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Congener-specific levels and patterns of polychlorinated biphenyls in edible fish tissue from the central Red Sea coast of Saudi Arabia

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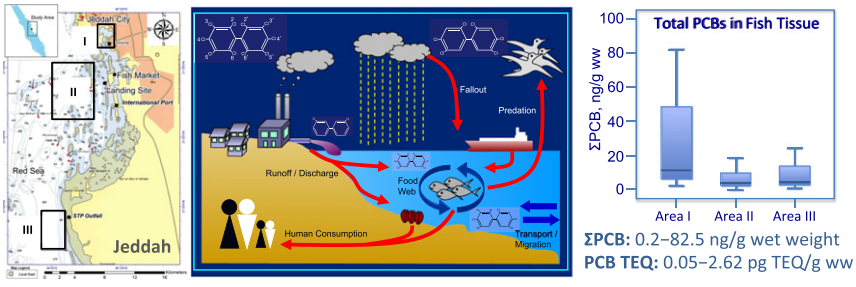
HIGHLIGHTS

- All 209 PCB congeners in fish tissue from coastal areas were analyzed.
- Total PCB levels were at the lower end of reported global range.
- Total PCB toxic equivalencies were within tolerable levels.
- Congener profiles were dominated by hexachlorobiphenyl isomers.
- Selected indicator congeners were identified for future biomonitoring.

GRAPHICAL ABSTRACT



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ABSTRACT

All 209 congeners of polychlorinated biphenyls (PCBs) in edible fish tissue from the central Red Sea coast (Jeddah region) of Saudi Arabia were analyzed by isotope dilution high-resolution gas chromatography–mass spectrometry. The upper-bound total PCB (ΣPCB) levels in nine commonly consumed fish species from three areas were 0.2–82.5 ng/g wet weight (17–8450 ng/g lipid weight), which were at the lower end of reported global range and far below international tolerance limits (500–3000 ng/g ww). Dioxin-like congeners contributed up to 12.8% (mean 6.5%) to ΣPCB in tissue samples, with the total PCB toxic equivalencies (TEQs) at a tolerable range (0.05–2.6 pg TEQ/g ww or 2–238 pg TEQ/g lw) for all species. PCB profiles were dominated by moderately chlorinated homologs, mainly hexachlorobiphenyls, but less chlorinated congeners were also consistently elevated, notably in *Siganus rivulatus* (Area III) and *Mugil cephalus* (Area I). It remains to be ascertained if the latter were breakdown products or due to fresh inputs. The top congeners based on dominance by both occurrence and abundance were identified as potential markers of ΣPCB in fish tissue, which can be used for future selective biomonitoring in case of reasonable constraints on full congener approach.

1. Introduction

Polychlorinated biphenyls (PCBs) are a class of man-made chlorinated aromatic hydrocarbons with various industrial and commercial

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applications due to their low flammability, chemical and thermal stability, and electric insulating properties. PCBs were widely used as dielectric fluids in transformers and capacitors, coolants, flame retardants, hydraulic oils, lubricants, printing ink and dye carriers, pesticide and wax extenders, and additives in paints, plastics, adhesives, sealants, and other products (ATSDR, 2000). PCBs have up to 10 chlorine (Cl) atoms connected to two phenyl rings and the variation in number and position of the Cl atoms results in 209 possible configurations, or congeners. The lateral substitutions of Cl atoms on the biphenyl molecule also force some PCB congeners into a coplanar conformation, in which the phenyl rings align to the same plane (Giesy and Kannan, 1998). Some coplanar PCBs are termed "dioxin-like" for having a structure similar to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD), recognized as the most toxic man-made organic compound (McFarland and Clarke, 1989). Twelve dioxin-like PCBs (DLPCBs) comprising four non-*ortho* (PCBs 77, 81, 126, 169) and eight mono-*ortho* (PCBs 105, 114, 118, 123, 156, 157, 167, 189) congeners are of particular concern due to their toxicity and are thus recommended for monitoring worldwide. Toxic equivalence (TEQ) was devised to express the potency of PCBs relative to 2,3,7,8-TCDD (Van den Berg et al., 2006). As only the DLPCBs share similar coplanar conformation and toxic activity with 2,3,7,8-TCDD, the total PCB TEQ is therefore based on the individual contributions of the 12 DLPCBs. The TEQ of each DLPCB is the product of its measured concentration and toxic equivalency factor (TEF) as set by the World Health Organization (WHO) (Van den Berg et al., 2006).

PCBs are resistant to biological and chemical degradation, which makes them highly persistent. They are ubiquitous in the environment (Risebrough et al., 1968; Fowler, 1990) as a result of widespread usage since their first commercial synthesis in 1929 and being prone to large-scale dispersal through aerial and oceanic transport and biologically mediated mechanisms (Bard, 1999; Weber and Goerke, 2003; Gouin et al., 2004). Several countries have unilaterally banned or imposed strict regulation on the production, marketing, and use of PCBs from the 1970s, as more evidence of their persistence and potential risks were increasingly recognized (Adeola, 2004). Cases of PCB contamination were thus attributed to legacy sources, i.e. as persisting residues from historical usage (Breivik et al., 2004; Lohmann et al., 2007). As the unintended uses of PCBs still persist, some contamination events were ascribed to recent releases from short-range sources, such as residential and industrial emissions, leaching from waste disposal sites, sewage and wastewater effluents, combustion, dumping, spills, and land runoff (Niimi, 1996; Chang et al., 1999; Nieuwoudt et al., 2009).

The marine environment serves as a sink for PCBs and other persistent organic pollutants (POPs). These contaminants occur in almost all water bodies, but often at elevated levels in industrialized and densely populated coastal areas (Fowler, 1990). Being less soluble in water and with congeners that can bind to lipids, PCBs tend to accumulate in sediment and biota (Schulz et al., 1988). Marine species bioaccumulate PCBs from their environment and the contaminant levels are further biomagnified up in the food chain (Porte and Albaigés, 1993). The food-chain transfer of PCBs thus poses potential ecological and human health risks. A growing global concern over such risks has led to the adoption in 2001 and enforcement in 2004 of the Stockholm Convention on Persistent Organic Pollutants, an international treaty that seeks to eliminate or restrict the intentional production, distribution, and use of toxic POPs, including PCBs (UNEP, 2002). The treaty aims to protect human health and the environment from the deleterious effects of POPs based on precautionary principle (Adeola, 2004). The Stockholm Convention was ratified by Saudi Arabia in July 2012 and entered into force in October 2012. Saudi Arabia thus imposed prohibition on the use of PCBs and other POPs since its accession to the Convention in 2012.

The Red Sea is a semi-enclosed water body bordering the western side of Saudi Arabia. The multispecies fisheries along the Saudi Red Sea coast exploit dominant fish species that are associated with an extensive coral reef ecosystem, with average annual production of 24,000 metric tons, mostly (68%) from nearshore traditional fishing, during 2000–2012 (Ministry of Agriculture, 2000–2012). Despite the rapid industrialization and urbanization along the coastal zones of Saudi Arabia (Abdulaal, 2012), PCBs in the marine environment, particularly in the Saudi section of Red Sea, remain largely unexamined, although PCBs and other POPs have been investigated in mussels (Khaled et al., 2004) and coral reef (El Nemr et al., 2004) from the Egyptian waters and in fish and shellfish from the Yemeni waters (Al-Shwafi et al., 2009) of Red Sea. The Saudi coastal waters along the Red Sea are highly susceptible to POPs loadings, including PCBs, due to potential urban and industrial sources onshore, e.g. desalination plants, sewage treatment plants, and industrial complexes, and the high likelihood of deposition of atmospheric emissions due to frequent dust storm events. The ambient urban air in Saudi Arabia has elevated levels of PCBs and other POPs bound to particulate matter (El-Mubarak et al., 2015) and the entire Red Sea receives about 6 megatons of dust deposition annually (Jish Prakash et al., 2015), suggesting a potentially significant POPs input via dust fallout although the overall precipitation is low (0.15–0.5 mm/yr, Sofianos et al., 2002) as an arid region.

