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Distribution and bioaccumulation of POPs and mercury in the Ga-Selati River (South Africa) and the rivers Gudbrandsdalslågen and Rena (Norway)

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ABSTRACT

Biomagnification of Hg and persistent organic pollutants (POPs: polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs)) in aquatic food chains can lead to high pollutant concentrations in top predators, including humans. Despite this threat to human health, research concerning bioaccumulation is still underrepresented in the southern hemisphere and in (sub)arctic and (sub) tropical areas, emphasizing the need for research in these locations. In this study, samples of water, sediment and aquatic biota were analyzed to determine concentrations of POPs and total mercury (THg) in the Ga-Selati river (South Africa) and two rivers Rena and Gudbrandsdalslågen in Norway. Trophic magnification factors (TMFs) were determined to evaluate and compare the biomagnification and the threat to human health due to consumption of the fish was assessed.

Concentrations of POPs in sediment and biota samples were generally low except for relatively high concentrations of ΣDDX (dichlorodiphenyltrichloroethane and metabolites) in aquatic biota from the Ga-Selati river (ranging from 1.9 to 133 ng/g ww in invertebrates and 1.9 to 5643 ng/g ww in fish). Dissolved THg concentrations were high in the Ga-Selati river (ranging from 0.009 to 0.036 µg/l) but THg concentrations in sediment and biota were low in studied rivers compared to other studies. Biomagnification occurred for THg, several DDT-metabolites and PCB compounds, TN and CN. Biomagnification of *p,p′*-DDT and THg differed significantly between the two countries, supporting existing patterns found in literature, although more data is needed to attribute these differences to climatic or other factors. Concentrations in fish from the rivers Ga-Selati and Rena were under the threshold levels reported for THg and POPs, but caution should be taken when consuming Northern pike (*Esox Lucius*) from the subarctic river Gudbrandsdalslågen, to avoid harmful effects due to both elevated THg and PBDE exposure.

1. Introduction

The release of Persistent Organic Pollutants (POPs), such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs)) and mercury (Hg), into the environment is of great concern due to their toxicity, their ability to accumulate in organisms and their persistence (Jones and de Voogt, 1999; Lavoie et al., 2013). Both POPs and mercury have been shown to biomagnify across the food chain, leading to high concentrations and potential effects in top predators, including humans. (El-Shahawi et al., 2010; Borgå et al., 2012; Lavoie et al., 2013). Despite the urgent need for understanding the impact and global patterns of bioaccumulation in aquatic food webs, the majority of studies on bioaccumulation of POPs and Hg have been conducted in temperate regions, and in the case of POPs predominantly in the northern hemisphere (Lavoie et al., 2013; Walters et al., 2016). This emphasizes the need to investigate bioaccumulation in underrepresented areas, such as countries in the southern hemisphere and (sub)tropical regions. To properly investigate bioaccumulation across different aquatic systems, Trophic Magnification Factors (TMFs) are used. TMFs represent the average prey to predator transfer of pollutants through food webs and have proven to be a reliable and conclusive tool to quantify biomagnification (Walters et al., 2016).

The present study investigates bioaccumulation in river ecosystems

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from two different hemispheres: the Ga-Selati River located in South Africa and the rivers Gudbrandsdalslågen (further on referred to as Laagen) and Rena located in Norway. The Ga-Selati river is heavily impacted by agricultural activities, local communities and a mining activity (King et al., 2008). Furthermore, although the use of DDT has been banned in most parts of the world (NPIC (National Pesticide Information Center), 1999; PAN UK, 2008), it is permitted to produce and use DDT in countries where the transmission of malaria and visceral leishmaniasis can be efficiently prevented by the use of DDT, in accordance with WHO (World Health Organization) recommendations and guidelines (van den Berg, 2008). Several African countries, including South Africa, still regularly use DDT. In Norway, the river Laagen flows in the Gudbrandsdalen valley and is influenced by surrounding villages and farms. Few people live in the drainage basin of the Rena river, so the latter is regarded as relatively little impacted by human activities (NINA (Norwegian Institute for Nature Research) and Eastern Norway Research Institute (ENRI), 2000). Because fish is an important food source for local people, especially around the Ga-Selati river, it is important to monitor the bioaccumulation of pollutants in the fish to assess whether they form a threat for human health.

The specific aims of this study are to (1) assess and compare POP and total mercury (THg) concentrations in water, sediment and aquatic biota originating from the Ga-Selati river and the rivers Laagen and Rena (2) examine the degree of biomagnification of the analyzed POPs and THg in the aquatic biota using TMFs, and (3) assess the potential human health risk due to consumption of contaminated fish from this study using Minimum Risk Levels and converting this to a maximum edible amount per day.

2. Materials and methods

2.1. Study area

The Ga-Selati River is located in the northeastern province of Limpopo in South Africa and merges with the larger Olifants river at the boundary of the Kruger National Park, near the Phalaborwa mine (Chapman, 2006) (Fig. 1). Extensive farming takes place along the river and several rural communities, commercial game farms and nature reserves depend on the water flows of the river for irrigation. At the confluence of the Ga-Selati River with the Olifants River, mining activity is very intense with the Palabora Mining Company extracting copper and other by-products, and Foskor, one of the world's largest producers of phosphate and phosphoric acid, as key players (King, 2008). Samples were collected at three locations along the river (Fig. 1). The first one, Harmonie, is situated in an agricultural area. The second sampling site, Namakgale, is located near the low-income area of Namakgale and the third collection of samples was at Lepelle Bridge (LB), a bridge that is situated in the vicinity of a fertilizer factory and a phosphate mine. The climate of the Ga-Selati River catchment is subtropical, with mean temperatures of 25.8 °C in the summer and 18.0 °C in the winter, and an annual rainfall of 450–600 mm (Chapman, 2006).

Sampling in Norway was carried out in two of the larger river systems of the country: Gudbrandsdalslågen (Laagen) and Rena. The river Laagen flows through the Gudbrandsdalen valley in the southeast of Norway, and runs about 250 km south through glacially sculptured rural valleys before merging into Lake Mjøsa, the largest lake in Norway (NINA (Norwegian Institute for Nature Research) and Eastern Norway Research Institute (ENRI), 2000). Several villages and farms are found in the valley. Few people live in the drainage area of Rena, and only minor agricultural areas occur in the vicinity of the river. Both rivers encounter a continental climate in contrast to coastal rivers in south Norway, and are located in an area characterized by a subarctic climate with high precipitation and cold temperatures, with an average annual rainfall of 719 mm and mean temperatures of 13.8 °C in the summer and −8.1 °C in the winter (Yr, 2017).

2.2. Sample collection

Samples of sediment, water, fish and invertebrates were collected at each location in South Africa between the17th and 19th of September 2014 and during the first week of November 2014 in Norway.

