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Biological responses of midge (*Chironomus riparius*) and lamprey (*Lampetra fluviatilis*) larvae in ecotoxicity assessment of PCDD/F, PCB and Hg contaminated river sediments

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Abstract We evaluated utility of chironomid and lamprey larval responses in ecotoxicity assessment of polychlorinated dibenzo-*p*-dioxins, dibenzofurans (PCDD/F)-, polychlorinated biphenyls (PCB)- and mercury (Hg)-contaminated river sediments. Sediment samples were collected from the River Kymijoki with a known industrial pollution gradient. Sediment for the controls, and lamprey larvae were obtained from an uncontaminated river nearby. Contamination levels were verified with sediment and tissue PCDD/F, PCB and Hg analyses. Behaviour of sediment exposed chironomid and lamprey larvae were measured with Multispecies Freshwater Biomonitor© utilizing quadrupole impedance conversion technique. In addition, mortality, growth and head capsule deformity incidence of chironomids were used as ecotoxicity indicators. $WHO_{PCDD/F+PCB}$-TEQ in the R. Kymijoki sediments ranged from the highest upstream 22.36 ng g^{-1} dw to the lowest 1.50 ng g^{-1} near the river mouth. Sum of PCDD/Fs and PCBs correlated strongly with Hg sediment concentrations, which ranged from <0.01 to 1.15 µg g^{-1}. Lamprey tissue concentrations of PCDD/Fs were two orders and PCBs one order of magnitude higher in the R. Kymijoki compared to the reference. Chironomid growth decreased in contaminated sediments, and was negatively related to sediment ∑PCDD/Fs, $WHO_{PCDD/F+PCB}$-TEQ and Hg. There were no significant differences in larval mortality or chironomid mentum deformity incidence between the sediment exposures. The distinct behavioural patterns of both species indicate overall applicability of behavioural MFB measurements of these species in sediment toxicity bioassays. Chironomids spent less and lampreys more time in locomotion in the most contaminated sediment compared to the reference, albeit statistically significant differences were not detected. Lamprey larvae had also a greater activity range in some of the contaminated sediments than in the reference. High pollutant levels in lamprey indicate risks for biomagnification in the food webs, with potential health risks to humans consuming fish.

Keywords: polychlorinated dibenzo-*p*-dioxins and -furans; polychlorinated biphenyls; mercury; sediment toxicity; *Chironomus riparius*; *Lampetra fluviatilis*; behaviour

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INTRODUCTION

Sediment as a repository of different persistent chemicals may act as significant pollutant source to aquatic biota. Risk assessment of sediments typically relies on chemical concentration measurements. However, sediment concentrations of individual substances do not provide conclusive information on bioavailability and potential combined toxic effects of chemicals. Therefore, biological assays are needed to more comprehensively characterize potential ecological risks of exposure. Among bioassays, early warning systems measuring behavioural changes of fish and invertebrates have been increasingly used in ecotoxicity assessments of pollutants in water (e.g. van der Schalie et al. 2001; Amiard-Triquet 2009), whereas behavioural responses of sediment dwelling species are used less often, and standardized bioassays using sediment-dwelling organisms do not include behavioural responses as endpoints, in spite of numerous encouraging studies (Gerhardt et al. 2002; Petrauskienė 2003; Kirkpatrick et al. 2006; Sardo and Soares 2010). Chemical exposure may alter animal behaviour in multiple ways which often emerge at lower concentrations and earlier than mortality, and major changes at population or community level (Heinis et al. 1990; Vuori 1994). The altered behaviour may however, be later reflected to population, community and ecosystem levels if changes in activity results in e.g., impaired foraging and thereby reduces growth, reproduction and survival (Weiss et al. 2001; Dell'Omo 2002; Riddell et al. 2005). Hence, quantitative behavioural measurements may be relevant to evaluate ecological risks of contaminated sediments (Sardo and Soares 2010).

Organisms inhabiting sediments are potentially exposed to contaminants through feeding on sediment particles, or via direct epidermal contact with the sediment, and sediment pore water or overlying water (Hill et al. 1993). Larvae of both invertebrate chironomids and vertebrate lampreys are tube dwellers in soft sediments. They feed mainly on detritus and associated microbes (Johnson 1987; Mundahl et al. 2005), and are potentially exposed to sediment contaminants via all the routes mentioned above. Chironomids, especially laboratory reared *Chironomus riparius*, are frequently used in ecotoxicological assessments with mortality, growth and emergence as the endpoints in standard tests. Also behavioural responses have been occasionally measured in water but not in sediment (Gerhardt and Janssens de Bisthoven 1995; Langer-Jaesrich et al. 2010; Azevedo-Pereira and Soares 2010). Several studies have shown that due to their high lipid contents, lamprey larvae effectively bioaccumulate mercury and organic contaminants, such as PCDD/Fs and PCBs (e.g. Renaud et al. 1999; MacEachen et al. 2000; Soimasuo et al. 2004; Isosaari et al. 2006). Also some toxicity studies with lamprey eggs and larvae have been conducted (Myllynen et al. 1997; Andersen et al. 2010), but behavioural responses of lampreys to polluted sediments appear to be unexplored. A potential technique for measuring behavioural responses of aquatic animals is the Multispecies Freshwater Biomonitor©, MFB, which is a biomonitor quantitatively recording different behavioural patterns of animals by quadrupole impedance conversion technique (Gerhardt 2001). Each MFB test chamber has one pair of electrodes creating an electrical field within a chamber, and another pair of electrodes functioning as an impedance sensor. The data are analysed via a stepwise discrete Fast Fourier Transform (FFT), an algorithm converting the original periodic sinusoidal signal into its component frequencies. The frequency analysis yields a percentage of time an animal is spending on movements with frequencies that can be associated with specific behavioural patterns like ventilation or locomotion. MFB is able to record also behavioural responses of sediment burrowing species (Gerhardt et al. 2002; Sardo and Soares 2010), due to the non-optical technology. Possible behavioural deviations associated with the exposure can be detected by comparing behavioural profiles across exposure and control treatments.

Polychlorinated dibenzo-*p*-dioxins, dibenzofurans (PCDD/Fs) and biphenyls (PCBs) from anthropogenic sources have contaminated soils and sediments widely. These compounds are of special concern in risk assessment and management due to their global dispersal, low rate of degradation and high solubility in lipids, which make them very persistent and enable their accumulation into organisms and biomagnification along food webs, including humans (e.g. Sinkkonen and Paasivirta 2000; Kiviranta et al. 2001, 2004; Pereira 2004; Karjalainen et al. 2012). The half-life times of these compounds in sediment vary from years to hundreds of years (Sinkkonen and Paasivirta 2000). TCDD (2,3,7,8-tetrachlorodibenzo-*p*-dioxin) is one of the most toxic synthetic compounds and classified as a human carcinogen. Other toxic effects of dioxin include teratogenic,

hormonal and immunotoxic responses (Mandal 2005). Of all the 210 PCDD/F and the 209 PCB congeners, those structurally similar to TCDD (17 PCDD/Fs and 12 dioxin-like PCBs) are considered the most toxic. On the other hand, recent laboratory exposure of Péan et al. (2013) indicate that also other than dioxin-like PCBs can induce behavioural disruptions in fish at environmentally relevant concentrations. Low environmental concentrations and variation in toxic potency among congeners complicate risk assessment of these persistent compounds (Walker et al. 1991; Barber et al. 1998; Abnet et al. 1999). Also mercury (Hg) is ubiquitous in the environment, and in aquatic food webs it biomagnifies readily as organic methylmercury (e.g. Munthe et al. 2007; Lehnherr 2014). Hg exposure is known to induce neurotoxic effects along with behavioural, hormonal and reproductive changes in fish, birds, and mammals (Boening 2000; Scheuhammer et al. 2007).

Our aim in this study is to assess potential ecotoxicity of multi-contaminated river sediments via biological responses of sediment-dwelling chironomid (*Chironomus riparius*) and European river lamprey (*Lampetra fluviatilis*) larvae with multiple lines of evidence approach. More specifically, impacts of PCDD/F, PCB and Hg contamination gradient on traditional biological responses (mortality, growth, morphological deformities and bioaccumulation) and novel behavioural indicators are assessed and compared. Further, we evaluate suitability of the behavioural responses of midge and lamprey larvae as sediment ecotoxicity indicators.

MATERIALS AND METHODS

The study sediments

In our laboratory experiments we used sediment samples collected across a pollution gradient in River Kymijoki, and from River Urpalanjoki not affected by industrial effluents. Pollution history (PCDD/Fs, PCBs, Hg) and gradients in the R. Kymijoki are described by Salo et al. (2008) and Verta et al. (2009). Surface sediment samples were collected in two replicates from six sites, Kuusaansaari (sample codes 1A and 1B), Keltti (2A, 2B), Lopotti (3A, 3B), Koskenalusjärvi (4A, 4B), Ahvionkoski (5A, 5B) and Kyminlinna (6A, 6B) along a longitudinal contamination gradient in R. Kymijoki, and from one site in R. Urpalanjoki (7A, 7B) in July 2012 (Fig. 1). Sediment samples were taken using an Ekman-grab or kick net from shallow water near river banks where soft, silty sediments were found. Distance between the two replicate samples was approximately 5–10 m. Sediments were kept in dark at +4°C and homogenized using a 1.0 mm sieve to remove indigenous macrofauna prior to the exposures.

17 toxic PCDD/F congeners, 12 dioxin-like PCB congeners and 6 marker-PCBs along with 19 other PCB-congeners were analysed from frozen sediment and lamprey larval samples with an accredited method in the National Institute for Health and Welfare. Individual congeners are listed in the footnote of Table 1. The quantification was performed by gas chromatography with high resolution mass spectrometry (GC-HRMS). Measurement uncertainty for PCDD/Fs and for PCBs were ± 50% when WHO-TEQ < 1 pg g^{-1} dw, ± 40% when WHO-TEQ 1–5 pg g^{-1} dw, and ± 30% when WHO-TEQ > 5 pg g^{-1} dw. Mercury (Hg) concentrations of sediments were analysed by cold vapour atomic absorption spectrometry (CVAAS) with sample specific measurement uncertainties that ranged from ± 0.02 to ± 0.2 mg kg^{-1}. Also loss on ignition (LOI %) (SFS 3008) and dry weight of sediments were analysed. Sediment organic carbon (SOC) content was estimated from LOI according to Pajunen (2004), as y = 0.435*x-0.847 when LOI < 15 %, and y = 0.505*x-1.8 when LOI > 15 %, based on sediment data from 140 Finnish lakes.

Sediment PCDD/F and PCB mass concentrations (pg g^{-1} or ng g^{-1}) were normalized to molar concentrations (nmol g^{-1}) using molecular weight data from the review article of Mannetje et al. (2012). Mass concentrations were also normalized by calculating toxic equivalencies (TEQ) of World Health Organization ($WHO\text{-}TEQ_{1998}$ and $WHO\text{-}TEQ_{2005}$) (Van den Berg et al. 1998; Van den Berg et al. 2006). $WHO\text{-}TEQ_{1998}$ and $WHO\text{-}TEQ_{2005}$ indicate the overall toxic potency for fish and mammals, respectively. Upper bound values for ∑PCDD/F, ∑PCB, $WHO\text{-}TEQ_{1998}$ and $WHO\text{-}TEQ_{2005}$ were calculated assuming those congener concentrations below the limit of quantification (LOQ) equal to LOQ, and lower bound values excluding congener concentrations < LOQ.

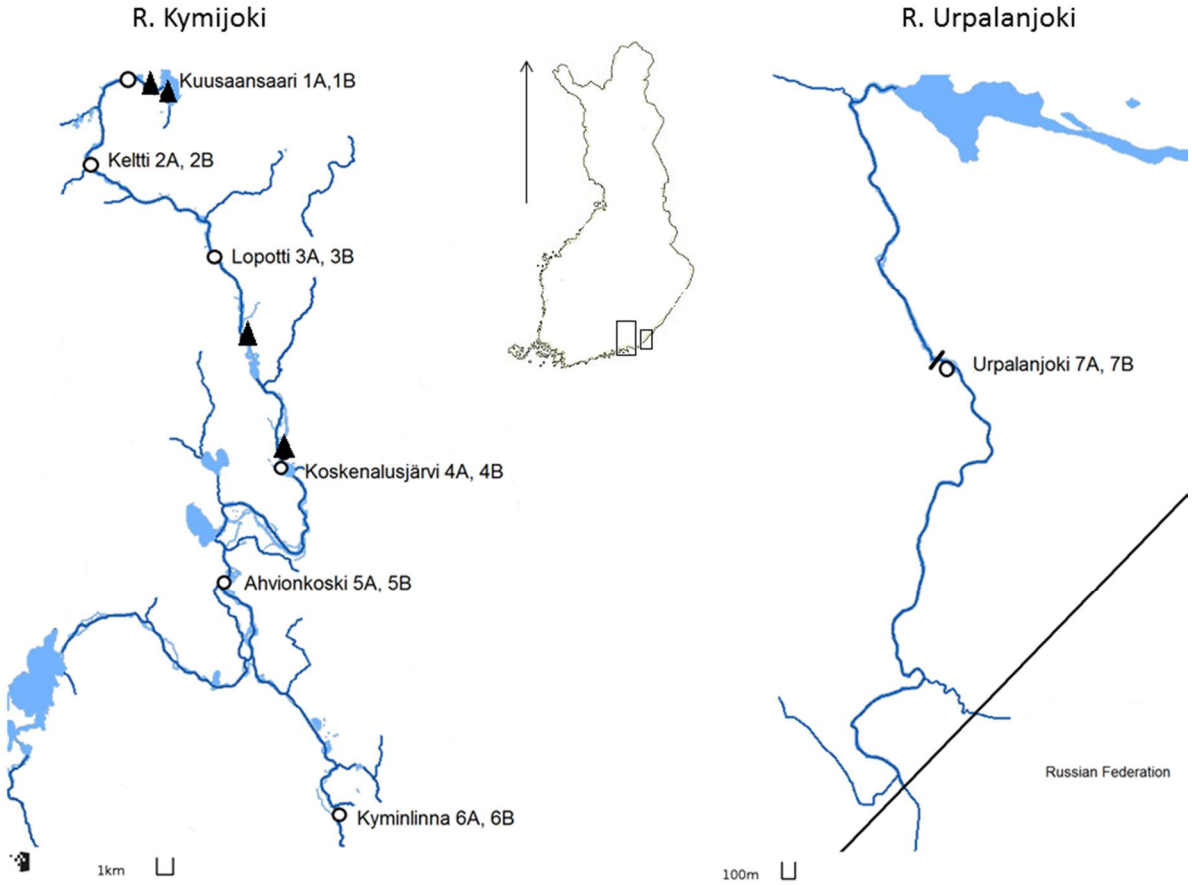

Figure 1. Study area, the contaminated River Kymijoki and the reference River Urpalanjoki in south-eastern Finland. Open circles indicate sediment sample sites. From the lowest downstream sites in R. Kymijoki, (sites 5 and 6), and from R. Urpalanjoki, also river lamprey larvae were sampled for tissue analysis. Black triangles indicate chemical or paper and pulp factories. Black transverse bar upstream from R. Urpalanjoki sampling site denotes a dam. Base map © National Land Survey of Finland.