This study is the first attempt to determine the levels of all PCB congeners in edible fish tissue from the central Red Sea coast of Saudi Arabia. We have earlier reported the levels of heavy metals in tissue of the major commercial fishes from the same region (Jeddah) (Burger et al., 2014a,b), some of which were also analyzed for PCBs and form the subject of this paper. Our previous findings showed interspecific and locational differences in metal burdens of the major fishes, with human health risk levels exceeding allowable limits for some species at current fish consumption rates in the region. Here, we elucidate the degree of PCB contamination in fishes from the same coastal region, focusing on congener-specific trends and their implications to future biomonitoring.

2. Materials and methods

2.1. Collection of fish samples

Samples of nine fish species (Table 1) were collected from three areas designated as: (a) Area I: a semi-enclosed, soft-bottom lagoon with several drainage outlets and close to city roads and hotels; (b) Area II: a reef complex off the Jeddah International Port, which handles large cargo ships; and (c) Area III: an open-water fringing reef near the outfall of the largest sewage treatment plant in the Jeddah area (Fig. 1). The analyzed fishes were collected by experimental fishing with hook and lines or gillnet by local fishermen in the presence of researchers for quality assurance. All fishing activities were conducted with permits from the Saudi Ministry of Agriculture and Coast Guard. The differences in feeding habits and trophic levels of all analyzed fish species were shown in Burger et al. (2014a) and the rates of fish consumption in the study area were examined in Burger et al. (2014c).

During fishing, the captured fishes were washed with onsite seawater, wrapped individually in aluminium foil, placed in Ziploc zipper bags, and kept in coolers with cooling packs until reaching the laboratory. All fishes were immediately dissected at the laboratory within the same day of capture. After scaling, each fish was rinsed with deionized water and cut for the required flesh fillets (skin-on), which were separately wrapped in aluminium foil, placed in pre-labeled Ziploc bags, and kept frozen until chemical analysis. Field sampling and sample preparations prior to chemical analysis followed quality assurance protocols (USEPA, 2000).

2.2. Tissue sample preparations and chemical analysis

All 209 PCB congeners were determined by isotope dilution high-resolution gas chromatography–mass spectrometry using Method 1668A (USEPA, 2003). For the sample preparation, tissue portions of

Table 1
Body size and lipid content of fish species analyzed for polychlorinated biphenyls (PCBs) from the central Red Sea coast of Saudi Arabia.

Species	Common name	Local name	Area	n	Statistic	Length, cm	Weight, g	Lipid, %
Chanos chanos	Milkfish	Salmani	I	11	Min – Max	32–40	643–936	0.5–8.2
					Mean ± SD	35 ± 3	776 ± 107	3.3 ± 2.9
Mugil cephalus	Mullet	Arabi	I	5	Min – Max	34–39	605–821	6.4–13.1
					Mean ± SD	36 ± 2	669 ± 88	8.9 ± 2.6
Carangoides bajad	Trevally	Bayad	II	5	Min – Max	32–39	718–1085	0.2–1.8
					Mean ± SD	35 ± 3	864 ± 194	0.7 ± 0.7
Epinephelus tauvina	Grouper	Tauvina	II	5	Min – Max	39–59	690–2130	0.6–1.8
					Mean ± SD	47 ± 8	1226 ± 602	1.2 ± 0.5
Plectropomus pessuliferus	Grouper	Najil	II	5	Min – Max	36–69	658–3895	0.2–2.0
					Mean ± SD	48 ± 14	1765 ± 1390	1.0 ± 0.7
Siganus rivulatus[a]	Rabbitfish	Sijan	III	4	Min – Max	17–18	94–120	1.6–3.2
					Mean ± SD	18 ± 1	111 ± 12	2.4 ± 0.7
Cephalopholis argus	Grouper	Hamour	II	5	Min – Max	32–38	543–689	0.3–0.8
					Mean ± SD	35 ± 2	604 ± 60*	0.5 ± 0.2*
			III	4	Min – Max	32–39	810–1110	1.1–2.1
					Mean ± SD	36 ± 3	913 ± 139*	1.6 ± 0.4*
Lethrinus lentjan	Emperor	Sheiry	II	5	Min – Max	33–46	646–1658	0.7–4.8
					Mean ± SD	36 ± 6	976 ± 431	2.5 ± 1.9
			III	5	Min – Max	33–61	646–3085	0.3–7.6
					Mean ± SD	41 ± 11	1272 ± 1028	2.2 ± 3.1
Variola louti	Grouper	Louti	II	9	Min – Max	33–48	690–1851	0.5–2.1
					Mean ± SD	40 ± 5	1090 ± 356	1.4 ± 0.5
			III	5	Min – Max	31–43	496–1124	0.6–1.9
					Mean ± SD	36 ± 5	754 ± 274	1.0 ± 0.5

Min—minimum, Max—maximum, SD—standard deviation, n—number of samples.
[a] Analyzed in composite of 5 individuals each; all other species were analyzed individually.
* Significantly different (Mann-Whitney *U* test) between areas at 0.05 level.

