Interspecies comparisons of brominated flame retardants in relation to foraging ecology and behaviour of gulls frequenting a UK landfill

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**Keywords:** Ecotoxicology, Flame Retardants, Persistent Organic Pollutants, Birds, Landfill, Gulls

**Abstract**

This study quantifies and compares concentrations and profiles of legacy and alternative (alt-) brominated flame retardants (BFRs) in the eggs of three gull (Laridae) species of international/UK conservation concern – great black-backed gulls (*Larus marinus; n* = 7), European herring gulls (*L. argentatus; n* = 16) and lesser black-backed gulls (*L. fuscus; n* = 11) in relation to their foraging ecology and behaviour in order to investigate potential exposure pathways at a remote landfill in western Scotland, UK. Egg concentrations of sum (∑) polybrominated diphenyl ethers (∑8PBDEs) in all three species exceeded those for most reported avian species using landfill, except for those in North America. Despite relatively high detection frequencies of ∑hexabromocyclododecanes (∑3HBCDDs) (94–100%), concentrations of ∑8PBDEs exceeded ∑3HBCDDs and ∑5alt-BFRs, with ∑8PBDE levels similar in all three species. Egg carbon isotopic (δ13C) values highlighted a greater marine dietary input in great black-backed gulls that was consistent with their higher BDE-47 levels; otherwise, dietary tracers were minimally correlated with measured BFRs. ∑3HBCDD egg concentrations of herring gulls markedly exceeded those reported elsewhere in Europe. Decabromodiphenylethane (DBDPE) was the only alt-BFR detected (6–14% detection rate), in a single egg of each species. The great black-backed gull egg contained the highest concentration of DBDPE measured in biota to date globally and provides strong evidence for its emerging environmental presence as a BDE-209 replacement in UK wildlife. Correlations between δ13C (dietary source) and some measured BFRs in eggs suggest multiple routes of BFR exposure for gulls frequenting landfill through their diet, behaviour, preening, dermal exposure and likely inhalation. The frequent use of landfill by herring gulls and their increased egg FR burdens suggest that this species may be an important bioindicator of BFR emissions from such sites.

**Keywords**

Birds, PBDEs, HBCDD, DBDPE, Stable isotopes, Behaviour

**1. Introduction**

Brominated flame retardants (BFRs) are synthetic organohalogens that have been applied to many manufactured polymeric products to impart fire retardancy (De Wit, 2002; Jenssen et al., 2007). Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD) were widely used BFRs historically but are now strictly regulated given their toxicity, persistence and capacity for bioaccumulation, biomagnification, and long-range atmospheric transportation. As a result, both of these ‘legacy’ BFRs are classed as persistent organic pollutants (POPs) under the Stockholm Convention (Stockholm Convention, 2019). In addition, and often in response to the regulation of legacy BFRs, manufacturers have developed alternative BFRs (alt-BFRs), which have been subject to recent research into their environmental presence and effects on biota (e.g., Marteinson and Fernie, 2019; Marteinson et al., 2017; Verreault et al., 2018).

 Given that they are repositories of obsolete consumer products, waste management facilities such as municipal solid waste landfill/dump sites (hereafter ‘landfill’) often contain elevated concentrations of PBDEs, HBCDD, and alt-BFRs in abiotic media such as air, soil, and leachate (Eguchi et al., 2013; Harrad et al., 2019a; Morin et al., 2017) as a result of weathering and abrasion of polymeric items (Stubbings and Harrad, 2014). Consequently, landfills are well documented as major sources of these contaminants. Because of the large quantities of human food refuse that can also enter landfill, various bird species routinely forage at such sites (Oro et al., 2013; Plaza and Lambertucci, 2017). Increasingly, it has been recognised that birds associated with landfill exhibit elevated legacy BFR and alt-BFR burdens (Tongue et al., 2019), which is of concern since legacy and some alt-BFRs have been demonstrated to affect birds adversely through studies of reproductive success, behaviour and growth of free-living and captive species (Guigueno and Fernie, 2017). Although attention is now being paid to landfill as a source of organohalogen exposure in wild birds (Chen et al., 2013; Tongue et al., 2019), ours is the first study to examine such exposure in the UK and among different bird species using landfill.

 The exposure of free-living birds to flame retardants (FRs) and other contaminants can occur through multiple routes, including their diet, feather preening, dermal exposure and inhalation. In studies of the avian diet, nitrogen (δ15N) stable isotope values have been examined to characterise trophic position (Bearhop et al., 2002; Hobson et al., 1994). By comparison, carbon (δ13C) tracers determine relative contributions of marine and terrestrial food sources (Hobson, 1987; Inger and Bearhop, 2008), while sulphur (δ34S) values allow more precise determination of these nutrient sources in the same marine *vs*. terrestrial environments (Eulaers et al., 2014; Hobson et al., 1997). Chen et al. (2012) and Roscales et al. (2016) successfully used these dietary tracers in studying avian trophodynamics of environmental contaminants. Direct ingestion of FR-treated items and their abraded particles may be an important route of exposure and uptake by birds (Seif, 2017) that is rarely explored (Brusseau et al., 2019), including in birds utilising landfill (Tongue et al., 2019). Avian behaviours can influence exposure to FRs, including feather preening, when preen oil traps contaminant particulates on the plumage which is inadvertently ingested during feather maintenance (Jaspers et al., 2007). Exposure may also result from inhalation of airborne contaminants by birds because of their specialised respiratory system (Brown et al., 1997). Finally, dermal contact may also be a major exposure route (Mineau, 2011), particularly in gulls that forage on landfill substrate, thereby potentially exposing the skin of their webbed feet, legs, and facial bare parts to FRs.

