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Source identification of human exposure to lead in nine Cree Nations from Quebec, Canada (*Eeyou Istchee* territory)

Eric N. Liberda[a,*], Leonard J.S. Tsuji[b], Ian D. Martin[b], Pierre Ayotte[c], Elizabeth Robinson[d], Eric Dewailly[c,1], Evert Nieboer[e]

[a] School of Occupational and Public Health, Ryerson University, 350 Victoria St, Toronto, Ontario M5B 2K3, Canada
[b] Health Studies, and the Department of Physical and Environmental Sciences, University of Toronto Scarborough, Toronto, Ontario, Canada
[c] Axe Santé des populations et pratiques optimales en santé, Centre de recherche du CHU de Québec - Université Laval, Québec, Québec, Canada
[d] Public Health Department of the James Bay Cree Territory, Quebec, Canada
[e] Department of Biochemistry and Biomedical Sciences, McMaster University, Hamilton, Ontario, Canada

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ABSTRACT

While policies to decrease lead in the environment have been implemented to reduce human exposure to various lead sources, the use of lead ammunition to harvest wild game may continue to contribute significantly to human blood lead levels (BLLs). As part of a multi-community environment-and-health study, BLLs representing all age groups were evaluated in the nine Cree Nations located in the James and Hudson Bay regions of Quebec, Canada. Personal, market food, traditional food and 24-h recall questionnaires were administered. Predictor variables were assessed for various exposure sources, including diet and hunting practices. Elevated BLLs were observed in association with increased hunting status, use of firearms and leaded ammunition, and consumption of traditional foods. Significant differences were observed between all communities, age groups and sexes. Recommendations include educational campaigns that promote switching to non-lead ammunition and, if lead ammunition continues to be used, careful removal from tissues of pellets, bullet fragments and ammunition paths.

1. Introduction

Lead is a naturally occurring non-essential metal found in soil, water and air, and is a ubiquitous environmental contaminant found in food and drinking water. It poses a health risk to both humans and wildlife alike. In addition to its deleterious neurobehavioural effects, this contaminant adversely affects the renal and reproductive systems, blood formation, the cardiovascular system (via the autonomic nervous system), as well as other organ systems (Health Canada, 2013; Green and Pain, 2012; Peters et al., 2012; Bradman et al., 2001; Srianujata, 1998; Loghman-Adham, 1997; World Health Organization WHO, 1995; Landrigan and Todd, 1994; Hammond and Dietrich, 1990; Grandjean, 1978, 1993). The primary sources of exposure for the general population have been identified as drinking water because of old lead plumbing/service lines, lead-based paint and associated indoor/outdoor dust, contaminated soils in playing areas and gardens, and dietary intake (Nieboer et al., 2013a). Although the significant reduction of lead use over the last several decades has resulted in diminished human body burdens, the acceptable levels in whole blood have also been reduced (Nieboer et al., 2013a). Nevertheless, some individuals may still be exposed to environmental lead such as among populations who live a subsistence lifestyle, particularly indigenous peoples (Iqbal et al., 2009; Johansen et al., 2006; Tsuji and Nieboer, 1997; Tsuji et al., 1999).

Lead ammunition is found in two forms. Lead pellets are used in shotgun shells (shotshells) primarily for hunting birds and small game; each shell can contain more than 100 projectile pellets, with the actual number dependent upon pellet size and intended use. Lead bullets are single projectiles typically fired from a rifle and are used for hunting large and small mammals. Consumers of hunted birds and mammals are exposed to lead through ingestion of whole lead pellets from shotshells, fragments of either pellets or bullets, and lead contaminated muscle tissue around the wounds from these projectiles (Johansen et al., 2001; Hunt et al., 2009; Taggart et al., 2011). Ingestion of lead is more commonly associated with shotshell pellets than with rifle bullets, though the latter will fragment into smaller particles in some cases (Fachehoun et al., 2015). An additional inhalation exposure route of lead may exist for hunters if the ammunition uses lead-based primers or

* Corresponding author.
E-mail address: eric.liberda@ryerson.ca (E.N. Liberda).
[1] Deceased.

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detonators (e.g., lead styphnate; Tsuji et al., 1999).

While lead shotshell has been banned nationwide in Canada since September 1999, this ban only applies to migratory birds, and hence lead shotshell can still be purchased for the hunting of upland game birds and small mammals (Tsuji et al., 2008a). This is important because our group has demonstrated by comparing stable lead-isotope ratios in blood with those of the lead pellets used in subsistence hunting that gun use was a major source of lead exposure for First Nation Cree people of the James Bay region of Ontario, Canada (Tsuji et al., 2008b,c). More specifically, 88.9% of the First Nation subjects had an isotopic ratio in blood that reflected the ammunition profile, compared to 10% of the Southern Ontario control population. In the northern Ontario Cree study, we also obtained radiographic evidence of lead pellet abdominal entrapment, as well as in bagged game-bird tissues (Tsuji and Nieboer, 1997; Tsuji et al., 1999). Chan et al. (2011) have remarked that consumption of game meat contaminated with lead may be associated with an increase in human body burden and estimated a mean daily intake of 0.23 μg/kg body weight in British Columbia on-reserve indigenous community residents. Comparatively, the Total Diet Study estimated a dietary intake of lead among non-Indigenous populations (all ages) of 0.1 μg/kg body weight (Health Canada, 2013). Clearly, Indigenous peoples are at an increased exposure to lead when compared to non-Indigenous populations. Indeed some health concerns may remain due to the lack of a threshold response for developmental effects (Binns et al., 2007; Centers for Disease and Prevention, 2005; Federal-Provincial Advisory Committee on et al., 1994). In Canada, a tiered BLL action guideline starting at 0.48 μmol/L (10 μg/dL) was established in 1994 (Health Canada, 2013). However, the Centers for Disease Control and Prevention's (CDC) Advisory Committee for Childhood Lead Poisoning Prevention (ACCLPP) reduced the previous BLL "level of concern" of 10 μg/dL to 5 μg/dL (0.24 μmol/L); the latter constitutes a new reference level at which public health interventions for children (and thus also for women of reproductive age) should be initiated (Centers for Disease Control and Prevention CDC, 2012; for a review of the evidence for adverse effects in children at blood lead levels below 10 μg/dL see Centers for Disease Control and Prevention CDC, 2005). In the Province of Quebec, laboratories were required to report blood lead concentrations above 0.5 μmol/L to public health authorities, but this threshold has recently been reduced to 0.25 μmol/L for children under age 11 (Gouvernement du Québec, 2016).

