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HOUSE DUST AND INORGANIC URINARY ARSENIC IN TWO ARIZONA MINING TOWNS

by

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ABSTRACT

Residents of copper mining and smelting towns may have increased risk of arsenic exposure from elevated arsenic contained in environmental media. To determine the relationship 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; eighty-five 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 10μg/L (N=106). The generalized estimating equation was used to determine the relationship between urinary and house dust arsenic concentrations, allowing adjustment for the correlation of measurements obtained from the same home. Seafood consumption during the past three 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.

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(1). Exposure to arsenic through drinking water at levels exceeding 30μg/L is associated with cutaneous effects including skin cancer(2-4). Studies of skin cancer and arsenic exposure in the United States show little or no association at levels around 10μg/L (5). In addition, many studies report significant associations between arsenic in drinking water and several other cancer types including lung, bladder, and liver(6-10). Arsenic is associated with lung cancer in populations with occupational exposure to inhaled arsenic (11, 12).

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)(1). The presence of high arsenic concentrations in environmental and biological media near copper smelters is well documented(13-18). Several studies evaluate exposure to arsenic and resultant health effects in populations living near copper smelters(19-25). These health effects include lung cancer, low birth weight, spontaneous abortion, and congenital malformations(26).

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 imply that inorganic arsenic is absorbed across the gastrointestinal tract, as evidenced by the fact that 60-80% of daily oral intake of these compounds is excreted in urine(27-29). Arsenic exposure via inhalation is more complex. During inhalation, arsenic in the air is deposited on the lung surface. Arsenic deposited in the lung can be both directly absorbed and carried by the mucociliary escalator back into the throat and then swallowed. Arsenic is then absorbed through the gastrointestinal tract. No evidence exists for the uptake of inorganic arsenic through dermal absorption (1). In an attempt to separate environmental exposures from dietary exposures, urine is often speciated, and the concentration of inorganic arsenic, MMA and DMA are used as biomarkers of exposure.

Understanding the potential contribution of 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

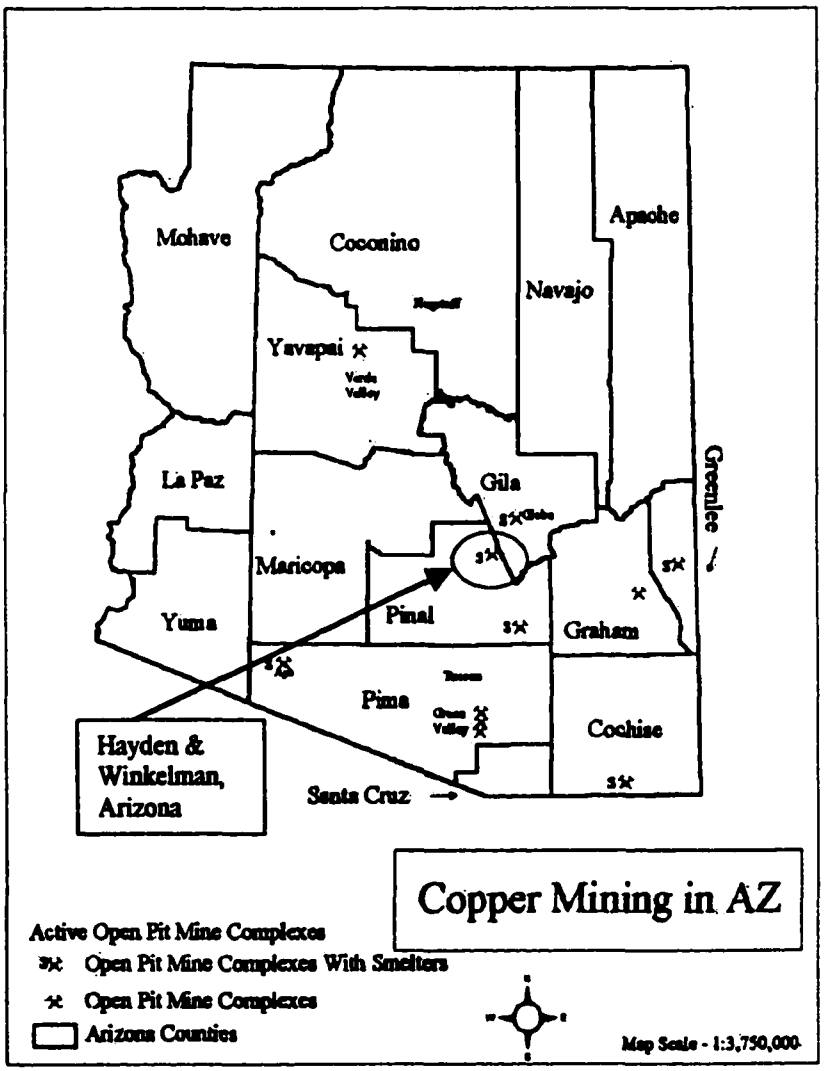
We conducted a door to door survey of all residents in Hayden and Winkelman, Arizona (Figure 1) from June to October 1999. Hayden and Winkelman have a combined population of 1585 people according to 1990 census data. Prior to the survey, a campaign to educate residents about lead and arsenic and possible 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.

