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EFFECT OF SEDIMENT ON THE BIOACCUMULATION OF A COMPLEX MIXTURE OF POLYCHLORINATED DIBENZO-*p*-DIOXINS (PCDDs) AND POLYCHLORINATED DIBENZOFURANS (PCDFs) BY FISH

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ABSTRACT

Guppies (*Poecilia reticulata*) were exposed to a complex mixture of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) in the presence and absence of sediment for 21 days. Since the guppies were separated from sediment to prevent uptake of sediment into the intestines, uptake was possible only from water. The accumulation kinetics, bioconcentration factors and biota-sediment accumulation factors were determined for 13 toxic PCDD and PCDF congeners.
PCDDs and PCDFs were bioavailable to fish. In the presence of sediment, however, the accumulation was reduced compared to the accumulation in the system without sediment. Bioconcentration factors were reduced between 15% for 2,3,7,8-TCDF and 82% for OCDD by the presence of sediment. Reduction is probably caused by the introduction of organic matter in the water. Biota-sediment accumulation factors ranged from 0.155 for 2,3,7,8-TCDD to 0.003 for OCDD. The effects of sediment increased with increasing hydrophobicity of the compounds.

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INTRODUCTION

Persistent hydrophobic compounds are ubiquitously present in the aquatic environment due to pollution from industrial and municipal sources. As these compounds are not readily biodegradable they accumulate in sediment. The fate of hydrophobic organic compounds in the aquatic environment is largely determined by their sorption to sediment. It is possible that sediment acts as a source of contaminants to organisms living in direct contact with the polluted sediment, such as benthic invertebrates. Considering organisms that do not live in direct contact with the sediment, it is generally found that the bioavailability of hydrophobic compounds is reduced due to the sorption to sediment [1,2]. In the short term, sediment can act as a sink for hazardous chemicals and reduce the uptake of these compounds by pelagic organisms. However, concerning a longer period of time, sediment can act as a secondary source to these organisms. If the immissions into the aquatic environment are reduced, previously sorbed chemicals may desorb into the water phase and become bioavailable again, a process that can continue for many decades.

The extent of sorption to sediment depends on characteristics of both the sediment and the chemical involved. Sorption to sediment is amongst other factors determined by the sediments organic carbon content and particle size distribution. The affinity of a nonpolar compound for sediment is primarily governed by its hydrophobicity.

Hydrophobicity is also an important factor in the bioaccumulation process. It was demonstrated in laboratory studies [3,4] that the extent of bioaccumulation of persistent organic micropollutants can be predicted from the compounds hydrophobicity. Linear relationships were observed between log K_{ow} (1-octanol/water partition coefficient, a measure of hydrophobicity) and log bioconcentration factors in laboratory studies [5]. However, these relationships do not hold for extremely hydrophobic compounds (log $K_{ow} > 6$). In the field situation the process of bioaccumulation is complicated by the presence of suspended and dissolved organic matter and sediment which effects the bioavailability of these compounds.

In the current study the bioaccumulation of a mixture of polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) is investigated. PCDDs and PCDFs are extremely hydrophobic compounds with log K_{ow} values > 6. In the aquatic environment a large fraction of PCDDs and PCDFs will be sorbed to organic matter and sediment compared to the truly dissolved fraction. However, PCDDs and PCDFs are observed in a wide variety of organisms [6,7] and must therefore be bioavailable to a certain extent. Reduced bioaccumulation of PCDDs and PCDFs in the presence of sediment or dissolved humic materials has been reported before [8,9]. In these studies fish lived in direct contact with the sediment.

The aim of the present study was to investigate whether sediment sorbed PCDDs and PCDFs are bioavailable and if so, to what extent bioaccumulation is affected. The bioaccumulation of a complex mixture, containing almost all the tetra- and more highly chlorinated PCDD and PCDF congeners (extracted from fly-ash) was compared in the presence and absence of sediment. Sediment is usually spiked in the laboratory by adding the contaminants in an organic solvent. The application of a carrier solvent may effect the behaviour of very hydrophobic compounds. In the current study, the sediment was spiked through the water phase, which gives a better approach to the field situation, providing for a homogeneous loading of the sediment. To prevent uptake of significant amounts of sediment in the gastro-intestinal tract, guppies were separated from direct contact with the sediment. Bioconcentration factors and biota-sediment accumulation factors were determined for most of the toxic, laterally substituted congeners.

