Urinary Inorganic Arsenic in Residents Living in Close Proximity to a Nickel and Copper Smelter in Ontario, Canada

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ABSTRACT

Objectives: The current study determines whether internal exposure to inorganic arsenic for residents living in close proximity to an operating smelter is any different from that for residents living in a similar community without anthropogenic sources of inorganic arsenic. Furthermore, the study examines factors that may contribute to internal inorganic arsenic exposure in these populations.

Methods: A cross-sectional study was used to determine whether recent internal exposure to inorganic arsenic for residents living in close proximity to an operating smelter with known elevated soil arsenic content is any different from exposure for residents living in a similar socio-demographic community without point sources of arsenic. Personal information was collected from participants during in-home interviews, and first morning void urine samples were obtained to assess recent internal exposure to arsenic.

Results: The mean concentration of urinary inorganic arsenic of residents living in the exposed community was 7.11 ug/L (Standard deviation (SD) 4.53, N=368) and in the non-exposed community was 7.19 ug/L (SD 5.63, N=321). The difference in urinary inorganic concentration was not statistically significant ($p=0.83$). Regression analysis showed that age and sex were significant predictors of internal exposure, with males and younger age groups having higher exposure levels.

Conclusions: Based on analyses of data collected, the results did not demonstrate a statistically significant difference in recent internal exposure to inorganic arsenic between the community with and the one without industrial or other point sources of environmental arsenic contamination. However, individual characteristics such as age and sex are important contributors to urinary inorganic arsenic levels.

Key words: Urinary arsenic; exposure assessment; biological monitoring; community health

La traduction du résumé se trouve à la fin de l'article.

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Arsenic occurs naturally in low concentrations throughout the environment.¹ However, mining and smelting operations are important anthropogenic sources of inorganic arsenic and represent a major health concern among residents living within close proximity of such operations. Inorganic forms of arsenic are associated with diabetes^{2,3} and cardiovascular diseases.⁴ Inorganic arsenic is also a known human carcinogen^{5,6} and long-term exposure has been associated with cancers of the bladder, kidney, lung and liver.⁷ Due to the potential serious adverse health effects, prolonged exposure to air emissions and soil arsenic from anthropogenic sources represents a source of concern among local residents of mining and smelting communities.

Although studies in the past have been conducted to examine the relationship between environmental arsenic and urinary inorganic concentrations, results have been inconsistent for soil arsenic.⁸⁻¹² The current study was conducted to determine whether recent internal exposure to inorganic arsenic for residents living in close proximity to an operating smelter is any different from that for residents living in a similar community without anthropogenic sources of arsenic. Furthermore, the study examines factors that may contribute to internal inorganic arsenic exposure in these populations.

METHODS

Study design and population

A cross-sectional study was used to compare urinary inorganic arsenic concentrations in residents living in the exposed commu-

nity of Falconbridge with those in residents from the control community of Hanmer. Average soil arsenic concentration for the exposed community (FC) was 78.5 ug/g (SD 84.9, n=879) and for the control community (HC) was 3.7 ug/g (SD 3.47, n=143).¹³ As part of the Sudbury Area Risk Assessment, 24-hour air samples showed that in most cases, atmospheric arsenic was below detection limit for both HC and FC.¹⁴ No anthropogenic sources of arsenic were identified in the control community and this was confirmed by the observed low soil arsenic concentrations.

The study population consisted of residents living in Falconbridge, a small community of approximately 250 households with 700 residents of all ages living within 500 metres from the smelter. The community is named after the Falconbridge smelter that is owned and has been operated by Falconbridge Limited over the past 40 years. The Community of Hanmer (HC), located approximately 15 kilometres northwest of FC, was chosen as a comparison community as it has a similar socio-demographic profile as FC but with no history of mining or smelting in the community.

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Conflict of Interest: None to declare.

Ethics approval was obtained from the Research Ethics Review Committee of the Sudbury District Health Unit. All FC residents and a simple random sample of households from HC were invited to participate in the study. Informed consent was obtained from all study participants prior to data collection.

