Exponent®

Middleport Environmental Exposure Investigation

Prepared for

FMC Corporation
Philadelphia, Pennsylvania

Exponent®

Middleport Environmental Exposure Investigation

Prepared for

FMC Corporation
Corporate Administrative Offices
1735 Market Street
Philadelphia, PA 19103

Prepared by

Exponent
15375 SE 30th Place, Suite 250
Bellevue, WA 98007

August 2004

Doc. no. 8602390.001 0901 0704 JT16

Contents

List of Figures

List of Tables

Acronyms and Abbreviations

ANOVA	analysis of variance
AsB	arsenobetaine
AsC	arsenocholine
ATSDR	Agency for Toxic Substances and Disease Registry
CAP	Community Advisory Panel
CDC	Centers for Disease Control and Prevention
DMA	dimethylarsinic acid
EPA	U.S. Environmental Protection Agency
FMC	FMC Corporation
MMA	monomethylarsonic acid
NYSDEC	New York State Department of Environmental Conservation
NYSDOH	New York State Department of Health
SOMA	Sandler Occupational Medicine Associates, Inc.
TMAO	trimethylarsine oxide

Executive Summary

At the request of the Middleport Community Advisory Panel, an environmental exposure investigation was conducted by Exponent during the summer of 2003 to provide information on exposure to arsenic in soil for the Village of Middleport, New York. The study was funded by FMC Corporation (FMC). The results of the study were reviewed by an independent panel of scientific experts from universities, health institutes, and the Centers for Disease Control and Prevention (CDC). This voluntary biomonitoring study examined levels of arsenic in urine of young children less than age 7, the target population of most concern for soil exposure. Older individuals were also allowed to participate at their request. In addition, arsenic levels in soil, house dust, and garden produce were measured, and information was obtained from participants via questionnaires on potential arsenic sources and factors affecting exposure.

Study Design

This investigation 1) evaluated urinary arsenic levels of young children and other participants in the Village of Middleport; 2) examined the correlation between biomarkers of arsenic exposure (i.e., urinary arsenic levels) and arsenic in soil and in house dust; 3) assessed indirect indicators of arsenic exposure based on questionnaire responses; and 4) provided study participants with individual biomarker and environmental arsenic levels and the community of Middleport with group-level information on biomarker levels. The study area was larger than the Village of Middleport boundaries and was bounded by Carmen Road, Pearson Road, Mountain Road, and County Line.

The primary analysis focused on young children less than age 7 who are more likely to be exposed to soil than children of older ages. Additional analyses were expanded to include children less than age 13 and to all ages. This study differed from the 1987 New York State Department of Health biomonitoring study of Middleport school children by focusing on preschool children during the summer months when exposure to soil would be highest, examining the effect of environmental arsenic levels and other potential sources and indicators of exposure, and analyzing urine samples for the specific forms of arsenic in the urine that would result from ingestion of arsenic in soil. A within-community study design, in this case, is more informative for examining exposure to arsenic in soil than is a comparison of group mean urinary levels between Middleport and another community.

Participation

To estimate the population size and recruit children within the target age range, a door-to-door census was conducted of every house within the study area. Nearly half of the children within the target age range (77 out of an estimated 164 young children in the study area) and 362 older participants provided urine samples and survey information. The total population size within the study area was estimated to be 1,930. Toenail sampling was offered with the instruction that feet must not be exposed to soil or dust for a month prior to sampling to avoid surface

contamination of nails. Of the 67 (out of 84 submitted) toenail samples with sufficient mass for arsenic analysis, none were from young children.

Fewer households elected to have their soil or house dust sampled. Unlike urinary sampling, participation in soil sampling required reporting the soil results to the New York State Department of Environmental Conservation.

Sample Collection and Analysis

Two first morning void urine samples from each participant were combined into one sample and analyzed for total arsenic forms as well as specific arsenic forms (i.e., speciated arsenic) related to ingestion of inorganic arsenic, the form present in soil. Because total arsenic can be highly affected by organic arsenic compounds found in the diet, speciated urinary arsenic levels and inorganic arsenic in urine were the primary indicators of potential excess soil arsenic exposure. A subset of samples analyzed by the primary contract laboratory was also analyzed by the CDC laboratory as a part of quality control for analyses of arsenic in urine. The results of these two laboratories for total and speciated arsenic showed reasonable agreement, given the differences in the analytical techniques and detection limits.

Soil samples were a composite of four separate locations in each yard for which residents elected to have such sampling. Additional composite samples were taken for larger yard areas (i.e., >11,000 ft^2). Separate composite samples were also taken from play areas and gardens. House dust was collected by a vacuum technique that allowed capture of fine dust particulates. Measurements were presented as arsenic concentration in dust and arsenic surface loading in mass per area. Garden vegetables were sampled at the request of participants.

Biomarker Results

Urinary arsenic results were compared to reference levels that are intended to represent the upper limit of background levels of arsenic in urine in the U.S. population, above which elevated arsenic exposure is indicated along with a need for individual follow-up. Program reference levels were based on CDC recommendations and other arsenic biomonitoring studies. Speciated urinary arsenic levels of all participants were well below the program reference level of 40 μg/L (all samples were less than 20 μg/L and averaged 4.7 μg/L). Inorganic arsenic levels in urine were also low for all participants and below the program reference level of 20 μg/L (all samples were less than 3 μg/L and averaged 0.83 μg/L). Mean levels and ranges of these urinary arsenic levels were similar for children less than age 7 and older participants. Total arsenic levels in urine were elevated (i.e., above 50 μg/L) in some samples; however, the low speciated and low inorganic arsenic levels in these individuals indicate that the elevated total arsenic levels were most likely due to dietary arsenic sources such as organic compounds from fish or other seafood. Seafood arsenic compounds are considered relatively nontoxic.

Levels of speciated and inorganic arsenic in urine of Middleport participants were also low in comparison to levels detected in urine of participants in arsenic exposures studies of other

communities and consistent with levels detected in unexposed or control populations in other studies.

Toenail arsenic data were not available for young children. Arsenic levels in toenail samples from older participants were below the CDC reference level for nails of 1 mg/kg, or ppm. The toenail data showed an association between higher toenail arsenic levels and nail discoloration, indicative of external surface contamination. Toenail arsenic concentrations were thus likely to have been increased by such contamination and were considered not to be a reliable indicator of ingested arsenic exposure.

Effect of Arsenic in Soil on Urinary Arsenic Levels

Soil arsenic exposure was assessed using several approaches. Direct indicators of arsenic exposure from soil were:

- Correlation of speciated or inorganic urinary arsenic levels with average or maximum soil arsenic levels
- Correlation of speciated or inorganic urinary arsenic levels with house dust arsenic concentration or surface loading of arsenic in house dust, and correlation of arsenic in soil with arsenic in house dust.

Analysis of whether urinary arsenic levels are elevated via uptake of arsenic from soil into vegetables was complicated by the number of different vegetable types collected, most of which were not widely grown in the community and thus had few samples; by differences in sample preparation (washed versus unwashed samples); and by the fact that only a subset of participants with vegetable samples elected to have their garden soil sampled. All vegetables had arsenic levels (less than <0.6 mg/kg) that were well below background levels of arsenic in soil. The most prevalently grown vegetables (tomatoes) had very low arsenic levels (e.g., ≤0.01 mg/kg) that were near and below the limit of detection.

Other indirect indicators of soil arsenic exposure were also examined, including:

- Correlation and association of greater concentration of speciated or inorganic arsenic in urine with various factors that increase soil exposure such as mouthing behavior, amount of time playing outdoors, eating garden vegetables, digging projects, taking food or drink outdoors, playing with outdoor pets, frequency of washing, and other behaviors or characteristics. These factors were also examined for potential confounding of the direct correlations between urinary arsenic and soil and dust arsenic levels.
- The geographic distribution of speciated urinary arsenic levels in relation to the FMC plant and areas with elevated soil arsenic levels (e.g., creeks).

These indirect indicators involved the full group of participants in the target age range and were less limited by sample size than the correlation analysis of urinary arsenic with soil (N=41) or

house dust data (N=52). The direct and indirect analyses were repeated in the expanded age group of children less than 13 years old and in the total study population. Including older age groups increased the sample size, but was considered a secondary analysis because older ages are less exposed to soil and participation rates were lower (i.e., smaller percentage of the available population participated) than for young children. It is also important to note that the older age groups were a self-selected voluntary group that was not targeted for participation.

Because of the number of correlations and associations examined, it is possible that some statistically significant results might occur due to chance. For this reason, it is important to evaluate consistency of results across various direct and indirect indicators of soil exposure.

Overall, no evidence of a relationship between urinary arsenic levels and environmental arsenic levels was found in our target population (i.e., children less than 7 years) or in the other age groups evaluated (i.e., children less than 13 years and the total study population). All measures of association between speciated urinary arsenic levels and soil or house dust arsenic levels were weak and nonsignificant. There was also no association between outdoor soil arsenic levels and house dust arsenic concentration or surface loading.

There was no consistent evidence of elevated soil arsenic exposure for the indirect indicators of soil exposure such as the geographic distribution of urinary arsenic data in relation to locations with elevated arsenic levels in the community, garden vegetable consumption, playing in or near creeks, or other behaviors that could be related to increased soil exposure. Expanding the age range to children less than 13 years or to all participants did not change these results.

The only factors showing significant associations related to higher urinary arsenic levels were visits to local fruit orchards (speciated urinary arsenic levels for all three age groupings examined; not related to Middleport soil), visits to a house undergoing renovation (speciated urinary arsenic levels for children less than 7 years only), and the presence of a digging project in the yard in the last 12 months (only inorganic urinary arsenic levels for children less than age 13 and all participants). All of these associations are based on a small number of individuals engaged in these activities whose urinary arsenic levels were still low in comparison with the reference level. Thus, the actual impact of these activities on arsenic exposure appears negligible.

The results of this study are consistent with studies at other sites involving much larger populations and ranges in arsenic soil concentrations. At these sites, the relationship of soil arsenic levels with speciated urinary arsenic levels is at best small (i.e., very small increase in urinary arsenic level with increase in soil arsenic level) and weak (i.e., most of the variation in urinary arsenic levels is not associated with soil arsenic levels).

Conclusions

No clear evidence of elevated exposure from arsenic in soil was found for participants in this investigation. The overall low urinary arsenic levels and lack of relationship between arsenic in soil and arsenic in urine indicate that sources of inorganic arsenic other than soil (likely background levels in water and diet) are the primary contributors to inorganic arsenic exposure in this community.

1 Introduction

An environmental exposure investigation was conducted at the request of the Middleport Community Advisory Panel (CAP) to provide the community and interested health agencies with information regarding residential exposure to arsenic from soil in the Village of Middleport, New York. The study involved biomonitoring of residents for arsenic exposure; environmental sampling for arsenic in soil, house dust, and garden produce to correlate with biomonitoring results; and collection of survey responses on potential arsenic sources and factors affecting exposure.

