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Distribution Patterns of Polychlorinated Biphenyl Congeners in Water, Sediment and Biota from Midway Atoll (North Pacific Ocean)

BRUCE HOPE*, SUSAN SCATOLINI, ERIC TITUS† and JEFF COTTER
Ogden Environmental and Energy Services Company, Inc., 680 Iwilei Road, Suite 660, Honolulu, HI 96817, USA

To increase our understanding of critical pathways of polychlorinated biphenyl (PCB) transfer from abiotic media into marine organisms, this study quantified 20 PCB congeners in surface water, sediment and tissues of marine biota (macrophytes, snails, urchins, bivalves, sea cucumbers, fishes) taken from Midway Atoll. PCBs 138, 153, 170, 180 and 187 were the most abundant congeners in all samples analysed. Distribution of PCB congeners was shifted in favour of higher (hexa- and above) chlorinated congeners in all species; only aquatic macrophytes displayed significant bioaccumulation of lower (tri- and tetra-) chlorinated congeners. Evidence is presented for the differential metabolism of congeners by marine species. Non-*ortho* substituted congeners (PCBs 77, 126) with elevated toxic potency were not present at significant levels in the sampled species. Certain mono-*ortho* congeners (PCBs 105, 118), implicated in marine mammal toxicity, comprised only ≈4.5% of total congener load in prey for piscivorous birds and marine mammals. © **1997 Elsevier Science Ltd**

Midway Atoll is in the North Pacific Ocean 1100 miles northwest of Pearl Harbour, Hawaii, approximately 178°W longitude, 28°N latitude (Fig. 1). The atoll consists of two main islands, Sand and Eastern, surrounded by a fringing coral reef. Although heavily modified by human activity over the previous 90 years, the islands provide breeding and feeding habitat for more than one million migratory seabirds; a total of 45 migratory bird species have been recorded at the atoll. Midway also provides habitat for terrestrial and marine mammals, sea turtles and other reptiles, and a rich diversity of reef fishes and invertebrates. Pacific green sea turtles (*Chelornia mydas agasszi*) and the protected spinner dolphin (*Stenella longirostris*) both frequent the lagoon. US Federally-listed endangered Hawaiian monk seals (*Monachus schauinslandi*) are present in small numbers and known to pup on the atoll's beaches. The atoll is considered critical habitat for this species. In 1988, the atoll was designated as an overlay National Wildlife Refuge, under the full control of the US Fish and Wildlife Service in May 1996.

As a result of historical use in industrialized countries and current use in the rapidly industrializing countries of low-latitude tropical and subtropical Asia and Oceania, PCBs have entered and are still being eased into the global environment (Iwata *et al.*, 1994; Tanabe *et al.*, 1994). Atmospheric transport and ocean circulation are primary mechanisms for circum-global dispersion of these organochlorine compounds (Tanabe and Tatsukawa, 1986). Their wide dispersal, along with their great capacity to bioaccumulate, have allowed PCBs to attain 'steady-state' in some ecological and biological systems (Bauman and Whittle, 1988; Falandysz *et al.*, 1994; Harris *et al.*, 1993). They are currently recognized as ubiquitous in marine, estuarine and freshwater food chains, with the largest biological concentrations occurring in fish-eating aquatic seabirds and marine mammals, making them an ecotoxicological concern to these species (Tanabe *et al.*, 1994).

Air samples collected in the North Pacific Ocean, along a track that passed immediately south of Midway Atoll, contained ΣPCB concentrations in the range of 0.012–0.390 ng m^{-3}, with a mean of 0.130 ng m^{-3} (Iwata *et al.*, 1993). Iwata *et al.* (1993, 1994) also investigated PCB contamination in seawater along the route followed by the Kuroshio current from the low-latitude rapidly industrializing regions, where PCBs are still used, past the Northwestern Hawaiian Islands (including Midway Atoll), and northward into the Gulf of Alaska and the Bering Sea. Seawater samples taken along this route contained ΣPCB levels ranging from 0.0091 to 0.063 μg l^{-1}, with a mean of 0.024 μg l^{-1}.

*Current address: Oregon Department of Environmental Quality, Waste Management and Cleanup Division, P.O. Box 8099, Portland, OR 97207-8099, USA.

†Current address: Ogden Environmental and Energy Services Company, Inc., 343 Sansome Street, Suite 700, San Francisco, CA 94104, USA.

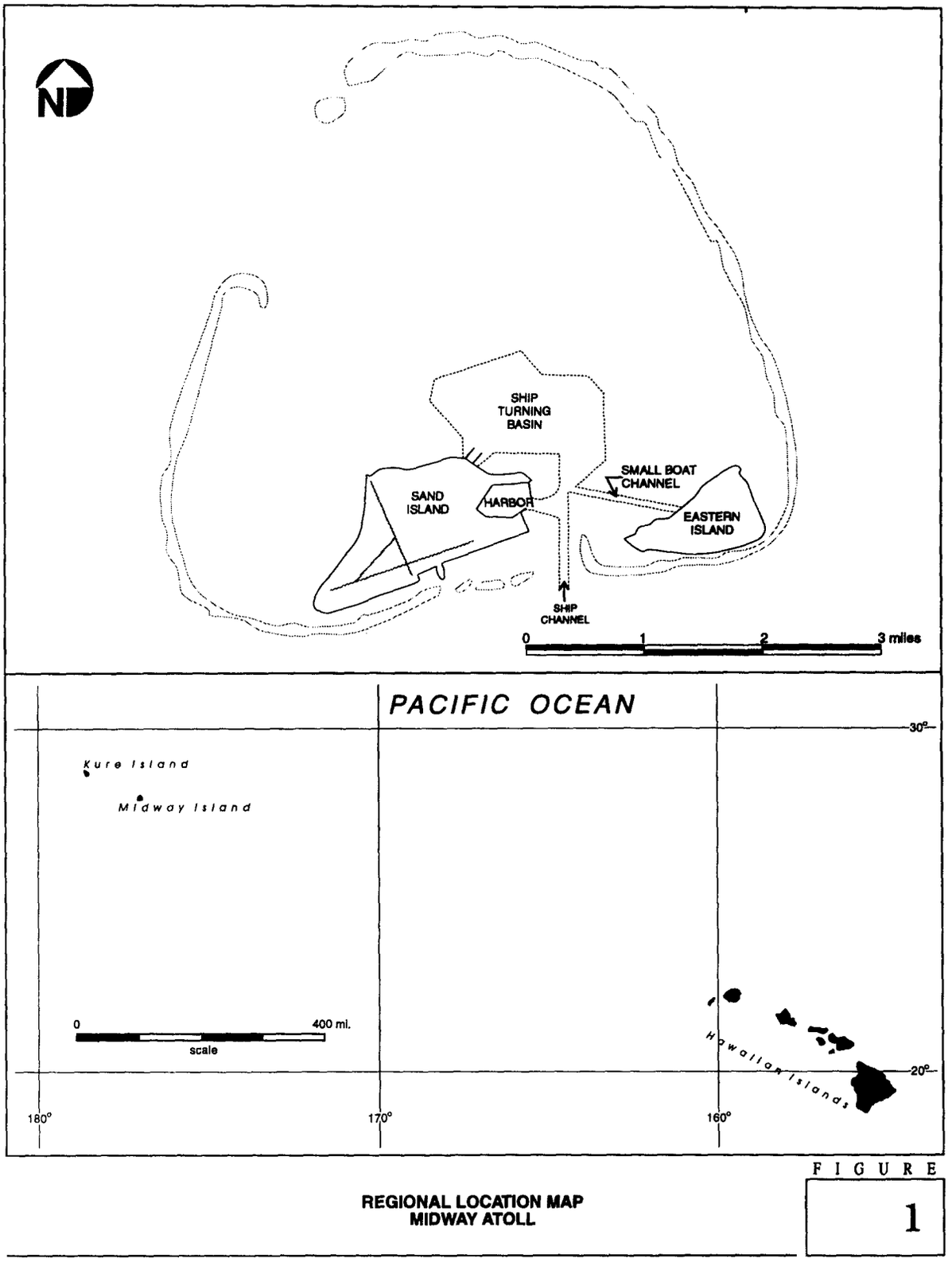


Fig. 1 Regional location map Midway Atoll.

These findings suggest that Midway Atoll may be receiving ambient environmental PCB exposures from atmospheric deposition and ocean surface water circulation.

To increase our understanding of critical pathways of PCB transfer from sediment or water into lower trophic level organisms and, subsequently, to higher order consumers, particularly for the North Pacific Ocean, this study measured the concentration and distribution of 20 PCB congeners (PCBs 8, 18, 28, 44, 52, 66, 77, 101, 105, 118, 126, 128, 138, 153, 170, 180, 187, 195, 206 and 209 (congeners numbered according to Ballschmiter

and Zell (1980)) in marine sediments, surface waters, and tissues of 12 species of marine biota from near-shore waters at Midway Atoll. PCB congeners 28, 52, 101, 118, 138, 153 and 180 are among those recommended by the International Council for Exploration of the Seas (ICES) for assessing marine pollution by PCBs (Duinker *et al.*, 1988; Porte and Albaigés, 1993).

PCBs are lipophilic and thus are expected to associate with benthic and suspended sediments, phytoplankton, zooplankton and natural surface oil films. Assimilation of PCBs into aquatic food chains generally begins with these sediments, planktons, or films being incidentally ingested by fish and invertebrates, or taken up by aquatic macrophytes. Therefore, species analysed in this study included two macroalgaes (*Dictyota acutiloba*; *Giffordia breviarticulata*), a sea grass (*Halophila ovalis*), a bivalve mollusk (*Chama iostoma*), a sea urchin (*Echinometra mathaei*), a snail (*Nerita picea*), two holothurians (*Bohadschia obesus*; *Holothuria atra*), and four fishes: striped convict tang (*Acanthurus triostegus*), goatfish (*Mulloidichthys flavolineatus*), Pacific gregory (*Stegastes fasciolatus*) and blacktail wrasse (*Thalassoma ballieui*). Based on limited life history information, these species were grouped into broad trophic levels as follows: primary producers (*D. acutiloba*; *G. breviarticulata*, *H. ovalis*); herbivores (*C. iostoma*, *E. mathaei*, *A. triostegus*, *S. fasciolatus*); omnivores (*B. obesus*; *H. atra*); and carnivores (*N. picea*, *M. flavolineatus*, *T. ballieui*).