Exposure setup and measured responses of Chironomus riparius

The midge larvae originated from the breeding stock of Finnish Environment Institute, Jyväskylä, Finland. Egg ropes ≤ 24 h from ovipositioning were collected on three consecutive days and placed in artificial freshwater on small Petri dishes with few droplets of fish food suspension (finely ground Tetramin® in artificial fresh water). Two days after hatching twenty 1st instar larvae were collected for each sediment treatment conducted in 600 ml borosilicate glass beakers. Each beaker contained 140 g of wet-sieved sediment with sediment:water ratio of 1:4, sediment layer of approximately 1.5 cm, and 2.0 cm^2 surface area per larva. The artificial freshwater used in all exposures was prepared according to the standard ISO 6341 (1996) ($CaCl*2\ H_2O$, $MgSO_4*7\ H_2O$, $NaHCO_3$, KCl) had total hardness of 0.5 mmol L^{-1}, and was buffered prior to exposures with phosphate buffer ($Na_2HPO_4*H_2O$, $Na_2H_2PO_4*H_2O$), with a final concentration of 1 mM, to pH 7 ± 0.1. After buffering, the water was gently aerated overnight and pH adjusted with 1 M HCl if necessary. Gentle aeration of the overlying water was started 24 h after transferring larvae into the beakers to allow the larvae to settle into the sediment. The larvae were fed with suspended aquarium fish food Tetramin® (0.25 mg larva^{-1} d^{-1}) and kept with photoperiod of 16:8 h light:dark (light intensity approx. 530 lx) at 20 ± 1°C during the exposures. Three replicates of six contaminated sediments (sites 1A, 2B, 3A, 4B, 5B and 6B) and six replicates of the reference sediment (7B) were used. Exposure replicates were started on three successive days with larvae hatched from three different egg ropes to get larvae of similar age and exposure history for the behavioural measurements, which could not be accomplished during one day.

Total number of exposed larvae was 480. Exposure duration was 10 days ending before the onset of pupation. Temperature (C°), oxygen (saturation %) and pH of the overlying water were measured at 0 d and at 10 d. Semi-static system was used and overlying water was partially replaced (1/4) three times during the exposure to avoid toxic concentrations of ammonia evolving associated with field sediments containing high levels of decaying organic material. Ammonia levels (NH_3/NH_4^+) were assessed approximately by an aquarium test kit in all test vessels in the 2nd exposure day before water renewals, and in the 10th exposure day from the reference and 6B exposures.

After the 10 d exposure, sediments were sieved with 0.5 mm mesh size sieve to find living chironomid larvae, and larval mortality (%) for each replicate was calculated. Larval wet weight (mg) was measured individually from samples preserved in 70 % ethanol. Just prior to weighting, larvae were held 10 min in clean tap water and surface-dried on tissue paper. Average larval wet weight (mg) per replicate was calculated. Also deformity response of chironomid larvae was measured and deformity incidence DI (%) (Hämäläinen 1999) calculated for each replicate. Only mentum deformities were considered. Missing or additional teeth, and Köhn gaps were counted as deformities, and to avoid a preconception bias, deformity analysis was conducted blind (Salmelin et al. 2015).

Larval behaviour types were evaluated before the exposures via oscilloscope-function of MFB, which allows the amplitude and frequency of the behavioural signals to be compared simultaneously with the visual observation of the larva. After the 10 d exposure, larvae were placed individually in the MFB-test chambers with the corresponding sediment and artificial test water (1:4). The larvae were acclimated to the test chambers for 30 min before the measurements started. Behaviour was measured for 2 h with 4 min periods at 10 min intervals, resulting in 12 measuring periods per larva.

Fourth instar chironomid larvae were used in behavioural measurements (n = 9 per exposure) and their behaviour was measured once after the 10 d exposure. In addition to this, chironomid larvae grown in the reference sediment were placed in the MFB-test chambers with contaminated sediment from site 2A, and their behaviour was measured.

Exposure setup and measured responses of Lampetra fluviatilis

River lamprey larvae used in the exposures were sampled in August 2013 from the River Urpalanjoki, and transported to the laboratory with the river sediment and water, and with continuous aeration. These 0+ larvae, hatched in early June, with length of 17–22 mm, were acclimated to the laboratory conditions for one week before exposures. Lamprey larvae were exposed individually (n = 6 larvae per exposure) to three differently contaminated sediments (2A, 4A and 6B) and to the reference sediment (7A). The same sediments as in *C. riparius* exposure could not be used due to insufficient amount of sediments and number of lamprey larvae. The larvae (n = 24) were allocated randomly into each test vessel, a 600 ml borosilicate glass beaker. Each beaker contained 100 g of wet-sieved sediment of approximately 1.5 cm in depth and sediment to water -ratio of 1:4. Gentle aeration of the overlying water was maintained during the exposures and larvae were fed with 0.75 mg $larva^{-1}$ d^{-1} of suspended mixture (1:1) of aquarium fish food (Tetramin®) and yeast and kept with photoperiod of 16:8 h light:dark (light intensity approx. 530 lx) at 20 ± 1°C. Exposure duration was 28 days. Mortality of lamprey larvae was checked five times when conducting the behavioural measurements at days 0, 3, 7, 14 and 28. Behaviour was measured with a similar set up as described above for *C. riparius*, except that larger MFB-test chambers were used for the lamprey larvae. Length and wet weight of anesthetized (clove oil at 100 mg l^{-1}) lamprey larvae was measured at d 28 after behavioural measurements, and larvae were decapitated.

Temperature (C°), oxygen (saturation %) and pH of overlying water were measured four times during the exposure at days 0, 3, 7, 14 and 28. Semi-static system was used and overlying water was partially (1/4) replaced every third day to avoid toxic ammonia levels. Ammonia levels (NH_3/NH_4^+) were assessed approximately by an aquarium test kit twice in the reference (7A) and 6B sediment exposures at days 2 and 9 immediately before water renewals, and ammonium ion (NH_4^+) concentration was analysed once at day 9 from 6B sediment exposure with an accredited method SFS-EN ISO 11732:2005.

River lamprey larvae for tissue residue analysis were present in and collected from the two lowest downstream sites of the R. Kymijoki (Kyminlinna and Ahvionkoski) and R. Urpalanjoki. The size of larvae ranged from 45 to 130 mm indicating a few years residence time in the river sediment. One

composite larval sample was formed for each site to ensure sufficient amount of tissue for PCDD/F and PCB analyses, which were conducted as described for the sediments above. Biota Sediment Accumulation Factor (BSAF) was calculated for PCDD/F congeners, PCB congeners, ∑PCDD/F and ∑PCBs from upper bound values as

$$BSAF = \frac{C_O / f_l}{C_S / f_{SOC}}$$

where C_O is the chemical concentration in the organism, f_l is the lipid fraction of the organism, C_S is the chemical concentration in sediment and f_{SOC} is fraction of the sediment as organic carbon estimated from LOI as described earlier (g organic carbon/g dry weight) (Ankley *et al.* 1992). Only congeners with the measured concentration above the LOQ were included in the calculations.

Statistical analyses

The association of response variables (chironomid wet weight, mortality and deformity incidence, lamprey larvae body burden, sediment loss on ignition, MFB data) with sediment PCDD/Fs, PCBs and Hg was studied using Pearson or Spearman (for variables not normally distributed) correlation.

Kolmogorov-Smirnov or Shapiro-Wilk test were used to evaluate normality of distribution and Levene test to examine among treatment homogeneity of variances of the behavioural, growth and mortality data. If the data could not be normalized using arcsin (for frequency data) or logarithmic transformations, non-parametric Kruskal-Wallis test was used to examine differences among sediment exposures, followed if necessary with pairwise comparisons by Mann-Whitney test. Chironomid larval behaviour was analysed using fixed factor univariate general linear model, egg rope as a block factor and sediment as a treatment factor. Additionally, non-parametric Friedman test (Related-Samples Friedman's Two-Way Analysis of Variance by Ranks) was used to test if lamprey larval behaviour differed between exposure days within a certain sediment treatment.

IBM SPSS Statistics 22 software was used in all statistical analyses.

RESULTS

Pollutant concentrations, congener profiles and bioaccumulation

Sediment dry weight ranged from 15.7 to 67.4 %. Loss on ignition ranged from 3.5 to 26.9 % indicating the lowest organic matter content in the reference sediment, where also dry matter content was high (Table 1). The lower-bound concentrations for the ∑PCDD/Fs, ∑PCBs and marker PCBs in the R. Kymijoki sediments were on average 100.0 %, 99.0 % and 98.5 % of those measured upper bound, respectively. In R. Urpalanjoki the lower-bound ∑PCDD/Fs were on average 6.9 % of the upper bound values. All PCBs in R. Urpalanjoki sediments were < LOQ except for CO-PCB-81 in 7A sample. The lower-bound sediment $WHO_{PCDD/F+PCB}$-TEQs were on average 99.9 % of those measured upper bound in R. Kymijoki, but 0.9 % in the reference site. The lower-bound ∑PCDD/Fs, ∑PCBs and marker PCBs in lamprey larvae tissue (lipid) were the same as upper bound concentrations, and only in the larvae from the reference site the lower bound ∑PCDD/Fs were 99.5 % of the upper bound. The lower-bound tissue $WHO_{PCDD/F+PCB}$-TEQs were 100.0 % of those measured upper bound in R. Kymijoki, and 99.1 % in the reference site. Hereafter all the sum and TEQ-concentrations are reported as the upper bound values.

The highest concentrations of ∑PCDD/Fs in R. Kymijoki were found from the upper reach of the river (site 2A 19.33 nmol g^{-1} or 8 423 ng g^{-1} dw), and the lowest concentration near the river mouth (site 6A 0.86 nmol g^{-1} or 371 ng g^{-1} dw) (Table 1, Fig. 1). At the reference site ∑PCDD/Fs were 6.38×10^{-5} nmol g^{-1} or 0.027 ng g^{-1} dw.

The sediment ∑PCBs in R. Kymijoki ranged from 10.51 nmol g^{-1} dw (2 971 ng g^{-1} dw) at site 4B to 0.13 nmol g^{-1} (38 ng g^{-1} dw) at site 6A (Table 1). Two distinct PCB concentration peaks were detected, in sites 2B and 4B. In the reference site the lower and upper bound values ranged from 0.0 to 0.03 nmol g^{-1} dw (9.1 ng g^{-1} dw) since most congeners were below the LOQ.

Table 1. Sediment dry matter content (%), loss on ignition (LOI %), organic carbon content (SOC %) and concentrations of ∑PCDD/Fs, ∑PCBs (nmol g^{-1}), $WHO_{PCDD/F-PCB}$-TEQ_{1998} for fish and $WHO_{PCDD/F-PCB}$-TEQ_{2005} for mammals (ng g^{-1}) and Hg (µg g^{-1}) in sediment dry weight (dw) in R. Kymijoki and R. Urpalanjoki.

Site	Dry matter	LOI	SOC[a]	∑PCDD/F[b]	∑PCB[c]	$WHO_{PCDD/F+PCB}$-TEQ_{1998}	$WHO_{PCDD/F+PCB}$-TEQ_{2005}	Hg
	%	% (±SD)	% (±SD)	nmol g^{-1}	nmol g^{-1}	ng g^{-1}	ng g^{-1}	µg g^{-1} (±SD)
R. Kymijoki								
1A*	20.4	15.7 (0.1)	6.2 (0.05)	3.69	1.77	7.11	7.33	0.54 (0.07)
1B	20.2	16.6 (0.1)	6.6 (0.03)	2.13	2.99	3.67	3.83	0.58 (0.03)
2A#	23.6	18.2 (0.8)	7.4 (0.42)	19.33	3.28	20.97	22.36	1.00 (0.00)
2B*	28.9	17.1 (0.1)	6.8 (0.05)	18.07	6.96	20.30	21.59	1.15 (0.05)
3A*	21.4	13.2 (0.4)	4.9 (0.16)	12.21	1.23	18.24	18.99	0.64 (0.06)
3B	44.5	6.5 (0.1)	2.0 (0.04)	2.74	2.89	4.57	4.74	0.13 (0.03)
4A#	35.1	8.6 (0.2)	2.9 (0.08)	3.50	4.31	5.77	6.00	0.78 (0.32)
4B*	43.8	7.6 (0.1)	2.5 (0.04)	1.73	10.51	3.03	3.14	0.65 (0.08)
5A	20.2	20.7 (0.3)	8.7 (0.15)	0.99	2.43	1.69	1.75	0.34 (0.03)
5B*	15.7	26.9 (0.9)	11.8 (0.46)	0.92	2.06	1.58	1.64	0.46 (0.13)
6A	32.7	19.6 (1.8)	8.1 (0.90)	0.86	0.13	1.44	1.50	0.26 (0.02)
6B*#	33.9	19.7 (1.5)	8.1 (0.77)	1.25	0.19	2.07	2.15	0.31 (0.04)
R. Urpalanjoki								
7A#	56.6	5.3 (0.1)	1.5 (0.03)	$6.4x10^{-5}$	0.03	$8.0x10^{-4}$	$7.7x10^{-4}$	< 0.01
7B*	67.4	3.5 (0.2)	0.7 (0.10)	$6.4x10^{-5}$	0.03	$8.8x10^{-4}$	$8.6x10^{-4}$	< 0.01

[a] SOC estimate is calculated from LOI

[b] The sum of 17 toxic congeners (2,3,7,8-TCDD; 1,2,3,7,8-PeCDD; 1,2,3,4,7,8-HxCDD; 1,2,3,6,7,8-HxCDD; 1,2,3,7,8,9-HxCDD; 1,2,3,4,6,7,8-HpCDD; OCDD; 2,3,7,8-TCDF; 1,2,3,7,8-PeCDF; 2,3,4,7,8-PeCDF; 1,2,3,4,7,8-HxCDF; 1,2,3,6,7,8-HxCDF; 2,3,4,6,7,8-HxCDF; 1,2,3,7,8,9-HxCDF; 1,2,3,4,6,7,8-HpCDF; 1,2,3,4,7,8,9-HpCDF; OCDF)

[c] The sum of 37 analysed congeners (DL-PCBs 77, 81, 126, 169, 105, 114, 118, 123, 156, 157, 167, 189; marker-PCBs 28/31, 52, 101, 138, 153, 180; and PCBs 18, 33, 47, 49, 51, 60, 66, 74, 99, 110, 122, 128, 141, 170, 183, 187, 194, 206, 209)

[*] Sediments used in *C. riparius* exposures

[#] Sediments used in *L. fluviatilis* exposures

The total toxic potency of sediments as $WHO_{PCDD/F+PCB}$-TEQ_{1998} for fish ranged from 0.0008 ng g^{-1} dw (reference site 7A) to 20.97 (site 2A), and was similar with $WHO_{PCDD/F+PCB}$-TEQ_{2005} for mammals (Table 1) although the latter values were slightly higher due to estimated higher toxicity of dioxins and some dioxin-like PCBs to mammals than fish. R. Urpalanjoki proved to be an appropriate reference site with very low sediment PCDD/Fs, PCBs and Hg. Most of the TEQ load in R. Kymijoki sediments was due to PCDD/Fs, and dioxin-like PCBs have only minor contribution between 0.01–9.21 % to the overall toxic potency.

Sediment Hg correlated strongly with ∑PCDD/F (r_s = 0.82, $p < 0.001$) and with ∑PCB (r_s = 0.81, $p < 0.001$), the highest Hg concentration being in the upper river (site 2A, 1.2 µg g^{-1} dw) (Table 1). In the reference site sediment Hg concentration was very low (< 0.01 µg g^{-1}).

