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Trace element concentrations in harvested auks from Newfoundland: Toxicological risk of a traditional hunt

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ABSTRACT

Common (*Uria aalge*) and Thick-billed Murres (*Uria lomvia*) are apex predators in the North Atlantic Ocean, and are also subject to a traditional hunt in Newfoundland and Labrador during the winter months, along with small numbers of illegally harvested Razorbills (*Alca torda*). Because of their high trophic position, auks are at risk from high contaminant burdens that bioaccumulate and biomagnify, and could therefore pose a toxicological risk to human consumers. We analysed trace element concentrations from breast muscle of 51 auks collected off Newfoundland in the 2011–2012 hunting season. There were few differences in contaminant concentrations among species. In total, 14 (27%) exceeded Health Canada or international guidelines for arsenic, lead, or cadmium; none exceeded guidelines for mercury. Cadmium concentrations >0.05 μg/g have persisted in Newfoundland murres for the last 25 years. We urge the integration of this consumptive harvest for high-trophic marine predators into periodic human health risk assessments.

1. Introduction

Many remote and indigenous populations harvest marine predators for economic, cultural, and subsistence reasons (Dehn et al., 2006; Diamond, 1987; Kuhnlein and Chan, 2000; Merkel and Barry, 2008; Skira et al., 1985). Globally, there are few regulated harvests of marine birds, and those that remain unregulated are generally declining in popularity (Gaston and Robertson, 2010; Richardson, 1984; Skira, 1990) or are stable (Newman et al., 2009; Olsen and Nørrevang, 2005). As apex predators in the marine environment, seabirds are exposed to bioaccumulated and biomagnified contaminants (Burger and Gochfeld, 2002). Recently, there has been an increased awareness by researchers of the human health effects of the consumption of traditional foods, including apex predators (Dewailly et al., 1992; El-Din Bekhit et al., 2011; Hoekstra et al., 2005; O'Hara et al., 1999; Ostertag et al., 2009), but there has been little toxicological monitoring of traditional harvests (Burger et al., 2007; Lavers and Bond, 2013).

There is a legal, non-indigenous, traditional hunt for murres (*Uria* spp.), large seabirds in the family Alcidae, in Newfoundland and Labrador (Chardine et al., 2008). This harvest is unique in that it is North America's only legal non-indigenous harvest of seabirds; both Common (*Uria aalge*) and Thick-billed Murres (*Uria lomvia*) are harvested at sea between September and March (Elliot, 1991; Environment Canada, 2011). The practice of harvesting seabirds in general, and "turr hunting" in particular, is a longstanding tradition in Newfoundland and Labrador (Montevecchi et al., 2007; Pope, 2009; Tuck, 1952), and though its practice is generally decreasing (Gaston and Robertson, 2010), technological advances have allowed the remaining hunters to exploit the murre populations considerably (Montevecchi et al., 2007; Regular et al., 2010). In the 1970s and 1980s, upwards of 400,000–750,000 murres were shot annually (Chardine et al., 1999; Wendt and Cooch, 1984), but this declined in the 1990s to less than 200,000 through a combination of tighter hunting regulations and a decreasing number of hunters (Chardine et al., 1999; Gaston and Robertson, 2010). The majority of harvested birds are Thick-billed Murres, but Common Murres comprise 5–20% in some areas (Elliot, 1991; Hedd et al., 2011); up to 4–5% of the annual harvest is composed of Razorbills (*Alca torda*), a similar-looking auk for which there is no legal hunt (Blanchard, 1984; Lavers et al., 2009).

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In general, the entire murre is cooked and most is consumed, particularly breast muscle (Montevecchi et al., 2007), and often the viscera, known locally as the "lights" (PCR, pers. obs.).

Murres feed on fish, such as capelin (*Mallotus villosus*), Arctic cod (*Boreogadus saida*), or Atlantic cod (*Gadus morhua*) as well as invertebrates (Elliot et al., 1990; Moody and Hobson, 2007; Rowe et al., 2000). Furthermore, murres are harvested using lead (Pb) shot, which can become embedded or leave small fragments in edible portions, causing high Pb concentrations in foods (e.g., soup, meat) that are up to 25 times higher than other food sources of Pb (Johansen et al., 2001, 2004, 2006b).

