Environment International 71 (2014) 63–73

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Identification of environmental sources of lead exposure in Nunavut (Canada) using stable isotope analyses

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ARTICLE INFO

Article history:
Received 11 February 2014
Accepted 3 June 2014
Available online 25 June 2014

Keywords:
Lead
Blood
Environment
Dust
Exposure
Inuit

ABSTRACT

Background: Blood lead levels (BLLs) were measured in the adult Inuit population of Nunavut, Northern Canada, during the Inuit Health Survey (IHS) in 2007–2008. Approximately 10% of the adult participants had BLL over the Health Canada's guidance of 100 μg/L.
Objectives: 1) To repeat the measurement of BLL among the IHS participants with high BLL and household members including pregnant women and children under 10 years of age; 2) to measure lead (Pb) concentrations in environmental samples to identify potential sources and 3) to explore how Pb from environmental samples contributes to BLL using Pb stable isotopic analyses.
Methods: Blood samples were collected from 100 adults and 56 children in 2012. A total of 169 environmental samples (tap water, house dust, paint, country food, soil, and ammunition) were collected from 14 houses from three communities where the IHS participants had the highest BLL. Total Pb concentrations and Pb isotope mass balance were determined by inductively coupled plasma-mass spectrometry (ICP-MS).
Results: The geometric mean of BLL was 43.1 μg/L; BLL increased with age and was higher in adults than children (71.1 vs. 17.5 μg/L). Median Pb concentrations in water (1.9 μg/L) and dust (27.1 μg/m^2 for wiped dust, 32.6 mg/kg for vacuum dust coarse fraction, and 141.9 mg/kg for vacuum dust fine fraction) were generally higher than in other parts of Canada. Median Pb concentrations of food and soil coarse and fine fractions were low (36.6 μg/kg, 5.4 mg/kg and 11.8 mg/kg respectively); paint chips exceeded the Canadian guidelines in two houses (median: 3.8 mg/kg). Discriminant analyses and isotope ratio analyses showed that ammunition and house dust are major sources of Pb in this study population.
Conclusion: Analyses of Pb stable isotopes are useful to identify the routes of exposure to Pb. This approach can contribute to develop targeted public health programmes to prevent Pb exposure.

1. Introduction

Over the past decades, there has been a general decline in blood lead levels (BLLs) in the population of the countries that have implemented a phasing out of lead (Pb) from gasoline (Lanphear et al., 2003; Pirkle et al., 1994); in addition, the reduction of Pb content in Pb-based paints and the elimination of Pb solder in food cans also contributed to the decline of BLL (Health Canada, 2013a). However, human exposure to Pb is still a public health concern, as some segments of the population are still more exposed than others (Brown and Longoria, 2010; Couture et al., 2012; Meyer et al., 2003). This suggests that a variety of Pb sources are still present in the environment, contributing to an increased exposure of the groups in contact with these sources. Identifying the environmental sources contributing to blood Pb can contribute to the development of mitigation measures to further reduce human exposure to Pb. Stable isotopes of Pb have increasingly been used to assess the environmental sources of Pb contributing to human exposure (Chen et al., 2012; Oulhote et al., 2011; Tsuji et al., 2008).

Pb occurs naturally in the environment, but anthropogenic activities have been contributing significantly to the increased Pb levels in some environments (United Nations Environment Programme., International Labour Organisation. et al., 1995). Mining of Pb and other metals, industrial processes using Pb, and combustion of coal, oil, or waste all contribute to the release of Pb in the environment (WHO, 1989). The past use of Pb in gasoline has been a major contributor to Pb in air and soils, and higher Pb levels are found in soils near roadways. In urban settings, studies have shown that sources of Pb in dust and soil include previous automotive exhaust emitted when gasoline contained Pb, as well as weathering and chipping of Pb-based paint from buildings, bridges, and other structures (United

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http://dx.doi.org/10.1016/j.envint.2014.06.004
0160-4120/

Nations Environment Programme, International Labour Organisation et al., 1995). People living in areas where old houses have been painted with Pb paint may be exposed to higher Pb levels in dust and soil (Rabinowitz et al., 1985). In the aviation sector, Pb is still added to fuel for piston-engine aircraft (in general aviation or air taxi aircrafts) in the form of tetraethyl Pb (EPA, 2010). Pb is also found in ammunition, particularly for Pb pellets in gunshots. Food items, both from the market or from subsistence activities, usually contain low levels of Pb (WHO, 2003), but wild game shot using Pb ammunitions have been reported to have high Pb concentrations, especially after cooking (Mateo et al., 2007).

Humans can be exposed to Pb through a variety of pathways. In Canada, since the phasing out of Pb from gasoline, respiratory exposure from inhalation of Pb has significantly declined, and now exposure occurs mainly orally through water and diet (Health Canada, 2013a). Drinking water in houses containing leaded plumbing may contain Pb and the association between BLL and tap water Pb concentrations has been documented in Canada, despite the generally low Pb levels in municipal water supplies (Levallois et al., 2013). All food items from the diet can also represent a source of Pb for consumers, although Pb levels found in foods are generally low (Health Canada, 2013a). Food preparation can also represent a source of Pb, depending on how food is transformed, stored or prepared (Barbosa et al., 2009; Hernandez-Serrato et al., 2006). Human exposure to Pb can also occur from the ingestion of soil and dust that contain Pb (Bell et al., 2013), which is higher in children than in adults as a result of their hand-to-mouth behaviour (Lanphear and Roghmann, 1997). In areas where soils are heavily loaded with Pb, soil can be a primary source of Pb in both internal floor dust and hand-wipe dust (Zahran et al., 2011).