Every residence in the two towns was approached to participate, and homes where there was no initial response were approached a maximum of 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 (Appendix A). Questionnaire data at the household level included questions about the age of the home,

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



primary water source for individuals in the home, any recent construction, and how the home was cooled and heated (Appendix B).

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 –20 degrees 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 (HG-AAS) at Departmento de Farmacologia y toxicologia, Seccion de Toxicologia Ambiental. Those with total arsenic concentrations above 10 ug/L (N=106) were analyzed for As+5, MMA, and DMA. Originally, 108 samples had total urinary arsenic above 10μg/L, however, two of the samples were lost during processing.

Each dust sample was sieved to three sizes, [coarse (gravel, ≥2mm); medium (sand, >2mm and ≤62.5μm); fine, (clay & silt < 62.5μm)]. One gram of the finest fraction (<62.5μm) 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 seconds on each of three isotopes (^{109}Cd, ^{55}Fe, & ^{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 (MDL) for arsenic in this application was 50μg/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 2.25μg/L. Analysis for inorganic arsenic was completed in two phases, with different LOD's. 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 30μg/L, then the LOD for inorganic arsenic was 0.5 μg/L. Urine samples with total arsenic concentrations below 10μg/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 25μg/g for all statistical analysis (N=32).

Data were analyzed for the relationship between dust and inorganic urinary arsenic using STATA 6.0 (College Station, Texas). Generalized estimating equations (GEE) were 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 hours were forced into the model due to 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(30). 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 ($p < .05$).

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 $\geq 10\mu g/L$ and were therefore speciated are presented in Table 3. Subjects with total urinary arsenic $\geq 10\mu g/L$ (N=106) and $<10\mu g/L$ (N=118) did not differ by gender, smoking status, whether or not they smoked inside, race/ethnicity, occupation (working at mine), seafood consumption in last three 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 $<10\mu g/L$.

Table 1. Comparison of participants that completed the survey questionnaire with population data from the 1990 census in Hayden & 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*					
White Hispanic†	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)

*Significantly different ($p<.001$) in both Hayden and Winkelman

† More White Hispanics participated in our survey then 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=180 no urine
		N (%)	N (%)
Gender	Male	107 (48)	94 (52)
	Female	117 (52)	86 (48)
Age*	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)

*Significantly different ($p<.001$)

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

Characteristic		N=106
Gender	Male	57 (54)
	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 past three days	Yes	11 (10)
	No	95 (90)
Consumption of homegrown fruits and vegetables	Yes	20 (19)
	No	86 (21)
Water source	Tap	43 (41)
	Other source	63 (59)

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 six months, family income, whether the home was a one family home or mobile home, and the number of people that 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 ($p=.122$) or inorganic arsenic ($p=.783$). However, Hayden, the town closest to the smelter, had significantly higher concentrations of arsenic in floor dust than did Winkelman ($p=.004$).

Two of the homes in Hayden had vacuum dust concentrations considerably above the others (192 μg/g and 177 μg/g). Both of these homes were built before 1960 and residents had performed construction in the last six 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 seven-year old child (47μg/g). 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, personal smoking, consumption of homegrown fruits and vegetables, employment at mine, and water source. Terms that were significant

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

Hayden		Mean	Range	25th	50th	75th	90th
Total Urinary Arsenic (µg/L)	N=147	14.4±14.3	1.3-94.7	5.7	10.5	16.4	29.4
Inorganic Urinary Arsenic (µg/L)	N= 74	12.6±7.7	2.4-47.1	7.2	10.8	14.2	21.1
Total Dust Arsenic (µg/g)	N= 57	70.1±38.3	25-192	25	72		114.2