Although arsenic levels in soil throughout the Middleport community are generally low in comparison to soil levels at sites in which environmental exposure investigations (including biomonitoring) have been conducted (e.g., by the Agency for Toxic Substances and Disease Registry [ATSDR]), FMC Corporation (FMC) has agreed to fund this study due to public concerns regarding arsenic exposures in the Middleport community. The study was conducted by a contractor, Exponent, with outside review of the results by a scientific expert panel.

1.1 Site History

This summary of site history and remedial activities is based on the background section of the Resource Conservation and Recovery Act facility investigation (Conestoga-Rovers 1999) and a recent joint-agency fact sheet (U.S. EPA et al. 2003). The focus of this history is on arsenic in offsite (i.e., outside the plant property) soil related to the FMC Middleport plant. The approximately 91-acre plant property is located in the southeast corner of the Village of Middleport. Prior to manufacturing operations at the plant, the plant property and surrounding areas were used for agricultural purposes, which still occur in areas outside the village. Past agricultural use of arsenical pesticides is thus also a source of arsenic in soil in the general area.

Manufacturing at the site started about 1928 with the manufacture and/or formulation of pesticide products and spraying machines by Niagara Sprayer Company. In 1943, FMC purchased a controlling interest and acquired complete interest in the company in 1946. Activities conducted at the facility during its history included pesticide manufacturing, pesticide formulation and packaging, and research and development activities. Arsenical pesticides along with lime (i.e., calcium hydroxide, calcium oxide) were manufactured at the site between 1928 and 1974.

Environmental investigations and monitoring programs have been conducted at and around the Middleport facility since the early 1970s including soil and groundwater investigations. In 1991, FMC signed an Administrative Order on Consent with the New York State Department of Environmental Conservation (NYSDEC) and the U.S. Environmental Protection Agency (EPA) to conduct comprehensive environmental investigations related to the facility and interim corrective measures as necessary. Soil sampling and remedial activities at the site have focused on the plant site, nearby residential and school properties, and the adjacent properties, banks, and sediments of Tributary One of Jeddo Creek and Culvert 105 drainage ditch, which received

surface water flow from the plant property (Figure 1). The Niagara County Water District, which obtains its water from the Niagara River, provides drinking water to the community. The Niagara County Water District report for monitoring of drinking water contaminants in 2003 notes that arsenic was not detected and that its water met or exceeded state and federal regulations (NCWD 2004). Sampling of 48 private wells offsite in 2000 did not show evidence of arsenic contamination from the site (Conestoga-Rovers 2002).

Prior to construction of the surface water treatment plant in 1976–1977 and surface water impoundments in 1977–1978, process wastewater and storm water at the site were collected in several lagoons and discharged to Tributary One on a controlled basis to minimize impact (due to ammonia) to the receiving stream. This discharge was via a buried stormwater pipe that exited the property at the northwestern boundary and joined with the Village of Middleport storm water sewer system, which eventually discharged into Tributary One under the Francis Street Bridge. Prior to 1977, storm water runoff from a small portion of the facility also discharged to the Northern Conrail Ditches, which ran along the northern property boundary and eventually drained to Culvert 105 north of the facility. Culvert 105 runs in a northerly direction under residential properties and emerges as a surface ditch north of the Erie Canal (Barge Canal), before it discharges to Tributary One downstream of the discharge point for the Village of Middleport sewage treatment plant. These past discharges of water from the facility have been the source of elevated arsenic concentrations detected along these drainages and in sediments downstream of the facility.

Previous soil sampling of these areas show soil arsenic levels from discrete (point) sample locations in residential properties along these drainage areas that range from background levels to as high as 1,680 ppm with a mean of approximately 70 to 100 ppm (Table 1; U.S. EPA et al. 2003). By comparison, soil samples of residential properties in the nearby town of Gasport were 21 ppm and below; however, orchard properties in Gasport had levels up to 121 ppm (Table 1).

Table 1. Soil arsenic concentration data summary

Sample Location	Range (ppm)	Average (ppm)
Tributary One properties north of Francis Street to south of Barge Canal	2–1,680	109
Tributary One properties north of Barge Canal to Pearson Road	4–722	93
Culvert 105 properties along ditch north of Barge Canal	0.1–323	70
Off-site air deposition properties north of FMC plant and outside of flood zone	0.4–115	32
14 residential properties west of FMC plant	17–1,124	95
Gasport soils: residential/public property	3–21	10
Gasport soils: orchard property	3–121	33

Source: U.S. EPA et al. (2003)

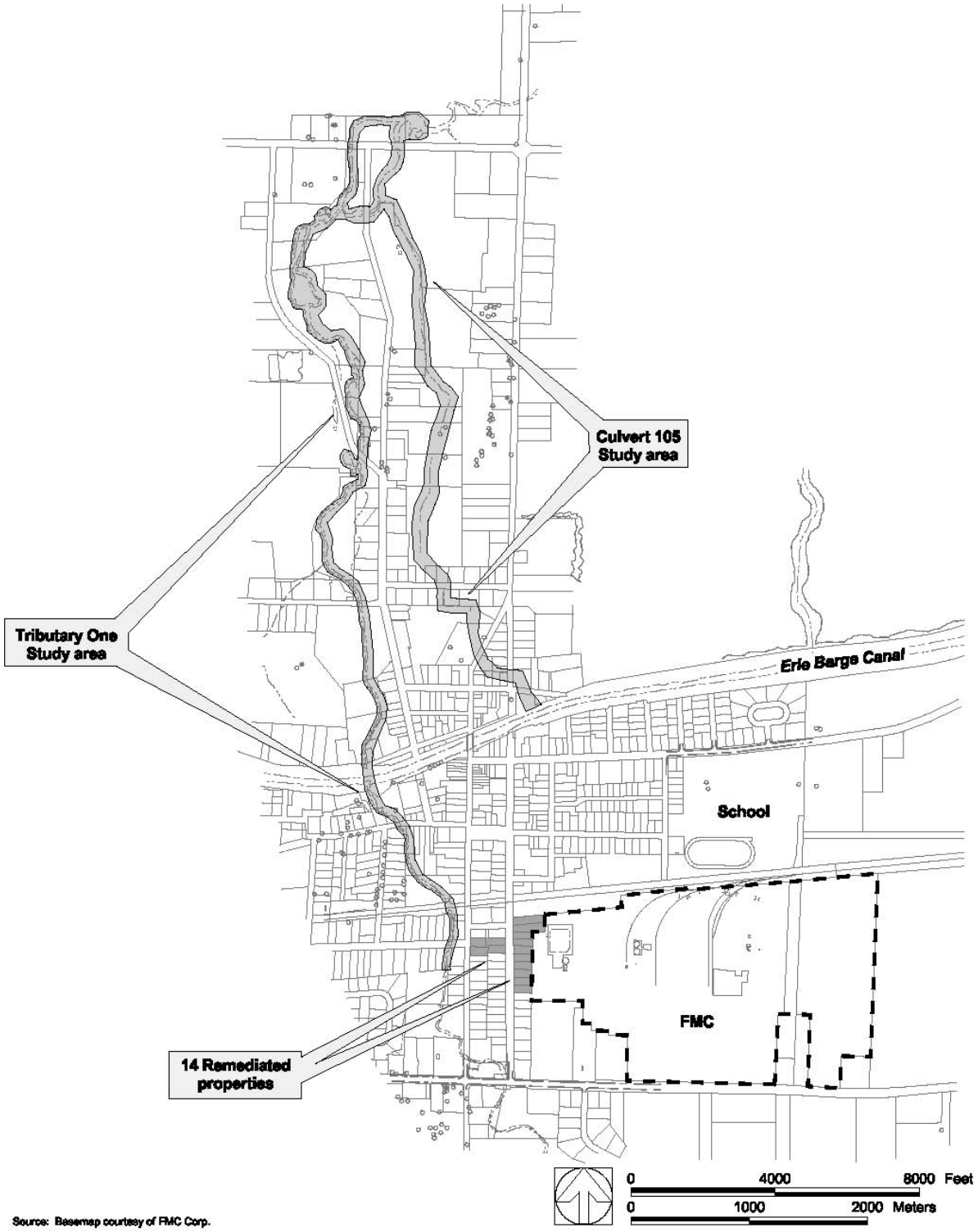


Figure 1. Tributary One and Culvert 105 sampling areas (2004) and remediated properties (summer and fall 2003)

The FMC plant also had air emission discharge permits for the unit operations (i.e., dryers, evaporator, dust collectors, storage tanks, process thermal oxidizer, boilers) associated with manufacturing of arsenical and other pesticides at the facility. Little information is available on air discharges or sources of air emissions at the facility prior to the 1970s. The available permits and other information were used to model historical releases from arsenic-based product manufacturing at the facility. Air dispersion modeling suggested general areas of past arsenic deposition to the west, north, and northeast of the FMC property. Properties sampled in this area (primarily north of the plant property and south of the Barge Canal) had surface soil arsenic levels of 0.4 to 115 ppm (Table 1; U.S. EPA et al. 2003).

In addition to actions on the FMC plant property, actions related to arsenic in soil offsite included remediation of the northern ditches bordering Royalton-Hartland School (in 1987–1988) and the school's south bleacher area (in 1996) and football field and track (in 1999–2000). In the summer of 2003, soil containing arsenic levels above 20 ppm was remediated on 14 properties that are located west of the plant site in the historical water drainage pathway (Figure 1). Ten properties are on the western border of the plant site and a sewer line that historically carried storm and other water from the plant site traversed four others. This sewer line and surrounding soils were removed as well. Residents of these properties who elected to participate in the 2003 biomonitoring study provided urinary samples before this soil remediation took place. The average and maximum surface soil samples on these properties were similar to those sampled along Tributary One (Table 1; U.S. EPA et al. 2003).

The New York State Department of Health (NYSDOH) conducted a cancer incidence study that examined cancer registry data from 1976 through 1984 for the Village of Middleport (NYSDOH 1987a). The study concluded that the cancer incidence for 17–19 different organs or for all cancers combined in men, women, or children was similar to the expected incidence for areas of similar population density for 1978–1982 in New York. A more recent study of the registry data is not available at this time.

NYSDOH also conducted a biomonitoring study in May and June of 1987 with students from Royalton-Hartland school to evaluate whether students had increased arsenic exposure from the arsenic soil levels detected on school property (NYSDOH 1987b). Samples were also collected from students from the Albany area (East Greenbush Central School District) as a control group. This voluntary study focused on students with the greatest potential for soil exposure: kindergarten and first-grade students and participants in high school athletics. The study included 104 kindergarten and first-grade students from Middleport and 84 from East Greenbush, and 30 athletes from Middleport and 15 from East Greenbush. No significant differences were found between the Middleport students and those from East Greenbush; however, only total arsenic levels were measured in urine, which can be confounded by dietary organic arsenic sources. None of the total arsenic levels in urine for Middleport students exceeded 50 μg/L; whereas, five students from East Greenbush had levels exceeding 50 μg/L and were retested. Questionnaires from these five students indicated recent ingestion of seafood. Thus, because urinary arsenic levels below 50 μg/L are likely confounded by dietary arsenic as well, the ability of this study to detect differences in arsenic exposure from soil was limited.

