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ORIGINAL RESEARCH

House dust and inorganic urinary arsenic in two Arizona mining towns

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Residents of copper mining and smelting towns may have increased risk of arsenic exposure from elevated arsenic contained in environmental media. To determine the relation of arsenic in house dust to inorganic urinary arsenic concentrations, a door-to-door survey was conducted in Hayden and Winkelman, Arizona. A total of 122 households (404 individuals) participated; 85 provided dust samples. Urine was collected at first morning void and analyzed for total and speciated arsenic. Speciation of arsenic was performed in samples with total arsenic above 10 μg/l ($N = 106$). The generalized estimating equation was used to determine the relation between urinary and house dust arsenic concentrations, allowing adjustment for the correlation of measurements obtained from the same home. Seafood consumption during the past 3 days and smoking contributed significantly to inorganic urinary arsenic, after adjusting for age and gender. Arsenic in house dust was not significantly associated with inorganic urinary arsenic measurements in this population.
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Introduction

Arsenic is a naturally occurring element in the environment that may be released from industrial processes, including copper smelting. Arsenic is a known human carcinogen (ATSDR, 1993). Exposure to arsenic through drinking water at levels exceeding 30 μg/l is associated with cutaneous effects, including skin cancer (Tseng et al., 1968; Tseng, 1977; Tondel et al., 1999). Studies of skin cancer and arsenic exposure in the United States show little or no association at levels around 10 μg/l (Morton et al., 1976). In addition, many studies report significant associations between arsenic in drinking water and several other cancer types, including lung, bladder, and liver (Chen et al., 1985, 1986, 1992; Bates et al., 1995; Smith et al., 1998). Arsenic is associated with lung cancer in populations with occupational exposure to inhaled arsenic (Lee-Feldstein, 1983; Jarup and Pershagen, 1991).

Arsenic is found in many kinds of rock, particularly in ores containing copper or lead. When the ores and flux are heated at smelters, much of the arsenic is released as arsenic trioxide (As_2O_3) (ATSDR, 1993). The presence of high arsenic concentrations in environmental and biological media near copper smelters is well documented (Milham and Strong, 1974; Baker et al., 1977; Hartwell et al., 1983; Binder et al., 1987; Pollisar et al., 1990; Hwang et al., 1997) . Several studies evaluate exposure to arsenic and resultant health effects in populations living near copper smelters (Blot and Fraumeni, 1975; Pershagen et al., 1977; Greaves et al., 1981; Rom et al., 1982; Pershagen, 1985; Bates et al., 1992; Marsh et al., 1998). These health effects include lung cancer, low birth weight, spontaneous abortion, and congenital malformations (Hughes et al., 1988).

Arsenic exists in many forms, including inorganic (As^{3+}/As^{5+}), monomethyl arsonic acid (MMA), dimelthylarsinic acid (DMA), and arsenicals including arsenic containing proteins and sugars. In general, inorganic arsenic is the most toxic form followed by MMA and DMA, with much less toxicity from naturally occurring organic arsenic. Studies of arsenic ingestion in humans demonstrate that 60–80% of daily oral intake of these compounds is excreted in urine (Crecelius, 1977; Tam et al., 1979; Buchet et al., 1981). Following inhalation, arsenic deposited in the lung can be both directly absorbed and carried by the mucociliary escalator back into the throat and swallowed, where it is absorbed through the gastrointestinal tract. No evidence exists for the uptake of inorganic arsenic through dermal absorption (ATSDR, 1993). In an attempt to separate

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environmental exposures from dietary exposures, urine is often speciated, and the concentration of inorganic arsenic, MMA, and DMA is used as a biomarker of exposure.

Understanding the potential contribution to total arsenic exposure from different routes is essential to formulate community arsenic reduction strategies. This study evaluates urinary arsenic among all residents of the copper mining and smelting towns of Hayden and Winkelman, Arizona. House dust samples were collected to evaluate the arsenic distribution in environmental media from homes where residents provided urinary samples, and to evaluate the association between dust arsenic and inorganic urinary arsenic concentrations.