Previous assessments of contaminants in murres involved pooled samples (which eliminates individual variation), considered one (or few) elements or compounds, had small sample sizes, or did not discuss the human health implications of the murre harvest directly (Braune et al., 1999; Donaldson et al., 1997). All previous sampling was conducted at least 20 years ago, and global concentrations of some contaminants, such as mercury (Hg) have increased substantially since then (Lamborg et al., 2014; Streets et al., 2009; UNEP, 2013).

Current Health Canada guidelines limit mercury (Hg) and lead (Pb) in meat to 0.5 μg/g, and arsenic (As) to 3.5 μg/g in meat products and fish protein on a wet weight basis (Government of Canada, 2013; Health Canada, 2007). There are no current Canadian national guidelines for cadmium (Cd), but both the European Commission, and Australia and New Zealand have adopted a recommendation of 0.05 μg/g in meat and meat products (Department of Health and Ageing, 2011; European Commission, 2006).

Our objectives were to (1) assess the toxicological threat posed by the consumed portions of harvested murres in Newfoundland, (2) compare these results with previous studies of contaminants in murre muscle, and (3) place the results in a broader context of wildlife toxicology and human health interactions.

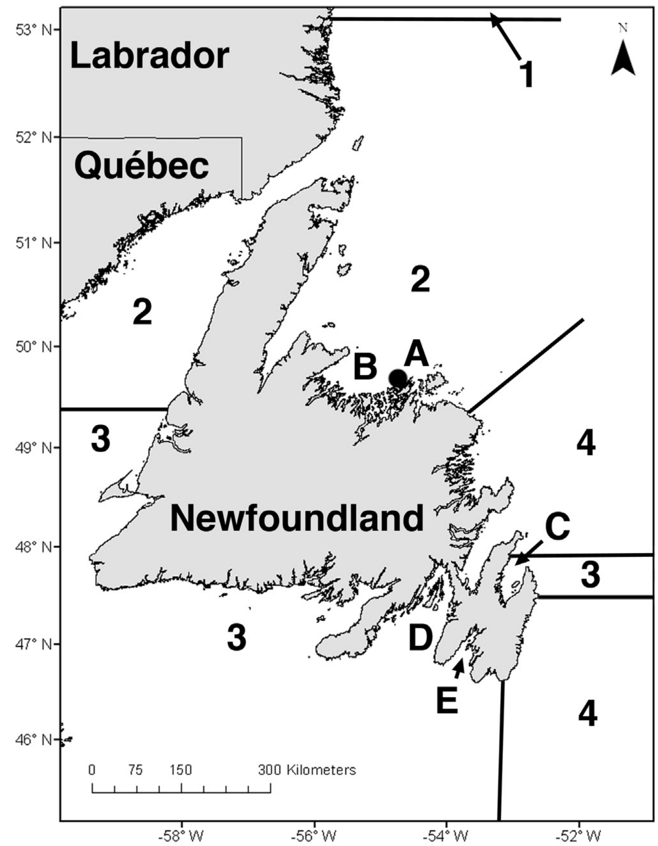

Fig. 1. We collected murres (*Uria* spp.) from Twillingate (A, circle), Conception Bay (C), and St. Mary's Bay (E). Confiscated Razorbills (*Alca torda*) were shot in Notre Dame Bay (B), and murres examined for lead shot also came from Placentia Bay (D) and Conception Bay. The approximate boundaries of murre hunting zones (1–4) are also indicated (Environment Canada, 2011).

2. Methods

2.1. Sample collection and preparation

Birds were collected under permit from the Canadian Wildlife Service off the Newfoundland coast near Twillingate (49.67°N, 54.79°W, $n=34$) in November 2011, St. Mary's Bay in January 2012 (46.93°N, 53.69°W, $n=6$) and Conception Bay in January 2012 (47.75°N, 53.00°W, $n=4$). Razorbills ($n=7$ from Notre Dame Bay (49.83°N, 55.37°W)) were obtained from seizures of illegally shot birds (Environment Canada enforcement personnel; Fig. 1). Birds were frozen within 8 h of collection, and shipped frozen to the National Wildlife Research Centre in Ottawa, Ontario for dissection. Birds were thawed, and the species identified. The left breast muscle from each bird was removed and placed in a sterile glass jar covered with aluminium foil to prevent moisture loss, and frozen. Prior to analysis, 3–5 g (wet weight) was transferred to a small vial, and freeze-dried for 48 h.