1.2 Purpose and Study Design

The purpose of this investigation is to 1) quantify the correlation between arsenic in soil and arsenic in biomarkers (e.g., urine), and 2) provide Middleport residents with information on their biomarker levels of arsenic in relation to reference levels used by health agencies or in other biomonitoring studies to denote the upper limit of background levels for the general population, above which individual follow-up is warranted. To minimize dietary influences of non-soil-related arsenic forms in urine, urine samples were analyzed for the type of urinary metabolites of ingested arsenic related to the form of arsenic in soil (i.e., inorganic arsenic). Toenail samples were also analyzed at participants' request, but in the end, toenail analysis was considered to be an unreliable method for assessing internal arsenic exposure because of problems encountered with external surface contamination of nails by contact with soil and dust.

This study is a cross-sectional biomarker-based evaluation of Middleport residents. Recruitment of the study population focused on children less than 7 years old (i.e., less than 84 months and had not reached their 7th birthday as of August 1, 2003, the commencement of the biomonitoring phase of the study), who have a greater likelihood of exposure to soil. However, older children and adults were also allowed to participate if they desired. Soil, house dust, and vegetables (if requested) from participants' residences were collected and analyzed for arsenic.

The relationship between environmental arsenic concentrations and selected biomarkers was examined to evaluate the contribution of soil arsenic to the overall arsenic exposure. Because the study focused on Middleport residents exposed to a range of arsenic soil concentrations, an unexposed reference population is unnecessary and potentially problematic because of the difficulty in controlling for differences between communities in background sources of arsenic and other factors that might affect exposure. A cross-sectional evaluation of exposure within a community is also more informative than a comparison of group means between two different communities. The within-community design provides information on whether urinary arsenic levels are increased by higher soil arsenic levels (i.e., strength of correlation), how much of the variation in urinary arsenic levels is due to soil arsenic exposure, and the amount of increase in urinary arsenic levels per increase in soil arsenic level.

1.3 Study Elements

The overall program included the following elements, which were conducted in 2003 to 2004 (Figure 2):

- **Study Communication, Census, and Recruitment—**Presentation of the program to the community and census and recruitment of participants
- **Arsenic Biomonitoring—**Biomarker (urine, toenail) sample collection for arsenic analysis
- **Environmental Assessment**—Soil, house dust, and garden produce collection at participants' residences for arsenic analysis

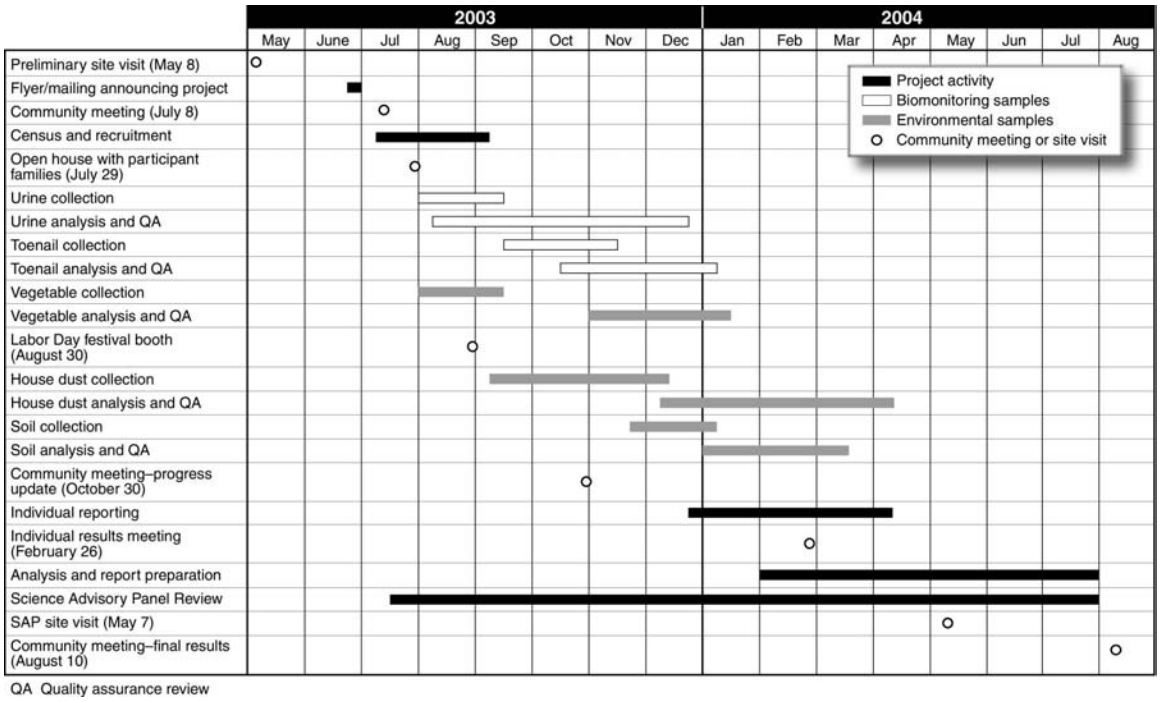


Figure 2. Time line for study activities

- **Reporting of Individual Results and Case Follow-up—**Interpretation and reporting of individual biomarker and environmental results and follow-up with participants, as appropriate

- **Analysis of Results—**Statistical analysis of biomonitoring and environmental data to examine the relationship between arsenic in soil and overall arsenic exposure.

1.4 Scope of Study

The term biomonitoring as used in this study refers to testing of a population for clinical measures of potential exposure. The primary purpose is to investigate the potential for excess exposures due to soil arsenic levels in the community. This type of study should be distinguished from a medical screening test whose primary purpose is to detect previously unrecognized disease in asymptomatic individuals and thereby improve their likely health outcome. This study also does not examine the incidence of health effects in the community. Exposure is important to measure because in the absence of excess exposure, increased health effects are not a concern.

Biomonitoring for arsenic measures recent exposure but does not identify the source of exposure or potential for disease. Arsenic is ubiquitous in the environment, and water and diet are the main sources for the general population. For this reason, biomonitoring is indicative of a person's exposure relative to general population ranges, but determining the source of exposure requires detailed assessment of dietary composition and habits and conditions affecting arsenic

exposure. The identification of any particular source of arsenic as being responsible for elevating exposure often requires examination of a number of individuals with measurements of environmental arsenic levels and habits affecting exposure.

This investigation was conducted in response to a request by the CAP for biomonitoring for exposure to soil arsenic and as a service to residents of Middleport who wished to have such biomonitoring. This environmental exposure investigation using biomonitoring does not estimate health risks, and its goals should be distinguished from those of other tools used in assessing environmental health such as risk assessment. Risk assessment is a separate effort that will be conducted for Middleport as a part of the environmental investigations under the Administrative Order on Consent between FMC and NYSDEC and EPA. Risk assessment allows investigators to compare findings or extrapolate either between human populations or from laboratory animals to humans. Accordingly, risk assessment is a frequently used regulatory tool in the protection of public health. Risk assessments calculate exposure and estimate health risks associated with a specific source, regardless of whether such risks are present in a community. EPA guidance holds that risk calculations are intended to estimate exposures for a potential high-end segment of the population so that decisions based on the estimated risks are protective of virtually all possible exposures in a population. In reality, very few people in a community would have the combination of upper-bound behaviors and characteristics assumed for this high-end segment of the population.

Risk assessments also typically focus on long-term exposure from a particular source (e.g., arsenic in soil) and do not consider risks associated with this source in context with risks from background sources (e.g., arsenic in water, diet). Measurement of biomarkers (e.g., arsenic in urine) includes all sources of recent exposure both from the source of interest (e.g., soil) as well as from background (e.g., diet, water). This biomonitoring study attempts to statistically distinguish whether the source of interest increases exposure above what would be expected from background, and if so, to what degree. The results of a biomonitoring study and risk assessment may therefore differ. A human health risk assessment will be part of the regulatory process that guides decision-making for the protection of health in Middleport. It is important to recognize, however, that risk assessment is distinct from the biomonitoring study reported here.

2 Study Methods

2.1 Program Roles

Exponent has conducted this exposure investigation with the assistance of Geomatrix for soil sampling and Sandler Occupational Medicine Associates, Inc. (SOMA) for house dust sampling. Battelle Pacific Northwest National Laboratory analyzed arsenic in urine, toenail, and vegetable samples and the Centers for Disease Control and Prevention (CDC) analyzed a subset of split samples of urine. Lockport Memorial Hospital analyzed urinary creatinine and specific gravity, and H2M Labs analyzed soil and house dust (Figure 3). Additional input and review of results was provided by an external technical advisory panel composed of toxicology, epidemiology, and public health experts (Figure 3). The study work plan was also provided to the lead regulatory agency (i.e., NYSDEC) for the site.