MATERIALS AND METHODS

Chemicals

Dioxins and furans were soxhlet extracted with toluene from 190 g fly-ash from a municipal incinerator near Zaanstad, the Netherlands. The fly-ash extract was cleaned up as described below for the fish samples and concentrated in 5 ml hexane. In the fly-ash extract 104 different peaks were separated by GC-MSD analysis of the tetra- and more highly chlorinated PCDDs and PCDFs. [^{13}C]-labelled dioxins and furans (quantification standards) were obtained from Cambridge Isotopes Laboratories. Chromosorb (P-AW 60-80 mesh) was obtained from CHROMPACK. All solvents used (hexane, toluene, n-nonane, dichloromethane, chloroform, and tetrachloromethane) were of glass distilled quality. Reagents used were silica gel (kieselgel 60, 70-230 mesh), aluminium oxide (Al_2O_3, basic 70-230 mesh), silver nitrate ($AgNO_3$ p.a.), potassium hydroxide (NaOH p.a.) and concentrated sulphuric acid (H_2SO_4 p.a).

Sediment

Field sediment from Lake Schoonrewoerdse Wiel (the Netherlands) was used. The organic carbon content was 10.1%. The particle size distribution was as follows: 83% > 60 μm; 1.2%: 50 - 60 μm; 7.2%: 16 - 50 μm; 2.4%: 10 - 16 μm; 4.4%: 2 - 10 μm and 1.4% < 2 μm. The sediment was freeze dried before application in the experiment. In the original sediment no PCDDs and PCDFs were detected. The sediment was loaded with PCDDs and PCDFs in the laboratory (see under exposure systems).

Organisms

Adult female guppies (*Poecilia reticulata*) were used with an average wet weight of 0.84 ± 0.17 g. The average lipid content was 8.0 ± 1.7%.

Exposure systems

The experiment was performed in two different systems. In system I guppies were exposed to water contaminated with PCDDs and PCDFs. In system II guppies were exposed to water that was circulated through a column with sediment contaminated with PCDDs and PCDFs. In both experiments clean water and clean sediment were loaded in the laboratory with a mixture of PCDDs and PCDFs that was extracted from fly-ash. The water in system I was contaminated by the generator column technique as described by Veith and Comstock [10]. Chromosorb had been coated with fly-ash extract in hexane. The hexane was evaporated under nitrogen. The coated chromosorb was placed in the generator column. Water was circulated through the generator column with chromosorb for 8 days before introduction of the organisms. The water contamination procedure has been described in detail before [11]. The construction was adapted in system II, in order also to contaminate sediment. The contamination of sediment was performed as follows. Water was circulated through two glass columns in series for 7 days. The first column contained chromosorb coated with fly-ash extract, the second column contained 0.237 g of sediment (dry weight). After 7 days of water circulation, the column with chromosorb was disconnected from the system and water was pumped through the column with sediment for one additional day for further equilibration of the sediment/water system before introduction of the fishes.

Accumulation experiments

The water in system I was circulated continuously over chromosorb during the exposure period. The water in system II was circulated continuously over sediment during exposure of guppies. The aquaria were maintained at a temperature of 25.0 ± 1.2°C. Water was aerated through glass capillaries. Aquaria were lit 16 hours a day with fluorescent daylight lamps. Fish were fed dried fish food three times a week. PCDDs and PCDFs were not detectable in food. Food and faeces residues were removed from the aquaria 3 times a week by an aquarium vacuum cleaner device. Fish were sampled at different time intervals in duplicate or triplicate. Organisms were killed in liquid nitrogen and stored at -18°C until analysis. Water (250 ml) was also sampled in duplicate or triplicate. Extraction of water was started immediately after sampling by adding 5 ml hexane.