Data collection

Data collection occurred in the months of September and October of 2004. Eligible participants were first sent a letter describing the study and indicating that a member of the study team would be visiting their house to enquire about their interest in participating in the study. Once interest was confirmed, a time convenient for the participants was set up for an in-house interview. All interviewers were trained in the questionnaire administration and explanation of the urine collection protocol. Interviews were conducted by a two-member team using a structured questionnaire to collect information from each member of the household. For practical reasons, data were collected from the person most knowledgeable of household activities. Information collected included residential history, occupational history, diet (garden vegetables, fish and seafood consumption), smoking history, medication use, and history of medical problems, and children's indoor and outdoor activities.

Urine samples collection, storage, transport and analysis

In addition to the interviews, participants were also asked to provide urine samples for laboratory analysis of arsenic content. The interviewers reviewed the urine collection protocol with participants to ensure that they understood the collection procedure. Each household was provided with a urine collection kit containing written instructions, a cooler bag and icepack to keep the sample cold. Each participant was provided with a 125 mL Nalgene screw cap bottle labeled clearly with the participant's name. These bottles were pre-tested to ensure that they did not contain traces of arsenic. First morning void urine was collected into the Nalgene bottle by each study participant. For toddlers, potties were used to facilitate urine collection. Potties were pre-rinsed in mild acid in the laboratory to remove contaminants and were kept sealed in plastic wraps until ready for use. The participant was instructed to place the sample in the cooler bag in the refrigerator until pickup by the study team the same morning. Subsequently, the urine samples were kept at 4°C in a monitored and temperature-controlled refrigerator. No power outages or malfunction of refrigeration equipment was experienced throughout the study. Samples were shipped to the laboratory in insulated containers containing icepacks to maintain temperature.

Laboratory analytic methods

The Trace Element Laboratory of London Laboratory Services Group conducted all analyses of urine: creatinine, total and inorganic arsenic. Creatinine in urine was measured with the Jaffe colorimetric method on a Beckman Synchron LX20 analyzer (Beckman Coulter, Fullerton, CA 92834-3100).

Total urinary arsenic was determined by high-resolution inductively coupled plasma-mass spectrometry (HR-ICP-MS) using a Finnigan MAT Element instrument (Bremen, Germany) directly, after appropriate dilution of the urine. This consisted of inorganic (As^{3+} , As^{5+} , monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA)) and organic species (arsenobetaine + arsenocholine).

Table 1. Descriptive Statistics of Selected Subject Characteristics

Characteristics	Falconbridge n (%)	Hanmer n (%)
Sex		
Males	189 (51.2)	156 (48.9)
Females	180 (48.8)	165 (51.4)
Age (years)		
Mean (SD)	37.9 (22.5)	36.1 (21.2)
Co-morbidity		
High cholesterol	70 (19.0)	60 (18.7)
Diabetes	21 (5.7)	20 (6.2)
Gastrointestinal conditions	38 (10.3)	43 (13.4)
Cancer (any type)	19 (5.1)	10 (3.1)
Skin rashes or lesions	63 (17.1)	53 (16.5)
Other conditions	133 (36.0)	127 (39.9)
None	151 (40.9)	126 (39.3)
Smoking status		
Current tobacco use	85 (32.2)	54 (24.7)
Cigarettes	82 (31.1)	50 (22.8)
Other tobacco	7 (2.7)	2 (0.9)
Do not use tobacco	179 (67.8)	165 (75.3)
Exposure to second-hand smoke (Within the past 7 days)		
In the house	61 (21.6)	49 (18.4)
In the car	24 (8.7)	35 (13.7)
Other locations	52 (19.3)	51 (19.2)
Handwashing practices before eating (12 years and under)		
Usually (75-100%)	43 (60.6)	53 (68.8)
Sometimes (25-75%)	18 (25.4)	16 (20.8)
Rarely (0-25%)	10 (14.1)	8 (10.4)
Handwashing practices before going to sleep (12 years and under)		
Usually (75-100%)	63 (90.0)	56 (74.7)
Sometimes (25-75%)	4 (5.7)	9 (12.0)
Rarely (0-25%)	3 (4.3)	10 (13.3)
Education		
< High school	16 (6.1)	23 (10.5)
High school	95 (36.3)	78 (35.5)
Post secondary	143 (54.6)	108 (49.1)
Other	8 (3.1)	11 (5.0)
Years in current residence		
Mean (SD)	15.5 (12.8)	12.3 (12.3)
Years in community		
Mean (SD)	21.9 (17.8)	21.8 (16.6)