 In north-western Europe, several gull species are associated with landfill, including great black-backed gulls (*Larus marinus*), European herring gulls (*L. argentatus*) (hereafter ‘herring gulls’) and lesser black-backed gulls (*L. fuscus*) (Coulson, 2019; Greig et al., 1986; Horton et al., 1983) (Table S1). If landfill is a considerable source of exposure for such gulls to accumulate FRs (and other environmental pollutants), this may potentially be of conservation significance. Herring gulls are designated as ‘Near Threatened’ in the EU (BirdLife International, 2015), while great black-backed gulls and lesser black-backed gulls are of conservation concern in the UK (Eaton et al., 2015). The exposure and accumulation of BFRs by these species while frequenting landfill have previously not been considered in a conservation context, nor has any study to date examined BFR profiles and concentrations across an avian species assemblage associated with landfill. Here, we address the following objectives: (i) to assess and compare the concentrations and profiles of PBDEs, HBCDD, and five alt-BFRs accumulated in eggs laid by great black-backed gulls, herring gulls, and lesser black-backed gulls breeding adjacent to a landfill frequented by gulls; (ii) to investigate BFR trophodynamics using stable isotope analyses of δ13C, δ15N and δ34S in egg contents; and (iii) to identify and characterise possible (behavioural) routes of BFR contamination in birds using landfill.

**2. Materials and methods**

This study was conducted with the necessary authorisation from Scottish Natural Heritage (SNH; licence numbers 77830, 92331 & 112381) for egg collection; birds were not directly handled and so no other licences or permits were required.

*2.1.* *Study site*

 The study was conducted at an active landfill (behavioural observations) with eggs collected from a nearby (~2 km distant) mixed breeding colony of herring gulls, great black-backed gulls and lesser black-backed gulls, located in western Scotland, UK (Fig. 1). During 2016–2018, the landfill received on average 16,000 metric tonnes of household and commercial waste annually from a human population of ~20,000 (Scottish Environmental Protection Agency information request response).

*2.2.* *Egg sampling*

Locally breeding gulls are known to begin egg laying in late April (Kim et al., 2010) when maternal deposition of BFR burdens into eggs occurs, since gulls rely on exogenous nutrients gathered immediately prior to egg formation (Drent and Daan, 1980). Table S1 details the number of eggs of each species collected during our study and provides general information about diet and local movements of each species. Eggs were collected between April and May 2016 from a mixed colony of gulls containing approximately 250 pairs of herring gulls, 30 pairs of lesser black-backed gulls, and three pairs of great black-backed gulls. Due to the comparatively low number of great black-backed gull eggs collected in 2016 (*n* = 4), an additional three eggs of this species were collected from the same colony in 2017 (*n* = 2) and 2018 (*n* = 1) and analysed. From randomly-selected individual nests containing full clutches of three eggs, the estimated largest egg was collected between ~1–6 d following incubation onset as assessed via flotation testing (OSPAR, 2000) and individually identified. Egg length and breadth were recorded to the nearest 0.1 mm using digital Vernier calipers (MachineMart, Nottingham, UK), allowing the calculation of volume (Narushin, 2005). Eggs were weighed (to the nearest 0.1 g) (On Balance, Liverpool, UK), wrapped in aluminium foil, placed into individually-labelled Whirlpak™ sample bags (Nasco, Fort Atkinson, WI, USA), and stored securely in foam-lined Peli Storm iM2300™ cases (Pelican Products, Torrance, CA, USA) until transportation to the University of Birmingham, UK for laboratory analysis. Egg contents were collected, homogenised and frozen at -70°C until chemical analysis.