Lamprey larval concentration of ∑PCDD/Fs in R. Kymijoki ranged from 0.01 to 0.03 nmol g^{-1} dw, and in the reference site it was only $5.40x10^{-5}$ (Table 2). Lamprey tissue ∑PCBs varied from 0.69 to 1.22 nmol g^{-1} dw in R. Kymijoki, and was ten or twenty times lower, 0.07 nmol g^{-1} dw, in R. Urpalanjoki. Sediment PCDD/F congener concentration (dw) correlated strongly with corresponding lamprey larvae tissue congeners (r_s = 0.90 $p < 0.001$) as well as sediment and tissue PCB congeners (r_s = 0.83, $p < 0.001$). BSAF for ∑PCDD/F were on average 0.021 (± 0.003) and for ∑PCB 2.437 (± 2.623) across two R. Kymijoki sites (Table 2). Based on BSAFs, accumulation was lower for highly chlorinated PCDD/Fs. For example, in R. Kymijoki BSAFs for OCDD and OCDF were on average 0.033 (±SD 0.019) compared to BSAFs 1.708 for 2,3,7,8-TCDD, and 2.308 (±SD 1.37) for 2,3,7,8-TCDF (Table S5).

Table 2. Sum of PCDD/Fs and PCBs (nmol g^{-1} and ng g^{-1}), $WHO_{PCDD/F+PCB}$-TEQ (ng g^{-1}) for fish (1998) and for mammals (2005) and in lamprey larvae tissue dry weight (dw) and lipid, and biota-sediment accumulation factor (BSAF) for ∑PCDD/F_S and ∑PCBs from sediment organic carbon to larval lipid in R. Kymijoki. ∑PCDD/Fs are the sum of 17 toxic congeners and ∑PCBs the sum of 37 analysed congeners. Ratio of larval tissue contaminants (dw) indicated in contaminated Ahvionkoski and Kyminlinna sites in relation to the reference site Urpalanjoki.

Site	**R. Kymijoki Ahvionkoski**			**R. Kymijoki Kyminlinna**			**R. Urpalanjoki**		**Ratio[a]**	**Ratio[b]**
Fat (%)	6.3			11.1			7.4			
Dry matter (%)	23.5			26.6			25.8			
	dw	lipid	BSAF	dw	lipid	BSAF	dw	lipid	dw	dw
∑**PCDD/Fs** ng g-1	4.75	17.74	0.023	14.60	34.88	0.019	0.02	0.07	238	730
∑**PCDD/Fs** nmol g-1	0.011	0.042		0.034	0.082		$5.4x10^{-5}$	$1.9x10^{-4}$		
∑**PCBs** ng g-1	205.0	766.0	0.583	356.0	850.0	4.291	21.00	74.00	10	17
∑**PCBs** nmol g-1	0.695	2.580		1.217	2.914		0.065	0.227		
$\mathbf{WHO_{PCDD/F+PCD}TEQ_{1998}}$ ng g^{-1}	0.051	0.191		0.122	0.291		0.002	0.008	26	61
$\mathbf{WHO_{PCDD/F+PCB}TEQ_{2005}}$ ng g^{-1}	0.056	0.209		0.129	0.307		0.003	0.011	19	43

[a]Ratio of larval tissue contaminants (dw) indicated in contaminated Ahvionkoski in relation to the reference site Urpalanjoki
[b]Ratio of larval tissue contaminants (dw) indicated in contaminated Kyminlinna in relation to the reference site Urpalanjoki

The most abundant PCDD/F congeners in all sediment samples were OCDF and 1,2,3,4,6,7,8-HpCDF accounting for 36–78 % and 21–40 % of the total sum of PCDD/Fs, respectively (Fig. 2, Table S1). The most abundant PCB congener was PCB-153 in the upper section of R. Kymijoki, but PCB-28/131 in site 4A and sites downstream (Table S2). Also co-planar PCB-77 and PCB-81 peaked in site 4A and 4B, and were higher in sites downstream than in the upper river. Other abundant PCB congeners were 138, 170 and 180 throughout the river. The congener profile in lamprey larvae followed that of the sediments, so that the two most abundant congeners were OCDF and 1,2,3,4,6,7,8-HpCDF both in sediments and in lamprey larvae tissue, except in the reference site R. Urpalanjoki where OCDD appeared in the larvae with slightly higher proportion than OCDF (Fig. 2, Table S3). Also 2,3,7,8-TCDF and 2,3,4,7,8-PeCDF were abundant in larvae in the reference site. The most abundant DL-PCBs both in the sediment and in larvae were PCB-118 and PCB-105 (Fig. 2).

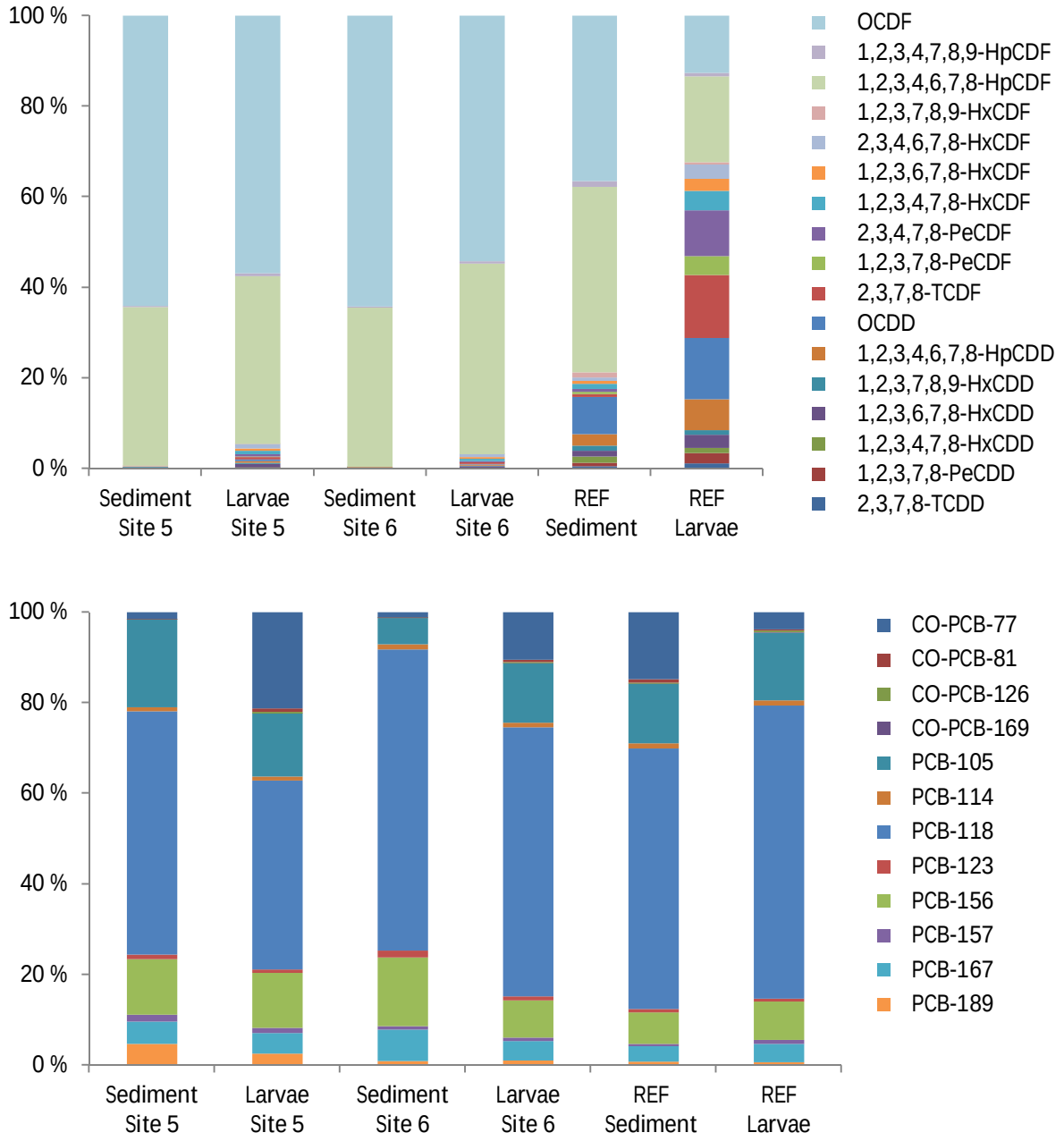

Figure 2. Proportion of 17 PCDD/F congeners in sediment dry weight and lamprey larvae tissue dry weight and proportion of dioxin-like PCBs in contaminated R. Kymijoki site 5 (Ahvionkoski) and site 6 (Kyminlinna), and in the reference site R. Urpalanjoki (REF). Sediment concentrations in the reference site < LOQ except for 1,2,3,4,6,7,8-HpCDD and OCDD.

Chironomid larval behaviour, growth, mortality and deformities

Water temperature during chironomid exposure tests varied from 20.2 to 21.5 C°, oxygen saturation from 66 to 101%, pH from 6.4 to 7.7 and ammonium approximately from 0.25 to 9.0 mg l^{-1}.

Three main behaviour types were detectable in MFB measurements even though they produced slightly overlapping frequencies: (i) ventilation i.e., dorsoventral, undulating, regular movements approximately at the range of 1.0–3.5 Hz, (ii) other activity, mainly slower movements associated with foraging and crawling (0.5–1.0 Hz), and (iii) inactivity (Fig. 3). Typical figure-of-eight swimming style produced signals with approximately similar frequency as ventilation, but signal amplitude was more irregular and variable than in ventilation. In the actual MFB-measurements also

some higher frequency signals were recorded (4.0–8.5 Hz) indicating faster movements in some measurement periods, and these were included in the data analysis, since exposure may not only affect the time spent in ventilating but possibly also the frequency of ventilation.

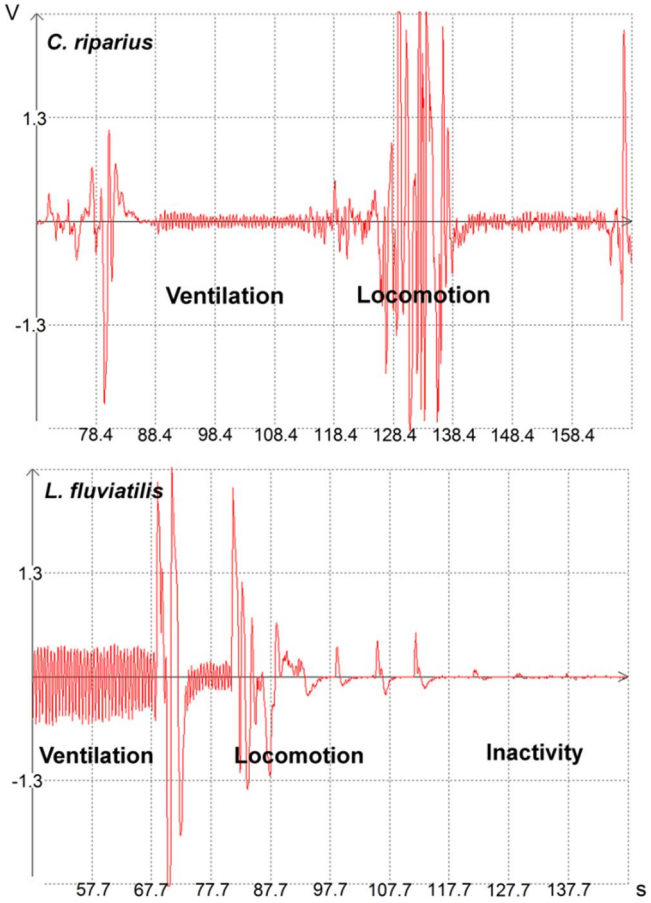

Figure 3. Behaviour of chironomid and lamprey larvae showing regular high frequency and low amplitude signals of ventilation, and high amplitude signals of locomotory behaviour.

Chironomid larvae spent on locomotion and ventilation approximately 40 % and 10 % of the measurement time, respectively (Fig. 4) with no differences among treatments in locomotion ($F = 1.055$, $p = 0.41$) or ventilation ($F = 1.760$, $p = 0.12$), nor interaction effects with the block factor (egg rope the larvae were hatched from). The blocks differed from each other for both locomotion ($F = 6.633$, $p < 0.05$) and ventilation ($F = 4.986$, $p < 0.05$), hence this block factor was relevant to include to reduce error variation in the test. However, time spent in locomotion was lowest in the most contaminated sediment both for the larvae originating from the reference sediment and those grown in that same contaminated sediment (Fig. 4). Larvae were approximately 10 % less active in this most contaminated sediment than in the reference, although statistically significant differences were not found.

Larval wet weight differed among the sediment exposures (Kruskal-Wallis $X^2 = 30.112$ df = 6, $p < 0.001$), being smaller in the contaminated sediments 1A, 2B and 4B than in the reference sediment (Mann-Whitney $p < 0.05$) (Fig. 5). The wet weight showed a tendency to decrease with increasing sediment ∑PCDD/Fs and $TEQ_{PCDD/F+PCB}$ ($r_s = –0.75$, $p = 0.052$), and Hg ($r = –0.65$, $p = 0.113$). The mortality of larvae in the reference sediment was 10.0% on the average, and it varied from 6.7% (4B) to 26.7% (1A) but with no statistically significant differences among the treatments (Kruskal-Wallis $X^2 = 2.342$, df = 6, $p = 0.87$).

Deformity incidence, DI, in the reference sediment was 1.8 %. In the contaminated R. Kymijoki sediments DI was the highest (8.0 %) in site 3A, and otherwise varied from 1.7 % (site 4B) to 4.2%

(2B) with no differences among the exposures (Kruskal-Wallis $X^2 = 2.418$, df = 6, p = 0.88), and no association with chironomid wet weight ($r_s = –0.06$, p = 0.806). The most common deformity type was a missing tooth and some larvae had extra mentum teeth. Köhn gaps were detected only in the reference sediment and in the least contaminated Kymijoki sediment (6B), one in each.

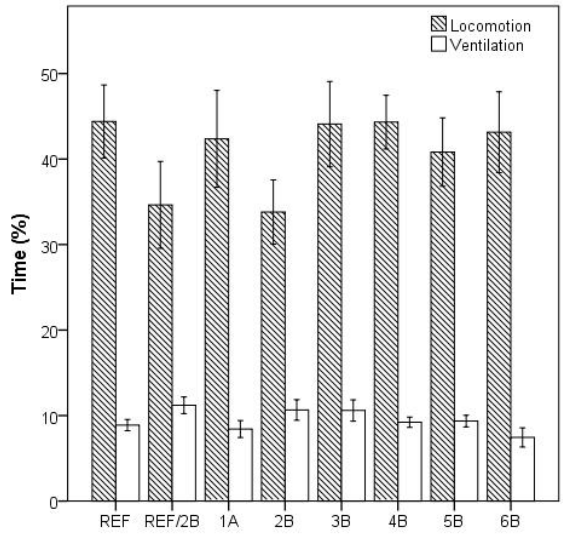

Figure 4. Mean percentage time (± SE) chironomid larvae spent on locomotion or ventilating in the reference sediment from the R. Urpalanjoki (REF), and six contaminated sediments from the R. Kymijoki (sites 1A–6B). REF/2B denote larvae grown in the reference sediment, but their behaviour measured in contaminated 2B sediment.