Water samples were taken in triplicates by manually filling sterile 50 ml poly propylene (PP) tubes at about 10cmn below water surface at all locations. Three sediment samples were collected at each location along the Ga-Selati river and later pooled to obtain one mixed sample per location. Sediment was scooped from the side of the river at Namakgale and Lepelle Bridge and a Van Veen Grab sampler was used at Harmonie to collect the sediment samples onboard a boat. Sediments in Norway were collected with the invertebrate catching net from at least three points down to 1.5 m deep in both rivers. Gomphidae (Odonata) larvae were collected by hand and stored in 5 ml PP tubes at the riverbank of the Ga-Selati river, with an average of 10 individuals per site. In Norway, snails (Lymneae sp.) were collected by net-sweeps just above the bottom of the river and by manually selecting them among sediments and plants with a metal pincher. Since snails were not found in the Rena River, they were sampled 400 m up the river mouth in lake Løpsjøen, where the river runs through. In South Africa eight fish species were caught: African sharptooth catfish (*Clarias gariepinus*), leaden labeo (*Labeo molybdinus*), plain squeaker (*Synodontis zambeziensis*), redbreast tilapia (*Tilapia rendalli*), Mozambique tilapia (*Tilapia*

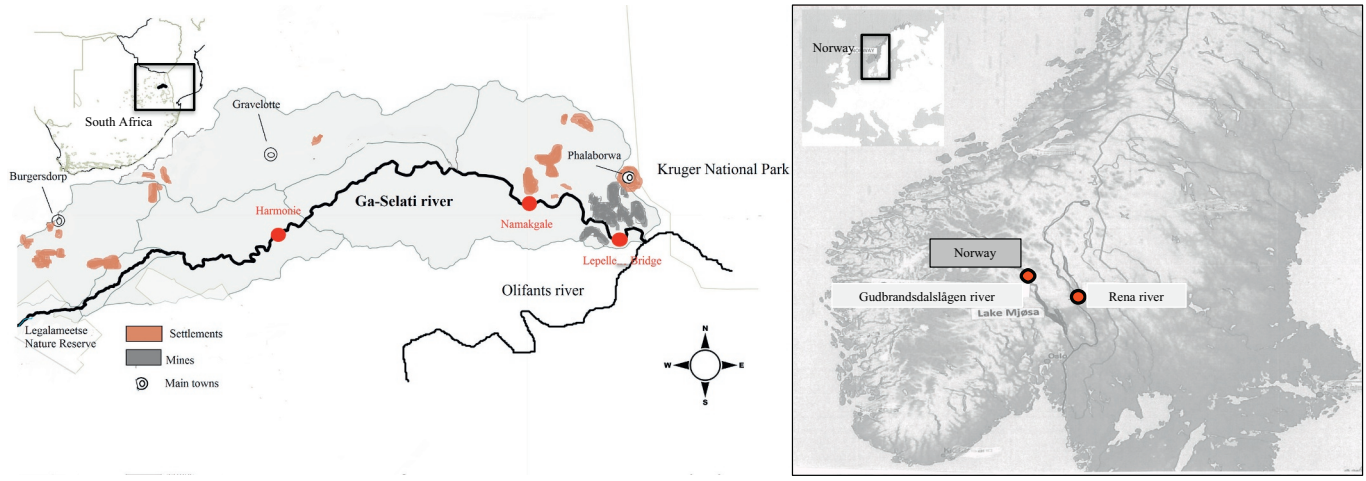

Fig. 1. Map of the Ga-Selati catchment, South Africa, and the rivers Laagen and Rena, Norway, with sampling sites indicated in red. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

mossambicus), sawfin suckermouth (*Chiloglanis paratus*), river goby (*Glossogobius callidus*) and threespot barb (*Barbus trimaculatus*). The fish were caught and dissected the same day. The tissues were stored in 50 ml PP tubes and kept with the other samples in a liquid nitrogen tank before being stored in freezers at −20 °C in the lab in Belgium. In Norway, five species of fish were collected: Brown trout (*Salmo trutta*), Eurasian ruffe (*Gymnocephalus cernuus*), common minnow (*Phoxinus phoxinus*), alpine bullhead (*Cottus poecilopus*) and the piscivorous Northern pike (*Esox Lucius*). All species were present in the river Laagen, but only the brown trout and the Northern pike were present in the Rena river. All water, sediment and biota samples were stored in a liquid nitrogen tank during field work before being stored in freezers at −20 °C in the lab in Belgium.

2.3. POPs

2.3.1. Sample preparation

Samples were extracted following the protocol described in Verhaert et al. (2013). Concerning aquatic biota samples, 29 South African fish and 24 Norwegian fish were used as well as pooled samples of invertebrates for each site in both Norway and South Africa. Accurate amounts of fish (0.4–3.4 g wet weight (ww)), invertebrate samples (2.5–4.2 g ww) and sediment samples(3.1–3.2 g) were extracted using an automated hot Soxhlet extractor as described by Verhaert et al. (2013) (see SI1).

2.3.2. Analysis

The following persistent organic pollutants were targeted: PCB congeners (IUPAC numbers 28, 49, 52, 74, 95, 99, 101, 105, 110, 118, 128, 138, 146, 149, 151, 153, 156, 170, 171, 174, 177, 180, 183, 187, 194, 195, 196/203, 199, 206 and 209), dichlorodiphenyltrichloroethane (*p,p′*-DDT and *o,p′*-DDT) and metabolites (*p,p′*-DDD, *p,p′*-DDE, *o,p′*-DDE and *o,p′*-DDD), *β*- and γ- HCH (hexachloro-cyclohexane) isomers, chlordane metabolites (oxychlordane (OxC), cis-nonachlor (CN) and trans-nonachlor (TN)), hexachloro-benzene (HCB), and PBDEs (BDE 28, 47, 99, 100, 153, 154, and 183). PBDEs, CHLs, and higher PCBs were measured with an Agilent 6890-5973 gas chromatograph coupled with a mass spectrometer system (GC–MS). The GC was equipped with a 30 m × 0.25 mm × 0.25 μm DB-5 ms capillary column (J&W Scientific, Folsom, CA, USA) and the MS was operated in electron capture negative ionisation (ECNI) mode and used in the selected ion-monitoring (SIM) mode. For measuring the levels of lower chlorinated PCBs, DDXs (different DDT compounds), and HCB, an Agilent 6890 GC – 5973 MS system operated in electron ionisation (EI) mode equipped with a 25 m × 0.22 mm × 0.25 μm HT-8 capillary column was used in the selected ion-monitoring (SIM) mode with 2 ions monitored for each PCB homologue group or individual OCP.

Procedural blanks were included for quality assurance and quality control. The limit of quantification (LOQ) was calculated as three times the standard deviation of the mean of the blank measurements. The analytical procedures were validated through the analysis of certified material SRM 1945 (PCBs, PBDEs and OCPs in whale blubber) for which deviations from certified values were < 10%. The QC scheme is assessed through participation in the inter-laboratory Comparison Exercise Program for Organic Contaminants in Marine Mammal Tissues, which is organized by the National Institute of Standards and Technology (NIST, USA).

2.4. Mercury

2.4.1. Sample preparation

A 0.5 g wet weight (ww) of sediment and biota samples was accurately weighed using a Mettler AT261 DeltaRange® sensitive balance. The samples were then freeze dried at −55 °C for 48 h to a constant weight using a freeze drier (Heto PowerDry® LL3000), and weighed again to obtain the dry weight (dw). Samples were digested as described by Mataba et al. (2016) (see SI2). After digestion, samples were stored in a freezer (−20 °C) until analysis. Reference materials and blanks were handled in a similar manner to ensure quality control. Lyophilized Cod Muscle (BCR 422) from the Institute for Reference Materials and Measurements (IRMM, Geel Belgium), freeze dried blue mussel tissue (no 2976) from NIST and channel sediment BCR 320-R were used as reference materials to verify for recoveries. Prior to analysis, all sediment and biota samples were thawed and diluted 2 times with Milli-Q water.

To determine the dissolved THg concentrations, 10 ml of each water sample was filtered through a filter with a pore size of 0.2 μm and subsequently acidified with 150 μl HNO_3 (67%). In total, three replicas were prepared per site. Three blanks were used as control, carrying out the same procedure but utilizing MilliQ water. Samples were then stored at −20 °C until analysis.

2.4.2. Analysis

Analysis of total mercury (THg) was carried out in cold plasma mode (to prevent the Hg to become volatile) by High Resolution Inductively Coupled Mass Spectrometry (HR-ICP-MS) (Thermo scientific Finnigan element 2, Waltham, MA, USA) with a detection limit of 0.001 μg/l. Obtained concentrations agreed well with the certified values, with a mean recovery of 112%.