*2.3.* *Sample extraction and clean-up*

Egg samples underwent a combined pressurised liquid extraction (PLE) and clean-up process for all target BFRs, which comprised the following PBDE congeners: BDEs -28, -47, -99, -100, 153, -154, -183 and -209, α-, β- and γ- HBCDD diastereomers, and five alt-BFRs: (1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenylethane (DBDPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), pentabromobenzene (PBB), and pentabromoethylbenzene (PBEB). Aliquots (~1 g wet weight) of homogenised egg sample were accurately weighed and loaded into a pre-cleaned 66 mL Dionium™ extraction cell pre-packed from the bottom upwards with: two glass fibre filters (GFFs), 3 g of pre-cleaned hydromatrix, 2 g of 1% deactivated silica, 1 GFF, 10 g of 44% acid impregnated silica, and 4 g of Florisil. Cells were then spiked with known quantities of internal standards (BDE-77, BDE-128, 13C12-BDE-209, 13C12- α-, β- and γ-HBCDD, and 13C6-BTBPE). Cells underwent PLE on a Dionex Accelerated Solvent Extractor (ASE) 350 at a pressure of 1,500 psi with hexane/dichloromethane ((DCM); 3:1, v/v ratio) as the extraction solvents. The oven temperature was 90°C, with a heating time of 5 min. Each sample underwent three static cycles with a static time of 4 min, purge time of 90 sec and flush volume of 40%. Clean extracts were transferred to 200 mL tubes and concentrated to near-dryness at 40℃ under a gentle stream of nitrogen. The sample was reconstituted in 50 µL of toluene containing 10 ng each of PCB-129 and d18-γ-HBCDD as recovery determination standard (RDS), sonicated for 10 sec and transferred to a labelled glass-inserted vial prior to analysis.

*2.4.* *Instrumental analysis*

HBCDD levels in eggs were determined on a Shimadzu LC-20AB liquid chromatograph (LC) (Shimadzu Corporation, Kyoto, Japan), coupled to an AB Sciex API 2000 triple quadropole mass spectrometer (MS/MS) (Applied Biosystems, Foster City, CA, USA). Full details of the LC-MS/MS methodology have been published previously (Abdallah et al., 2008). Concentrations of PBDEs and alt-BFRs were determined on a Thermo Scientific Trace 1310 gas chromatograph (GC) coupled to a Thermo Scientific ISQ MS. Full GC/MS parameters are provided in Abdallah et al. (2017).

*2.5.* *Lipid analysis of egg samples*

The lipid content of egg samples was determined gravimetrically. One gramm of sample was weighed into an ASE cell containing clean hydromatrix (*W1*). The cell was then extracted following the same conditions as for BFR extraction (see Section 2.3). An empty Turbovap™ tube was weighed (*W2*) and the entire extract was transferred to the tube. The extract was concentrated to dryness at 40°C under a gentle stream of nitrogen and the Turbovap™ tube was re-weighed (*W3*). The lipid content was determined using the equation:

$$Lipid content \left(\%\right)= \frac{(W3-W2)}{W1} ×100$$

*2.6.* *Quality Assurance/Quality Control*

A reagent blank was analysed with every batch of nine samples. In the majority of sample batches, none of the target compounds was measured above the limit of detection (LOD). In these cases, the samples were assigned limits of quantification (LOQs) based on a signal to noise ratio of 10:1. However, in three batches of samples, BDE-209 was detected in the blank above the LOD (0.95, 0.90 and 0.95 ng/g). In these instances, the LOQ was reported as the average blank plus three times its standard deviation (i.e., 1.0 ng/g). For the three sample batches where BDE-209 was detected in reagent blank samples, the concentration of BDE-209 in each sample was corrected by subtracting from the concentration in the sample the average blank concentration plus three times its standard deviation. If the blank concentration was > 50% of the sample concentration (which occurred in the case of four samples), the sample was reported as < 1.0 ng/g.

 In the absence of an appropriate certified reference material, the full analytical method was validated by replicate analysis (*n* = 10) of domestic chicken (*Gallus gallus*) egg spiked with known concentrations of target compounds. Concentrations of target analytes were then measured according to the above analytical protocols. All measured concentrations were 80–120% of their spiked concentration, with a relative standard deviation of < 15%. For ongoing accuracy and precision, a control sample (*n* = 9) spiked with target compounds was analysed every twentieth sample and was required to be within 80–120% of the spiked concentration for the sample batch to be accepted. Full details of the method validation and ongoing accuracy and precision are provided in the Supplementary Information (SI) (Tables S2 and S3).

*2.7.* *Stable isotope analysis* *(SIA)*

Analysis of homogenised egg (i.e., yolk and albumen) contents for δ13C, δ15N and δ34S (using lipid extracted aliquots in the case of δ13C ) was carried out at the Scottish Universities Environmental Research Centre (SUERC) at East Kilbride, UK, using standardised methods (Bell et al., 2017; Webb et al., 2017) for a subset of analysed eggs laid by great black-backed gulls (*n* = 5; collected 2016–2018), herring gulls (*n* = 14; 2016) and lesser black-backed gulls (*n* = 7; 2016). Prior to SIA, aliquots of egg (~2 mL) were freeze-dried for 24 hours using a Christ Beta 1–8 LSCplus freeze-dryer (Martin Christ, Osterode am Harz, Germany). Further details of SIA methodology are provided in the SI.