Once in the bloodstream, Pb accumulates in the bones, where it can be sequestered for a long time, contributing to the body burden; as humans age, and during certain health events, decalcification progresses, releasing Pb into the bloodstream and therefore increasing BLL (Health Canada, 2013a).

The main target for Pb toxicity is the nervous system, both in adults and children. Past occupational exposure to Pb of adults, assessed by Pb in the bones, has been associated with a longitudinal decline in cognitive function (Schwartz et al., 2000). Children are more sensitive than adults to the health effects of Pb (Bellinger, 2004; Landrigan and Todd, 1994). In children, pre-clinical neurotoxicity of Pb includes cognitive deficits (measured by IQ), reduction of attention and memory, alterations of a large spectrum of behaviours, fatigue and persistent brain lesions (Bellinger et al., 1994; Hu et al., 2006; Landrigan et al., 1993; Lanphear et al., 2005; Stewart et al., 2006). At maternal BLL of 100 to 150 μg/L, in-utero exposure to Pb has been associated with impaired mental development, such as decreased IQ scores (ATSDR and A. f. T. S. a. D. R., 2007; U.S.EPA, 2006). To date, no toxicity threshold has been identified and there is sufficient evidence that BLLs below 50 μg/L, and as low as 10–20 μg/L, are associated with adverse health effects (Bellinger, 2004; Gilbert and Weiss, 2006; Lanphear et al., 2005; Tellez-Rojo et al., 2006). Based on the 97.5th percentile of the population BLL in children ages 1–5, the United States Centers for Disease Control (CDC) recently lowered the reference value for BLL of children to 50 μg/L (CDC, 2012).

BLLs in the Canadian Inuit population have been documented since the mid-1980s. A recent review of the environmental contaminants and health in the western Canadian Arctic showed that Pb levels in Inuvialuit and Dene/Métis mothers were almost two times the levels of non-Aboriginal mothers (13 vs. 6.9 μg/L) from the Inuvik region (Donaldson et al., 2010). During the International Polar Year Inuit Health Survey (IHS) in 2007–2008, elevated BLLs were reported in the adult Inuit participants from Nunavut ($N = 1628$). The average (geometric mean) BLL was 37.1 μg/L (unpublished data), which is higher than the levels reported in the Canadian Health Measures Survey Cycle 2, which was 12 μg/L (Health Canada, 2013b). Almost 10% (155 participants) had BLL exceeding the Canadian blood Pb intervention level for general unexposed population of 100 μg/L, which is currently under review; among these, three women of childbearing age had BLL higher than 100 μg/L. The mean BLL significantly increased with age and male participants had significantly higher mean BLL than female participants. At that time, no data were available regarding children's BLL in Nunavut or possible sources specific to this region (Laird et al., 2013).

The objective of this paper is to describe the BLL in the participants of this follow-up study, which included past IHS participants with elevated BLL (EBLL) and any household members who were pregnant or children under the age of 10, and to explore the potential environmental sources of exposure in the Inuit population of Nunavut, northern Canada, using Pb stable isotope analyses.

2. Materials and methods

2.1. Study design and sampling

Recruitment was organized by the Government of Nunavut Department of Health, who prepared invitation letters for the IHS participants who presented BLL of 100 μg/L or higher in 2007–2008. These letters were sent to all local Health Centres in Nunavut (Fig. 1), where a nurse personally met these individuals to present them the follow-up project and the letter of invitation.

Children 10 years of age and younger as well as pregnant women from the identified participants' houses were also invited to take part in the follow-up. All adult participants signed a consent form; for the children, the parent or guardian signed an assent form. The letters of invitation and the consent/assent forms were available in both English and Inuktitut, and translation to Inuktitut was provided during the interviews when necessary. This project obtained the ethics approval from the Office of Research Ethics and Integrity from the University of Ottawa.

2.2. Data collection

2.2.1. Human exposure questionnaires

For every adult participant, a nurse from the local Health Centre administered an Exposure Questionnaire; for children, the parent or guardian provided the answers. The questionnaire included information regarding the participants' dietary habits and potential occupational and/or residential sources of exposure to Pb.

2.2.2. Environmental samples information

Environmental samples were collected in 14 houses from three communities of Nunavut, where the participants had the higher BLL during the 2007–2008 IHS. Environmental Health Officers (EHO) visited the communities, where they sampled up to five households from the list of participants in the blood-sampling programme. Environmental samples collected included tap water, house dust, house paint, country food, and soil. For each house visited, the EHOs completed a Visit Record Sheet to collect information on the house (e.g. conditions, history, construction, renovations), samples collected, types of ammunition used by the participants, and soil sampling locations.

2.3. Sample collection, preparation, and digestion

2.3.1. Blood

For each participant, a nurse from the local Health Centre collected one 6 mL tube of blood by venipuncture. Samples were kept refrigerated and sent to the University of Ottawa for Pb concentration and Pb isotope mass balance analyses. Blood samples were kept at 4 °C until analysis. For each blood sample, a volume of 0.5 mL was diluted to 45 mL with Milli-Q H_2O. Diluted samples were mixed rigorously and centrifuged at 4 °C for 5 min at 3000 rpm, before being transferred to 15 mL centrifuge tubes in a clean room.