SD=Standard Deviation

Figure 1. Box plot of inorganic arsenic concentration for Falconbridge (exposed) and Hanmer (comparison) communities

The latter concentration was determined by the difference between the total amount and the inorganic arsenic concentration. The reference range for total arsenic is 0-0.53 $\mu\text{mol/L}$ (0-70 $\mu\text{mol/mol}$ creatinine) in non-occupational subjects. The detection limit is 0.5 $\mu\text{g/L}$.

Table 2. Comparison of Arithmetic Mean of Urinary Inorganic Arsenic Concentration (ug/L) by Community and Age Groups

Age Group (years)	Communities				Parametric		Non-parametric	
	Falconbridge (Exposed)		Hanmer (Control)		t-value	p-value	Mann-Whitney	p-value
	n	Mean (SD)	n	Mean (SD)				
All	368	7.11 (4.53)	321	7.19 (5.63)	0.21	0.83	58,019	0.64
0-4	18	8.66 (4.65)	17	7.53 (3.63)	0.79	0.44	131	0.46
5-12	53	9.51 (6.45)	61	9.30 (6.24)	0.18	0.86	1578	0.83
13-17	29	7.77 (4.32)	17	7.89 (4.18)	0.09	0.69	236	0.80
18+	268	6.46 (3.87)	226	6.54 (5.56)	0.20	0.84	29,378	0.52

Table 3. Multiple Linear Regression Analyses of Urinary Inorganic Arsenic Levels

Model	Independent Variables	Falconbridge & Hanmer			Falconbridge (Exposed Group)			Hanmer (Comparison Group)		
		Beta	SE	p-Value	Beta	SE	p-Value	Beta	SE	p-Value
Overall	Sex	-0.176	0.377	0.000	-0.224	0.450	0.000	-0.131	0.625	0.019
	Age	-0.246	0.009	0.000	-0.260	0.011	0.000	-0.215	0.015	0.000
	Community	0.015	0.383	0.682	—	—	—	—	—	—
	Exposed to smoke	-0.064	0.446	0.146	-0.087	0.554	0.163	-0.031	0.723	0.626
	Smoking status	-0.018	0.550	0.678	0.008	0.647	0.900	-0.037	0.945	0.563
	Above average exposure	0.074	0.414	0.054	0.055	0.519	0.286	0.085	0.658	0.134
	Ate food over camp fire (past 7 days)	0.049	0.724	0.202	0.026	0.922	0.628	0.063	1.164	0.262
	Ate food from garden	-0.024	0.438	0.525	0.032	0.575	0.530	-0.078	0.679	0.173
	Handled pressure-treated wood	0.016	0.838	0.662	0.017	0.952	0.745	0.023	1.516	0.679
	Ate outside	-0.019	0.466	0.610	0.030	0.579	0.575	-0.062	0.765	0.268
	Ate fish (past 7 days)	-0.089	0.397	0.022	-0.023	0.482	0.658	-0.149	0.651	0.009
		R-square = 0.33			R-square = 0.37			R-square = 0.32		
Children (0-18 years)	Wash hands before eating	0.042	0.0851	0.654	0.127	1.360	0.382	-0.031	1.114	0.805
	Eat with fork or spoon	-0.061	1.403	0.527	-0.149	0.806	0.326	-0.019	2.647	0.887
	Hands/face washed before bed	0.049	1.100	0.593	0.037	2.348	0.795	0.060	1.249	0.630
	Chew on object or toys	0.180	1.228	0.057	0.037	2.061	0.801	0.272	1.557	0.036
	Eat dirt	-0.111	1.981	0.245	-0.264	2.688	0.061	0.052	3.084	0.704
		R-square = 0.04			R-square = 0.09			R-square = 0.32		