2.5. Total organic carbon (TOC)

Sediment samples were pooled per location and approximately 10 g of sediment of each site was transferred to pre-weighted crucibles and subsequently weighed. The samples were then placed in a muffle oven at 150 °C for 24 h and thereafter allowed to cool down to room temperature. The filled crucibles were then weighed again, placed in the muffle oven and heated up to 550 °C in a span of an hour, after which they were kept at this temperature for another 4 h. After the samples were allowed to cool down to room temperature, they were weighed to obtain the final dry weight. The organic content is inferred through loss of ignition (LOI) from the weight difference between the wet and dry state of the sample as described by Heiri et al. (2001) and the total amount of organic carbon was calculated as described by Nelson and Sommers (1996) and Schumacher (2002) (see SI3).

2.6. Stable isotopes

To assess the isotopic composition, a small amount of each sample was transferred to an Eppendorf tube (2 mL), freeze-dried, and homogenized. The fish and snail samples were encapsulated in pre-weighted 5 × 8 mm Sn capsules and weighted with an accuracy of three decimal places with a Mettler AT261 scale. A similar procedure was used for the sediment and gomphid samples, with the exception of the samples being encapsulated in Ag cups instead of Sn cups, since HCl must be added to samples that contain sediment or invertebrates with an exoskeleton in order to remove traces of non-dietary carbonates (Verhaert et al., 2013). C and N concentrations were determined at the Department of Earth and Environmental Sciences, KU Leuven (Belgium) using a Thermo Flash HT/EA coupled to a Thermo DeltaV Advantage IRMS with a Conflo IV interface. Isotopic composition is expressed using following formula:

$$\delta^{13}C,\ \delta^{15}N = [(R_{sample}/R_{reference}) - 1] \times 1000$$

with $R = {}^{13}C/{}^{12}C$ and ${}^{15}N/{}^{14}N$ for respectively carbon and nitrogen.

Data were calibrated by using a combination of IAEA-C6, IAEA-N1 and acetanilide, which had been calibrated in the lab for both $\delta^{13}C$ and $\delta^{15}N$. To relate the pollutant concentration to the trophic level (TL) of the fish, the TL was calculated for all fish and invertebrates using following formula (Borgå et al., 2012):

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

with δ^{15}Nconsumer = ^{15}N concentration of the consumer, δ^{15}Nprimary consumer = ^{15}N concentration of the primary consumer, Δ^{15}N = fractionation of ^{15}N into predator which has a value of 3% because the abundance of ^{15}N in tissues of consumers is typically enriched by 3% relative to their prey (Pinnegar and Polunin, 1999), and 2 is the trophic level of the primary consumer. The ^{15}N concentration of the primary consumer was based on concentrations in *Lymnea* snail samples for Norway and, since snails could not be collected at South African sites and Gomphids cannot be considered as primary consumers, snails (*Tarebia granifera*) collected by Verhaert et al. (2017) from the Olifants River for South Africa. The latter were collected a couple of kilometers downstream of its confluence with the Ga-Selati river and are also present in the Ga-Selati river (Rasifudi, and Addo-Bediako1, A., Bal1, K., Swemmer, T.M., 2018).

To quantify the Trophic Magnification Factor (TMF), a regression analysis was carried out between the trophic level of a fish/invertebrate and the corresponding concentration of the contaminant. The TMF is calculated as the antilog of the regression slope with base 10 (Borgå et al., 2012):

$$\text{Log[Contaminant]} = a + b\,TL \rightarrow TMF = 10^{b}$$

2.7. Statistical analysis

Statistical analysis was carried out using RStudio (Version 0.98.1103) with a level of statistical significance set at $p < 0.05$. Concentrations below LOQ were given a value of LOQ/2 (Bervoets et al., 2005; Custer et al., 2000). The data was tested for normality and homogeneity of variance using a Shapiro-Wilk test and Levene's test respectively, and data were log-transformed since these criteria were not met. Linear regression was used to examine (1) the relation between the concentrations in the TOC-normalized sediment and in lipid-normalized biota (2) the relation between trophic level and corresponding pollutant concentration (see TMF section). To test the relationships between trophic level and concentration of pollutants, as well as to investigate whether this relationship differs between climates, ANCOVA was used to make a regression of the trophic level and the log of the pollutant concentration and then compare the slopes.

2.8. Human health risks

The Agency for Toxic Substances and Disease Registry (ATSDR) has developed a list of commonly used hazardous substances and determined significant human exposure levels, which are used to assess whether certain concentrations of pollutants in food items can cause significant acute, subacute, and chronic health effects on humans (ATSDR, Agency for Toxic Substances and Disease Registry, 2017). Minimum Risk Levels (MRL) (ng/day) were calculated for a person of 70 kg. Using following formula, the maximum daily intake (g/day) without risk of negative effects due to ingestion of pollutants was calculated for a person of 70 kg, based on the mean concentration of the pollutants found in the muscle tissue of every fish species (ng/g):

$$\text{Maximum edible amount} = (\text{MRL for 70 kg person}) / (\text{mean concentration in fish})$$

3. Results and discussion

3.1. POPs

3.1.1. Sediment

TOC values ranged from 0.52–2.98% in South Africa and 2.66–10.4% in Norway and are shown in Table 1, together with the mean ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs concentrations measured in the sediment samples.

ΣPCBs concentrations in both South Africa and Norway were lower than in other countries in Europe, such as Spain and Belgium (Fernandez et al., 1999; Van Ael et al., 2012), Asian countries including China, Korea and Vietnam (Koh et al., 2004; Zhang et al., 2004; Minh et al., 2007) and in studies from Egypt and another region in South Africa (El-Kady et al., 2007; Quinn et al., 2009). Concentrations were higher than values from a study in Congo (Verhaert et al., 2013) and comparable to ΣPCBs values in the Olifants River Basin in South Africa (Verhaert et al., 2017). The most frequently occurring PCB congeners were CB110 (18%), CB149 (14%), CB95 (13%) and CB153 (9%) for South Africa and CB149 (14%), CB153 (14%), CB95 (13%) and CB138 (11%) for Norway.

ΣPBDEs values were significantly higher in sediment originating from the river Laagen. Values of the present study were however low compared to concentrations reported in sediments from rivers in Belgium, Congo, China and other South African rivers (Hu et al., 2010; Olukunle et al., 2012; Van Ael et al., 2012; Verhaert et al., 2013; Verhaert et al., 2017). The compounds that made up most part of ΣPBDEs were BDE 209 (48%) and BDE 47 (35%) for South Africa, and BDE 209 (52%) and BDE 99 (21%) for Norway.

All six analyzed DDX metabolites were detected in South African samples, but only *p,p′*-DDE and *p,p′*-DDD were found in sediment from Norway. ΣDDX values were significantly lower in Norwegian samples. Concentrations in South African sediments were also higher compared to rivers in Belgium and Congo, but comparable to concentrations reported in studies in China, Spain, Vietnam and other South African rivers (Fernandez et al., 1999; Zhang et al., 2004; Quinn et al., 2009; Van Ael et al., 2012; Verhaert et al., 2013; Verhaert et al., 2017), with the exception of the very high concentrations at Lepelle Bridge. The high concentrations at Lepelle Bridge are possibly a result of government sponsoring of DDT spraying to kill mosquitoes in the town and lodges nearby, although it is surprising that it is significantly higher than at the Namakgale site, which is closer to said town. Norwegian sediments contained less or similar amounts of DDX compared to previously mentioned studies. The most dominant DDX were *p,p′*-DDT (70%) and *p,p′*-DDE (15%) in South African sediment and *p,p′*-DDE (67%) and *p,p′*-DDD (33%) in Norwegian sediment. DDD and DDE are formed by various chemical breakdowns of DDT, therefore a higher content of DDE and DDD compared to DDT indicates aged DDT that has already been broken down (Aislabie et al., 1997). Only DDE- and DDD-isomers have been found in Norwegian samples, which is not surprising since the use of DDT has been banned from Europe since the 80's (PAN UK, 2008). The same was observed for sediments from the Scheldt river in Belgium and the Netherlands (Van Ael et al., 2012). However, DDT was present in sediment from South Africa and Congo (Verhaert et al., 2013) which suggests DDT is still used in those areas. In countries where the transmission of malaria and visceral leishmaniasis can be efficiently prevented by the use of DDT, it is permitted to produce and use DDT, in accordance with WHO (World Health Organization) recommendations and guidelines (van den Berg, 2008). The results from the present study are consistent with the fact that African countries, as opposed to European countries, still regularly use DDT as a pesticide.

