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High human exposure to lead through consumption of birds hunted with lead shot

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"Capsule": *Lead shot contaminates the edible parts of birds so that tolerable human lead intake is exceeded.*

Abstract

We assess lead contamination of Greenland seabirds killed with lead shot having studied thick-billed murre and common eider, the two most important species in the diet. The lead concentration is very high in meat of eiders killed with lead shot (mean 6.1 μg/g-wet wt, 95% CL 2.1–12). This level is about 44 times higher than in drowned eiders and eight times higher than in shot murres. Analyzing whole breasts instead of sub-samples reveals about seven times higher lead levels in birds' meat. We conclude that in some cases the lead intake by Greenland bird eaters will largely exceed the FAO/WHO tolerable lead intake guideline and that lead shot is a more important source of lead in the diet than previously estimated.

Keywords: Lead shot; Contamination; Birds; Human exposure; Greenland

1. Introduction

Although lead shot has been banned or has restricted use in some countries, it is still widely used in bird hunting, and the replacement of lead with less toxic alternatives is progressing only slowly (Beintema, 2001). In Denmark for example, this ban is total whereas in North America the use of lead shot is restricted only for migratory game birds while lead shot can still be used to harvest indigenous game birds such as grouse. In Greenland there are no restrictions.

Lead shot ingestion and poisoning of waterfowl is a well-documented effect of the use of lead shot, and secondary poisoning in raptors is also a recognized environmental impact (see review in Scheuhammer and Norris, 1995). This has been the main reason for replacing lead in ammunition, whereas less attention has been given to human lead exposure from eating game hunted with lead shot.

Lead shot pellets eaten unintended may result in increased lead exposure and intoxication in humans (Hillman, 1967; Madsen et al., 1998; Johansen and Nygård, 1987). For some groups lead shot from the consumption of game are common in the gastro-intestinal system as found in people from the western James Bay region (Tsuji and Nieboer, 1997) or people from Newfoundland (Reddy, 1985).

However, not only whole pellets remain in tissue from species killed by lead shot. High lead concentrations have been attributed to the presence of lead fragments 1–2 mm in length to very fine dust and were judged to be the result of the disruption of lead shot pellets upon collision with bone (Frank, 1986). Thus tissue may become contaminated with high concentrations of lead through this mechanism, as found in Canada (Scheuhammer et al., 1998) and Greenland (Johansen et al., 2001). In Canada, 11% of lead levels in breast muscle of waterfowl exceeded the national residue guideline value of 0.5 μg/g-wet wt for fish protein (Scheuhammer et al., 1998). In Greenland, we found that 11% of lead levels in breast muscle of thick-billed murre (*Uria lomvia*) exceeded the Danish residue guideline value of 0.3 μg/g-wet wt

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(Johansen et al., 2001). Since seabirds are important in the local diet, we concluded that birds hunted with lead shot are probably the most important lead source for many people in Greenland. Other studies have also pointed to game hunted with lead shot as a lead source in the diet, e.g. Tsuji et al. (1999; 2001) for the First Nation Cree in Northern Ontario, Canada, and Kosatsky et al. (2001) for Montrealers consuming sportfish and waterfowl. In Greenland, a relationship between bird intake and blood lead has been documented. In a cross sectional population survey in West Greenland, Bjerregaard et al. (in prep.) found that participants reporting less than weekly intake of seabirds had blood lead levels around 75 μg/l, while those reporting to eat seabirds several times per week had more than 50% higher blood lead levels.

In an earlier study (Johansen et al., 2001) we found that lead was not homogeneously distributed in the breast muscle samples analyzed. Even after removal of visible pellets and homogenization of the whole breast, duplicate analyses in meat from the same bird showed differences between duplicates far exceeding the analytical variability of <25% estimated relatively from inter-laboratory comparison programs. For example, in two sub-samples of the homogenate from the same breast, 0.95 μg/g lead wet wt was measured in one subsample and 0.16 μg/g in the other. Consequently the uncertainty of the estimated lead concentration in the meat was high in this study. Scheuhammer et al. (1998) also found highly variable lead concentrations in different sub-samples from the same tissue in some cases.