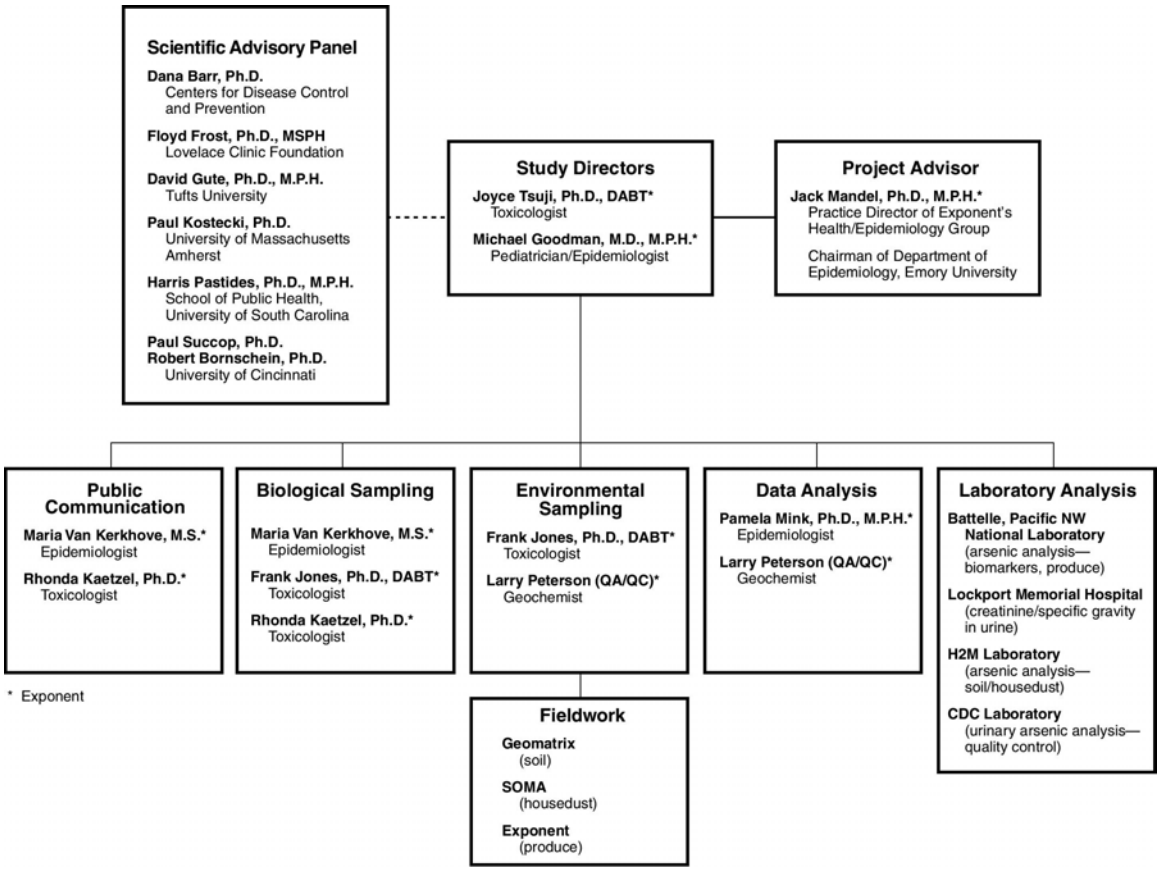


Figure 3. Organizational chart of the study team

2.2 Study Population

Middleport is located in Niagara County in western New York State. According to the 2000 U.S. Census, the total population of the Village of Middleport was 1,917 (47.4 percent male, 52.6 percent female; predominantly Caucasian [98.2 percent]). There were 756 total households in 2000. Of these households, 268 (35.4 percent) were households with children under 18 years of age (Table 2).

Table 2. Demographic characteristics of the Village of Middleport, New York: U.S. Census 2000

	Number of People	Percent of Total
Total Persons	1,917	100
Population by Sex		
Male	908	47.4
Female	1,009	52.6
Population by Age		
Under 5 years	141	7.4
5–9	129	6.7
10–14	172	9.0
15–19	155	8.1
20 and over	1,320	68.9
Population by Race[a]		
White	1,882	98.2
Black or African American	24	1.3
American Indian and Alaska Native	6	0.3
Asian	11	0.6
Native Hawaiian and other Pacific Islander	0	0.0
Other race	11	0.6
Total Households	756	100
Family households with own children under 18	268	35.4
Households with individuals under 18	286	37.8
Household with Income	757	100
Households with 1999 annual income < $35,000	358	47
Households with 1999 annual income ≥ $35,000	399	53
1999 annual median income	$36,464	--
1999 annual mean income	$40,239	--
School Enrollment		
Population 3 years and over enrolled in school	495	100
Nursery school, preschool	37	7.5
Kindergarten	15	3.0
Elementary school (grades 1–8)	277	56.0
High school (grades 9–12)	107	21.6
College or graduate school	59	11.9

Source: U.S. Census (2000).

[a] In combination with one or more other races listed. The six numbers add to more than the total population and the six percentages may add to more than 100 percent because individuals may report more than one race.

The target population for the study was Middleport children younger than 7 years old. Figure 4 shows the Village of Middleport and indicates the study area. Based on experience from other studies (e.g., Polissar et al. 1990; Hwang et al. 1997a,b), young children are the most appropriate study group because of their higher soil exposure. There were 194 children less than 7 years old identified in this area in the 2000 U.S. Census. During recruitment, we attempted to identify all children less than 7 years old in the study area. Although young children were the focus, anyone within the study area who desired testing was allowed to participate. The study area boundaries were intentionally set to be overly inclusive of areas with potentially elevated arsenic levels in soil, to provide a greater opportunity for participation to potentially exposed or concerned residents.

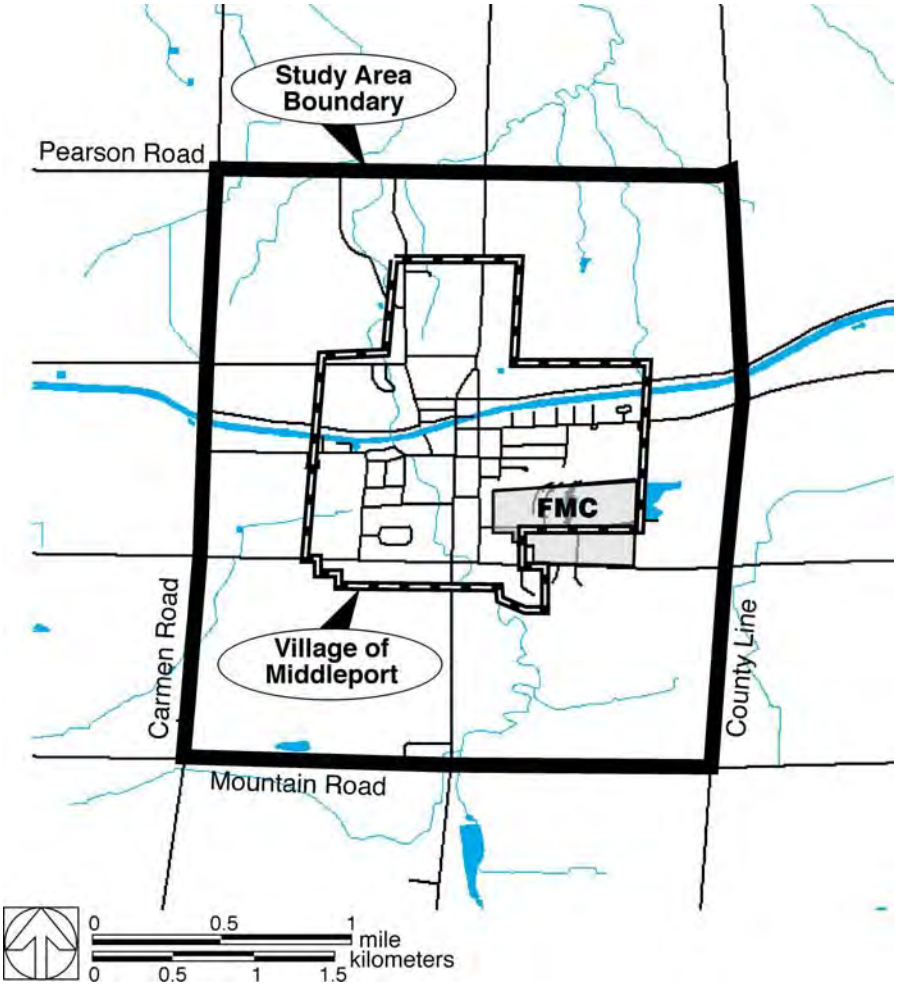


Figure 4. Study area of the Middleport environmental exposure investigation

2.3 Study Communication, Census, Recruitment, and Assessment of Participants

The biomonitoring program was announced in the FMC Middleport community newsletter in late May 2003, and a general announcement was mailed in late June to approximately 750 homes in the 14105 zip code (which included the study area boundaries). A section of the Middleport community website (www.teapothollow.com/middleport/, see Biomonitoring Program), maintained by FMC, was created to communicate information about the biomonitoring program.

In addition to informational mailings and flyers and posters displayed throughout the village, meetings were held in Middleport to announce the study, recruit participants, provide instructions and answer questions, and communicate results. The first meeting to introduce the program and begin recruitment of participants was held on July 8, 2003. Two local physicians who treat children were also contacted at this time and provided with information about the program. An Exponent field office was opened in downtown Middleport during recruitment and biomarker sample collection, and a toll-free hotline was available through the duration of the study to answer participants' and community questions. The study communication plan was described in detail in the work plan for the study (Exponent 2003).

Other presentations in the community included an open house on July 29, 2003, to continue to recruit participants and provide specific directions on urine and toenail collection; an informational booth at the Labor Day Festival in Middleport (August 30, 2003) to answer questions about the biomonitoring program; a meeting on October 30, 2003, to summarize biomonitoring participation rates and progress on soil and house dust collection and sample analysis; and a meeting on February 26, 2004, after participants received their results, to summarize sample results and answer questions. An open house to present the final study results is planned for August 10, 2004. Exponent also answered questions about the biomonitoring study during a local AM radio show on September 2, 2003, and during an FMC community meeting on June 2 and 3, 2004. Members of the Science Advisory Panel visited Middleport on May 7, 2004, and met with community representatives.

To estimate the number of children less than 7 years old, a door-to-door census was conducted during recruitment within the study area. Exponent personnel visited each home in the study area and provided them with a brochure about the study with contact information (Appendix A). Participation in the program was voluntary. Every dwelling within the study area was visited at least once. If no one was at home, information about the program was left at the door. Throughout the recruitment and sampling period, surveyors attempted to return to homes where no contact was made, particularly if there was an indication that young children in the target age range resided there. Despite these efforts, no contact was ever made at 16 percent of homes. However, most of these homes were believed not to have small children residing there, based on observations of the yard and premises (e.g., absence of toys and play equipment) and conversations with neighbors and village officials. Unfortunately, because of the time of the year, vacations made recruitment and scheduling for sampling more difficult.

Parents who agreed to allow their children to participate and adults who wanted to participate were asked to sign a study consent form (Appendix E in the work plan [Exponent 2003]) and complete a background questionnaire and child background questionnaire (Appendix C in the work plan [Exponent 2003]). Residents who chose not to participate were asked to answer some of the questions on the census form to assist in the evaluation of the potential number of children less than 7 years old (Appendix B in the work plan [Exponent 2003]). Reasons for not participating in the study included belief that "arsenic risks are overblown," reported distrust of FMC or anyone else involved with the study, feelings that the study was too invasive, reported hearing that the study "is not valid," or sample collection or scheduling difficulties. If the resident refused to release any information, census takers completed the census form according to their observations (e.g., whether young children were present, name on mailbox).

Incentives were offered to help publicize the program and encourage participation, especially by families with young children, and included a raffle for three outdoor gas grills (all participants) and five U.S. savings bonds (young children only).

Two questionnaires, a background questionnaire (Appendix C in the work plan [Exponent 2003]) and an exposure questionnaire (Appendix D in the work plan [Exponent 2003]) were administered to the participants to characterize the study population demographics and identify potential sources of arsenic exposure and possible factors that affect biomarker levels. The background questionnaire, which was administered at the time of participant recruitment, consisted of two parts: a survey of demographic information and a survey of soil exposure locations for child participants. Ideally, the demographic portion of the background questionnaire should have been administered to those who declined to participate as well to those participating to help evaluate potential self-selection bias; however, it was not administered because nonparticipants were not inclined to provide personal information. The exposure questionnaire, which was administered at the time of biomarker sampling, focused on the recent time period prior to biomonitoring to identify specific areas of outdoor exposure and other factors affecting arsenic exposures, such as diet. Both questionnaires included questions about soil and dust exposure (e.g., nature, location, and frequency) and the responses were used in the evaluation of individual biomarker results and in the analysis of potential covariates or modifying factors in the relationship between biomarker (i.e., urine and toenail samples) and environmental (i.e., soil, house dust, vegetable samples) data.