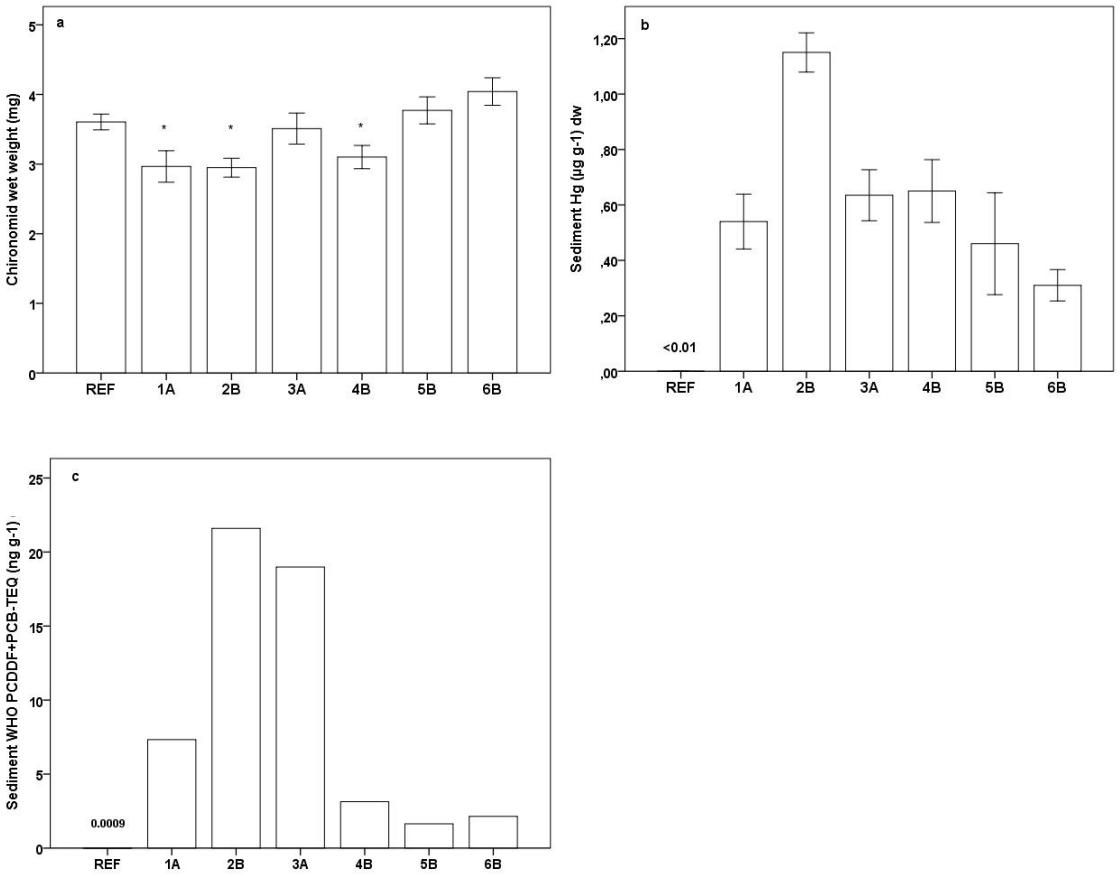

Figure 5. Chironomid larval wet weight (mg) (±SE) in the R. Urpalanjoki reference sediment (REF), and in the six contaminated R. Kymijoki sediments from sites 1A - 6B (a). Asterisks denote the sediments where larval wet weight differed statistically significantly from those grown in the reference sediment. Mean Hg (µg g^{-1} dw) (b) and $WHO_{PCDD/F+PCB}$-TEQ_{2005} (ng g^{-1} dw) of the same sediments (c).

Lamprey larval behaviour, growth and mortality

Water temperature during lamprey exposure tests varied from 18.6 to 19.8 C°, oxygen saturation from 82 to 101 %, pH from 6.3 to 7.4 and ammonium from 3.0 to > 5 mg l^{-1}.

Behaviour of lamprey larvae consisted of three types detectable by MFB: (i) locomotion activity like burrowing and swimming at ≤ 2.0 Hz, (ii) ventilation at approximately 2.0–3.5 Hz and (iii) inactivity (Fig. 3). Some movements associated to swimming and burrowing were also quite fast with high frequency signals apparently overlapping with the ventilation signals. Due to overlapping, also all signals from 0.5 Hz to 8.5 Hz were analysed as a one composite measure of larval activity, although majority of the behavioural signals were < 4.0 Hz. In the reference sediment (7A), larvae spent on locomotion approximately 2.6–5.0 % and on ventilation 0.7–3.6 % of the time (Fig. 6). There was no difference in larval locomotion activity or ventilation in the reference sediment between different exposure days during the entire 28 d exposure (for locomotion Friedman $p = 0.79$, and for ventilation $p = 0.86$, $df = 4$). Temporal differences were not found in any of the contaminated sediments (2A, 4A, or 6B) either (Friedman $p > 0.05$). There was also no difference in either locomotory or ventilatory activity across the sediment treatments within the certain measurement day (Kruskal-Wallis $p > 0.05$).

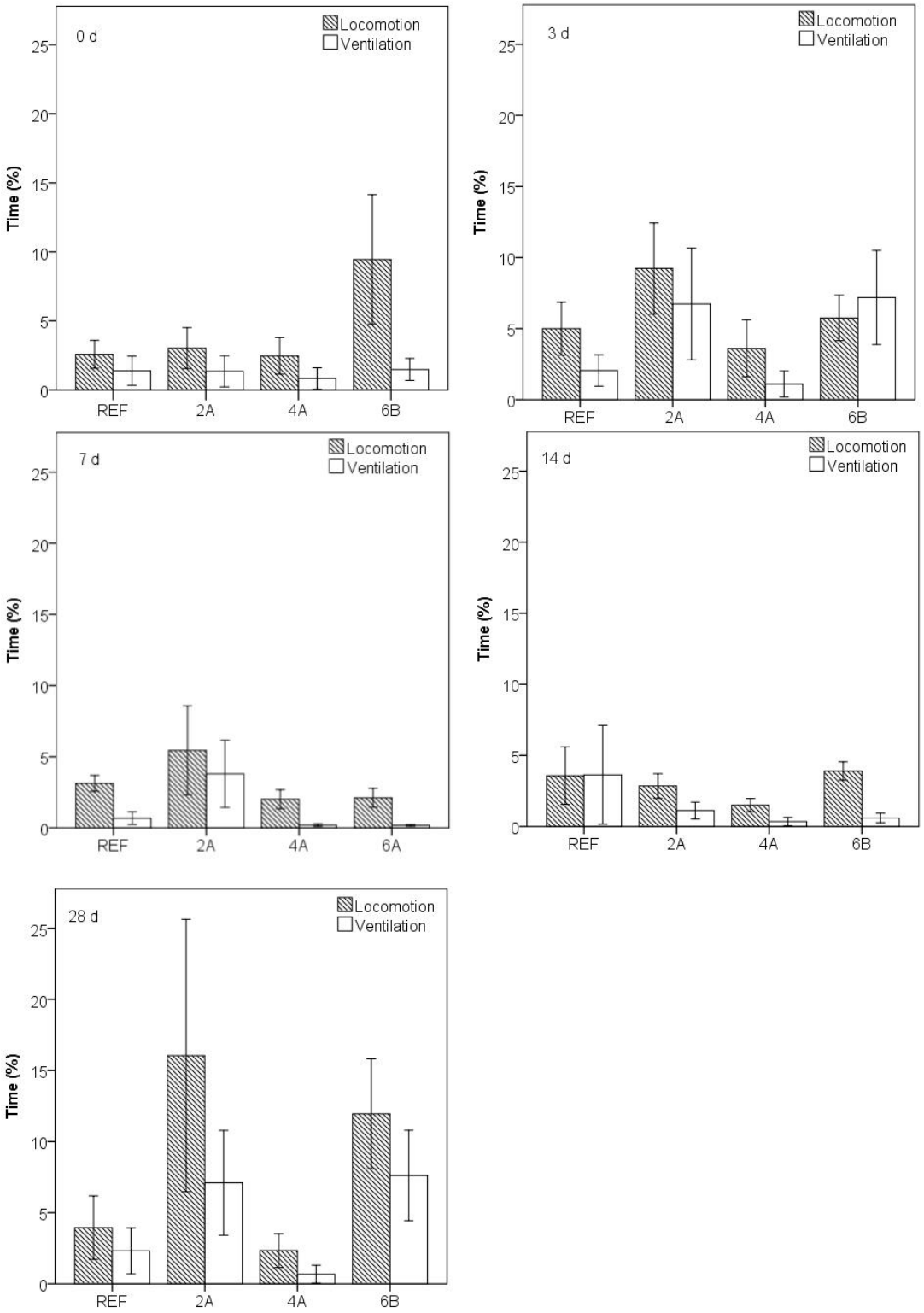

Figure 6. Mean percentage time (± SE) on locomotion and ventilation of *L. fluviatilis* larvae during two hour measurement period (n = 6) at exposure day 0, 3, 7, 14 and 28 in the reference sediment (REF) and in contaminated R. Kymijoki sediments from sites 2A, 4A and 6B.

When all activity detected by MFB in the range of 0.5–8.5 Hz was combined, no differences were found during exposure duration (Friedman all tests $p > 0.05$, $df = 4$ for separate tests) nor across sediment treatments within a certain measurement day (Kruskal-Wallis all tests $p > 0.05$, $df = 3$ for separate tests). Lamprey larvae seemed to be more active in contaminated 2A and 6B sediments (Fig. 6) spending on locomotion approximately 2.8–16.1 % and 0.2–11.9 % of their time, respectively, however with no statistically significant differences to the reference. In these contaminated sediments larval activity range was greater than in the reference sediment: some larvae were really inactive while some were highly active within the same sediment exposure and measurement period. The activity range increased especially in the most polluted sediment (2A) after 28 d exposure. In the sediments 4B the larvae were quite inactive throughout the exposure, and their activity varied between 1.5 to 3.6 %.

All lampreys (n = 24) survived the sediment exposures. Mean larval wet weight in the reference sediment after the exposures was 20.2 (± 4.9) mg, and mean length 20.1 (± 2.1) mm. Larval weight or length did not differ among the sediments (ANOVA $F = 0.981$, $df = 3$, $p = 0.42$, and $F = 0.865$, $df = 3$, $p = 0.48$, respectively).

DISCUSSION

The surface sediments of R. Kymijoki were still heavily contaminated with PCDD/Fs, PCBs and Hg although the loading of dioxins and Hg has been significantly reduced over three decades ago, due to closing of the industrial plants and changes in the industrial processes. In the present study, the most abundant PCDD/F-congeners were OCDF and 1,2,3,4,6,7,8-HpCDF, the latter being impurity of the previous manufacture of wood-preservative (Ky-5), and the most common congener in R. Kymijoki sediments also in the studies of Salo et al. (2008) and Verta et al. (2009).

PCBs in R. Kymijoki sediments are less studied, but PCB28/31 and PCB153 were among those congeners that formed the majority of the sediment load also according to Koistinen et al. (2010). Heterogeneity of river sediments was evident especially in one site (3A and 3B) where two samples taken quite near each other yielded highly differing contaminant and organic matter content.

Lamprey larvae tissue concentrations of PCDD/Fs and PCBs were over hundredfold and tenfold higher, respectively, in the R. Kymijoki than in the reference site. This is a clear sign of contamination, and indicates risk for the river ecosystem, biomagnification in food webs and for humans consuming the river fish. The maximum concentration permitted in edible fish in European Union (EC 2011) is 6.5 pg g^{-1} wet weight as WHO-$_{PCDD/F-PCB}$-TEQ. This was clearly exceeded in R. Kyminjoki lamprey larvae, although Finland has exceptionally permission for marketing and consumption of river lamprey exceeding this level (EC 2011). Isosaari et al. (2006) reported maximum tissue concentrations of 14 pg g^{-1} ww WHO-$TEQ_{PCDD/F+PCB}$ in the adult river lamprey caught in Bothnian Bay, Baltic Sea. In our study larval lampreys' tissue concentration was 13–34 pg g^{-1} ww $WHO_{PCDD/F+PCB}$-TEQ, which is higher than the earlier measured adult lampreys' body burdens by Isosaari et al. (2006) and of 8.6 pg g^{-1} ww by Hallikainen et al. (2011), indicating that uptake of sediment-bound PCDD/Fs and DL-PCBs during lamprey larval stage may considerably account for the species' total body burden of these compounds in an adult phase. Indicator-PCBs in larval lamprey in this study were at the same range than in adult lamprey caught from the Gulf of Finland, near R. Kymijoki river mouth, where indicator-PCBs was 57 ng g^{-1} ww, compared to 24–46 ng g^{-1} ww in the present study. However, lamprey larvae in R. Urpalanjoki had accumulated indicator-PCBs only 3.0 ng g^{-1} ww. Lampreys had accumulated PCDD/F and PCB congeners in relation to the congener concentrations in sediment. This is in agreement with the results of Soimasuo et al. (2004) from Kyminlinna area in the R. Kymijoki. They concluded that generally the same major congeners were abundant both in lamprey larvae and sediment, although larvae and sediment congener profiles were not exactly identical. Biota-sediment accumulation factors, BSAFs, for 1,2,3,4,7,8-HxCDD and 1,2,3,4,6,7,8-HpCDF that were also calculated in Soimasuo et al. (2004) in R. Kymijoki, Kyminlinna area for lamprey larvae were similar compared to our study. BSAFs for PCBs were within the same

range but on average considerably lower at the present study than in Soimasuo et al. (2004), and only BSAF for PCB-52 was similar in both studies. Lamprey larvae in the present study were analyzed with their gut contents, which as a rule may lead to overestimation of the true BSAFs. It has been observed with oligochaetes that if worms are not purged, error on BSAF estimates increases when BSAF is substantially less than one (Mount et al. 1999). To be more precise, without purging the gut contents of the larvae, we can expect BSAFs to be overestimated for congeners, which real BSAF would be below one, because the concentration of the congener under question in the gut sediment is higher than in the larvae. For congeners which real BSAF would be above one, we would expect underestimation of the real BSAFs by the same logic (because the congener concentration in the larvae is higher than in the sediment). Two major congeners, OCDF and 1,2,3,4,6,7,8-HpCDF, at the present study in R. Kymijoki sediments and lamprey larvae were found to be typical to Kymijoki sediments and also to mussel tissues according to Koistinen et al. (2010). Although we did not measure bioaccumulation of PCDD/Fs and PCBs into chironomids, obviously we can expect what earlier studies show that TCDD accumulates also into chironomid larval tissue (West et al. 1996), potentially leading to biomagnification of these compounds in the food webs.

Behavioural responses are often considered more sensitive than other ecotoxicity endpoints, such as mortality. Both of our study species yielded distinct behavioural patterns in the MFB measurements, indicating that their behaviour can be consistently quantified in sediment bioassays. Making the measurements in the natural habitat of sediment dwelling-animals increases ecological relevance and is particularly important for such animals as lamprey larvae, which are highly sensitive to light and express photophobic behaviour (Binder et al. 2013). Our multi-contaminated sediments did not affect mortality of midge and lamprey larvae in laboratory exposures. Consistent differences in larval behaviour were not detected between sediment exposures. However, differences in activity range and time spent in locomotion in the most contaminated sediments compared to reference sediments were apparent for both species, albeit these effects were not clear cut and should be interpreted with caution. To our knowledge, this was the first time these species were used in behavioural MFB measurements in the sediment. However, in order to develop the approach, our understanding of the effects of measurement conditions, intervals, duration etc. need to be improved.

Ventilation behaviour of larval chironomids consists of regular undulatory movements of their body, which brings oxic water into their burrows (Roskosch et al. 2012). Previous studies show that chironomid ventilation is affected by xenobiotics like insecticides and waterborne Hg (Langer-Jaesrich et al. 2010; Azevedo-Pereira and Soares 2010), but also by naturally varying conditions like temperature and oxygen saturation (Roskosch et al. 2012). In the present study, chironomid larval mortality did not differ between the exposures, this result being consistent with West et al. (1996), who did not detect any effects of TCDD on chironomid growth, emergence rate or fecundity. However, we noticed a decreased larval growth in some of the contaminated sediments. The reduced growth observed in the present study was associated with dioxins and Hg. Also Verta et al. (1999) found that *C. riparius* growth was slower in the most contaminated R. Kymijoki sediments. Hg impaired chironomid growth in the study of Azevedo-Pereira and Soares (2010). Chibunda (2009) found that the growth of the chironomid larvae was inhibited at sediment Hg concentration of 2.42 mg kg^{-1} dw, which is two times higher than in the present study at the most contaminated site.