*2.8. Behavioural observations*

Behavioural observations (undertaken only for gulls in adult plumage) coincided with the likely period of yolk formation in large gulls (Roudybush, 1979) and further details are provided in the SI. Briefly, data were obtained in 2018 by video recording of gull foraging behaviour on the active tipping area of the landfill and observing the incidence of preening at a separate area within the landfill where gulls gathered to loaf (i.e., rest). Data were collected during landfill operational hours (i.e., Monday to Friday; 09.00–16.00hrs GMT; 9–20 April 2018). In addition, counts and observations of loafing birds were made at 30-minute intervals and the following recorded: total numbers of birds of each species resting and the incidence and duration of preening performed by individual adult (i.e., potentially breeding-age) gulls. Given resource and time constraints, it was not possible to collect eggs from individuals clearly identified and known to frequent the landfill, although the constant movement of birds between the landfill and the colony suggested that the landfill was an important and regularly-used site for these birds. Data were extracted from video footage following (Greig et al., 1985, 1986). For foraging observations, an observation period of 15 sec per bird was defined, during which the number of (i) pecks made into the substrate, (ii) swallowing events, and (iii) paces across the substrate (scored from 0 to 3: 0 = 0 paces, 1 = 1–4 paces, 2 = 5–10 paces, and 3 = > 10 paces) were recorded. The mean number of paces was estimated and scored from 1 to 3: 1 = 2.5 paces, 2 = 7.5 paces, 3 = 15 paces (approx.). To provide further insight into potential dermal exposure we also estimated and ranked the mean length of time each bird stood stationary while foraging (0 = 0 sec, 1 = 1–5 sec, 2 = 6–10 sec, and 3 = 11–15 sec) or not foraging (1 = 2.5 sec, 2 = 7.5 sec, and 3 = 12.5 sec) within the 15 sec observational period. These behaviours were identified as some of the most likely behaviours that would result in birds being exposed to FRs (Table S5). We analysed both the number of paces made and time spent stationary because either behaviour may potentially expose birds to FRs via dermal contact (Alharbi et al., 2016; Henderson et al., 1994; Mineau, 2011).Review of video footage yielded 2,329 observations of foraging birds, consisting of 255 observations of great black-backed gulls, 1,961 of herring gulls and 113 of lesser black-backed gulls. Over the course of a given day, gulls regularly loafed (rested) on a plastic and gravel-covered former waste pile on site, located ~400 m from the active tip face. Counts and observations of loafing birds were made at 30-min intervals and recorded on pre-printed field datasheets.

*2.9.* *Data visualisation, manipulation and statistical methods*

For statistical purposes, in samples where the detection frequency (DF) was lower than 50%, “zero” values were replaced with the DF multiplied by the LOQ. For samples where the DF was ≥ 50%, values of zero were replaced with 0.5 × LOQ. Those measured compounds for which detection frequencies were < 30% for all three species were excluded from statistical analyses (Table 1). All statistical analyses were undertaken using R (R Core Team, 2018). Data were checked for normality via visual inspection and Shapiro-Wilk tests. Homogeneity of variance was assessed using the Levene’s test. The BFR data were not normally distributed and could not be successfully transformed to normality. Therefore, the non-parametric Kruskal-Wallis (K-W) test was used to compare egg BFR concentrations and relative contributions between species with subsequent pairwise Mann-Whitney (M-W) post-hoc tests using a Holm correction. Stable isotope data were normally distributed and therefore interspecies comparisons of these data were made using one-way analysis of variance (ANOVA), with post-hoc testing undertaken using the Games Howell test. The ‘Corrplot’ package in R (Wei and Simko, 2017) with Spearman’s correlation coefficient was used to examine relationships between egg BFR concentrations and isotope values. Confidence limits were set to 95% and an alpha threshold of 0.05 was used for statistical comparisons, except for behavioural data when this was adjusted to 0.01 (after Grant and Grant, 2002; Portugal et al., 2010) since statistical independence of behavioural data could not be guaranteed as gulls were not individually identifiable.

**3. Results**

*3.1.* *Profiles and concentrations of PBDEs in gull eggs*

Detection frequencies of each of the eight measured PBDE congeners (i.e., BDEs -28, -47, -99, -100, -153, -154, -183, and -209) varied amongst the three gull species observed on the landfill and were most frequently detected in the eggs of great black-backed gulls (71–100%), followed by lesser black-backed gulls (45–90%) and then herring gulls (62–88%) (Table 1). The PBDEs with the highest detection rates were similar in the eggs of lesser black-backed gulls (BDE -47 ≈ -153 ≈ -209) and herring gulls (BDE-47 ≈ -153 ≈ -154 ≈ -209) in contrast to the eggs of great black-backed gulls (BDE-47 > -99 ≈ -153 ≈ -183) (Table 1).

There were no significant interspecies differences in egg concentrations of ∑8PBDEs (K-W test: *H*= 0.2, df = 2, *P* = 0.91) or of ∑7PBDEs (i.e., excluding BDE-209; K-W test: *H*= 0.01, df = 2, *P* = 0.99) (Table 1). However, there was a significant interspecies difference for egg concentrations of BDE-47 (K-W test: *H* = 7.3, df = 2, *P* = 0.02). Post-hoc (M-W) testing showed BDE-47 concentrations to be significantly higher in great black-backed gulls compared to the other two species (pairwise post-hoc tests: both *P*s ≤ 0.03) that were statistically similar to each other (Fig. 2). There were no other significant interspecies differences for concentrations of the remaining BDE congeners (pairwise post-hoc tests: all *P*s≥ 0.20). In terms of relative contributions to ∑8PBDE burdens, BDE-99 followed by BDE-209 were the most prevalent congeners in the eggs of the great black-backed gulls and lesser black-backed gulls in contrast to BDE-209 and, to a lesser extent, BDE-99 in herring gulls (Fig. 3). In addition, the eggs of great black-backed gulls showed a significantly greater relative mean contribution of BDE-47 to their ∑8PBDE burdens compared to the eggs of herring gulls and lesser black-backed gulls (K-W test: *H*= 9.7, df = 2, *P* = 0.007; pairwise post-hoc M-W tests: *P*s ≤ 0.03) (Fig. 3). The relative contributions to ∑8PBDEs of the other individual congeners were comparable across the three species (K-W tests: all *P*s≥ 0.10).