HCB concentrations in this study were lower than those reported in sediments from rivers in Belgium, China, Vietnam and other South African rivers (Nakata et al., 2005; Minh et al., 2007; Quinn et al., 2009; Van Ael et al., 2012; Verhaert et al., 2017;) but higher or comparable to concentrations in sediments from rivers located in Spain and Congo (Fernandez et al., 1999; Verhaert et al., 2013).

Concentrations of CHLs and HCHs were low compared to other studies. OxC, TN and CN were not detected in any of the sites, with the exception of Lepelle Bridge (South Africa) where low concentrations of TN and CN were found. CHLs seem to be rarely detected in sediment as they were also under the detection limit in several other studies from Belgium and China (Nakata et al., 2005; Van Ael et al., 2012). ΣHCHs were only detected in South African samples, with γ-HCH as the only compound present. South African HCH concentrations in the present study were lower than concentrations in sediments from rivers in Belgium, Congo, China, Vietnam and other rivers in South Africa (Zhang

Table 1
Values of total organic carbon (TOC%) and mean sediment concentrations (ng/g) of ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs for sites in South Africa and Norway in this analysis compared to other studies.

	TOC (%)	ΣPCBs	OxC	TN	CN	HCB	ΣDDX	ΣHCHs	ΣPBDEs
South Africa									
Harmonie	2.98	0.16	< LOQ	< LOQ	< LOQ	0.0059	0.17	0.0044	0.077
Namakgale	2.09	0.25	< LOQ	< LOQ	< LOQ	0.0057	1.5	0.0056	0.049
Lepelle Bridge	0.52	0.22	< LOQ	0.0062	0.0026	0.0062	80	0.0064	0.042
Norway									
Laagen	2.66	0.21	< LOQ	< LOQ	< LOQ	0.0072	0.051	< LOQ	0.406
Rena	10.36	0.09	< LOQ	< LOQ	< LOQ	0.0046	0.019	< LOQ	0.033
Congo River, Congo[a]		0.08	–	–	–	< LOQ	0.12	0.036	0.23
Scheldt estuary, Belgium[b]		6.43	< LOQ	< LOQ	< LOQ	0.01	0.03	0.02	6.9
Tonghui River, China[c]		3.3 ± 2.6	–	–	–	–	1.1 ± 1.1	0.17 ± 0.10	–
Ebro River, Spain[d]		14 ± 7.6	–	–	–	0.007 ± 0.009	3.1 ± 3.1	–	–
Hyeongsan River, Korea[e]		62							
Vaal River, South Africa[f]		3.02*	–	–	–	0.12	1.65	0.52	–
Nile River, Egypt[g]		0.22 ± 0.04							
Lake Tai, China[h]		< LOQ	< LOQ	0.03 ± 0.01	0.03 ± 0.02	0.08 ± 0.05	0.72 ± 0.18	0.43 ± 0.11	–
Fuhe River, China[i]		–	–	–	–	–	–	–	2.3
Jukskei River, South Africa[j]		–	–	–	–	–	–	–	3.4
Mekong River, Vietnam[k]		0.89	–	–	–	0.016	6.5	0.1	–
Olifants River Basin, South Africa[l]		0.16	–	–	–	0.031	0.64	< LOQ	1.5

< LOQ = below Limit of Quantitation, * ΣPCBs (7 indicator PCBs).
[a] Verhaert et al., 2013.
[b] Van Ael et al., 2012.
[c] Zhang et al., 2004.
[d] Fernandez et al., 1999.
[e] Koh et al., 2004.
[f] Quinn et al., 2009.
[g] El-Kady et al., 2007.
[h] Nakata et al., 2005.
[i] Hu et al., 2010.
[j] Olukunle et al., 2012.
[k] Minh et al., 2007.
[l] Verhaert et al., 2017.

et al., 2002; Minh et al., 2007; Quinn et al., 2009; Van Ael et al., 2012; Verhaert et al., 2013) but higher than concentrations in sediments from the Olifants river in South Africa (Verhaert et al., 2017). It should be mentioned that α-HCH results were not included in the analysis due to interferences, so we can only say β-HCH was absent but cannot conclude anything concerning α-HCH.

3.1.2. Invertebrates

The lipid content of the invertebrates varied from 1.3% to 1.7% for *Gomphidae* larvae and from 1.3% to 1.6% for *Lymnaea* sp. snails. Concentrations of ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs (ng/g ww) for aquatic invertebrates are given in Table 2. PCBs were only found in the snails collected in Norway, and concentrations were low compared to a Belgian study but similar to a study in Congo (Van Ael et al., 2012; Verhaert et al., 2013). CB 153 was the most dominant congener (35%), followed by CB 110 (23%), CB 101 (22%) and CB 138 (20%). Concentrations of OxC, TN, CN, HCB and HCHs were not detected in any of the invertebrates, except for a small amount of TN present in *Gomphidae* larvae at Namakgale. DDX were the most abundant pollutants in South African invertebrates from this study, but were not detected in *Lymnaea* snails in Norway. ΣDDX concentrations in *Gomphidae* larvae from this study very high compared to other invertebrates from Congo, Belgium and another South African river (Van Ael et al., 2012; Verhaert et al., 2013; Verhaert et al., 2017), especially at Namakgale. As stated before, the use of DDT is very common in towns as a way to prevent malaria. The most frequently occurring metabolite was *p,p*'-DDE (73%), followed by *p,p*'-DDT (17%), *p,p'*-DDD (8%) while the remaining congeners contributed < 1% to the total. Only two PBDE congeners were

Table 2
ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs concentrations (ng/g ww invertebrate) in invertebrates from this study compared to other studies.

	Lipid %	ΣPCB	sOxC	TN	CN	HCB	ΣDDX	ΣHCHs	ΣPBDEs
South Africa (*Gomphidae* sp.)									
Harmonie	1.7	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	3.9	< LOQ	0.51
Namakgale	1.3	< LOQ	< LOQ	0.13	< LOQ	< LOQ	133	< LOQ	< LOQ
Lepelle Bridge	1.6	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	1.9	< LOQ	< LOQ
Norway (*Lymnaea* sp.)									
Laagen	1.6	2.0	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	0.33
Rena	1.3	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	0.43
Congo River Basin, Congo (*Pila* sp.)[a]	1.2	2.1	–	–	–	< LOQ	0.14	0.060	0.040
Scheldt estuary, Belgium (Polychaeta)[b]	1.3	53	< LOQ	0.03	< LOQ	0.14	0.55	0.06	1.2
Olifants River Basin, South Africa (Gomphidae)[c]	0.82	< LOQ	–	–	–	< LOQ	0.33	< LOQ	< LOQ

< LOQ = below Limit of Quantitation.
[a] Verhaert et al., 2013.
[b] Van Ael et al., 2012.
[c] Verhaert et al., 2017.

Table 3
Means ± SD of ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs concentrations (ng/g ww) in fish from this study compared to other studies.

