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# Tracking the sources of polybrominated diphenyl ethers in birds: Foraging in waste management facilities results in higher DecaBDE exposure in males

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ABSTRACT

Differences in feeding ecology are now recognized as major determinants of inter-individual variations in contaminant profiles of free-ranging animals, but exceedingly little attention has been devoted to the role of habitat use. Marked inter-individual variations and high levels of polybrominated diphenyl ethers (PBDEs) (e.g., DecaBDE) have previously been documented in ring-billed gulls (*Larus delawarensis*) breeding in a colony near Montreal (QC, Canada). However, the environmental sources of these compounds, and thus the reasons causing these large inter-individual variations remain unidentified. In the present study, we used GPS-based telemetry ($\pm$5 to 10 m precision) to track ring-billed gulls from this colony to reconstruct their movements at the landscape level. We related habitat use of individual gulls ($n=76$) to plasma concentrations (ng/g ww) and relative contributions (percentages) to $\Sigma_{38}$PBDEs of major congeners in the internationally restricted PentaBDE and current-use DecaBDE mixtures. Male gulls that visited waste management facilities (WMFs; i.e., landfills, wastewater treatment plants and related facilities; 25% of all GPS-tracked males) exhibited greater DecaBDE (concentrations and percentages) and lower PentaBDE (percentages) relative to those that did not. In contrast, no such relationships were found in females. Moreover, in males, DecaBDE (concentrations and percentages) increased with percentages of time spent in WMFs (i.e., ~5% of total foraging time), while PentaBDE (percentages) decreased. No relationships between percentages of time spent in other habitats (i.e., urban areas, agriculture fields, and St. Lawrence River) were found in either sex. These findings suggest that animals breeding in the vicinity of WMFs as well as mobile species that only use these sites for short stopovers to forage, could be at risk of enhanced DecaBDE exposure.



## 1. Introduction

Despite steady improvements in the understanding of mechanisms modulating contaminant exposure and transfer dynamics in food webs, explaining variations in body burden of free-ranging animals remains a challenge. Numerous physiological factors may influence contaminant burden such as gender, age, biotransformation capacity, and reproductive status (Verreault et al., 2010). Variations in foraging strategies may also modify exposure pathways and affect contaminant uptake (Burgess et al., 2013; Eulaers et al., 2014; Hebert and Weseloh, 2006; Morrissey et al., 2010). Consequently, a poor understanding of the feeding ecology of bioindicator species may lead to critical misinterpretations of contaminant spatiotemporal trends and exposure-related effects. These knowledge gaps may ultimately compromise our ability to assess ecosystem health (Ramos et al., 2011).

In the last decade, increasing attention has been devoted to the role of foraging ecology in studies investigating contaminant dynamics in organisms and food webs (McKinney, 2012; Ramos et al., 2013; Schipper et al., 2012). However, contaminant uptake does not solely depend on dietary exposure. Anthropogenic chemicals

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can also be absorbed through inhalation, dermal contact, and ingestion of contaminated soil and dust (Mineau, 2011; Sample et al., 2013). Moreover, for compounds that are unevenly distributed in the environment (air, soil, water, and sediments), foraging location may be on some instances as important as diet composition. Disregarding the origin of food/prey items (i.e., foraging location) can thus be misguiding, as it does not capture the full realm of exposure pathways. This is particularly important for species that occupy large home ranges encompassing heterogeneous landscapes, and for generalist foragers whose patterns of habitat use may vary widely in space and time (Wickwire et al., 2011). However, characterizing habitat use represents a technical challenge, especially for highly mobile species such as birds. Consequently, birds have traditionally been categorized according to the geographical position of their nest or colony in order to compare individual contaminant burden and health-related endpoints across a gradient of exposure within a given area. A major shortcoming of this approach is the assumption that all individuals from a nesting site or breeding colony share similar patterns of habitat use. However, recent studies have shown that individuals within populations tend to selectively exploit certain resources within specific habitat components (Araújo et al., 2010; Wakefield et al., 2009; Woo et al., 2008). These inter-individual specializations can result in different patterns and regimes of exposure to certain contaminant types, occasionally leading to large inter-individual variations in body burden. Rapid advances in the miniaturization of animal tracking devices now allow the reconstruction of individual movements at a very fine scale ( $<10$ m precision). Consequently, a detailed characterization of habitat use is now feasible even for species with small body sizes such as birds. Few studies included assessments of bird movements in their investigation of contaminant exposure (Ackerman et al., 2007; Ito et al., 2013; Kendall et al., 2012; Yates et al., 2010). Yet, none of these studies characterized habitat use at the landscape level in omnivorous species that forage in mosaics of anthropogenic/natural and aquatic/terrestrial habitats, encompassing complex exposure profiles to contaminants.