*3.2. Profiles and concentrations of HBCDD in gull eggs*

We detected α-HBCDD in all great black-backed gull and lesser black-backed gull eggs and in 87% of herring gull eggs (Table 1). There was marked variation among the three species in the detection frequency of γ-HBCDD, which was 100% in great black-backed gull eggs, 87% in herring gull eggs, but only 27% in lesser black-backed gull eggs. There were significant interspecies differences in egg concentrations of ∑3HBCDDs (K-W test: *H*= 6.4, df = 2, *P* = 0.04), with pairwise post-hoc M-W tests showing that great black-backed gull eggs contained significantly higher concentrations than lesser black-backed gulls (*P* = 0.05) (Table 1). This appeared to be driven by the much lower detection rate (27%) of γ-HBCDD in lesser black-backed gull eggs compared to great black-backed gulls (100%). The mean relative contribution of α-HBCDD to ∑3HBCDD significantly varied among the three species (K-W test: *H* = 11.8, df = 2, *P* = 0.002) and was significantly higher in lesser black-backed gulls (90%) than in herring gulls (62%) or great black-backed gulls (49%) (pairwise M-W post-hoc tests: *P*s ≤ 0.01). Correspondingly, the mean relative contribution of γ-HBCDD to ∑3HBCDD varied among all three species (K-W test: *H* = 13.5, df = 2, *P* = 0.001) and was significantly lower in lesser black-backed gulls (10%) compared to both great black-backed gulls (51%) and herring gulls (37%) (pairwise M-W post-hoc tests: *P*s ≤ 0.01).

*3.3.* *Alt-BFR concentrations in Eggs*

DBDPE was detected in only three of the 34 eggs collected, being in one egg of each species. Specifically, the DBDPE concentration in the great black-backed gull egg (7,700 ng/g lw) exceeded significantly that in the egg of the herring gull (57 ng/g lw) and in the egg of the lesser black-backed gull (68 ng/g lw). This represents DBDPE detection frequencies of only 14%, 6%, and 9%, respectively. None of the other targeted alt-BFRs was detected in any of the eggs analysed.

*3.4. Stable isotope analysis*

The δ13C, δ15N and δ34S dietary tracer values measured in eggs laid by the three gull species are shown in Table 1. The only isotopic value that demonstrated significant interspecies differences was δ13C (dietary source) (one-way ANOVA: *F*2,23 = 8.0, *P* = 0.002) (Fig. 4), with no significant differences among the three species in egg δ15N (trophic position: *F*2,23 = 0.5, *P* = 0.60) andδ34S (dietary source: *F*2,23 = 1.6, *P* = 0.20). Post-hoc tests revealed that there were significantly higher δ13C values in eggs of great black-backed gulls (*P* = 0.01) and herring gulls (*P* = 0.02) than those of lesser black-backed gulls, with no significant difference in δ13C egg values between great black-backed gulls and herring gulls (*P* = 0.61) (Table 1; Fig. 4). A significant negative correlation was found between egg δ15N isotopic values and BDE-28 egg concentrations of herring gulls (*r* = -0.32, df =12, *P* = 0.02) only (Fig. 5), but no other significant correlations were found between isotopic values and other measured BFR concentrations in any of the three species.

*3.5.* *Behavioural observations*

Vantage point surveys revealed a regular movement of all three gull species between the breeding colony and the landfill (Fig. 1). At any one time we observed a maximum of ~800 birds at the landfill, with herring gulls comprising ~90% of all birds, while great black-backed gulls and lesser black-backed gulls made up ~7% and ~3% of the total birds observed on the landfill, respectively.