	Lipid %	ΣPCBs	OxC	TN	CN	HCB	ΣDDX	ΣHCHs	ΣPBDEs
South Africa									
Harmonie									
Labeo molybdinus	2.5	1.8	< LOQ	< LOQ	< LOQ	< LOQ	21	< LOQ	< LOQ
Tilapia mossambicus	0.97	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	4.4 ± 0.43	< LOQ	< LOQ
Clarias gariepinus	1.2	1.9 ± 0.40	< LOQ	< LOQ	< LOQ	< LOQ	64 ± 74	< LOQ	< LOQ
Namakgale									
Labeo molybdinus	3.6	2.6	< LOQ	3.7	1.3	0.13	5643	2.7	< LOQ
Clarias gariepinus	0.57	< LOQ	0.16 ± 0.16	0.76 ± 1.0	0.27 ± 0.31	< LOQ	361 ± 275	0.48 ± 0.091	0.29 ± 0.058
Lepelle Bridge									
Labeo molybdinus	3.0	1.7 ± 0.030	< LOQ	< LOQ	< LOQ	< LOQ	60 ± 55	< LOQ	0.27 ± 0.04
Tilapia rendalli	1.1	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	16 ± 4.4	< LOQ	< LOQ
Clarias gariepinus	1.3	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	21 ± 23	< LOQ	< LOQ
Synodontis zambezensis	5.5	2.7 ± 0.45	< LOQ	0.22 ± 0.08	0.17 ± 0.04	< LOQ	410 ± 141	0.17 ± 0.07	0.25 ± 0.08
Norway									
Laagen									
Gymnocaphalus cernuus	1.7	2.5 ± 1.3	< LOQ	< LOQ	< LOQ	< LOQ	1.2 ± 0.45	< LOQ	0.58 ± 0.34
Phoxinus phoxinus	1.4	3.1 ± 0.23	< LOQ	< LOQ	< LOQ	< LOQ	2.2 ± 0.53	< LOQ	0.47 ± 0.22
Salmo trutta	1.5	4.6 ± 1.7	< LOQ	0.11 ± 0.05	< LOQ	0.15 ± 0.03	2.8 ± 1.1	< LOQ	1.0 ± 0.30
Esox lucius	0.65	9.5 ± 7.7	< LOQ	0.069 ± 0.032	0.065 ± 0.026	< LOQ	4.5 ± 2.8	< LOQ	11 ± 14
Cottus poecilopus	1.5	7.1 ± 5.5	< LOQ	0.11 ± 0.10	0.077 ± 0.046	0.12 ± 0.13	2.0 ± 1.6	< LOQ	2.0 ± 1.6
Rena									
Salmo trutta	1.9	1.7 ± 0.01	< LOQ	< LOQ	< LOQ	0.13 ± 0.031	0.85 ± 0.016	< LOQ	0.19 ± 0.017
Esox lucius	0.57	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	0.83 ± 0.019	< LOQ	0.27 ± 0.14
Congo river, Congo (*Synodontis alberti*)[a]		1.5	–	–	–	0.078	0.090	0.16	1.2
Olifants River Basin, South Africa (*Clarias gariepinus*)[b]		0.14 ± 0.12	–	–	–	< LOQ	8.0 ± 11	< LOQ	< LOQ
Nile River, Egypt (*Oreochromis niloticus*)[c]		0.00082 ± 0.00011	–	–	–	–	–	–	–
Lakes of the Tibetan Plateau[d]		0.32	–	–	–	0.21	4.0	0.55	–
Lake Chad, Chad (*Synodontis schall*)[e]		2.6	–	–	–	–	0.79	0.69	–
Tiber River, Italy (*Leuciscus cephalus*)[f]		99	–	–	–	–	–	–	13
Scheldt estuary, Belgium (*Platichthys flesus*)[g]		105	0.00	0.01	0.00	0.16	0.56	0.06	1.2
Lake Tanganyika, Burundi (*Stolothrissa tanganikae*)[h]		64 ± 15	–	–	–	6.5 ± 1.2	124 ± 19	55 ± 12	–
Vaal River, South Africa (*Labeo capensis*)[i]		371 ± 113	< 0.5	5.8 ± 1.8	2.6 ± 0.6	257 ± 204	161 ± 47	10 ± 0.9	5.9 ± 1.0
Hadley Lake, US (*Pomoxis annularis, Lepomis macrochirus*)[j]		–	–	–	–	–	–	–	65 ± 8.0
Lan-Yang River, Taiwan[k]		–	–	–	–	–	–	–	25 ± 11
Erh-Jen estuary, Taiwan[k]		–	–	–	–	–	–	–	281 ± 210
Lake Mjøsa, Norway (*Salmo trutta*)[l]		–	–	–	–	–	–	–	342 ± 312
River Turia, Spain (*Salmo trutta*)[m]		6.9	–	–	–	–	4.3	–	–

< LOQ = below Limit of Quantification.

[a] Verhaert et al., 2013.
[b] Verhaert et al., 2017.
[c] El-Kady et al., 2007.
[d] Yang et al., 2010.
[e] Kidd et al., 2004.
[f] Miniero et al., 2011.
[g] Van Ael et al., 2012.
[h] Manirakiza et al., 2002.
[i] Wepener et al., 2011.
[j] Dodder et al., 2002
[k] Peng et al., 2007.
[l] Mariussen et al., 2008.
[m] Bordajandi et al., 2003.

Table 4
Mean concentration (± SD) of THg in water samples (μg/l) (n = 3), sediments (μg/g dw) (n = 2) and biota (μg/g ww biota) from the Ga-Selati river (South-Africa) (n = 5) and the rivers Laagen (n = 3) and Rena (n = 3) (Norway) compared to other studies.

	Water	Sediment	Invertebrates	Fish		Water	Sediment	Invertebrates	Fish
South Africa			*Gomphidae* sp.		Norway			*Lymnaea* sp.	
Namakgale	0.03 ± 0.01	0.02	0.03 ± 0.008	*Clarias gariepinus* 0.11 ± 0.02 *Labeo molybdinus* 0.03 *Chiloglanis paratus* 0.30 *Glossogobius callidus* 0.06 ± 0.02 *Clarias* gariepinus 0.04 ± 0.05	Rena	< LOQ	0.02	0.03 ± 0.003	*Salmo trutta* 0.06 ± 0.01 *Esox lucus* 0.13 ± 0.07
LB	0.02 ± 0.005	0.002	0.01 ± 0.005	*Labeo molybdinus* 0.01 ± 0.003 *Synodontis zambeziensis* 0.11 ± 0.03 *Tilapia rendalli* 0.02 ± 0.001 *Clarias gariepinus* 0.25 ± 0.14	Laagen	< LOQ	0.07	0.03 ± 0.004	*Gymnocaphalus cernuus* 0.24 ± 0.07 *Phoxinus phoxinus* 0.09 ± 0.006 *Salmo trutta* 0.05 ± 0.005 *Esox lucius* 0.55 ± 0.17 *Cottus poecilopus* 0.11 ± 0.03
Harmonie	0.02 ± 0.006	0.001	0.007 ± 0.004	*Labeo molybdinus* 0.03 *Tilapia mossambicus* 0.02 ± 0.004 *Barbus trimaculatus* 0.09 ± 0.008					
Lake Victoria[a]									
Napoleon Gulf, Uganda	0.0037 ± 0.0011	0.18	–	–					
Emin Pasha Gulf, Tanzania	0.88 ± 0.76	0.59	–	–					
Atatürk Dam lake, Turkey(*Capoetta trutta*)[b]	< LOQ	< LOQ	–	< LOQ					
Thigithe River, Tanzania (*Labeo victorianus*)[c]	< LOQ	0.077 ± 0.15	–	0.05–0.90					
Madeira River, Bolivia (*Pseudoplastystoma tigrinum*)[d]	0.0077 ± 0.0013	–	–	0.99					
Mekong River, Vietnam (*Macrobranchium equidens*)[e]	< LOQ	–	0.08 ± 0.03*	–					
Lake Murray, Papua New Guinea (*Arius berneyi*)[f]	0.0014 ± 0.0012	0.11 ± 0.06	–	0.23 ± 0.17					
Yangtze River, China[g]	–	0.19 ± 0.21	–	–					
Nikonga River, Tanzania (Gastropoda sp.)[h]	–	0.02	0.040	–					
Canon river, Taiwan[i]	–	3.41 ± 2.95	–	–					
Kwilu River, Congo (*Clarias pachynema*)[j]	–	0.22 ± 0.32	–	0.44 ± 0.49					
Lake Chad, Central Africa (*Etheria elliptica*)[k]	–	–	0.019 ± 0.009	–					
Lake Ontario, Canada (*Echinogammarus fasciatus*)[l]	–	–	0.0087 ± 0.0025	–					
Río Las Marías, Venezuela (*Astyanax integer*)[m]	–	–	–	0.37 ± 0.22					

< LOQ = below Limit of Quantitation. *Concentration in dry weight, compared to a mean THg concentration of 0.10 ± 0.080 and 0.16 ± 0.053 μg/g dw in samples from South Africa and Norway respectively.

