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Tetrabromobisphenol A (TBBPA) and hexabromocyclododecanes (HBCDs) in tissues of humans, dolphins, and sharks from the United States

Boris Johnson-Restrepo^{a,1}, Douglas H. Adams^b, Kurunthachalam Kannan^{a,*}

^a Wadsworth Center, New York State Department of Health, and Department of Environmental Health Sciences, School of Public Health, State University of New York at Albany, Empire State Plaza, P.O. Box 509, Albany, NY 12201-0509, USA ^b Cape Canaveral Scientific, Inc., 220 Surf Road, Melbourne Beach, FL 32951, USA

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Abstract

Concentrations of tetrabromobisphenol A (TBBPA) and α -, β -, and γ -isomers of hexabromocyclododecanes (HBCDs) were determined by liquid chromatography-tandem mass spectrometry (LC-ESI-MS/MS) in human adipose tissue obtained in New York City, and in three marine top-level predators - bottlenose dolphin, bull shark, and Atlantic sharpnose shark - collected from coastal waters of Florida, USA. The overall mean concentrations (mean \pm SD) of TBBPA and HBCDs were 0.048 \pm 0.102 and 0.333 \pm 0.571 ng/g lipid wt in human adipose tissue samples, 1.2 ± 3 and 7.38 ± 18 ng/g lipid wt in bottlenose dolphin blubber, 9.5 ± 12 and 77.7 ± 128 ng/g lipid wt in bull shark muscle, and 0.872 ± 0.5 and 54.5 ± 88 ng/g lipid wt in Atlantic sharpnose shark muscle. Overall mean concentrations of HBCDs were 5–10-fold higher than mean TBBPA concentrations, in all of the samples analyzed. The highest concentrations of TBBPA and HBCDs were detected in the bull shark muscle at concentrations of 35.6 and 413 ng/g, lipid wt, respectively. Concentrations of TBBPA and HBCDs, after log-transformation, were significantly correlated with each other in human adipose tissue and bottlenose dolphin blubber, but not in bull shark muscle samples. In the human adipose tissue samples, the concentrations of HBCDs were 3-4 orders of magnitude lower than the concentrations of polybrominated diphenyl ethers (PBDEs) previously reported for the same set of tissue samples. Concentrations of HBCDs in human samples from the United States were 1-5-fold lower than the concentrations reported from several European countries. HBCD concentrations in bottlenose dolphins from the United States were 1-2 orders of magnitude lower than the concentrations reported for other cetacean species from Europe. The present report is the first to determine levels of TBBPA and HBCDs in humans, bottlenose dolphins, and sharks from the United States. © 2007 Elsevier Ltd. All rights reserved.

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1. Introduction

Brominated flame retardants (BFRs) are incorporated as additives or by chemical reactions with polymers, for the purpose of reducing the flammability of the final manufactured product. Polymers that contain BFRs, including polyurethane foams, high-impact polystyrene, acrilonitrile-butadiene-styrene, and polyvinyl chloride, are extensively used in products such as building materials, electronic goods, carpets, upholstery textile, and car panels. The most widely used BFRs (Fig. 1; BSEF, 2005) in the USA and Canada are polybrominated diphenyl ethers (PBDEs), followed by tetrabromobisphenol A (TBBPA), and then by hexabromocyclododecanes (HBCDs). The market demand for BFRs in the USA and Canada was estimated to be 54000 metric tons per year (BSEF, 2005). PBDEs and HBCD occur at measurable levels in human and wildlife tissues due to their persistent and lipophilic

^{*} Corresponding author. Tel.: +518 474 0015; fax: +518 473 2895.

E-mail address: kkannan@wadsworth.org (K. Kannan).

¹ Present address: Environmental and Computational Chemistry Group, Department of Chemistry, University of Cartagena, Cartagena, Colombia.

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Fig. 1. Chemical structures of α -, β -, and γ -HBCD isomers and TBBPA.

properties (Loganathan et al., 1995; de Wit, 2002; Hites, 2004; Hoh and Hites, 2005; Law et al., 2005; Covaci et al., 2006).

There are eight HBCD diastereoisomers found in the technical HBCD mixture, comprising three pairs of racemic enantiomers of $(\pm) \alpha$ -(11.8%), β -(5.8%), and γ -(81.6%) isomers and two meso forms of δ -(0.5%) and ε -(0.3%) isomers (Heeb et al., 2005). When the HBCD technical mixture is heated at temperatures of between 160 and 200 °C, α-HBCD prevails due to thermal stereoisomerization (Peled et al., 1995). Liquid chromatography-tandem mass spectrometry (LC-MS/MS) is routinely used in the analysis of HBCD stereoisomers (Budakowski and Tomy, 2003). The occurrence of HBCDs has been reported in environmental and biological samples collected from several European countries (Remberger et al., 2004; Zegers et al., 2005; Covaci et al., 2006; Law et al., 2006). However, prior to this study, no information on the occurrence of HBCDs and TBBPA in humans, bottlenose dolphins, and sharks collected from the United States was available. In this study, we report the concentrations of HBCDs and TBBPA in human adipose tissue obtained in New York City, and in three top-level marine predators, namely, bull shark, Atlantic sharpnose shark, and bottlenose dolphin, all collected from Florida coastal waters between 1991 and 2004. Concentrations of HBCDs and TBBPA in humans, dolphins, and sharks were compared with PBDE concentrations reported earlier for the same set of samples (Johnson-Restrepo et al., 2005a, 2005b). Relationships among TBBPA, HBCDs, and PBDE concentrations in the human and marine predator samples were investigated. Also, the influence of biological/demographic factors (age, gender, and ethnicity of donor) on the concentrations of TBBPA and HBCDs in the human adipose tissues was examined.

2. Materials and methods

2.1. Sampling

Human adipose tissue samples (n = 20) were obtained in New York City during 2003–2004, from patients who underwent liposuction surgery. The samples were devoid of personal identifiers; the only demographic information available to this study was age, gender, ethnicity, occupation, and date of collection. Samples were stored in precleaned glass bottles at -20 °C until analysis. Institutional Review Board (IRB) approval for the analysis of human adipose samples was obtained from the New York State Department of Health. Further details on the human samples have been provided previously (Johnson-Restrepo et al., 2005a).

Blubber from bottlenose dolphin (*Tursiops truncatus*; n = 15) and muscle tissues from bull shark (*Carcharhinus leucas*; n = 13) and Atlantic sharpnose shark (*Rhizoprionodon terraenovae*; n = 3) were analyzed in this study. Bottlenose dolphins were collected from the coastal waters of Florida during 1991–2004 by Hubbs-Sea World Research Institute, and the Southeastern Marine Mammal Stranding Network. Details regarding gender, length, and weight of the dolphins and sharks analyzed in this study have been reported in a previous publication (Johnson-Restrepo et al., 2005b). Shark samples were collected from estuarine waters of the Indian River Lagoon and adjacent offshore coastal waters of Florida in 2004.

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