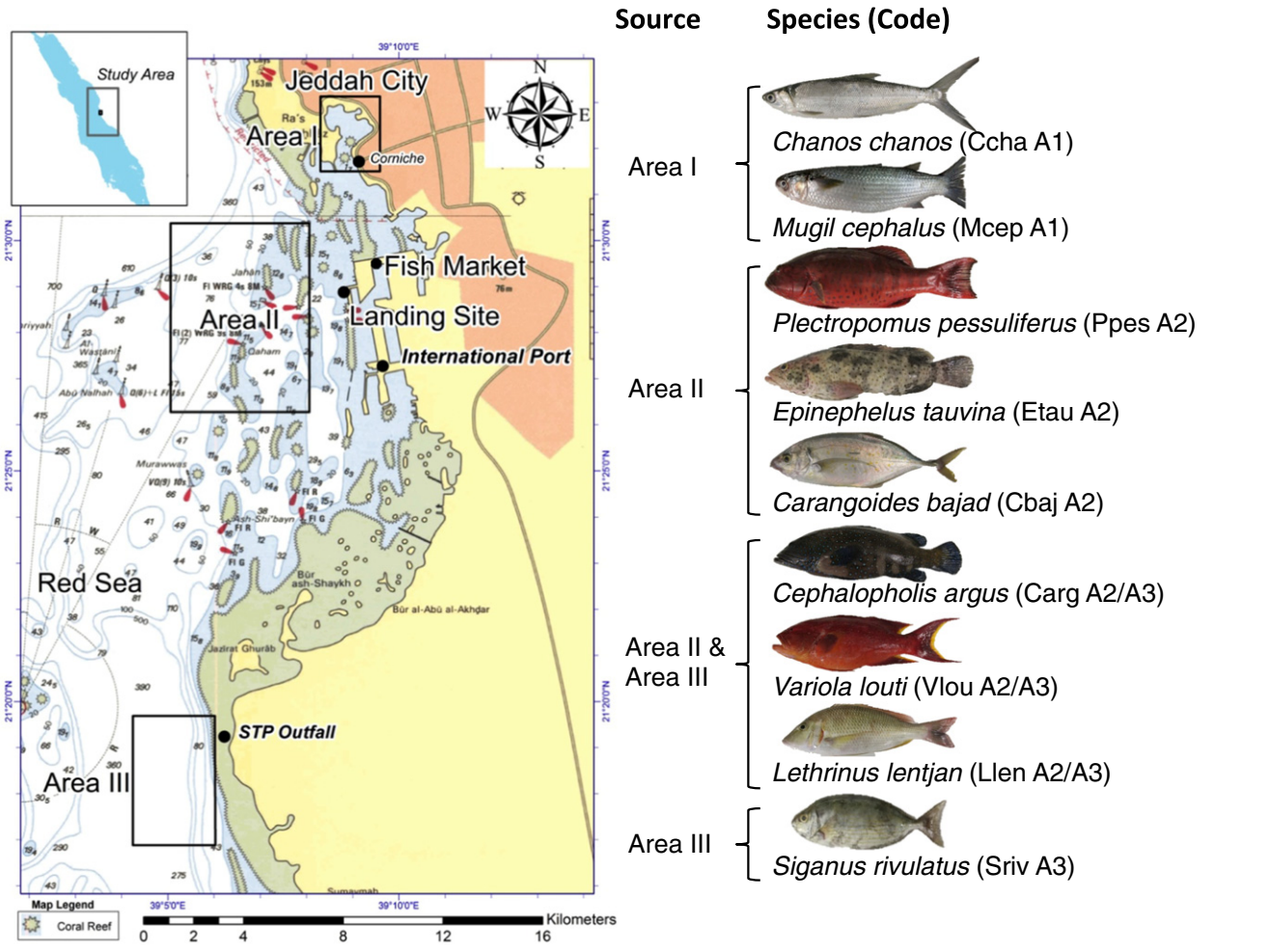

Fig. 1. Location of sampling areas (Area I–III) and respective sources of fish species analyzed for polychlorinated biphenyls (PCBs) in edible tissue from the central Red Sea coast (Jeddah region) of Saudi Arabia. Red Sea is shaded blue in inset.

about 1 cm^3 were cut from the frozen flesh fillets with clean scalpel on cutting board covered with aluminium foil, pre-cleaned with acetone. After homogenizing with blender to fine flour-like powder, a sample of 5–10 g was added with 80–100 g of anhydrous sodium sulfate (reagent grade, EMD Chemicals Inc., USA), just enough to absorb any moisture present in the sample but not to overload the Soxhlet tube, and then mixed with spatula until a uniform mixture was achieved. The sample and sodium sulfate mixture was left for at least 6 h until achieving a free-flowing consistency and no moisture is observed prior to extraction.

For extraction, 500-mL round bottom flask and Soxhlet apparatus for each sample were pre-cleaned thrice with dichloromethane (DCM) (LC/GC grade, EMD). The sample mixture was added with the extraction solvent 1:1 (v:v) DCM/hexane (GC grade, EMD), along with 5–7 boiling chips. Samples, as well as the method blanks and quality control samples (sodium sulfate), were added with 20 μL of $^{13}C_{12}$-labeled surrogate standards. Soxhlet walls were rinsed down with 5 mL DCM to ensure that the entire surrogate is well soaked into the sample. The sample mixture was then kept for 5 min and then placed on a heating mantle to boil, ensuring correct siphoning even when left unattended. The Soxhlets were monitored until they cycled once every 12 min by adjusting the heating mantle and water pressure. Samples were refluxed for 16–24 h after adding the appropriate surrogate and spiking solutions. Samples were syringe-filtered, an aliquot used for gravimetric lipid analysis (sodium sulfate drying and concentrating by Kudaena-Danish evaporation and nitrogen blowdown), and the split for PCBs was subjected to cleanups by gel permeation chromatography (GPC) using silica gel 60 (EMD Millipore) and multi-silica column (conditioned basic-acid silica layers separated by clean sodium sulfate). For multicolumn cleanup, the column was pre-eluted with 30 mL hexane and the sample was quantitatively transferred with hexane and then concentrated to ≤1.0 mL by rotary evaporator (TurboVap). Sample extract was then transferred with hexane to 12 g, 2% deactivated Florisil (pesticide residue analysis grade, Caledon) column and eluted with 35 mL of 5% ethyl acetate (HPLC grade, Fisher) in hexane. After the Florisil cleanup, the extract was placed in a GC vial with DCM, placed in a ventilated box, concentrated overnight, and brought to 90 μL final volume in nonane (distilled in glass grade, Caledon). The extracts were capped and internal standard added prior to instrumental analysis.

Analyte concentrations were measured by Thermo Scientific DFS high-resolution MS coupled to Agilent 6890 GC or Thermo TRACE Ultra GC using Phenomenex ZB-XLB columns (60 m × 250 μm × 0.25 μm), with deactivated fused silica as guard column (1 m × 0.53 mm) and helium as carrier gas. The instrument was operated in selected ion monitoring detection mode at 280 °C inlet temperature. Ramp rates were 140–205 °C at 40 °C/min, 205–220 °C at 3 °C/min, and 205–220 °C at 3 °C/min, with equilibration time of 1 min. The injection volume was 1 μL in pulsed splitless mode and flow rate at 1.60 mL/min constant. Individual components were quantified based on internal standard quantitation using the average relative response factors from a minimum of 5-point initial calibration. Analyte concentrations were corrected by the average recoveries (41–95%) of 24 labeled surrogate standards.

2.3. Data analysis

All statistical analyses were performed using IBM SPSS version 22 software. Non-parametric multiple and pairwise comparisons were based on Kruskal-Wallis *H* test and Mann-Whitney *U* test with Bonferroni correction (Shaffer, 1995), respectively, at 0.05 significance level. PCB nomenclature follows the shorthand numbering notation (PCB 1–209) of Ballschmiter and Zell (1980). Total PCB TEQ was derived using the WHO 2005 TEFs for human health (Van den Berg et al., 2006) assuming that ND = LOD (worst-case scenario).

3. Results

3.1. Body sizes and lipid contents of fish species

Body sizes and lipid contents of the analyzed fish species are shown in Table 1. Not all species were available from the sampling areas due to differences in habitat features. Three species (*Cephalopholis argus*, *Lethrinus lentjan*, and *Variola louti*) from both Areas II and III (Table 1) allowed for locational comparisons of the measured PCB levels. Among these species, only *C. argus* markedly differed in body weight and lipid content between areas.

3.2. Congener detection rates and concentrations

The present analysis resolved 178 PCB domains comprising 152 single congeners, 21 double coeluting isomers, and 5 triple coeluting isomers from a total of 68 tissue samples at the limits of detection (LOD) of 0.1–1.8 pg/g wet weight (ww). Overall, 16 domains had 100% detection rate, whereas 78 (including 8 DLPCBs) from 50 to 99% and 23 non-detects (ND). ΣPCB, as the sum of concentrations of dioxin-like (DLPCB) and non-dioxin-like (NDLPCB) congeners assuming ND = LOD (upper-bound estimate) (Bordajandi et al., 2003), was 0.2–82.5 ng/g ww (ppb ww) (17–8450 ng/g lipid weight), with the ΣDLPCB in the range of 0.01–3.8 ng/g ww (0.9–603 ng/g lw) or 0.5–13% as mass fraction of ΣPCB. The sample frequencies by class of ΣPCB levels ($N = 68$) were: 6% (>50 ng/g ww), 25% (11–50), 19% (6–10), 44% (1–5), and 6% (<1). Log-transformed ΣPCB per wet weight and lipid weight correlated significantly ($R = 0.57$, $P < 0.05$) and ΣPCB increased log-linearly with the number of detected domains per sample (Pearson correlation $R = 0.72$, $P < 0.05$). The ΣPCB variation, however, largely depended on a few dominant congeners. ΣPCB, ΣNDLPCB, and ΣDLPCB levels on wet weight basis for each species by area are shown in Table 2.