Sample analysis

Fish samples were freeze dried for 24 h and prior to extraction a mixture of 10 [^{13}C]-PCDDs and [^{13}C]-PCDFs was added as internal standard. The samples were soxhlet extracted with toluene for

24 h. The extract was cleaned up by open column chromatography. Silica was washed with dichloromethane and dried under a nitrogen stream for 24 h. Silica was activated under nitrogen at 180°C for 90 minutes. Activated silica was treated with H_2SO_4 (22 or 44% w/w), NaOH (33% w/w) or $AgNO_3$ (10% w/w). The sample extracts were passed over macro-columns (diameter 10 mm) with H_2SO_4 on silica (2 cm 22%; 10 cm 44%) and NaOH on silica (9 cm). The macro-columns were eluted with 50 ml hexane. After concentration of the samples under nitrogen at 50°C., the extracts were transferred to high aspect columns (diameter 6 mm) with $AgNO_3$ on silica (10 cm) and high aspect columns with Al_2O_3 (19 cm). Both high aspect columns were eluted simultaneously with 80 ml hexane. Al_2O_3 was further eluted with 20 ml 10% CCl_4 and 30 ml dichloromethane. The dichloromethane extracts were concentrated under nitrogen at 50 °C. and transferred with hexane to a small glass vial. Water samples were extracted with hexane (3 times 5 ml) and cleaned up by the same procedure with exception of the $AgNO_3$ column.

Quantification of fish and water extracts was performed by GC-MSD analysis (HP5890/HP5970) using a 60 m Supelco 2331 column; temperature program: 140°C, 40°C/min, 200°C, 4°C/min, 250°C. Prior to injection the extracts were evaporated to dryness and dissolved in n-nonane. Circa two microlitres were injected at an inlet pressure of 12 psi He and analysed with the SIM program (selected ion monitoring). Identification of PCDD and PCDF peaks in the chromatograms was based on retention times and isotope ratios. Detection limits were defined as the ratio of two times the background noise divided by the internal standard peak, multiplied by the amount of internal standard.

Data analysis

Data from an extensive experiment performed previously [11] were integrated with data from system I. The former experiment was performed under identical circumstances, resulting in comparable concentrations in water and fish.

Bioconcentration factors (BCFs) were calculated from the ratios of the concentrations in fish after 21 days exposure and the average concentrations in water during the experiment (eq. 1). Values for fish were expressed on a lipid weight base. Biota-Sediment Accumulation Factors (BSAFs) were defined as the ratio of the concentrations in fish at the end of the exposure period and the average concentrations in sediment (eq. 2). Concentrations in fish and sediment were expressed on lipid weight and organic carbon weight respectively in the calculation of BSAF values.

$$BCF = C_f / C_w \qquad \text{eq. 1}$$

$$BSAF = C_f / C_s \qquad \text{eq. 2}$$

BCF = bioconcentration factor (l kg^{-1}), BSAF = biota-sediment accumulation factors, C_f = concentration in fish at day 21 ($\mu g\ kg^{-1}$), C_w = average concentration in water during the exposure experiment (ng l^{-1}), C_s = average concentration in sediment ($\mu g\ kg^{-1}$).

RESULTS AND DISCUSSION

PCDDs or PCDFs were not detected in control fish samples. Detection limits ranged from 0.1 to 4.1 $\mu g\ kg^{-1}$ lipid weight in fish. In control water samples traces of hepta- and octachlorinated dioxins were observed. These concentrations were negligable compared to the concentrations in the exposure water.

Table I: Average concentrations of the toxic PCDDs in water (C_w), sediment (C_s) and fish (C_f at t= 21 d)(). I is system without sediment; II is system with sediment. Concentrations in water are expressed in ng l^{-1}, concentrations in sediment are based on organic carbon weight ($\mu g\ kg^{-1}$), in fish on lipid weight ($\mu g\ kg^{-1}$).