In this paper, we report on: *1.* concentrations of individual PCB congener associated with marine biota near the base of the food chain in near-shore, North Pacific waters; *2.* distribution of PCB congeners in these marine biota as a function of trophic level and congener chemistry; *3.* patterns of PCB bioconcentration and bioaccumulation in these biota with respect to surface water and sediments; *4.* the influence of stereochemistry on each species capacity to metabolize specific congeners; and *5.* the distribution and potential toxicity of non-*ortho*-substituted (PCBs 77 and 126) and selected mono-*ortho*-substituted (PCBs 105 and 118) congeners.

Materials and Methods

Sample collection

Samples of marine sediment were collected from locations in nearshore areas around Sand and Eastern Islands using horizontal and vertical cores. Sediment, water and biota samples were collected subtidally. Horizontal sediment cores were collected at all stations, vertical sediment cores were only collected at a small subset of the stations. Aluminium core tubes with a diameter of 5 cm and a length of 60 cm were used to collect both core orientations. Horizontal cores were collected by pushing the tubes though the upper 6 inches of sediment and capping both ends. Vertical samples were collected with the use of a weighted slide hammer to pound the full depth of the tube into the sediment. The top of the vertical core was marked at the time of sampling. Each core was processed by removing the cap, decanting the water, measuring the head space, and cutting the tube at a point 25% below the sediment surface. The lower 75% of the sleeve (45 cm of sleeve depending on head space) was retained as the sample, the top 25% was discarded. Resulting horizontal and vertical samples were homogenized in decontaminated stainless steel bowls, placed in new, laboratory-certified clean glass jars, sealed, cooled to 4°C, and shipped to the laboratory for analysis of low level PCBs.

All sediment sleeves were decontaminated prior to use by washing in Alconox®, rinsing twice with bottled water, spraying with isopropyl alcohol and rinsing again. Teflon® sheets were placed over the ends of the sleeves to reduce the possibility of outside contamination. The sleeves with Teflon® were then capped, taped at the ends and placed on ice until homogenization. Seawater samples were also collected from locations in nearshore areas around Sand and Eastern Islands. Samples were collected by removing the container lid just below the water surface, filling a 2-l amber glass bottle, and resealing lids with the bottle still submerged. All glassware used for water samples were new laboratory-certified clean bottles provided by the analytical laboratory. Samples were then cooled to 4°C, and shipped off-atoll for low level PCB analyses.

A biological reconnaissance of the lagoon at Midway Atoll and most nearshore areas was completed prior to initiating sampling for biota. All habitats, substrate types and species were identified to the extent feasible, and appropriate species for collection identified. Organisms collected for tissue analysis were selected based on trophic level, specificity of location, and availability. The goal was to collect representatives from a wide range of trophic levels, especially species that may be prey for endangered Hawaiian monk seals and threatened green sea turtles. Fish species were collected using a spear; invertebrates and algae were collected by hand. Species collected were placed in new, food grade, resealable plastic storage bags, and frozen at -10°C until shipped to the laboratory for analysis. Due to the mass requirements for PCB analysis, samples were sometimes composites of several individuals, depending on wet wt of the organisms.

All samples were shipped on ice in coolers, with custody seals and chain of custody forms to track sample access. Temperature of samples was recorded by the laboratory upon receipt and samples greater than 6°C were qualified as estimated values. All holding times were met for all samples.

Sample preparation and analysis

The off-atoll laboratory performing the PCB analyses on seawater, sediment and biotic material was Arthur D. Little, Inc., (ADL) Cambridge, Massachusetts. Low-level PCB analysis was performed in accordance with the National Oceanic and Atmospheric Administra-

tion's Status and Trends Program Methods. In preparation for low level analyses for PCBs, seawater samples were spiked with a calibration surrogate (PCB 103), then extracted with methylene chloride in a separatory funnel. This extract was dried with sodium sulphate, concentrated, and an aliquot of the sample weighed, then analysed by gas chromatography-electron detection (GC-ECD) technique (ADL, 1994). Sediment samples were first spiked with surrogates, then extracted with a solution of methylene chloride/acetone (1:1 ratio) using a sonication/shaking technique, then dried with sodium sulphate, followed by concentration, centrifugation and weighing. Sediment sample extracts were analysed with a GC-ECD technique (ADL, 1994).

Tissue samples were homogenized, an aliquot removed, weighed and placed in a Teflon® jar, after which they were dried with sodium sulphate and spiked with surrogates. Samples were then serially extracted with methylene chloride, the solvent and matrix having been mixed through maceration; the extract was then concentrated for clean-up and fractionation. Total extractable (lipid) weight was calculated as part of the clean-up process. Tissue sample extracts were analysed using the GC-ECD technique employed for surface water and sediment samples (ADL, 1994).

Method detection limits were determined in accordance with US Environmental Protection Agency criteria (40 CFR 136, Appendix B). Congener-specific detection limits were 0.00022–0.003 $\mu g\ l^{-1}$ for seawater, 0.011–0.21 $\mu g\ kg^{-1}$ for sediments and tissues. The gas chromatograph was calibrated prior to PCB analyses, then daily or after every 10 samples analysed. Concentrations of all analytes were determined relative to that of the internal standard spiked just prior to analysis. Recovery of surrogates were also determined relative to the internal standard. Individual compounds were then identified by their retention time in the gas chromatograph, and the concentration of each analyte computed on the basis of sample volume or weight. Standard reference materials (SRMs) were used for precision and accuracy tests and performance evaluation samples were submitted to laboratory. All data were validated to US Navy Level D criteria.

Results and Discussion

Individual congener concentrations

The geometric mean and the 95th percentile upper and lower confidence intervals about the mean for concentrations of individual congeners in surface water, sediments, and each species (on a dry wt basis) are summarized in Table 1. Means and confidence intervals were calculated using log-transformed data to normalize residuals; non-detects were included in these calculations at one-half the detection limit. All congeners were present in marine sediments at mean concentrations from 0.03 to 0.20 $\mu g\ kg^{-1}$. Mean congener concentrations in surface water were 0.0003 to 0.001 $\mu g\ l^{-1}$, and generally 1–2 orders of magnitude lower than concentrations in sediments. Individual congener mean concentrations in biota were 0.02 $\mu g\ kg^{-1}$ in *H. atra* to 3300 $\mu g\ kg^{-1}$ in *Mulloidichthys*; with the majority, however, in the range of 0.1–50 $\mu g\ kg^{-1}$. Three species, *Bohadschia*, *Mulloidichthys*, and *Stegastes*, had elevated ($> 50\ \mu g\ kg^{-1}$) mean concentrations of several congeners, particularly hexa- and heptachlorobiphenyls. Interpretations based on data for *Nerita* and *Mulloidichthys* were made with caution owing to the high heterogeneity in these data as reflected in the broad confidence intervals.

Congener distribution patterns

Shifts in the proportion of each individual congener concentration relative to the sum of the mean concentration of all 20 congeners are represented graphically (Figs 2–5). For biota, total congener concentrations were calculated on a dry wt basis. In all samples, contributions to total PCB concentrations were not evenly distributed across the 20 congeners. In both marine sediments and surface water (Fig. 2), congeners making the proportionally largest individual contributions to total congener concentration were the hexa- and heptachlorobiphenyls PCBs 138, 153, 170, 180 and 187. These five collectively accounted for approximately 52% and 36% of total congener concentration in sediments and surface water, respectively. The deca-chlorinated congener PCB 209 comprised approximately 3–5% of the total. Lower chlorinated congeners (PCB 101 and below) generally made small ($< 10\%$) individual contributions to total loading. This observed skewing of congener distribution towards more highly chlorinated compounds is consistent with previously published findings indicating the preferential retention of these less volatile, more lipophilic compounds in aquatic systems (De Voogt *et al.*, 1990).

Major congeners detected in all biota samples (Figs 3–5) were very similar to those dominating the abiotic media: two hexa- (PCBs 138 and 153) and three heptachlorobiphenyls (PCBs 170, 180 and 187). These five collectively accounted for approximately 46–87% and 67–80% of the total congener concentration in invertebrates and fishes, respectively (Figs 4 and 5). These isomers have in common, chlorine atoms in positions 2,4,5- in one (PCB 187) or both rings (PCB 153 and 180), *para* positions on both biphenyl rings, and a lack of adjacent unsubstituted *meta* and *para* positions, making these congeners particularly recalcitrant to degradation by invertebrates or fishes (Bright *et al.*, 1995; Zell *et al.*, 1978). Borlakoglu *et al.* (1990) have also shown that these same congeners are not readily metabolized by vertebrates and tend to accumulate in the adipose tissue of seabirds. In aquatic macrophytes (Fig. 3), these five congeners collectively accounted for 43–50% of the total congener concentration. However, plant species exhibited higher proportions of lower

TABLE 1

Mean (dry wt) concentrations (with 95% confidence limits) of individual PCB congeners sampled in abiotic media and marine species from Midway Atoll.

