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TRENDS AND PATTERNS OF POLYBROMINATED DIPHENYL ETHERS IN THICK-BILLED MURRE EGGS FROM THE CANADIAN ARCTIC

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Introduction

Polybrominated diphenyl ethers (PBDEs) are brominated flame retardants (BFRs) which have been used in a wide variety of consumer products including textiles, plastics and foams.¹ The three major PBDE technical products used were penta-BDE (primarily BDE-47, -99 and -100), octa-BDE (primarily BDE-183 and -153) and deca-BDE (almost entirely BDE-209).² The tetra- to hexa-BDEs are persistent, bioaccumulative and are found ubiquitously in Arctic biota.^{1,3} PBDEs, particularly the penta-BDE product, were heavily used in North America¹ and, as a result, PBDE concentrations in North American marine biota are the highest in the world.⁴ Braune et al.⁵ examined temporal trends and congener patterns of PBDEs in seabird eggs sampled from a colony in the Canadian high Arctic during 1975-2014. Here we compare data for PBDEs in eggs of thick-billed murres (*Uria lomvia*) sampled from two colonies in the eastern Canadian low Arctic with those from the high Arctic colony.

Materials and methods

Thick-billed murre eggs were sampled from two colonies situated within 200 km of each other in northern Hudson Bay, Canada: Coats Island (62°98'N, 82°00'W) from 1993 to 2015, and Digges Island (62°35'N, 77°50'W) from 2012 to 2015. Fifteen eggs were sampled per colony per year and egg homogenates were analyzed as pooled (composite) samples consisting of three eggs each. Egg homogenates were analyzed for 12 BDE congeners (BDE-17, -28, -47, -49, -66, -85, -99, -100, -138, -153, -183 and -209) by GC-MSD (ECNI) as described elsewhere.⁵ All reported residue levels were corrected for internal standard recoveries and method blanks. Since BDE-209 was less than the method detection limit in all samples, Σ PBDE was standardized to the sum of BDE-17, -28, -49, -47, -66, -100, -99, -85, -153, -138 and -183. Eggs were also analyzed for stable isotopes of nitrogen (¹⁵N/¹⁴N or δ^{15} N) following procedures described elsewhere.⁵

PBDE concentrations were lipid-normalized to facilitate comparisons among colonies. Statistical tests were performed using a significance level of $p < 0.05$. To account for potential influence of change in trophic position over time, the decreasing temporal trend from 2005 to 2015 at Coats Island was analyzed by backward stepwise regression with year and δ^{15} N as regressors. δ^{15} N was eliminated from the model for both BDE-47 and Σ PBDE. Concentrations of BDE-47 and Σ PBDE were compared between Coats and Digges Islands in 2013, 2014 and 2015 using t-tests. Results were compared with data for eggs of thick-billed murres collected from Prince Leopold Island (74°02'N, 90°05'W) in Lancaster Sound, Canada, from 1975 to 2014 reported in Braune et al.⁵

Results and discussion

The predominant BDE congeners found in eggs of thick-billed murres sampled from Coats and Digges Islands over the years sampled were BDE-47, -99 and -100 which together comprised an average of 78% and 90% of Σ PBDE, respectively, for the two colonies (Figure 1). BDE-138 was also relatively prominent in the murre eggs from Coats Island during 1993-2008 and in 2013 (Figure 1). The predominant BDE congeners over most years in thick-billed murre eggs from Prince Leopold Island were also BDE-47, -99 and -100 which together comprised over 80% of Σ PBDE over 1975 to 2014,⁵ although BDE-138 also contributed high proportions during 2005-2008 and in 2013 (Figure 1; Braune et al.⁵). BDE-47, -99 and -100 are major BDE congeners found in the commercial penta-BDE product² for which demand in North America was historically very high.¹

Braune et al.⁵ showed that concentrations of Σ PBDE increased significantly in eggs of thick-billed murre from Prince Leopold Island from 1975 to 2003 followed by a significant and rapid decline after 2003 (Figure 2). Concentrations of BDE-47 also increased significantly from 1975 to 2005 followed by a rapid decline from 2005 to 2007 (Figure 2). As the dataset from Coats Island covered a shorter time period, the change from increasing to decreasing trend was not as clearly delineated. However, it is clear that rapid and significant declines in concentrations of Σ PBDE ($p < 0.001$) and BDE-47 ($p < 0.001$) occurred after 2005 in murre eggs from Coats Island with levels plateauing in more recent years, which reflects a trend similar to that seen in the murre eggs from Prince Leopold Island (Figure 2). No significant differences were found for concentrations of Σ PBDE and BDE-47 in murre eggs between Coats and Digges Islands in 2013, 2014 or 2015.