SE = Standard Error

Inorganic arsenic (As) was determined by solvent extraction with toluene of the acidified urine after reduction with potassium iodide (KI) that excludes the organic species. Following back extraction of the inorganic As with 1% HNO₃, it was quantified by high-resolution inductively coupled plasma mass spectroscopy (HR-ICP-MS). The concentration of inorganic As was determined from a calibration curve of National Institute of Standards Technology similarly treated, and analyzed by HR-ICP-MS. The reference range for inorganic arsenic is 0-0.13 µmol/L (0-25 µmol/mol creatinine) in non-occupational subjects. The detection limit is 0.5 µg/L.

Statistical methods

Descriptive statistics, including frequency distributions and measures of central tendency, were used to examine the distribution of the variables and to characterize the study population. Differences in urinary arsenic levels between communities were compared using both parametric (independent t-test) and non-parametric (Mann-Whitney U) statistical tests. Statistical significance was based on a 0.05 significance level. Modelling for predictors of internal exposure began by selecting survey responses that, based upon current knowledge, would be an indication of the level of exposure to arsenic. The association between the selected survey response and the level of the chemical of concern (COC, or inorganic arsenic) was examined graphically through box plots or scatter grams and tested for statistical significance. Those survey questions that were found to have p-values below 0.15 in the bivariate analyses were considered for inclusion in a multivariate regression model. A regression of variables in the multivariate model that were found to be statistically significantly related to urinary arsenic were deemed to be predictors of internal exposure to a given COC. SPSS (version 11) was used for statistical analyses.

RESULTS

In Falconbridge Community, of the 273 households invited to participate in the study, 148 (54%) agreed. In addition, there were 20 households (7%) willing to participate but not available during the data collection period due to vacation, illness, or work schedules. Ninety-three households (34%) refused to participate in the study. Reasons for refusal varied but included “not interested in study”, “doesn’t perceive there is a problem”, “believes that they are too old”, or “not enough time available”. The study team was unable to contact any adult residents of the remaining 12 households (5%). Overall, information was collected for 393 participants in the interview portion of the study and, of these, 368 participants provided a urine sample. In HC, of the 360 households approached, 129 (36%) agreed to participate in the study. Interviews captured information on 335 respondents, and 321 participants provided urine samples.

Characteristics of study population

As shown in Table 1, sex distribution of the two communities was very similar with approximately equal proportions of males and females participating from each community. The age distribution of the two samples also followed a similar pattern with a slightly higher proportion of 6-12 year olds and 50-64 year olds in Hanmer, and slightly higher proportions of 18-34 year olds and those 65 years or older in Falconbridge. The average for FC was 37.9 years and for HC was 36.1 years. However, the differences were not statistically significant ($p > 0.05$). Most residents reported some form of health problem. The most common problems were high blood pressure (19%), skin rashes or lesions (17%) and gastrointestinal problems (12%). The prevalence of current tobacco use is slightly higher in Falconbridge Community (32%) in comparison to Hanmer (25%).

Inorganic urinary arsenic

In total, 689 samples were collected from study participants, 368 from Falconbridge Community and 321 from the control community of Hanmer (Table 2). Of these, 149 (22%) samples were collected from children under the age of 13 years. The arithmetic mean for the total group was 7.11 µg/L and 7.19 µg/L for FC and HC, respectively. The median (Figure 1) was the same for each community (5.99 µg/L). The whiskers of the two box plots show that the distributions of the urinary inorganic arsenic are also similar for both communities. The highest average urinary inorganic concentration was seen in the 5-12 year old age group for both communities.

Table 2 also shows the results of statistical comparison of using both parametric and non-parametric approaches. Comparisons were made for all participants combined as well as stratified by age groups. For all comparisons, there was no statistically significant difference seen for inorganic arsenic levels (overall or by age groups) in urinary inorganic levels for residents of Falconbridge Community (exposed group) and Hanmer Community (control group).