As part of the multi-community environment-and-health study in *Eeyou Istchee*, northern Quebec, Canada, our research team investigated the human body burdens of lead in nine Cree Nation communities and examined possible sources of exposure. Subsistence hunting occurs frequently in these communities and residents may be at an elevated risk for lead exposure when consuming lead-contaminated hunted game birds and using lead ammunition. Lead shot is still being used even though the Migratory Birds Regulations (C.R.C., c. 1035, Section 15.1) requires the use of non-toxic alternatives (e.g., bismuth and steel shot) to hunt migratory birds with a few exceptions: *Zenaida macroura* (Mourning Dove), *Patagioenas fasciata* (Band-tailed Pigeon) and *Scolopax minor* (American Woodcock) (Government of Canada, 2016). In addition, lead shot is not illegal for ptarmigan and grouse and non-toxic alternatives are not easily available. In this context, we have investigated the contribution to BLL of various routes of lead exposure, including diet (store bought and traditional foods), smoking, personal characteristics (age and gender) and hunting practices (ammunition type and usage).

2. Methods

2.1. Study sites, participant recruitment, and sample collection

The multi-community environment-and-health study database includes participants from all nine Cree First Nation communities located in the James and Hudson Bay region of Quebec, Canada (known as *Eeyou Istchee*; for map and location of communities A-I, see Liberda et al., 2014). The homes and community facilities in the Cree communities are relatively modern and were mostly built during the 1975–1995 period. Participants from these communities were recruited by random sampling without replacement, except for Community A which was purposefully oversampled at the request of the locally-elected administration. Participants were stratified according to age groups (8–14 y, 15–39 y, and 40 y and over), by sex, and by community of residence. Of the 1776 total participants, data for 1429 persons aged 8 and older (809 female, 620 male) were used for analysis with the remaining 347 participants being removed due to lack of participation in certain segments of the study. The number of participant observations per analysis was dependent on compliance with different aspects of the study. The < 8 y old age group were necessarily excluded from many aspects of this article except BLL exceedances as their participation was limited to the donation of blood.

Whole blood samples (16 ml) were drawn into EDTA (ethylenediamine-*N,N,N′,N′*-tetraacetate) coated Vacutainer tubes (BD Medical, NJ, USA) and were frozen and shipped to the Toxicology Laboratory at the Institut National de Santé Publique du Québec (INSPQ); it serves as the Arctic Monitoring and Assessment Program (AMAP) reference laboratory. Ethics approval was granted by the research ethics committees of Université Laval, McGill University, McMaster University and the Cree Board of Health and Social Services of James Bay (CHBSSJB). Written informed consent was obtained from all participants or their guardians. Six questionnaires were administered, namely individual, clinical, market food frequency, 24-h recall, traditional food frequency and zoonoses, which are available online (Nieboer et al., 2013b). Cree speaking (*Eeyou Ayimuwin*) translators assisted participants who did not speak English or French.

2.2. Laboratory analysis

The analytical methods employed for the determination of lead in whole blood have been described previously (Nieboer et al., 2011; Bonnier-Viger et al., 2007). Briefly, lead content was analyzed from thawed whole blood samples using inductively coupled plasma mass spectrometry as part of a suite of contaminants (Perkin Elmer Sciex Elan 6000 ICP-MS). Blood samples were diluted in ammonium hydroxide and converted to their elementary form by aspirating the sample into an argon plasma followed by mass spectrometry identification. The detection limit for lead was 0.001 μmol/L.

2.3. Statistical methods

2.3.1. Characterization of BLL

Blood lead values were transformed as log10(x +1) to aid in normalizing the distribution. These values were analyzed by 3-way ANOVA (Community X Sex X Age-group). The ANOVA was bootstrapped (n = 1000) to minimize effects of outlier values and to provide greater confidence in estimated test statistic p-values. Estimated marginal means (EMMs) were used to characterize mean BLL values adjusted for effects of interactions and variable group size; these were compared between levels of the group factors at Bonferroni-adjusted p-values of 0.05 to test multiple comparisons between various communities, age-groups, and between males and females.

2.3.2. Hunting and use of lead ammunition

Participants also provided answers to survey questions regarding hunting practices and tobacco smoking. We evaluated these categorical responses and assessed associations (ANOVA) with participant-matched BLLs. Sample sizes varied slightly from one analysis to another as not all participants answered every question. To quantify the BLL effects of exposure attributable to various hunting practices, we used bootstrapped 2 × 2 contingency table analysis to calculate the relative risk (RR) of exceeding 0.24 μmol/L BLL for individuals who hunted or did