Chironomid mentum deformity incidence (DI) did not differ among the sediments, and was in all exposures within the range suggested to represent a background incidence of 2.8 % to less than 8.0 % (Dickman et al. 1992; Vermeulen 1995; Salmelin et al. 2015) found in minimally disturbed sites in the field. DI seemed not to be related to the chironomid growth, which was reduced in 1A, 2B and 4B sediments, but not in 3A sediment, where the DI was the highest. In contrast, high DI up to 50 % has been observed in field-collected *Chironomus* spp. from R. Kymijoki (Heikki Hämäläinen, unpublished).

Lamprey larvae were quite inactive in our reference sediment. This is in accordance with the study of Mallatt (1982), who stated that lamprey larvae are normally very inactive in sediments, and that increased activity is a sign of stress. Larval lampreys' regular pumping movements of ventilatory muscles in velum serves both in gas exchange and feeding (Hill and Potter 1971; Yap and Bowen 2003). It seemed that the range of behavioural activity among individuals increased in our most contaminated sediments, in which some larvae were inactive, others being highly active at the same day. The weak or inconsistent behavioural response of the lamprey larvae to contamination might be

due to the relatively short 28 d exposure of our study relative to the typically 4–6 years duration of lamprey larval phase (Potter 1980). On the other hand, behavioural changes may appear rapidly after a chemical exposure (Dell'Omo 2002), which is an obvious advantage of using behavioural responses of animals as biological early warning systems of environmental hazards.

The main exposure route of lipophilic substances to benthic animals is supposed to be through intestines from assimilated food. In the present study, the additional feeding by fish food probably decreased the exposure of larvae to the contaminants. Artificial feeding was however considered essential, since the lack of feeding has been shown to increase mortality of larval *C. riparius* (Ristola et al. 1999) and because organic content and thus potential amount of food varied among the sediments. Another potential confounding factor in the present study was the high total ammonia levels of overlying water in some of the exposures despite frequent water renewals. However, this apparently did not affect chironomid or lamprey larvae, since no consistent associations of behavioural, growth or mortality responses to ammonia were detected. According to Monda et al. (1995), *C. riparius* LC_{50} for un-ionized ammonia (NH_3-N) is 9.4 mg L^{-1}, but fish are much more sensitive (Craig & Laming 2004). In the present study, total ammonia (NH_3/NH^{4+}) was at the highest approximately 9.0 mg L^{-1}. Water renewals likely have no effect on contaminant concentrations in the sediments, since these hydrophobic substances are considered to be tightly bound to the sediment organic matter (Hill et al. 1993).

CONCLUSIONS

Surface sediment were sampled in formerly gross polluted River Kymijoki and a reference river and used in whole-sediment bioassays, with chironomid and lamprey larvae as the study species. The surface sediments of R. Kymijoki were still heavily contaminated with PCDD/Fs, PCBs and Hg although pollutant discharges practically ceased several decades ago. Our field investigation showed that lamprey larvae accumulate PCDD/Fs and PCBs in their natural habitats, and congener concentrations correlated strongly between the sediment and larval tissue of lampreys both for PCDD/Fs and PCBs. High pollutant levels in the lamprey larvae indicate risks for biomagnification in the food webs. Chironomid mortality and morphological deformities as well as mortality and growth of lamprey larvae appeared to be insensitive endpoints in laboratory exposure to PCDD/F, PCB and Hg contaminated sediments. Chironomid growth was reduced in the contaminated sediments, and their wet weight was negatively related to sediment $\sum$PCDD/Fs, $WHO_{PCDD/F+PCB}$-TEQ and Hg. The distinct behavioural patterns of chironomid and lamprey larvae, measured via Multispecies Freshwater Biomonitor, indicate their applicability in sediment toxicity bioassays in general.

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Table S1, 1/4. Concentrations (pg g^{-1}) of 17 toxic PCDD/F-congeners in sediment wet weight (ww), dry weight (dw) and proportion of sediment dry matter (%) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCDD/F-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki**			**Kymijoki**			**Kymijoki**			**Kymijoki**		
	Kuusansaari (1A)			**Kuusansaari (1B)**			**Keltti (2A)**			**Keltti (2B)**		
Sampling date	24.7.2012			24.7.2012			24.7.2012			24.7.2012		
Dry matter (%)	20.4			20.2			23.6			28.9		
PCDD pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
2,3,7,8-TCDD	2.2	11	(<0.01)	2.0	10.0	(<0.01)	1.0	4.3	(<0.01)	1.2	4.3	(<0.01)
1,2,3,7,8-PeCDD	4.0	20	(<0.01)	2.3	11	(<0.01)	2.3	9.9	(<0.01)	3.2	11	(<0.01)
1,2,3,4,7,8-HxCDD	3.1	15	(<0.01)	0.73	3.6	(<0.01)	2.2	9.5	(<0.01)	2.0	6.9	(<0.01)
1,2,3,6,7,8-HxCDD	270	1 322	(0.08)	95	471	(0.05)	347	1 468	(0.02)	437	1 511	(0.02)
1,2,3,7,8,9-HxCDD	67	330	(0.02)	34	169	(0.02)	98	417	(<0.01)	127	438	(0.01)
1,2,3,4,6,7,8-HpCDD	673	3 296	(0.21)	239	1 185	(0.13)	1 247	5 275	(0.06)	1 537	5 311	(0.07)
OCDD	559	2 736	(0.17)	333	1 651	(0.18)	2 382	10 077	(0.12)	2 399	8 286	(0.11)
PCDF pg g-1												
2,3,7,8-TCDF	34	165	(0.01)	29	146	(0.02)	16	68	(<0.01)	21	72	(<0.01)
1,2,3,7,8-PeCDF	42	207	(0.01)	10	52	(0.01)	41	176	(<0.01)	61	212	(<0.01)
2,3,4,7,8-PeCDF	155	757	(0.05)	28	136	(0.01)	165	699	(0.01)	221	763	(0.01)
1,2,3,4,7,8-HxCDF	746	3 654	(0.23)	204	1 009	(0.11)	2 260	9 561	(0.11)	3 146	10 868	(0.14)
1,2,3,6,7,8-HxCDF	350	1 712	(0.11)	93	461	(0.05)	915	3 870	(0.05)	1 088	3 757	(0.05)
2,3,4,6,7,8-HxCDF	598	2 929	(0.18)	147	730	(0.08)	1 550	6 560	(0.08)	1 713	5 916	(0.08)
1,2,3,7,8,9-HxCDF	16	78	(<0.01)	6.4	32	(<0.01)	52	222	(<0.01)	63	219	(<0.01)
1,2,3,4,6,7,8-HpCDF	115 376	564 960	(35.6)	65 643	325 059	(35.4)	415 532	1 758 208	(20.9)	487 049	1 682 471	(21.4)
1,2,3,4,7,8,9-HpCDF	1 290	6 316	(0.40)	500	2 475	(0.27)	6 993	29 587	(0.35)	9 958	34 398	(0.44)
OCDF	203 744	997 666	(62.9)	118 226	585 448	(63.7)	1 559 002	6 596 477	(78.3)	1 770 043	6 114 462	(77.7)
∑PCDD/Fs pg g-1												
sum (lower bound)	323 929	1 586 174		185 594	919 047		1 990 607	8 422 689		2 277 869	7 868 705	
sum (upper bound)	323 929	1 586 174		185 594	919 047		1 990 607	8 422 689		2 277 869	7 868 705	
WHO1998 TEQ (lower bound)	1 488	7 286		755	3 740		5 006	21 182		5 940	20 520	
WHO1998 TEQ (upper bound)	1 488	7 286		755	3 740		5 006	21 182		5 940	20 520	
WHO2005 TEQ (lower bound)	**1 497**	**7 330**		**773**	**3 829**		**5 284**	**22 360**		**6 249**	**21 588**	
WHO2005 TEQ (upper bound)	**1 497**	**7 330**		**773**	**3 829**		**5 284**	**22 360**		**6 249**	**21 588**	
WHO1998 fishTEQ (lower bound)	**1 451**	**7 106**		**740**	**3 666**		**4 955**	**20 965**		**5 875**	**20 296**	
WHO1998 fishTEQ (upper bound)	**1 451**	**7 106**		**740**	**3 666**		**4 955**	**20 965**		**5 875**	**20 296**	

Table S1, 2/4. Concentrations (pg g^{-1}) of 17 toxic PCDD/F-congeners in sediment wet weight (ww), dry weight (dw) and proportion of sediment dry matter (%) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCDD/F-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki**			**Kymijoki**			**Kymijoki**			**Kymijoki**		
	Lopotti (3A)			**Lopotti (3B)**			**Koskenalusjärvi (4A)**			**Koskenalusjärvi (4B)**		
Sampling date	27.7.2012			27.7.2012			27.7.2012			27.7.2012		
Dry matter (%)	21.4			44.5			35.1			43.8		
PCDD pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
2,3,7,8-TCDD	1.2	5.7	(<0.01)	<0.93	<2.1	(<0.01)	<1.4	<3.9	(<0.01)	1.4	3.2	(<0.01)
1,2,3,7,8-PeCDD	1.4	6.5	(<0.01)	<0.85	<1.9	(<0.01)	8.4	24	(<0.01)	2.7	6.2	(<0.01)
1,2,3,4,7,8-HxCDD	0.77	3.6	(<0.01)	<1.6	<3.5	(<0.01)	4.2	12	(<0.01)	<1.8	<4.1	(<0.01)
1,2,3,6,7,8-HxCDD	128	595	(0.01)	81	181	(0.02)	175	499	(0.03)	72	165	(0.02)
1,2,3,7,8,9-HxCDD	37	174	(<0.01)	24	53	(<0.01)	59	168	(0.01)	20	46	(0.01)
1,2,3,4,6,7,8-HpCDD	438	2 044	(0.04)	293	658	(0.06)	372	1 060	(0.07)	200	456	(0.06)
OCDD	839	3 911	(0.07)	422	949	(0.08)	433	1 234	(0.08)	469	1 071	(0.14)
PCDF pg g-1												
2,3,7,8-TCDF	15	68	(<0.01)	6.8	15	(<0.01)	7.8	22	(<0.01)	9.0	21	(<0.01)
1,2,3,7,8-PeCDF	20	94	(<0.01)	9.6	22	(<0.01)	9.3	26	(<0.01)	7.4	17	(<0.01)
2,3,4,7,8-PeCDF	68	317	(0.01)	40	90	(0.01)	49	138	(0.01)	36	83	(0.01)
1,2,3,4,7,8-HxCDF	963	4 489	(0.09)	706	1 587	(0.13)	484	1 379	(0.09)	369	842	(0.11)
1,2,3,6,7,8-HxCDF	368	1 714	(0.03)	271	609	(0.05)	311	888	(0.06)	244	556	(0.07)
2,3,4,6,7,8-HxCDF	601	2 803	(0.05)	451	1 014	(0.09)	473	1 349	(0.09)	381	869	(0.12)
1,2,3,7,8,9-HxCDF	17	80	(<0.01)	11	24	(<0.01)	7.9	23	(<0.01)	8.5	19	(<0.01)
1,2,3,4,6,7,8-HpCDF	354 474	1 652 595	(31.3)	181 511	408 295	(34.6)	180 601	514 944	(34.2)	117 258	267 653	(36.0)
1,2,3,4,7,8,9-HpCDF	5 560	25 919	(0.49)	1 809	4 070	(0.34)	1 439	4 104	(0.27)	1 024	2 338	(0.31)
OCDF	767 845	3 579 771	(67.9)	339 775	764 295	(64.7)	344 336	981 798	(65.1)	205 680	469 486	(63.1)
∑PCDD/Fs pg g-1												
sum (lower bound)	1 131 376	5 274 591		525 408	1 181 862		528 770	1 507 669		325 782	743 632	
sum (upper bound)	1 131 376	5 274 591		525 412	1 181 870		528 771	1 507 673		325 784	743 636	
WHO1998 TEQ (lower bound)	3 932	18 332		2 046	4 601		2 044	5 828		1 338	3 055	
WHO1998 TEQ (upper bound)	3 932	18 332		2 048	4 606		2 045	5 832		1 339	3 055	
WHO2005 TEQ (lower bound)	**4 072**	**18 984**		**2 105**	**4 736**		**2 103**	**5 996**		**1 372**	**3 132**	
WHO2005 TEQ (upper bound)	**4 072**	**18 984**		**2 107**	**4 740**		**2 104**	**6 000**		**1 372**	**3 133**	
WHO1998 fishTEQ (lower bound)	**3 913**	**18 242**		**2 033**	**4 573**		**2 021**	**5 762**		**1328**	**3 031**	
WHO1998 fishTEQ (upper bound)	**3 913**	**18 242**		**2 036**	**4 579**		**2 022**	**5 766**		**1329**	**3 033**	

Table S1, 3/4. Concentrations (pg g^{-1}) of 17 toxic PCDD/F-congeners in sediment wet weight (ww), dry weight (dw) and proportion of sediment dry matter (%) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCDD/F-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki**			**Kymijoki**			**Kymijoki**			**Kymijoki**		
	Ahvionkoski (5A)			**Ahvionkoski (5B)**			**Kyminlinna (6A)**			**Kyminlinna (6B)**		
Sampling date	26.7.2012			26.7.2012			26.7.2012			26.7.2012		
Dry matter (%)	20.2			15.7			32.7			33.9		
PCDD pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
2,3,7,8-TCDD	<0.35	<1.7	(<0.01)	<0.21	<1.3	(<0.01)	0.58	1.8	(<0.01)	0.64	1.9	(<0.01)
1,2,3,7,8-PeCDD	0.84	4.1	(<0.01)	0.59	3.8	(<0.01)	1.2	3.6	(<0.01)	1.1	3.3	(<0.01)
1,2,3,4,7,8-HxCDD	<0.61	<3.0	(<0.01)	<0.35	<2.2	(<0.01)	0.56	1.7	(<0.01)	0.51	1.5	(<0.01)
1,2,3,6,7,8-HxCDD	22	106	(0.02)	17	110	(0.03)	29	88	(0.02)	31	93	(0.02)
1,2,3,7,8,9-HxCDD	6.9	34	(0.01)	6.1	39	(0.01)	11	34	(0.01)	10	31	(0.01)
1,2,3,4,6,7,8-HpCDD	57	283	(0.07)	49	311	(0.08)	75	229	(0.06)	83	245	(0.05)
OCDD	94	465	(0.11)	109	693	(0.17)	121	370	(0.10)	153	450	(0.08)
PCDF pg g-1												
2,3,7,8-TCDF	3.3	16	(<0.01)	2.6	17	(<0.01)	5.6	17	(<0.01)	6.5	19	(<0.01)
1,2,3,7,8-PeCDF	2.1	10	(<0.01)	1.8	11	(<0.01)	2.5	7.7	(<0.01)	3.5	10	(<0.01)
2,3,4,7,8-PeCDF	9.5	47	(0.01)	7.3	47	(0.01)	9.9	30	(0.01)	15	44	(0.01)
1,2,3,4,7,8-HxCDF	102	502	(0.12)	78	497	(0.13)	93	285	(0.08)	164	482	(0.09)
1,2,3,6,7,8-HxCDF	57	284	(0.07)	39	245	(0.06)	45	138	(0.04)	74	219	(0.04)
2,3,4,6,7,8-HxCDF	90	445	(0.10)	65	415	(0.10)	84	257	(0.07)	137	403	(0.07)
1,2,3,7,8,9-HxCDF	<2.5	<12	(<0.01)	<1.4	<9.2	(<0.01)	0.77	2.4	(<0.01)	1.4	4.2	(<0.01)
1,2,3,4,6,7,8-HpCDF	30 172	149 228	(35.1)	21 954	139 569	(35.2)	43 022	131 398	(35.4)	63 708	187 891	(34.9)
1,2,3,4,7,8,9-HpCDF	241	1 192	(0.28)	154	982	(0.25)	450	1 374	(0.37)	584	1 721	(0.32)
OCDF	55 194	272 987	(64.1)	39 877	253 506	(63.9)	77 433	236 492	(63.8)	117 770	347 333	(64.5)
∑PCDD/Fs pg g-1												
sum (lower bound)	86 051	425 604		62 361	396 445		121 384	370 727		182 742	538 951	
sum (upper bound)	86 054	425 621		62 363	396 458		121 384	370 727		182 742	538 951	
WHO1998 TEQ (lower bound)	344	1 701		251	1 594		477	1 457		707	2 086	
WHO1998 TEQ (upper bound)	345	1 705		251	1 596		477	1 457		707	2 086	
WHO2005 TEQ (lower bound)	**353**	**1 747**		**257**	**1 635**		**490**	**1 498**		**728**	**2 147**	
WHO2005 TEQ (upper bound)	**354**	**1 750**		**258**	**1 638**		**490**	**1 498**		**728**	**2 147**	
WHO1998 fishTEQ (lower bound)	**341**	**1 685**		**248**	**1 577**		**473**	**1 444**		**703**	**2 073**	
WHO1998 fishTEQ (upper bound)	**342**	**1 690**		**249**	**1 580**		**473**	**1 444**		**703**	**2 073**	