Foraging strategy differed among the three gull species, although data were zero-inflated (i.e., containing a preponderance of zero observations) for many of the behaviours. The frequency of pecks at the substrate differed significantly between species overall (K-W test: *H*= 58.3, df = 2, *P* < 0.001; Fig. 6a). Pairwise post-hoc comparisons showed that there were significant differences between great black-backed gulls and herring gulls and between lesser black-backed gulls and herring gulls (M-W tests: *P*s < 0.001). Herring gulls were observed pecking the landfill substrate more often (i.e., the lowest percentage of zero pecks per bird-observation) (23%) than lesser black-backed gulls (30%) or great black-backed gulls (51%), and while highly zero inflated, the number of observed swallowing events was nearly significantly different among species (K-W test: *H*= 8.3, df = 2, *P* = 0.02). There was also a significant difference overall in the number of paces taken across the landfill substrate (K-W test: *H* = 46.3, df = 2, *P* < 0.001; Fig. 6b), with lesser black-backed gulls pacing (median: 7.5 paces) significantly more than the other two species (medians: 2.5 for each species; pairwise M-W post-hoc test: *P*s< 0.0001). The time spent stationary when foraging on the landfill differed significantly overall (K-W test: *H* = 93.5, df = 2, *P* < 0.0001; Fig. 6c), with great black-backed gulls (median: 12.5 sec) significantly more stationary than herring gulls (median: 7.5 sec) or lesser black-backed gulls (median: 2.5 sec) (pairwise M-W post-hoc tests: *P*s < 0.0001). Episodes of preening by birds on the landfill were rare across all species (15% of 129 observations) and in each species (13–15%), with similar duration of preening for each species (100–116 sec).

**4. Discussion**

In this study we observed interspecies differences in *in ovo* detection rates, profiles and concentrations of BDE-47 and ∑3HBCDDs among three gull species frequenting a Scottish landfill. Of the five alt-BFRs targeted, DBDPE was the only one detected and with one of the three eggs having the highest concentration reported in biota globally to date. The diets of great black-backed gulls and herring gulls had greater marine input (as reflected in egg δ13C isotopic values) compared to the more terrestrially-based diet of the lesser black-backed gulls, although finer marine-terrestrial dietary details (δ34S) and trophic position (δ15N) were similar among the three species. Interestingly, dietary trophic level (δ15N) and BDE-28 were negatively correlated in herring gulls. There were interspecies behavioural differences for gulls foraging on landfill which are potentially relevant as possible FR exposure pathways.

*4.1.* *PBDE and HBCDD concentrations in gull eggs*

 Compared to BFR burdens reported in other birds known to frequent landfill, ∑8PBDE egg concentrations of our great black-backed gulls, herring gulls, and lesser black-backed gulls (i.e., 35, 54 and 61 ng/g ww, respectively; Table 1) exceed those reported for white storks (*Ciconia ciconia*; 4.4 ng/g ww) in Spain (Muñoz-Arnanz et al., 2011), and African sacred ibis (13.2 ng/g ww) in South Africa (Polder et al., 2008). Furthermore, ∑8PBDE egg concentrations of herring gulls and lesser black-backed gulls in our study exceeded ∑10PBDE egg concentrations (also including BDE-209) of yellow-legged gulls (*L. michahellis*) (i.e., 38 ng/g ww) in Spain (Roscales et al., 2016). Such differences are likely influenced by various factors, including the history of FR use in the respective countries concerned, as well as species- and congener-specific toxicokinetics and metabolism (Chen and Hale, 2010). Our egg samples were obtained from a landfill in a relatively remote location in rural Scotland where there were likely to be fewer alternative sources of environmental exposure to FRs (Fig. 1). In contrast, Polder et al. (2008) and Roscales et al. (2016) studied birds laying eggs in predominantly urban/industrial locations. In this context, our findings emphasise the importance of landfill (however remote) as a source of BFR exposure and accumulation by birds frequenting such locations. Moreover, the gull eggs in our study contained ∑PBDE concentrations that exceeded the 29 ng/g ww effects-level threshold set by the Canadian Federal Environmental Quality Guidelines for avian eggs (Environment and Climate Change Canada, n.d.), suggesting that the birds in our study could have been exposed to potentially deleterious effects following PBDE exposure; nevertheless, species’ differences in sensitivity to the toxicity of PBDEs must also be acknowledged.

Some studies have found higher ∑PBDE egg concentrations in birds associated with landfill across Canada, with concentrations of 268 ng/g ww (excluding BDE-209) in eggs of common starlings (*Sturnus vulgaris*) (Eens et al., 2013), and 230 ng/g ww (including BDE-209) in those laid by American herring gulls (*L. smithsonianus*) (Chen et al., 2012). Great black-backed gulls in the Gulf of St Lawrence (Canada) laid eggs containing comparatively elevated ∑5PBDE (BDEs -47, -99, -100, -153, and -154) concentrations (i.e., 1,643 ± 234 ng/g lw) (Lavoie et al., 2010), although this study did not reveal whether that population was associated with landfill. Elevated PBDE egg concentrations in these Canadian gulls likely reflect the greater and longer period of use of PBDEs (particularly lower-brominated commercial mixtures) in North America compared with in Europe (BSEF, 2003). However, *in ovo* BDE-209 concentrations of up to 137 ng/g ww were reported in herring gulls breeding in Canada (Chen et al., 2012), and indeed some of the highest avian liver concentrations of BDE-209 ever recorded were in ring-billed gulls that frequently fed on landfill in Montreal, PQ, Canada (57.2 ± 12.2 ng/g ww; Gentes et al., 2012).In terms of individual PBDE congeners, the comparatively broad foraging niche and elevated trophic position of the great black-backed gulls in the present study likely explain the high concentrations and relative contribution of BDE-47 to the total PBDE burden found in their eggs compared to those of herring gulls and lesser black-backed gulls (Fig. 2). Certainly, the predominance of BDE-47 in the PBDE congener egg profile is commonly associated with birds feeding predominantly in aquatic rather than terrestrial food webs (Chen and Hale, 2010). Similarly, high concentrations of BDE-47 were reported in eggs laid by piscivorous Audouin’s gulls (*Ichthyaetus audouinii*) compared to those laid by yellow-legged gulls that are dietary generalists (Roscales et al., 2016).

