Environmental Exposure and Fingernail Analysis of Arsenic and Mercury in Children and Adults in a Nicaraguan Gold Mining Community

JOEL B. WICKRE Department of Biological Sciences Dartmouth College Hanover, New Hampshire CAROL L. FOLT **Department of Biological Sciences** and **Center for Environmental Health Sciences Dartmouth College** Hanover, New Hampshire STEFAN STURUP **Center for Environmental Health Sciences** and **Department of Earth Sciences Dartmouth College** Hanover, New Hampshire

MARGARET R. KARAGAS Center for Environmental Health Sciences Dartmouth College Hanover, New Hampshire and Section of Biostatistics and Epidemiology Department of Community and Family Medicine Dartmouth Medical School Lebanon, New Hampshire

ABSTRACT. Gold mining can release contaminants, including mercury, into the environment, and may increase exposure to naturally occurring elements such as arsenic. The authors investigated environmental and human tissue concentrations of arsenic and mercury in the gold mining town of Siuna, Nicaragua. The study involved 49 randomly selected households in Siuna, from whom a questionnaire along with environmental and fingernail samples were collected. Environmental samples indicated that mercury concentrations in drinking water, although generally low, were higher near the mine site. Arsenic concentrations were elevated in water and soil samples, but their distribution was unrelated to the mining site. Mercury concentrations in fingernail samples were correlated with residential proximity to the mine, drinking water concentrations, occupation, and, among children, with soil concentrations. Fingernail arsenic concentrations correlated with drinking water concentrations among adults who consumed higher levels, and with soil concentrations among children. Fingernail analysis helped to identify differential exposure pathways in children and adults. Mercury and arsenic uptake via soil exposure in children warrants further consideration.

<Key words: arsenic, artisanal mining, biomarker, children, environmental exposure, fingernail, gold mining, mercury>

GOLD MINING can severely damage the environment and may pose a hazard to human health, both by introducing contaminants from the mining process and by enhancing concentrations of naturally present minerals. Traditional industrial methods for processing gold ore use mercury to separate gold from crushed rock, with consequent release of mercury into the surrounding air, tailings, soil, sediment, and water.¹ In developing countries, artisanal miners extract gold by a process that involves burning off elemental mercury to isolate gold from a gold-mercury amalgam. Once in the environment, mercury persists.² Among naturally present minerals, elevated arsenic concentrations are also found in surface water and sediment in gold mining watersheds,^{3,4} and have been found in surface soil as far as 13 km from gold mining sites.⁵

This investigation focused on 2 toxic metals of health concern-mercury and arsenic-in Siuna, a small town at the hub of "Las Minas," a gold mining area in remote north-Atlantic Nicaragua. Gold mining began in Siuna in the 1890s, was fully mechanized by the 1930s, and continued on an industrial scale until the mine closed in 1987. By that time, the hill at the center of Siuna had become a deep pit, which filled with groundwater to form an approximately 15-acre lake. From the 1930s to the 1980s, the mine produced a tailings heap more than 1 km long, 0.3 km wide, and approximately 10 m deep. Waste water containing mercury and cyanide was released into a stream along the tailings known as El Cianuro (The Cyanide), which joins with a river commonly used for washing and bathing. Since the closure of the mine, many unemployed miners have turned to artisanal mining, locally known as guiriseria. The owner of the mine concession commissioned an environmental impact study in the 1990s, but no data were publicly released regarding heavy metal concentrations in Siuna. To our knowledge, the extent of human exposure to potential mining contaminants has not been investigated in Siuna. Therefore, we conducted an environmental assessment of mercury and arsenic in drinking water and soil samples in relation to the mining site, and evaluated the extent of human uptake by examining levels of these metals in fingernail tissue as a biologic marker of internal dose of exposure. This approach has been used elsewhere to assess metal exposure in adults,^{6,7} but data for children are extremely limited. Therefore, we specifically examined the association between fingernail concentrations and potential sources of mercury and arsenic exposure according to age, as well as other factors.

Materials and Method

In May 2002, we surveyed 49 households from 4 study areas in Siuna, which we classified on the basis of distance from the mine. Specifically, the *tailings* study site included only families who lived in residences constructed directly on the tailings. The *close* site included the neighborhood Carlos Fonseca, which is on the opposite bank of El Cianuro from the tailings, and portions of the neighborhoods Luis Delgadillo and Sol de Libertad, which border the pit mine lake. Our medium site included homes in the neighborhoods Rigoberto Lopez and Pedro Joaquin Chamorro, which ranged in distance from 0.3-0.8 km south of the pit mine lake. Our far site included households in Pedro Joaquin Chamorro, Sector 1, and Olivero, which ranged from 1.3-2.5 km south of the pit mine lake. In each neighborhood, we performed a census of the number of homes and then determined a set interval (e.g., every 4th house) that provided us approximately 7 to 8 homes

per neighborhood. We obtained verbal informed consent from an adult member of the selected household to obtain a household water sample from the source used for drinking and cooking, a soil sample from the yard, fingernail clippings from each family member present, and a brief interview in accordance with a protocol approved by the Committee for the Protection of Human Subjects at Dartmouth College. Of the 51 households we approached, 49 (96%) agreed to participate in the study.