	Abiotic media or species sampled							Species sampled						
Congener	Marine sediment μg kg^{-1}[a]	Surface water μg l^{-1}	*Dictyota acutiloba* μg kg^{-1}	*Giffordia breviarticulata* μg kg^{-1}	*Halophila ovalis* μg kg^{-1}	*Chama iostoma* μg kg^{-1}	*Echinometra mathaei* μg kg^{-1}	*Nerita picea* μg kg^{-1}	*Bohadschia obesus* μg kg^{-1}	*Holothuria atra* μg kg^{-1}	*Acanthurus triostegus* μg kg^{-1}	*Mulloidichthys flavolineauts* μg kg^{-1}	*Stegastes fasciolatus* μg kg^{-1}	*Thalassoma ballieui* μg kg^{-1}
8	0.040.03–0.05	0.00040.0003–0.0004	0.310.16–0.6	0.360.1–1.0	0.180.08–0.41	0.450.2–1.03	0.110.05–0.29	0.080.02–0.33	0.260.12–0.54	0.150.12–0.2	0.480.21–1.08	2.40[c]	0.730.21–2.57	0.210.08–0.52
18	0.080.06–0.09	0.00050.0005–0.0005	2.141.43–3.2	1.030.55–1.95	2.200.34–14.22	0.490.31–0.79	0.080.05–0.12	0.440–401108.75	0.460.14–1.47	0.200.15–0.26	0.440.38–0.52	3.07[c]	1.390.39–4.93	0.270.11–0.66
28	0.060.05–0.08	0.00100.0009–0.001	0.200.17–0.25	0.360.2–0.65	0.290.11–0.82	1.120.42–2.98	0.120.07–0.19	0.210–323.48	0.450.1–1.99	0.190.15–0.25	1.310.22–7.87	2.93[c]	3.290.44–24.4	0.260.1–0.66
44	0.030.02–0.04	0.00030.0003–0.0003	0.400.16–1.01	0.430.24–0.75	0.150.11–0.2	0.310.25–0.38	0.060.04–0.08	0.180–106.53	0.410.13–1.28	0.160.12–0.21	0.360.31–0.42	2.30[c]	2.610.38–17.86	0.220.09–0.54
52	0.040.03–0.05	0.00040.0004–0.0004	7.352.87–18.83	0.970.53–1.79	0.360.18–0.71	0.770.57–1.04	0.380.18–0.84	0.240.11–0.53	1.040.25–4.33	0.320.22–0.47	4.552.0–10.0	7.05[c]	7.651.06–54.95	0.900.02–36.14
66	0.030.02–0.03	0.00030.0003–0.0003	0.280.23–0.35	0.630.38–1.06	0.240.18–0.32	0.480.41–0.57	0.150.09–0.24	0.190.1–0.37	0.260.22–0.31	0.270.2–0.36	0.850.31–2.35	4.13[c]	2.440.63–9.43	0.360.2–1.0
77	0.060.04–0.08	0.00060.0005–0.0006	0.380.3–0.46	0.850.5–1.42	0.480.24–0.99	0.640.55–0.76	0.110.11–0.12	0.250.09–0.69	0.340.29–0.41	0.360.27–0.47	1.140.39–3.32	5.47[c]	2.190.6–8.03	0.420.11–1.61
101	0.060.04–0.1	0.00040.0004–0.0004	1.140.75–1.74	2.731.2–6.2	0.420.23–0.77	1.821.28–2.58	0.960.5–2.0	0.500.18–1.38	5.120.95–27.52	0.810.62–1.05	4.701.54–14.35	32.98[c]	28.013.55–221.05	1.360.23–8.01
105	0.030.02–0.04	0.00050.0005–0.0006	0.470.14–1.6	0.690.33–1.46	0.200.13–0.3	0.720.42–1.23	0.250.12–0.5	0.260–1737.42	2.891.03–8.06	0.240.15–0.39	1.760.36–8.55	7.82[c]	5.510.69–43.91	0.750.05–10.76
118	0.070.05–0.1	0.00030.0003–0.0004	0.800.4–1.6	2.511.46–4.34	0.770.6–1.0	1.871.22–2.86	0.670.39–1.16	0.700.18–2.72	7.701.73–34.36	0.940.72–1.22	7.181.62–31.86	30.66[c]	24.043.6–160.49	1.600.21–12.11
126	0.050.05–0.05	0.00050.0004–0.0005	0.310.25–0.38	0.690.41–1.14	0.260.19–0.34	0.530.45–0.62	0.090–0	0.200.08–0.52	0.280.24–0.34	0.290.22–0.38	0.650.56–0.76	4.52[c]	1.450.38–5.51	0.400.16–1
128	0.040.03–0.06	0.00030.0003–0.0003	0.520.31–0.89	1.140.68–1.9	0.330.24–0.45	0.770.6–0.99	0.280.16–0.47	0.160–18 439.84	0.580.31–1.1	0.360.19–0.7	2.090.84–5.21	13.21[c]	5.951.5–23.66	0.740.21–2.55
138	0.160.1–0.26	0.00060.0005–0.0007	6.384.16–9.8	3.350.78–14.35	1.300.69–2.47	7.264.04–13.03	1.890.62–5.78	0.610–53 785.97	43.384.51–417.13	1.610.66–3.92	17.373.31–91.16	111.98[c]	89.4011.28–708.32	4.030.32–50.32
153	0.150.09–0.27	0.00030.0002–0.0004	6.013.85–9.37	4.151.14–15.14	1.941.05–3.6	12.226.79–21.97	2.420.85–6.89	0.21[c]	59.064.81–725.18	0.740.18–3	24.290.92–641.72	105.74[c]	132.8610.93–1615.02	2.060–382.74
170	0.200.14–0.3	0.00170.0001–0.0026	1.671.07–2.59	0.690.09–5.41	0.310.13–0.73	3.411.65–7.08	0.190.1–0.38	0.280–11 517.5	2.070.26–16.11	0.230.14–0.38	5.120.13–201.17	40.64[c]	47.823.21–712.88	2.070.03–169.9
180	0.150.08–0.26	0.00070.0006–0.0009	4.222.57–6.92	2.690.49–14.7	0.460.08–2.75	8.534.05–17.96	0.960.49–1.88	2.190–4597.36	4.770.74–30.67	0.290.17–0.47	16.110.77–338.88	74.83[c]	142.2210.52–1922.19	5.820.07–483.3
187	0.180.11–0.28	0.00060.0005–0.0007	2.700.84–8.7	4.211.18–15.08	0.980.45–2.12	3.401.72–6.73	1.340.44–4.07	0.960–19 728.38	52.152–1358	1.520.37–6.35	11.660.66–207.04	48.73[c]	91.537.55–1109.22	3.850.1–155.93
195	0.060.04–0.09	0.00050.0005–0.0006	0.990.7–1.39	1.290.46–3.62	0.360.24–0.53	0.750.32–1.77	0.160.11–0.21	0.660.5–0.89	1.110.23–5.46	0.290.2–0.43	2.230.6–8.26	12.30[c]	20.181.59–256.75	0.470.01–29.81
206	0.050.03–0.08	0.00040.0003–0.0004	1.430.87–2.11	0.570.16–2.11	0.190.11–0.32	1.130.67–1.91	0.080.06–0.11	0.250–1327.93	0.990.2–5.03	0.270.2–0.36	2.850.4–20.48	13.56[c]	14.060.93–212.6	0.310–21.76
209	0.050.03–0.08	0.00050.0004–0.0006	6.922.04–23.51	0.280.13–0.61	0.110.06–0.19	0.880.3–2.59	0.090.05–0.19	0.220–4035.87	0.440.06–3.05	0.120.06–0.24	2.570.06–106.58	1.75[c]	1.770.35–8.87	0.210.08–0.52
n[b]	79	41	8	4	6	10	13	2	7	7	4	2	7	3

[a] Geometric mean (lower 95% confidence interval–upper 95% confidence interval), dry wt

[b] Number of samples.

[c] Broad 95% confidence interval (< 0.000001– > 1 000 000) due to heterogeneity in these data.

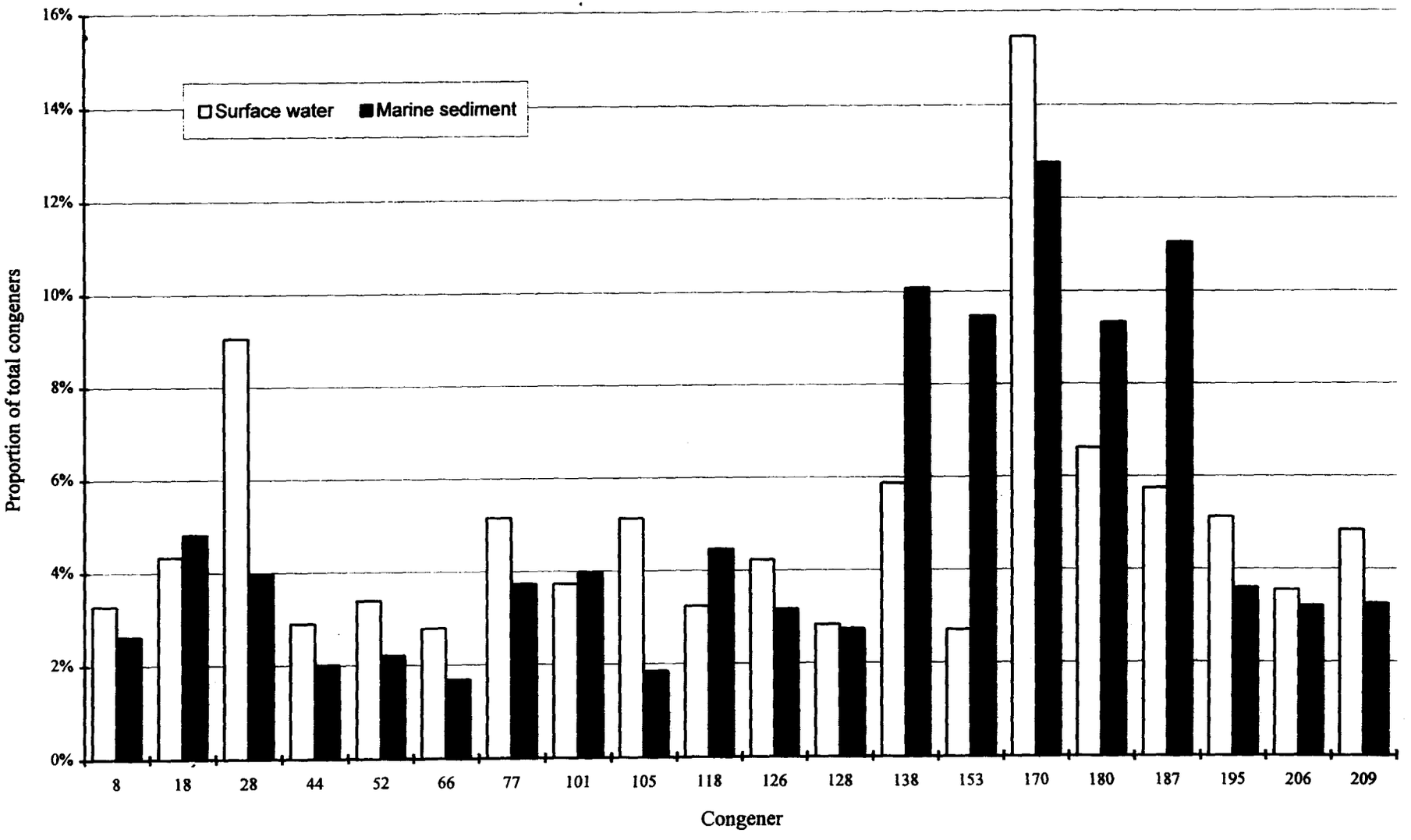


Fig. 2 Distribution of individual congeners in marine sediments and surface water.

