

Do Polychlorinated Biphenyls (PCBs) Banned in 1970s Still Persist in the Marine Environment? Levels, Patterns, and Possible Toxicity Manifestations in Stranded Cetaceans Found in the Philippines

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ABSTRACT

Polychlorinated biphenyls (PCBs) which were commercially produced in the 1930s to 1970s have been reported to bioaccumulate in lipid tissues of apex predators. However, there is apparent paucity in information regarding PCBs especially dioxin-like PCB accumulation, their corresponding toxic equivalents (TEQs) and associated histopathologies in dolphins and whales particularly found in the Philippines. Cetacean stranding events in the country provide unique opportunities to probe into various research questions, particularly towards One Health. Thus, to elucidate the possible connection between PCB toxicological profiles with risk assessment and pathological changes in tissues, we initiated a sampling of blubber and liver tissues from 30 stranded individual cetaceans. The present paper uncovers important and significant results from 17 individuals, comprising seven species, out of the 30 sampled cetaceans found stranded along Philippine coastal waters during 2013-2015. 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 38 PCB congeners (ΣPCBs_{38} congeners) in the blubber tissues ranged from 21.7 ng/g lipid weight of an adult female dwarf sperm whale to 1850 ng/g lipid weight of an adult male spinner dolphin, both found in Camarines Sur. These two stranders also revealed the lowest (21.7 ng/g lipid weight) and highest (1851 ng/g lipid weight) ΣPCBs_{38} congeners from combined blubber and liver concentrations per individual. Total PCB concentrations (ΣPCBs_{38} congeners) in the liver ranged from 1.0 ng/g lipid weight of an adult male spinner dolphin (Camarines Sur) to 143.4 ng/g lipid weight of an adult female Fraser's dolphin (La Union). Significant differences in total PCB concentrations were observed among categories of age class and gender, i.e., ΣPCBs_{38} congeners, adult male > sub-adult male > sub-adult female > adult female > calf male (Kruskal Wallis, $H = 0.011$). For species category, ΣPCBs_{38} congeners were found to be in the order of spinner dolphin > Blainville's beaked whale > rough-toothed dolphin > Fraser's dolphin > Cuvier's beaked whale > Risso's dolphin > dwarf sperm whale ($H = 0.006$). For species type, ΣPCBs_{38} congeners was in the order of delphinids > beaked and sperm whales ($H = 0.007$). Tetra- to penta-chlorinated biphenyls were the dominant homologs, accounting for about 60% of the total PCBs. The most abundant congeners were PCBs 44 and 52 (tetra-CBs) and PCB 99 (penta-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 0.94 pg/g lipid weight of a sub-adult male spinner dolphin (Ilocos Sur) to 6,131 pg/g lipid weight of a sub-adult female spinner dolphin (Cebu) indicating the presence of the more toxic PCB congeners in the sub-adult female albeit the lesser number of total PCB congeners found in the mammal. In this analysis, the greatest TEQ contributors were the mono-*ortho* congener PCB 126 ($\bar{x} = 87\%$) and the non-*ortho* congener PCB 169 ($\bar{x} = 43\%$). Furthermore, 59% (10/17 individuals) have TEQs exceeding the minimum threshold of 160 pg/g lipid weight for the onset of physiological effects in marine mammals while 24% (4/17) have

higher TEQs than the maximum threshold of 1400 pg/g lipid weight. Generally, PCB concentrations and TEQs were higher in adult males than adult females suggesting maternal toxic burden transfer to calves during gestation and lactation before possible occurrence of contaminant re-accumulation later in their lives. Significant result was observed ($H = 0.001$) from the analysis of relationship between prevalence of lesions (none, moderate, severe) and TEQs. Lesions observed were edema, 12% (2 out of 17 individuals), hemorrhages, 18% (3/17), hypercontracted myofibres, 12% (2/17), plerocercoid cyst-like formations, 12% (2/17), embolism and bile duct proliferation, 0.06% (1/17), hemosiderosis, 35% (6/17), hepatic periportal fibrosis, 12% (2/17), hepatic sinusoids and fatty infiltration, 0.06% (1/17), hepatocytic and pancreatic vacuole accumulation, 0.06% (1/17), hepatocytic lipid accumulation, 0.06% (1/17), inflammation and infection, 0.06% (1/17), hepatic hyperplasia, 0.06% (1/17), and steatosis, 0.06% (1/17). Individuals with severe histopathological changes had the highest TEQs. Note, however, that essential to establishing a more conclusive account on the cause and effect of PCBs in cetaceans, conduct of clinical trials are warranted. Overall, results of this study provide empirical evidence of potential adverse effects of PCB contamination in cetaceans to human health and contribute a larger impact to Philippine locals who are reported to be using these stranded cetacean species as human food source.

Keywords: polychlorinated biphenyls cetacean histopathology Philippines