 In the present study, the mean ∑3HBCDD egg concentrations of great black-backed gulls and herring gulls (i.e., 9.3 and 30 ng/g ww, respectively) exceeded those of glaucous-winged gulls (*L. glaucescens*) associated with landfill in Canada (i.e., 4.5 ng/g ww). Herring gulls in our study also had higher ∑3HBCDD concentrations compared to herring gulls (i.e., 16.6 ng/g ww) using landfill in Canada (Chen et al., 2012). In comparison with Europe, HBCDD was generally used to a lesser extent in North America (BSEF, 2003; Law et al., 2014). There are relatively few studies of egg HBCDD concentrations for known populations of our target species that use landfill, with which we can compare our findings. However, for illustrative purposes only we found that the ∑3HBCDD concentrations in eggs of lesser black-backed gulls (i.e., 2.2 ng/g ww) were comparable with those reported in eggs of African sacred ibises using landfill in South Africa (1.9 ng/g ww; Polder et al., 2008), although these ∑3HBCDD concentrations were both exceeded by those in our great black-backed gull eggs (Table 1).

 As reported previously in other biota, α-HBCDD is the most frequently found HBCDD diastereomer in avian tissues (Letcher et al., 2015). The substantial relative contribution of γ-HBCDD to ∑3HBCDD burdens in the eggs of herring gulls and especially great black-backed gulls in our study, conflicts with other published findings. For example, in great black-backed gulls and herring gulls breeding in Norway, α-HBCDD comprised 97% and 100% of egg ∑3HBCDD concentrations, respectively (Haukås et al., 2009; Helgason et al., 2009), and > 90% of the egg burdens of herring gulls breeding in Germany (Esslinger et al., 2011). The three gull species frequenting landfill in our study may have been exposed to more recent sources of HBCDD such as waste materials treated with this BFR, with -HBCDD being the dominant diastereomer in the commercial formulation (Harrad et al., 2019b; Law et al., 2005). In addition, the lower relative contribution of γ-HBCDD in the lesser black-backed gull eggs may reflect species- or diastereomer-specific toxicokinetics (Head et al., 2008; de Wit et al., 2019). Dietary differences may also play a role, since lesser black-backed gulls appeared to have a more terrestrial diet compared with the other two species (Table S1; Fig. 4). Moreover, maternal transfer of HBCDD and other measured BFRs in our study may also account for differences in the respective profiles among the three gull species.

*4.3.* *Alt-BFR* *concentrations in gull eggs*

 Of the five alt-BFRs measured in our study, we detected only DBDPE in one egg of each of the three species. This is consistent with other studies that have found its emerging use as a replacement for Deca-BDE (summarised by Betts, 2009). While the detection frequency of DBDPE in gull eggs in our study was low (i.e., 10% for all species combined), the maximum concentration of 7,700 ng/g lw in a single egg laid by a great black-backed gull (Table 1) is the highest recorded in biota worldwide. This is suggestive of an upward trend in DBDPE usage as also highlighted by its elevated environmental concentration in indoor air and dust in the Republic of Ireland (Wemken et al., 2019). The highest concentration of DBDPE previously reported by Gauthier et al. (2007) was 505 ng/g lw in herring gull eggs in the Great Lakes in a study that had a similar detection frequency (i.e., 9%) to ours. The apparent high concentrations of DBDPE measured in some eggs may warrant further research into the possible embryonic toxicity of this alt-FR (see also Egloff et al., 2011; Zheng et al., 2014, 2015). Indeed, the findings of these previous studies and our study, collectively suggest that further research concerning DBDPE is required (de Wit et al., 2019; Guo et al., 2019; Stubbings et al., 2019; Wemken et al., 2019).

 The lack of detection of the other four targeted alt-BFRs in our gull eggs is broadly consistent with the existing literature (reviewed in Tongue et al., 2019). Most alt-BFRs are detected in birds at orders of magnitude less than legacy BFRs, possibly reflecting their limited usage over time and space and/or possibly reduced bioavailability (Chen et al., 2013; Covaci et al., 2011). Nevertheless, compared with other birds known to use landfill, BTBPE and PBEB have been detected in eggs of herring gulls in the Great Lakes at low concentrations of up to 0.7 ng/g ww and 1.4 ng/g ww, respectively (Gauthier et al., 2007), and white storks in urban areas of Spain laid eggs containing PBEB at concentrations of up to 9.79 ng/g ww (detection frequency: 20%) (Munoz-Arnanz et al., 2010). Detection rates of alt-FRs were also low in eggs of tree swallows (*Tachycineta bicolor*) nesting downstream of waste water treatment plants, but concentrations of six alt-FRs including BTBPE were greater than HBCDD or BDE-209 (Fernie and Letcher, 2018). The production and use of both BTBPE and PBEB appear to be greater in North America compared to Europe (Covaci et al., 2011; Harju et al., 2009).