Materials and methods

Study Area and Selection of Households

We conducted a door-to-door survey of all residents in Hayden and Winkelman, Arizona (Figure 1) from June to October 1999. Hayden and Winkelman have a combined population of 1585 people according to 1990 census data. Prior to the survey, a campaign to educate residents about lead and arsenic and possible adverse health effects was initiated. Multicultural Spanish-speaking field crews administered the questionnaire in the participants' language of choice (English or Spanish) and collected all samples.

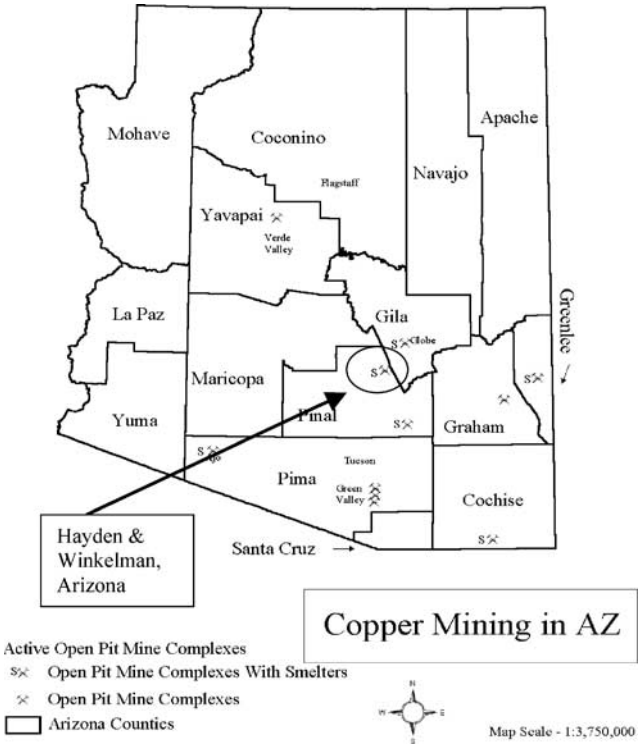

Figure 1. Location of the Hayden and Winkelman mining district and other mining operations in Arizona.

Every residence in the two towns was approached to participate, and homes where there was no initial response were approached no more than five times. All members of each household were asked to participate in the survey.

Sample Collection

All participating subjects agreed to fill out a questionnaire ascertaining sociodemographic characteristics. Each subject willing to provide urine answered an additional set of questions regarding time lived in current home and community, present occupation (mine and/or smelter worker), consumption of seafood and mushrooms, and whether the respondent ate home or locally grown fruits and vegetables. Questionnaire data at the household level included questions about the age of the home, primary water source for individuals in the home, any recent construction, and how the home was cooled and heated.

Subjects agreeing to urine analysis provided a urine sample from the first morning void. Urine was collected in three-ounce acid-washed containers and frozen by the participant immediately upon collection of the sample. Urine was collected by the field teams and stored at $-29^{\circ}C$ ($-20^{\circ}F$) until analyzed.

Environmental sampling included obtaining the bag from the vacuum cleaner used primarily indoors. One bag from each household willing to participate was placed in a sealed ziplock bag ($N = 85$). Dust samples were stored in a freezer until the time of analysis.

Laboratory Analysis

All urine was analyzed for total arsenic ($N = 224$) using hydride generation-atomic absorption spectrophotometry (Departmento de Farmacologia y Toxicologia, Seccion de Toxicologia Ambiental, Mexico City). Those with total arsenic concentrations above 10 μg/l ($N = 106$) were analyzed for combined As^{3+} and As^{5+}, MMA, and DMA. Originally, 108 samples had total urinary arsenic above 10 μg/l; however, two of the samples were lost during processing.