2.4 Biomarker Sample Collection

The principal biomarker for inorganic arsenic exposure is the concentration of arsenic in the urine, and in particular, the urinary forms of arsenic related to ingestion of inorganic arsenic. Sampling of arsenic in toenails was also offered to provide a longer-term measure of exposure than arsenic in urine (reflects exposure over the past few days); however, because of external contamination problems in this study and others (Hinwood et al. 2003a) and low participation and insufficient sample mass received in this study from some participants, toenail sampling was not the primary focus of the investigation. Urinary arsenic collection occurred from August 1 to September 13. Toenail collection occurred over the months of August through October.

Two first-morning void samples were collected from Middleport participants to even out potential day-to-day differences and as a backup in the case of the unavailability of one of the samples. Participants were asked not to eat seafood for 3 days prior to sample collection to avoid confounding urinary measurements with dietary organic arsenic. Asking participants to not eat seafood for more than 3 days (e.g., a week) was judged to be too onerous. Participants were asked on the exposure questionnaire whether they had eaten seafood within the week prior to sampling.

A community meeting was held on July 29, 2003, after initial recruitment of participants to discuss the instructions for collection and to provide a forum for responding to any questions. After providing verbal instructions to residents during scheduling for sampling, collection kits with written instructions (Appendix F in the work plan [Exponent 2003]) and the exposure questionnaire were distributed at participant's houses the day before and collected the following morning. Collection kits included a small cooler with freezable gel to keep samples cool after collection and before pick-up the same morning by field personnel. Pediatric urine bags with instructions for use were provided for parents of non-toilet-trained children.

Urine samples were kept chilled using blue ice (during transport) and refrigeration (at field office and Lockport Memorial Hospital) until delivered to the Battelle Pacific Northwest National Laboratory for arsenic analysis. Samples collected were held under refrigeration at the field office in Middleport until delivery to the Lockport Memorial Hospital laboratory (generally within 3 days) for measurement of specific gravity and creatinine. At least two deliveries a week were made to Lockport Memorial Hospital.

Toenail sampling and analysis were offered to participants only at their request and with the caution that they not contact soil with their feet for at least a month prior to toenail sampling and that a sufficient amount for analysis (possibly requiring several weeks of clipping) needed to be collected (0.5 g). Participants who enlisted in toenail sampling were provided with a pair of nail clippers, collection bag(s), and instructions for collection (Appendix F in the work plan [Exponent 2003]).

Toenail samples were received from late August through October. Some toenail samples were collected during the urine collection activities. Participants who enrolled toward the latter part of the sampling program were provided with self-addressed, stamped shipping packages to mail their toenail samples to Exponent when they had collected a sufficient mass. Toenail samples received were inspected and scored by Exponent and Battelle for the presence of visible dirt or discoloration before arsenic analysis. Most samples were also photographed.

2.5 Urine and Toenail Sample Analysis

Urine samples were delivered each week to the Lockport Memorial Hospital laboratory where an aliquot was removed from each sample for specific gravity and creatinine analysis by Lockport Memorial Hospital Laboratory's standard operating procedures. The samples were then shipped overnight to Battelle Pacific Northwest National Laboratory, in Sequim, Washington, for analysis of total and speciated arsenic. Samples were shipped at 4°C under chain of custody following the standard operating procedures in the work plan (Exponent 2003).

Quality control samples for urine samples consisted of analysis of split samples at a frequency of one in 20 samples. Sample splitting was performed at Lockport Memorial Hospital Laboratory as directed by the field personnel. Split samples were submitted blind to Battelle for the arsenic analysis. Splits consisted of both morning void samples for the same individual. Bottle blanks (empty bottles shipped from the field office in Middleport) at a rate of one in 20 samples were also sent to Battelle for use in control sample analysis.

For each participant, Battelle created a 20 mL composite, 10 mL from the two consecutive daily urine samples. The composite was analyzed for arsenic, and the original urine samples were archived at −20°C. Urine composites were stored frozen until batch analysis for total arsenic and speciated arsenic (sum of inorganic arsenic, dimethylarsinic acid [DMA], and monomethylarsonic acid [MMA]) using EPA Methods 1638 and 1632A, respectively (see details in quality assurance review in Appendix C). The method detection limit for total arsenic is 0.2 μg/L, and the method detection limits for arsenic species are 0.06, 0.08, and 0.4 μg/L for inorganic arsenic, MMA, and DMA, respectively. Quality control samples (e.g., blank, standard reference material, and matrix spike) are described in detail in the quality assurance report (Appendix C). No preservative was added to the sample because such preservatives convert organic arsenic forms into inorganic arsenic.

For one in 20 samples with detectable arsenic, an additional composite was prepared and submitted to CDC's laboratory for analysis of total and speciated arsenic for comparison with the results reported by Battelle.

Toenail samples were prepared as described by Karagas et al. (2000), which included an acetone wash to remove nail polish, if necessary, followed by sonification in deionized water for 10 minutes and a deionized water rinse. The samples were then acid-digested per American Public Health Association Standard Method 3030E (APHA 1995) and analyzed by inductively coupled plasma-mass spectrometry per EPA Method 6020 (U.S. EPA 1986). The method detection limit for toenail arsenic is 0.02 μg/g (dry weight), or 0.02 ppm. Laboratory control samples included a blank, control sample, and matrix spike sample for every 20 samples as described in Appendix C.

2.6 Environmental Assessment

Environmental samples for correlation with biomarker measures included yard and garden soil, house dust, and homegrown produce. Soil and household dust were collected from residences that participated in the biomonitoring study and whose occupants provided permission and access for collection of representative soil and dust samples. Homegrown produce was sampled at residences where the owners expressed an interest in having the produce sampled and analyzed.

2.6.1 Surficial Soils Collection and Analyses

Residential soil samples were collected by Geomatrix from November 2003 to January 2004, in accordance with Appendix H of the work plan (Exponent 2003) with the primary exception that the sampling strategy was changed from discrete sampling to composite sampling of yard soils

to be more representative of average exposure over a yard (see Geomatrix [2004] for updated procedures). Discrete sampling was used to characterize arsenic concentrations in soil as part of the environmental investigations conducted under the Administrative Order on Consent with NYSDEC. However, composite sampling of areas (e.g., yard, play area, and garden) provide a more representative measure of ongoing exposure over time and has been the method of choice by other environmental arsenic studies (e.g., Hwang et al. 1997a).

Of the 79 properties sampled, soil data from 77 families participating in soil sampling were used. Unfortunately, participation in soil sampling by homeowners was likely limited by the requirement that soil sampling results be shared with NYSDEC.

To create the yard composite sample, properties smaller than 11,000 ft^2 were divided laterally into a minimum of four sampling sectors, and one soil sampling location was randomly selected in each sampling sector to provide coverage of the area. Sampling locations were located at least 10 ft apart. If yard areas exceeded 11,000 ft^2, two to six additional composite samples were taken to represent these larger areas. Low areas on any property near drainage areas of concern (e.g., Tributary One or Culvert 105) were sampled as a separate yard composite sample.

Separate composite samples at properties were taken of children's play areas and vegetable gardens. Soil was sampled from a minimum of four locations within the play area and composited. One additional surface soil sampling location was added to the composite for every 625 ft^2 in excess of 2,500 ft^2. Yard soil and play area samples were taken from 0–3 in. depth below any vegetative cover.

Vegetable garden soil composites were composed of one soil sample from each garden plot with additional sample locations added for every 25 ft^2 of vegetable garden area. At each location, separate samples were collected for the 0–6 in. depth and the 0–12 in. depth to represent the root zone of different types of plants. Samples from these two depths were composited separately, resulting in two garden soil sample composites per property.

Standard field control samples were also collected as noted in Geomatrix (2004). Soil samples were analyzed by H2M Laboratories in Melville, New York (certified under the NYSDOH Environmental Laboratory Approval Program), for total arsenic using trace inductively coupled plasma-atomic emission spectroscopy by Methods 3050B/6010B, as specified in EPA SW-846 (U.S. EPA 1986), with a targeted limit of quantitation of 1 mg/kg. Conestoga-Rovers & Associates validated the soil sample laboratory results as detailed in Geomatrix (2004).

In addition to these samples, arsenic soil concentrations from discrete sample locations were available from 14 properties remediated during and after the biomonitoring study. Of these 14 properties, soil data for eight families participating in the biomonitoring study were used. None of these families had children less than age 7. These soil concentrations were provided by Geomatrix and collected, for the most part, during 2002 (Lachell 2004, pers. comm.). To make these samples more similar to the composite samples, the discrete samples for each yard were averaged. The maximum yard samples, however, are not similar to the maximum yard composite sample (i.e., highest sample among yard, play area, and garden composite samples) for the other yards sampled as part of the biomonitoring study. The highest soil sample reported

in this study is thus a discrete sample from a point location and is not representative of daily exposure even for this yard.

2.6.2 Interior Household Surface Dust Collection and Analyses

SOMA conducted house dust sampling for participants in the biomonitoring study who gave permission to sample. Due to scheduling and logistical difficulties, sampling occurred between September 3 and December 11, 2003. Residents were instructed not to sweep or vacuum their houses for a week prior to sampling; however, not all complied. Other observations during the sampling period were the soil remediation work in progress for the 14 properties in the community during September and October and recent or ongoing remodeling or renovation work at some of the houses. A short questionnaire that was administered at the time of sampling asked for information such as recent cleaning activities, type of heating (e.g., fireplace), the presence of pets that go in and out of the house, the presence of treated wood inside or outside the house, and renovation activities (see Appendix I of work plan [Exponent 2003]).

Based on protocol of Que Hee et al. (1985), dust samples were collected by vacuum technique using a personal air sampling pump and a piece of tubing attached to a sample cassette fitted with a filter provided by H2M Laboratories. (See Appendix I of the work plan [Exponent 2003] and text of SOMA house dust report in Appendix D of the current report. Tables of the SOMA report are not included because they contain personal information that is confidential according to the study consent form.)

For each house, a composite sample was obtained from at least three measured surface areas (625 cm^2 each) within each residence, including floor areas directly inside the entrance most often used by residents, the most frequently occupied room (living room, kitchen, or family room), and the residents' bedroom (child's bedroom if present). Additional areas were vacuumed in each of the three areas when the weight of the sample collected was approximately less than 60 to 80 percent of the target mass of target mass (0.5 g) for sample analysis.