2.2. Trace element analysis

The resulting 1.0–1.5 g dried muscle was subsampled for analysis using inductively coupled plasma mass spectrometry (ICP-MS) at Memorial University of Newfoundland, or for analysis of total Hg using atomic absorption spectrometry at the University of New Brunswick. For ICP-MS analysis, between 0.3 and 0.7 g of each sample was placed in an acid-cleaned screw cap Teflon vessel with 1 ml of 8 M HNO_3 on a 70 °C hotplate. After the first hour, another 1 ml of 8 M HNO_3 was added. After digesting for 24 h, the hotplate temperature was lowered to 50 °C, and 1 ml H_2O_2 was added, and the caps removed. When the tissue was completely digested, the vessels were recapped, and the hotplate temperature was raised to 70 °C for 3 h. The digested samples were transferred to clean, sealed containers, and diluted 500× with Millipure water. For ICP-MS analysis, 1 ml of this solution was pipetted into a clean 10 ml tube with 4 ml of Millipure water, making the final solution a 2500× dilution. All tools were either cleaned with 95% ethanol or were single-use; all chemicals were ACS analytical grade.

ICP-MS analysis was conducted on a PerkinElmer ELAN DRCCII, and instrument set-up followed Bond and Lavers (2011) and Friel et al. (1990). Procedural blanks and certified reference materials were run every 15–20 samples. The reference materials used were NIST 2976 and NIST 2977 (mussel tissue), and recovery details are presented in Table 1. We restricted our analysis to those elements that could be measured reliably as indicated by recovery of the reference materials: Ca, Fe, Mn, Co, Cu, Zn, As, Br, Rb, Sr, and Pb. Elemental concentrations are presented as mean ± S.D. in parts per million (μg/g) based on wet weight.

Samples were analysed for total Hg using a Milestone DMA-80 direct mercury analyser (Haynes et al., 2006) and calibrated using procedural blanks, and reference materials DORM-2 and TORT-3 (National Research Council, Ottawa) every 10 samples; recovery of reference materials was 90.5 ± 5.8% and 104.5 ± 8.3% respectively. Approximately 10% of samples were analysed in duplicate, and the mean standard deviation of duplicates was 0.015 ± 0.009 μg/g. Detection limits for all elements are presented in Table 2.

2.3. X-ray procedures for detecting lead shot

Because murres are hunted with lead shot, and shot or fragments of shot can contaminate tissue (Johansen et al., 2001), muscle samples were x-rayed to determine the quantity of lead

Table 1
Recovery of two certified reference materials (CRMs; NIST 2976 and 2977) using inductively coupled plasma mass spectrometry (ICP-MS). $N=3$ for each CRM. Measured and assigned concentrations are presented as μg/g. CV=(S.D./mean)× 100.

NIST 2976	Mean ± SD	CV	Assigned	% Recovery
Ca	7417 ± 929	12.53	7600	98
Fe	217 ± 17	8.05	171	127
Mn	36 ± 1	1.53	33	108
Co	0.57 ± 0.03	4.72	0.61	94
Cu	3.99 ± 0.75	18.81	4.02	99
Zn	137 ± 4	2.78	137	00
As	12.47 ± 0.55	4.40	13.3	94
Br	288 ± 35	12.18	329	88
Rb	4.85 ± 0.38	7.90	4.14	117
Sr	69 ± 2	2.56	93	74
Cd	0.82 ± 0.08	9.56	0.82	100
Pb	1.27 ± 0.21	16.17	1.19	107
NIST 2977	**Mean ± SD**	**CV**	**Assigned**	**% Recovery**
Ca	7633 ± 211	2.77	8300	92
Fe	285 ± 30	10.38	274	104
Mn	21.76 ± 1.30	5.97	23.93	91
Co	0.34 ± 0.08	24.55	0.48	72
Cu	8.4 ± 0.7	8.33	9.42	89
Zn	126 ± 5	3.57	135	93
As	7.19 ± 0.63	8.83	8.83	81
Br	260 ± 35	13.36	215	121
Rb	6.7 ± 0.6	8.18	6.7	100
Sr	61.8 ± 3.6	5.8	69.3	89
Cd	0.27[a]		0.18	153
Pb	2.02 ± 0.19	9.17	2.27	89

[a] $n=1$ where the measured concentration > level of detection.

Table 2
Detection limits of elements measured by inductively coupled plasma mass spectrometry and direct mercury analysis, and the number of samples (out of 51 tested) that were below each element's threshold. Detection limits are presented as μg/g.