Table S1, 4/4. Concentrations (pg g^{-1}) of 17 toxic PCDD/F-congeners in sediment wet weight (ww), dry weight (dw) and proportion of sediment dry matter (%) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCDD/F-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Urpalanjoki**			**Urpalanjoki**		
	Urpalanjoki (7A)			**Urpalanjoki (7B)**		
Sampling date	25.7.2012			25.7.2012		
Dry matter (%)	56.6			67.4		
PCDD pg g-1	ww	dw	(%)	ww	dw	(%)
2,3,7,8-TCDD	<0.062	<0.11	(0.41)	<0.090	<0.13	(0.48)
1,2,3,7,8-PeCDD	<0.12	<0.21	(0.78)	<0.17	<0.25	(0.93)
1,2,3,4,7,8-HxCDD	<0.19	<0.33	(1.22)	<0.25	<0.36	(1.33)
1,2,3,6,7,8-HxCDD	<0.18	<0.32	(1.19)	<0.24	<0.36	(1.33)
1,2,3,7,8,9-HxCDD	<0.17	<0.30	(1.11)	<0.22	<0.33	(1.22)
1,2,3,4,6,7,8-HpCDD	0.45	0.80	(2.96)	0.39	0.58	(2.15)
OCDD	1.3	2.3	(8.52)	<1.4	<2.1	(7.78)
PCDF pg g-1						
2,3,7,8-TCDF	<0.082	<0.14	(0.52)	<0.11	<0.17	(0.63)
1,2,3,7,8-PeCDF	<0.093	<0.17	(0.63)	<0.10	<0.15	(0.56)
2,3,4,7,8-PeCDF	<0.086	<0.15	(0.56)	<0.093	<0.14	(0.52)
1,2,3,4,7,8-HxCDF	<0.19	<0.33	(1.22)	<0.23	<0.33	(1.22)
1,2,3,6,7,8-HxCDF	<0.083	<0.15	(0.56)	<0.12	<0.18	(0.67)
2,3,4,6,7,8-HxCDF	<0.10	<0.18	(0.67)	<0.15	<0.23	(0.85)
1,2,3,7,8,9-HxCDF	<0.15	<0.27	(1.00)	<0.23	<0.34	(1.26)
1,2,3,4,6,7,8-HpCDF	<6.4	<11	(40.7)	<7.6	<11	(40.7)
1,2,3,4,7,8,9-HpCDF	<0.17	<0.31	(1.15)	<0.23	<0.34	(1.26)
OCDF	<5.6	<9.8	(36.3)	<6.6	<9.8	(36.3)
∑PCDD/Fs pg g-1						
sum (lower bound)	1.8	3.1		0.39	0.58	
sum (upper bound)	15	27		18	27	
WHO1998 TEQ (lower bound)	0.005	0.008		0.004	0.006	
WHO1998 TEQ (upper bound)	0.41	0.73		0.55	0.82	
WHO2005 TEQ (lower bound)	**0.005**	**0.009**		**0.004**	**0.006**	
WHO2005 TEQ (upper bound)	**0.40**	**0.70**		**0.53**	**0.79**	
WHO1998 fishTEQ (lower bound)	**0.001**	**0.001**		**0.0004**	**0.001**	
WHO1998 fishTEQ (upper bound)	**0.451**	**0.790**		**0.599**	**0.876**	

Table S2, 1/4. PCBs pg g^{-1} (coplanar PCBs and TEQ-values) or ng g^{-1} (all other values) in sediment wet weight (ww) and dry weight (dw) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCB-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki Kuusansaari (1A)**			**Kymijoki Kuusansaari (1B)**			**Kymijoki Keltti (2A)**			**Kymijoki Keltti (2B)**		
Sampling time	24.7.2012			24.7.2012			24.7.2012			24.7.2012		
Coplanar PCBs pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
CO-PCB-77	<15	<73	(0.01)	<15	<77	(0.01)	<15	<62	(0.01)	<18	<62	(<0.01)
CO-PCB-81	<0.26	<1.3	(<0.01)	0.82	4.1	(<0.01)	0.31	1.3	(<0.01)	0.41	1.4	(<0.01)
CO-PCB-126	<1.2	<6.1	(<0.01)	<1.3	<6.4	(<0.01)	<1.2	<5.2	(<0.01)	2.2	7.6	(<0.01)
CO-PCB-169	1.3	6.6	(<0.01)	1.5	7.3	(<0.01)	1.8	7.5	(<0.01)	3.0	10	(<0.01)
Other PCBs ng g-1												
PCB-18	0.67	3.3	(0.52)	1.0	5.1	(0.48)	0.53	2.2	(0.19)	0.66	2.3	(0.09)
PCB-28/31	3.9	19	(3.00)	6.2	31	(2.94)	2.8	12	(1.01)	3.5	12	(0.47)
PCB-33	0.76	3.7	(0.58)	1.3	6.5	(0.62)	0.56	2.4	(0.20)	0.68	2.4	(0.09)
PCB-47	0.80	3.9	(0.62)	2.0	10	(0.95)	0.74	3.1	(0.26)	1.2	4.2	(0.16)
PCB-49	1.8	8.7	(1.37)	4.3	21	(1.99)	1.4	5.9	(0.50)	2.0	7.0	(0.27)
PCB-51	0.042	0.20	(0.03)	0.072	0.36	(0.03)	0.034	0.15	(0.01)	0.044	0.15	(0.01)
PCB-52	2.3	11	(1.74)	3.5	17	(1.61)	4.1	17	(1.44)	6.2	22	(0.86)
PCB-60	0.22	1.1	(0.17)	0.36	1.8	(0.17)	0.25	1.1	(0.09)	0.37	1.3	(0.05)
PCB-66	2.0	9.9	(1.56)	3.7	18	(1.71)	1.9	7.9	(0.67)	2.9	10.0	(0.39)
PCB-74	0.64	3.1	(0.49)	1.2	5.7	(0.54)	0.61	2.6	(0.22)	0.95	3.3	(0.13)
PCB-99	2.7	13	(2.05)	9.1	45	(4.27)	3.8	16	(1.35)	11	39	(1.52)
PCB-101	9.2	45	(7.11)	16	81	(7.68)	24	102	(8.61)	39	135	(5.27)
PCB-105	0.77	3.8	(0.60)	1.3	6.6	(0.63)	1.1	4.6	(0.39)	2.6	8.9	(0.35)
PCB-110	6.8	33	(5.21)	11	57	(5.40)	15	64	(5.41)	22	77	(3.00)
PCB-114	<0.030	<0.15	(0.02)	0.043	0.21	(0.02)	<0.022	<0.092	(0.01)	0.14	0.48	(0.02)
PCB-118	4.2	21	(3.32)	7.8	39	(3.70)	9.3	40	(3.38)	21	74	(2.89)
PCB-122	<0.031	<0.15	(0.02)	<0.023	<0.12	(0.01)	<0.023	<0.098	(0.01)	0.061	0.21	(0.01)
PCB-123	<0.030	<0.15	(0.02)	<0.022	<0.11	(0.01)	0.081	0.34	(0.03)	0.29	0.99	(0.04)
PCB-128	2.1	10	(1.58)	3.5	17	(1.61)	4.5	19	(1.60)	12	43	(1.68)
PCB-138	15	75	(11.9)	26	127	(12.04)	38	161	(13.6)	101	350	(13.7)
PCB-141	3.5	17	(2.69)	5.1	25	(2.37)	13	54	(4.56)	30	103	(4.02)
PCB-153	22	107	(16.9)	38	188	(17.8)	52	220	(18.9)	141	487	(19.0)
PCB-156	1.2	6.0	(0.95)	1.7	8.3	(0.79)	3.9	16	(1.35)	15	51	(1.99)
PCB-157	0.13	0.65	(0.10)	0.16	0.80	(0.08)	0.31	1.3	(0.11)	1.4	5.0	(0.20)
PCB-167	0.57	2.8	(0.44)	0.86	4.3	(0.41)	1.7	7.1	(0.60)	7.1	24	(0.94)
PCB-170	9.6	47	(7.42)	13	67	(6.35)	24	103	(8.70)	84	290	(11.3)
PCB-180	18	91	(14.4)	26	129	(12.2)	42	178	(15.0)	141	487	(19.0)
PCB-183	3.3	16	(2.53)	6.1	30	(2.84)	8.7	37	(3.13)	24	84	(3.28)
PCB-187	6.7	33	(5.21)	12	60	(5.69)	15	65	(5.49)	42	146	(5.70)
PCB-189	0.28	1.4	(0.22)	0.38	1.9	(0.18)	0.73	3.1	(0.26)	3.5	12	(0.47)
PCB-194	3.0	15	(2.37)	4.0	20	(1.90)	4.9	21	(1.77)	17	59	(2.30)
PCB-206	2.3	11	(1.74)	1.8	9.0	(0.85)	1.7	7.2	(0.61)	2.8	9.6	(0.37)
PCB-209	4.0	20	(3.16)	4.4	22	(2.09)	2.3	9.9	(0.84)	3.2	11	(0.43)
∑PCBs ng g-1												
sum (lower bound)	129	632		213	1 054		280	1 184		742	2 563	
sum (upper bound)	129	633		213	1 055		280	1 184		742	2 563	
∑Indicator-PCBs ng g-1												
Indicator-PCBs (lower bound)	71	348		116	573		163	690		432	1 493	
Indicator-PCBs (upper bound)	71	348		116	573		163	690		432	1 493	
∑PCB-TEQ pg g-1												
WHO1998 TEQ (lower bound)	1.2	6.0		1.9	9.5		3.2	14		11	39	
WHO1998 TEQ (upper bound)	1.4	6.7		2.1	10		3.4	14		11	39	
WHO2005 TEQ (lower bound)	**0.26**	**1.3**		**0.41**	**2.0**		**0.57**	**2.4**		**1.8**	**6.4**	
WHO2005 TEQ (upper bound)	**0.38**	**1.9**		**0.55**	**2.7**		**0.69**	**2.9**		**1.8**	**6.4**	
WHO1998 fishTEQ (lower bound)	**0.04**	**0.18**		**0.06**	**0.34**		**0.09**	**0.36**		**0.27**	**0.92**	
WHO1998 fishTEQ (upper bound)	**0.04**	**0.22**		**0.07**	**0.35**		**0.09**	**0.40**		**0.27**	**0.93**	

Table S2, 2/4. PCBs pg g^{-1} (coplanar PCBs and TEQ-values) or ng g^{-1} (all other values) in sediment wet weight (ww) and dry weight (dw) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCB-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki**			**Kymijoki**			**Kymijoki**			**Kymijoki**		
	Lopotti (3A)			**Lopotti (3B)**			**Koskenalusjärvi (4A)**			**Koskenalusjärvi (4B)**		
Sampling time	27.7.2012			27.7.2012			27.7.2012			27.7.2012		
Coplanar PCBs pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
CO-PCB-77	<16	<73	(0.02)	<28	<63	(0.01)	405	1 154	(0.09)	1 430	3 263	(0.11)
CO-PCB-81	<0.21	<0.97	(<0.01)	<0.31	<0.71	(<0.01)	15	44	(<0.01)	63	144	(<0.01)
CO-PCB-126	<1.3	<6.1	(<0.01)	6.5	15	(<0.01)	2.2	6.3	(<0.01)	7.4	17	(<0.01)
CO-PCB-169	0.85	4.0	(<0.01)	1.9	4.2	(<0.01)	2.4	6.7	(<0.01)	1.5	3.5	(<0.01)
Other PCBs ng g-1												
PCB-18	0.42	2.0	(0.45)	0.32	0.71	(0.07)	29	83	(6.56)	99	226	(7.61)
PCB-28/31	2.3	10	(2.26)	1.5	3.3	(0.31)	123	352	(27.8)	420	959	(32.3)
PCB-33	0.51	2.4	(0.54)	0.30	0.67	(0.06)	37	106	(8.37)	135	308	(10.4)
PCB-47	0.49	2.3	(0.52)	0.47	1.1	(0.10)	15	41	(3.24)	52	120	(4.04)
PCB-49	0.78	3.6	(0.81)	0.49	1.1	(0.10)	20	57	(4.50)	70	160	(5.39)
PCB-51	0.035	0.16	(0.04)	0.034	0.076	(0.01)	1.2	3.5	(0.28)	4.6	11	(0.37)
PCB-52	1.9	8.7	(1.96)	1.9	4.4	(0.41)	26	73	(5.77)	85	195	(6.56)
PCB-60	0.19	0.90	(0.20)	0.31	0.69	(0.06)	1.8	5.2	(0.41)	6.9	16	(0.54)
PCB-66	1.2	5.8	(1.31)	0.95	2.1	(0.20)	35	98	(7.74)	120	274	(9.22)
PCB-74	0.49	2.3	(0.52)	0.49	1.1	(0.10)	15	43	(3.40)	54	124	(4.17)
PCB-99	1.5	7.0	(1.58)	0.77	1.7	(0.16)	4.7	13	(1.03)	14	33	(1.11)
PCB-101	7.5	35	(7.90)	24	54	(5.03)	14	41	(3.24)	31	70	(2.36)
PCB-105	0.56	2.6	(0.59)	0.63	1.4	(0.13)	1.3	3.7	(0.29)	3.2	7.4	(0.25)
PCB-110	4.8	22	(4.97)	12	27	(2.51)	12	33	(2.61)	25	58	(1.95)
PCB-114	0.025	0.11	(0.02)	<0.035	<0.079	(0.01)	0.18	0.51	(0.04)	0.47	1.1	(0.04)
PCB-118	3.4	16	(3.61)	6.5	15	(1.40)	10	29	(2.29)	24	56	(1.88)
PCB-122	<0.022	<0.10	(0.02)	<0.037	<0.084	(0.01)	0.11	0.31	(0.02)	0.35	0.79	(0.03)
PCB-123	0.034	0.16	(0.04)	0.067	0.15	(0.01)	0.10	0.29	(0.02)	0.55	1.2	(0.04)
PCB-128	1.4	6.6	(1.49)	5.7	13	(1.21)	2.0	5.7	(0.45)	2.7	6.1	(0.21)
PCB-138	12	57	(12.9)	69	155	(14.4)	18	51	(4.03)	27	61	(2.05)
PCB-141	3.5	16	(3.61)	17	37	(3.45)	5.5	16	(1.26)	8.6	20	(0.67)
PCB-153	16	75	(16.9)	102	230	(21.4)	23	65	(5.13)	37	84	(2.83)
PCB-156	1.1	5.3	(1.20)	5.1	11	(1.02)	1.6	4.6	(0.36)	2.6	6.0	(0.20)
PCB-157	0.096	0.45	(0.10)	0.24	0.54	(0.05)	0.14	0.41	(0.03)	0.27	0.63	(0.02)
PCB-167	0.50	2.3	(0.52)	1.9	4.3	(0.40)	0.80	2.3	(0.18)	1.3	2.9	(0.10)
PCB-170	7.5	35	(7.90)	55	123	(11.5)	9.9	28	(2.21)	16	36	(1.21)
PCB-180	13	62	(14.0)	103	232	(21.6)	18	52	(4.11)	29	66	(2.22)
PCB-183	2.7	13	(2.93)	21	47	(4.38)	3.6	10	(0.79)	6.1	14	(0.47)
PCB-187	5.4	25	(5.64)	27	61	(5.68)	6.7	19	(1.50)	11	26	(0.88)
PCB-189	0.22	1.0	(0.23)	1.2	2.7	(0.25)	0.32	0.91	(0.07)	0.50	1.2	(0.04)
PCB-194	1.7	8.1	(1.83)	16	37	(3.45)	2.6	7.4	(0.58)	4.7	11	(0.37)
PCB-206	0.94	4.4	(0.99)	2.5	5.5	(0.51)	2.3	6.5	(0.51)	2.6	5.9	(0.20)
PCB-209	1.9	8.8	(1.99)	0.35	0.79	(0.07)	4.3	12	(0.95)	4.3	9.8	(0.33)
∑PCBs ng g-1												
sum (lower bound)	95	442		477	1 074		444	1 266		1 302	2 971	
sum (upper bound)	95	443		478	1 074		444	1 266		1 302	2 971	
∑Indicator-PCBs ng g-1												
Indicator-PCBs (lower bound)	53	249		302	679		222	634		628	1 434	
Indicator-PCBs (upper bound)	53	249		302	679		222	634		628	1 434	
∑PCB-TEQ pg g-1												
WHO1998 TEQ (lower bound)	1.1	4.9		4.2	9.4		2.5	7.0		5.5	12	
WHO1998 TEQ (upper bound)	1.2	5.5		4.2	9.5		2.5	7.0		5.5	12	
WHO2005 TEQ (lower bound)	**0.20**	**0.95**		**1.2**	**2.6**		**0.78**	**2.2**		**1.9**	**4.4**	
WHO2005 TEQ (upper bound)	**0.34**	**1.6**		**1.2**	**2.7**		**0.78**	**2.2**		**1.9**	**4.4**	
WHO1998 fishTEQ (lower bound)	**0.03**	**0.14**		**0.11**	**0.25**		**0.13**	**0.38**		**0.38**	**0.87**	
WHO1998 fishTEQ (upper bound)	**0.04**	**0.18**		**0.11**	**0.26**		**0.13**	**0.38**		**0.38**	**0.87**	

