Environmental Research 146 (2016) 331–339

Contents lists available at ScienceDirect

Environmental Research

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

Multimedia exposures to arsenic and lead for children near an inactive mine tailings and smelter site☆

Miranda M. Loh[a,1,*], Anastasia Sugeng[a], Nathan Lothrop[a], Walter Klimecki[b], Melissa Cox[c], Sarah T. Wilkinson[d], Zhenqiang Lu[e], Paloma I. Beamer[a]

[a] Department of Community, Environment and Policy, Mel and Enid Zuckerman College of Public Health, University of Arizona, 1295 N. Martin Avenue, P.O. Box 245163, Tucson, AZ 85718, USA

[b] Department of Pharmacology and Toxicology, College of Pharmacy, The University of Arizona, P.O. Box 210207, Tucson, AZ 85724, USA

[c] Hospital Medicine and Outreach, Department of Pediatrics, Diamond Children's Medical Center, The University of Arizona, 1501 N. Campbell Ave. Tucson, AZ 85724, USA

[d] Superfund Research Program, The University of Arizona, 1110 E. South Campus Dr., Tucson, AZ 85721, USA

[e] BIO5 Institute, The University of Arizona, 1657 E. Mabel St., Tucson, AZ 85721, USA

ARTICLE INFO

Article history:
Received 26 September 2015
Received in revised form
6 December 2015
Accepted 10 December 2015

Keywords:
Metals
Exposure assessment
Multimedia
Biomarkers
Hazardous waste

ABSTRACT

Children living near contaminated mining waste areas may have high exposures to metals from the environment. This study investigates whether exposure to arsenic and lead is higher in children in a community near a legacy mine and smelter site in Arizona compared to children in other parts of the United States and the relationship of that exposure to the site. Arsenic and lead were measured in residential soil, house dust, tap water, urine, and toenail samples from 70 children in 34 households up to 7 miles from the site. Soil and house dust were sieved, digested, and analyzed via ICP-MS. Tap water and urine were analyzed without digestion, while toenails were washed, digested and analyzed. Blood lead was analyzed by an independent, certified laboratory. Spearman correlation coefficients were calculated between each environmental media and urine and toenails for arsenic and lead. Geometric mean arsenic (standard deviation) concentrations for each matrix were: 22.1 (2.59) ppm and 12.4 (2.27) ppm for soil and house dust (<63 μm), 5.71 (6.55) ppb for tap water, 14.0 (2.01) μg/L for specific gravity-corrected total urinary arsenic, 0.543 (3.22) ppm for toenails. Soil and vacuumed dust lead concentrations were 16.9 (2.03) ppm and 21.6 (1.90) ppm. The majority of blood lead levels were below the limit of quantification. Arsenic and lead concentrations in soil and house dust decreased with distance from the site. Concentrations in soil, house dust, tap water, along with floor dust loading were significantly associated with toenail and urinary arsenic but not lead. Mixed models showed that soil and tap water best predicted urinary arsenic. In our study, despite being present in mine tailings at similar levels, internal lead exposure was not high, but arsenic exposure was of concern, particularly from soil and tap water. Naturally occurring sources may be an additional important contributor to exposures in certain legacy mining areas.

1. Introduction

Mining and associated industries, such as smelting, generate waste materials that can contaminate the surrounding environment and be a source of exposure for nearby communities even after the facilities close. Exposure to various metal(loid)s from mining waste can occur via inhalation and ingestion of windblown soil and dust, ingestion of contaminated drinking water, and even ingestion of foods grown in personal gardens with contaminated soil.

Adverse health effects of exposure to metal(loid)s include cancer, neurodevelopmental impairment, and other systemic effects. Children are particularly susceptible to the effects of these

☆This work was supported by the University of Arizona Superfund Research Program (NIEHS P42 ES04940), the Southwest Environmental Health Sciences Center (NIEHS P30 ES006694). P. Beamer was supported by NHLBI (K25 HL103970). This publication's contents are solely the responsibility of the authors and do not necessarily represent the official views of the National Institutes of Health. This project was approved by the Human Subjects Protection Program of the University of Arizona.

* Corresponding author.
E-mail addresses: mloh@email.arizona.edu, miranda.loh@iom-world.org (M.M. Loh).

[1] Present address: Institute of Occupational Medicine, Research Avenue North, Riccarton, Edinburgh EH14 4AP, United Kingdom.

http://dx.doi.org/10.1016/j.envres.2015.12.011
0013-9351/© 2016 Published by Elsevier Inc.

contaminants because their bodies are still developing (Hines et al., 2010). Lead has been associated with a decrease in IQ and various chronic diseases (Bellinger et al., 1986; Lanphear, 2005; Lanphear et al., 2005; Spanier and Lanphear, 2005). Arsenic exposure at an early age may also lead to disease later in life (Dauphine et al., 2011). Arsenic is a known carcinogen which has also been associated with decreased lung function and increased susceptibility to respiratory infections and cardiovascular effects (Ahsan et al., 2006, 2000; Bates et al., 2004; Chen et al., 1995; Dauphine et al., 2011; Navas-Acien et al., 2005; Lantz et al., 2009).