3.3. Locational and interspecific comparisons

The sampling areas, after combining data for constituent species, significantly differed (Kruskal-Wallis *H* test) in wet weight-adjusted levels of ΣPCB ($P = 0.004$), ΣDLPCB ($P = 0.008$), and ΣNDLPCB ($P = 0.003$). Both ΣPCB and ΣNDLPCB per wet weight were significantly higher in fishes from Area I than II or III, while ΣDLPCB markedly differed between I and II only (Fig. 2). These locational differences in weight-adjusted PCB levels coincided with the spatial trend in fish lipid content, as expected (Fig. 2). In contrast, there were no significant differences (Kruskal-Wallis *H* test) in lipid-adjusted levels of ΣPCB ($P = 0.696$), ΣDLPCB ($P = 0.362$), and ΣNDLPCB ($P = 0.671$) in pooled tissue samples, regardless of species, between areas.

The fish species, regardless of source, significantly differed (Kruskal-Wallis *H* test, $P < 0.05$) in weight- and lipid-normalized ΣPCB, ΣNDLPCB, and ΣDLPCB. Grey mullet *Mugil cephalus* had the highest ΣPCB per wet weight (34–82 ng/g), whereas the grouper *C. argus* per lipid weight (663–8450 ng/g) (Fig. 3). Weight-normalized ΣPCB correlated positively, although not significantly due to small sample size, with the high lipid content (7.3–13.1%) in *M. cephalus* (Tables 1, 4). Lipid contents in pooled samples of *C. argus* were lower (0.3–2.1%) but more enriched in ΣPCB than in *M. cephalus* (Table 1, Fig. 3). ΣPCB per lipid weight declined with increasing body weight for both species and also the emperor *L. lentjan*, probably indicating dilution or reduced retention of PCBs with growth as lipid content increased with body weight for these species (Table 3).

ΣPCB levels in the grouper *Plectropomus pessuliferus*, locally called "najil" and the most popular but high-priced fish in Saudi Arabia, was consistently low and of fewer congeners. However, ΣPCB in najil increased with body weight, coinciding with an increase in lipid content with size (Table 3). Although both obtained from Area I, the milkfish *Chanos chanos* had fewer congeners and lower weight-adjusted ΣPCB

Table 2
Statistical summary of total PCB (ΣPCB) and its dioxin-like (ΣDLPCB) and non-dioxin-like (ΣNDLPCB) components in fish tissue.

				ΣPCB		ΣNDLPCB		ΣDLPCB	
Species	Area	n	Statistic	Domains[a]	ng/g ww	Domains[a]	ng/g ww	Domains[a]	ng/g ww
C. chanos	I	11	Min – Max	61–109	0.22–44.3	55–99	2.04–42.4	6–10	0.12–1.82
			Mean ± SD	86 ± 16	13.7 ± 14.9	78 ± 15	12.9 ± 14.3	8 ± 1	0.68 ± 0.58
M. cephalus	I	5	Min – Max	124–137	33.9–82.5	114–126	32.0–79.6	10–11	1.89–3.83
			Mean ± SD	129 ± 5	61.2 ± 20.7	118 ± 5	58.3 ± 20.2	11 ± 0	2.84 ± 0.69
C. bajad	II	5	Min – Max	104–133	1.85–16.0	96–122	1.73–15.0	8–11	0.10–0.99
			Mean ± SD	119 ± 12	6.81 ± 5.93	110 ± 11	6.37 ± 5.56	9 ± 1	0.42 ± 0.37
E. tauvina	II	5	Min – Max	95–126	4.20–22.9	86–115	3.90–20.8	8–11	0.28–2.04
			Mean ± SD	104 ± 13	9.16 ± 8.02	94 ± 12	8.37 ± 7.23	9 ± 1	0.75 ± 0.76
P. pessuliferus	II	5	Min – Max	31–40	0.20–24.8	29–85	0.18–23.3	2–9	0.08–1.40
			Mean ± SD	36 ± 4	7.26 ± 10.68	56 ± 26	6.80 ± 10.0	6 ± 3	0.42 ± 0.60
S. rivulatus[b]	III	4	Min – Max	56–87	3.93–9.72	52–80	3.86–9.61	4–7	0.04–0.11
			Mean ± SD	68 ± 14	6.83 ± 3.07	62 ± 13	6.76 ± 3.06	6 ± 1	0.07 ± 0.03
C. argus	II	5	Min – Max	83–112	3.49–25.3	76–101	3.21–23.5	7–11	0.24–1.81
			Mean ± SD	97 ± 12	11.1 ± 1.22	89 ± 10	10.2 ± 9.46	9 ± 2	0.87 ± 0.78
	III	4	Min – Max	81–101	10.0–23.0	72–91	9.04–20.7	7–10	0.97–2.22
			Mean ± SD	92 ± 9	15.8 ± 5.5	83 ± 8	14.2 ± 4.96	9 ± 1	1.53 ± 0.52
L. lentjan	II	5	Min – Max	69–121	2.48–9.26	61–111	2.27–8.45	7–10	0.16–0.79
			Mean ± SD	93 ± 21	4.11 ± 2.90	84 ± 20	3.74 ± 2.65	9 ± 1	0.35 ± 0.26
	III	5	Min – Max	66–113	1.60–24.3	59–102	1.50–22.1	6–11	0.08–2.16
			Mean ± SD	86 ± 19	6.97 ± 9.73	78 ± 17	6.37 ± 8.83	8 ± 2	0.57 ± 0.89
V. louti	II	9	Min – Max	62–106	1.90–14.8	55–98	1.76–13.5	6–9	0.12–1.24
			Mean ± SD	84 ± 17	5.47 ± 4.26	76 ± 16	5.04 ± 3.91	8 ± 1	0.41 ± 0.37
	III	5	Min – Max	40–107	0.80–14.7	38–97	0.79–13.7	2–10	0.01–0.96
			Mean ± SD	78 ± 25	5.10 ± 5.52	72 ± 22	4.73 ± 5.14	7 ± 3	0.33 ± 0.37

Min—minimum, Max—maximum, SD—standard deviation.
[a] Refers to number of single congeners and coeluting isomers, each counted as one.
[b] Analyzed in composite of 5 individuals each; all other species were analyzed individually.

than *M. cephalus*, but the latter had much fatter tissues (cf. Tables 1–3). The rabbitfish *Siganus rivulatus*, captured near the large sewage outfall at Area III, had relatively low ΣPCB composed mostly of PCB 11 (73–88%) (Table 2, Fig. 3).

PCBs accumulate in adipose tissue and thus tend to increase with lipid content. The trend in lipid content generally decreased (*C. chanos*, *C. bajad*, *S. rivulatus*, *V. louti*) or increased (*M. cephalus*, *E. tauvina*, *C. argus*, *P. pessuliferus*, *L. lentjan*) with increasing body weight, but significant correlations were exhibited only in *C. chanos*, *C. argus*, *P. pessuliferus*, and *L. lentjan* (Table 3, Fig. 3). Lipid-adjusted ΣPCB rose with body weight in *C. chanos*, *S. rivulatus*, and *V. louti*, indicating higher retention of PCB residues in bigger sizes of these species. As expected, lipid-adjusted ΣPCB and lipid content positively correlated for all species, although significant only in *C. chanos*, *C. bajad*, and *L. lentjan* (Table 4). The lack of statistical significance for the other species is attributed to small sample sizes.