[a] Campbell et al., 2003.
[b] Karadede and Erhan, 2000.
[c] Mataba et al., 2016.
[d] Maurice-Bourgoin et al., 2000.
[e] Ikemoto et al., 2007.
[f] Bowles et al., 2001.
[g] Yi et al., 2011.
[h] Taylor et al., 2005.
[i] Chen et al., 2007.
[j] Ngelinkoto et al., 2014.
[k] Kidd et al., 2004.
[l] Zhang et al., 2012.
[m] Kwon et al., 2012.

present in *Lymnaea* snails (BDE 183 (90%) and BDE 47 (10%)), while only BDE99 was found in *Gomphidae* larvae.

3.1.3. Fish

Lipid content (%) ranged from 0.6 ± 0.1% (*Esox lucius*) to 5.5 ± 1.7% (*Synodontis zambezensis*). An overview of the ΣPCBs, OxC, TN, CN, HCB, ΣDDX, ΣHCHs and ΣPBDEs concentrations (ng/g ww aquatic biota) in the analyzed fish is given in Table 3. Relative distribution of indicator PCBs, DDX and PBDE congeners in sediment, invertebrates and fish samples are given in Fig. S1.

Dominant congeners in South African samples were CB 153 (18%), CB 74 (16%), CB 138 (13%), CB 118 (11%) and CB 180 (10%). In Norwegian fish, the most dominant congeners were CB 153 (25%), CB 138 (13%), CB 118 (7%) and CB 180 (7%). ΣPCBs concentrations in muscle from South African specimens in this analysis were comparable to studies of relatively pristine rivers in Central Africa and rivers located in Egypt and Central Asia, but were higher in Norwegian specimens from the present study (Kidd et al., 2004; El-Kady et al., 2007; Yang et al., 2010; Verhaert et al., 2013). Concentrations in fish from both South Africa and Norway were much lower compared to a lake located in Burundi and in rivers from more industrialized areas such as in Italy and Belgium (Manirakiza et al., 2002; Miniero et al., 2011; Van Ael et al., 2012).

Chlordanes (OxC, TN, CN) were only detected in a few fish from this study, and comparable to concentrations reported in a Belgian study (Van Ael et al., 2012), but lower than in fish from another South African river, the Vaal River, which is heavily influenced by the runoff of surrounding cities, industries and mining activities (Wepener et al., 2011). Detectable concentrations of HCB were only reported in one South African fish sample, while present in all but one of the Norwegian species. Concentrations in Norwegian fish from this study were comparable to those reported in studies on fish from rivers in Belgium and the Tibetan plateau (Yang et al., 2007; Van Ael et al., 2012), but lower than in fish from rivers in Congo (Verhaert et al., 2013) and the heavily polluted Vaal River in South Africa (Wepener et al., 2011).

BDE 99 was the only congener present in South African samples from this study. PBDE concentrations were low in South African fish samples compared to lakes in the United States, rivers in Belgium, Congo, Italy, Taiwan and another South African region and especially compared the heavily polluted Lake Mjøsa in Norway (Dodder et al., 2002; Peng et al., 2007; Mariussen et al., 2008; Miniero et al., 2011; Wepener et al., 2011; Van Ael et al., 2012; Verhaert et al., 2013). PBDEs were more abundant in fish from Norway, although they were still generally lower than aforementioned studies. The most dominant congeners were BDE 47 (42%), BDE 99 (35%) and BDE 100 (13%).

All six DDT metabolites were detected in fish from this study, except for *o,p'*-DDE being absent in Norwegian fish. ΣDDX concentrations in fish from South African sites were much higher than studies carried out in Belgium and Spain and water bodies from Congo and Chad, but comparable to other African countries such as Burundi and another region in South Africa; concentrations in Norwegian fish were lower than South African fish but still higher than those reported in a study from Belgium and Chad (Kidd et al., 2001; Manirakiza et al., 2002; Bordajandi et al., 2003; Wepener et al., 2011; Van Ael et al., 2012; Verhaert et al., 2013). The most present DDX metabolites in South African fish were *p,p'*-DDE (70%), *p,p'*-DDT (17%) and *o,p'*-DDT (5%), and *p,p'*-DDE (64%), and *p,p'*-DDT (26%) in Norwegian fish. *p,p'*-DDE is very soluble in lipids compared to other congeners which explains its abundance in animal tissue (ATSDR, Agency for Toxic Substances and Disease Registry, 2002), as found in other studies (Van Ael et al., 2012; Verhaert et al., 2013).

Finally, ΣHCHs concentrations in South African fish from this study were comparable to those reported in studies from Tibet, Congo, Chad and Belgium but lower than other studies from Burundi and the Vaal River in South Africa (Kidd et al., 2001; Manirakiza et al., 2002; Yang et al., 2007; Wepener et al., 2011; Van Ael et al., 2012; Verhaert et al., 2013). Similarly to the sediment samples, only γ-HCH was detected. ΣHCHs concentrations were below the detection limit in Norwegian fish in this study.

Relationships between concentrations of POPs in TOC normalized sediment and lipid normalized biota were investigated in South African samples. Concentrations in *Clarias gariepinus* were used as this fish species occurred at each site. No significant relationships could be found between POP concentrations in biota and sediment.

3.2. Mercury

3.2.1. Surface water

Mean dissolved THg concentrations (μg/l) (Table 4) show no detectable amount of Hg in the water from the Norwegian rivers. Dissolved THg concentrations in the Ga-Selati river however were higher than concentrations reported in studies on the more pristine parts of lake Victoria (Napoleon Gulf, Uganda) and studies from Turkey, Tanzania, Bolivia, Vietnam and Papua New Guinea but still lower than the more polluted areas of the Lake Victoria (Emin Pasha Gulf, Tanzania) (Karadede and Erhan, 2000; Maurice-Bourgoin et al., 2000; Bowles et al., 2001; Campbell et al., 2003; Mataba et al., 2016).

3.2.2. Sediment

Mean concentrations of THg in sediments (Table 4) indicate concentrations of THg in both South African and Norwegian sediments lower than those reported for in sediments from the Yangtze river (China), Kwilu river (Congo), Thigithe River (Tanzania) and Lake Victoria (Uganda and Tanzania) but comparable to concentrations in the sediments of the Malagarasi River (Tanzania) (Campbell et al., 2003; Taylor et al., 2005; Yi et al., 2011; Ngelinkoto et al., 2014; Mataba et al., 2016; Chen et al., 2007). Concentrations of THg in sediment from the Norwegian rivers in the present study are higher than dissolved concentrations, which supports the fact that the sediment can act as a reservoir (Foster and Charlesworth, 1996). However, for the South African sites this was not the case. The fact that in general dissolved THg concentrations in the Ga-Selati river were higher in the water compared to sediment is rather surprising since organic matter has been shown to increase the adsorption of metals on sediments (Lin and Chen, 1997). Furthermore, the high pH of water from the Ga-Selati river, ranging from 8.5 to 9.8 across all sites, should positively influence the adsorption on sediment because a higher pH leads to a higher binding of Hg to organic material (Haitzer et al., 2003). Dissolved THg concentrations were comparable among the three South African sites, but sediment concentration was notably higher at Harmonie, which had the highest TOC value (3%) (Table 1). A higher input of pesticides at that site could also play a role, as several commonly used pesticides contain substantial amounts of metals such as Hg (Wuana and Okieimen, 2011).