Environmental samples. Water samples from each household's drinking water storage vessels were collected using techniques designed by the U.S. Geological Survey for testing ambient metal concentrations in environmental water.8 Each sample was collected in a 125-ml I-CHEM certified trace-metal clean polyethylene bottle (Chase Scientific Glass, Inc., Rockwood, Tennessee) after a single rinse with sample water. Water samples were poured directly from the storage vessel into the sample container and preserved by acidifying with 0.5 ml of trace-metal clean concentrated nitric acid and 0.05 ml of trace-metal clean concentrated hydrochloric acid. Surface soil samples were collected from the yard of each home at least 2 m away from any potential point sources of contamination, such as painted surfaces. These samples were collected and stored for transport in resealable polyethylene bags.

In addition to household water samples, we collected samples from lakes, rivers, streams, and reservoirs commonly used for drinking, washing, bathing, and artisanal mining. We collected 12 samples from the stream El Cianuro, which collects runoff and sewage from the center of town and the tailings and-although not used for washing or bathing-is used for artisanal mining. In addition, we collected 2 water samples from the lake formed by the pit mine and 12 samples total from rivers, reservoirs, and public well-fed cisterns. We identified these supplemental water sources with the aid of a local engineer and geologist and included 2 of the 3 reservoirs that feed the antiguated town water system, major rivers used for drinking water (the Wani, Uli, and Yaoya), and another river used for washing and bathing (the Matis). Finally, 4 additional surface soil samples were collected from the tailings. All samples were geographically referenced using a Garmin GPS 12 MAP handheld global positioning system unit (Garmin International, Inc., Olathe, KS). These points were later overlain on a geo-referenced topographical map of the area using arcGIS 8.2 (ESRI; Redlands, CA).

Interview data and tissue collection. After informed consent was obtained, we administered a brief questionnaire to an adult member of each of the 49 selected households regarding water use, purification (i.e., with chlorine), and storage (e.g., type of vessel and whether it was covered). The questionnaire was designed for this study and administered by two teams, each consisting of one researcher from the U.S. and one university student from Siuna. In addition to the age and gender of each household member, we asked about the typical amount of fish consumed by the family per week, whether artisanal mining was practiced in the household, and other factors. We requested fingernail samples from each family member present at the time of our visit. The fingernail samples were obtained using a standard fingernail clipping device that we provided. The samples were collected and stored for transport in paper envelopes.

Laboratory analysis. All samples were prepared and analyzed at the Trace Element Analysis Core Facility at Dartmouth College, using either hydride-generation/cold vapor inductively coupled plasma-mass spectrometry (ICPMS) (Element 1, Finnigan MAT, GmbH, Bremen, Germany) or Octopole Reaction System ICPMS (Agilent 7500c, Agilent Technologies, Wilmington, DE), as described in the following paragraphs. Sample digestions were performed in closed Teflon[®] vessels using a Mars 5 microwave oven system (CEM Corp., Matthews, NC).

Elemental mercury formed on reaction with sodium-borohydride (NaBH₄) was separated from the sample matrix and led directly into the ICPMS torch for measurement of ²⁰⁰Hg and ²⁰²Hg isotopes. Calibration was performed with matrix-matched external calibration using 8 standards in the 0.0–0.500 µg/l range. Standards were bought from SCP Science, Champlain, NY. Results were all blank-subtracted and reported as the average between the 2 isotopes. The detection limit was 0.002 µg/l, and the measurement uncertainty was 5% residual standard deviation (*RSD*).

Water sample analysis. The water samples were analyzed undiluted, and calibrations were performed using an external standard curve (5 standards in the 0–10 µg/l range). Ga-71 and In-115 were used as internal standards. The following isotopes were measured using the instrument standard mode (nonpressurized octopole reaction cell): ⁶³Cu, ⁶⁵Cu, ⁶⁶Zn, ⁸²Se, ¹¹¹Cd, ¹¹²Cd, ¹¹⁴Cd, ¹²¹Sb, ¹²³Sb, ¹³⁸Ba, ²⁰⁶Pb, and ²⁰⁸Pb. A number of elements were analyzed with the octopole reaction cell pressurized with helium gas to remove polyatomic interferences by collision (e.g., ⁴⁰Ar³⁵Cl⁺ on ⁷⁵As⁺). The following isotopes were analyzed under these conditions: ⁵¹V, ⁵⁵Mn, ⁵⁹Co, ⁶⁰Ni, and ⁷⁵As.