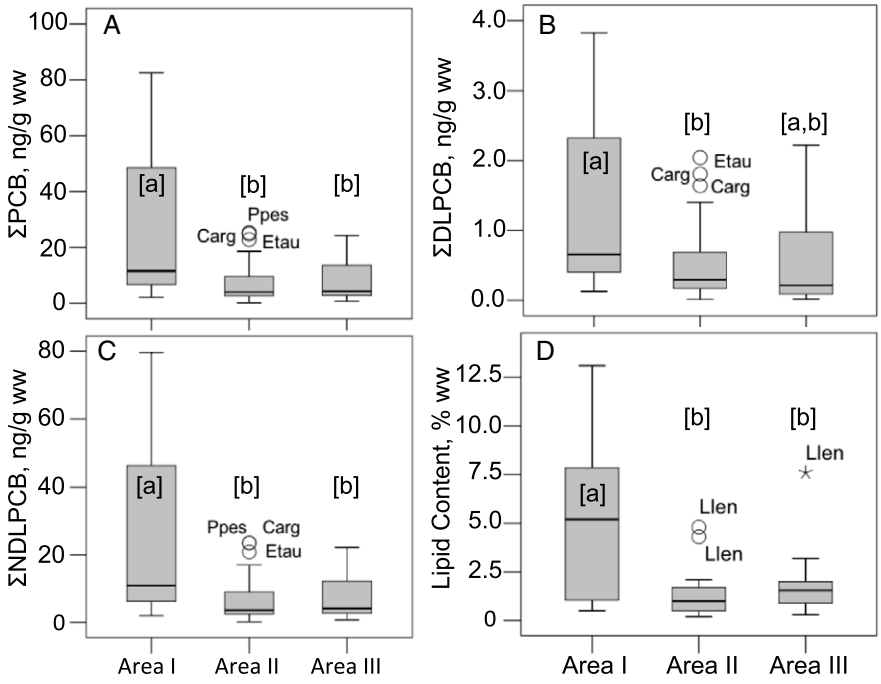

Fig. 2. Weight-adjusted levels of A) total (ΣPCB), B) dioxin-like (ΣDLPCB), and C) non-dioxin-like (ΣNDLPCB) PCBs, compared with D) lipid content in fish tissue by sampling area. Different letters in bracket indicate statistical difference at 0.05 significance level by Mann-Whitney *U* test. Outliers are indicated by circle (1.5–3 times the interquartile range) and asterisk (>3 times the IQR), labeled by species code as in Fig. 1.

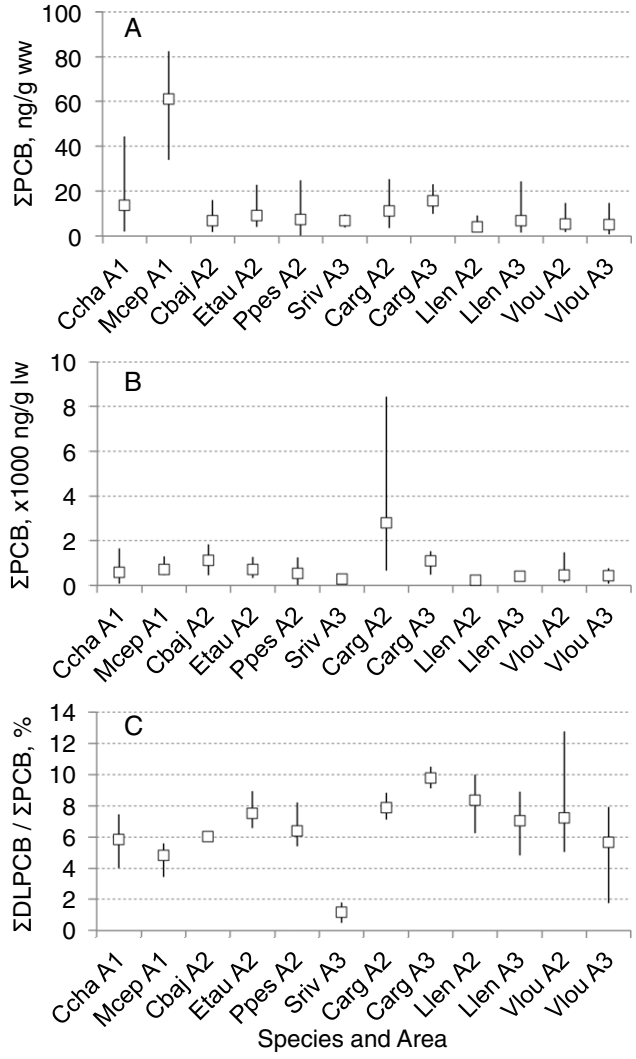

Fig. 3. Total PCB (ΣPCB) levels relative to A) wet weight and B) lipid weight, and C) dioxin-like PCBs as a percentage of total PCB (ΣDLPCB/ΣPCB) among species by area. Vertical bar and square tick indicate range and mean of concentrations in tissue samples, respectively. Species code as in Fig. 1.

3.4. Toxic equivalence (TEQ) relative to 2,3,7,8-TCDD

Total PCB TEQs, using the WHO 2005 TEFs for human health, were derived for each species by source (Table 4), with the highest in *M. cephalus* (Area I) per wet weight (1.3–2.6 pg TEQ/g ww) and *C. argus* (Area II) per lipid weight (35–238 pg TEQ/g lw). The least TEQs were in *S. rivulatus* (Area III) by wet weight (0.05–0.07 pg TEQ/g ww) and lipid weight (2–4 pg TEQ/g lw), due to consistently low ΣPCB. Non-*ortho* DLPCBs (PCBs 77, 81, 126, 169) contributed 84–100% (mean 96%) to the total PCB TEQs for all samples. The PCB TEQs were dominated by PCB 126, being the most potent and thus having the highest TEF = 0.1 among DLPCBs, with relative contributions of 58–92% (mean 74%), followed by PCB 169 with 4–34% (21%) (TEF = 0.03).

Table 4
Statistical summary of total PCB TEQs using WHO 2005 TEFs by species.

			pg TEQ/g wet weight				pg TEQ/g lipid weight			
Species	Area	n	Min	Max	Mean	SD	Min	Max	Mean	SD
C. chanos	I	11	0.10	1.06	0.47	0.34	4	51	20	13
M. cephalus	I	5	1.28	2.62	2.09	0.50	18	33	24	7
C. bajad	II	5	0.09	0.58	0.26	0.22	22	60	41	15
E. tauvina	II	5	0.12	1.29	0.43	0.49	12	71	31	23
P. pessuliferus	II	5	0.08	0.78	0.28	0.29	6	74	35	25
S. rivulatus[a]	III	4	0.05	0.07	0.06	0.01	2	4	3	1
C. argus	II	5	0.17	0.96	0.44	0.37	35	238	101	83
	III	4	0.37	1.72	0.94	0.58	18	114	66	44
L. lentjan	II	5	0.13	0.48	0.27	0.14	7	25	15	9
	III	5	0.14	1.57	0.47	0.62	18	49	31	16
V. louti	II	9	0.10	0.64	0.23	0.16	9	64	20	18
	III	5	0.11	0.66	0.26	0.23	12	35	24	10

Min—minimum, Max—maximum, SD—standard deviation, n—number of samples.
[a] Analyzed in composite of five individuals each; all other species were analyzed individually.

3.5. Congener and homolog patterns

PCB 153/168 (coeluting isomers) was the most abundant on average among tissue samples (1.6 ng/g ww), followed by PCB 180 (1.0 ng/g ww) and PCB 138 (0.9 ng/g ww). However, PCB 11 had the highest individual level (8.2 ng/g ww in *S. rivulatus*) among all congeners, but was detected in only 65% of samples and highly positively skewed (mean 0.4 ng/g ww). ΣDLPCB accounted for 0.5–12.8% of ΣPCB, being the lowest in *S. rivulatus* and highest in *C. argus* on average (Fig. 3). ΣDLPCB consisted mostly (83–99%) of mono-*ortho* congeners, among which only PCBs 118 and 105 were detected in all samples. PCB 118 contributed 37–57% to ΣDLPCB by species, followed by PCBs 105 (4–25%) and 156 (4–20%). Apparently, the predominance of PCBs 105, 118, and 156 among DLPCBs in the present samples is a typical pattern in fish (Bhavsar et al., 2007a).

