ABSTRACT

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Polychlorinated biphenyls (PCBs) which are man-made chemicals produced in the 1930s to 1970s have been reported to bioaccumulate in lipid-rich tissues of apex predators, making them ubiquitous threat to the marine environment. However, there remains an apparent paucity in PCB information especially dioxin-like PCB accumulation, their corresponding toxic equivalents (TEQs) and associated pathologies in dolphins and whales particularly found in the Philippines. Similarly, the unique opportunities that cetacean stranding events offer must be maximized to probe into various research questions, particularly towards One Health. Thus, to elucidate the possible link between PCB toxicological profiles with risk assessment and pathological changes, sampling and analyses of blubber and liver tissues from 30 stranded individual cetaceans, comprising seven species, found along Philippine coastal waters during January 2013 – February 2016, were conducted.

The PCB concentration levels in the cetacean tissues were determined using a pre-validated modified method that subjected homogenized tissues to extraction, series of cleanup procedures and macro- and micro-concentration steps prior to analysis by gas chromatography/mass spectrometry. Total PCB concentrations, i.e., the sum of 40 PCB congeners ($\Sigma PCBs_{40\ congeners}$) in the blubber tissues ranged from 22 ng/g lipid weight (ng/g lw) of an adult female dwarf sperm whale to 2,375 ng/g lw of an adult male Fraser's dolphin. In the liver, $\Sigma PCBs_{40\ congeners}$ ranged from 1.0 ng/g lw of an adult female Fraser's dolphin to 656 ng/g lw of an adult male false killer whale. Overall, the $\Sigma PCBs_{40\ congeners\ (blubber\ +\ liver)}$ ranged from 22 ng/g lw (in an adult female dwarf sperm whale) to 2,802 ng/g lw (in an adult male spinner dolphin), both found in Camarines Sur. Significant differences were observed among categories of sex (F (1, 25) = 32.12, p = .000, α = 0.05) and combined age class and sex (F (1,25) = 41.28, p = .000, α = 0.05), using factorial ANOVA. Tetra- to hexa-chlorinated biphenyls were the major homologs, accounting for 50 - 90% of the total PCBs. The most predominant congeners were PCBs 44 and 52 (tetra-CBs), PCB 99 (penta-CB) and PCB 153 (hexa-CB) all of which were important components of the technical formulations of the toxic Aroclor compounds. The toxicity risk measured as 2,3,7,8-tetrachlorodibenzo-*p*-dioxin

equivalents (TEQs) ranged from 0.00 to 4,351 *pg*/g lw (adult male Fraser's dolphin (Pangasinan)). In this analysis, the greatest TEQ contributors were the non-*ortho* congeners PCBs 126 and 169 (detected in 19/30 animals, 63%). Furthermore, 19/30 animals (63%) have TEQs exceeding the minimum threshold of 160 *pg*/g lw for the onset of physiological effects in marine mammals while 18/30 (60%) have higher TEQs than the maximum threshold of 1400 *pg*/g lw. Generally, PCB concentrations and TEQs were higher in adult males than adult females suggesting maternal toxic burden transfer to calves during gestation and lactation. Significant result was observed (Kruskal Wallis $H = 0.007$; $\alpha = 0.05$) from the analysis of relationship between presence of lesions/TEQs across categories of age class/sex. Lesions observed were fibrosis (58%, 11/19), edema (53%, 10/19), hemosiderosin or porphyrin accumulation (47%, 9/19), plerocercoid cyst-like formations (47%, 9/19), microhemorrhages (42%, 8/19), hypercontracted myofibers (26%, 5/19), steatosis (16%, 3/19), hepatic hyperplasia (11%, 2/19), hepatic sinusoidal congestion (11%, 2/19), necrosis (11%, 2/19), granulomatous panniculitis (11%, 2/19), endomysial atrophy (5%, 1/19), bile duct proliferation/embolism (5%, 1/19), endomysial atrophy (5%, 1/19), granuloma (5%, 1/19), scattered vascular channels (5%, 1/19) and steatitis (5%, 1/19). All 19 individuals with high TEQs had severe histopathological changes due to combined lesional findings, implying high risk of cetaceans found in the country to dioxin-like PCBs. Overall, this work was successful in establishing and validating an easy-to-follow, environmental-friendly and cost-effective analytical method for quantifying PCB concentrations, at trace levels, in lipid-rich tissues of cetaceans. Furthermore, complete elucidation of and correlation between TEQs of the most potent non- and mono-*ortho* PCB congeners and pathological findings were accomplished. With the obtained baseline toxico-pathological information for the Philippines, empirical evidence of potential adverse effects of PCB contamination in cetaceans to human health are evident. These data contribute a larger impact to some Philippine locals who are reported to be using these stranded cetacean species as food source. Future ecotoxicological research efforts should consider in-depth investigation of dioxin-like PCBs and other endocrine disrupting compounds in wildlife and humans where conduct of clinical trials is also warranted.

Keywords: polychlorinated biphenyls cetacean toxic equivalents lesions Philippines