Samples were submitted to H2M Laboratories for analysis of arsenic and total weight of dust. One blank sample was prepared and submitted once per day or for 10 percent of the samples collected. H2M Laboratories analyzed samples using the same methods as for soil arsenic analysis. Based on the arsenic content of each filter, the weight of dust on each filter, and the surface area vacuumed, arsenic sample results were expressed as concentration by weight (i.e., mg arsenic/kg dust, or ppm) and by surface loading (i.e., μg arsenic/100 cm^2).

House dust samples were collected and analyzed in five batches. For a subset of houses (31 residences) during the second sample round, only arsenic loading but not concentration in house dust was quantified because the mass of dust sample was not measured by the laboratory. Of these 31 residences, 16 were resampled to quantify arsenic loading and concentration. Surface loading from the earlier sampling round was used in the analysis. Because permission to resample was not obtained from all 31 residences lacking arsenic concentration results, 111 households have measurements of surface loading, but only 96 households have a measurement of arsenic concentration in house dust.

A quality assurance review conducted by SOMA on the quality of data produced by H2M Laboratories is detailed in SOMA's report (Appendix D). Exponent conducted an additional quality review of the information and data reported by SOMA.

2.6.3 Homegrown Vegetable Collection and Analyses

Homegrown produce was sampled and analyzed at participants' request. Exponent sampled vegetables during August and early September. Sampling focused on produce grown in gardens rather than tree fruits because leafy and root vegetables are of most concern for metals uptake from soil. This part of the program was provided as a service to concerned residents and was not intended to be a comprehensive survey of arsenic in Middleport vegetables, which was beyond the scope of the study objectives and what could be accomplished within the time line.

Because residents may grow more than one category of produce in their gardens, samples were collected from several categories of produce. Thus, gardens, for example, with leafy and fruiting vegetables and root plants, had samples from each of these vegetable categories collected. Sample quantities and other protocols are described in the work plan (Exponent 2003).

Limited samples of produce similar to each type of produce collected from Middleport residential home gardens were purchased from local farm stands or grocery stores. Results of the farm-stand produce analyses provided some comparison of arsenic concentrations in produce grown outside the Middleport study area; however, because only one or a few samples of each produce type were collected from stands or stores, the purchased samples were not representative of the range of arsenic concentration possible in commercial produce.

After picking, produce samples were brought back to the field office, refrigerated, processed, and frozen. Frozen samples were shipped to the laboratory on blue ice in coolers in four shipments over the duration of sampling. Procedures for processing and washing samples changed midway through the collection period. The work plan specified that samples would be washed with tap water in a manner similar to preparation for consumption. Initially, it was assumed that the laboratory would wash the vegetables and the field team would not wash vegetable samples unless they were noticeably dirty. However, field logistics were such that it was necessary to store samples longer than 48 hours, which required freezing to preserve the sample, and the frozen samples could not subsequently be washed at the laboratory without compromising sample integrity. In addition, for earlier samples that were washed, arsenic-free water (i.e., distilled or deionized water) may not always have been used because it was assumed that the samples would later be washed and rinsed with arsenic-free water. The protocol was later changed to having all vegetables washed and rinsed with arsenic-free water in the field office prior to freezing (except onion and garlic samples, which were brushed to preserve sample integrity). Vegetables were also scrubbed with a brush if necessary. None of the vegetables (e.g., carrots, turnips, cucumbers) were peeled, although the laboratory analyzed edible portions, which required removing the outer layers of onion and garlic and tops of root vegetables. Because of these differences in vegetable sample cleaning protocols, vegetable sample results were identified as "washed" or "unwashed" in the data analysis.

Purchased samples were obtained toward the end of vegetable collection to match the collected types of vegetables. As a result, all purchased samples from local farms stands or stores were thoroughly washed. The purchased samples are therefore not comparable to the unwashed or less thoroughly washed vegetable samples. Purchased samples were also not necessarily locally grown and may have been more thoroughly washed and trimmed of outer leaves, etc., prior to purchase. Another potential difference between Middleport vegetable garden samples and purchased samples is that some types of produce such as lettuce in Middleport gardens were sampled toward the end of their growing season, potentially resulting in collection of older plants with possibly higher mineral content than, for example purchased lettuce, much of which was commercially grown out of state.

Battelle analyzed produce samples for total arsenic according to the methods and detection limits specified in the work plan and as noted in the quality assurance review for biological samples (Appendix C). A limited number of samples with higher total arsenic levels were analyzed for speciated arsenic.

2.7 Data Management

Environmental data (i.e., soil and house dust) were obtained in electronic format from Geomatrix and SOMA. Biological data (i.e., urine, toenail, vegetable) were obtained in electronic format from Battelle, Lockport Memorial Hospital, or CDC. All data were entered into a computerized database. Questionnaire responses were also loaded into the database. The relational database included location of participant, urine or toenail arsenic levels, environmental sampling results, and questionnaire results.

The data management procedures were designed to ensure integrity, security, and reliability of the data, including:

- Backup procedures to maintain long-term integrity of the data
- Password-protected access to the database to prevent unauthorized access to, or modification of, the data, and to ensure confidentiality of personal information
- Independent verification to ensure the correctness of all key-entered data
- Maintenance of a comprehensive record of all changes made to the authoritative data set
- Use of automated procedures for rapid, repeatable, and quality-assured selection and summarization of data for interpretive analyses.

Data collected by Exponent are confidential to protect the identity of residents. The documents pertaining to biological samples, environmental sampling, and questionnaires are housed at Exponent and available to designated project staff. Each participant, family, and yard was assigned an identification number for tracking urine, toenail, and environmental samples to protect confidentiality and to track individuals, families, and yards separately from each other

(e.g., to track a child who plays in another part of Middleport). Some information was therefore shared with other involved parties at the site (i.e., Geomatrix and SOMA) but did not include any identifiers unless necessary (e.g., contact with Geomatrix or SOMA for environmental sampling of participants). Release of identifying information requires written authorization from the participant or parent.

The quality assurance program evaluated the quality of data generated by laboratories analyzing biomarker and environmental samples; blanks, blind, and bench quality control samples; and duplicate or split samples. This quality assurance program also involved checking of individual data reporting for identification of potential data entry errors. This performance evaluation helped ensure early detection and correction of any quality control problems.

2.8 Data Analysis

The focus of the study analysis was on the relationship between environmental arsenic levels (e.g., soil, dust) and arsenic concentrations in urine among the study population. To the extent possible, the results were controlled for the potentially confounding factors of other environmental and dietary sources and mediators of arsenic exposure.

As described in previous sections, the outcome measures of primary interest (dependent variables) were speciated arsenic and inorganic arsenic as measured in urine of the children (less than 7 years old) participating in the study. Other age groups were also examined, specifically children less than 13 years old and all participants, because of the relatively small population of young children and the large number of older participants in the study. Older age groups were also more likely to engage in some behaviors of interest (i.e., playing in creeks and eating garden vegetables). These results must be interpreted with caution, however, because of the potential for selection bias for older participants. Although more older individuals in total participated in the study, the participation rate among the older ages was lower than for young children. The effects of correction of the urinary data for hydration state using creatinine levels were also examined. The exposure measures of primary interest (independent variables) were the levels of arsenic in the soil and house dust. Other potential sources of arsenic exposure (e.g., diet), mediators of soil arsenic exposure (e.g., mouthing behaviors), and other covariates were ascertained through the questionnaire responses. Data were analyzed using the statistical software SPSS® for Windows (Version 7.0) and Microsoft® Excel.

2.8.1 Descriptive Statistics

The comparison of demographic characteristics of respondents and non-respondents with census data for this population was limited by insufficient information obtained from those who declined to participate and those with whom contact could not be established. Participation rates were estimated by comparison to census information collected in this study. Age demographics of participants were also compared to U.S. 2000 Census information.

We derived summary statistics (median, mean, and standard deviation) for continuous variables and frequency distributions (percent in each category) for the categorical variables. Variables with little or no variation were excluded from the inferential analyses discussed below (except

for those of particular interest, e.g., playing in creeks) or, where possible, categories were collapsed to increase the number of observations in some cells. Histograms were created and tests for normality were conducted with the environmental and biomarker data. The environmental and biomarker data were log-transformed based on their distribution as also reported in other studies (Hwang et al. 1997a). The distributions of all log-transformed variables were not significantly different from a normal distribution with the exception of speciated urinary arsenic data. However, transforming this variable did improve the fit with respect to the normal distribution with a change in the *p*-value from 0.007 to 0.013 (Kolmogorov-Smirnov Test of Normality). Geometric means and geometric standard deviations are also presented along with the arithmetic means.

For the purposes of evaluating exposure, soil arsenic data within yards were characterized as 1) overall mean arsenic level (all areas including 0–6 in. depth garden samples, play area, and yard samples), and 2) maximum arsenic level among these areas. We evaluated house dust level in terms of arsenic concentration and arsenic surface loading. Arsenic levels in vegetables (to the extent available) were categorized separately as "washed" or "unwashed."

2.8.2 Inferential Statistics

The primary purpose of these analyses was to evaluate whether measures of arsenic in the soil (i.e., mean arsenic level, maximum arsenic level) and in house dust (i.e., concentration in house dust, surface loading into house dust) are related to speciated arsenic and inorganic arsenic in the urine of young children. To address this question, the association between the dependent and independent variables were examined graphically and through correlation and regression analyses among children less than 7 years. As a secondary analysis, these associations were also examined for children less than 13 years, and the total study population. We estimated simple bivariate Pearson correlation coefficients among the dependent variables, exposure variables of primary interest, and continuous variables derived from the questionnaires (e.g., number of days the individual spent in or near a creek), and constructed a correlation coefficient matrix to present these estimates. Analysis of variance (ANOVA) and *t*-tests were conducted, where applicable, to evaluate associations between the primary outcome and independent variables (i.e. arsenic levels in urine, soil, and house dust) and the categorical variables derived from the questionnaires. Prior to conducting regression analyses, we visually examined a series of scatter plots, showing a dependent variable on the y-axis (e.g., arsenic in urine) and an independent variable (e.g., arsenic in soil) on the x-axis to determine graphically whether the association between the dependent and independent variables is indeed linear.

Linear regression models were constructed using the linear regression command in SPSS®. All models were initially adjusted for age. Age-adjusted regression models that included speciated arsenic and inorganic arsenic in urine as the dependent variables with each of the environmental variables (i.e. soil and house dust arsenic levels) were run to identify a "base" model from which to build multiple regression models. These models included the dependent and independent variables that appeared to best characterize and describe the exposure-outcome association. We also ran a series of age-adjusted linear regression models evaluating the association between each of the questionnaire variables and each of the dependent variables. To

be conservative, variables with a *p*-value of less than 0.15 in the age-adjusted models were considered to meet our inclusion criteria.