To obtain a more reliable basis for estimating human lead intake, we have re-analyzed whole breast muscles of the murres and have included common eider (*Somateria mollissima*) which also is important in the local diet. Here we present these new data and conduct an assessment human lead intake from eating seabirds hunted with lead shot in Greenland. We also compare lead levels in eiders shot with levels in drowned birds, so that the significance of lead shot to the lead contamination of the edible portions can be evaluated.

2. Materials and methods

Southwest Greenland waters are important wintering grounds for seabirds, in particular thick-billed murre and common eider. During winter, substantial numbers of about 3000,000 of these two species are killed here annually with lead shot.

Birds killed with lead shot were collected during winter hunting in the Nuuk region (64°N, 51°W): 35 murres in 1998 and 25 common eiders in 2000. Twenty five eiders not killed with lead shot (e.g. drowned in fishing net) were also collected. Each bird was examined for embedded lead shots by taking high resolution X-ray photographs (35 × 40 cm) at the local hospital in Nuuk or at the Royal Veterinary School in Copenhagen. From the X-ray photographs shot pellets were located and counted. Birds were frozen whole and send deep-frozen Greenland to the National Environmental Research Institute in Denmark.

The birds were thawed at room temperature for about 1 day and aged and sexed based on plumage character and on sexual organs (Falk and Merkel, 2001). Wings, head, tarsi and toes were cut off, the bird was skinned and the viscera were removed. Then each skinned carcass without viscera was boiled according to a recipe commonly used in Greenland. After boiling, the right pectoral muscle was cut out with stainless steel blades for chemical analysis. If the X-ray photograph had shown presence of shot pellets in the muscle, visible pellets were located and removed. We used this procedure to simulate the human lead exposure from eating seabirds in Greenland, assuming that the consumer would detect and not ingest whole pellets.

The chemical analyses were performed at the National Environmental Research Institute, Department of Arctic Environment (DAE). After ashing of the entire pectoral muscle in a porcelain crucible at 550 °C, the ash was dissolved in Merck Suprapur nitric acid. After dissolution, samples were transferred to polyethylene bottles and diluted to approx. 25 g with milli Q water. Lead analyses were performed directly in these solutions. High lead concentrations were determined by flame AAS (Perkin Elmer AAnalyst 300) and low concentrations by graphite furnace AAS (Perkin-Elmer AAnalyst 800). The standard addition method was used in the graphite furnace measurements. Analytical quality was checked regularly by analyzing certified reference materials (Dorm-1, Tort-1, Bovine-liver and Sewage-sludge), and is also evaluated by interlaboratory comparison programs, in which DAEs chemical laboratory has participated. These have shown that for lead concentrations above 0.02 μg/g, the deviation by DAEs chemical laboratory is within 25% of the assigned lead values. For lead concentrations below 0.02 μg/g QUASIMEME, the organizer of the inter-laboratory comparisons, presented only indicative values, because of disagreement on concentration between too many of the participating laboratories (Wells and Cofino, 1997a; 1997b). In these cases DAEs chemical laboratory always found lower lead values than those given by QUASIMEME.

The method was checked by analyzing spiked whole chicken breasts, as there is a risk of both lead contamination and lead loss during ashing. Less than 10% lead was lost during ashing and in unspiked chicken breast the mean lead concentration was 0.034 μg/g (wet wt). Based on this we conclude that the detection limit of the method ashing whole breast is less than 0.04 μg/g-

wet wt. Two values from eiders and five from murres were below this detection limit, but the chemical laboratory reported a value (between 0.008 and 0.035 μg/g-wet wt.) that we used in calculating means and confidence limits.

As the frequency distribution of the lead concentration is very skewed, the "bootstrap method"(Efron and Tibshirani, 1993) using 1000 're-samples' was chosen to estimate confidence limits of the mean concentrations. Paired *t*-test on logarithmic transformed data was used to test differences of mean lead concentrations between sub-samples and whole breasts from the same birds. Linear regression analyses were applied to test for relationship between the lead concentration in the breast and number of lead shot pellets.