Each dust sample was sieved to three sizes: coarse (gravel, ≥ 2 mm); medium (sand, > 2 mm and $\leq 62.5\,\mu$m); fine (clay and silt $< 62.5\,\mu$m). One gram of the finest fraction ($< 62.5\,\mu$m) was placed in a plastic X-ray fluorescence (XRF) cup, covered with mylar film, and set aside for XRF analysis. Samples were analyzed for several metals using the fine particle Soil Application on the Spectrace 2000. The samples were analyzed for 200 s on each of three isotopes (^{109}Cd, ^{55}Fe, and ^{241}Am). (Arsenic was evaluated with ^{109}Cd.) The XRF was calibrated using a Teflon plug; the values returned had to be within three standard deviations of zero to ensure calibration. Calibration was performed before and after each batch ($n = 10$) of samples was run. The

minimum detection limit for arsenic in this application was 50 μg/g.

Statistical Analysis

Urine samples with arsenic levels below the limit of detection were assigned values of one-half the limit of detection (LOD) for all statistical analysis ($N = 15$). The LOD for total urinary arsenic was 2.25 μg/l. Analysis for inorganic arsenic was completed in two phases, with different LODs. If the total urinary concentration was above 30 μg/l, then the LOD for inorganic arsenic was 1 μg/l. If the total urinary arsenic concentration was less than 30 μg/l, then the LOD for inorganic arsenic was 0.5 μg/l. Urine samples with total arsenic concentrations below 10 μg/l were not analyzed for inorganic arsenic ($N = 118$). Total and inorganic urinary arsenic measurements were log transformed so that parametric statistics could be used. Dust samples below the LOD were assigned a value of 25 μg/g for all statistical analysis ($N = 32$).

Data were analyzed for the relation between dust and inorganic urinary arsenic using STATA 6.0 (College Station, TX, USA). The generalized estimating equation (GEE) was used to adjust for multiple urine samples collected from a single household. The GEE enables use of the individual urinary arsenic values rather than taking an average for a household or using one person to represent the household. We assumed that the correlation between individual measurements in any single home is the same (exchangeable correlation matrix). An alternative model making no assumptions about the within-household correlation structure (unstructured correlation matrix) yielded similar results (not shown).

Terms significant bivariately at the $P = 0.10$ level using GEE were included in the model. Age, gender, and consumption of seafood in the past 72 h were forced into the model because of past studies showing significant associations between these variables and inorganic urinary arsenic measurements.

Results

Enrolled subjects were compared to the general population of Hayden and Winkelman using data from the 1990 census (Census, 1990). Participants in Hayden and Winkelman did not differ by age or gender from data reported for the two towns in 1990 census data (Table 1). However, the race/ethnicity of the participants in our study differed significantly from the distribution of race/ethnicity in the two towns as reported in the 1990 census ($P < 0.05$).

All enrolled participants ($N = 404$) agreeing to complete the descriptive questionnaire were asked to provide a urine sample, and 224 agreed to do so (55%). No significant differences were found in gender, race/ethnicity, or smoking habits between the 224 who provided urine and the 180 who did not (Table 2). The age structure of the population providing urine was significantly different from that of the population who did not provide urine. Specifically, very few individuals under the age of 29 agreed to provide urine for this survey. Characteristics of the 106 subjects who had total urinary arsenic ≥ 10 μg/l and were therefore speciated are presented in Table 3. Subjects with total urinary arsenic ≥ 10 μg/l ($N = 106$) and < 10 μg/l ($N = 118$) did not differ by gender, smoking status, whether or not they smoked inside,

Table 1. Comparison of participants that completed the survey questionnaire with population data from the 1990 census in Hayden and Winkelman, Arizona

