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Bioaccumulation of anthropogenic contaminants by detritivorous fish in the Río de la Plata estuary: 2-Polychlorinated biphenyls

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Abstract

The temporal variability and bioaccumulation dynamics of individual PCBs were studied in a detritivorous fish (Sábalo: *Prochilodus lineatus*) collected from 1999 to 2005 in the polluted Buenos Aires coastal area. Fish muscles contain high concentrations of total PCBs (11 ± 7.2, 4.6 ± 3.4 or 19 ± 13 μg g^{-1}, dry, fresh and lipid weight, respectively) reflecting chronic bioaccumulation from sewage-industrial particulates. On a temporal basis, lipid normalized PCBs concentrations peaked by the end of 2001–2002 coincident with the rainiest period over the last four decades and shortly after PCB prohibition in the country, reflecting massive discharges to the coastal ecosystem. PCB composition in fish muscles show a prevailing contribution of hexachlorobiphenyls ($35 \pm 4.2\%$), followed by hepta ($23 \pm 3.0\%$), penta ($20 \pm 3.6\%$), tri-tetra ($16 \pm 4.8\%$) and minor proportions of octa-decachlorobiphenyls ($5.7 \pm 3.1\%$) similar to an Aroclor 1242–1254–1260 1:2:4 mixture. During 2001–2002 maxima fish showed an enrichment in tri-tetrachlorobiphenyls (~1242–1254–1260 1:1:1 mixture) denoting a fresher signature. Fish/settling material lipid-organic carbon accumulation factors (BSAFs: 2.4–46, average: 21 ± 10) plotted against k_{ow} showed a parabolic trend ($BSAFs = -0.38 \log k_{ow}^2 + 5.16 \log k_{ow} - 15.85$; $R^2 = 0.46$) maximizing at hexa, hepta and octachlorobiphenyl 203 with reduced bioaccumulation of a few hepta (170, 191) and most octa-decachlorobihenyls suggesting limited intestinal absorption.

Keywords: Sewage detritus; PCBs; Fish; Río de la Plata

1. Introduction

Sewage-derived organic matter is an important energy subsidy for aquatic food webs as well as prevailing contamination pathway for specialized detritus feeding organisms (deBruyn et al., 2003). Previous studies in the Río de la Plata designed to evaluate the impact of crude effluent discharges in the coastal ecosystem demonstrated the massive load of PCBs, aliphatic and aromatic hydrocarbons to bottom sediments of Metropolitan Buenos Aires coast (Colombo et al., 2005a,b, 2006) which constitute the feeding ground of a specialized detritivore fish: the Sábalo (*Prochilodus lineatus*). While optimizing the diet searching for organic-rich detritus, this fish feeds directly on flocculent matter deposited close to sewage and industrial outfalls which leads to a rapid accumulation of fat and associated organic pollutants such as hydrocarbons and PCBs (Colombo et al., 2000).

In a previous paper, data on the temporal variation and bioaccumulation dynamics of aliphatic hydrocarbons in Sábalos from Metropolitan Buenos Aires coast were reported (Colombo et al., in press). In this paper, the study of bioaccumulation of anthropogenic contaminants in this detritivore fish is continued analyzing individual PCB

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congeners in fish and settling particles. The temporal variability of PCB concentrations, the composition and individual congener bioaccumulation are discussed and correlated to physico-chemical properties.

2. Methods

Sampling methods and general analytical procedures have been previously described (Colombo et al., in press). Briefly, fish samples were obtained every 3–4 months from local fishermen in the coastal area of Metropolitan Buenos Aires affected by crude industrial and sewage discharges (Fig. 1) Settling particles were collected 1.5 m below the surface upstream and downstream the main Buenos Aires sewer at Berazategui (Fig. 1) during 13 trap deployments covering spring, summer, autumn and winter 2002–2006.

Fish dorso-lateral muscle samples were pooled (2–5 individuals) according to the size and weight of the fish discriminating small (<1 kg), medium ($1 < x < 2.5$ kg), large ($2.5 < x < 3.5$ kg) and very large fish (3.5–4.5 kg). Fish with dissimilar morphometric characteristics were processed individually. A total of 79 samples corresponding to 150 fish were analyzed (46 pools and 33 individual fish). The muscles were processed in glass jars using different 250 watts-blenders for small, medium and large fish.