Compound	C_{wI} avg	se	C_{wII} avg	se	C_s avg	se	C_{fI} avg	se	C_{fII} avg	se
2378 TCDD	0.08	0.02	0.12	0.07	62	-[1)]	5.1	1.3	9.6	0.6
2378 TCDF	0.12	0.02	0.19	0.09	184	-	2.2	0.3	2.5	-
12378 PCDD	0.21	0.04	0.29	0.10	351	-	36.0	8.2	28.2	4.2
12378 PCDF	0.14	0.04	0.14	0.05	168	82	3.0	0.3	0.3	-
23478 PCDF	0.16	0.04	0.22	0.08	207	72	22.0	2.2	18.1	2.7
123478 HxCDD	0.20	0.04	0.18	0.06	332	77	21.1	3.4	8.1	1.0
123678 HxCDD	0.23	0.07	0.22	0.08	459	113	24.1	3.1	11.2	1.0
123789 HxCDD	0.28	0.07	0.48	0.16	1259	598	24.2	3.1	10.5	1.8
123478 HxCDF	0.21	0.05	0.28	0.10	226	-	16.1	1.9	7.1	0.8
123678 HxCDF	0.22	0.04	0.20	0.07	292	-	18.0	2.1	6.2	0.8
1234678 HpCDD	1.31	0.17	0.91	0.30	1128	-	58.7	8.1	15.2	1.4
1234678 HpCDF	0.68	0.18	0.66	0.23	483	163	27.0	4.5	7.9	0.1
OCDD	0.80	0.16	0.76	0.27	1138	-	13.1	0.5	3.5	-

[1)] Congener was detected only once.