3.2.3. Invertebrates

Mean THg concentrations in invertebrates (Table 4) are comparable to THg concentrations reported in studies from Tanzania, Central Africa and Vietnam but higher than in invertebrates from a Canadian lake (Kidd et al., 2004; Taylor et al., 2005; Ikemoto et al., 2007; Zhang et al., 2012).

3.2.4. Fish

Mean THg concentrations in fish muscle tissue in the present study (Table 4) were in general slightly lower or comparable to concentrations reported in studies from Congo, Bolivia, Venezuela, Papua New Guinea and Tanzania, but higher than in fish from a lake in Turkey (Maurice-Bourgoin et al., 2000; Karadede and Erhan, 2000; Bowles et al., 2001; Chen et al., 2007; Kwon et al., 2012; Ngelinkoto et al., 2014; Mataba et al., 2016). In general, the uptake of most metals occurs primarily through water and to a lesser extent through food. Hg however occurs primarily as the organic form, methyl mercury (MeHg). Due to the methylation by bacteria, the properties of mercury alter in such a way that it becomes much more lipophilic and more mobile, entering any substrate that contains fat, especially aquatic organisms (Hodson, 1988). As methyl mercury is conserved while energy is lost through trophic transfer, this compound can be highly biomagnified through the

food chain resulting in high THg concentrations in toppredators (Campbell et al., 2003).

Again, relationships between THg concentrations in TOC-normalized sediment and lipid-normalized biota were investigated in South African samples. No significant relationships could be found between THg concentrations in biota and sediment either.

3.3. Biomagnification

3.3.1. Trophic web structure

The ranges of nitrogen stable isotopes ratios and trophic levels (Table S1) indicate a variation in trophic level between 1.77 (*Labeo molybdinus*) and 5.34 (*Synodontis zambezensis*). Trophic levels determined in the present study were comparable with those reported on Fishbase (www.fishbase.org) for the majority of fish. Trophic levels of *T. rendalli*, *S. zambesensis*, *C. paratus* and *G. cernuus,* however, were higher compared to levels reported on Fishbase, while the trophic level of *B. trimaculatus* was lower in the present study. The small sample size of aforementioned fish might account for these discrepancies. In general, trophic levels were higher in the subarctic rivers of Norway (Fig. S2).

3.3.2. Trophic transfer and magnification

To assess the biomagnification, the relation between the log of the concentration of a compound in biota and the corresponding trophic level was examined. One food web per climate was used, being Lepelle Bridge in South Africa and Laagen in Norway, as they had the highest number of species and samples and to exclude the risk of pooling distinctive food webs together. Significant relations were found between the TL and the log of THg, all DDT-metabolites, several PCB compounds (CB 110, 118, 138 and 153), TN and CN concentrations for South African biota, and between the TL and the log of THg, CB153 and 138, TN, CN, *p,p′*-DDE and *p,p′*-DDT for Norwegian biota (Fig. 2). Calculation of TMFs

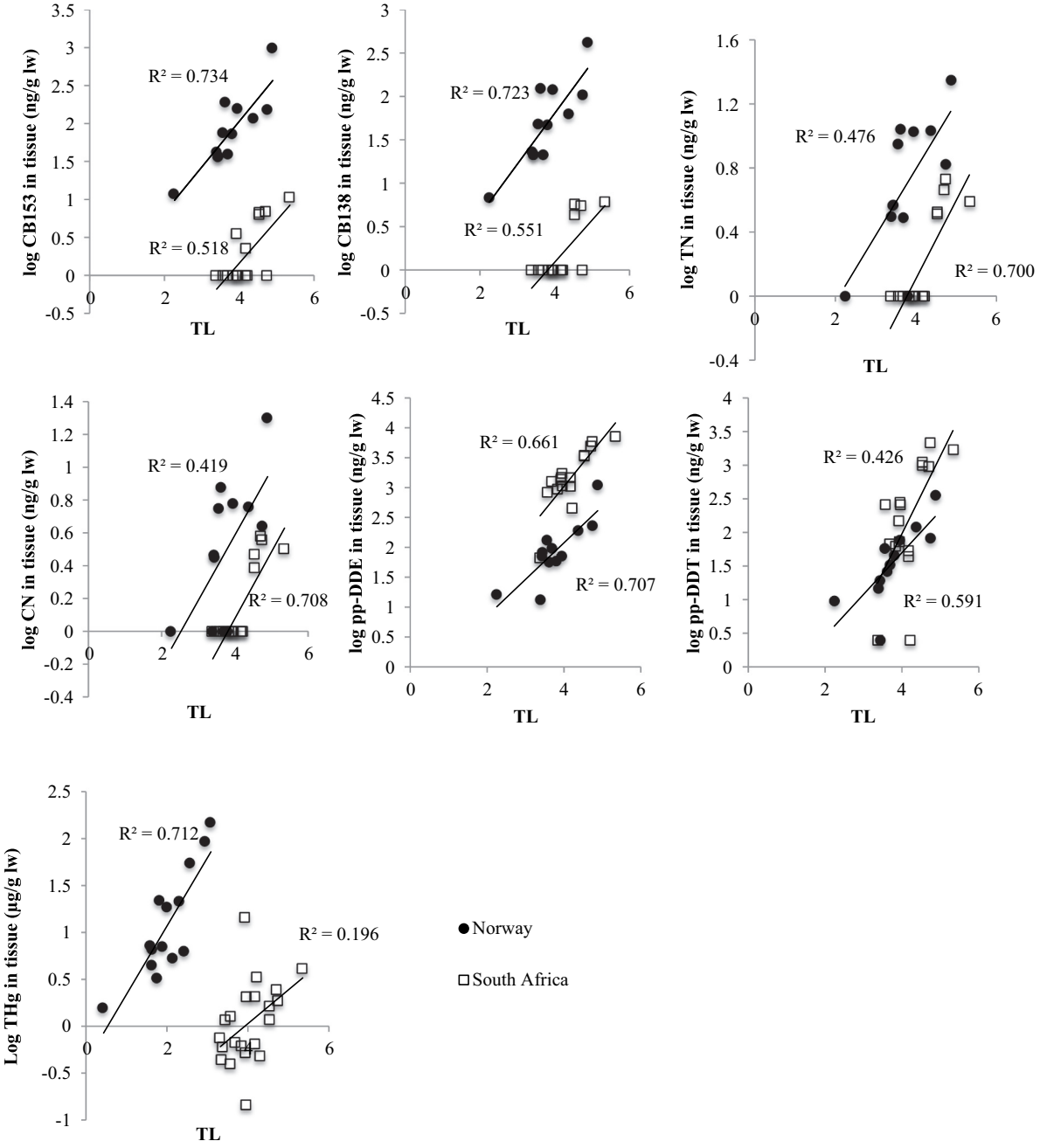

Fig. 2. Trophic transfer for CB153, CB138, TN, CN, *p,p′*-DDE, *p,p′*-DDT and THg in aquatic biota from South Africa and Norway.

Table 5
TMFs of significant relationships between the log of compound concentration and trophic level in aquatic biota from the present study, compared to other studies. (*) depicts a significant difference between climates.