We followed generally accepted statistical practice and used regression diagnostics and techniques to guide our decisions regarding how to transform variables and how to consider potential covariates. Because some of the samples came from subjects living in the same household, we evaluated the assumption of independence by randomly selecting one subject from each household and comparing the results of these analyses to the results obtained from including all subjects. There was no difference in these results and therefore all results presented in this report are based on all subjects, regardless of household.

2.8.3 Reporting of Individual Results and Follow-Up

Sample results were evaluated for potential individual case follow-up to offer education and exposure assessment for individuals with potentially elevated arsenic exposures. The results were first compared to target screening levels (i.e., program reference levels), which are potential indicators of the upper range of background levels (assuming no excess dietary arsenic consumption) and the need for case follow-up. Any results exceeding these levels were also verified with the laboratory. The entire urinary profile of total arsenic and the different arsenic species was also examined for evidence of potential excess inorganic arsenic exposure. Responses to the exposure questionnaire were also available to obtain information about potential arsenic sources and exposures (e.g., recent seafood ingestion). Follow-up with individual participants was to include discussing potential sources of arsenic with residents and offering resampling. Individual study participants were sent their biomonitoring results by first class mail only after data validation and quality assurance. Those with results in need of follow-up were to be contacted directly by telephone; however, none required such follow-up. The nature of the data and the wording of letters communicating results were also provided to the scientific expert advisory panel for review (see Appendix B for example communication letters).

Because of ongoing exposure to background levels of arsenic through diet and water, reference levels were considered an initial screening for potential elevated exposures. Individual comparisons, however, do not necessarily indicate excess arsenic exposure to a specific source (e.g., soil). The evaluation of arsenic in soil as a source of exposure was based on the statistical analysis of the biomonitoring results paired with environmental data and other modifying factors for the community.

CDC and international health agencies have set reference levels for total arsenic in urine and nails. These levels generally range from 50 to 200 μg/L for total arsenic in urine. Levels of total arsenic in urine that exceed 50 or 100 μg/L do not necessarily constitute excess exposure to inorganic arsenic; a seafood meal may increase total urinary arsenic levels greatly above this range because of relatively nontoxic organic arsenic compounds in seafood. For example, 4 hours after eating a lobster tail, human volunteers showed an increase in total urinary arsenic level from 30 μg/L to 1,300 μg/L (ATSDR 2000a). CDC (2003a,b) and ATSDR (2000a,b) note that normal levels of total arsenic in urine are less than 50 μg/L (in absence of eating seafood in the past 48 hours), that follow-up monitoring should occur for levels between 50 to 200 μg/L,

and that a level over 200 μg/L is considered abnormal (assuming no recent large dietary source of arsenic) and may require medical treatment if symptoms of arsenic poisoning are present.

None of the federal or international health agencies have set standard reference levels for speciated or inorganic arsenic in urine, although several biomonitoring studies including those conducted by ATSDR have examined speciated or inorganic arsenic in urine in an attempt to avoid measuring nontoxic organic forms from the diet. In addition to total arsenic in urine, speciated arsenic forms (i.e., the metabolites of inorganic arsenic ingestion) were examined in relation to a reference level. Speciated arsenic levels in urine are a more complete measure of inorganic arsenic ingestion than only inorganic arsenic (ASTDR 2000b; WHO 2001). Even speciated arsenic (especially DMA) profiles, however, can be affected by dietary arsenic sources, particularly seafood (WHO 2001), and thus, participants were instructed not to eat seafood prior to testing. Reference levels for elevated exposure and need for case follow-up for speciated urinary arsenic have ranged from 40 to 50 μg/L in other studies (Table 3). A level as low as 20 μg/L has been used for inorganic arsenic (Table 3). Reports by ATSDR for the Spring Valley site in Washington, DC, refer to several speciated arsenic levels for urine that are considered not elevated or typically expected in unexposed individuals or the general population (e.g., 10 μg/L, 20 μg/L, 29 μg/L); however, a specific reference level for distinguishing normal from abnormal or elevated arsenic exposure for case follow-up was not stated (ATSDR 2002, 2003).

Table 3. Reference levels for arsenic in urine used in other exposure studies

Site/Study	Reference Level	Arsenic Form	Citation
Anaconda, Montana	50 μg/L (total)	Speciated	Hwang et al. (1997a,b)
Hayden, Arizona	30 μg/L	Inorganic	ADHS and ATSDR (2002)
Tacoma, Washington	40 μg/L 20 μg/L (children) 40 μg/L (adults)	Speciated Inorganic Inorganic	TPCHD (1988) Holland (2002)
Everett, Washington	50 μg/g creatinine	Inorganic	Sanderson and Kess (1995)
Nova Scotia	20 μg/L	Inorganic	NSDH and CBDHA (2001)
Denver, Colorado	50 μg/g creatinine	Total	WGI (2001)
Salmon, Idaho	50 μg/g creatinine	Inorganic	ATSDR (1998)
Fallon, Nevada	<50 μg/L (normal) >200 μg/L (abnormal)	Total	CDC (2003a,b)

Note: Units of micrograms of arsenic per gram of creatinine (μg/g creatinine) are approximately equivalent to micrograms of arsenic per liter of urine assuming an average urinary creatinine content of 1 g/L.

Reference levels for the Middleport program were selected on the basis of available health guidance and levels used by other studies. The Middleport program first used the CDC reference level of 50 μg/L (or 50 μg/g creatinine) for initial screening of total arsenic levels in urine. Because of the dietary interference problems with this level, speciated arsenic was considered a better measure than total arsenic for evaluating elevated exposure to inorganic arsenic. Speciated arsenic (and in particular the urinary profile of the different arsenic forms in relation to total arsenic levels) provides more information than total arsenic on the potential contribution of different sources of arsenic exposure. Based on reference levels used in other

studies, a speciated arsenic level of 40 μg/L was used to screen the Middleport results for elevated arsenic exposure and potential need for follow-up actions.

The inorganic arsenic level was also compared to 20 μg/L, the lowest of the levels used for inorganic arsenic in other studies reviewed (Table 3).

Normal levels for total arsenic in nails are considered to be less than 1 ppm (ATSDR 2000b); however, a control group with low arsenic exposure had an average of 1.7 ppm (Hinwood et al. 2003a). Thus, toenail arsenic levels somewhat above this level may still be considered normal. A level of 1 ppm in toenails was used as the initial reference level for comparison of this biomarker. Because of the potential for external contamination of toenail samples and apparent variation in what is considered a normal arsenic level (e.g., Hinwood et al. 2003a), toenail arsenic levels that do not differ substantially from 1 ppm may still be normal.

3 Results

3.1 Community and Participant Demographics

The results of the study census indicate that there are 826 households in the study area (including individual apartments). Of these 826 households, 39 were observed as vacant, 55 were never interviewed (i.e., no contact with residents other than the study team leaving information), and 480 provided only partial census information (either the resident refused to give information or partial information was obtained by a third party). Households with no contact were assumed to have at least one adult resident. Demographics of the study area (Table 4) show an overall similarity to the 2000 U.S. Census for the Village of Middleport (Table 2) even though the study area included outlying areas. The population density outside the Village of Middleport but within the study area is relatively low. A total of 1,930 people reside within the study area boundaries. The study census identified 164 children under the age of 7 years old from 106 households.

Table 4. Demographic characteristics of the study area for the Middleport environmental exposure investigation, 2003

	Number of People	Percent of Total	2000 U.S. Census Number of People	2000 U.S. Census Percent of Total
Total Persons	1,930	100	1,917	100
Population by Sex				
Male	874	45	908	47
Female	981	51	1,009	53
Unknown	75	4	--	--
Population by Age				
Under 5 years	104	5	141	7
Under 7 years (i.e., younger than 84 months)	164	8	--	--
5–9	116	6	129	7
10–14	105	5	172	9
15–19	128	7	155	8
20 and over	997	52	1,320	69
Age unknown	465	25	--	--
Total Households	826	100	756	100
Family households with children under 7	106	13	--	--
Households with individuals under 18	227	27	286	37.8

Note: The values for 2000 U.S. Census represent only the Village of Middleport.

There were 439 people from 167 families who participated in urine sampling. Of these 439 people, 77 were less than 7 years old, the target age range for this exposure study (Table 5). These 77 children (from 55 families) represent 47 percent of the target population in the study area. There were 101 children ages 7–18 years old and 261 adults who participated in urine sampling. Of the 84 individuals from 42 families who participated in toenail sampling, only 6 were children of the target age. Only 67 toenail samples had a sufficient mass for the arsenic concentration to be quantified. None of these toenail samples were from young children.

Table 5. Demographic characteristics of the participants in the Middleport environmental exposure investigation, 2003

	All Participants	Children 7 Years and Older and Adults	Children < 7 Years Old	Children < 13 Years Old
All Persons Participating in Urine Sampling	**439**	**362**	**77**	**142**
Male	206	169	37	68
Female	233	193	40	74
Urine and Environmental Sampling				
Persons with urine, soil, and house dust data[a]	185	151	34	57
Persons with urine and soil data only	64	57	7	19
Persons with urine and house dust data[a] only	93	75	18	31
Persons with urine data only	97	79	18	35
Population by Age				
Under 7 years	77	NA	77	NA
Under 13 years	142	NA	NA	142
7–18 years	101	101	NA	NA
18–92 years	261	261	NA	NA
Total Persons Participating in Toenail Sampling[b]	**67 (84)**	**67 (78)**	**0 (6)**	**2 (8)**
Male	32 (38)	32 (35)	0 (3)	1 (4)
Female	35 (46)	35 (43)	0 (3)	1 (4)
Toenail and Environmental Sampling				
Persons with toenail, soil, and house dust data[a]	34	34	0	1
Persons with toenail and soil data only	9	9	0	1
Persons with toenail and house dust data[a] only	14	14	0	0
Persons with toenail data only	10	10	0	0

Note: NA - not applicable

[a] House dust data refers to arsenic concentration in dust only (not surface loading).

[b] Numbers in parentheses indicate the total number of samples submitted, including those with insufficient mass for quantification.

The overall study population is generally representative of the demographics of the Village of Middleport with respect to age and income as compared to the 2000 U.S. Census (Table 6; Table 2).

3.2 Biomonitoring Results

3.2.1 Urine Sample Results

The urine results of the participants who provided samples during the biomonitoring study showed no indication of excess exposure from inorganic arsenic above background sources such as diet and water. There were 26 participants with urinary levels of total arsenic greater than 50 μg/L; however, in all cases, speciated arsenic levels were well below the 40 μg/L program reference level and inorganic arsenic levels in urine were very low (i.e., below 3 μg/L) compared to the program reference level of 20 μg/L. Thus, the more reliable indicator of inorganic arsenic ingestion, speciated urinary profile, for all participants does not indicate elevated exposure to inorganic arsenic.