Table S2, 3/4. PCBs pg g^{-1} (coplanar PCBs and TEQ-values) or ng g^{-1} (all other values) in sediment wet weight (ww) and dry weight (dw) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCB-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki Ahvionkoski (5A)**			**Kymijoki Ahvionkoski (5B)**			**Kymijoki Kyminlinna (6A)**			**Kymijoki Kyminlinna (6B)**		
Sampling time	26.7.2012			26.7.2012			26.7.2012			26.7.2012		
Coplanar PCBs pg g-1	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)	ww	dw	(%)
CO-PCB-77	91	450	(0.06)	68	430	(0.07)	181	552	(1.45)	180	530	(0.91)
CO-PCB-81	4.3	21	(<0.01)	3.2	20	(<0.01)	6.6	20	(0.05)	6.7	20	(0.03)
CO-PCB-126	<1.3	<6.5	(<0.01)	<0.82	<5.2	(<0.01)	2.4	7.2	(0.02)	2.4	7.2	(0.01)
CO-PCB-169	0.37	1.8	(<0.01)	0.20	1.3	(<0.01)	0.48	1.5	(<0.01)	0.49	1.4	(<0.01)
Other PCBs ng g-1												
PCB-18	5.0	25	(3.16)	6.6	42	(7.02)	0.51	1.6	(4.21)	0.79	2.3	(3.97)
PCB-28/31	27	134	(17.0)	28	178	(29.8)	3.3	10	(26.3)	4.7	14	(24.1)
PCB-33	6.6	33	(4.18)	8.1	51	(8.53)	0.79	2.4	(6.32)	1.3	3.8	(6.55)
PCB-47	2.9	14	(1.77)	1.8	11	(1.84)	0.25	0.75	(1.97)	0.22	0.65	(1.12)
PCB-49	4.0	20	(2.53)	2.7	17	(2.84)	0.42	1.3	(3.42)	0.33	0.97	(1.67)
PCB-51	0.18	0.91	(0.12)	0.19	1.2	(0.20)	0.013	0.040	(0.11)	0.017	0.050	(0.09)
PCB-52	5.3	26	(3.29)	3.9	25	(4.18)	0.50	1.5	(3.95)	0.44	1.3	(2.24)
PCB-60	1.4	6.9	(0.87)	1.9	12	(2.01)	0.33	1.0	(2.63)	0.29	0.85	(1.47)
PCB-66	8.2	41	(5.19)	7.9	50	(8.63)	1.3	4.1	(10.8)	1.4	4.0	(6.90)
PCB-74	3.8	19	(2.41)	2.8	18	(3.01)	0.42	1.3	(3.42)	0.42	1.2	(2.07)
PCB-99	1.4	6.7	(0.85)	0.88	5.6	(0.94)	0.15	0.46	(1.21)	0.13	0.38	(0.66)
PCB-101	3.8	19	(2.41)	2.6	17	(2.84)	<0.47	<1.4	(3.68)	0.60	1.8	(3.10)
PCB-105	1.3	6.5	(0.82)	0.79	5.0	(0.84)	0.13	0.38	(1.00)	0.11	0.33	(0.57)
PCB-110	3.5	17	(2.15)	4.4	28	(4.68)	<0.43	<1.3	(3.42)	0.82	2.4	(4.14)
PCB-114	0.072	0.36	(0.05)	<0.027	<0.17	(0.03)	0.0071	0.022	(0.06)	0.0073	0.022	(0.04)
PCB-118	3.5	17	(2.15)	2.3	15	(2.51)	0.31	0.93	(2.45)	0.39	1.2	(2.07)
PCB-122	0.035	0.17	(0.02)	<0.029	<0.18	(0.03)	<0.0066	<0.020	(0.05)	0.0048	0.014	(0.02)
PCB-123	0.065	0.32	(0.04)	0.044	0.28	(0.05)	<0.0067	<0.021	(0.06)	<0.0041	<0.012	(0.02)
PCB-128	1.4	6.8	(0.86)	0.56	3.6	(0.60)	0.069	0.21	(0.55)	0.17	0.50	(0.86)
PCB-138	8.8	44	(5.57)	4.0	26	(4.35)	0.44	1.4	(3.68)	1.3	3.9	(6.72)
PCB-141	2.4	12	(1.52)	0.96	6.1	(1.02)	<0.14	<0.43	(1.13)	0.38	1.1	(1.90)
PCB-153	9.6	48	(6.08)	4.7	30	(5.02)	<0.64	<1.9	(5.00)	1.7	5.0	(8.62)
PCB-156	0.92	4.6	(0.58)	0.42	2.7	(0.45)	0.066	0.20	(0.53)	0.14	0.42	(0.72)
PCB-157	0.11	0.55	(0.07)	0.050	0.32	(0.05)	0.0094	0.029	(0.08)	0.011	0.034	(0.06)
PCB-167	0.35	1.7	(0.22)	0.19	1.2	(0.20)	0.023	0.071	(0.19)	0.055	0.16	(0.28)
PCB-170	16	79	(10.0)	2.0	12	(2.01)	0.32	0.98	(2.58)	0.94	2.8	(4.83)
PCB-180	22	108	(13.7)	3.4	22	(3.68)	0.52	1.6	(4.21)	1.6	4.7	(8.10)
PCB-183	3.8	19	(2.41)	0.56	3.6	(0.60)	0.067	0.20	(0.53)	0.23	0.67	(1.16)
PCB-187	6.7	33	(4.18)	1.2	7.4	(1.24)	0.16	0.49	(1.29)	0.50	1.5	(2.59)
PCB-189	0.47	2.3	(0.29)	0.078	0.50	(0.08)	0.011	0.035	(0.09)	0.031	0.091	(0.16)
PCB-194	7.4	37	(4.68)	0.45	2.9	(0.48)	0.11	0.33	(0.87)	0.22	0.64	(1.10)
PCB-206	1.2	5.9	(0.75)	0.36	2.3	(0.38)	0.065	0.20	(0.53)	0.12	0.34	(0.59)
PCB-209	0.66	3.3	(0.42)	0.42	2.7	(0.45)	0.084	0.26	(0.68)	0.097	0.29	(0.50)
∑PCBs ng g-1												
sum (lower bound)	160	790		94	598		11	32		20	58	
sum (upper bound)	160	790		94	598		12	38		20	58	
∑Indicator-PCBs ng g-1												
Indicator-PCBs (lower bound)	76	378		47	296		4.8	15		10	31	
Indicator-PCBs (upper bound)	76	378		47	296		5.9	18		10	31	
∑PCB-TEQ pg g-1												
WHO1998 TEQ (lower bound)	1.1	5.4		0.57	3.6		0.35	1.1		0.40	1.2	
WHO1998 TEQ (upper bound)	1.2	6.1		0.66	4.2		0.35	1.1		0.40	1.2	
WHO2005 TEQ (lower bound)	**0.22**	**1.1**		**0.13**	**0.83**		**0.29**	**0.88**		**0.30**	**0.89**	
WHO2005 TEQ (upper bound)	**0.36**	**1.8**		**0.21**	**1.4**		**0.29**	**0.88**		**0.30**	**0.89**	
WHO1998 fishTEQ (lower bound)	**0.05**	**0.22**		**0.03**	**0.18**		**0.04**	**0.11**		**0.04**	**0.11**	
WHO1998 fishTEQ (upper bound)	**0.05**	**0.26**		**0.03**	**0.20**		**0.04**	**0.11**		**0.04**	**0.11**	

Table S2, 4/4. PCBs pg g^{-1} (coplanar PCBs and TEQ-values) or ng g^{-1} (all other values) in sediment wet weight (ww) and dry weight (dw) in contaminated R. Kymijoki (6 sites with 2 replicate surface sediment samples: Kuusaansaari, Keltti, Lopotti, Koskenalusjärvi, Ahvionkoski ja Kyminlinna) and in reference R. Urpalanjoki sediments. Percentage of each congener of upper bound sum of concentrations in parentheses. Sediment overall PCB-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Urpalanjoki**			**Urpalanjoki**		
	Urpalanjoki (7A)			**Urpalanjoki (7B)**		
Sampling time	25.7.2012			25.7.2012		
Coplanar PCBs pg g-1	ww	dw	(%)	ww	dw	(%)
CO-PCB-77	<3.5	<6.2	(0.07)	<4.2	<6.2	(0.07)
CO-PCB-81	0.15	0.27	(<0.01)	<0.035	<0.051	(<0.01)
CO-PCB-126	<0.30	<0.52	(0.01)	<0.35	<0.52	(0.01)
CO-PCB-169	<0.031	<0.055	(<0.01)	<0.035	<0.052	(<0.01)
Other PCBs ng g-1						
PCB-18	<0.033	<0.059	(0.65)	<0.040	<0.059	(0.65)
PCB-28/31	<0.12	<0.20	(2.20)	<0.14	<0.20	(2.20)
PCB-33	<0.047	<0.084	(0.92)	<0.057	<0.084	(0.92)
PCB-47	<0.021	<0.037	(0.41)	<0.025	<0.037	(0.41)
PCB-49	<0.013	<0.023	(0.25)	<0.015	<0.023	(0.25)
PCB-51	<0.0038	<0.0067	(0.07)	<0.0043	<0.0064	(0.07)
PCB-52	<0.089	<0.16	(1.76)	<0.11	<0.16	(1.76)
PCB-60	<0.014	<0.025	(0.27)	<0.017	<0.025	(0.27)
PCB-66	<0.032	<0.056	(0.62)	<0.038	<0.056	(0.62)
PCB-74	<0.0083	<0.015	(0.16)	<0.0098	<0.015	(0.16)
PCB-99	<0.018	<0.033	(0.36)	<0.022	<0.033	(0.36)
PCB-101	<0.81	<1.4	(15.4)	<0.96	<1.4	(15.4)
PCB-105	<0.017	<0.031	(0.34)	<0.021	<0.031	(0.34)
PCB-110	<0.74	<1.3	(14.3)	<0.88	<1.3	(14.3)
PCB-114	<0.0037	<0.0066	(0.07)	<0.0044	<0.0065	(0.07)
PCB-118	<0.21	<0.36	(3.96)	<0.25	<0.36	(3.96)
PCB-122	<0.0040	<0.0070	(0.08)	<0.0047	<0.0069	(0.08)
PCB-123	<0.0050	<0.0088	(0.10)	<0.0054	<0.0080	(0.09)
PCB-128	<0.063	<0.11	(1.21)	<0.075	<0.11	(1.21)
PCB-138	<0.69	<1.2	(13.2)	<0.83	<1.2	(13.2)
PCB-141	<0.24	<0.43	(4.73)	<0.29	<0.43	(4.73)
PCB-153	<1.1	<1.9	(20.9)	<1.3	<1.9	(20.9)
PCB-156	<0.046	<0.082	(0.90)	<0.055	<0.082	(0.90)
PCB-157	<0.0027	<0.0048	(0.05)	<0.0026	<0.0039	(0.04)
PCB-167	<0.021	<0.037	(0.41)	<0.025	<0.037	(0.41)
PCB-170	<0.15	<0.26	(2.86)	<0.17	<0.26	(2.86)
PCB-180	<0.34	<0.60	(6.59)	<0.41	<0.60	(6.59)
PCB-183	<0.10	<0.18	(1.98)	<0.12	<0.18	(1.98)
PCB-187	<0.19	<0.33	(3.63)	<0.22	<0.33	(3.63)
PCB-189	<0.0028	<0.0050	(0.05)	<0.0034	<0.0050	(0.05)
PCB-194	<0.012	<0.022	(0.24)	<0.015	<0.022	(0.24)
PCB-206	<0.0037	<0.0066	(0.07)	<0.0047	<0.0069	(0.08)
PCB-209	<0.0026	<0.0045	(0.05)	<0.0014	<0.0021	(0.02)
∑PCBs ng g-1						
sum (lower bound)	0,0002	0,0003		0,0	0,0	
sum (upper bound)	5.1	9.1		6.1	9.1	
∑Indicator-PCBs ng g-1						
Indicator-PCBs (lower bound)	0,0	0,0		0,0	0,0	
Indicator-PCBs (upper bound)	3.1	5.6		3.8	5.6	
∑PCB-TEQ pg g-1						
WHO1998 TEQ (lower bound)	1,52E-05	2,686E-05		0,00	0,00	
WHO1998 TEQ (upper bound)	0.080	0.14		0.095	0.14	
WHO2005 TEQ (lower bound)	**4,559E-05**	**8,058E-05**		**0,000**	**0,000**	
WHO2005 TEQ (upper bound)	**0.040**	**0.071**		**0.048**	**0.071**	
WHO1998 fishTEQ (lower bound)	**0.0001**	**0.0001**		**0.000**	**0.0000**	
WHO1998 fishTEQ (upper bound)	**0.0035**	**0.006**		**0.004**	**0.0059**	

