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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

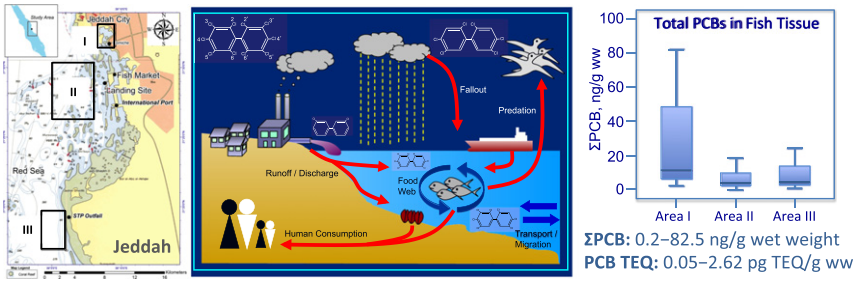
Zenon B. Batang [a,*], Nabeel Alikunhi [a], Michael Gochfeld [b], Joanna Burger [c,d], Ramzi Al-Jahdali [a], Haitham Al-Jahdali [a], Mohammed A.M. Aziz [f], Dalal Al-Jebreen [e], Abdulaziz Al-Suwailem [a]

[a] Coastal and Marine Resources Core Laboratory, King Abdullah University of Science and Technology, Thuwal 23955, Saudi Arabia
[b] Environmental and Community Medicine, Robert Wood Johnson Medical School, Rutgers University, Piscataway, NJ 08854, USA
[c] Division of Life Sciences, Rutgers University, Piscataway, NJ 08554, USA
[d] Environmental and Occupational Health Sciences Institute, Rutgers University, Piscataway, NJ 08854, USA
[e] Department of Nutrition and Food Science, Princess Nora bint Abdul Rahman University, Riyadh 11564, Saudi Arabia
[f] Directorate of Aquatic Environment, Ministry of Environment, Water and Agriculture, Riyadh 11564, Saudi Arabia

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

* Corresponding author.
E-mail address: zenon.batang@kaust.edu.sa (Z.B. Batang).

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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