For quality control, a blank solution and certified standard reference material (NIST SRM 1640, Trace Elements in Natural Water, National Institute of Standards and Technology, Gaithersburg, MD) were analyzed for every 10 samples. Good agreement between certified and measured element concentrations was found. The overall method uncertainty was 5–10% *RSD*. Detection limits were < 0.1 μ g/l for all elements except Zn (1.12) and Se (0.20).

Soil sample analysis. The soil samples were prepared for analysis in accordance with U.S. Environmental Protection Agency method 3051 for microwave-assisted digestion of soils,⁹ and analyzed using the method described previously for water analysis, except that an in-house soil reference material was used for quality control. Good agreement between expected and measured values was obtained.

Fingernail sample analysis. Fingernail samples were washed carefully, using procedures shown in previous studies to remove external contamination.¹⁰ First, samples were washed in 1% Triton X-100 solution in an ultrasonic bath for 20 min. (A single sample received with nail polish was rinsed briefly in acetone.) Samples were then rinsed 5 times with deionized water, agitating between each rinse. Each sample was digested with 1 ml of concentrated nitric acid, then diluted 20 times with ultrapure water. Calibration was performed as standard addition calibration; 1 in every 7 sample solutions was split into 4 subsolutions, to which were added 0, 1, 2, and 3 μ g/l each of arsenic and mercury. From these solutions, calibration curves were calculated and used for quantification. For quality control, a certified reference material (GBW 07601 Human Hair Powder, Geophysical and Geochemical Exploration, Langfang, China) was used. Good agreement between certified and measured values was obtained. The detection limit was 0.10 ng/g for mercury and 0.35 ng/g for arsenic in the 1-ml digested solution. This detection limit was multiplied by the dilution factor (1/sample weight) to obtain the detection limit for each sample.

Statistical analysis. We performed statistical tests using JMP version 4 statistical software (SAS Institute, Inc., Cary, NC). Log-transformed mercury and arsenic concentrations in water and soil approximated normal distributions. One-way analysis of variance (ANOVA) was used to detect significant differences across sites. The Tukey-Kramer honest significant difference (HSD) test, based on a significance level of 0.05, was used to determine differences among sites. Correlations between environmental concentrations and fingernail levels were performed assuming linear relationships. All *p* values presented herein were determined with 1-way ANOVA.

Results

Environmental samples. The mean level of mercury in household water was 0.017 µg/l (median = 0.010 µg/l; range = 0.002-0.074 µg/l). Household well-water concentrations of mercury varied according to distance from the mine (p = 0.0033; Fig. 1), with higher concentrations closer to the mine. Mercury levels in drinking water from homes on the tailings were significantly higher than levels in water from medium and far homes (Tukey-Kramer HSD, Fig. 1). Of the 12 samples collected along the length of El Cianuro, concentrations were highest around its confluence with a second stream that



passes directly through the tailings (Fig. 1), but none of the samples exceeded the World Health Organization (WHO) guideline of 1 μ g/l for mercury in drinking water.¹¹

The mean concentration of mercury in soil samples from around the houses was 0.49 μ g/g (median = 0.30 μ g/g; range = 0.09–2.60 μ g/g). Soil concentrations were higher in households closer to the mining site (mean [standard deviation] = 0.650 [0.533], 0.616 [0.805], 0.402 [0.382], and 0.396 [0.307] μ g/g for the tailings, close, medium, and far sites, respectively); however, the differences were not statistically significant (Tukey-Kramer HSD was not statistically significant for the tailings vs. far site; 1-way ANOVA for all sites, p =0.47). Mercury concentrations in the 4 soil samples taken from the surface of the tailings ranged from 0.15 to 0.19 $\mu g/g.$

The mean level of arsenic in drinking water was 7.12 μ g/l (median = 1.78 μ g/l, range = < 0.03–73.63 μ g/l). Ten (20%) of the 49 households had levels of arsenic in drinking/cooking water that exceeded 10 μ g/l, the WHO provisional guideline value¹¹ (Fig. 2), with arsenic concentrations highest at the far site. Overall, concentrations were not statistically significantly different across sites (p = 0.14).

Of the other water samples we collected, we found 1 public well that exceeded 10 μ g/l arsenic (14.03 μ g/l). This well was built by a local nongovernmental organization (NGO) with international funding and was used primarily for washing; it was one of the



deepest wells in the area (approximately 10 m). Additionally, 6 of 12 stream-water samples from El Cianuro exceeded 10 µg/l arsenic (Fig. 2), with a mean arsenic level of 10.48 µg/l (median = 9.88 µg/l, range = 6.94-17.44 µg/l). Arsenic concentrations in El Cianuro, like mercury concentrations, were highest around its confluence with a stream that passes directly through the tailings. Neither of the samples from the lake formed by the pit mine exceeded 10 µg/l arsenic (Fig. 2).