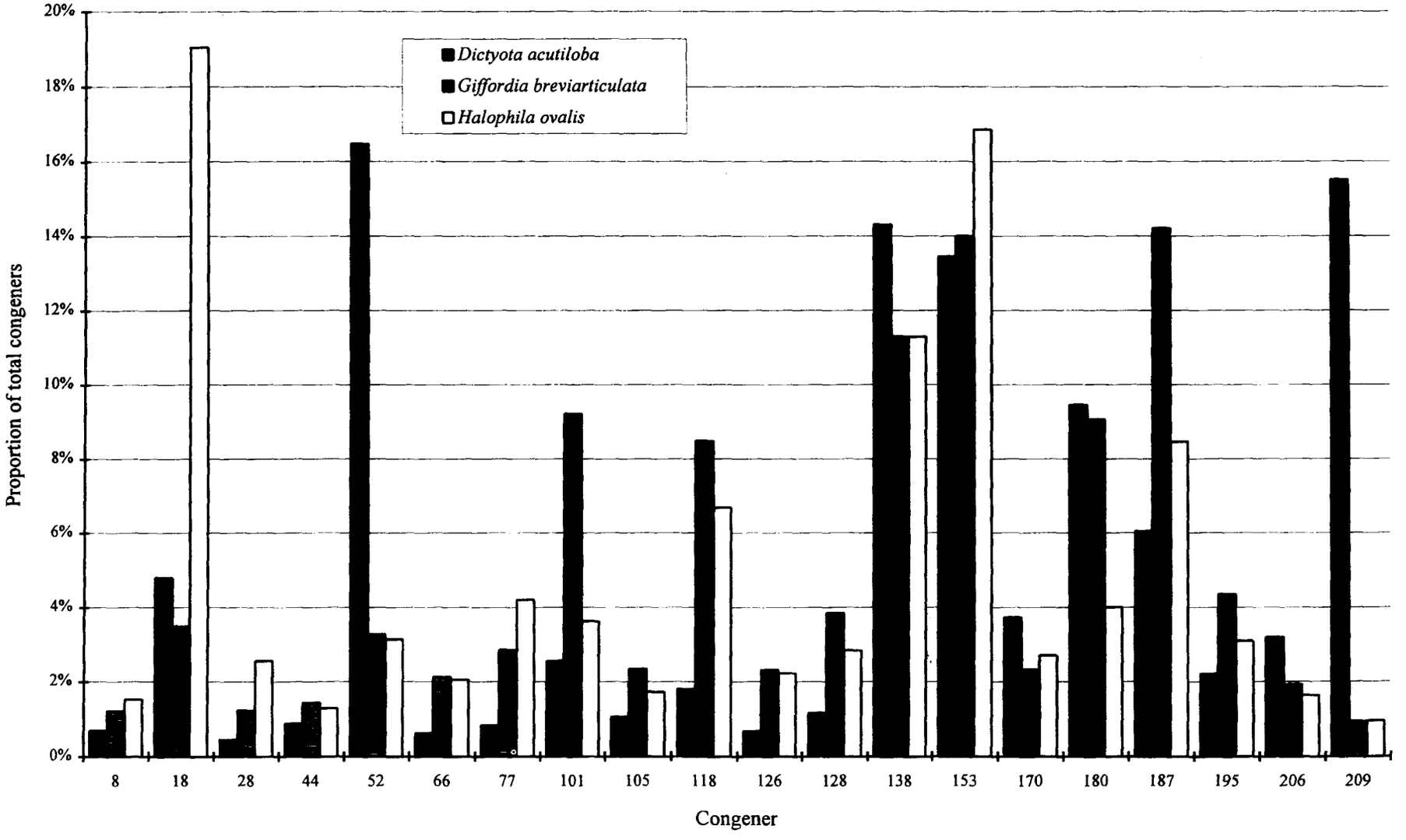


Fig. 3 Distribution of individual congeners in tissues of marine macrophytes.

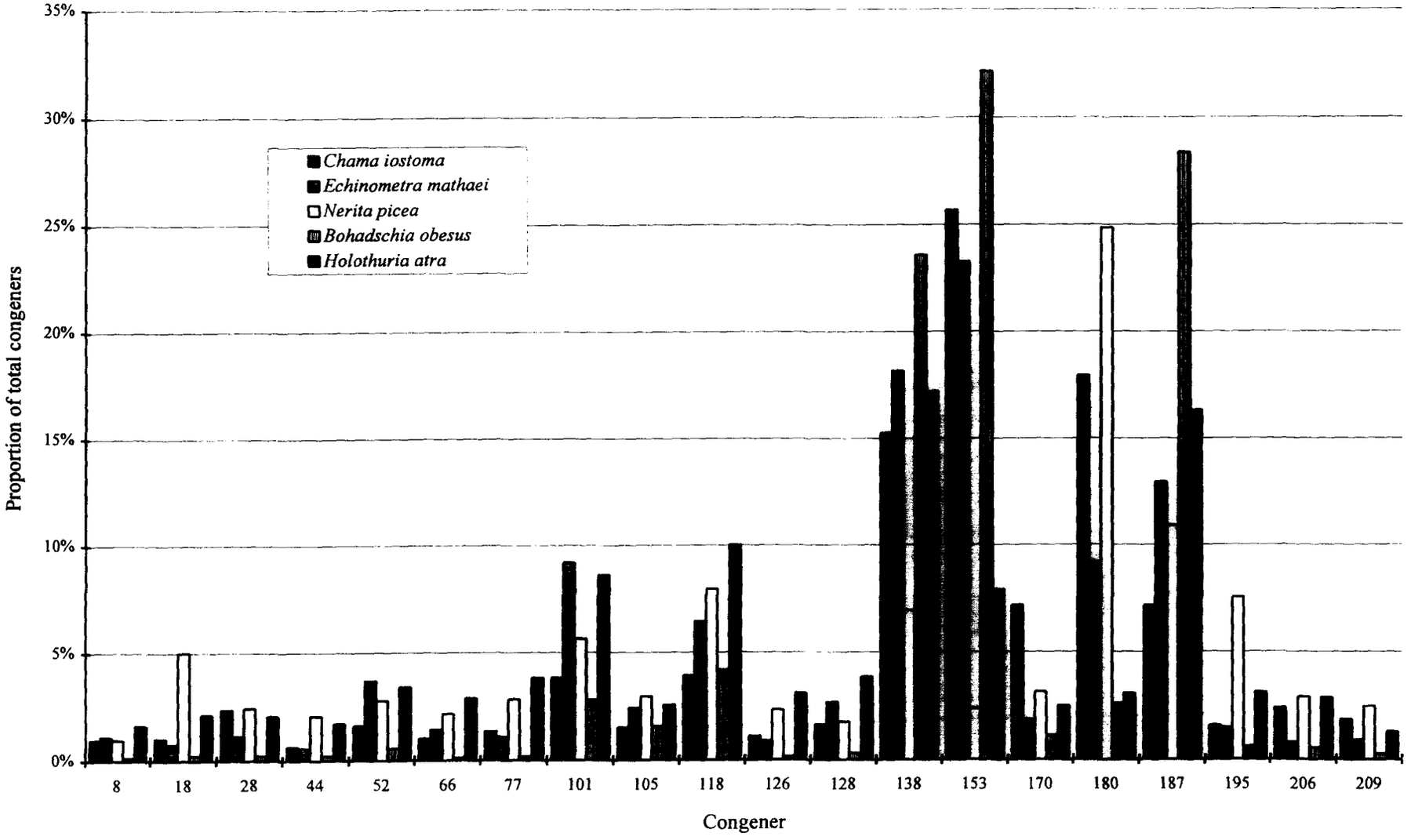


Fig. 4 Distribution of individual congeners in tissues of marine invertebrates.