Factors contributing to internal exposure

Regression models were constructed to determine whether place of residence (FC vs. HC) and other factors were significant predictors of urinary inorganic arsenic concentration. Separate models were also constructed for adults and children. Results of the multiple regression analyses are shown in Table 3. The overall models were significant for all participants, and for each community. For the overall model with all participants included, the main variables associated with urinary inorganic arsenic levels were age (younger people have higher levels), sex (males tend to have higher levels), and fish consumption within the past seven days. Place of residence was not a predictor of urinary arsenic level. The overall model for Falconbridge demonstrated that the variables significantly associated with urinary arsenic included age and sex. Hanmer had the same pattern as that found in the overall model.

DISCUSSION

Inorganic arsenic has been associated with a number of adverse health effects. Potential exposure to high inorganic arsenic levels from soil caused by historical smelting practices is a legitimate source of concern for residents living in this area. In response to these concerns, this study used a population-based exposure survey designed to examine the recent arsenic exposures. Most of the arsenic ingested is excreted in urine within a few days.¹⁵⁻¹⁸ Urinary inorganic arsenic is considered a reliable biomarker for recent arsenic exposure and was the primary outcome of interest in this survey. This was justified and considered in discussions with community residents on the design of the study.

Our results were consistent with findings from Tsuji and colleagues¹² in that soil arsenic concentration was not significantly related to urinary levels. These results suggest that environmental arsenic in this study has minimal impact on the overall internal dose. However, these findings are different from some other studies that have found evidence of increased exposure associated with soil arsenic.⁹⁻¹¹ The discrepancy may be attributable to a number of factors such as soil arsenic concentration, bioaccessibility and lifestyle. With respect to soil concentration, soil arsenic in the study conducted by Hinwood and colleagues (2004) ranged from

9-9000 µg/g while soil arsenic concentrations in our study were significantly lower, ranging from 2.5 to 620 µg/g.¹⁹ Furthermore, evidence suggests that the bioaccessibility of the soil arsenic in Falconbridge was low.¹⁹ Soil speciation analyses of the Greater Sudbury Area sampling indicated that the predominant form of arsenic present in the soil was arsenopyrite (FeAsS), a sulphidic form of the arsenic derived from the ore processed in the Greater Sudbury Area.¹⁹ Based upon the results of the sequential leach analyses, the arsenic present in the soil leached out primarily in the organic step (the most aggressive of the leaching phases).¹⁹ This indicates that the majority of the arsenic present within the soil matrix was tightly bound to the organic portion of the soil, with little being available to the type of influences that would typically occur in the gastrointestinal tract.¹⁹ As a result, estimates of the upper-end bioaccessibility in soil were 37.3%. This is lower than that observed in other studies (46.5%).⁸ Finally, exposure may have been influenced also by the level of individuals' interaction with the environment. Results from our questionnaire showed that many study participants engaged in hunting, fishing and other outdoor activities. While these are typical of the lifestyle of Ontario's northern communities, frequent interactions with an environment with high levels of arsenic can also contribute to seasonal arsenic exposure, especially in the summer months. Results from our questionnaire showed that levels of outdoor physical activity were similar for both communities; therefore it is unlikely that differences in lifestyle (i.e., fishing, hunting and other outdoor activities) biased study results.

Our regression models showed that age and sex were important predictors of urinary inorganic arsenic level. This observation was consistent with studies conducted elsewhere. Children are of particular concern for greater risk of direct exposure to environmental arsenic. The highest average level of urinary inorganic arsenic was found among participants 5-12 years of age in both communities. Furthermore, average urinary inorganic arsenic in children from the exposed community was slightly higher than in children from the comparison community of Hanmer, although the differences were not statistically significant (Table 2). Children under the age of 2 had similar levels in exposed and unexposed communities, and levels increased commensurate with age. Higher urinary arsenic levels among younger children may reflect soil ingestion as the exposure source and hand-to-mouth behaviours at early ages. Urine collection from toddlers was a challenge and families were assisted by our team physician in the collection process in order to ensure compliance with the procedures.