PCBs are grouped into homologs, herein labeled 1CB–10CB, each comprising congeners with equal numbers but different positions of Cl

Table 3
Correlations (Pearson *R*) between fish body weight, lipid content, and PCB levels per wet weight in fish tissue.

			Body weight (g ww) vs.				Lipid content (% ww) vs.		
Species	Area	n	Lipid	ΣPCB	ΣDL	ΣNDL	ΣPCB	ΣDL	ΣNDL
C. chanos	I	11	−0.626*	−0.420	−0.418	−0.419	0.809**	0.789**	0.809**
M. cephalus	I	5	0.905*	0.562	0.873	0.547	0.362	0.801	0.344
C. bajad	II	5	−0.559	−0.616	−0.630	−0.615	0.974**	0.975**	0.974**
E. tauvina	II	5	0.507	0.562	0.585	0.560	0.834	0.836	0.834
P. pessuliferus	II	5	0.887*	0.981**	0.984**	0.981**	0.839	0.831	0.839
S. rivulatus[a]	III	4	−0.653	−0.324	0.802	−0.332	0.923	−0.071	0.926
C. argus	II + III	9	0.814**	0.418	0.599	0.399	0.211	0.398	0.193
L. lentjan	II + III	10	0.912**	0.977**	0.980**	0.977**	0.852**	0.860**	0.851**
V. louti	II + III	14	−0.253	0.256	0.434	0.239	0.286	0.264	0.287

n—number of samples; ΣPCB, ΣDL, and ΣNDL indicate the total concentrations of all PCB congeners, 12 dioxin-like congeners, and all non-dioxin-like congeners, respectively, normalized to wet weight.
[a] Analyzed in composite of 5 individuals each, all other species were analyzed individually.
* Significant at 0.05 level.
** Significant at 0.01 level.

atoms (isomers). The moderately chlorinated homologs (5CB–7CB), mainly hexachlorobiphenyls (6CB), were the most abundant, but some less chlorinated isomers (1CB–3CB), notably PCB 11, 18, 28, and 31, were surprisingly elevated in *S. rivulatus* and *M. cephalus* (Fig. 4).

We combined the detection rate and mean relative concentration of each domain across samples and ranked them in decreasing order to identify the dominant PCBs by both occurrence and abundance. The top 12 (iPCB-12) and 20 (iPCB-20) domains were assessed for their relative contributions to ΣPCB. iPCB-12 comprised (in order of decreasing rank) PCBs 153/168, 180, 138, 28, 11, 43/52, 18, 31, 187, 90/101, 48/49, and 118; while iPCB-20 included iPCB-12 plus PCBs 44, 147/149, 170, 146, 70, 110, 183, and 22. iPCB-12 and iPCB-20 accounted for 45–89% and 60–91% of ΣPCB on average by species, respectively, and both highly correlated with ΣPCB ($R = 0.986$ for iPCB-12, $R = 0.998$ for iPCB-20), well representing the ΣPCB in *S. rivulatus* but were highly variable in *V. louti* and *P. pessuliferus*.

4. Discussion

4.1. Comparisons of total PCB levels and TEQs in fish tissue

ΣPCB levels (0.2–82.5 ng/g ww) in this study were as much as three orders of magnitude lower than those reported in fishes from chronically polluted freshwater (Jackson et al., 2001; Šalgovičová and Zmetáková, 2006; Bhavsar et al., 2007b; Roosens et al., 2008; de Boer et al., 2010), estuarine (Fu and Wu, 2005; Colombo et al., 2007), and coastal (Porte and Albaigés, 1993; Roach and Runcie, 1998) sites worldwide. Specifically, ΣPCB averaged up to 108,752 ng/g ww in fishes from a lake impacted by leaks from a PCB production plant in Slovakia (Šalgovičová and Zmetáková, 2006). ΣPCB levels were up to 5000 ng/g ww in trout from lakes in Canada (Bhavsar et al., 2007b) and 15,000 ng/g ww in a detritus-feeding fish from an estuary in Argentina (Colombo et al., 2007). ΣPCB averaged up to 10,140 ng/g ww in fishes, including *M. cephalus*, from Georges/Cooks Rivers and Sidney Harbor, Australia (Roach and Runcie, 1998).

ΣPCB levels in this study were comparable to those found in fishes and bivalves, by converting from dry to wet weight basis, from the United Arab Emirates, Bahrain, Kuwait, Qatar, and Oman (de Mora et al., 2010) and in fishes from harbors in Guam (Denton et al., 2006); an estuary and aquaculture farms in China (Nie et al., 2005, 2006); Ise Bay (Matsuo et al., 2009) and Ariake Sea (Kobayashi et al., 2010), Japan; Moreton Bay, Australia (Matthews et al., 2008); and rivers in Spain (Bordajandi et al., 2003) and Pakistan (Eqani et al., 2013). The present ΣPCB levels were within the range of data for fish species "from marine ecosystems with no known source of local contamination", as compiled by Niimi (1996). Our ΣPCB levels, however, exceeded those found in marine fishes (<1 ng/g ww) from the Yemeni waters of Red Sea (Al-Shwafi et al., 2009).

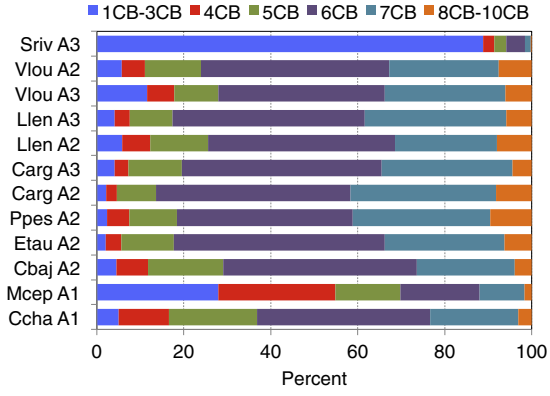

Fig. 4. Percent composition of PCB homologs (1CB–10CB) among species by area. Species code as in Fig. 1.

The above comparisons suggest that the ΣPCB levels in this study were generally at the lower end of global range. In addition, the present ΣPCB levels were below the tolerance levels for seafood in the United States, Canada, and China (2000 ng/g ww), Japan (3000 ng/g ww for coastal species), and Australia and New Zealand (500 ng/g ww) (USFDA, 2001; CFIA, 2005; FSANZ, 2006; Wu et al., 2008). Exceedance of these limits triggers necessary actions, such as further investigation to ascertain levels, impacts, and sources; issuance of seafood consumption advisories; or trade regulation, depending on the country.

Our range of total PCB TEQs (0.05–2.6 pg TEQ/g ww or 2–238 pg TEQ/g lw) was comparable to those found in fish and shellfish from the South Korean markets (0.004–2.7 pg TEQ/g ww range of means by seafood category) (Lee et al., 2007). PCB TEQs in fish and invertebrate species from the Atlantic southwest coast of Spain (0.04–0.97 pg TEQ/g ww) (Bordajandi et al., 2006) were exceeded entirely by our TEQs in *M. cephalus* (Area I) and partly in *C. chanos* (I), *E. tauvina* (II), *C. argus* (II–III), and *L. lentjan* (III). PCB TEQs measured by Matthews et al. (2008) in fishes from Moreton Bay (up to 110 pg TEQ/g lw) were higher than our study, although the ΣPCB levels were comparable between studies. Our PCB TEQs were also far lower than in fishes from certain locations in the US (4.5–107.2 pg TEQ/g ww, as cited in Judd et al., 2004) and Germany (3.1–44.3 pg TEQ/g ww in fishes from surface waters in the inner city of Hamburg, Götz, 2012). The global data on PCB TEQs in seafood compiled by Domingo and Bocio (2007) were mostly comparable to our study, except for the higher levels in salmon (3.7–15.6 pg TEQ/g ww) and herring (up to 7.36 pg TEQ/g ww) from the Baltic region (Isosaari et al., 2006). Note that the TEQ values cited above were based on the WHO 1998 TEFs, except for those by Matthews et al. (2008) who used the revised WHO TEFs in 2005 (Van den Berg et al., 1998, 2006).