Characteristic	Hayden subjects $N = 282$ (%)	1990 census $N = 909$ (%)	Winkelman subjects $N = 122$ (%)	1990 census $N = 676$ (%)
Gender				
Male	138 (49)	425 (47)	63 (52)	345 (51)
Female	144 (51)	484 (53)	59 (48)	331 (49)
Age				
0–9	42 (15)	142 (16)	21 (17)	105 (16)
10–20	65 (23)	210 (23)	25 (20)	133 (20)
21–29	26 (9)	82 (9)	6 (5)	71 (11)
30–39	33 (12)	108 (11)	16 (13)	69 (10)
40–49	28 (10)	123 (14)	16 (13)	93 (13)
50–59	38 (13)	88 (10)	14 (11)	82 (11)
60–69	20 (7)	79 (9)	14 (11)	71 (11)
70+	30 (11)	77 (8)	10 (8)	52 (8)
Race/ethnicity[a]				
White Hispanic[b]	237 (84)	210 (23)	101 (83)	172 (25)
White non-Hispanic	26 (9)	116 (13)	21 (17)	144 (21)
Other	19 (7)	583 (64)	0 (0)	360 (54)

[a]Significantly different ($P < 0.001$) in both Hayden and Winkelman.

[b]More White Hispanics participated in our survey than were enumerated in the 1990 census reflecting immigration into this area.

Table 2. Comparison among participant groups based on provision of urine samples

Characteristics	$N = 224$ urine N (%)	$N = 180$ no urine N (%)
Gender		
Male	107 (48)	94 (52)
Female	117 (52)	86 (48)
Age[a]		
0–10	22 (10)	49 (27)
11–20	30 (13)	52 (29)
21–30	9 (4)	26 (15)
31–40	33 (15)	15 (8)
41–50	39 (17)	11 (6)
51–60	38 (17)	7 (4)
61–70	28 (13)	7 (4)
71+	25 (11)	13 (7)
Race/ethnicity		
White Hispanic	186 (83)	152 (84)
White non-Hispanic	29 (13)	18 (10)
Other	9 (4)	10 (6)
Smoke		
Yes	38 (17)	20 (11)
No	186 (83)	154 (89)
Smoke inside		
Yes	26 (12)	12 (7)
No	197 (88)	165 (93)

[a]Significantly different ($P < 0.001$).

Table 3. Characteristics of the 106 individuals evaluated for inorganic urinary arsenic concentrations

Characteristic	$N = 106$
Gender	57 (54)
Male	
Female	49 (46)
Age	
0–10	13 (12)
11–20	21 (20)
21–30	6 (6)
31–40	20 (19)
41–50	21 (20)
51–60	11 (10)
61+	14 (13)
Race/Ethnicity	
White Hispanic	91 (85)
White non-Hispanic	9 (8)
Other	6 (6)
Smoke	
Yes	19 (8)
No	87 (82)
Work at mine	
Yes	17 (18)
No	89 (82)
Seafood consumption in the past 3 days	
Yes	11 (10)
No	95 (90)
Consumption of home grown fruits and vegetables	
Yes	20 (19)
No	86 (81)
Water source	
Tap	43 (41)
Other source	63 (59)

race/ethnicity, occupation (working at the mine), seafood consumption in the last 3 days, consumption of local fruits and vegetables, or source of drinking water. More participants with total urinary arsenic $\geq 10\,\mu g/l$ were under the age of 30 compared to participants with total urinary arsenic values $< 10\,\mu g/l$. By design, data regarding smoking, occupation, seafood consumption, consumption of local fruits and vegetables, and source of drinking water were not collected from all 404 individuals in an attempt to minimize participant burden. A comparison between all 404 participants and the 106 with inorganic urinary arsenic measurements is therefore not possible. Of the 122 primary respondents agreeing to provide descriptive data about their home, 85 (70%) provided a dust sample. Characteristics of the homes with dust samples did not differ significantly from homes without dust samples by age of the home, means of cooling/heating the home, construction in the past 6 months, family income, whether the home was a one-family home or a mobile home, and the number of people who lived in the home.