The homogenized muscle samples were split for the determination of water content (100 °C, 24 h) and for Soxhlet extraction of lipids with acetone, dichloromethane and petroleum ether (1:2:2) on tissue-sodium sulphate (1:3) slurries. The extracts were concentrated to constant weight under nitrogen to determine total lipids, partially cleaned with sulphuric acid, and fractionated by silica-gel chromatography. Individual PCB congeners were quantified by high resolution gas chromatography with ECD detection (Agilent 6890 N; Fig. 2). Quantification was performed by an external standard of 41 PCBs (IUPAC Nos.: 17, 18, 28, 31, 33, 44, 49, 52, 70, 74, 82, 87, 95, 99, 101, 105, 110, 118, 128, 132, 138, 149, 151, 153, 156, 158, 169, 170, 171, 177, 180, 183, 187, 191, 194, 195, 199–201, 205, 206, 208, 209; C-QME-01, AccuStandard Inc.). Linear responses were obtained with 4-point calibration curves (2, 10, 50, and 250 pg μl^{-1}). PCBs 103 and 198 (Ultra) were added to control recovery yields. Blanks included in every 8 sample batch gave negligible results. Method detection limits for the trap material ranged from 50 to 150 pg g^{-1} depending on the congener and the amount of material available. Method precision evaluated through repeated analysis of certified reference material (cod liver oil; COD1588a, National Institute of Standards & Technology, Gaithersburg MD 20899, USA) averaged 87 ± 16% for 22 certified congeners.

3. Results and discussion

3.1. Total PCB concentrations

Table 1 presents individual PCB average concentrations in fish from 1999 to 2005 and in settling particulates collected in the sewer area. Fig. 3 shows the full range variability of total PCB lipid-based levels in fish muscles and the ratio of tri to hexachlorobiphenyls (31 + 28/153). Sábalos contain large amounts of PCBs ranging from 0.98 to 36 μg g^{-1} dry weight (mean: 11 ± 7.2 μg g^{-1} dw), 0.25–15 μg g^{-1} fresh weigh (4.6 ± 3.3 μg g^{-1} fw) and 4.8–65 μg g^{-1} lipids (19 ± 13 μg g^{-1} lip). The average fresh weight PCB concentration in Sábalos is well above the 2 μg g^{-1} fw USFDA tolerance level for human consumption (USFDA, 2006) with individual values as high as 15 μg g^{-1}, indicating that consumption of this fish represents severe risk for human health. Considering the average

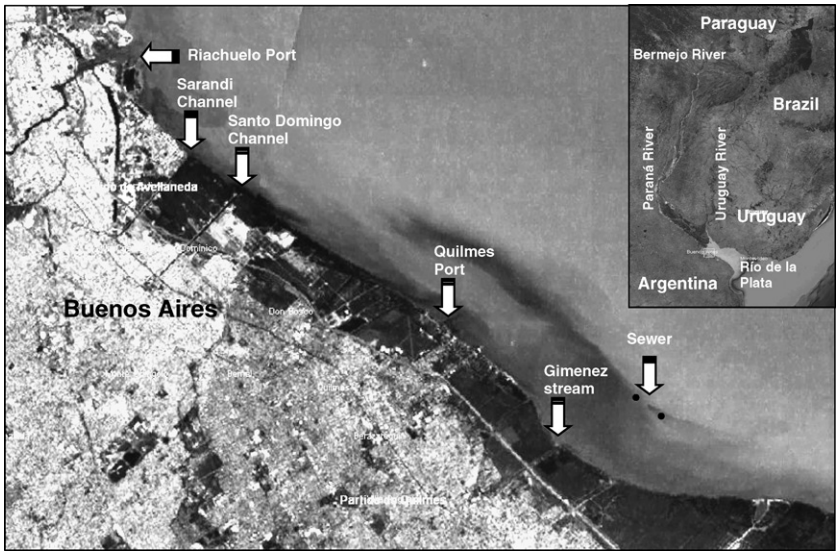

Fig. 1. Study area and sampling sites in Metropolitan Buenos Aires coast (image from NASA World Win). The white arrows indicate major sources of pollutants. The black points close to the sewer show the position of sediment traps.