The European Union (EU) legislated the maximum TEQ limit in muscle of wild caught fish at 3.5 pg TEQ/g ww for polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) and 6.5 pg TEQ/g ww when including the PCB TEQ (EFSA, 2012). The upper-bound TEQs for PCDDs/PCDFs that we simultaneously measured in the same fish species during this study (data not shown) had a maximum of 0.63 pg TEQ/g ww using WHO 2005 TEFs. This suggests that the total TEQs from PCBs and PCDDs/PCDFs in the present fish species were substantially lower than the EU limit. Many previous studies found that the total TEQs in seafood are mostly due to PCBs. In the study by Lee et al. (2007) as cited above, PCBs contributed 51–100% to total TEQs in 31 fish and shellfish items examined. The global data on total TEQs in fish and fish products compiled by Alcock et al. (1998) showed higher contributions from PCBs (50–92%) than PCDDs/PCDFs by wet and lipid weights. PCBs accounted for 51–81% of the total TEQs in most fish species studied by Bordajandi et al. (2003), whereas 81% in mussel from Catalonia, Spain by Eljarrat et al. (2002). These results indicate that seafood has significant contribution to the human dietary intake of PCBs, along with PCDDs/PCDFs.

4.2. Congener patterns and implications to monitoring

Most ΣPCB levels cited above were based on a few selected congeners. Matsuo et al. (2009) analyzed 205 congeners (1CB–8CB) in coastal food web and found that the sums of selected congeners in other studies underestimated the ΣPCB estimates by 16 to >60%. Hence, the differences in number of analyzed congeners led to ambiguous comparisons of ΣPCB between studies. Congener-specific analysis is vital to health risk assessment, as ΣPCB toxicity depends on concentration and conformation of the component congeners (Jones, 1988; Connor et al., 2005). However, congener-specific method entails high cost and effort, potentially constraining the full analysis of PCBs in many samples. For practical and economic reasons, therefore, PCB monitoring is frequently based on a few selected congeners (Henry and DeVito, 2003).

In Saudi Arabia, researchers and regulators have interest in selective biomonitoring of PCBs in major fisheries resources, as the high number

of species of concern over a large geographic area entails tremendous cost. The monitoring of all PCB congeners remains a more desirable approach as it provides greater informative value and better use for decision-making, but the cited constraints may limit future initiatives. When resorting to selective monitoring, the top congeners in this study could adequately serve the purpose but subject to interpretative caution. In fact, iPCB-12 includes the seven congeners, herein termed ICES-7 (PCBs 28, 52, 101, 118, 138, 153, and 180), which are used as marker congeners for PCB monitoring and regulation in Europe. Using our present data, the ICES-7 congeners (along with the coeluents PCBs 52, 101, and 153) accounted for 6–55% of ΣPCB on average by species. With iPCB-12, the ΣPCB representation is increased to 45–89% on average by species (Fig. 5). This suggests that a selective monitoring using ICES-7 substantially underestimates the ΣPCB in fishes from our study area. The lower ΣPCB representation by ICES-7 is due to its failure to account for the less chlorinated congeners, mainly PCBs 11, 18 and 31, which were dominant in some fishes, notably *S. rivulatus* and *M. cephalus*. The additional congeners (PCBs 22, 44, 70, 110, 146, 147/149, 170, and 183) in iPCB-20 further improved the mean ΣPCB representation to 60–91% by species (Fig. 5).

McFarland and Clarke (1989) recommended 36 PCB domains, herein termed MC-36, for monitoring due to their persistence and potency, which were used for PCB assessment in estuarine and aquaculture fishes in China (Nie et al., 2005, 2006). With our data, MC-36 (including 9 coeluents) accounted for 9–79% of ΣPCB on average by species, largely underrepresenting the ΣPCB in *S. rivulatus* by 73–89% and *M. cephalus* by 6–14% relative to iPCB-20. MC-36 shared 14 domains with iPCB-20 and better accounted for ΣPCB by as much as 8% in *C. argus*, *E. tauvina*, *L. lentjan*, and *P. pessuliferus*. Miao et al. (2000) used the sum of PCBs 28, 31, 44, 52, 101, 118, 138, 153, 180, and 194, herein termed FFS-10, to compare the ΣPCB levels between marine species from the French Frigate Shoals, North Pacific Ocean. Using our data, FFS-10 amounted to 7–58% of ΣPCB on average by species, underestimating the ΣPCB in *S. rivulatus* by 74–89% (2–10% for all other species) relative to iPCB-12.

The high correlations of iPCB-12 ($R = 0.986$) and iPCB-20 (0.998) with ΣPCB suggest that both congener sets serve as good signatures of PCB trends in fish tissue from the present study area. Except for PCB 11, the component congeners of these indicator PCBs also individually correlated significantly with ΣPCB ($R = 0.64$–0.93), thus indicating their relative consistency as major constituents of ΣPCB burdens in the examined fishes. While these results imply the potential utility of iPCB-12 and iPCB-20 for selective monitoring, a full congener approach remains necessary and more advantageous given the need for greater information to enable a more reliable health risk assessment. Thus, a selective congener set comprising at least the NDLPCBs in iPCB-12, plus all DLPCBs, may prove useful for future biomonitoring when the full congener approach is reasonably constrained. Such a set of indicator PCBs allows for comparisons with monitoring results based on ICES-7 and also enables the conduct of TEQ-based human health risk assessment.