Concentrations of arsenic from the environmental and biological samples (total urinary arsenic, inorganic urinary arsenic, and arsenic in floor dust) are presented in Table 4. There were no significant differences between Hayden and Winkelman in mean urinary arsenic concentrations for total arsenic ($P = 0.122$) or inorganic arsenic ($P = 0.783$). However, Hayden, the town closest to the smelter, had significantly higher concentrations of arsenic in floor dust than Winkelman ($P = 0.004$).

Two of the homes in Hayden had vacuum dust concentrations considerably above the others (192 and $177\,\mu g/g$). Both these homes were built before 1960 and residents had performed construction in the last 6 months. Residents of the homes with elevated house dust arsenic did not have elevated total or inorganic urinary arsenic measurements. There was one case of elevated inorganic urinary arsenic in a 7-year-old child ($47\,\mu g/l$). There had been no recent construction in the home and, unfortunately, dust measurements were not available for this household.

For the inorganic urinary arsenic model, terms that were considered for the model in bivariate analysis included race/ethnicity, personal smoking, consumption of home grown fruits and vegetables, employment at the mine, and water

Table 4. Concentrations of arsenic in urine and house dust by town

	N	Mean	Range	25th	50th	75th	90th
Hayden							
Total urinary arsenic (μg/l)	147	14.4 ± 14.3	1.3–94.7	5.7	10.5	16.4	29.4
Inorganic urinary arsenic (μg/l)	74	12.6 ± 7.7	2.4–47.1	7.2	10.8	14.2	21.1
Total house dust arsenic (μg/g)	57	70.1 ± 38.3	25–192	25	72		114.2
Winkelman							
Total urinary arsenic (μg/l)	77	12.3 ± 15.0	1.3–114.3	4.8	8.7	13.2	24.3
Inorganic urinary arsenic (μg/l)	32	11.7 ± 3.5	4.9–19.7	9.9	11.8	13.1	15.5
Total house dust arsenic (μg/g)	28	46.7 ± 29.8	25–130	25	25		88
Hayden and Winkelman							
Total urinary arsenic (μg/l)	224	13.6 ± 14.6	1.3–114.3	5.4	9.6	16.1	27.5
Inorganic urinary arsenic (μg/l)	106	12.3 ± 6.7	2.4–47.1	7.8	11.4	14	19.7
Total house dust arsenic (μg/g)	85	62.4 ± 37.2	25–192	25	62	86	108

Table 5. Regression coefficients of a predictive model for log inorganic urinary arsenic measurements using the general estimating equation

Variable	Coefficient ± SE	*P*-value	95% CI
As in house dust	0.0006 ± 0.001	0.627	[−0.002, 0.003]
Age	−0.003 ± 0.002	0.221	[−0.007, 0.002]
Gender	−0.013 ± 0.087	0.884	[−0.183, 0.157]
Seafood in past 72 h	0.247 ± 0.102	0.016	[0.047, 0.447]
Personal smoking	0.623 ± 0.120	0.000	[0.387, 0.860]
Constant	2.17 ± 0.150	0.000	[1.87, 2.46]

source. Terms that were significant bivariately at the $P \leq 0.10$ level were personal smoking and consumption of home grown fruits and vegetables. Age was included in the model because previous work has shown children to have higher urinary arsenic concentrations (Pollisar et al., 1990). In all 74 inorganic urinary arsenic measurements from 47 homes were used for this analysis, with a mean of two urinary arsenic measurements per household with a dust measurement. After adjusting for age, gender, seafood consumption, and personal smoking, there was no significant association between arsenic in floor dust and inorganic urinary arsenic concentrations (Table 5). Both seafood consumption in the past 3 days and personal smoking were significantly related to inorganic urinary arsenic measurements. Those that consumed seafood in the past 3 days had significantly higher inorganic urinary arsenic measurements than those that did not (coefficient = 0.247, 95% CI 0.047, 0.447) as did personal smokers (coefficient = 0.623, 95% CI 0.387, 0.860). Results did not change when those that work at the mine ($N = 17$) were excluded from the analysis.