Polybrominated diphenyl ethers (PBDEs) are a class of widely used halogenated flame retardants (HFRs) that delay the ignition of consumer products, including electronic or electric devices, upholstered furniture, and construction materials (Covaci et al., 2011). They bioaccumulate in humans as well as in aquatic and terrestrial biota worldwide (Law et al., 2014). Among the three main PBDE commercial formulations, only DecaBDE remains on the international market, while PentaBDE and OctaBDE were banned internationally in 2009 under the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2010). Nonetheless, recent research established that BDE-209, the main congener (i.e., $>97\%$) in DecaBDE mixture can bioaccumulate in humans (Hardy et al., 2009), terrestrial birds and mammals (Barón et al., 2014; Voorspoels et al., 2007), as well as in certain marine species (Shanmuganathan et al., 2011; Wan et al., 2013). Despite growing concern over its endocrine disrupting potential (Li et al., 2014; Noyes et al., 2013) and suspected developmental toxicity (Costa and Giordano, 2011), DecaBDE is still the most widely used halogenated flame retardant in Asian countries, where the bulk of consumer goods intended to international export are produced (Ni et al., 2013).

Recent studies reported unexpectedly high concentrations and relative contributions (to $\sum$PBDEs) of BDE-209 in ring-billed gulls (*Larus delawarensis*) breeding in a colony located in the St. Lawrence River near the city of Montreal (QC, Canada). These findings suggest that this region might be a local hotspot of DecaBDE exposure for those birds (Chen et al., 2012; Gentes et al., 2012). Ring-billed gulls are generalist foragers that use a wide array of habitat types including urban areas, landfills, waste transhipment sites, wastewater treatment plant ponds, agricultural fields, water bodies, and riparian zones (Patenaude-Monette et al., 2014), where they may be exposed to different concentrations and profiles of PBDEs. This gull population thus provides a unique opportunity to investigate the relationship between habitat use and exposure to PBDEs in a heterogeneous landscape.

In the present study, we used high-resolution (GPS-based) telemetry to reconstruct the movements of individual gulls and related their plasma PBDE concentrations and profiles to their habitat use patterns. More specifically, we investigated two hypotheses related to the effects of foraging in facilities associated with management of liquid and solid waste (i.e., wastewater treatment plants, wastewater lagoons, landfills, and waste transhipment centers). First, we tested whether plasma DecaBDE varied between birds that visited waste management facilities (WMFs) and those that did not, and second, whether plasma DecaBDE increased linearly with percentages of time spent by gulls on WMFs. We focused on WMFs because high levels of DecaBDE have been documented in wastewater effluents (Kim et al., 2013) and sewage sludge (Gorga et al., 2013), as well as in landfill air (St-Amand et al., 2008) and leachates (Li et al., 2012). We also investigated whether DecaBDE increased with usage of urban areas (e.g., cities, towns, villages, and residential agglomerations), because DecaBDE emissions from consumer goods are highest in urban air (Besis and Samara, 2012). To highlight the contrasts between current-use DecaBDE and internationally restricted PentaBDE formulations, we also present comparative relationships between habitat use and plasma PentaBDE.

## 2. Materials and methods

### 2.1. Study area

Fieldwork was conducted from April through June 2010, 2011, and 2012 on Deslauriers Island (45.717°N, 73.433°W). This 600-m long island is located in the St. Lawrence River, 3 km downstream from Montreal (QC, Canada). This ring-billed gull colony is among the largest in Canada and supported approximately 44,000 breeding pairs annually during the course of the study (P. Brousseau, Environment Canada, pers. comm.). This colony is surrounded by a mosaic of high and low density urban areas, agricultural fields, as well as riparian habitats along the St. Lawrence River and its tributaries (Fig. S1). WMFs in our area included four landfills, two waste material transhipment stations, 14 wastewater treatment plants, and one wastewater disposal site for recreational vehicles. All these are located within flying distance ( $<63$ km) from the Deslauriers Island colony (Patenaude-Monette et al., 2014).

### 2.2. Field sampling

Birds ($n=153$) were captured on their nest while incubating using a dip net or a nest trap triggered with a remote control system. Each bird was banded with a US Fish and Wildlife Service stainless steel ring and a color-coded plastic band. Birds were equipped with a miniature GPS data logger (GiPSy2, TechnoSmArt, Rome, Italy) affixed on the two central rectrices using waterproof tape (TESA, Charlotte, NC, USA). The data loggers (12–16 g) represented less than 3% of ring-billed gull body mass (mean $\pm$ SEM: $481 \pm 4$ g; $n=153$). Birds were recaptured 2–12 days later to recover the GPS units. A blood sample (4–10 mL; $n=127$) was collected from the brachial vein immediately after recapture, and birds were euthanized by cervical dislocation as part of companion studies (Aponte et al., 2014; Caron-Beaudoin et al., 2013; Plourde et al., 2013). Sex was determined through gonad examination.