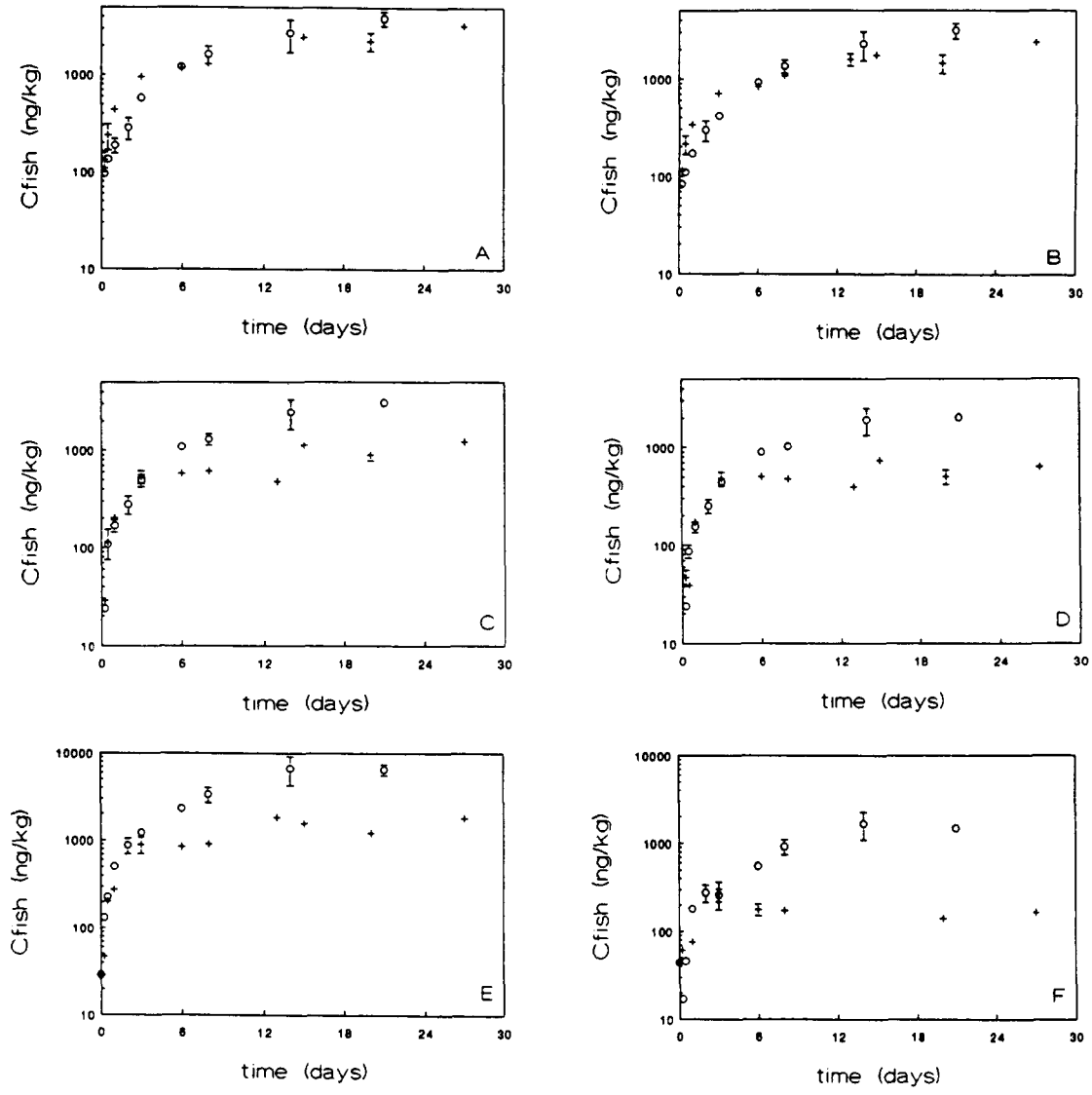

Fig. 1: Concentrations of PCDDs and PCDFs in fish versus time in the presence ( ) and absence ( ) of sediment. A = 1,2,3,7,8-PCDD, B = 2,3,4,7,8-PCDF, C = 1,2,3,6,7,8-HxCDD, D = 1,2,3,6,7,8-HxCDF, E = 1,2,3,4,6,7,8-HpCDD and F = OCDD.

The average concentrations of the toxic PCDDs and PCDFs in water for system I (system without sediment, C_{wI}) and system II (system with sediment, C_{wII}) are presented in Table I. The average concentrations in water ranged from 0.08 to 1.31 ng l^{-1} in system I for the different 2,3,7,8-substituted PCDDs and PCDFs. The average concentrations determined in the water in system II (C_{wII} = 0.12 to 0.91 ng l^{-1}) were comparable to the concentrations in system I. During the experiment a slow decline in water concentrations was observed in both systems. In general the concentrations in water were below the maximum water solubilities [12-14]. However, the water concentrations of hepta- and octachlorinated congeners were close to or even above their reported water solubilities, indicating that possibly not the whole fractions of these compounds were truly dissolved in the water phase, due to the presence of organic matter or the formation of micelles.

The concentrations of PCDDs and PCDFs in sediment were determined only twice, namely at the beginning and at the end of the exposure period. Because no significant change in concentrations was observed, these two values were averaged. The concentrations of the toxic PCDD and PCDF congeners ranged from 62 to 1250 μg kg^{-1} sediment (organic carbon weight) (Table I). The fractions of PCDDs and PCDFs that were sorbed to sediment were 40 to 75% of the total amount present in system II.

Altogether, 104 peaks of the tetra- and more highly chlorinated PCDDs and PCDFs were distinguished with GC analysis of the original fly-ash extract. Almost all of these compounds were present in sediment and water (data not included here). However, in fish tissue only a limited number of PCDDs and PCDFs was detected. A comparable selective accumulation of PCDDs and PCDFs was previously reported in accumulation studies with exposure from food or water [11,15,16]. It can be concluded that PCDDs and PCDFs are accumulated selectively if they are substituted at least at the 2,3,7, and 8 positions. Because of this selective accumulation and because of their toxic activity, only results for 2,3,7,8 substituted congeners are presented in this paper. As 1,2,3,7,8,9- and 2,3,4,6,7,8-HxCDF and 1,2,3,4,7,8,9-HpCDF were present in fly-ash in small amounts only, they were not detected in water and sediment. Therefore these substances were not taken into account. OCDF was not analysed. The results for the remaining 13 2,3,7,8-substituted PCDDs and PCDFs are presented in this paper.

The concentrations in fish after 21 days exposure are shown in Table I. PCDDs and PCDFs were observed in fish exposed in both the presence and absence of sediment. However, the concentrations in organisms from the system with sediment were lower than those in organisms from the system without sediment for most congeners. The average concentrations (lipid weight) in fish at the end of the exposure period ranged from 2.2 to 58.7 μg kg^{-1} for the 2,3,7,8-substituted PCDDs and PCDFs in the system with water only and from 0.3 to 28.2 μg kg^{-1} in the system with sediment. Transferred to toxic 2,3,7,8-TCDD equivalents [17] these concentrations become 5.4 μg kg^{-1} and 3.0 μg kg^{-1} (wet weight) respectively. In the current study exposed fish did turn pale and showed somewhat