*4.4.* *Trophic ecology of gulls on landfill*

 Lesser black-backed gulls are the most widespread breeding gull species in urban centres in western Scotland (Forrester and Andrews, 2012). Consistent with that, the decreased δ13C egg isotope values of lesser black backed gulls compared with the other two gull species (Fig. 4) indicate that lesser black-backed gulls may have consumed more food from terrestrial than from marine sources, notwithstanding potential confounding effects from the consumption of anthropogenic items *vs*. ‘natural’ terrestrial food which may also have influenced δ13C values (Caron-Beaudoin et al., 2013). In the current study, lesser black-backed gull eggs contained comparatively higher ∑8PBDE concentrations than the other two gull species, and compared to great black-backed gull eggs, they also had a significantly lower relative contribution of BDE-47 to ∑PBDEs, indicating a relatively limited marine component in their diet. BDE-47 tends to dominate the PBDE egg profile of aquatic feeding birds compared to more terrestrial species (Chen and Hale, 2010). The comparatively low δ34S isotope values in herring gull eggs (Table 1) provide evidence of the reliance of this species on landfill for foraging. Ramos et al. (2009) found especially low δ34S isotopic values in regurgitant of yellow-legged gull nestlings whose parents were dietary specialists on landfill. The negative relationship between δ15N isotope values and BDE-28 concentrations in herring gull eggs (Fig. 5) suggests that for those individuals foraging at higher trophic levels and thus consuming less refuse, they may be less exposed to this congener and/or differentially metabolize and transfer BDE-28 to eggs compared to the other gull species. This hypothesis warrants further investigation since the current study appears to be the first to report this negative congener-isotope association in free-living birds.

 *4.5.* *Behavioural observations*

 Our findings suggest that there were multiple routes of BFR exposure for these adult-plumage gulls when frequenting the study landfill, including their diet (Section 4.4), behaviour and potentially also inhalation, that likely contributed to the observed differences amongst species in egg BFR concentrations and profiles.

 Behavioural observations allowed us to identify four potential routes of BFR exposure to gulls on landfill: pecking at the substrate, swallowing items, walking across the substrate and/or standing stationary on the substrate. There were considerable variations among the three gull species in their major behaviours on the landfill, i.e., number of pecks, number of paces, time stationary. However, the extent to which these behavioural differences among the gulls may have contributed to the variations in egg contaminant burdens warrants further research, ideally with observations of individuals for which egg burdens are known. Such future studies should also investigate other possible routes of exposure to environmental chemicals for gulls, including preening of contaminated plumage while loafing (Jaspers et al., 2007) and inhalation (Gentes et al., 2015; Sorais et al., 2020). Compared with mammals, the avian respiratory system requires a larger air volume to be inhaled to supply a near-constant airflow through the lungs and air sacs (Brown et al., 1997), potentially making birds especially susceptible to inhaled gaseous/particulate phase BFRs and other contaminants.

 We studied three gull species that are of national/international conservation concern and that frequented a remote landfill in Scotland, UK. Their egg concentrations of BFRs, reflecting maternal exposure, transfer and deposition, are at some of the highest levels of PBDEs and HBCDDs for gull species compared to those in mainland Europe or North America also using landfill. We also identified the highest concentration of DBDPE ever reported in biota to date. Given previous avian studies demonstrating adverse reproductive changes in relation to similar egg concentrations of some of these FRs (Guigueno and Fernie, 2017), it is important for future research to address possible reproductive and/or physiological implications for these species as a result of their contaminant burdens and association with landfill. Our results, in conjunction with those of previous studies, identify landfill as an important point source of FRs (and other environmental contaminants) for birds. Our study also highlights that further research is required to characterize and understand better the relative importance of direct ingestion, inhalation, dermal contact and preening as pathways of exposure to BFRs and other environmental contaminants that birds are exposed to at landfill. Given the far greater numbers of herring gulls observed on landfill in the present study, and the considerable FR concentrations measured in their eggs, we suggest that herring gulls may be a highly appropriate bioindicator species for similar (landfill) studies.

**Declarations of interest**

None.

**Acknowledgements**

We gratefully acknowledge provision of a CENTA studentship award to ADWT by the UK Natural Environment Research Council (NERC ref. NE/L002493/1), NERC Core Facility funding to SJR and ADWT for analytical support at the SUERC (NERC ref. EK290-13/17), and Environment and Climate Change Canada (ECCC) for support to KJF. This research also received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement no. 734522 (INTERWASTE) project. We are also grateful to the Waterbird Society and the Seabird Group for funding made available to ADWT.

**Appendix A. Supplementary Information**

Supplementary Information to this article can be found online at [PLEASE INSERT WEB ADDRESS]

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