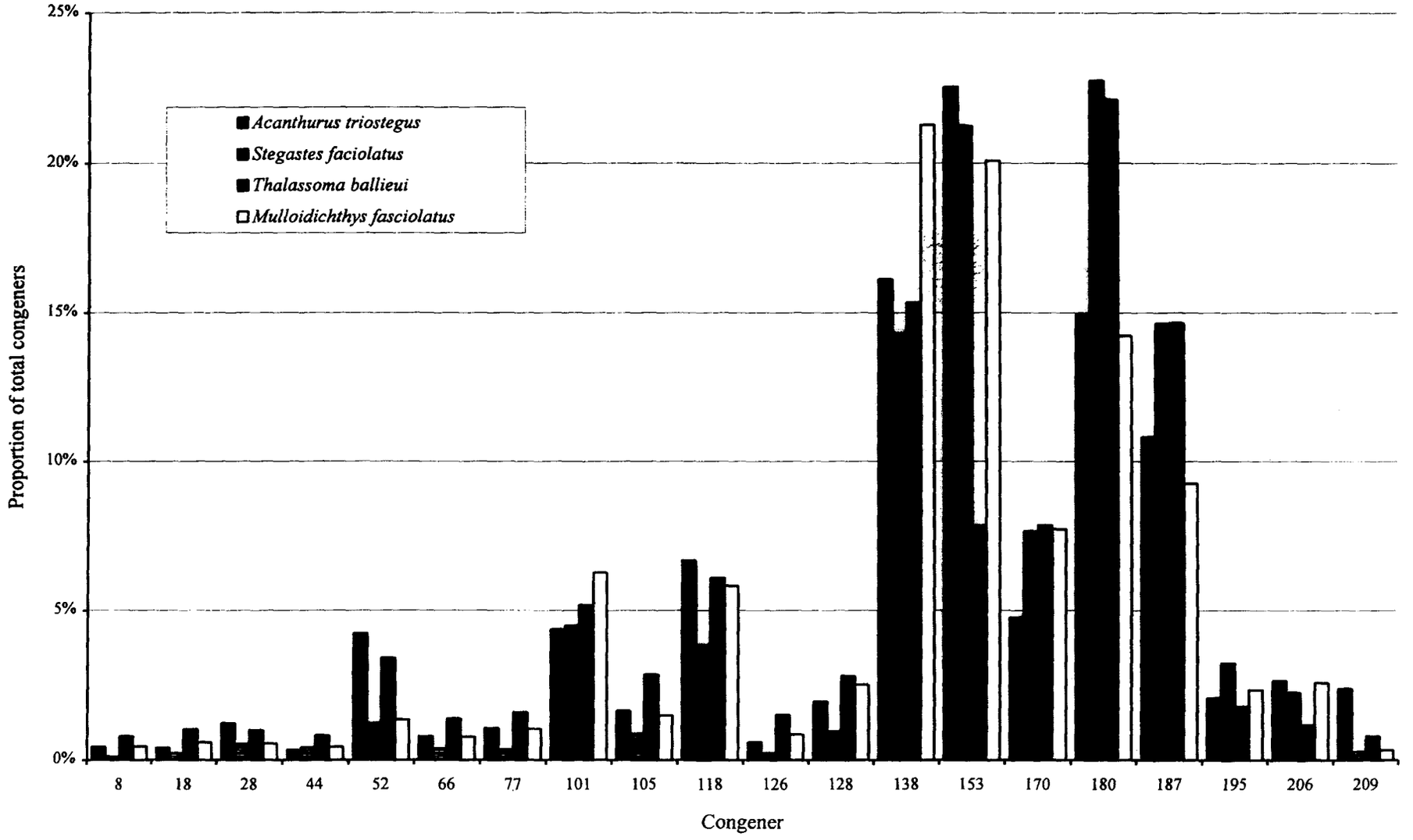


Fig. 5 Distribution of individual congeners in tissues of marine fish.

chlorinated congeners, as evidenced by the elevated proportion (19%) of a trichlorobiphenyl (PCB 18) in sea grass (*H. ovalis*) and similarly (16%) for a tetrachlorobiphenyl (PCB 52) in the macroalgae, *D. acutiloba*. This effect is possibly attributable to the greater aqueous solubility of the lower chlorinated congeners (Bright *et al.*, 1995; Gobas *et al.*, 1991) and the mechanism (passive sorption) by which aquatic plants accumulate PCBs from water. Proportions of tri- and tetrachlorobiphenyls are generally low (< 10%) in animal species. In all biota, a reduction in the proportion of PCB 170 was a common pattern.

Oliver and Niimi (1988) and MacDonald *et al.* (1992) reported a relative constancy of congener proportions for biota in freshwater systems. In this marine system, however, collective proportions of five prominent congeners (PCBs 138, 153, 170, 180, 187) increased slightly with increasing trophic level-in sediments, and surface water collective proportions ranged from 36 to 52%, in plants from 43 to 50%, in invertebrates from 46 to 73% (excluding *Bohadschia*), and in fish from 67 to 80%. The nearly three-fold increase between abiotic media and fish suggests enrichment of these congeners. However, for all congeners with five or more chlorines (PCB 105 and above), any increases with trophic level were less apparent, with collective proportions ranging from 69 to 78% in sediments and surface water, from 66 to 84% in plants, from 81 to 91% in invertebrates (excluding *Bohadschia*), and from 89 to 96% in fish.

Changes in congener composition in some aquatic systems may be attributed to a decline in the proportion of less chlorinated PCBs that are more susceptible to losses through volatilization, sedimentation, and possibly microbial degradation (Brown *et al.*, 1987; MacDonald *et al.*, 1992; Quensen *et al.*, 1988). Moderately and higher chlorinated PCBs may remain at constant levels in the aquatic environment because they are less volatile, more soluble in lipids, adsorb readily to sediments, and are more resistant to metabolic and microbial degradation (Connell, 1988; Shiu and Mackay, 1986). Results of the preceding analysis of alterations in congener concentration and composition in various species and trophic levels for this large aquatic system suggest that: (a) proportions of some, but not all, higher chlorinated (≥5 Cl) congeners increase through the food web and (b) less chlorinated congeners constitute a relatively small proportion of congeners in biota that accumulate PCBs by absorption from the aqueous phase.

Pearson product–moment correlation assessed whether congener proportions among the different species moved together, i.e. did different species have similar values for proportions of each congener. As shown in Table 2, significant relationships ($p < 0.01$) in congener patterns were identified. Of note is the lack of significant correlation between congener patterns in seawater and any biota. Given the tendency for PCBs to sorb on macroalgal surfaces and partition into algal

TABLE 2

Between species Pearson product–moment correlation coefficients based on mean congener proportions.

Media/species	Surface water	Sediment	*Dictyota acutiloba*	*Giffordia breviarticulata*	*Halophila ovalis*	*Bohadschia obesus*	*Holothuria atra*	*Chama iostoma*	*Echinometra mathaei*	*Nerita picea*	*Acanthurus triostegus*	*Stegastes fasciolatus*	*Mulloidichthys flavolineauts*	*Thalassoma ballieui*
Surface water														
Sediment	**0.605**													
Dictyota acutiloba	−0.025	0.377												
Giffordia breviarticulata	−0.096	**0.662**	0.402											
Halophila ovalis	−0.106	0.464	0.358	**0.600**										
Bohadschia obesus	−0.052	**0.664**	0.473	**0.864**	**0.627**									
Holothuria atra	−0.052	0.557	0.303	**0.844**	0.448	**0.798**								
Chama iostoma	0.122	**0.734**	0.556	**0.759**	0.542	**0.751**	0.462							
Echinometra mathaei	−0.088	**0.648**	0.548	**0.924**	**0.612**	**0.916**	**0.767**	**0.882**						
Nerita picea	0.148	0.475	0.214	0.510	0.133	0.180	0.279	0.469	0.329					
Acanthurus triostegus	0.040	**0.735**	**0.620**	**0.865**	**0.561**	**0.841**	**0.633**	**0.969**	**0.947**	0.489				
Stegastes fasciolatus	0.180	**0.812**	0.500	**0.830**	0.465	**0.747**	0.546	**0.942**	**0.845**	**0.679**	**0.945**			
Mulloidichthys flavolineatus	0.145	**0.786**	0.539	**0.839**	0.544	**0.807**	**0.689**	**0.939**	**0.929**	0.465	**0.960**	**0.915**		
Thalassoma ballieui	0.275	**0.783**	0.427	**0.747**	0.291	**0.569**	**0.606**	**0.741**	**0.680**	**0.831**	**0.786**	**0.893**	**0.807**	

For sample size = 20 (degrees of freedom = 18), critical value of R is 0.561 at the 1% level.
bold = Correlation between variables is significantly different than zero at the 1% level.

lipids (Rohrer *et al.*, 1982), some significant degree of correlation might be expected between the macroalgaes (*D. acutiloba* and *G. breviarticulata*) and seawater. Although congener concentrations in biota are undoubtedly influenced by ambient concentrations of PCBs in seawater, this lack of correlation suggests that seawater measurements alone may not be reliable predictors of congener concentrations in marine biota.

Congener profiles in biota result from the interaction of ecological factors, such as foraging range, food preferences, behaviour, etc., that influence exposure to PCBs. Profiles are also affected by species-specific and congener-specific metabolic activity rates that apparently influence the post-exposure form of PCBs (Barron, 1990; Boon *et al.*, 1984). Differences in species lipid content, depuration rates, and uptake of the different congeners in the diet as well as biotransformation also undoubtedly influence these profiles (Porte and Albãigés, 1993). Given the potential complexity of such interactions, this degree of significant correlation is remarkable.

Bioconcentration and bioaccumulation patterns

The degree of chlorination of the PCB molecule is known to have an effect on bioconcentration of PCBs, with the tetra-, penta- and hexachlorobiphenyls being accumulated to the greatest extent by fish (Fox *et al.*, 1994). Bioconcentration of PCBs by aquatic organisms is also correlated with lipophilicity (Fox *et al.*, 1994). Log bioconcentration factors (log BCF = $\log_{10}$\{mean tissue concentration/mean surface water concentration\}) and log bioaccumulation factors (log BAF = $\log_{10}$\{mean tissue concentration/mean sediment concentration\}) were calculated to indicate whether PCBs were accumulated in biota to concentrations greater than those in surrounding surface waters and whether any such accumulation displayed a characteristic pattern.