	South Africa	Norway	Congo[a]	China[b]	U.S.[c]	Uganda[d]	Burkina Faso[e]	Vietnam[f]
THg	2.3*	5.4*	–	–	–	1.9–5.6	2.9–6.5	1.3
CB153	3.7	4.0	2.5	3.6	3.4	–	–	–
CB138	3.1	3.9	2.6	2.3	3.3	–	–	–
CB 118	2.8	–	–	3.6	–	–	–	–
CB 110	1.9	–	2.4	1.8	–	–	–	–
TN	3.1	2.6	–	–	3.6	–	–	–
CN	2.6	2.5	–	–	–	–	–	–
o,p′-DDE	24	–	–	–	–	–	–	–
p,p′-DDE	6.2	4.1	–	1.4	4.0	–	–	–
o,p′-DDD	9.1	–	–	–	–	–	–	–
p,p′-DDD	8.8	–	–	1.7	–	–	–	–
o,p′-DDT	10.3	–	–	–	–	–	–	–
p,p′-DDT	14*	4.2*	1.7	1.1	–	–	–	–

[a] Verhaert et al., 2013.
[b] Zhang et al., 2010.
[c] Houde et al., 2017.
[d] Poste et al., 2015.
[e] Ouédraogo et al., 2015.
[f] Ikemoto et al., 2007.

(Table 5) was only done for significant relationships. When biomagnification occurred for a compound in biota from both climates, it was possible to compare both slopes and assess the influence of climate.

In general, TMF values for PCBs were comparable to values reported from Congo, China and the U.S. (Zhang et al., 2010; Verhaert et al., 2013; Houde et al., 2017). TMFs of DDT compounds were found to be high in the South African food web compared to food webs from Norway (present study), Congo, China and the U.S. (Zhang et al., 2010; Verhaert et al., 2013; Houde et al., 2017). The initial concentrations of DDT compounds, which are usually higher in African countries, should not play a role in calculating the TMFs, as TMFs are not related to exposure concentrations at the base of the food webs (Borgå et al., 2012). It has been previously reported that the biomagnification of POPs, with the exception of DDT-compounds, didn't follow a clear relationship with trophic level and that consequently $\delta^{15}N$ wasn't effective to predict the accumulation of other POPs than DDT (Campbell et al., 2000; Guo et al., 2008; Deribe et al., 2011). In this study, only *p,p′*-DDT bioaccumulated differently in the different climates ($p = 0.03$). A global analysis of studies on trophic magnification of POPs suggests that latitude doesn't influence biomagnification in food webs, and that differences in TMF values likely result from differences in food web composition rather than temperature dependent processes such as growth and physiological changes (Walters et al., 2016). However, major data gaps still exist as a result of overrepresentation of studies mainly conducted in temperate climates of the northern hemisphere, which emphasizes the need to investigate biomagnification in the southern hemisphere and other types of climates (Walters et al., 2016).

In the current study, the TMF value for THg was significantly higher in Norway than in South Africa ($p = 0.0005$). TMF values from studies in lakes located in Uganda and Burkina Faso and a river in Vietnam were in general comparable to the TMF value in the South African river from the present study (Ikemoto et al., 2007; Ouédraogo et al., 2015; Poste et al., 2015). In contrast to POPs, it has been shown that the global biomagnification of mercury is positively related to latitude. Indeed, research shows that Trophic Magnification Slopes are significantly higher in polar and temperate regions as opposed to tropical ones (Lavoie et al., 2013). Although there is not enough data in this study to attribute the differences in biomagnification of THg to climatic or other factors, values from this study do follow the pattern found in literature. Several factors related to climatic conditions would explain this lower biomagnification of Hg in temperate or arctic food webs. The subtropical climate of the Ga-Selati river experiences higher temperatures throughout the year, resulting in a high primary productivity, which stimulates growth rates in the fish. This can lead to growth biodilution of Hg, as the amount of Hg per unit of body mass decreases with growth. Fish from the Norwegian rivers however have growth rates limited by the low temperatures and have to starve for long periods of the year. Shorter life spans of fish living in tropical conditions and a higher species diversity in tropical food webs can also play a role in reducing the trophic transfer of Hg (Lavoie et al., 2013; Poste et al., 2015). However, a few of the aforementioned African lakes had a TMF value comparable or even higher than the one reported for Norwegian rivers in the present study (Ouédraogo et al., 2015), depicting trophic status as a more influencing factor rather than latitude. Indeed, the lakes with highest TMF values were described as eutrophic and mesotrophic compared to the other hypereutrophic lakes. The fact that higher TMF values can also be found in more tropical systems shows that latitude is just one of many factors that influence the biomagnification of Hg, and that trophic status also plays a key role in the biomagnification of Hg in an aquatic food web. Indeed, it has been shown that in African lakes trophic status has a negative relationship with Hg TMF and thus appears to be an important driver of TMFs (Poste et al., 2015). Further research needs to be carried out to better understand the determining factors influencing bioaccumulation of Hg.

3.4. Implications for human health

The mean concentrations of POPs and THg in fish captured in the Ga-Selati River and the Norwegian rivers have been compared with the corresponding Minimum Risk Levels for oral intake (ATSDR, Agency for Toxic Substances and Disease Registry, 2017). This information has been translated into the maximal amount of fish a human with the average weight of 70 kg can consume before experiencing negative effects due to exposure to excessive concentrations of pollutants. Taking into account that, according to the FAO, the South African population consumes an average of 21 g of fish a day (Fishery and Aquaculture Country Profiles, FAO, South Africa, 2010), it is unlikely that the recommended limits to avoid adverse health effects is exceeded when consuming fish from this study. The average fish consumption for the Norwegian population is 52 g of fish a day, as reported by the Norwegian Scientific Committee for Food Safety (VKM, Vitenskapskomiteen for mattrygghet, 2014). Considering this, caution should be taken when consuming Northern pike (*Esox lucius*) from the river Laagen to avoid harmful effects due to an elevated THg and PBDE exposure (Table 6). A

Table 6
Minimal Risk Levels for *p,p′*-DDT, ΣPBDEs (lower brominated), ΣPCBs, γ-HCH, CHLs and THg (ATSDR 2017), the corresponding mean concentrations in *Esox lucius* river Laagen (Norway), and the maximum amount of fish recommended to avoid risk of adverse non-cancer health effects due to the ingestion of the pollutants. (*) Depicts an amount exceeded by the average daily fish consumption of the Norwegian population.

	p,p′-DDT	ΣPBDEs (lower brominated)	ΣPCBs	γ-HCH	CHLs	THg
Minimal risk level (ng/kg body weight/day)	500	3	30	10	600	230
MRL (ng/day) for a 70 kg person	35,000	210	2100	700	42,000	16,100
Mean concentration in *Esox lucius* (ng/g ww)	0.78	8.1	7.1	< LOQ	0.14	550
Maximum edible amount of *Esox lucius* for a person of 70 kg (g)	45,000	30*	300	–	300	30*

detailed overview of all maximal amounts per fish species can be found in Table S2 for the Ga-Selati river and Table S3 for the rivers Laagen and Rena.

4. Conclusion

Overall, concentrations of POPs and mercury in water, sediment and aquatic biota were relatively low in the studied rivers. However, caution should be taken when consuming Northern pike from the river Laagen as PBDEs and THg concentrations exceeded the Minimum Risk Levels.

Biomagnification was detected for THg, all DDT-metabolites, several PCB compounds (CB 110, 118, 138 and 153), TN and CN in the South African river system while THg, CB153 and 138, TN, CN, *p,p′*-DDE and *p,p′*-DDT were found to be biomagnified in the Norwegian river system. The biomagnification of THg and *p,p′*-DDT differed significantly between both river systems, THg being more biomagnified in the Norwegian river and *p,p′*-DDT being more biomagnified in the South African river. Although this study doesn't have enough data to attribute these differences to climatic or other factors, biomagnification of THg has been shown in various studies to be higher in polar and temperate regions as well as being influenced by trophic status of the aquatic system. Biomagnification of POPs, however, does not seem to be significantly influenced by climate. Differences in biomagnification of POPs might be more influenced by the composition of the food web rather than by differences in temperature. Further research in underrepresented areas is essential to investigate global patterns and to identify the factors influencing bioaccumulation of POPs and Hg.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.10.058.

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