The mean level of arsenic in household soil was 55.9 μ g/g (median = 25.0 μ g/g, range = 6.5–1263.4 μ g/g). Arsenic concentrations in soil were higher at the medium site than at other sites (p = 0.0027). Levels of arsenic

in the 4 soil samples from the mine tailings ranged from 20.7 to 47.8 μ g/g.

Characteristics of study subjects. A total of 130 individuals participated from the 49 selected household (i.e., 2.7 individuals per household on average). Their characteristics are given in Table 1. Females comprised 54% of the study group. Participants ranged in age from 11 mo to 85 yr, with 27% under the age of 10 yr. Regular fish consumption was uncommon in the study population; no individuals reported eating more than 1 fish meal per week. Overall, 5% of participants were involved in artisanal mining, all of whom were over the age of 20 yr. Eighty-two percent of households used plastic storage vessels for water, and 88% kept them

covered; 39% reported chlorinating their drinking water.

Fingernail tissue concentrations. The mean level of mercury in subjects' fingernails was $0.254 \ \mu$ g/g (median = $0.164 \ \mu$ g/g, range = $0.007-2.724 \ \mu$ g/g). Fingernail levels of mercury did not differ by age (p = 0.46) or gender (p = 0.93). Artisanal miners had higher fingernail

Characteristic	п	Percentage
Gender		
Female	70	54
Male	60	46
Age (yr)		
0-9	35	27
10–19	28	22
20–39	38	29
40-85	29	22
Artisanal mining		
No	123	95
Yes	7	5
Fish consumption		
<1 meal/mo	70	54
≥1 meal/mo	52	40
Unknown	8	6
Chlorination of water		
No	79	61
Yes	51	39
Water storage vessel material		
Metal	12	9
Plastic	106	82
Other	12	9
Storage vessel kept covered		
No	16	12
Yes	114	88

levels of mercury than nonminers (p = 0.0058). Concentrations of mercury in fingernails did not vary by fish consumption; however, this activity was rare (i.e., no individual reported eating more than 1 fish meal per week). Fingernail levels of mercury were higher in individuals who chlorinated their water vs. those who did not, for reasons unknown (p = 0.0012), but did not differ by type of storage vessel (p = 0.31) or storage vessel coverage (p = 0.14).

Fingernail mercury levels were correlated positively with log-transformed mercury levels in drinking/cooking water (r = .32, p = 0.0003). The correlation was stronger in young children (< 10 yr of age; r = .54, p = 0.0009) than in older children and adults (≥ 10 yr of age; r = .29, p = 0.0041). Also, among young children, mercury levels in fingernails correlated with mercury in soil (r = .33, p = 0.053), but this was not the case for older children or adults (r = .033, p = 0.75). Fingernail mercury levels were significantly higher in residents of the tailings than of the close, medium, and far sites (Tukey-Kramer HSD) and increased with proximity to the mine (all subjects: p = 0.0001; young children: p = 0.0022; older children and adults: p = 0.0013; Fig. 3).

The mean level of arsenic in fingernails was 0.825 μ g/g (median = 0.511 μ g/g, range = 0.036–5.115 μ g/g), and was slightly higher in males than females (p = 0.072). Arsenic levels in fingernails were inversely related to age, and on average were highest in children younger than 10 yr (p < 0.0001; Fig. 3). Fingernail concentrations of arsenic were unrelated to artisanal mining (p = 0.79), fish consumption (p = 0.58), water chlorination (p = 0.76), storage vessel material (p = 0.66), and storage vessel coverage (p = 0.40).



Arsenic levels in fingernails correlated with arsenic concentrations in soil among young children (< 10 yr of age; r = .34, p = 0.049) but not among older children or adults (≥ 10 yr of age; r = .015, p = 0.88). There was a trend of decreasing fingernail arsenic concentrations with increasing distance from the mine, but the differences by site were not statistically significant (p =0.080; Fig. 3). This trend appeared stronger among young children (p = 0.11) than in older children and adults (p = 0.40). Arsenic levels in fingernails did not correlate with log-transformed arsenic levels in household water samples (young children: r = .16, p = 0.36; older children and adults: r = .048, p = 0.65) overall. However, among individuals with drinking water concentrations \geq 5 µg/l, there was a positive correlation between fingernail arsenic and log-transformed arsenic levels in household water among older children and adults (r = .43, p = 0.010), but not among young children (r = .14, p = 0.69).