Table S3. PCDD/Fs (pg g^{-1}) in lamprey larvae wet weight (ww), dry weight (dw) and fat (lipid) in contaminated R. Kymijoki (2 sites Ahvionkoski and Kyminlinna) and in the reference R. Urpalanjoki. Percentage of each congener of upper bound sum of concentrations in parentheses. Tissue overall PCDD/F-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki**				**Kymijoki**				**Urpalanjoki**			
	Ahvionkoski				**Kyminlinna**				**Muurikkalan Myllypato**			
Sampling date	26.7.2012				26.7.2012				25.7.2012			
Lipid (%)	6.3				11.1				7.4			
Dry matter (%)	23.5				26.6				25.8			
PCDD pg g-1	ww	dw	lipid	(%)	ww	dw	lipid	(%)	ww	dw	lipid	(%)
2,3,7,8-TCDD	0.77	3.3	12	(0.07)	1.5	5.5	13	(0.04)	0.059	0.23	0.79	(1.10)
1,2,3,7,8-PeCDD	1.3	5.7	21	(0.12)	2.3	8.6	21	(0.06)	0.12	0.47	1.6	(2.22)
1,2,3,4,7,8-HxCDD	0.28	1.2	4.4	(0.02)	0.43	1.6	3.9	(0.01)	0.065	0.25	0.87	(1.21)
1,2,3,6,7,8-HxCDD	9.1	39	144	(0.81)	16	60	142	(0.41)	0.16	0.60	2.1	(2.92)
1,2,3,7,8,9-HxCDD	2.0	8.6	32	(0.18)	3.6	14	32	(0.09)	0.052	0.20	0.70	(0.97)
1,2,3,4,6,7,8-HpCDD	5.0	21	79	(0.45)	9.3	35	83	(0.24)	0.37	1.4	5.0	(6.94)
OCDD	3.7	16	59	(0.33)	7.0	26	62	(0.18)	0.73	2.8	9.8	(13.6)
PCDF pg g-1												
2,3,7,8-TCDF	6.1	26	98	(0.55)	11	41	99	(0.28)	0.76	2.9	10	(13.9)
1,2,3,7,8-PeCDF	1.2	5.2	19	(0.11)	2.1	7.8	19	(0.05)	0.22	0.86	3.0	(4.17)
2,3,4,7,8-PeCDF	4.9	21	78	(0.44)	9.3	35	84	(0.24)	0.54	2.1	7.3	(10.1)
1,2,3,4,7,8-HxCDF	8.4	36	134	(0.76)	22	81	193	(0.55)	0.22	0.87	3.0	(4.17)
1,2,3,6,7,8-HxCDF	6.8	29	108	(0.61)	15	54	130	(0.37)	0.14	0.56	1.9	(2.64)
2,3,4,6,7,8-HxCDF	10	43	160	(0.90)	23	88	210	(0.60)	0.17	0.66	2.3	(3.19)
1,2,3,7,8,9-HxCDF	0.14	0.60	2.3	(0.01)	0.33	1.2	3.0	(0.01)	<0.028	<0.11	<0.38	(0.53)
1,2,3,4,6,7,8-HpCDF	415	1 765	6 589	(37.1)	1 638	6 153	14 696	(42.1)	1.0	3.9	14	(19.4)
1,2,3,4,7,8,9-HpCDF	6.3	27	99	(0.56)	21	78	186	(0.53)	0.046	0.18	0.62	(0.86)
OCDF	637	2 706	10 102	(56.9)	2 107	7 914	18 903	(54.2)	0.66	2.6	9.0	(12.5)
∑PCDD/Fs pg g-1												
sum (lower bound)	1 118	4 752	17 741		3 888	14 604	34 880		5.3	21	72	
sum (upper bound)	1 118	4 752	17 741		3 888	14 604	34 880		5.4	21	72	
WHO1998 TEQ (lower bound)	13	56	211		34	130	309		0.63	2.5	8.5	
WHO1998 TEQ (upper bound)	13	56	211		34	130	309		0.64	2.5	8.6	
WHO2005 TEQ (lower bound)	**12**	**53**	**197**		**33**	**124**	**296**		**0.52**	**2.0**	**7.0**	
WHO2005 TEQ (upper bound)	**12**	**53**	**197**		**33**	**124**	**296**		**0.52**	**2.0**	**7.1**	
WHO1998 fishTEQ (lower bound)	**12**	**51**	**190**		**32**	**121**	**290**		**0.60**	**2.32**	**8.03**	
WHO1998 fishTEQ (upper bound)	**12**	**51**	**190**		**32**	**121**	**290**		**0.60**	**2.33**	**8.06**	

Table S4. PCBs pg g^{-1} (coplanar PCBs and TEQ-values) or ng g^{-1} (all other values) in lamprey larvae wet weight (ww), dry weight (dw) and fat (lipid) in contaminated R. Kymijoki (2 sites Ahvionkoski and Kyminlinna) and in the reference R. Urpalanjoki. Percentage of each congener of upper bound sum of concentrations in parentheses. Tissue overall PCB-toxicity TEQ is estimated using WHO Toxic Equivalency Factors for mammals (1998, 2005) and for fish (1998).

Site	**Kymijoki, Ahvionkoski**				**Kymijoki, Kyminlinna**				**Urpalanjoki**			
Sampling time	26.7.2012				26.7.2012				25.7.2012			
Lipid (%)	6.3				11.1				7.4			
Dry matter (%)	23.5.				26.6				25.8			
Coplanar PCBs pg g-1	ww	dw	lipid	(%)	ww	dw	lipid	(%)	ww	dw	lipid	(%)
CO-PCB-77	283	1 202	4 486	(0.59)	685	2 574	6 148	(0.72)	20	76	264	(0.36)
CO-PCB-81	14	58	215	(0.03)	33	125	299	(0.04)	1.0	3.9	14	(0.02)
CO-PCB-126	6.2	26	98	(0.01)	9.6	36	86	(0.01)	2.4	9.4	33	(0.04)
CO-PCB-169	0.97	4.1	15	(<0.01)	1.2	4.5	11	(<0.01)	0.54	2.1	7.3	(0.01)
Other PCBs ng g-1												
PCB-18	3.2	13	50	(6.53)	5.5	21	49	(5.76)	0.17	0.65	2.3	(3.11)
PCB-28/31	11	48	177	(23.1)	23	87	207	(24.4)	0.50	1.9	6.8	(9.19)
PCB-33	3.0	13	47	(6.14)	5.7	21	51	(6.00)	0.14	0.53	1.9	(2.57)
PCB-47	1.2	5.1	19	(2.48)	3.6	13	32	(3.76)	0.050	0.19	0.67	(0.91)
PCB-49	1.9	8.0	30	(3.92)	5.4	20	49	(5.76)	0.075	0.29	1.0	(1.35)
PCB-51	0.11	0.45	1.7	(0.22)	0.27	1.0	2.4	(0.28)	0.0058	0.023	0.079	(0.11)
PCB-52	2.7	12	43	(5.61)	6.8	26	61	(7.18)	0.17	0.67	2.3	(3.11)
PCB-60	0.96	4.1	15	(1.96)	1.5	5.7	14	(1.65)	0.073	0.28	0.98	(1.32)
PCB-66	4.1	17	65	(8.49)	9.0	34	81	(9.53)	0.19	0.75	2.6	(3.51)
PCB-74	1.8	7.5	28	(3.66)	4.1	15	37	(4.35)	0.087	0.34	1.2	(1.62)
PCB-99	0.74	3.2	12	(1.57)	1.5	5.7	14	(1.65)	0.12	0.45	1.6	(2.16)
PCB-101	2.5	11	40	(5.22)	4.9	19	44	(5.18)	0.60	2.3	8.1	(11.0)
PCB-105	0.36	1.5	5.7	(0.74)	0.62	2.3	5.6	(0.66)	0.077	0.30	1.0	(1.35)
PCB-110	2.2	9.2	34	(4.44)	3.7	14	33	(3.88)	0.61	2.3	8.2	(11.1)
PCB-114	0.029	0.12	0.46	(0.06)	0.055	0.20	0.49	(0.06)	0.0055	0.021	0.074	(0.10)
PCB-118	1.6	6.8	25	(3.26)	2.7	10	25	(2.94)	0.34	1.3	4.6	(6.22)
PCB-122	0.011	0.048	0.18	(0.02)	0.022	0.083	0.20	(0.02)	0.0019	0.0073	0.026	(0.04)
PCB-123	0.026	0.11	0.41	(0.05)	0.039	0.14	0.35	(0.04)	0.0033	0.013	0.045	(0.06)
PCB-128	0.17	0.71	2.7	(0.35)	0.30	1.1	2.7	(0.32)	0.047	0.18	0.64	(0.86)
PCB-138	2.1	8.7	33	(4.31)	3.3	12	30	(3.53)	0.53	2.1	7.2	(9.73)
PCB-141	0.48	2.0	7.6	(0.99)	0.87	3.3	7.8	(0.92)	0.13	0.51	1.8	(2.43)
PCB-153	4.5	19	71	(9.27)	6.3	24	57	(6.71)	0.92	3.6	12	(16.2)
PCB-156	0.22	0.94	3.5	(0.46)	0.32	1.2	2.8	(0.33)	0.044	0.17	0.59	(0.80)
PCB-157	0.018	0.077	0.29	(0.04)	0.027	0.10	0.24	(0.03)	0.0046	0.018	0.062	(0.08)
PCB-167	0.12	0.50	1.9	(0.25)	0.16	0.60	1.4	(0.16)	0.021	0.080	0.28	(0.38)
PCB-170	0.48	2.0	7.6	(0.99)	0.76	2.9	6.8	(0.80)	0.091	0.35	1.2	(1.62)
PCB-180	1.3	5.7	21	(2.74)	1.9	7.0	17	(2.00)	0.24	0.91	3.2	(4.32)
PCB-183	0.24	1.0	3.8	(0.50)	0.37	1.4	3.3	(0.39)	0.061	0.23	0.82	(1.11)
PCB-187	0.56	2.4	8.9	(1.16)	0.84	3.1	7.5	(0.88)	0.12	0.46	1.6	(2.16)
PCB-189	0.025	0.11	0.40	(0.05)	0.033	0.12	0.29	(0.03)	0.0034	0.013	0.045	(0.06)
PCB-194	0.11	0.45	1.7	(0.22)	0.14	0.52	1.3	(0.15)	0.013	0.052	0.18	(0.24)
PCB-206	0.082	0.35	1.3	(0.17)	0.083	0.31	0.75	(0.09)	0.0070	0.027	0.095	(0.13)
PCB-209	0.10	0.43	1.6	(0.21)	0.094	0.35	0.84	(0.10)	0.0092	0.036	0.12	(0.16)
∑PCBs ng g-1												
sum (lower bound)	48	205	766		95	356	850		5.5	21	74	
sum (upper bound)	48	205	766		95	356	850		5.5	21	74	
∑Indicator-PCBs ng g-1												
Indicator-PCBs (lower bound)	24	103	386		46	174	415		3.0	11	40	
Indicator-PCBs (upper bound)	24	103	386		46	174	415		3.0	11	40	
∑PCB-TEQ pg g-1												
WHO1998 TEQ (lower bound)	0.99	4.2	16		1.6	6.0	14		0.32	1.2	4.3	
WHO1998 TEQ (upper bound)	0.99	4.2	16		1.6	6.0	14		0.32	1.2	4.3	
WHO2005 TEQ (lower bound)	**0.75**	**3.2**	**12**		**1.2**	**4.5**	**11**		**0.28**	**1.1**	**3.7**	
WHO2005 TEQ (upper bound)	**0.75**	**3.2**	**12**		**1.2**	**4.5**	**11**		**0.28**	**1.1**	**3.7**	
WHO1998 fishTEQ (lower bound)	**0.08**	**0.33**	**1.24**		**0.15**	**0.57**	**1.38**		**0.02**	**0.07**	**0.23**	
WHO1998 fishTEQ (upper bound)	**0.08**	**0.33**	**1.24**		**0.15**	**0.57**	**1.38**		**0.02**	**0.07**	**0.23**	

Table S5. Biota-sediment accumulation factors (BSAFoc) of PCDD/Fs for congeners measured above LOQ, from sediment organic carbon to lamprey larvae lipid in R. Kymijoki Ahvionkoski and Kyminlinna.

Site	R. Kymijoki Ahvionkoski, site 5	R. Kymijoki Kyminlinna, site 6
	BSAFoc	BSAFoc
2,3,7,8-TCDD	-	1.708
1,2,3,7,8-PeCDD	2.882	1.482
1,2,3,4,7,8-HxCDD	-	0.594
1,2,3,6,7,8-HxCDD	0.734	0.381
1,2,3,7,8,9-HxCDD	0.489	0.240
1,2,3,4,6,7,8-HpCDD	0.148	0.085
OCDD	0.059	0.037
2,3,7,8-TCDF	3.279	1.337
1,2,3,7,8-PeCDF	1.004	0.521
2,3,4,7,8-PeCDF	0.909	0.550
1,2,3,4,7,8-HxCDF	0.147	0.122
1,2,3,6,7,8-HxCDF	0.219	0.176
2,3,4,6,7,8-HxCDF	0.202	0.154
1,2,3,7,8,9-HxCDF	-	0.220
1,2,3,4,6,7,8-HpCDF	0.025	0.022
1,2,3,4,7,8,9-HpCDF	0.049	0.029
OCDF	0.021	0.016

Table S6. Biota-sediment accumulation factors (BSAFoc) of PCBs for congeners measured above LOQ, from sediment organic carbon to lamprey larvae lipid in R. Kymijoki Ahvionkoski and Kyminlinna.

Site	R. Kymijoki Ahvionkoski BSAFoc	R. Kymijoki Kyminlinna BSAFoc
CO-PCB-77	5.549	2.765
CO-PCB-81	5.705	3.636
CO-PCB-126	-	2.905
CO-PCB-169	5.077	1.846
PCB-18	0.878	6.093
PCB-28/31	0.646	4.183
PCB-33	0.651	3.986
PCB-47	0.806	11.134
PCB-49	0.869	10.529
PCB-51	0.917	12.947
PCB-52	0.919	10.612
PCB-60	0.938	3.687
PCB-66	0.804	4.866
PCB-74	0.823	7.205
PCB-99	1.043	8.122
PCB-101	1.199	-
PCB-105	0.524	3.842
PCB-110	0.886	-
PCB-114	-	5.418
PCB-118	0.841	5.697
PCB-122	-	-
PCB-123	0.735	-
PCB-128	0.262	1.837
PCB-138	0.483	2.730
PCB-141	0.422	-
PCB-153	0.938	-
PCB-156	0.490	2.183
PCB-157	0.341	1.851
PCB-167	0.685	2.928
PCB-170	0.076	0.868
PCB-180	0.150	1.301
PCB-183	0.155	1.828
PCB-187	0.206	1.817
PCB-189	0.133	1.111
PCB-194	0.038	0.648
PCB-206	0.155	0.673
PCB-209	0.284	0.742