3. Results and discussion

Table 1 summarizes the results from our study. Four single high lead concentrations (above 1000 μg/g-wet wt.) were omitted from data treatment, because it was suspected that in these four cases whole pellets or large pellet fractions were not detected and removed before analysis. We did this to simulate that normally people would detect and not eat whole pellets or large pellet fractions. There is a possibility that these high concentrations are caused by small fragments or dust, which would not be detected, and therefore eaten by the consumer, which would cause an under-estimate of the human lead exposure. However the lead concentration is so high in the four cases that it seems unlikely that lead in these breasts would not be present as whole pellets or large pellet fractions. We have computed that in these cases the amount of lead present is equal to the weight of one shot pellet.

3.1. Implications of method on estimates

The mean lead concentration, 0.73 μg/g-wet wt., in whole murre breasts is almost seven times higher than that found when we analyzed 0.5–1 gram sub-samples from the same birds (Table 2), even after freeze-drying and fine-grinding of the whole breast before taking a sub-sample for lead analysis (Johansen et al., 2001). This difference is statistically significant (paired t-test on log-transformed data, $t=-5.29$, $P<0.01$). In Fig. 1 we

Table 2
Lead concentration (μg/g-wet wt) in two sample types from thick-billed murre. Confidence limits of 95% are computed from the "bootstrap method"

Sample type	*n*	Mean	S.D.	95% conf. limit of mean
0.5–1 g sub-sample	32	0.111	0.254	0.032–0.210
Whole breast	32	0.729	2.963	0.155–1.937

Table 1
Lead concentration (μg/g-wet wt) in whole breasts of thick-billed murre and common eider. Confidence limits of 95% are computed from the "bootstrap method"

Species	Killing method	*n*	Mean	S.D.	95% conf. limit of mean
Thick-billed murre	Shot	32	0.73	2.9	0.16–1.9
Common eider	Shot	25	6.1	13	2.1–12
Common eider	Drowned	24	0.14	0.13	0.09–0.19

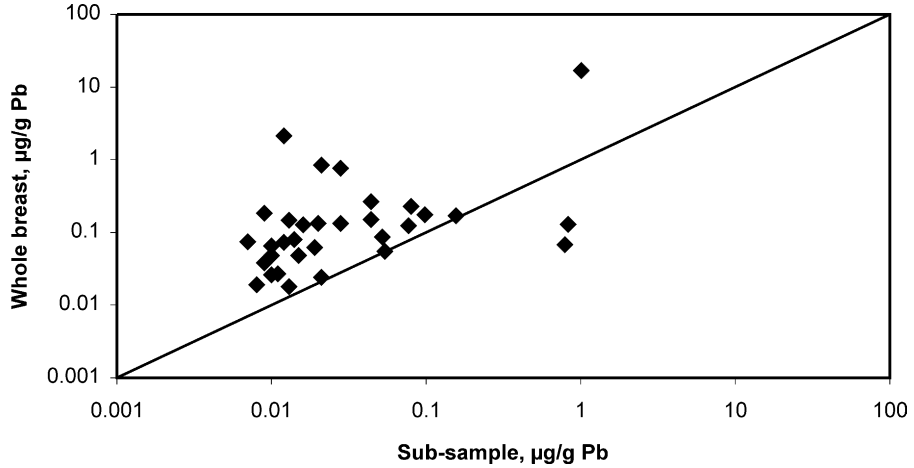

Fig. 1. Lead concentration (μg/g-wet wt) in breast meat of thick-billed murre. Concentration in whole breast plotted against 0.5–1 gram sub-sample from the same bird. Line shows expected correlation between the two variables.

have plotted corresponding concentrations in the whole breast and a sub-sample from the same bird. The lead concentration in the sub-sample is lower than that found in the whole breast in only two cases. We think this is caused by an inhomogeneous distribution of lead (as fragments or dust) in the breast, so that all non-visible lead fragments will be included when the whole breast is analyzed, whereas only a fraction of these fragments is included in the sub-sample. In the two cases where we find a higher lead concentration in the sub-sample than in the whole breast, the likely explanation is that one or more lead fragments was in the sub-sample by chance. That lead has an inhomogeneous distribution in bird's meat is supported by the findings of Scheuhammer et al. (1998) and Johansen et al. (2001).

Our finding has implications for other regions where lead shot are used since it indicates that human lead exposure from hunting with lead shot is significantly larger than previously estimated.