Discussion

In our study of Siuna, Nicaragua, the mercury concentrations observed in water-and to a lesser extent soil-correlated with distance from the tailings site. Mercury concentration in fingernail tissue was also related to proximity to the mining site and to drinking water concentrations, as well as to soil concentrations among children. However, levels of mercury did not exceed the current WHO guideline of 1 µg/l for drinking water, and soil concentrations of mercury were generally in line with those found in virgin or cultivated soil (i.e., 0.02–0.625 ppm).¹² Arsenic concentrations, on the other hand, exceeded the current WHO guideline of 10 µg/l for drinking water in several water samples. Also, levels in soil around a number of households were found to exceed the normal range of 1 to 40 $\mu g/g^{13}$ Of particular interest, arsenic concentrations exceeded 10 µg/l in one of the town's deepest wells, which was built by an NGO for the community. We found that arsenic concentrations in soil and water were unrelated to distance from the mine, but did vary across sites. Fingernail concentrations of arsenic appeared higher closer to the mining site, although the difference was not statistically significant. In older children and adults, fingernail arsenic concentrations correlated with drinking water concentrations among those with water concentrations above 5 μ g/l, and with soil arsenic concentrations among younger children (< 10 yr of age).

Metals emitted from mining processes represent an important source of environmental contamination. Metal contaminants in tailings can disperse through soil, water, and river sediments several kilometers from the mining site, and perhaps farther via airborne travel of volatilized particles.^{14,15} Mercury, used in gold ore

extraction, biomagnifies in the food web and becomes methylated into more toxic forms.¹⁶ As such, potentially hazardous levels of mercury have been detected in higher order fish^{17,18} and in other edible animals and plants¹⁹ near gold mines.

Gold mine tailings often contain concentrated levels of elements that are naturally present in gold-containing rock, including arsenic, aluminum, antimony, cadmium, copper, chromium, iron, lead, manganese, selenium, and zinc.^{1,20} Elevated arsenic concentrations are also found in surface water and sediment in gold mining watersheds,^{3,4} including at appreciable distances from gold mining sites.⁵ Although our study focused on environmental levels and human uptake of mercury and arsenic, we did measure other elements, including lead, cadmium, manganese, antimony, barium, copper, selenium, and zinc in water and soil samples. We did not detect elevated levels of these elements in any of the household drinking water samples we tested (data not shown). However, we did find elevated levels of lead $(3,280.5 \mu g/g)$ in the soil of 1 household, and elevated levels of manganese and lead in El Cianuro, the stream draining the mine tailings (215.10-5,388.00 µg/l for manganese and 5.34–238.65 μ g/l for lead). Also, the public well at the far site that contained high arsenic also contained a high level of manganese (60.21 µg/l).

Importantly, our study sought to understand the extent to which the mine site affected mercury and arsenic uptake in the community. To accomplish this, we used fingernail tissue as a biomarker of body burden or internal dose of exposure. Elevated mercury concentrations in hair,^{14,15,19,21-34} urine,^{19,21,22,24,32,35} and blood^{14,19,21,27,35-39} have been found in individuals living near mines or people occupationally exposed as miners. To our knowledge, only 1 previous study examined nail tissue as a biomarker for mercury exposure around a mining site.²¹ In that study, relatively high mercury concentrations were detected in the fingernails of individuals 12 to 18 yr of age from 4 mining communities in southwestern Ghana; however, the study did not attempt to correlate biomarker concentrations with environmental samples (e.g., water or soil concentrations) or exposure history (e.g., diet). We are not aware of any biomarker studies of arsenic exposure around gold mines.

Nails are a reliable biomarker for mercury⁶ and arsenic^{6,7} exposure among adults, but data are sparse for children. Nail tissue has been shown to be resistant to external contamination with proper washing of tissue samples.^{7,40} Soil ingestion is a known route of metal exposure in children, and children also may experience higher inhalation of soil contaminants through playing in dirt. Moreover, the significantly positive correlation we found between drinking water and fingernail mercury concentrations suggests that ingestion plays a role in determining mercury levels in these samples, and that

fingernails reflect endogenous deposition rather than exogenous contamination. Nail samples are easy to collect, store, and transport. This makes them an attractive biomarker for investigations, particularly in remote areas and developing communities.

Examination of fingernail concentrations permitted the assessment of multiple exposure pathways. Drinking water concentrations of mercury predicted fingernail concentrations. Although dietary ingestion,⁴¹ especially of fish, 23, 26, 28, 33, 35, 42 is a major contributor to human mercury exposure in many populations, fish was not a typical part of the diet among the household members we tested in Siuna, and therefore was not a predictor of mercury uptake in our study population. The correlation we observed between soil and fingernail concentrations of mercury suggests that soil exposure is an exposure pathway for children, either through dust inhalation or soil ingestion,²³ and a more intimate interaction with the physical environment. Mercury vapor exposure appeared to be a source of fingernail mercury for miners, similar to a result reported by Ikingura and Akagi³² in miners' hair in Tanzania. Likewise, Joshi et al.⁴² found elevated mercury in the fingernail tissue of dentists who were exposed to mercury vapor from dental amalgam. Thus, in the Siuna population, fingernail mercury concentrations likely represented exposure through drinking water, soil, and artisanal mining.