We were also concerned about the potential confounding effect of non-soil arsenic such as arsenic from drinking water, household dust, and seafood consumption. Although it was not possible to obtain individual water exposure information, we examined water quality records of both communities. At the time of the study, both communities were on separate municipal water sources. Public utility records revealed no differences in drinking water arsenic in both communities, levels being compliant with (usually much lower than) regulatory limits of 10 µg/L.²⁰ Therefore, potential biases caused by different arsenic concentration in drinking water were not likely. Data on individual household dust were not available at the time of the study. However, previous studies have shown that house dust is not a significant predictor of urinary arsenic.^{21,22} We also examined the potential confounding effect of fish and seafood intake up to 7 days before the urine sample was obtained. Although

most fish and seafood contain primarily (>90%) organic arsenic.²³ up to 10% may be inorganic and bioavailable. In our survey, the amount of seafood consumed in the 7 days prior to urine collection was not significantly different between the two communities.

Timing of the study was also subject to detailed discussions prior to its implementation. Summer season might have been an ideal window for data collection in order to maximize contact with soil arsenic exposure, particularly among children. Our data collection for the current study occurred in September and October. This decision was made in order to minimize absences due to typical summer travel and to better reflect activities of children from both communities. At the time of the study, all ground surfaces were still exposed, with no snow cover or any other impediments to potential exposure from soil. For this study, we used urine samples from first morning void as a proxy measure of current internal exposure to environmental arsenic. While it is possible that a 24-hour sample would be a better assessment of internal exposure than first morning void, the added burden on the study participants would significantly reduce the response rate.

There are limitations to this study. This cross-sectional study was designed to obtain a snapshot of the current exposure. Unless all environmental and behavioural factors remain constant, results of the current study cannot be generalized to past or future burden of internal inorganic arsenic exposure. The smelter is still in operation, and the degree to which residents will be exposed in the future depends in part on future emissions from these operations. Within the FC sample, distance from smelter was not considered as a variable because the entire town is built within approximately 500 metres from the smelter. In addition, the town has one park, one arena, and all children tended to play in the same areas. Misclassification of exposure would occur if we had attempted to categorize families using distance from smelter, given that the conditions would have been very similar.

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RÉSUMÉ

Objectifs : Notre étude vise à déterminer si l'exposition interne à l'arsenic inorganique chez les résidents vivant à proximité d'une fonderie en activité diffère de celle des résidents d'une localité semblable, mais sans source anthropogénique d'arsenic inorganique. De plus, nous examinons les facteurs pouvant contribuer à une exposition interne à l'arsenic inorganique dans ces populations.

Méthode : Nous avons mené une étude transversale pour déterminer si l'exposition interne récente à l'arsenic inorganique chez les résidents vivant à proximité d'une fonderie en activité ayant des niveaux connus élevés d'arsenic dans le sol diffère de l'exposition des résidents vivant dans une localité semblable sur le plan sociodémographique, mais sans source ponctuelle d'arsenic. Des renseignements personnels ont été recueillis auprès des participants à la faveur d'entrevues à domicile, et nous avons prélevé des échantillons de la première urine du matin pour évaluer l'exposition interne récente à l'arsenic.

Résultats : La concentration moyenne d'arsenic inorganique dans les urines des résidents vivant dans la localité exposée était de 7,11 µg/L (écart-type [ET] 4,53, N=368); dans la localité non exposée, elle était de 7,19 µg/L (ET 5,63, N=321). La différence de concentration en matières inorganiques dans l'urine n'était pas significative (p=0,83). Une analyse de régression a montré que l'âge et le sexe étaient des prédicteurs significatifs d'une exposition interne, les niveaux d'exposition étant plus élevés chez les hommes et dans les groupes d'âge plus jeunes.

Conclusion : D'après les analyses des données recueillies, les résultats ne démontrent pas de différence significative dans l'exposition interne récente à l'arsenic inorganique entre la localité ayant une source industrielle ou autre source ponctuelle de contamination à l'arsenic présent dans l'environnement et la localité n'ayant pas de telle source. Cependant, les caractéristiques individuelles comme l'âge et le sexe contribuent de façon importante aux niveaux d'arsenic inorganique dans les urines.

Mots clés : arsenic urinaire; évaluation du risque; biosurveillance; santé communautaire