Winkelman		Mean	Range	25th	50th	75th	90th
Total Urinary Arsenic (µg/L)	N=77	12.3±15.0	1.3-114.3	4.8	8.7	13.2	24.3
Inorganic Urinary Arsenic (µg/L)	N=32	11.7±3.5	4.9-19.7	9.9	11.8	13.1	15.5
Total Dust Arsenic (µg/g)	N=28	46.7±29.8	25-130	25	25		88

Hayden & Winkelman		Mean	Range	25th	50th	75th	90th
Total Urinary Arsenic (µg/L)	N=224	13.6±14.6	1.3-114.3	5.4	9.6	16.1	27.5
Inorganic Urinary Arsenic (µg/L)	N=106	12.3±6.7	2.4-47.1	7.8	11.4	14	19.7
Total Dust Arsenic (µg/g)	N=85	62.4±37.2	25-192	25	62	86	108

bivariately at the $p \leq .10$ level were personal smoking and consumption of homegrown fruits and vegetables. Age was included in the model because previous work has shown children to have higher urinary arsenic concentrations(14). Seventy-four 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 three days and personal smoking were significantly related to inorganic urinary arsenic measurements. Those that consumed seafood in the past three days had significantly higher inorganic urinary arsenic measurements than did those that did not (coefficient=.247, 95% CI .047,.447) as did personal smokers (coefficient=.623, 95% CI .387, .860). Results did not change when those that work at the mine (N=17) were excluded from the analysis.

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	.0006±.001	.627	[-.002, .003]
Age	-.003±.002	.221	[-.007, .002]
Gender	-.013±.087	.884	[-.183, .157]
Seafood in past 72 hours	.247±.102	.016	[.047, .447]
Personal smoking	.623±.120	.000	[.387, .860]
Constant	2.17±.150	.000	[1.87, 2.46]

DISCUSSION

Results of this study demonstrated that homes in Hayden, the town closer to the smelter, had significantly higher level of arsenic in house dust than did 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. O'Rourke et al.(31) reported a median house dust value of 6.7 μg/g (range 0.3-50.6) for a representative sample of homes throughout Arizona, including mining and non-mining towns. Pollisar et al.(14) 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 milliion (ppm) in dust of the homes closest to the smelter (median distance 483 meters) and median 30.5 ppm in homes further from the smelter (median distance 10100 meters). Milham et al.(13) also found a decreasing level of arsenic in vacuum cleaner dust with increasing distance from a copper smelter when he measured homes downwind of the smelter. Hartwell et al.(15) 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 (32) measured levels of arsenic in house dust in three sectors (within 600 m, 600-1200 m, and 1200-1800m) of the town of Anapra which is 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 dust of homes closest to the smelter (38.7 ppm) was higher than in homes furthest away (18.7

homes very close to a smelter (mean= 264ppm) compared to homes further away (range 4-62 ppm) in Anaconda, Montana. The consistency of these findings indicates homes nearer smelters are more heavily contaminated with arsenic.

Although there may be an increased risk of exposure to arsenic through house dust for residents living closest to the smelters, urinary biomarkers only show such an association for children. Milham et al.(13) reported elevated urinary arsenic measurements amongst 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 eight miles from the smelter. Almost twenty years later, Pollisar et al.(14) conducted an extensive survey in the same area to assess pathways of exposure to arsenic from the smelter. He found 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.(15) also reported higher urinary arsenic concentrations in children living near the smelter in Anaconda, Montana compared with children living further away. Total and inorganic urinary arsenic concentrations did not differ between the residents of Hayden and Winkelman in this survey. However, few children were sampled in this survey. Only four children under the age of six provided urine, and their total and inorganic urinary arsenic measurements are not significantly different than the rest of the population.

In an attempt to understand the route of exposure for the population in Tacoma, Washington, Pollisar et al.(14) 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, only indoor coarse air (2.5-10.0μm aerodynamic diameter), outdoor coarse air (2.5-10.0μm aerodynamic diameter), and outdoor fine air (≤2.5μm aerodynamic diameter) were significantly associated with inorganic urinary arsenic concentrations. Indoor fine air (≤2.5μm aerodynamic diameter) and settled house dust were not significantly associated with inorganic urinary arsenic concentrations.