less activity than control fishes. Kleeman et. al. [18] reported LD_{50} values ranging from 3 - 16 $\mu g\ kg^{-1}$ 2,3,7,8-TCDD for different fish species after 80 days. In the present experiment concentrations in fish were in this range (3 to 5 $\mu g\ kg^{-1}$ TCDD equivalents), but no increased mortality was observed. This is not in contradiction with the study of Kleeman et al., because in that study increased mortality was reported after at least 25 days, while the current experiment lasted for 21 days. Other effects that were described after exposure to 2,3,7,8-TCDD, such as loss of body weight and fin necrosis, were not observed during the present study. Plots of the concentrations of six PCDDs and PCDFs in fish with time during exposure to water or water and sediment are shown in Fig. 1. These congeners are illustrative for the other 2,3,7,8-substituted congeners. The uptake of tetra- and pentachlorinated PCDDs and PCDFs is apparently not substantially effected by the presence of sediment. Initially, similar uptake rates were also observed for the hexa-, hepta- and octachlorinated congeners in both systems. However, an increasing reduction of accumulation with time was observed for these higher chlorinated compounds if sediment was present. This phenomenon can be explained by the relation between hydrophobicity and affinity for sediment. Large partition coefficients between sediment and water were reported for highly hydrophobic compounds resulting in small dissolved fractions in water. Only this dissolved fraction in water is directly bioavailable for uptake by fish through the gills [19]. From the results in the present study it is obvious that the bioavailable fraction of higher chlorinated PCDDs and PCDFs is reduced more by the presence of sediment than the lower chlorinated congeners.

Bioconcentration factors

Bioconcentration factors (BCFs) were calculated from the ratios of the fish concentrations at the end of the exposure period and the average concentrations in water. The concentrations in fish are expressed on a lipid weight base. Log BCF values are presented in Table II. BCF values in the system without sediment were always larger than those in the system with sediment. The reductions of BCFs for system II compared to BCFs for system I range between 15% and 82% for the different congeners. As bioconcentration is considered to be a partioning process between water and fish, it was expected that the C_f/C_w ratios would be independent of the presence of sediment. The reduction of BCF values in the presence of sediment was probably the result of a fraction of the compounds in water not being truly dissolved in the water phase in system II. These congeners were probably partly sorbed to organic matter that was introduced in the water phase from the sediment and by faeces from the fishes. This organic matter consists of small suspended particles and dissolved organic macromolecules such as humic substances. Because total concentrations in water were determined (including dissolved or suspended organic matter), the truly dissolved concentration in water was overestimated, which results in underestimation of BCF values. It has been shown that dissolved organic matter (DOM) passes through the gill without being taken up, because the concentration of DOM in water is the same before

and after passing the gills [19]. Although the dissolved organic macromolecules together with sorbed compounds are pumped through the gills, they are probably unable to permeate the gill membrane because of their large size and the presence of polar functional groups. A similar reduction (84%) of the bioconcentration factor of benzo-a-pyrene in the presence of 20 mg l^{-1} organic carbon was reported previously [20].

Table II: Log bioconcentration factors (BCF_1) in the absence (system I) en presence (system II) of sediment (l kg^{-1} lipid weight) and biota-sediment accumulation factors (BSAF) (concentrations in fish in lipid weight and sediment concentrations in organic carbon weight).

Compound	log BCF_1 I		log BCF_1 II		BSAF	
	avg	se	avg	se	avg	se
2378 TCDD	5.24	0.22	4.91	0.04	0.155	0.014
2378 TCDF	4.19	0.07	4.12	-[1)]	0.014	-
12378 PCDD	5.27	0.07	4.98	0.08	0.080	0.017
12378 PCDF	4.59	0.15	3.29	-	0.002	-
23478 PCDF	5.14	0.18	4.92	0.08	0.088	0.019
123478 HxCDD	5.01	0.06	4.65	0.07	0.024	0.004
123678 HxCDD	4.94	0.19	4.70	0.05	0.024	0.003
123789 HxCDD	4.93	0.09	4.34	0.09	0.008	0.002
123478 HxCDF	4.91	0.09	4.40	0.06	0.031	0.005
123678 HxCDF	4.95	0.05	4.49	0.07	0.021	0.004
1234678 HpCDD	4.68	0.06	4.23	0.05	0.014	0.002
1234678 HpCDF	4.46	0.07	4.08	0.01	0.016	0.000
OCDD	4.13	0.02	3.38	-	0.003	-