Discussion

The results of this study demonstrated that homes in Hayden, the town closer to the smelter, had significantly higher levels of arsenic in house dust than homes further away in Winkelman. Several studies have documented higher arsenic levels in environmental media in homes around current and former copper smelters relative to homes in towns where there is no arsenic-emitting industry. Pollisar et al. (1990) measured arsenic in vacuum bag contents in homes near a smelter in Tacoma, Washington. They reported a median value for arsenic of 375.3 parts per million (ppm) in dust of the homes closest to the smelter (median distance 483 m) and 30.5 ppm in homes further from the smelter (median distance 10,100 m). Milham and Strong (1974) also found a decreasing level of arsenic in vacuum cleaner dust with increasing distance downwind from a copper smelter. Hartwell et al. (1983) evaluated homes in Ajo, Arizona and Anoconda, Montana and reported higher levels of arsenic in the dust of homes closest to the smelters. Diaz-Barriga et al. (1997) measured levels of arsenic in house dust in three sectors (within 600, 600–1200, and 1200–1800 m) of the town of Anapra adjacent to the copper smelter in El Paso, Texas. They reported no statistically significant association between distance lived from the smelter and arsenic in house dust. However, the mean arsenic level in the dust of homes closest to the smelter (38.7 ppm) was higher than in homes furthest away (18.7 ppm). Binder et al. (1987) found significantly higher levels of arsenic in homes very close to a smelter (mean = 264 ppm) compared to homes further away (range 4–62 ppm) in Anaconda, Montana. The consistency of these findings indicates that homes nearer smelters are more heavily contaminated with arsenic.

Despite higher levels of arsenic in house dust in Hayden, total and inorganic urinary arsenic concentrations did not differ significantly between the residents of Hayden and Winkelman in this survey. Although the results of our study indicated a lack of association between house dust and inorganic urinary arsenic in an adult population, there is evidence from other studies that children living near smelters are more heavily exposed to arsenic. Milham and Strong

(1974) reported elevated urinary arsenic measurements among third and fourth grade students in a school about 300 yards from the smelter in Tacoma, Washington compared to the same-age children in a school about 8 miles from the smelter. Almost 20 years later, Pollisar et al. (1990) conducted an extensive survey in the same area to assess pathways of exposure to arsenic from the smelter. They found that urinary arsenic concentrations did not differ greatly between residents living near the smelter (median urinary As = 11.4 parts per billion (ppb)) and those living further away (median urinary arsenic = 9.1 ppb). Children 0–6 years of age living within one-half mile of the smelter had elevated urinary arsenic levels compared to other children in the study. Hartwell et al. (1983) also reported higher urinary arsenic concentrations in children living near the smelter in Anaconda, Montana compared with children living further away. Not enough children in Hayden and Winkelman, Arizona provided urine to evaluate the association between house dust arsenic and inorganic urinary arsenic concentrations.

In an attempt to understand the route of exposure for the population in Tacoma, Washington, Pollisar et al. (1990) evaluated environmental media predictive of urinary arsenic levels in both the total and child population. In the total population, they found, after adjusting for gender, age, and fish consumption, that only indoor coarse air (2.5–10.0 μm aerodynamic diameter), outdoor coarse air (2.5–10.0 μm aerodynamic diameter), and outdoor fine air ($\leq$2.5 μm aerodynamic diameter) were significantly associated with inorganic urinary arsenic concentrations. Indoor fine air ($\leq$2.5 μm aerodynamic diameter) and settled house dust were not significantly associated with inorganic urinary arsenic concentrations.