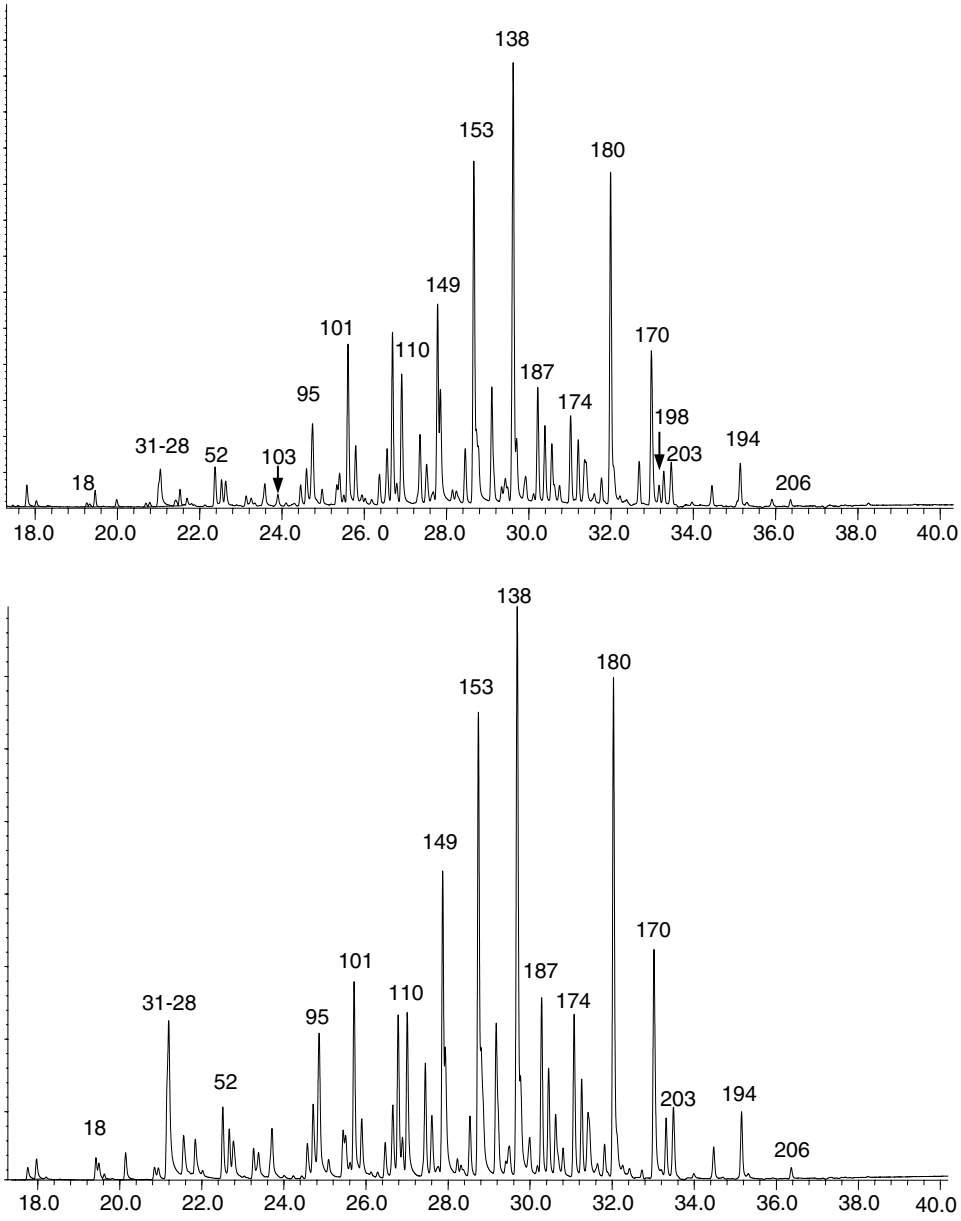

Fig. 2. PCB chromatograms in muscle of a small (a: 1.9 kg, 26% lipids, 0.7 μg g^{-1} fw PCBs) and large Sábalo (b: 3.6 kg, 68% lipids, 7.1 μg g^{-1} fw PCBs). Note the higher proportion of lower chlorinated PCBs (i.e. 31–28) in more polluted fish.

PCB concentration of Sábalos (4.2 ± 2.0 μg g^{-1} fw) and the reference dose of PCBs (2×10^{-5} mg kg^{-1} d^{-1}; EPA IRIS, 2007), the consumption limit of fish for a 70 kg person would be 0.33 g day^{-1} or 10 g $month^{-1}$, i.e. not recommended for consumption. These high PCB levels forced the closure of the Sábalo fishery in the Río de la Plata in 2001 by the Buenos Aires State government. However, due to its reproductive migrations this species transports the pollutant load upstream through the Paraná River potentially affecting remote human populations and natural predators such as the shovelnose catfish (*Pseudoplatystoma* sp.) and the dorado (*Salminus maxillosus*).