Lipophilicity is commonly measured as the partition coefficient (K_{ow}) between *n*-octanol and water. The log K_{ow} values of the PCB congeners, determined by Hawker and Connell (1988), are as follows (PCB #, log K_{ow}): 8, 5.07; 18, 5.24; 28, 5.67; 44, 5.75; 52, 5.84; 66, 6.20; 77, 6.36; 101, 6.38; 105, 6.65; 118, 6.74; 126, 6.89; 128, 6.74; 138, 6.83, 153; 6.92; 170, 7.27; 180, 7.36; 187, 7.17; 195, 7.56; 206, 8.09; 209, 8.18). Following the work of Fox *et al.* (1994) and Hansch and Clayton (1973), relationships between log K_{ow}, log BAF and log BCF values for marine biota at selected trophic levels were described by a parabolic relationship, where log BAF or log BCF first increases then decreases with increasing lipophilicity. This relationship takes the form: log BAF or log BCF = $a(\log K_{ow})^2 + b(\log K_{ow}) + c$. Such a relationship between log BAF, log BCF and log K_{ow} for a representative organism from each trophic level are shown graphically in Fig. 6a,b and as equations below:

- *G. breviarticulata* (GB) – primary producer

$$\log BAF = -0.19(\log K_{ow})^2 + 2.49(\log K_{ow}) - 6.85 \quad r^2 = 0.31, \; n = 20$$

$$\log BCF = -0.24(\log K_{ow})^2 + 3.21(\log K_{ow}) - 7.22 \quad r^2 = 0.23, \; n = 20$$

- *E. mathaei* (EM) – herbivorous invertebrate

$$\log BAF = -0.26(\log K_{ow})^2 + 3.55(\log K_{ow}) - 11.00 \quad r^2 = 0.31, \; n = 20$$

$$\log BCF = -0.32(\log K_{ow})^2 + 4.26(\log K_{ow}) - 11.31 \quad r^2 = 0.26, \; n = 20$$

- *T. ballieui* (TB) – carnivorous vertebrate

$$\log BAF = -0.26(\log K_{ow})^2 + 3.54(\log K_{ow}) - 10.72 \quad r^2 = 0.51, \; n = 20$$

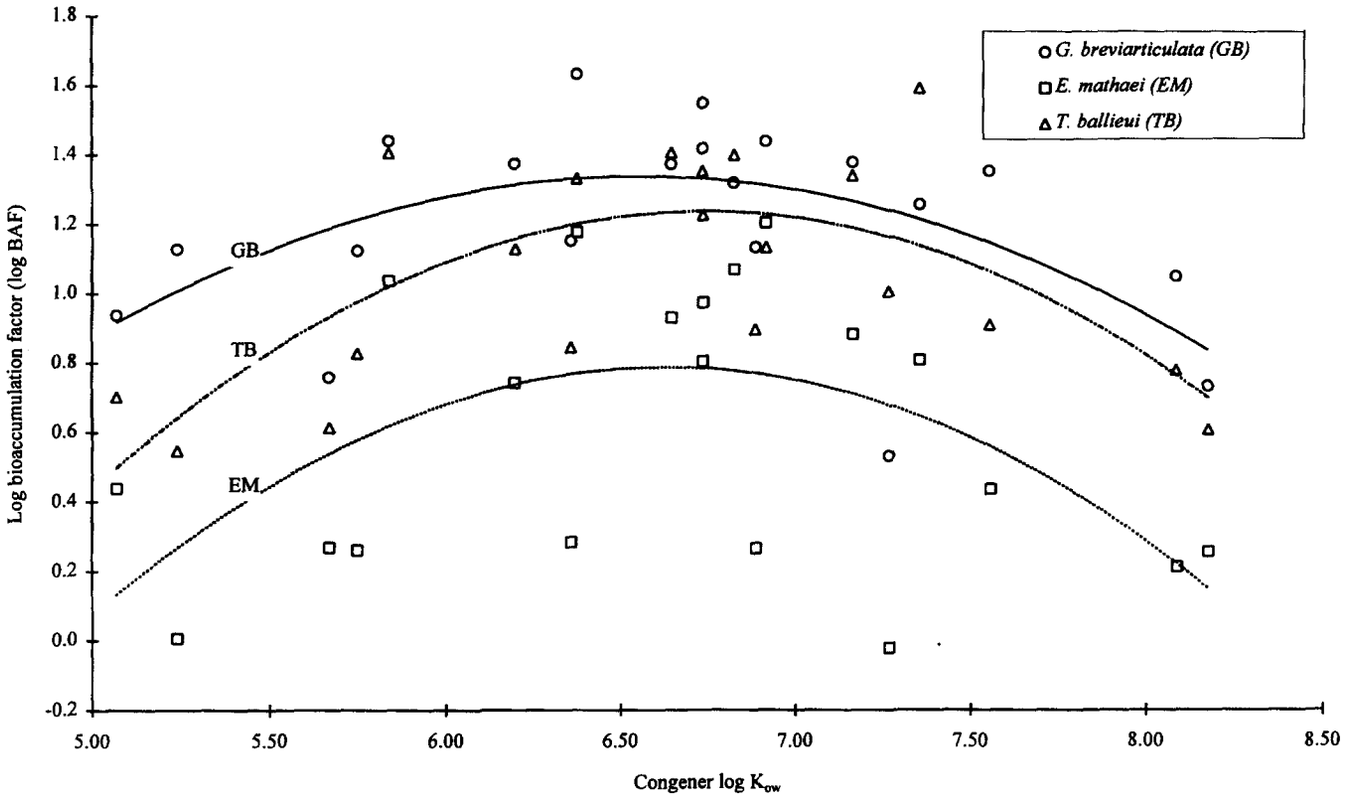
$$\log BCF = -0.31(\log K_{ow})^2 + 4.25(\log K_{ow}) - 11.09 \quad r^2 = 0.39, \; n = 20$$

For all biota sampled, the strength (as indicated by the correlation coefficient, r^2) of the parabolic relationship, although never strong, does increase slightly with increasing trophic level. Relationships similar to these have also been reported by Fox *et al.* (1994) and may be based in part on the increased lipid content of higher trophic level species.

Boese *et al.* (1995) reported bulk sediment BAF values, for individual PCB congeners in the deposit-feeding marine clam (*Macoma nasuta*), from 0.9 (PCB 209) to 30.3 (PCB 118), with an average BAF value of 15.6 for 13 congeners. In the present study, BAF values for the sediment-dwelling bivalve *C. iostoma* ranged from 6.39 (PCB 18) to 80.85 (PCB 153), with an average BAF value of 23.7 for 20 congeners.

Influence of congener stereochemistry on distribution

Borlakoglu *et al.* (1990) and Bright *et al.* (1995) have suggested that the stereochemistry of PCBs can influence their differential metabolism by marine animals, including invertebrates, fish, mammals and birds. Not readily metabolized or persistent, congeners share the characteristics of chlorines at the *para*-positions on both phenyl rings and no adjacent unsubstituted *meta*- and *para*-sites (i.e. chlorine substituents at adjacent *meta-para* carbon atoms). Borlakoglu *et al.* (1990) have indicated that the absence of H atoms at adjacent C atoms is essential for accumulation of PCBs. In addition, there is no evidence for metabolism of *ortho*- and *meta*-unsubstituted congeners. Both Borlakoglu *et al.* (1990) and Bright *et al.* (1995)



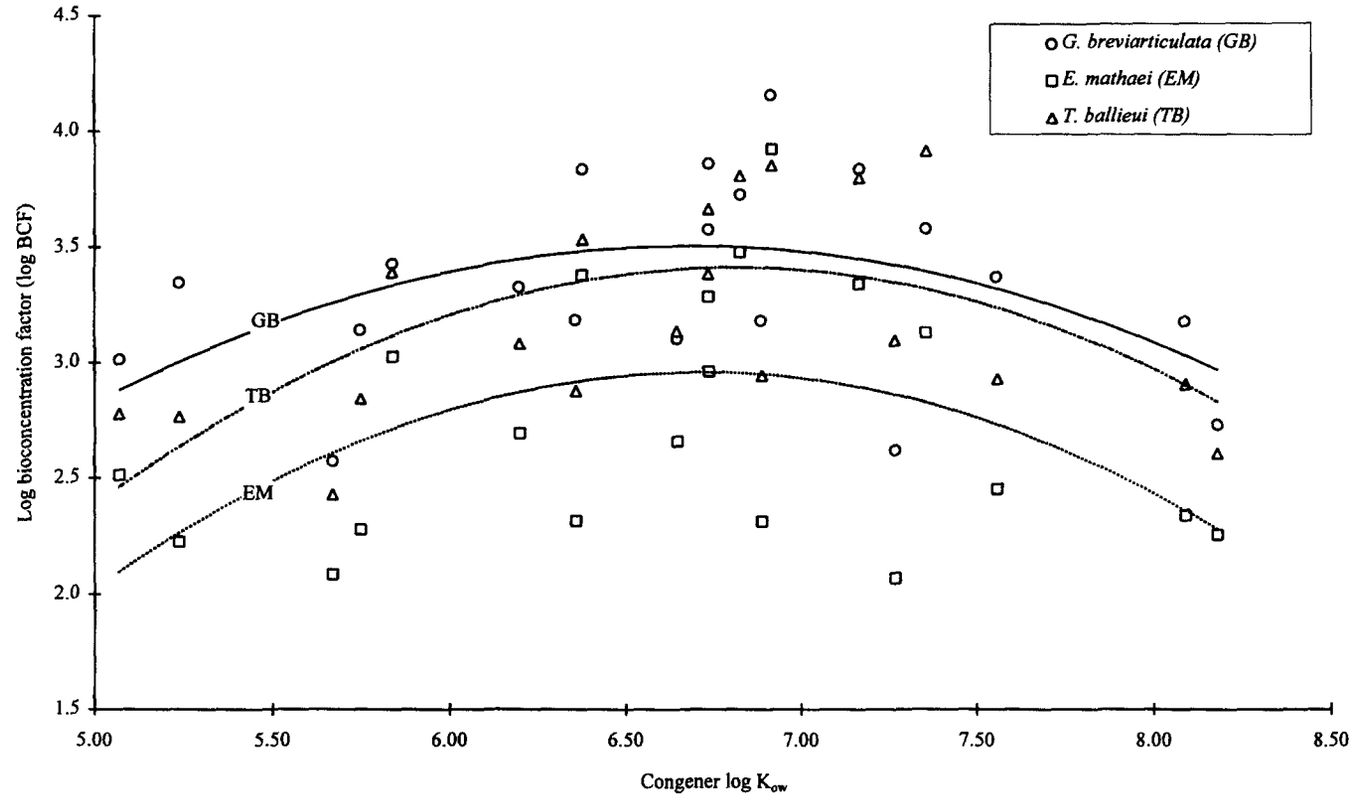


Fig. 6 (a) Bioaccumulation from marine sediment in relation to congener octanol–water partition coefficient (log K_{ow}). (b) Bioconcentration from marine surface water in relation to congener octanol-water partition coefficient (log K_{ow}).

have suggested that metabolic modification is facilitated by adjacent unsubstituted *meta-* and *para*-carbons on one or both biphenyl rings. Thus more readily metabolized or non-persistent congeners have one pair of adjacent unsubstituted *meta-para* carbon atoms in their rings.