Similar trends as seen in the murre eggs from Canadian Arctic colonies have been reported for other North American birds.^{6,7,8} These trends are consistent with the removal of the penta- and octa-PBDE products from the North American market in the mid-2000s. Use of the penta- and octa-BDE technical products was voluntarily discontinued in the United States in 2005¹ before being added to the Stockholm Convention on Persistent Organic Pollutants in 2009.⁹ However, despite these actions to reduce emissions of PBDEs, it is predicted that these compounds will continue to be released from consumer products to which they have been added, and then from disposed waste, for decades to come.^{1,4,10}

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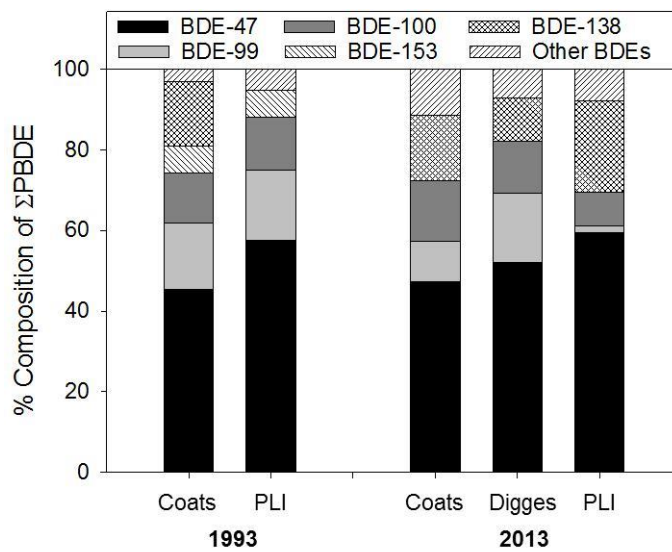


Figure 1. Mean percent contributions of BDE congeners to Σ PBDE in eggs of thick-billed murres from Coats Island, Digges Island and Prince Leopold Island (PLI) in 1993 and 2013. Data for Prince Leopold Island from Braune et al.⁵

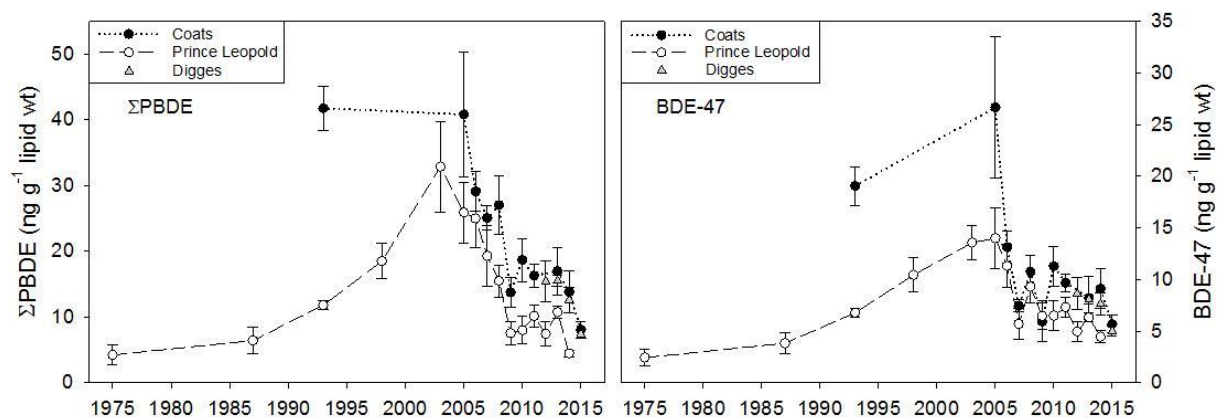


Figure 2. Mean annual concentrations of Σ PBDE and BDE-47 (lipid weight \pm standard error) in eggs of thick-billed murres from Coats Island (1993-2015), Prince Leopold Island (1975-2014) and Digges Island (2012-2015). Σ PBDE = sum of BDE-17, -28, -49, -47, -66, -100, -99, -85, -153, -138 and -183. Data for Prince Leopold Island from Braune et al.⁵