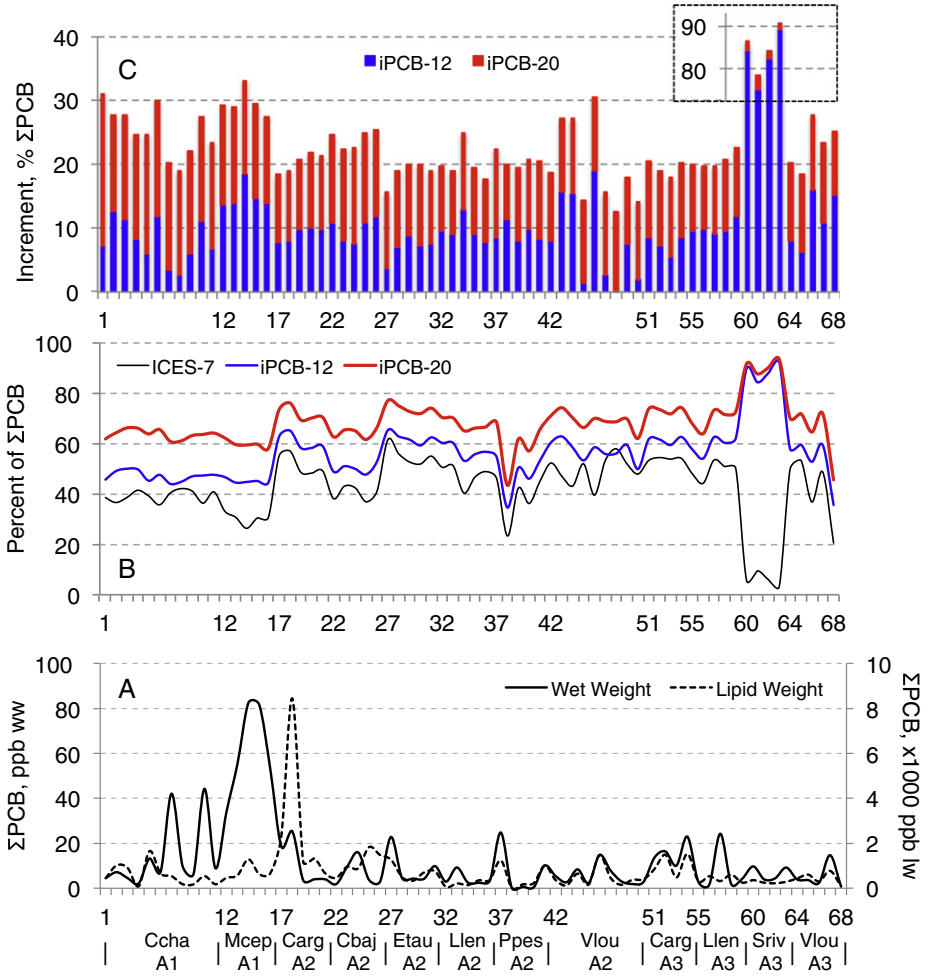

Fig. 5. Total PCB (ΣPCB) representation of top 12 (iPCB-12) and 20 (iPCB-20) congeners ranked by both abundance and occurrence. A) ΣPCB by species and area, B) ΣPCB representation by iPCB-12 and iPCB-20 compared with ICES-7 (7 marker congeners used in Europe), and C) Mean percent increments in ΣPCB representation by iPCB-12 and iPCB-20 relative to ICES-7 by species and area. Species code as in Fig. 1.

4.3. Potential sources and future concerns

The top PCBs in this study, except for the coeluents PCBs 43, 90, and 168, are the most frequently reported congeners in the environment and biota (Jones, 1988; Hansen, 1998). PCBs 138, 153, and 180 are generally dominant in fish (Eisler and Belisle, 1996) and human tissue (Černá et al., 2008; Fernandez et al., 2008). PCB 153 likely constituted all, if not most, of PCB 153/168 in our samples, as PCB 168 is not known to concentrate in fish tissue (Nie et al., 2005, 2006). A previously identified coelution of PCB 153/168 in liver of a coastal fish was later found to be composed entirely of PCB 153 (Zeng et al., 2002). PCB 153 is not metabolized, thus it is highly persistent and bioaccumulates in animal and human tissue (Eisler and Belisle, 1996). In contrast, the less chlorinated congeners accumulate rapidly but only transiently in lipid-rich tissue (Karjalainen et al., 2006). The elevated levels of PCB 11 in *S. rivulatus* from Area III, which has the largest sewage outfall in the region, deserve particular notice. PCB 11 is a non-legacy signature of PCB contamination, being absent in commercial PCB formulations, but largely originates as a byproduct of paint and pigment production and a residue in printed products (Hu and Hornbuckle, 2010; Hu et al., 2014) although PCBs 28, 52, 77 and 9CB–10CB congeners have also been detected in pigments and consumer products (Vorkamp, 2016). It was deemed that PCB 11 was commonly overlooked in previous PCB analyses (Vorkamp, 2016), but is now known to be a ubiquitous airborne contaminant and widely present in aquatic systems by way of atmospheric fallout and wastewater discharges (Rodenburg et al., 2010; Yao et al., 2014). Like the other less chlorinated congeners that have been in use as constituents of commercial PCB products, PCB 11 does not bioaccumulate in biota due to rapid metabolism but its biotransformation can yield metabolites that may persist and cause adverse effects in animals and humans (Zhu et al., 2013; Hu et al., 2014).

In most fish species we examined, the homolog profiles were dominated by 5CB–7CB, but mainly 6CB (Fig. 4), which are the most bioaccumulating PCBs (McFarland and Clarke, 1989). Apparently, the dominant congeners in the present tissue samples were also key constituents of commercial PCB mixtures, e.g. PCBs 18, 28, and 31 in Aroclors 1016 and 1242; 52 in Aroclor 1248; 101, 118, and 138 in Aroclor 1254; and 153, 180, and 187 in Aroclor 1260 (Jones, 1988; Ishikawa et al., 2007). To our knowledge, there is no available information on PCBs inventory in Saudi Arabia to allow for comparative reference. In addition, the present congener profiles might not reflect those of contaminant sources due to PCB transformations in the environment and biota. While the less chlorinated congeners, being volatile and also readily metabolized and excreted, have been considered as indicators of recent PCB inputs (Colombo et al., 2007), it remains to be ascertained if such congeners in our samples were breakdown products or due to fresh loadings in the study area.

There are no known natural sources of PCBs, thus contamination of fish tissue is due to anthropogenic causes. PCBs, mostly higher chlorinated congeners, in human adipose tissue, blood serum, and breast milk are mainly derived from food of animal origin, with significant contributions from contaminated seafood (Dougherty et al., 2000; Černa et al., 2008; Dórea, 2008; Goncharov et al., 2008; Ludewig et al., 2008). The less chlorinated PCBs, although transient, should not be dismissed to a lesser concern for human health risks due to the potential adverse effects of their metabolites (Li et al., 2010). Hence, PCB biomonitoring should afford attention to the dominant congeners, regardless of degree of chlorination. This justifies the utility of our iPCBs, in addition to all DLPCBs, for any future selective monitoring. PCB carcinogenicity is due to the potency of DLPCBs that is mediated by binding to the aryl hydrocarbon receptor (AhR) similar to the mode of action of PCDDs/PCDFs (WHO, 2003; Lauby-Secretan et al., 2013). Hence, the inclusion of all DLPCBs in selective monitoring should adequately serve the minimum needs for future PCB assessments.

There are various potential sources of chemical contaminants, including PCBs, along the Saudi Red Sea coast, such as industrial complexes producing metals and plastics, paints and pigments, construction materials, pharmaceuticals, electric appliances, and various other products. Municipal and industrial wastewater treatment plants discharge effluents at 300,000 m^3 daily, of which 42% are released through the submerged outfall in Area III (RPI/PTL, 2008). A previous study has revealed the historical rise in sewage discharges into the coastal zone in the Jeddah area (Risk et al., 2009). Reject water is also discharged from desalination plants, with a total capacity of 3.2 million m^3 along the Saudi Red Sea coast, of which 17% come from the study area (Lattemann and Höpner, 2008). The largest port in the Red Sea operates in Jeddah, which handled a total throughput of 4700 vessels, mostly large container ships, in 2011 (SPA, 2012). Interesting to note that the PCBs detected in atmospheric particulates at an inland metropolitan area (Riyadh) in Saudi Arabia were also dominant congeners in the tissue of the examined fishes. Further study to identify the actual sources of PCBs is thus vital to avert any further buildup of contaminants to deleterious levels in comestible marine resources. An expanded spatial scale of future monitoring is necessary to ascertain the extent and test the reliability of the marker congeners in tracking the trends of PCB contamination along the Saudi Red Sea coast.

5. Conclusion

For the first time, this study determined the full PCB profiles in edible fish tissue from the Saudi Red Sea coast. ΣPCB levels were at the lower end of reported global range and far below the prescribed tolerance thresholds for PCBs in other countries. Moderately chlorinated congeners, mostly hexachlorobiphenyls, dominated the PCB profiles, but some fish species were also exposed to elevated levels of less chlorinated congeners. It remains to be ascertained if the latter congeners are breakdown products of legacy PCB residues or due to fresh inputs. Our results identified a few dominant congeners that could serve as markers of PCB contamination in fish tissue within the study area. While the monitoring of all PCBs is vital and thus recommended, such a set of indicator congeners, in addition to all DLPCBs, may be useful for selective biomonitoring in case of reasonable constraints on full congener approach. Human health risk assessment of PCBs and PCDDs/PCDFs in seafood, including the present species, is underway to reinforce the results and implications of this study.

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