The concentrations of PCBs in the Sábalo are among the highest reported for fish, only comparable to those found in polluted Great Lakes and Hudson River fish, i.e. 4.3 μg g^{-1} fw in Lake Ontario Salmonids (Oliver and Niimi, 1988); 0.9–10.6 μg g^{-1} fw in Lake Trout from Great Lakes (Rasmussen et al., 1990); 0.04–5.6 μg g^{-1} fw in Lake Trout and Carp from Great Lakes (Kannan et al., 2000); 1.3–1.9 μg g^{-1} fw in salmonids from Lake Michigan (Jackson et al., 2001) and 1.2–4.0 μg g^{-1} fw in Hudson River Stripped Bass (Ashley et al., 2000). PCB concentrations in fish from less impacted freshwater and coastal marine environments are usually 1 to >3 orders of magnitude lower: e.g. 0.001–0.095 μg g^{-1} fw in Carp and Barbel from the Ebro River (Lavado et al., 2006); 0.002–0.07 μg g^{-1} fw in Largemouth bass from the Gulf of Mexico (Lewis et al., 2002); 0.06–0.3 μg g^{-1} fw in Black crappie, Carp and Smallmouth from Portland Harbor (Sethajintanin et al., 2004); 0.2–0.4 μg g^{-1} fw in Bluefin tuna and Swordfish

Table 1
Annually-averaged PCB concentrations (μg g^{-1} dw) in Sábalo and trap material

	1999	2000	2001	2002	2003	2004	2005	Traps
Fish muscle								
18	0.01	0.01	0.05	0.10	0.08	0.02	0.01	0.0006
17–15	0.02	0.00	0.06	0.06	0.03	0.01	0.01	0.0003
31	0.37	0.20	0.59	1.26	0.96	0.60	0.19	0.0024
33–53–20	0.02	0.02	0.23	0.38	0.19	0.08	0.02	0.0005
52	0.22	0.20	0.36	0.55	0.41	0.27	0.11	0.0023
49	0.15	0.15	0.18	0.21	0.22	0.15	0.06	0.0009
44	0.03	0.01	0.08	0.09	0.08	0.04	0.02	0.0011
74	0.10	0.09	0.18	0.28	0.18	0.14	0.06	0.0007
70	0.16	0.15	0.30	0.57	0.29	0.20	0.09	0.0012
95–66	0.29	0.19	0.30	0.73	0.60	0.33	0.14	0.0033
101–90	0.57	0.43	0.45	0.93	0.89	0.61	0.28	0.0044
99	0.24	0.23	0.24	0.30	0.28	0.18	0.08	0.0016
87–115	0.23	0.05	0.42	0.24	0.20	0.14	0.07	0.0014
110–77	0.24	0.21	0.36	0.58	0.47	0.32	0.15	0.0039
151	0.14	0.12	0.25	0.45	0.34	0.24	0.12	0.0019
149–123	0.37	0.36	0.50	1.01	1.11	0.75	0.31	0.0055
118	0.32	0.24	0.29	0.46	0.51	0.35	0.18	0.0031
153	0.68	0.55	0.59	1.50	2.10	1.55	0.67	0.0069
132–105	0.21	0.16	0.24	0.39	0.13	0.04	0.03	0.0032
138–160	0.67	0.71	0.71	1.67	1.77	1.31	0.57	0.0080
158	0.11	0.08	0.06	0.19	0.22	0.14	0.06	0.0012
178	0.11	0.12	0.12	0.21	0.18	0.10	0.07	0.0007
187	0.17	0.17	0.28	0.60	0.48	0.34	0.17	0.0019
183	0.11	0.12	0.19	0.41	0.29	0.21	0.11	0.0014
128–167	0.15	0.14	0.15	0.25	0.18	0.13	0.07	0.0009
174	0.15	0.19	0.22	0.56	0.44	0.23	0.14	0.0019
177	0.11	0.13	0.15	0.34	0.26	0.18	0.09	0.0012
171–156–202	0.17	0.15	0.23	0.40	0.24	0.19	0.10	0.0009
180	0.37	0.38	0.42	1.19	1.12	0.83	0.40	0.0039
191	0.01	0.02	0.04	0.10	0.04	0.02	0.01	0.0001
170–190	0.22	0.25	0.30	0.58	0.49	0.34	0.17	0.0026
199	0.04	0.07	0.14	0.27	0.15	0.10	0.05	0.0007
203–196	0.09	0.14	0.14	0.35	0.29	0.14	0.55	0.0008
189	–	–	–	0.03	0.03	0.01	0.01	0.00005
195–208	0.02	0.03	0.06	0.09	0.07	0.05	0.03	0.0005
194	0.06	0.08	0.13	0.18	0.14	0.10	0.05	0.0008
205	0.002	0.01	0.02	0.03	0.02	0.01	0.01	0.0002
206	0.01	0.01	0.02	0.03	0.02	0.02	0.01	0.0002
209	0.004	0.003	0.00	0.0001	0.005	0.004	0.003	0.0002
Total (μg g^{-1} dw)	6.90	6.12	8.73	17.43	15.49	10.47	5.24	0.073
SD	2.51	2.71	9.16	4.86	6.92	4.60	2.93	0.034
Total (μg g^{-1} fw)	3.09	2.72	3.73	7.08	6.88	3.77	2.09	
SD	1.40	1.45	3.93	1.98	3.95	2.15	1.54	
Total (μg g^{-1} lip)	8.48	8.74	17.04	31.93	22.49	20.20	12.22	0.90[a]
SD	1.42	2.32	19.73	12.50	8.21	4.97	2.87	0.42[a]