[1)] Congener was detected only once

The effect of sediment on bioaccumulation increased with increasing chlorination of the PCDDs and PCDFs. This is in agreement with the larger hydrophobicities of the latter congeners and hence the larger K_{oc} values. Comparable compoundspecific effects of sediment were reported for

polychlorinated biphenyls and polychlorinated benzenes [21,22]. In all cases the effect of sediment on the bioaccumulation of hydrophobic compounds was related to the hydrophobicity and K_{oc} values of the chemicals.

A relatively small BCF value was determined for 1,2,3,7,8-PCDF in both systems. Obviously other factors are involved in the bioaccumulation behaviour of this congener, such as fast biotransformation [23]. In general the BCF values are lower than would be predicted by the high hydrophobicity of the PCDDs and PCDFs in both exposure systems. Except for the reduced bioavailability due to organic matter, other reasons have been discussed to explain this lack of bioaccumulation [11], such as biotransformation [23], reduced lipid solubility [24] and reduced membrane permeability [25]. Our results show that reduced bioavailability will have an additional limiting effect.

Biota-sediment accumulation factors

Biota-Sediment Accumulation Factors (BSAFs) were calculated from the ratios of the concentrations in fish at the end of the exposure period and the average concentrations in sediment. Concentrations in fish and sediment were based on lipid weight and organic carbon weight, respectively. After 3 weeks exposure the BSAF values were always smaller than one. It has been hypothesized that BSAF values should be approximately one for a large series of stable nonpolar chemicals [26]. This is deduced from the fact that the biota-sediment accumulation factor reflects a partitioning process between fish lipid and organic carbon in sediment with water as the intermediary medium. If the affinities of a chemical are supposed to be equal for lipid and organic carbon, a BSAF value approximating one is expected at steady state and the BSAF value should be independent of the hydrophobicity of the compound. In the present study BSAF values are smaller than one and decrease with increasing number of chlorine atoms. The same phenomenon was reported before for PCDDs and PCDFs. Kuehl et al [8] reported comparable BSAF values ranging from 0.27 for 2,3,7,8-TCDD to 0.003 for 1,2,3,4,6,7,8-HCDF for a mixture of PCDDs and PCDFs after exposure to contaminated sediment in the laboratory. The same pattern of decreasing values with increasing chlorination was also observed in C_{fish}/C_{food} ratios and $C_{fish}/C_{fly\text{-}ash}$ ratios of 2,3,7,8-substituted PCDDs and PCDFs [16,27]. The relatively low BSAF values may be the result of equilibrium not being reached during the experiment. As a result the BSAF values may increase with increasing exposure periods. However, comparable BSAF values were calculated after prolonged exposure periods (205 days) by Kuehl et. al. [8]. Furthermore, the data in fig. 1 show that no further increase of concentrations in fish is observed for the hexa- to octachlorinated compounds after 13 days, hence a further increase in BSAF would not be expected after a prolonged exposure period. The observations that the BSAF ratios become smaller with increasing hydrophobicity (expressed as K_{ow} values) might be attributed to the affinity for lipid of

the compounds with increasing K_{ow} [24,28]. Little is known about the affinities of PCDDs and PCDFs for lipid and sediment. If the affinity for lipid decreases while the affinity for sediment is not, or less, influenced, the BSAFs will be reduced.

CONCLUSIONS

The presence of sediment alters the fate of PCDDs and PCDFs in aquatic systems. Although PCDDs and PCDFs accumulate in organisms in the presence of sediment, small amounts of sediment (10 mg l^{-1}) reduce the biovailability of PCDDs and PCDFs for fishes. The effect of sediment is compound specific: increasing influence of sediment is observed with increasing hydrophobicity of the PCDDs and PCDFs. Three processes may be involved in the effects of sediment. The bioconcentration factors were probably reduced by the presence of organic matter in water, the biota-sediment accumulation factors may become smaller with increasing hydrophobicity due to relatively declining affinities for fish lipids of the highly chlorinated PCDDs and PCDFs and due to a lack of equilibrium between sediment and fish.

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