/j.chemosphere.2018.02.146.
- Rust, A.J., Burgess, R.M., McElroy, A.E., Cantwell, M.G., Brownawell, B.J., 2004. Role of source matrix in the bioavailability of polycyclic aromatic hydrocarbons to depositfeeding benthic invertebrates. Environ. Toxicol. Chem. 23, 2604. https://doi.org/ 10.1897/03-353.
- Ruus, A., Øverjordet, I.B., Braaten, H.F.V., Evenset, A., Christensen, G., Heimstad, E.S., Gabrielsen, G.W., Borgå, K., 2015. Methylmercury biomagnification in an Arctic pelagic food web. Environ. Toxicol. Chem. https://doi.org/10.1002/etc.3143 n/a-n/a.
- Stogiannidis, E., Laane, R., 2015. Source characterization of polycyclic aromatic hydrocarbons by using their molecular indices: an overview of possibilities. In: Whitacre, D.M. (Ed.), Reviews of Environmental Contamination and Toxicology, Reviews of Environmental Contamination and Toxicology. Springer International Publishing, pp. 49–133 https://doi.org/10.1007/978-3-319-10638-0_2.
- Stout, S.A., Uhler, A.D., Emsbo-Mattingly, S.D., 2004. Comparative evaluation of background anthropogenic hydrocarbons in surficial sediments from nine urban waterways. Environ. Sci. Technol. 38, 2987–2994. https://doi.org/10.1021/es040327q.
- Sun, Z., Brittain, J.E., Sokolova, E., Thygesen, H., Saltveit, S.J., Rauch, S., Meland, S., 2018. Aquatic biodiversity in sedimentation ponds receiving road runoff – what are the key drivers? Sci. Total Environ. 610–611, 1527–1535. https://doi.org/10.1016/j. scitotenv.2017.06.080.
- US National Library of Medicine, n.d. HSDB hazardous substances data bank [WWW document]. URL http://toxnet.nlm.nih.gov (accessed 10.24.11).
- Vignet, C., Le Menach, K., Mazurais, D., Lucas, J., Perrichon, P., Le Bihanic, F., Devier, M.-H., Lyphout, L., Frère, L., Bégout, M.-L., Zambonino-Infante, J.-L., Budzinski, H., Cousin, X., 2014. Chronic dietary exposure to pyrolytic and petrogenic mixtures of PAHs causes physiological disruption in zebrafish - part I: survival and growth. Environ. Sci. Pollut. Res. 21, 13804-13817. https://doi.org/10.1007/s11356-014-2629-x.
- Villalobos-Jiménez, G., Dunn, A.M., Hassall, C., 2016. Dragonflies and damselflies (Odonata) in urban ecosystems: a review. EJE 113, 217–232. https://doi.org/10.14411/ eje.2016.027.
- Wayland, M., Headley, J.V., Peru, K.M., Crosley, R., Brownlee, B.G., 2008. Levels of polycyclic aromatic hydrocarbons and dibenzothiophenes in wetland sediments and aquatic insects in the oil sands area of Northeastern Alberta, Canada. Environ. Monit. Assess. 136, 167–182. https://doi.org/10.1007/s10661-007-9673-7.
- Yoon, Y., Ryu, J., Oh, J., Choi, B.-G., Snyder, S.A., 2010. Occurrence of endocrine disrupting compounds, pharmaceuticals, and personal care products in the Han River (Seoul, South Korea). Sci. Total Environ. 408, 636–643. https://doi.org/10.1016/j. scitotenv.2009.10.049.
- Zhang, J., Zhang, X., Wu, L., Wang, T., Zhao, J., Zhang, Y., Men, Z., Mao, H., 2018. Occurrence of benzothiazole and its derivates in tire wear, road dust, and roadside soil. Chemosphere 201, 310–317. https://doi.org/10.1016/j.chemosphere.2018.03.007.