For arsenic, drinking water concentrations predicted fingernail concentrations among adults who consumed water with higher arsenic levels. Several other studies have correlated drinking water arsenic concentrations7,43-46 and food arsenic concentrations41 with nail concentrations. Additionally, as with mercury, soil exposure appears to be an exposure pathway for children. Hinwood et al.43 found a strong correlation between soil and toenail arsenic in a study of 83 Australian individuals exposed to high arsenic in their residential environment, although the authors suggested that this correlation might have been exaggerated as a result of contamination of the nail samples. However, their washing procedure consisted of only 2 water rinses and 1 methanol rinse, and did not include the use of any soaping agents. Agahian et al.⁴⁰ identified a correlation between arsenic inhalation and nail concentrations in an occupational setting; the relationship between soil ingestion and arsenic uptake has not previously been studied. Children in our study exhibited higher arsenic body burdens than adults, as reflected by fingernail concentrations. This finding is consistent with our previous study of adults, in which we observed decreasing concentrations of arsenic in toenail tissue with increasing age.⁴⁶ In contrast to our study, however, Rodushkin and Axelsson¹⁰ found no age-specific differences in nail arsenic concentrations in a Swedish population that included 10 children under the age of 10 yr and 11 children between the ages of 10 and 20 yr out of a total sample of 96 individuals. However, body burdens of metals in the Swedish population likely reflect dietary sources more than other environmental exposures, such as those that appear to be important in the Siuna population.

Mercury and arsenic concentrations in fingernails in the Siuna population generally were higher than those reported in other populations. For mercury, the median concentration of 0.163 μ g/g was nearly twice the median in fingernails reported by a study of 96 subjects in Sweden (0.098 μ g/g).¹⁰ The maximum published value reported for mercury in nails was 2.80 µg/g, approximately equal to our maximum value. For arsenic, the median of 0.511 μ g/g was more than twice the median value for arsenic in fingernails (0.223 μ g/g) reported by Rodushkin and Axelsson.¹⁰ Our maximum value for arsenic was approximately twice the maximum published value they reported for arsenic in nails (2.570 μ g/g). These nail tissue levels—coupled with increasing evidence that mercury and arsenic are neurotoxic and carcinogenic, even at very low exposure levels-suggest that there may be detrimental health effects in Siuna as a result of toxic metal contamination.

Conclusions

Despite the long-term release of mercury from mining activities that had occurred in our study area, we detected relatively low environmental concentrations of mercury in Siuna, although these concentrations did reflect origin at the mining site. We found evidence of human uptake of mercury via drinking water and occupational exposure, and via soil exposure among children. Arsenic was present in the local environment at relatively high levels, although its distribution did not reflect origin at the mining site. Human uptake of arsenic also appeared to occur via drinking water exposure and via soil exposure among children. Measurement of mercury and arsenic levels in fingernail tissue helped us to identify differential exposure pathways in children vs. adults. As is the case for Bangladesh and other developing regions, Siuna has drinking water that is biologically contaminated. Our findings highlight the need to consider the likelihood of toxic metal exposure when introducing new drinking water sources in this community, and the need to better understand the potential role of soil as a pathway for arsenic and mercury exposure, particularly among children.

* * * * * * * * * *

The authors wish to thank Madeline Dalton, Joshua Hamilton, Angeline Andrew, Katie Martin, Stuart Lord, Bridges to Community, and the students of la Universidad de la Región Autónoma de la Costa Caribeña del Atlántico del Norte.

This study was supported by NIEHS grant ES07373, by the Center for Environmental Health Sciences at Dartmouth, and by the Dartmouth College Tucker Foundation.

Submitted for publication August 19, 2003; revised; accepted for publication July 24, 2004.

Requests for reprints should be sent to Dr. Margaret R. Karagas, Section of Biostatistics and Epidemiology, Department of Community and Family Medicine, Dartmouth Medical School, 7927 Rubin Bldg., One Medical Center Drive, Lebanon, NH 03756-1000.