Element	Detection limit	Samples < detection limit
Ca	61	2
Fe	41	0
Mn	0.07	0
Co	0.06	40
Cu	0.45	0
Zn	1.05	0
As	0.23	0
Br	3.48	0
Rb	0.05	0
Sr	0.02	1
Cd	0.17	31
Hg	0.0055	1
Pb	0.06	31

shot at the University of Saskatchewan's Western College of Veterinary Medicine. Because embedded lead shot could also originate from our own collection, we examined 91 Thick-billed and four Common Murres that were oiled and found dead on beaches in Placentia Bay, Newfoundland, in November 2004 (Robertson et al., 2006), and an additional 57 Thick-billed Murres that were found dead on beaches in Conception Bay, Newfoundland in April 2007 and March 2009 (McFarlane Tranquilla et al., 2010), using a fluoroscope (Fig. 1). In each case, whole birds were examined, and we counted whole or large fragments of lead shot.

2.4. Statistical analysis

Concentrations were not normally distributed, and because we had samples below the level of detection for some elements, data were censored (Helsel, 2012). For Ca, Sr, and Hg, only one or two samples were below the detection level, so we used a maximum likelihood estimation function in the package *NADA* (Lee, 2013) in R 3.1 (R Development Core Team, 2014) for summary statistics. The majority of Co, Cd, and Pb samples were below the detection limit, which impeded our ability to use a known distribution to estimate censored values (Helsel, 2012), so we used regression on order statistics to produce summary statistics (Helsel, 2012).

For elements without censored data, we tested for differences in log-transformed concentrations among species using a general linear model with Tukey's honest significant difference post-hoc test. Differences among species in concentrations of elements with censored values were assessed using a Generalized Wilcoxon Score Tests (Helsel, 2012). When there were significant differences among species, we used Gao's generalized Campbell–Skilling's test (Campbell and Skillings, 1985; Gao et al., 2008) in the package *nparcomp* (Konietschke, 2012) as a post-hoc test.

Results were considered significant when $p<0.05$, and are presented as μg/g on a wet weight (ww) basis based on measured percent moisture for comparison with food safety standards.

3. Results

3.1. Trace elements

We found species differences for Fe (Common Murre > Razorbill; $p=0.03$), Rb (Thick-billed Murre > Razorbill, $p=0.03$), and Pb (Razorbill > Common Murre; Table 3). We did not have adequate samples to test for age effects.

Of the 51 muscle samples, none exceeded the Canadian guidelines of 0.5 μg/g for Hg, and only one exceeded the Canadian guidelines of 0.5 μg/g for Pb (a Thick-billed Murre; 1.37 μg/g). One Thick-billed Murre also had muscle concentration of As of 4.50 μg/g, above the Canadian guideline of 3.5 μg/g. There were 14 samples that exceeded the European, New Zealand, and Australian guideline of 0.05 μg/g for Cd, including a sample that had 1.20 μg/g Cd. These 14 birds represented all three species (Common Murre: 4; Thick-billed Murre: 9; Razorbill: 1). In total, 14/51 birds (27%) exceeded established food safety guidelines for one or more trace elements (Table 4).

3.2. Lead shot

Lead shot was detected in 3/51 samples (6%); there were two cases of a single piece of shot, and one case of two pieces in the left breast muscle. Of the murres from Placentia Bay, 8/95 (8%) had

Table 3
There were few significant differences in the concentrations of elements among species. Samples below the detection limit were not included, so some comparisons were not made; results are from a general linear model.

Element	$\log_{10}$-transformed?	df	*F*	*p*
Ca	Yes	2, 46	0.13	0.88
Fe	No	2, 48	3.74	0.03
Mn	Yes	2, 48	0.13	0.88
Co	Yes	2, 8	0.84	0.47
Cu	Yes	2, 48	0.20	0.82
Zn	Yes	2, 48	1.22	0.31
As	Yes	2, 48	0.58	0.56
Br	No	2, 48	1.49	0.24
Rb	Yes	2, 48	3.82	0.03
Sr	Yes	2, 47	0.18	0.84
Cd	Yes	2, 17	5.89	0.01
Hg	Yes	2, 47	1.11	0.34
Pb	Yes	2, 15	0.41	0.67

Table 4
Concentrations of trace elements in muscle of harvested auks in Newfoundland. Values are presented as mean ± S.D. in μg/g on a wet weight basis; %*M* = percent moisture.