3.2. Lead levels

The lead concentration is significantly higher (by a factor of about eight) in eider than in murre, and it is much higher (by a factor of 44) in eiders killed with lead shot than in drowned birds (Table 1). Although low, lead levels in drowned eiders are also elevated, by a factor of about three above the detection limit. We think the reason is that the breast meat is contaminated by lead from embedded lead shot (whole or fractions) hitting but not killing the bird earlier. Of the 25 drowned birds analyzed in this study, 11 birds carried between one and three pellets. Twenty four percent of the common eiders in Greenland are reported to carry embedded lead shot, referred to as "crippling" (Falk and Merkel, 2001).

We have tested if there is a relation between the lead concentration in the breast and the number of pellets found in the whole bird or the number of pellets in the breast analyzed. It could be suspected that the lead concentration would increase with the number of pellets. The results of linear regression analyses are summarized in Table 3, and the regression coefficient (positive) was only significant at the 5% level for eiders shot.

For eiders killed by lead shot there is no relationship between the age of the birds and the lead concentration in their breast, whereas there is a tendency of increasing lead concentration with increasing age in drowned eiders (Table 4). The reason could be that the oldest birds are those most exposed to crippling, because they have lived longest. Thus for eiders in Greenland, crippling increases with the age of the birds: 13% of eiders less than 1 year old carry embedded shot pellets, and the similar figure is 16% for 1–3 year olds and 30% for birds older than 3 years (Falk and Merkel, 2001).

3.3. Human lead exposure

The residue guideline for lead for bird's meat is 0.3 μg/g-wet wt in Denmark (Anon., 2002). In Canada no such guideline exists, but Scheuhammer et al. (1998) use a residue guideline for fish protein of 0.5 μg/g-wet wt. for evaluating levels in birds. Thirteen percent of our observations in murres exceed these residue guidelines, whereas in eiders this is the case for more than half of the birds analyzed in our study.

FAO/WHO (1993) have established a "Provisional Tolerable Weekly Intake" (PTWI) for lead of 25 μg per kg body weight for both adults and children. In general, lead levels in Greenland biota are very low, and consequently human lead intake from local food items is also estimated to be low, on average only 15 μg per adult per week (Johansen et al., 2000). However, this estimate does not include any significant contribution from shot

Table 4
Lead concentration (μg/g-wet wt) in whole breasts of common eiders, related to age and killing method

Age (years)	*n*	Mean	Standard error	Average pellet number and range
Drowned birds				
<1	1	0.095		1
1–2	4	0.059	0.007	0.25 (0–2)
>3	19	0.156	0.032	1 (0–3)
Shot birds				
<1	9	5.73	2.94	11.9 (2–42)
1–2	3	0.363	0.178	5.3 (2–11)
2–3	1	4.76		11
>3	12	7.85	5.03	10.5 (1–27)

Table 3
Linear regression analysis between number of pellets in whole bird or breast analyzed and lead concentration in breast. p: Probability of the null hypothesis (regression coefficient = 0) to be true

	Murre shot ($n=32$)		Eider shot ($n=25$)		Eider drowned ($n=24$)	
	Whole bird	Breast	Whole bird	Breast	Whole bird	Breast
Range of pellets found	0–12	0–5	1–42	0–3	0–3	0–1
Coefficient of determination (R^2)	0.04	0.04	0.29	0.20	0.0002	0.06
Regression coefficient	0.08	0.19	0.14	1.25	0.01	1.06
p	0.30	0.29	0.005	0.02	0.94	0.26

pellets, and the use of lead shot in Greenland may increase human lead intake considerably.

To illustrate the importance of birds as a dietary lead source we have calculated the lead intake from our analysis of whole breasts. If, as an example, we consider a meal being 200-gram of bird meat, the resulting mean lead intake from such a meal will be 146 μg for murre and 1220 μg for eider. This implies that only one eider meal in average will result in a lead intake close to the PTWI. For murre the lead intake from one meal in average will be about 10% of the PTWI.

The actual lead intake over longer periods from birds hunted with lead shot will depend on (1) the amount of bird meat per meal, (2) the frequency of bird meals and (3) the lead concentration in the meals. There are not sufficient data to quantify this, but our calculation in the example illustrates that birds hunted with lead shot are a significant lead source and probably the most important single source. The example also indicates that the lead intake must be expected to exceed the PTWI in a number of cases, as both murre and eider are important in the diet, particularly in Southwest Greenland during winter.

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