Results of the Hayden and Winkelman survey indicate that in this population there is no association between house dust arsenic and inorganic urinary arsenic measurements. Although age and inorganic urinary arsenic measurements were inversely related, the coefficient for age was not significant, probably indicative of the small number of children that participated in this survey. Consumption of seafood within the past 3 days was significantly associated with inorganic urinary arsenic. This finding is consistent with the results of Pollisar et al. (1990), who also reported higher inorganic urinary arsenic concentrations for seafood consumers. Cigarette smokers had significantly higher inorganic urinary arsenic measurements than non-smokers, which to our knowledge has not been previously reported in epidemiological studies. According to a report published in 1934, six different brands of cigarettes showed a range of 7.5–26.4 parts arsenic trioxide per million (mean 13.9 ppm). After 1950, pressure from federal agencies discouraging the use of arsenical pesticides for control of tobacco hornworms apparently reduced the amount of arsenic in cigarettes (CDC, 1964). A report published in 1950 analyzed 17 brands of cigarettes and reported an average of 6.2 ppm arsenic trioxide per cigarette (CDC, 1964). A 1984 Environmental Protection Agency Report indicated that cigarettes have, on average, 1.5 μg of arsenic per cigarette (ATSDR, 1993). The significant association between seafood, cigarettes, and inorganic urinary arsenic concentrations implies that variables at the individual level in addition to arsenic in environmental media must be considered when assessing arsenic exposure.

The use of urine as a biomarker of exposure to arsenic is well documented. The Agency for Toxic Substances and Disease Registry reports that arsenic in urine is the most reliable way to detect exposure within the past few days (ATSDR, 1993). Crecelius (1977) reports a biologic half-life for arsenic of 30–60 h. Much of the work done on the excretion of arsenic in urine is in copper mine and smelter employees, where concentrations of arsenic trioxide are generally higher than in the ambient environment. Additionally, samples are collected immediately after the exposure has occurred in the workplace. Both Pinto et al. (1976) and Smith et al. (1977) report significant correlations between air arsenic trioxide levels and excretion of arsenic in urine.

One of the limitations of this study arises from the unknown time course of potential exposures and urinary arsenic collection. Considering the short half-life of arsenic in the body and the unknown association between chronic low-level environmental exposures and excretion of arsenic in urine, some authors have looked at hair as a possible indicator of arsenic exposure. Hartwell et al. (1983) found significant correlations between arsenic in hair and distance lived from the smelter and house dust arsenic and hair. In the same population, he reported no correlation between distance lived from the smelter and urine nor house dust and urine. The degree to which arsenic in hair reflects body burden or even urinary arsenic is unclear. For environmental exposure assessment studies, however, knowing to what extent hair reflects body burden of arsenic would be useful.

Another of the limitations in this study is the power to detect an association between inorganic urinary arsenic and arsenic in house dust. As is frequently the case in many environmental epidemiologic investigations, participants are not willing to provide all of the data requested. In the case of this study, many of the participants that provided dust were not willing to provide urine. Therefore, in the analysis of the data, it becomes very difficult to relate environmental samples taken from participants' homes to biological markers of exposure. Every attempt was made in the analysis of the data collected in this study to use all of the data points. The GEE was used to address the panel nature of this data, that is, multiple urine samples from a household where one dust sample was collected. This enabled us to use all of the urine measurements from a single household. Frequently, in these types of studies, a primary respondent is used to represent the household. Linear regression is then used to relate arsenic in

urine to arsenic in environmental media. The utility of using a primary respondent to represent the entire household is questionable. Nevertheless, the power of this study to detect an association between inorganic urinary arsenic and house dust was limited by the number of households from which we had both types of data.

In conclusion, in the residents of Hayden and Winkelman, Arizona, arsenic in house dust is not significantly associated with inorganic urinary arsenic concentrations. Other studies indicate that children living closer to smelters have higher body burdens of arsenic as evidenced by elevated urinary arsenic measurements. Not enough children provided urine in this survey to conduct an analysis in the child population. Recent ingestion of seafood and cigarette smoking were associated with increased levels of inorganic urinary arsenic, and these variables should be included in population surveys evaluating arsenic exposure.

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