To explore whether similar relationships exist for the marine biota sampled at Midway, changes in congener composition for each species relative to both surface water and sediment were compared within groups defined by the stereochemical characteristics of each congener. To facilitate comparison to previous investigations, the stereochemical categories developed by Bright *et al.* (1995) were employed, so that there were: (A) no adjacent *ortho-, meta-* or *meta-, para*-unsubstituted sites (PCBs 153, 180, 187, 206, 209); (B) adjacent *ortho-, meta*-unsubstituted sites only (PCBs 28, 105, 118, 128, 138, 170, 195); (C) adjacent *meta-, para*-unsubstituted sites only (PCBs 52, 66, 77, 101, 126); and (D) both *ortho-, meta-* and *meta-, para*-unsubstituted sites (PCBs 8, 18, 44).

The results shown in Fig. 7a,b for the 12 Midway species are in good agreement with those presented by Bright *et al.* (1995) for four-horn sculpins alone. Persistent category (A) congeners are generally accumulated in these marine species (the exceptions being *H. ovalis* and *B. obesus*), while non-persistent category (C) and (D) congeners are generally (with several exceptions) not accumulated. The most striking difference between these results and those of Bright *et al.* (1995) is that here the category (B) congeners are apparently being metabolized by a majority of species (with the exception of *B. obesus* and *A. triostegus*).

Distribution of non-ortho- *and mono*-ortho-*substituted congeners*

Only 20 of the 209 possible PCB congeners have non-*ortho* chlorine substitutions in the biphenyl rings. These congeners can attain a planarity which makes their structure similar to the highly toxic dibenzo-*p*-dioxins and dibenzofurans (McKinney *et al.*, 1976, 1985). Particularly important within this group are the PCBs having four, five, or six chlorines in non-*ortho* positions, for example, PCBs 77, 126 and 169. These are very potent mimics of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) and 2,3,7,8-tetrachlorodibenzofuran (TCDF) both in P-450 induction and toxic effects (Safe, 1990). Certain mono-*ortho*-substituted PCBs (PCBs 105, 114, 118, 123, 156, 157, 167 and 189) induce aryl hydrocarbon hydroxylase (AHH) and resemble mixed-type inducers (De Voogt *et al.*, 1990). It has been suggested that some of these mono-*ortho* congeners (specifically, PCBs 105, 118 and 156) have a greater toxic impact on marine mammals, while non-ortho congeners are more prominent in terrestrial mammals (Kannan *et al.*, 1989; Tanabe *et al.*, 1994). The levels of the mono-*ortho* congener 105 and the planar congener 126, and possibly also the mono-ortho congeners 118 and 156 appear to contribute substantially to the total toxicity of PCB residues with respect to TCDD (De Voogt *et al.*, 1990). Thus, the presence of significantly elevated levels of these congeners into marine food chains at Midway could have possible long-term toxic potential on marine mammals or certain piscivorous birds (i.e. terns and noddies) that forage in the near-shore waters of the atoll.

In marine sediments, PCBs 77 and 126 respectively accounted for 3.7% and 3.1% of total congeners measured; in surface water 5.1% and 4.2%, respectively. In aquatic macrophytes, PCB 77 accounted for 0.8–4.2% and PCB 126 for 0.6%–2.3% of the total congeners, with the higher percentage for PCB 77, possibly due to its higher aqueous solubility. In invertebrates these congeners were present in the range of 0.1–3.8% for PCB 77 and 0.1–3.0% for PCB 126. In fish species, PCBs 77 and 126 accounted for 0.3–1.5% and 0.2–1.5% of the total congener concentration, respectively. These data suggest that both PCB 77 and 126 generally comprise a smaller percentage of total PCBs with increasing trophic level. Bright *et al.* (1995) also reported that congener 77 comprised a smaller percentage of total PCBs with increasing trophic level in Cambridge Bay (Canada); from 0.64% of total PCBs in sediment to 0.12–0.18% in sea urchins and 0.03–0.06% in four-horned sculpins.

In marine sediments, PCBs 105 and 118 accounted for 1.8% and 4.4% of the total congener in sediments; in surface water 5.1% and 3.2%, respectively. In aquatic macrophytes, these two congeners accounted for 1.0–2.3% and 1.8–8.4% of the total congener concentration, respectively. In invertebrate species, PCB 105 contributed 1.5–2.9% and PCB 118 contributed 3.9–10.0% of the total congener concentration. In fish, their respective contributions ranged from 0.8 to 2.8% and 3.8 to 6.6%. Both PCBs 105 and 118 displayed an approximately constant percentage of total PCBs with increasing trophic level.

Toxic effects to wildlife appear to be correlated more with total TCDD toxicity equivalents than with absolute PCB concentrations. Toxic equivalency factor (TEF) values can be used to normalize PCB concentrations to their TCDD equivalents and Safe (1990) has proposed provisional TEF values of 0.01, 0.1, 0.001 and 0.001 for PCBs 77, 126, 105 and 118, respectively. Giesy *et al.* (1994) proposed the use of TCDD toxicity equivalents to determine levels of acceptable risk toward the protection of wildlife and human health. Total (sum of the four congeners) mean TCDD equivalent concentrations in marine biota collected at Midway ranged from 0.011 to 0.544 $\mu g\ kg^{-1}$, with a total (sum of 12 species) of 1.19 $\mu g\ kg^{-1}$. Although two higher trophic level species, *Stegastes* and *Mulloidichthys*, collectively contributed 0.74 $\mu g\ kg^{-1}$ or approximately 62% of the total, there was no indication of TCDD equivalent concentrations increasing with trophic level. TCDD equivalent concentrations for

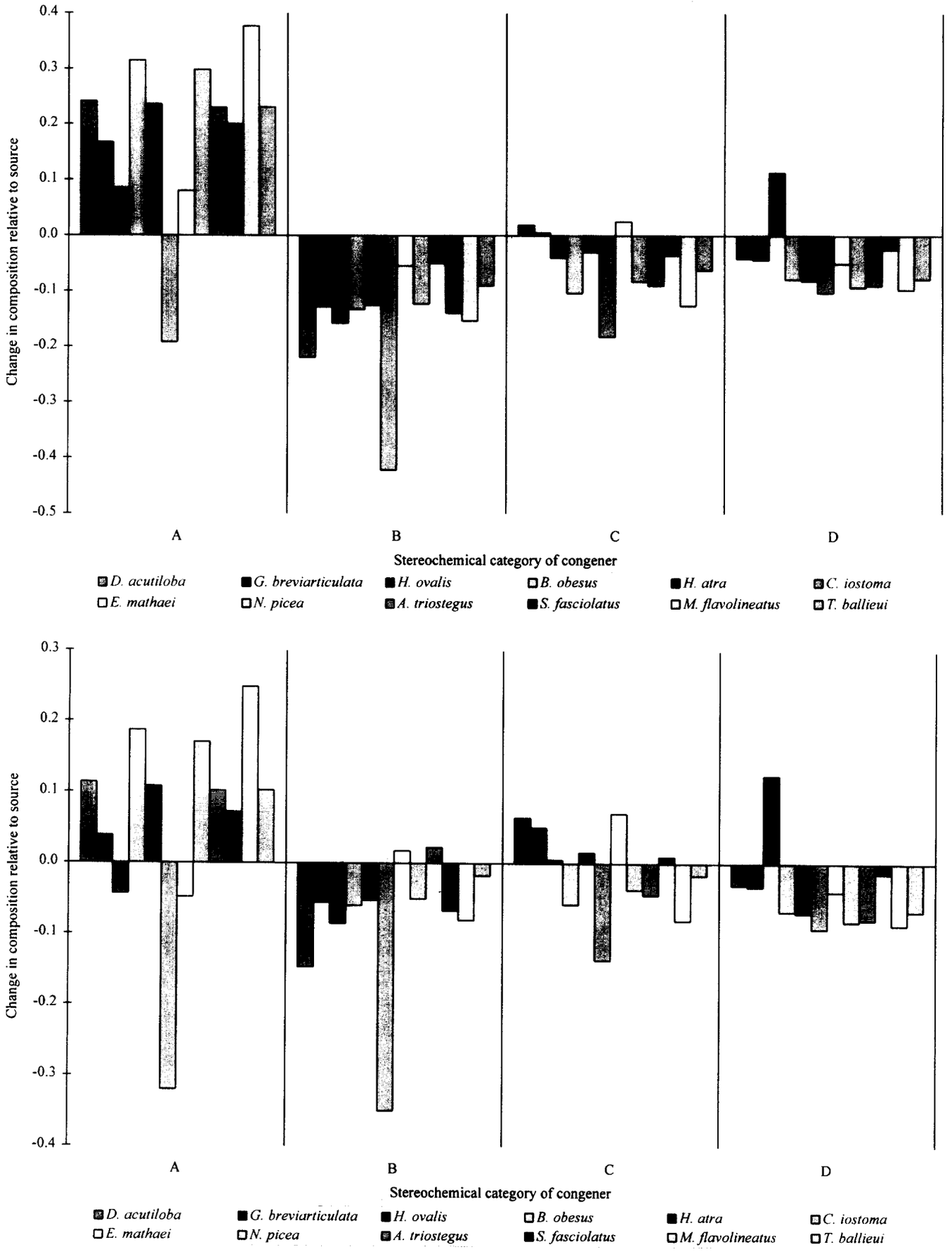


Fig. 7 (a) Differential metabolism (marine surface water) by species in relation to congener molecular characteristics. (b) Differential metabolism (marine sediment) by species in relation to congener molecular characteristics.

PCBs 105 and 118, those of specific interest with respect to marine mammals, ranged from 0.001 to 0.038 μg kg^{-1}, with *Stegastes* and *Mulloidichthys* again collectively contributing 0.06 μg kg^{-1} or approximately 67% of the total.

