Spatial trends of polybrominated diphenyl ethers (PBDEs) in loggerhead sea turtle eggs and plasma

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Introduction

Polybrominated diphenyl ethers (PBDEs) are flame retardants that have been in use since before 1970. PBDEs leach from consumer products and like polychlorinated biphenyls (PCBs), they have become global environmental contaminants bioaccumulating into animal tissues and showing toxic effects in laboratory animals [1]. PBDEs have been measured in several species of wildlife, including marine mammals and seabirds, but not in sea turtles or in any other reptile. The loggerhead sea turtle (*Carettacaretta*) is listed on the U.S. Endangered Species Act and feeds on crustaceans and bivalves. This species has been utilized in several recent heath assessment studies to monitor contaminant concentrations (mercury, PCBs, pesticides, perfluorinated compounds) and to assess the potential effects of these contaminants on its health [2-5].

Materials and Methods

Plasma samples were collected from 29 immature loggerhead turtles captured within 8 miles of the shoreline along the coasts of South Carolina (SC), Georgia (GA), and northeastern Florida (FL) in the summer of 2003. Unhatched eggs from 37 nests were collected from beaches in North Carolina (NC), eastern FL, and western FL in the summer of 2002. Yolks of eggs containing only early and middle stage embryos from each nest were pooled. Samples were spiked with internal standards prior to extraction. Plasma was extracted with formic acid, hexane, and dichloromethane (DCM). Yolk was extracted with DCM using pressurized fluid extraction. Extracts were cleaned up with size exclusion chromatography and alumina columns. Thirteen PBDE congeners (BDE 17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190) as well as approximately 85 PCB congeners and 20 pesticides were quantified using gas chromatography with electron capture detection (GC-ECD) and mass spectrometry (GC/MS). PBDEs were measured by GC/MS in electron impact mode using a 60 m x 0.25 mm column containing a non-polar phase. PBDE 209 will be quantified using a shorter column by GC/MS in negative chemical ionization mode.

Results and Discussion

Plasma concentrations of total predominant PBDEs (ΣBDE = sum of PBDE 47, 99, 100, 153, and 154) in the immature turtles averaged 85.8 ng/g lipid (131 pg/g wet mass). By comparison, the plasma concentrations of total predominant PCBs (ΣPCBs = sum of PCB 99, 118, 138, 153+132, 170, 180+193, 187, 196+203, and 199) were much higher and averaged 2780 ng/g lipid (2530 pg/g wet mass). ΣBDE concentrations were significantly but weakly correlated with ΣPCB concentrations (r_S=0.371; p=0.048). The ΣBDE concentrations in turtle plasma were higher than those measured in occupationally exposed humans (reviewed by Hites [6]). The PBDE congener pattern in plasma was 60% BDE 47, 15% BDE99, 15% BDE100, 5% BDE 153, and 5% BDE 154. This pattern is similar to the pattern seen in several species of fish and somewhat similar to human blood although BDE 153 often makes up a larger proportion in human blood (reviewed by Hites [6]). ΣBDE concentrations were not correlated with capture latitude (p>0.05), indicating that there was no north-south trend as was seen with perfluorinated compounds in these turtles [5]. However, the seven turtles with the highest ΣBDE concentrations (top 25% of turtles) were captured in two clustered areas offshore of Brunswick, GA (31.0 to 31.3 ^{o}N) and Charleston, SC (32.6 to 32.7 ^{o}N).

Geographical differences were seen in the ΣBDE concentrations in the loggerhead egg samples. The NC nests had

significantly higher concentrations of ΣBDE (13.5 ng/g lipid) compared to eastern FL (2.23 ng/g lipid) and western FL nests (0.815 ng/g lipid; ANOVA p=0.001). This geographical pattern was also observed with ΣPCBs and organochlorine pesticides [3]. The geographical difference is likely due to different habitat choices during the foraging seasons. Satellite tagging studies confirm that most females nesting in GA, SC, and NC (the northern subpopulation) migrate to northern Atlantic Ocean waters to forage during the non-nesting season [7,8], while the females nesting in FL migrate to more southern waters in the Gulf of Mexico, FL coastal waters, and the Caribbean Sea [9]. These geographical differences may be important because the northern subpopulation, the one with higher contaminant concentrations, has been declining over the last three decades compared to the stable or increasing south FL subpopulation.

Similar to the plasma samples, the ΣBDE concentrations measured in the egg samples were significantly correlated with ΣPCBs (r_S=0.693; p<0.0001). The egg ΣBDE concentrations were much lower than concentrations commonly reported for bird eggs (reviewed by Hites [6]). The pattern of individual BDE congeners in the NC eggs (those with the most detectable concentrations) was 34% BDE 47, 4% BDE 99, 27% BDE 100, 4% BDE 153, and 31% BDE 154. This pattern is unique compared to patterns reported for bird eggs (reviewed by Hites [6]) and was different from the pattern observed in the loggerhead plasma. The reason is unclear for these different patterns, but could include different sources of exposure (adult females feed in a different location than the immatures), different elimination rates, different biotransformation rates, or differential reproductive transfer into eggs.

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