Given the potentially higher concentrations of metal(loid)s in soils and dust near mining and smelting sites, it is important to understand the various sources and pathways of exposure in order to better target exposure reduction (Gulson et al., 1994a, 1994b). In particular, children are at greater risk of exposures as they are more likely to play outdoors or on the floor and inadvertently ingest dust adhered to hands or other objects (Cohen Hubal et al., 2000). For example, in the community near a former copper smelter in Montana, Hwang et al. (1997) found a significant correlation between arsenic concentrations in residential soils and urinary arsenic in children less than 72 months old, with the highest correlation for soils in bare yards. Polissar et al. (1990) found that indoor air and dust were associated with urinary arsenic in a community nearest a former copper smelter in Tacoma, Washington. They found a stronger relationship between soil arsenic and urinary arsenic in children compared to adults, indicating that soil ingestion may be a source of exposure in children, but not adults.

In Arizona, where copper mining has a long history, the former Iron King Mine and Humboldt Smelter site (Iron King) are now listed on the National Priorities List (a.k.a., a Superfund site). The Superfund program was established by the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA), which allows for the cleanup of listed sites by the United States Environmental Protection Agency (US EPA). Arsenic and lead concentrations in the mine tailings and smelter ash are well above 1000 ppm (US EPA, 2010), and arsenic and lead levels in soil near the mine could be impacted by windblown dust or rainwater runoff.

Arsenic, in particular, is known to be present in copper ores, and in Arizona, some groundwater wells are contaminated with naturally occurring arsenic as a result of the regional geology. Much of the drinking water in rural Arizona comes from private wells, providing an additional exposure source to arsenic, in addition to the soil and dust from the Iron King site. Gardenroots, a previous study looking at homegrown vegetables in the same community found arsenic in home irrigation water at levels ranging from 1.40 to 2030 ppb, and residential soil samples ranging from 3.07 to 322 ppm (Ramirez-Andreotta et al., 2013).

In response to the high levels of arsenic and lead at the Iron King site, the findings of the Gardenroots project, community interest, and the importance of early life exposures to the potential development of subsequent disease, we conducted the Metals Exposure Study in Homes (MESH) to examine the exposures of children in the community surrounding the Site. The aim of the study was to quantify the environmental levels and exposures of children and determine whether exposures were elevated compared to those of the general United States (US) population. Due to the high levels of arsenic and lead in the tailings, we hypothesized that levels of arsenic and lead in soil and house dust and exposures (as measured in urine and toenails) would be higher found in areas closer to the Iron King site than further away. We also include water ingestion as a pathway of exposure due the potential for groundwater wells in this area of Arizona to be high in arsenic (O'Rourke et al., 1999a, 1999b; Roberge et al., 2007).

2. Materials and methods

2.1. Study area and participants

We recruited households with children aged 1–11 years within a 5-mile radius of the geographic median between the Iron King Mine and Humboldt Smelter (Fig. 1). Recruitment methods included having a presence at local fairs, mailing flyers and postcards, and door-to-door canvassing from October 2011 through June 2013. A crew of local field technicians carried out the door-to-door recruitments and subsequent home visits. At least one child per household was enrolled in the study.

Upon enrollment, two home visits were scheduled at the participant's convenience, approximately one to two weeks apart. The first visit included a home walk-through and a questionnaire on the physical layout of the participant's home and property. Also during this visit, dust fall canisters were laid out, and participants were given biological sample collection materials and food and activity logs.

The parents or guardians were instructed to record food and activity/location information 4 days before the collection of a urine sample, which was to be the morning of the second visit by field staff. During the second visit, field staff collected environmental samples, and picked up the urine and toenails collected by the family and the activity and food logs.

2.2. Biological sample collection

Parents or guardians of participants were instructed to obtain urine samples the morning of the second home visit and to refrigerate them until the visit. Once transported to the field office, they were kept at −20 °C until analysis. The specific gravity of urine samples was determined using a refractometer (TS Meter Handheld Goldberg Series, Reichert Analytical Instruments). Samples were corrected for specific gravity before data analysis using the overall group mean according to Eq. (1):

$$U_{sg} = U\times(\frac{SG_{mean}-1}{SG_{measured}-1}) \quad (1)$$

where U_{sg}=specific gravity-corrected urine concentration of element (μg/L); U=uncorrected urine concentration of element (μg/L); SG_{mean}=the mean specific gravity of all individuals in the study group (1.02); and $SG_{measured}$=the specific gravity of each individual's urine sample. For summary statistics and data analyses, the specific gravity corrected urine concentrations were used.

All toenails were clipped with new nail clippers unique to each child and were collected in brown manila envelopes and kept at room temperature until processed. Toenails were sonicated in acetone for 20 min; rinsed with ultrapure water five times; and then sonicated in 1% Triton-X for an additional 20 min. They were then rinsed again with ultrapure water and dried in an oven at 60 °C for 24 h before being weighed. Toenails were then placed for 12 h in 2 mL of Optima® trace metal free nitric acid and microwave digested using a CEM MARS Xpress, set at 400 W at 75% power, with a 10 min ramp up to 105 °C, and held at that temperature for 15 min. The sample was allowed to cool before being transferred to trace metal-free centrifuge tubes and diluted to 10 mL before laboratory analysis. A certified reference material (INSPQ/Toxicologie QMEQAS10H-02, cheveux/hair) was used for quality control. For venous blood lead testing, participants went to a local Clinical Laboratory Improvements Amendments (CLIA) certified laboratory, Labcorp, Inc. (Burlington, NC), where samples were analyzed by ICP-MS. The blood lead LOD reported was 1 μg/dL.