The TCDD equivalent concentrations reported here are low in comparison to the lowest values (13 μg kg^{-1}) reported in oysters (*Crassostrea virginica*) taken from significantly contaminated regions of Galveston Bay, Texas (Sericano *et al.*, 1992). Murray *et al.* (1979) reported a NOAEL dose of 0.001 μg kg^{-1} day^{-1} for rats fed TCDD over three generations, including critical reproductive life stages, which extrapolates to a dietary NOAEL of ≈0.002 μg kg^{-1} for a 50-kg seal. Recognizing the significant uncertainties involved in such interspecies toxicological extrapolations, we nonetheless conclude that a large marine mammal which infrequently consumes PCB-contaminated food items from near-shore waters at Midway would not have a greater risk of experiencing adverse toxicological effects.

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ADL (1994) *Proposal for Off-site Laboratory Analytical Services, Statement of Qualifications.* Prepared by Arthur D. Little, Inc., Cambridge, Massachusetts for Ogden Environmental and Energy Services Company, Inc., RFP No. 4-32-R2-09492.

Ballschmiter, K. and Zell, M. (1980) Analysis of polychlorinated biphenyl (PCB) by glass capilliary gas chromatography. *Fresenius Z. Anal. Chem.* **302**, 20–31.

Barron, M. G. (1990) Bioconcentration. *Environmental Science and Technology* **24**, 1612–1618.

Bauman, P. C. and Whittle, D. M. (1988) The status of selected organics in the Laurentian Great Lakes: an overview of DDT, PCBs, dioxins, furans, and aromatic hydrocarbons. *Aquatic Toxicology* **11**, 241–257.

Boese, B. L., Winsor, M., Lee, H. Jr II, Echols, S., Pelletier, J. and Randall, R. (1995) PCB congeners and hexachlorobenzene biota sediment accumulation factors for *Macoma nasuta* exposed to sediments with different total organic carbon contents. *Environmental Toxicology and Chemistry* **14**, 303–310.

Boon, J. P., Oudejans, R. C. H. M. and Duinker, J. C. (1984) Kinetics of polychlorinated biphenyl (PCB) components in juvenile sole, *Solea solea*, in relation to their concentrations in food and to lipid metabolism. *Comparative Biochemistry and Physiology* **79C**, 131–142.

Borlakoglu, J. T., Wilkins, J. P. G., Walker, C. H. and Dils, R. R. (1990) Polychlorinated biphenyls (PCBs) in fish-eating sea birds – III. Molecular features and metabolic interpretations of PCB isomers and congeners in adipose tissues. *Comparative Biochemistry and Physiology* **97C**, 173–177.

Bright, D. A., Grundy, S. L. and Reimer, K. J. (1995) Differential bioaccumulation of non-*ortho*-substituted and other PCB congeners in coastal arctic invertebrates and fish. *Environmental Science and Technology* **29**, 2504–2512.

Brown, J. F. Jr, Bedard, D. L., Brennan, M. J., Caranhan, J. C., Feng, H. and Wagner, R. E. (1987) Polychlorinated biphenyl dechlorination in aquatic sediments. *Science* **236**, 709–712.

Connell, D. W. (1988) Bioaccumulation behavior of persistent organic chemicals with aquatic organisms. *Review of Environmental Contamination and Toxicology* **14**, 587–594.

De Voogt, P., Wells, D. E., Reutergårdh, L. and Brinkman, U. A. T. (1990) Biological activity, determination and occurrence of planar, mono- and di-ortho PCBs. *International Journal of Environmental Analytical Chemistry* **40**, 1–46.

Duinker, J. C., Schulz, S. E. and Petrick, G. (1988) Selection of chlorinated biphenyl congeners for analysis in environmental samples. *Marine Pollution Bulletin* **19**, 19–25.

Falandysz, J., Yamashita, N., Tanabe, S., Tatsukawa, R., Rucinska, L., Mizera, T. and Jakuczun, B. (1994) Congener-specific analysis of polychlorinated biphenyls in white-tailed sea eagles *Haliaeetus albicilla* collected in Poland. *Archives of Environmental Contamination and Toxicology* **26**, 13–22.

Fox, K., Zauke, G.-P. and Butte, W. (1994) Kinetics of bioconcentration and clearance of 28 polychlorinated biphenyl congeners in zebrafish (*Brachydanio rerio*). *Ecotoxicology and Environmental Safety* **28**, 99–109.

Giesy, J. P., Ludwig, J. P. and Tillitt, D. E. (1994) Deformities in birds of the Great Lakes regions: assigning causality. *Environmental Science and Technology* **28**, 129–135.

Gobas, F. A. P. C., McNeil, E. J., Lovett-Doust, L. and Haffner, G. D. (1991) Bioconcentration of chlorinated aromatic hydrocarbons in aquatic macrophytes. *Environmental Science and Technology* **25**, 924–929.

Hansch, C. and Clayton, J. M. (1973) Lipophilic character and biological activity of drugs. 2: The parabolic case. *Journal of Pharmacy Science* **62**, 1–21.

Harris, H. J., Erdman, T. C., Ankley, G. T. and Lodge, K. B. (1993) Measures of reproductive success and polychlorinated biphenyl residues in eggs and chicks for Forster's Terns on Green Bay, Lake Michigan, Wisconsin – 1988. *Archives of Environmental Contamination and Toxicology* **25**, 304–314.

Hawker, D. W. and Connell, D. W. (1988) Octanol-water partition coefficients of polychlorinated biphenyl congeners. *Environmental Science and Technology* **22**, 382–387.

Iwata, H., Tanabe, S., Sakai, N., Nishimura, A. and Tatsukawa, R. (1993) Distribution of persistent organochlorines in the oceanic air and surface seawater and the role of ocean in their global transport and fate. *Environmental Science and Technology* **27**, 1080–1098.

Iwata, H., Tanabe, S., Sakai, N. and Tatsukawa, R. (1994) Geographical distribution of persistent organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes. *Environmental Pollution* **85**, 15–33.

Kannan, K., Tanabe, S., Ono, M. and Tatsukawa, R. (1989) Critical evaluation of polychlorinated biphenyl toxicity in terrestrial and marine animals: increasing impact of non-*ortho* and mono-*ortho* coplanar of polychlorinated biphenyls from land and ocean. *Archives of Environmental Contamination and Toxicology* **18**, 850–857.

MacDonald, C. R., Metclafe, C. D., Metcalfe, T. and Balch, G. C. (1992) Temporal trends and distribution of PCB congeners in a small contaminated lake in Ontario Canada. In *Chemical Dynamics in Freshwater Ecosystems*, eds F. A. P. C. Gobas and J. A. McCorqodale, pp. 211–236. Lewis, Boca Raton, Florida.

McKinney, J. D., Chae, K., Gupta, B. N., Moore, J. A. and Goldstein, J. A. (1976) Toxicological assessment of hexachlorobiphenyl isomers and 2,3,7,8-tetrachlorodibenzofuran in chicks. I. Relationship of chemical parameters. *Toxicology and Applied Pharmacology* **36**, 65–80.

McKinney, J. D., Chae, K., McConnel, E. E. and Binbaum, L. S. (1985) Structure-induction versus structure-toxicity relationships for polychlorinated biphenyls and related aromatic hydrocarbons. *Environmental Health Perspectives* **60**, 57–68.

Murray, F. J., Smith, F. A., Nitschke, C. G., Humiston, R. J., Kociba, R. J. and Schwetz, B. A. (1979) Three-generation reproduction study of rats given 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) in the diet. *Toxicology and Applied Pharmacology* **50**, 241–252.

Oliver, B. G. and Niimi, A. J. (1988) Trophodynamic analysis of polychlorinated biphenyl congeners and other chlorinated hydrocarbons in the Lake Ontario ecosystem. *Environmental Science and Technology* **22**, 388–397.

Porte, C. and Albaigés, J. (1993) Bioaccumulation patterns of hydrocarbons and polychlorinated biphenyls in bivalves, crusta-

ceans and fishes. *Archives of Environmental Contamination and Toxicology* **26**, 273–281.

Quensen, J. F. Jr III, Tiedje, J. M. and Boyd, S. A. (1988) Reductive dechlorination of polychlorinated biphenyls by anaerobic microorganisms from sediments. *Science* **242**, 752–754.

Rohrer, T. K., Forney, J. C. and Hartig, J. H. (1982) Organochlorine and heavy metal residues in standard fillets of coho and chinook salmon of the Great Lakes – 1980. *Journal of Great Lakes Research* **8**, 623–634.

Safe, S. (1990) Polychlorinated biphenyls (PCBs), dibenzo-*p*-dioxins (PCDDs), dibenzofurans, (PCDFs) and related compounds, environmental and mechanistic considerations which support development of toxicity equivalency factors (TEFs). *Critical Reviews in Toxicology* **21**, 51–88.

Sericano, J. L., Wade, T. L., El-Husseini, A. M. and Brooks, J. M. (1992) Environmental significance of the uptake and depuration of planar PCB congeners by the American oyster (*Crassostrea virginica*). *Marine Pollution Bulletin* **24**, 537–543.

Shiu, W. Y. and Mackay, D. (1986) A critical review of aqueous solubilities, vapor pressures, Henry's Law constants and octanol-water partition coefficients of the polychlorinated biphenyls. *Journal of Physics and Chemistry Reference Data* **15**, 911–929.

Tanabe, S. and Tatsukawa, R. (1986) Distribution, behavior and load of PCBs in the oceans. In *PCBs and the Environment*, Vol. 1, ed. J. S. Waid, pp. 143–161. CRC Press, Boca Raton, Florida.

Tanabe, S., Iwata, H. and Tatsukawa, R. (1994) Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals. *The Science of the Total Environment* **154**, 163–177.

Zell, M., Neu, H. J. and Ballschmiter, K. (1978) Single component analysis of PCB and chlorinated pesticide residues in marine fish samples. *Fresenius Z. Anal. Chem.* **292**, 97–107.