dw: dry weight; fw: fresh weight; lip: lipid based.
[a] Organic carbon based.

from the Ionian and Tyrrhenian Sea (Kannan et al., 2002) and 0.07–0.7 μg g^{-1} fw in Mullet from Taiwan (Chung-Te and Shian-Chee, 2006).

The high PCB levels of Sábalos reflect a chronic exposure through crude effluent discharges to the Rio de la Plata and point out the notable ability of this fish to bioaccumulate hydrophobic substances from sewage-industrial particulates. As a specialized detritivore, the energy benefit of this feeding strategy is based on the significant organic enrichment of anthropogenic particles which allow an enhanced growth and accumulation of fat and associated pollutants in the Río de la Plata compared to the Northern basin where the diet is composed of organic-poor vegetal detritus (Speranza and Colombo, submitted).

3.2. Temporal variability

Annually-averaged lipid-normalized total PCB concentrations range from 4.8 to 65 μg g^{-1}, comparable to previous reports from fish collected in 1996 (2.2–28 μg g^{-1} lipids; Colombo et al., 2000). This reflects a long-term chronic PCB load to the estuary through industrial-sewer discharges which are used as feeding resource by the Sábalo. A significant amount of variability is observed in

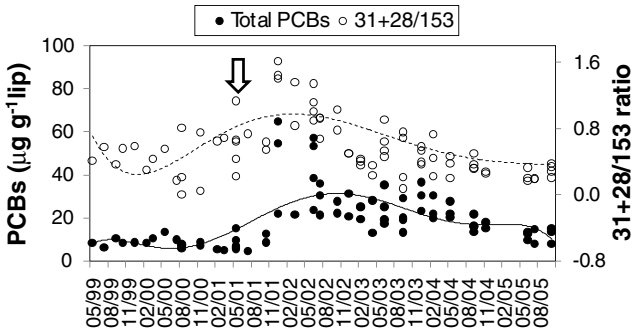

Fig. 3. Full range variability of lipid-normalized total PCB concentrations and tri/hexachlorobiphenyl ratios (31 + 28/153) in fish muscle. The vertical arrow indicate the date of PCB use prohibition in Argentina.

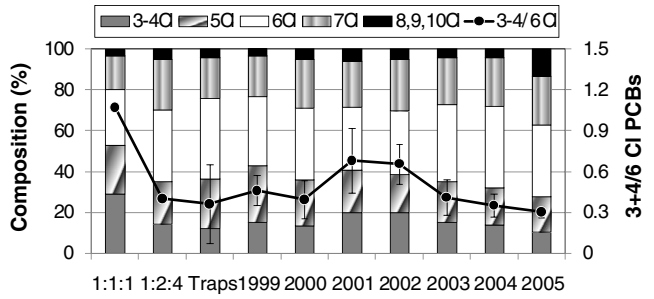

Fig. 4. Annually-averaged PCB composition and tri + tetra/hexachlorobiphenyl ratios in Sábalos compared to Aroclor 1242/54/60 mixtures (1:1:1 and 1:2:4) and trap material.