Element	Common Murre	Thick-billed Murre	Razorbill
Ca	155.24 ± 57.32	295.85 ± 773.22	159.98 ± 79.59
Fe	110.08 ± 24.35	103.34 ± 21.17	83.44 ± 20.03
Mn	0.69 ± 0.12	0.85 ± 1.00	0.69 ± 0.20
Co	–	0.04 ± 0.05	–
Cu	5.36 ± 0.44	6.25 ± 3.78	5.65 ± 1.11
Zn	11.29 ± 11.29	13.63 ± 13.63	13.03 ± 13.03
As	0.28 ± 0.18	0.56 ± 0.78	0.32 ± 0.13
Br	6.10 ± 1.46	5.77 ± 1.84	4.79 ± 1.25
Rb	1.35 ± 0.29	1.56 ± 0.41	1.20 ± 0.25
Sr	0.67 ± 0.38	1.49 ± 4.33	0.70 ± 0.52
Cd	0.13 ± 0.05	0.30 ± 0.42	0.04 ± 0.03
Hg	0.09 ± 0.05	0.12 ± 0.08	0.12 ± 0.03
Pb	0.02 ± 0.01	0.14 ± 0.40	0.02 ± 0.01
%M	72 ± 1	72 ± 2	71 ± 2
n	11	32	8

between 1 and 5 pieces of shot (2.4 ± 1.7 pieces/bird); all were Thick-billed Murres. Only 1/57 Thick-billed Murres from Conception Bay (2%) contained lead shot – a single piece in an adult.

4. Discussion

Murres are harvested throughout the circumpolar region (Merkel and Barry, 2008; Olsen and Nørrevang, 2005; Petersen, 2005), and there are a handful of toxicological studies, though none that have related concentrations in birds to regulations surrounding food safety. Donaldson et al. (1997), using pooled samples of muscle from Thick-billed Murres banded in the Canadian Arctic and collected (shot) off Newfoundland between 1987 and 1990 and found none exceeding Health Canada guidelines for Hg, and 1/10 exceeding guidelines for Pb. All pooled samples had Cd > 0.09 μg/g, above the 0.05 μg/g standard. Of four Thick-billed Murres collected from Newfoundland and the Canadian Arctic between 1990 and 1993, and 26 Common Murres collected in November 1988 from Scotland, none exceeded Health Canada's guidelines for Hg (Braune et al., 1999; Stewart et al., 1994). Common and Thick-billed murres from Hornøya, Norway also had low concentrations of Hg (0.42 ± 0.05 μg/g and 0.33 ± 0.14 μg/g dw, respectively), but mean Cd concentrations (assuming 71% moisture content of muscle; Table 3) were 0.05 μg/g and 0.15 μg/g, respectively (Wenzel and Gabrielsen, 1995), indicating that many birds exceeded the 0.05 μg/g guideline. Throughout the Barents Sea (Svalbard, Franz Josef Land, Novaya Zemlya, Seven Islands Archipelago), Thick-billed and Common Murres' muscle had > 0.05 μg/g Cd routinely, while As, Hg, and Pb were rarely above their respective consumption guidelines (Borgå et al., 2006; Savinov et al., 2003). The same pattern was observed in Thick-billed Murre muscle form northern Baffin Bay (Cd: 0.55 ± 0.13 μg/g ww; As, Hg, and Pb below food safety standards) (Borgå et al., 2006), and Greenland (Cd: 0.84 ± 0.57 μg/g ww; Hg: 0.30 ± 0.13 μg/g ww) (Nielsen and Dietz, 1989). Common Eiders throughout Arctic Canada also had high, and increasing concentrations of hepatic Cd (Mallory et al., 2014). Together, these results indicate that Cd has been a persistent contaminant in edible portions of murres and other hunted marine birds throughout their North Atlantic range (Falk and Durinck, 1992; Merkel and Barry, 2008; Olsen and Nørrevang, 2005; Petersen, 2005).