The use of urine as a biomarker of exposure to arsenic is well documented, however, its usefulness as a marker of chronic, low level exposure is questionable. 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(1). Crecelius et al.(28) report a biologic half-life for arsenic of 30-60 hours. 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. and Smith et al.(33, 34) report significant correlations between air arsenic trioxide levels and excretion of arsenic in urine.

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.(15) 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 unknown. The findings of Hartwell et al.(15) warrant further investigation of hair as a biomarker to chronic low-level arsenic exposure.

CONCLUSION

In the towns of Hayden and Winkelman, Arizona, there was no statistical association between inorganic urinary arsenic concentrations and house dust arsenic measurements in the general population. Age and inorganic urinary arsenic measurements are inversely related, a finding consistent in the literature showing children to have higher body burdens of arsenic. Our study indicates that residents who consumed seafood had significantly higher inorganic urinary arsenic concentrations. This finding is consistent with the results of Pollisar et al.(14) who also reported higher inorganic urinary arsenic concentrations for seafood consumers. For adults in these two towns, arsenic exposure through house dust does not appear to contribute significantly to total absorbed dose. Not enough children were available to analyze the association between house dust and urinary arsenic in this group.

APPENDIX A

URINE ARSENIC STUDY:

USE A SEPARATE SHEET FOR EACH PERSON WHO PROVIDES A URINE SAMPLE.

How long has NAME lived in this home? _____
How long has NAME lived in this community? _____
List prior residences, if any:

Dates	Address	Town/Country

What is NAME's present occupation? _____
Does NAME work at a mine or smelter? ☐ yes ☐ no
When was the last time (in days) NAME ate seafood? _____
This includes seafood such as tuna, tuna salad, shrimp, fish-soup, fish tacos, etc.
When is the last time (in days) NAME ate mushrooms? _____
This includes mushrooms in soups, salads, stews, etc.
Does NAME eat: ☐ locally or ☐ homegrown fruits or vegetables?

APPENDIX B

Descriptive Questionnaire Supplement:
Hayden-Winkelman Study

ADMINISTER THESE QUESTIONS TO ALL HOUSES: HHID_____

1. When was your house built?
   ()1990 – 1999 --- ()1980 – 1989 --- ()1970 – 1979 --- ()1960 – 1969 --- ()Prior to 1960

2. What is your primary source of water?
   [ ]Municipal/City [ ] Private Well [ ] Other: _____.

3. Do you have a separate source of water that you use for drinking or cooking?
   [ ] No. Tap = Drinking
   [ ] Yes. Check all that apply:
   [ ] buy bottled water for drinking and cooking (circle one or both).
   [ ] bottled water delivered by company for drinking and cooking (circle one or both).
   [ ] water from vending machine for drinking and cooking (circle one or both).
   [ ] In-line supply filter near or at tap for drinking and cooking (circle one or both).
   Type/Brand of filter: _____.
   [ ] filtered tap water (filtering device not fixed – e.g. Brita Carafe) for drinking and cooking (circle one or both). Type/Brand of Filter: _____.
   [ ] water from refrigerator dispenser for drinking and cooking (circle one or both).
   When was filter last replaced: _____.

4. How do you cool your home? (Check all that apply)
   [ ] Evaporative (swamp) cooling. () Central Unit or () Specific room(s) only.
   [ ] Air conditioning unit. () Central Unit or () Specific room(s) only.
   [ ] Fan(s) or other means: describe _____.

5. How do you heat your home? (Check all - 20 - that apply)
   [ ] Electric Heater/Furnace: () Central Unit or () Specific room(s) only.
   [ ] Gas Heater/Furnace: () Central Unit or () Specific room(s) only.
   [ ] Some other means: describe _____.

6. In the last six months, have any of the following been performed in your this home? (Fill in one bubble)

Adding a room	[ ] Yes	[ ] No
Putting up or taking down a wall	[ ] Yes	[ ] No
Replacing windows	[ ] Yes	[ ] No
Refinishing floors	[ ] Yes	[ ] No
Exterior painting	[ ] Yes	[ ] No
Interior painting	[ ] Yes	[ ] No

7. Family income is often used in scientific studies to compare groups of people who are similar. We do some analysis of the data using these groups. Please remember that all the answers you provide are held in strict confidence. Approximately what is the gross annual income for all family members in this household?
   ()< $9,999 - ()$10,000-19,999 - ()$20,000-29,999 - ()$30,000-39,999 - ()$40,000-49,999
   ()$50,000-74,999 - ()$75,000-99,999 - ()$100,000 or more - ()Don't know- ()Refuse

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