E-mail: margaret.karagas@dartmouth.edu

* * * * * * * * *

References

- 1. van Straaten P. Mercury contamination associated with small-scale gold mining in Tanzania and Zimbabwe. Sci Total Environ 2000; 259(1–3):105–13.
- 2. Pestana MHD, Formoso MLL. Mercury contamination in Lavras do Sul, south Brazil: a legacy from past and recent gold mining. Sci Total Environ 2003; 307(1–3):125–40.
- 3. Zheng J, Hintelmann H, Dimock B, et al. Speciation of arsenic in water, sediment, and plants of the Moira watershed, Canada, using HPLC coupled to high resolution ICP-MS. Anal Bioanal Chem 2003; 377(1):14–24. [Epub 2003 May 7].
- 4. May TW, Wiedmeyer RH, Gober J, et al. Influence of mining-related activities on concentrations of metals in water and sediment from streams of the Black Hills, South Dakota. Arch Environ Contam Toxicol 2001; 40:1–9.
- 5. Golow AA, Schlueter A, Amihere-Mensah S, et al. Distribution of arsenic and sulphate in the vicinity of Ashanti Goldmine at Obuasi, Ghana. Bull Environ Contam Toxicol 1996; 56:703–10.
- Garland M, Morris JS, Rosner BA, et al. Toenail trace element levels as biomarkers: reproducibility over a 6-year period. Cancer Epidemiol Biomarkers Prev 1993; 2:493–7.
- Karagas MR, Le CX, Morris S, et al. Markers of low level arsenic exposure for evaluating human cancer risks in a US population. Int J Occup Med Environ Health 2001; 14(2):171–5.
- Wilde FD, Radtke DB, Gibs J, Iwatsubo RT, editors. Collection of water samples: U.S. Geological Survey techniques of water-resources investigations, book 9, chapter A4. Available from: http://pubs.water.usgs.gov/twri9A4/. Accessed August 31, 2005.
- U.S. Environmental Protection Agency (EPA). Microwave assisted digestion of sediments, sludges, soils, and oils. Washington, DC: EPA, 1994; Method 3051A, Revision 1.
- Rodushkin I, Axelsson MD. Application of double focusing sector field ICP-MS for multielemental characterization of human hair and nails. Part II. A study of the inhabitants of northern Sweden. Sci Total Environ 2000; 262(1–2):21–36.
- World Health Organization. WHO Guidelines for Drinking Water Quality, 3rd ed. P186. Available from: http://www.who.int/water_sanitation_health/dwq/gdwq 3/en/index.html>. Accessed August 31, 2005.
- 12. Agency for Toxic Substances and Disease Registry. Toxicological Profile for Mercury. Atlanta, GA: U.S. Department of Health and Human Services, 1999.
- Agency for Toxic Substances and Disease Registry. Toxicological profile for arsenic. Atlanta, GA: U.S. Department of Health and Human Services, 2000.
- 14. Akagi H, Castillo ES, Cortes-Maramba N, et al. Health assessment for mercury exposure among schoolchildren residing near a gold processing and refining plant in

Apokon, Tagum, Davao del Norte, Philippines. Sci Total Environ 2000; 259(1–3):31–43.

- 15. Appleton JD, Williams TM, Breward N, et al. Mercury contamination associated with artisanal gold mining on the island of Mindanao, the Philippines. Sci Total Environ 1999; 228(2–3):95–109.
- 16. Morel FM, Kraepiel AM, Amyot M. The chemical cycle and bioaccumulation of mercury. Ann Rev Ecology Systematics 1998; 20:543–66.
- 17. Pfeiffer WC, de Lacerda LD, Malm O, et al. Mercury concentrations in inland waters of gold-mining areas in Rondonia, Brazil. Sci Total Environ 1989; 87–88:233–40.
- 18. Kambey JL, Farrell AP, Bendell-Young LI. Influence of illegal gold mining on mercury levels in fish of north Sulawesi's Minahasa Peninsula (Indonesia). Environ Pollut 2001; 114(3):299–302.
- 19. Palheta D, Taylor A. Mercury in environmental and biological samples from a gold mining area in the Amazon region of Brazil. Sci Total Environ 1995; 168(1):63–9.
- 20. Fields S. Tarnishing the earth: gold mining's dirty secret. Environ Health Perspect 2001; 109(10):474–81.
- 21. Adimado AA, Baah DA. Mercury in human blood, urine, hair, nail, and fish from the Ankobra and Tano river basins in southwestern Ghana. Bull Environ Contam Toxicol 2002; 68:339–46.
- 22. Anawar HM, Akai J, Mostofa KMG, et al. Arsenic poisoning in groundwater: health risk and geochemical sources in Bangladesh. Environ Int 2002; 27(7):597–604.
- 23. Campbell L, Dixon DG, Hecky RE. A review of mercury in Lake Victoria, East Africa: implications for human and ecosystem health. J Toxicol Environ Health B Crit Rev 2003; 6(4):325–56.
- 24. Chowdhury UK, Biswas BK, Chowdhury TR, et al. Groundwater arsenic contamination in Bangladesh and West Bengal, India. Environ Health Perspect 2000; 108(5):393–8.
- 25. Cordier S, Garel M, Mandereau L, et al. Neurodevelopmental investigations among methylmercury-exposed children in French Guiana. Environ Res 2002; 89(1): 355–9.
- 26. Crompton P, Ventura AM, de Souza JM, et al. Assessment of mercury exposure and malaria in a Brazilian Amazon riverine community. Environ Res 2002; 90(2):69–75.
- Dolbec J, Mergler D, Sousa Passos CJ, et al. Methylmercury exposure affects motor performance of a riverine population of the Tapajós river, Brazilian Amazon. Int Arch Occup Environ Health 2000; 73:195–203.
- 28. Frery N, Maury-Brachet R, Maillot E, et al. Gold-mining activities and mercury contamination of native amerindian communities in French Guiana: key role of fish in dietary uptake. Environ Health Perspect 2001; 109(5):449–56.
- 29. Grandjean P, White RF, Nielsen A, et al. Methylmercury neurotoxicity in Amazonian children downstream from gold mining. Environ Health Perspect 1999; 107(7):587–91.
- Harada M, Nakachi S, Cheu T, et al. Monitoring of mercury pollution in Tanzania: relation between head hair mercury and health. Sci Total Environ 1999; 227(2–3):249–56.
- 31. Harada M, Nakanishi J, Yasoda E, et al. Mercury pollution in the Tapajos River basin, Amazon: mercury level of head hair and health effects. Environ Int 2001; 27(4):285–90.
- 32. Ikingura JR, Akagi H. Monitoring of fish and human exposure to mercury due to gold mining in the Lake Victoria goldfields, Tanzania. Sci Total Environ 1996; 191(1–2):59–68.