We found a relatively low incidence of embedded lead shot, between 2% and 8%, or 3–4 birds per hunter per season. While direct ingestion of the shot is not common, cooked breast meat of Thick-billed Murres from Greenland had Pb concentrations 10 times higher than birds not killed with lead shot, and is a significant source of human Pb ingestion (Johansen et al., 2001). With a single exception, all of the murres in our study had Pb concentrations lower than the 95% confidence interval reported in Johansen et al. (2001) of 0.22 μg/g (wet weight); we recorded one bird with 1.37 μg/g (wet weight), but this bird contained no embedded lead shot in the left breast muscle. Though lead shot was rare in our sample of beached birds, this is likely an underestimate. The authors' experience suggests that the incidence of lead shot in harvest birds is roughly 20–25%, and we cannot discount this potential source (Johansen et al., 2001). Indeed, significant cases of lead shot in the appendices of residents of northern Newfoundland highlight the potential consequences of ingesting lead shot from murres and other game (Carey, 1977; Reddy, 1985). Regardless of the ultimate source of Pb (shot or prey), the vast majority of samples analysed had Pb concentrations below levels of concern.

While most murres fell below established food safety standards, 27% exceeded at least one (usually Cd). This is likely an underestimate, as an unknown number of murres below the detection limit for Cd (0.17 μg/g; Table 2) could exceed guidelines. This is a lower proportion than harvested Short-tailed Shearwaters (*Puffinus tenuirostris*) in Tasmania (57%), but higher than Sooty Shearwaters (*P. griseus*) from New Zealand (0%) (El-Din Bekhit et al., 2011; Lavers and Bond, 2013). All of the murres that were above food safety standards exceeded standards for Cd – 0.05 μg/g, or approximately 1.04×10^{-4} mg Cd/kg human body mass (assuming approximately 160 g of breast muscle per murre, and a 77 kg person) (McMahon et al., 2012). Significant kidney damage can occur with prolonged exposure to Cd at the rate of 0.05 mg Cd/day (Järup et al., 1998). Given the highly seasonal nature of the murre harvest, and relatively infrequency of consumption (a typical hunter might take 40 murres per season; authors' pers. obs.), the concentrations we observed are likely of low concern for most hunters; prolific consumers would of course be at greater risk. Furthermore, only three murres (6%) had Cd > 0.20 μg/g, which also suggests little risk from Cd ingestion. However, if kidneys and liver, which tend to have higher concentrations of Cd than muscle, are also consumed, the exposure, and risk would be greater (Mallory et al., 2014).

The main source of Cd exposure is tobacco and food, but Cd in human kidneys has a half-life of 15 years, meaning that any daily exposure from murres likely contributes relatively little to the overall body burden (Goyer, 1997; Mueller et al., 2011; Van Oostdam et al., 1999). Despite consuming a diet high in Cd, Greenlanders do not exhibit elevated liver or kidney Cd concentrations relative to northern Europeans or Canadians, suggesting that most Cd from traditional marine foods may not be bioavailable (Johansen et al., 2006a). A better understanding of acceptable intake of Cd, and its bioavailability is required (Johansen et al., 2006a; Mueller et al., 2011).

Arsenic exists in many forms that were not distinguishable in our analysis, but the predominant form in seabird muscle is arsenobetaine (Fujihara et al., 2004; Kunito et al., 2008). Arsenobetaine was thought to be relatively inert in humans (Brown et al., 1990), but more recent work suggests it may be resident for much longer than previously thought (Newcombe et al., 2010). Further research is needed both in terms of the human health effects of As species, and on their distribution in traditional foods.

Regardless, the rapidly changing Arctic and North Atlantic environments, coupled with an increase in offshore oil and gas development in Newfoundland waters and associated risks to marine predators highlights the importance of regular monitoring of contaminants, both metals, as well as legacy and novel organic compounds (Burke et al., 2012; Wiese and Ryan, 2003; Wilhelm et al., 2009).

5. Conclusions

Toxicological monitoring of wildlife subsistence hunts is often infrequent, owing to the remote regions in which the harvests occur, and in many cases, sensitivity to cultural traditions (Stephens et al., 2006). While regular monitoring of marine mammal consumption and toxicology by Arctic communities is more frequent (Hoekstra et al., 2005; Moses et al., 2009; O'Hara et al., 1999; Welfinger-Smith et al., 2011), assessments of consumed seabird toxicology are rare (Donaldson et al., 1997; El-Din Bekhit et al., 2011; Lavers and Bond, 2013). The harvest of murres in Newfoundland and Labrador is unique in that it is a non-indigenous consumptive harvest for apex marine predators. The harvest of murres and other seabirds in circumpolar countries is a generally heretofore unexplored method for obtaining samples, with the added benefit of informing populations of the risks associated with this consumptive harvest.

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