the data, i.e. 65–72% for the 1999–2005 dry, wet or lipid based grand mean relative standard deviations and 45–105% for the annual fresh weight variability. The variability of fresh weight concentrations include a small contribution from differences in fish size as indicate the poor relationship with weight (PCB fw = 1.52 weight (kg) – 0.17; $R^2 = 0.22$). This reflect the relatively uniform morphometric parameters of Sabalos analyzed (average standard length = 46 ± 5.3 cm; weight = 3.0 ± 1.1 kg; lipid contents: 59 ± 20% dw). Lipid normalized concentrations have no relationship with weight ($R^2 < 0.01$) but display large between-year fluctuations (Fig. 3). On a temporal basis, lipid normalized PCB concentrations in fish muscles peaked by the end of 2001 and beginning of 2002. This has been also observed for petrogenic aliphatic hydrocarbons and attributed to a more efficient wash out of residues from polluted Metropolitan Buenos Aires channels and harbors during 2001 which was the rainiest year over the last four decades (Colombo et al., in press). Thus, PCB data appear to confirm an enhanced exposition resulting from massive discharges of anthropogenic residues in wet relative to less impacted dry years. However, relative to the 1999-mid 2001 baseline, PCBs increase by a factor of 4.6 (8.4 ± 2.7 to 39 ± 16 μg g^{-1} lipids) as compared to ~2 times for aliphatic hydrocarbons (273 ± 87 to 501 ± 129 μg g^{-1} lipids; Colombo et al., in press). Since both pollutants have different sources, i.e. petrogenic inputs and transformer oils (Colombo et al., 2005a), these results suggest that overlapped to the rain washout effect, increased discharges of PCBs due to illegal dumping are apparent by the end of 2001 which coincides with the year of PCB phase out in Argentina. As has been also observed for aliphatic hydrocarbons, the composition of PCB in the fish indicates fresher signatures during the 2001–2002 maxima (see the 31 + 28/153 ratio in Fig. 3) compatible with a pulse of less degraded contaminants.

3.3. PCB composition

Fig. 4 presents the annually-averaged composition of PCBs in Sábalo muscles discriminated by homolog groups in tri-tetrachlorobiphenyls (3–4 Cl), penta (5 Cl), hexa (6 Cl), hepta (7 Cl) and octa to decachlorobiphenyls (8–10 Cl). The general composition of PCBs in Sábalos shows a dominant contribution of 6 Cl congeners (35 ± 4.2%) followed by 7 Cl (23 ± 3.0%), 5 Cl (20 ± 3.6%), 3–4 Cl (16 ± 4.8%) and 8–10 Cl PCBs (5.7 ± 3.1%). This general PCB pattern is comparable to the average trap composition and to an Aroclor 1242/1254/1260 1:2:4 mixture (Fig. 4). The significant proportion of lower chlorinated congeners suggests a relatively fresh signature (see Fig. 2) as has been also observed for aliphatic hydrocarbons which are more susceptible to degradation than PCBs. This reflects an efficient transfer of the trap signal to the fish which is favored by Sábalo's strategy of feeding on recently-decanted anthropogenic matter close to the sources. PCB signatures in underlying sediments suffer a consistent weathering with gradual loss of lower chlorinated congeners with distance offshore (Colombo et al., 2005a) providing further support to this interpretation.

On a temporal basis, PCB composition presents an enrichment of lower chlorinated congeners during the 2001–2002 maxima (Fig. 4). During both years the proportion of 3–4 Cl PCBs attains 20% as compared to 10–16% in the other years. This pattern is emphasized in fish with the highest PCB concentrations from December 2001 to July 2002 (3–4 Cl PCBs: 23 ± 3.5%). The tri/hexachlorobiphenyl ratio 31 + 28/153 captures this seasonal trend maximizing at 1.1 ± 0.3 from December 2001 to July 2002 over a general background of 0.45 ± 0.24 (Fig. 3). Similarly, the relation of all 3–4 to 6 Cl PCBs denotes a significant increase during the PCB maxima (0.8 ± 0.2; Fig. 4). A principal component analysis carried out with the relative abundance of the 5 homolog groups in all individual fish samples, settling particles and Aroclor mixtures shows the shift from prevailing 1254–1260 to the less chlorinated 1242 formulation in 2001–2002 (Fig. 5).