- Maurice-Bourgoin L, Quiroga I, Chincheros J, et al. Mercury distribution in waters and fishes of the upper Madeira rivers and mercury exposure in riparian Amazonian populations. Sci Total Environ 2000; 260(1–3):73–86.
- 34. Olivero J, Johnson B, Arguello E. Human exposure to mercury in San Jorge river basin, Colombia (South America). Sci Total Environ 2002; 289(1–3):41–7.
- 35. Silbergeld EK, Nash D, Trevant C, et al. Mercury exposure and malaria prevalence among gold miners in Para, Brazil. Rev Soc Bras Med Trop 2002; 35(5):421–9.
- Counter SA. Neurophysiological anomalies in brainstem responses of mercury-exposed children of Andean gold miners. J Occup Environ Med 2003; 45(1):87–95.
- 37. Counter SA, Buchanan LH, Laurell G, et al. Blood mercury and auditory neuro-sensory responses in children and adults in the Nambija gold mining area of Ecuador. Neurotoxicology 1998; 19(2):185–96.
- Counter SA, Buchanan LH, Ortega F, et al. Elevated blood mercury and neuro-otological observations in children of the Ecuadorian gold mines. J Toxicol Environ Health A 2002; 65(2):149–63.
- 39. Sing KA, Hryhorczuk DO, Saffirio G, et al. Environmental exposure to organic mercury among the Makuxi in the Amazon basin. Int J Occup Environ Health 1996; 2(3):165–71.

- 40. Agahian B, Lee JS, Nelson JH, et al. Arsenic levels in fingernails as a biological indicator of exposure to arsenic. Am Ind Hyg Assoc J 1990; 51(12):646–51.
- 41. Macintosh DL, Williams PL, Hunter DJ, et al. Evaluation of a food frequency questionnaire-food composition approach for estimating dietary intake of inorganic arsenic and methylmercury. Cancer Epidemiol Biomarkers Prev 1997; 6:1043–50.
- 42. Joshi A, Douglass CW, Kim HD, et al. The relationship between amalgam restorations and mercury levels in male dentists and nondental health professionals. J Public Health Dent 2003; 63(1):52–60.
- 43. Hinwood AL, Sim MR, Jolley D, et al. Hair and toenail arsenic concentrations of residents living in areas with high environmental arsenic concentrations. Environ Health Perspect 2003; 111(2):187–93.
- 44. Karagas MR, Morris JS, Weiss JE, et al. Toenail samples as an indicator of drinking water arsenic exposure. Cancer Epidemiol Biomarkers Prev 1996; 5(10):849–52.
- 45. Karagas MR, Stukel TA, Tosteson TD. Assessment of cancer risk and environmental levels of arsenic in New Hampshire. Int J Hyg Environ Health 2002; 205:85–94.
- Karagas MR, Tosteson TD, Blum J, et al. Measurement of low levels of arsenic exposure: a comparison of water and toenail concentrations. Am J Epidemiol 2000; 152(1):84–90.

Copyright of Archives of Environmental Health is the property of Heldref Publications. The copyright in an individual article may be maintained by the author in certain cases. Content may not be copied or emailed to multiple sites or posted to a listserv without the copyright holder's express written permission. However, users may print, download, or email articles for individual use.