The PCA accounts for 82% of the total variability, 53% corresponds to the first component which is determined by 3–4 Cl (positive) and 6–7 Cl PCBs (negative; i.e. Aroclors 1242 vs. 1260) and 29% to the second with a prevailing contribution of 5 Cl (positive) and 7 Cl, 8–10 Cl PCBs (negative; i.e. Aroclors 1254 vs. 1260). Most fish samples and the traps spread between 5 Cl, 6 Cl, 7 Cl and 8–10 Cl PCBs

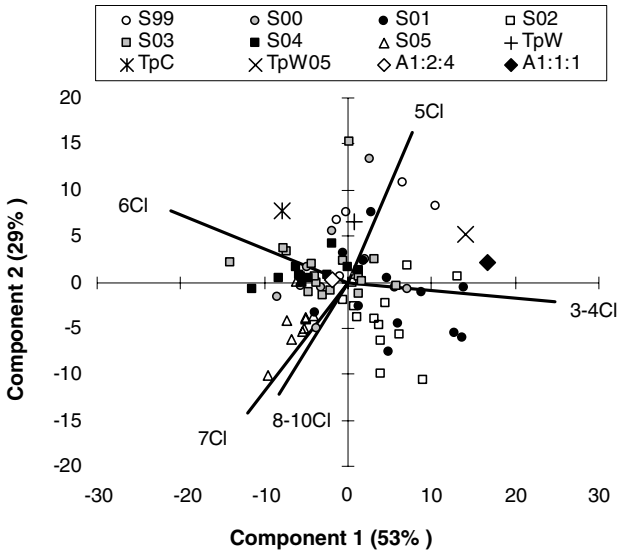

Fig. 5. Principal component analysis performed with the relative abundance of tri-tetra (3–4Cl), penta (5Cl), hexa (6Cl), hepta (7Cl), octadecachlorobiphenyls (8–10Cl) in individual fish samples collected annually from 1999 to 2005 (S99, S00, S01, S02, S03, S04, S05), settling material from warm (TpW), cold (TpC) periods and from December 2005 (TpW05) and Aroclor 1242/1254/1260 formulations (A1:2:4 and A1:1:1).

around the 1242/54/60 1:2:4 Aroclor mixture. Only Sábalos from 2001 and 2002 are significantly shifted with a higher contribution of 3–4 Cl PCBs closer to the 1:1:1 Aroclor mixture. The average composition of traps collected in warm-wet months (TpW: October–March) is also shifted to the right relative to the cold-dry average (TpC: April–September), reflecting a higher contribution of lower chlorinated congeners during the rainy season. This enrichment in 3–4 Cl and 5 Cl PCBs is exacerbated in traps collected in December–February 2005 (TpW05) which are very close to the 1:1:1 Aroclor mixture and to most polluted 2001–2002 fish. Considering the different time frame represented by fish and trap data (i.e. long-term integration vs. episodic events), these results are very consistent supporting an enhanced exposition to lower chlorinated PCBs during high discharge events. A similar trend was observed for aliphatic hydrocarbons which showed fresher signatures with higher C_{17}/pristane and C_{18}/phytane ratios during the 2001–2002 maxima (Colombo et al., in press).

3.4. Fish-settling particle bioaccumulation factors

The bioaccumulation dynamics of individual PCBs in this detritivorous fish was evaluated through the calculation of biota sediment accumulation factors (BSAFs) using the average concentration of each congener in fish muscle and the trap material on a dry weight and lipid–organic carbon basis (average fish lipids = 59 ± 20%; settling material organic carbon = 8.1 ± 5.8%).

Considering average PCB levels in the trap material the dry weight-based mean BSAFs is 147±68 ($\log = 2.11 \pm 0.25$) whereas the lipid-organic carbon BSAF is 21 ± 10 ($\log = 1.26 \pm 0.25$), an order of magnitude higher than the expected 1–2 value for equilibrium partitioning between body lipids and sediment organic carbon (Lee, 1992; Morrison et al., 1996). Lipid-organic carbon BSAFs of aliphatic hydrocarbons, in contrast, are lower than equilibrium values (0.41 ± 0.28; Colombo et al., in press). The increased bioaccumulation of PCBs in the Sábalo is probably related to a gastrointestinal magnification process during organic matter digestion (Gobas et al., 1999), enhanced by the higher persistence and more favorable molecular shape of PCBs during intestinal absorption compared to superhydrophobic ($\log k_{ow} = 9.2$–12.6) linear $C_{17–25}$ alkanes which showed very low BSAFs (Colombo et al., in press).

The episodic sampling of highly variable discharge events of settling material (Colombo et al., submitted) introduce some uncertainty in the dry-weight BSAFs but not in lipid-organic carbon based calculations. For example, if the organic and PCB-enriched traps (i.e. 20% organic carbon and 151 ng PCBs g^{-1} dw) collected in December 2005 are considered, the dry-weight based BSAFs are reduced by half (66 ± 44), but lipid-organic carbon values remain very similar (23 ± 15). Fig. 6 present a log–log plot of lipid-organic carbon BSAFs calculated with the organic-enriched December 2005 traps which presented more complete congener detections, vs. octanol–water partition coefficients (k_{ow}) of PCBs (Hawker and Connell, 1988). The BSAFs–k_{ow} relationship shows a parabolic pattern maximizing at highly hydrophobic ($k_{ow} = 6.9$–7.7), hexa (153, 128), hepta (178, 187, 183, 174, 180, 189) and octachlorobiphenyls (203) with a rapid decrease for superhydrophobic 8, 9 and 10 Cl PCBs, suggesting a reduction of intestinal assimilation for higher chlorinated congeners, e.g. steric hindrance or lower membrane permeability. Hexachlorobiphenyl 132 also exhibit a reduced bioaccumulation possibly reflecting its biotransformation as has been indicated by the higher depuration rates and lower half-lives of this congener in fish (Buckman et al., 2006).

Parabolic relationships of bioaccumulation factors with k_{ow} (BAFs declining at $\log k_{ow} \sim 6.7$–7.5) are well documented in the literature (e.g. Connell, 1988; Bremle et al., 1995; Morrison et al., 1996; Meylan et al., 1999; Zhou

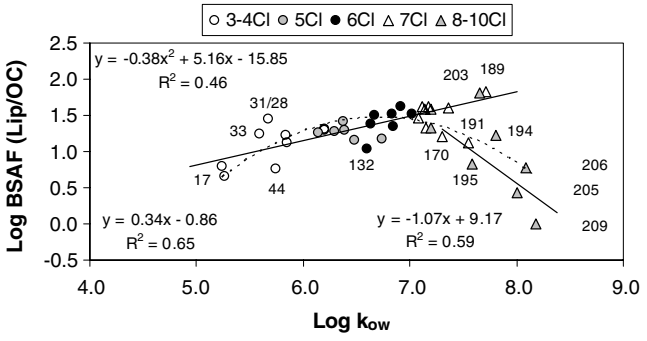

Fig. 6. Fish/trap lipid-organic carbon accumulation factors (BSAFs) vs. k_{ow} of individual PCBs. The quadratic regression for all data as well as the positive and negative linear regressions fitted are shown.

and Wong, 2000; Burkhard et al., 2004). Similar curvilinear trends have been also reported for the half-life vs. k_{ow} relationship of individual PCB congeners in fish (half-live declining at $\log k_{ow} > 7$; Buckman et al., 2006). The Sábalo/settling particle BSAFs of aliphatic hydrocarbons also displayed a bell-shaped curve maximizing at n-C_{15} alkane and isoprenoids ($\log k_{ow} = 7.71$–8.96), with reduced uptake of >n-C_{17} alkanes (Colombo et al., in press). The quadratic regression of the Sábalo BSAFs–k_{ow} relationship of PCBs ($\log \text{BAFs} = -0.38\ \log k_{ow}{}^2 + 5.16\ \log k_{ow}\ -15.85$; $R^2 = 0.46$; Fig. 6) is comparable to that reported for the membrane/water partitioning ($\log k_{mw}$) vs. $\log k_{ow}$ relationship of PCBs in C_{20} diacylphosphatidylcholine vesicles ($\log k_{mw} = -0.38\ \log k_{ow}{}^2 + 5.67\ \log k_{ow}\ -15.53$; $R^2 = 0.96$; Dulfer and Govers, 1995), supporting the key role of intestinal membrane-water partitioning in the uptake process.

Alternatively to the quadratic fit, the data could be modeled by two linear regressions between BSAFs and k_{ow}, the first positive and the second negative. Excluding two hepta (170, 191), three octa (194, 195, and 205), nona (206) and decachlorobiphenyls (209) from the first regression, the positive BSAF-k_{ow} relationship ($\log \text{BAFs} = 0.34\ \log k_{ow} -0.86$; $R^2 = 0.65$; Fig. 6) indicates increased bioaccumulation up to $\log k_{ow} \sim 7.7$. The second negative regression between the excluded 7–10 Cl PCBs and k_{ow} ($\log \text{BSAFs} = -1.07\ \log k_{ow} + 9.17$; $R^2 = 0.59$; Fig. 6) is similar to that reported by Meylan et al. (1999) for biota-water bioconcentration factor in the 7–10.5 $\log k_{ow}$ range ($\log \text{BCF} = -1.37\ \log k_{ow} + 14.4$), reflecting a reduced uptake of superhydrophobic PCBs.

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