Table 6. Demographic characteristics of the families who participated in the biomonitoring study

	Number of Households	Percent of Total	Households with Children <7 years Old	Percent of Total	Percent of 2000 U.S. Census
Families Participating in Biomarker Sampling					
Urine sampling	167	100	55	100	--
Toenail sampling[a]	41 (4)	25	0 (3)	0	--
Families Participating in Environmental Sampling					
Homegrown produce sampling	41	25	12	22	--
House dust sampling[b]	96/111	57/66	36/37	65/67	--
Total soil samples	85	51	29	53	--
Yard	84	50	28	51	--
Play area	28	17	21	38	--
Garden	23	14	8	15	--
Family Population by Race[c]					
White	154	92	52	95	98
African American	2	1.2	2	3.6	1.3
Native American	4	2.4	0	0	0.3
Asian	0	0	0	0	0.6
Other Race	3	1.8	1	1.8	0.6
Unknown	4	2.4	0	0	--
Household Income					
Less than $35,000 per year	--	--	--	--	47
Greater than or equal to $35,000 per year	--	--	--	--	53
Less than or equal to $40,000 per year	72	43	18	31	--
Greater than $40,000 per year	82	49	35	65	--
Unknown	13	8	2	4	--

[a] Values in parentheses represent the number of families who submitted toenail samples that had insufficient mass for quantification.

[b] Number of families with arsenic concentration in dust (ppm)/number of families with arsenic surface loading.

[c] In combination with one or more other races listed for the 2000 U.S. Census. The six numbers add to more than the total population and the six percentages may add to more than 100 percent because individuals may report more than one race.

A total of 439 composite urine samples were analyzed for total arsenic, inorganic arsenic, DMA, and MMA (Table 7). The total arsenic in urine samples averaged 22.7 μg/L, ranged from 2.1 to 773 μg/L among all participants, and was less than 60 μg/L for children less than 7 years old. Of the 26 participants who had a total arsenic level above 50 μg/L, 14 of them reported eating seafood in the 7 days prior to sampling and all of them had detectable levels of DMA, which can be affected by ingestion of seafood arsenic forms. Of the 439 participants, 29 percent (n=125, 16 children and 109 older children/adults) reported eating seafood in the 7 days prior to sampling.

The average speciated arsenic level (i.e., the sum of inorganic arsenic, MMA, and DMA values) for all participants was 4.7 μg/L, with all samples below 20 μg/L, or half the program reference level. The geographic distribution of the speciated urine data also shows little pattern with respect to proximity to the FMC facility or historical drainage from the plant (Appendix E). Speciated arsenic levels for young children versus older participants were not significantly different (*t*-test; p=0.665). The urinary species with the lowest measured levels was MMA for

which 62 percent of participants had levels that were undetectable or qualified as estimated below the limit of detection. Consistent with human metabolism of arsenic, the DMA species had the highest results among the arsenic species (Table 7), with only 12 percent undetected or qualified as estimated below the limit of detection.

Split samples of urine composites were sent to CDC to verify the values measured by Battelle. The comparison of these samples is provided in Appendix F. Overall, Battelle was able to achieve lower detection limits than the CDC, and the results of both laboratories showed a close 1 to 1 correlation for total arsenic levels and a reasonable agreement for speciated arsenic given differences in analytical techniques and detection limits (Figure 5). Differences in speciated arsenic levels were greatest at lower arsenic sample concentrations where analytical uncertainty is greater. Thus, such potential laboratory variation would have little effect on evaluating individual results for excess arsenic exposure.

Table 7. Summary of arsenic concentration in urine samples

	Total Arsenic		Speciated Arsenic		Individual Arsenic Species		
	µg/L	µg/g Creatinine	µg/L	µg/g Creatinine	Inorganic Arsenic (µg/L)	MMA (µg/L)	DMA (µg/L)
All Participants (n=439)							
Geometric Mean (±GSD)	15.7 (±2.0)	13.6 (±2.1)	3.9 (±1.9)	3.4 (±1.9)	0.78 (±1.4)	0.46 (±1.8)	2.5 (±2.4)
Arithmetic Mean (±SD)	22.7 (±44.7)	20.3 (±37.3)	4.7 (±3.0)	4.1 (±3.1)	0.83 (±0.32)	0.54 (±3.2)	3.5 (±2.6)
Median	14.0	11.9	4.2	3.4	0.77	0.50	3.1
Range	2.1–773	2.4–620	0.89–19.9	0.43–39.3	0.31–2.7	0.024–2.4	0.17–17.1
Children < 7 years (n=77)							
Geometric Mean (±GSD)	15.1 (±1.8)	19.6 (±2.0)	4.0 (±2.2)	5.2 (±1.9)	0.81 (±1.5)	0.54 (±1.9)	2.5 (±2.9)
Arithmetic Mean (±SD)	17.4 (±10.0)	25.9 (±27.2)	5.3 (±3.8)	6.4 (±5.1)	0.87 (±0.34)	0.65 (±0.43)	3.9 (±3.2)
Median	15.6	17.6	5.0	5.1	0.80	0.50	3.6
Range	2.1–59.6	3.0–174	0.89–17.7	0.76–39.3	0.31–2.1	0.12–2.1	0.27–13.8
Children < 13 years (n=142)							
Geometric Mean (±GSD)	15.7 (±1.7)	15.7 (±1.9)	4.6 (±2.1)	4.6 (±1.9)	0.83 (±1.4)	0.55 (±1.8)	3.0 (±2.6)
Arithmetic Mean (±SD)	18.2 (±10.8)	20.3 (±21.6)	5.8 (±3.8)	5.5 (±4.1)	0.89 (±0.35)	0.65 (±0.40)	4.3 (±3.2)
Median	15.0	14.1	5.1	5.7	0.80	0.52	3.8
Range	2.1–59.9	3.0–174.4	0.89–19.9	0.57–39.3	0.31–2.7	0.11–2.4	0.27–17.1
Older Children/Adults (n=362)							
Geometric Mean (±GSD)	15.8 (±2.1)	12.6 (±2.1)	3.8 (±1.9)	3.1 (±1.8)	0.78 (±1.4)	0.44 (±1.8)	2.5 (±2.3)
Arithmetic Mean (±SD)	23.8 (±48.9)	19.1 (±39.1)	4.6 (±2.8)	3.6 (±2.1)	0.82 (±0.31)	0.51 (±0.28)	3.4 (±2.4)
Median	13.8	11.0	4.2	3.2	0.75	0.50	3.0
Range	3.9–773	2.4–620	0.91–19.9	0.43–16.7	0.31–2.7	0.02–2.4	0.17–17.1

Note: DMA - dimethylarsinic acid
GSD - geometric standard deviation
MMA - monomethylarsonic acid
SD - standard deviation

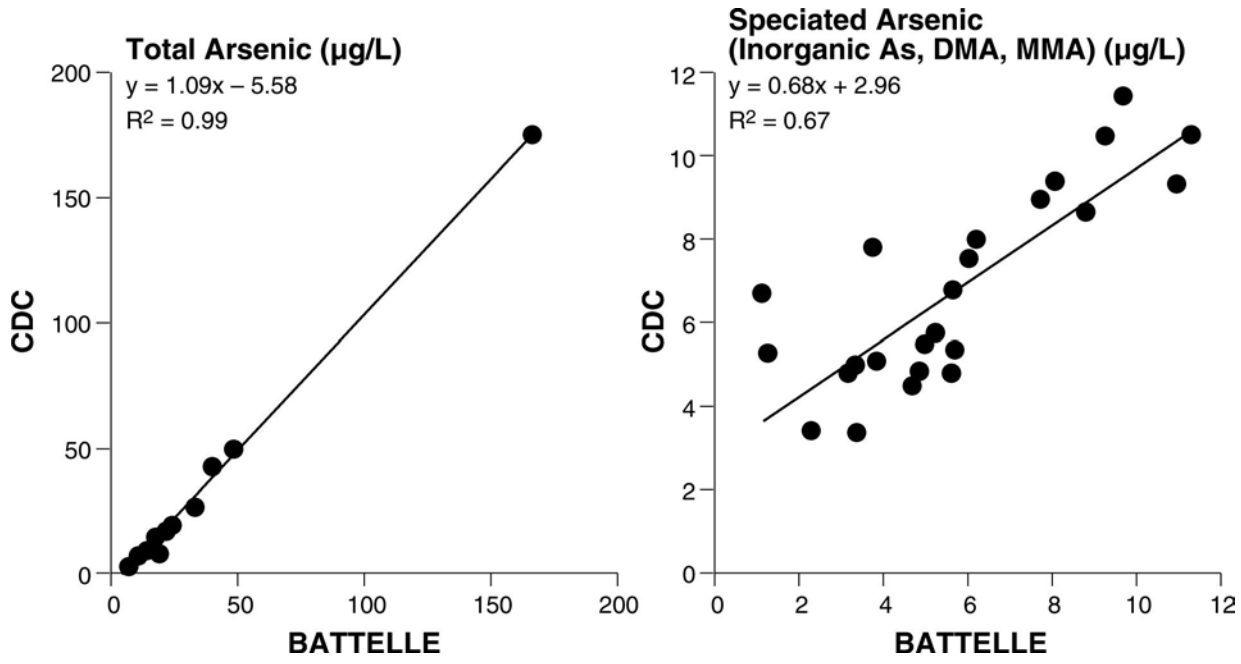


Figure 5. Comparison of CDC and Battelle laboratory results

Species of arsenic measured by Battelle included total inorganic arsenic, MMA, and DMA. CDC measured arsenous acid (III), arsinic acid (V), MMA, DMA, trimethylarsine oxide (TMAO), arsenobetaine (AsB), and arsenocholine (AsC). Arsenous acid (III), TMAO, and AsC were below the detection limits established by CDC in these samples (a detailed presentation of all the data is in Appendix F). AsB, a form of arsenic found in foods such as seafood, was detected in most of the samples.

3.2.2 Toenail Sample Results

Of the 67 samples with sufficient sample mass to quantify arsenic concentration, none exceeded the reference level of 1 ppm set by CDC. Though samples from six children under the age of 7 were submitted, none had sufficient sample mass; therefore, no data are available on arsenic toenail levels in children less than 7 years old. The average total arsenic in toenail samples was 0.19 ppm, with a range of 0.02 to 0.97 ppm (Table 8).

Surface contamination with dirt appears to have affected the measured arsenic concentration of toenail samples. Toenail samples were scored from 1 (clean) to 4 (all clippings with dirt or dark discoloration), with some intermediate scores (e.g., 2.5). A positive relationship was apparent between the level of discoloration and arsenic concentration of the toenail samples, with approximately a 75 percent relative increase in arsenic concentration per unit increase in discoloration score (p=0.0001, R^2=0.205) (Figure 6).

Table 8. Summary of arsenic concentration in toenail samples

	Toenail Arsenic (ppm)
All Participants (n=67)	
Geometric mean (±GSD)	0.13 (±2.53)
Arithmetic mean (±SD)	0.19 (±0.20)
Median	0.12
Range	0.02–0.97

Note: GSD - geometric standard deviation
SD - standard deviation

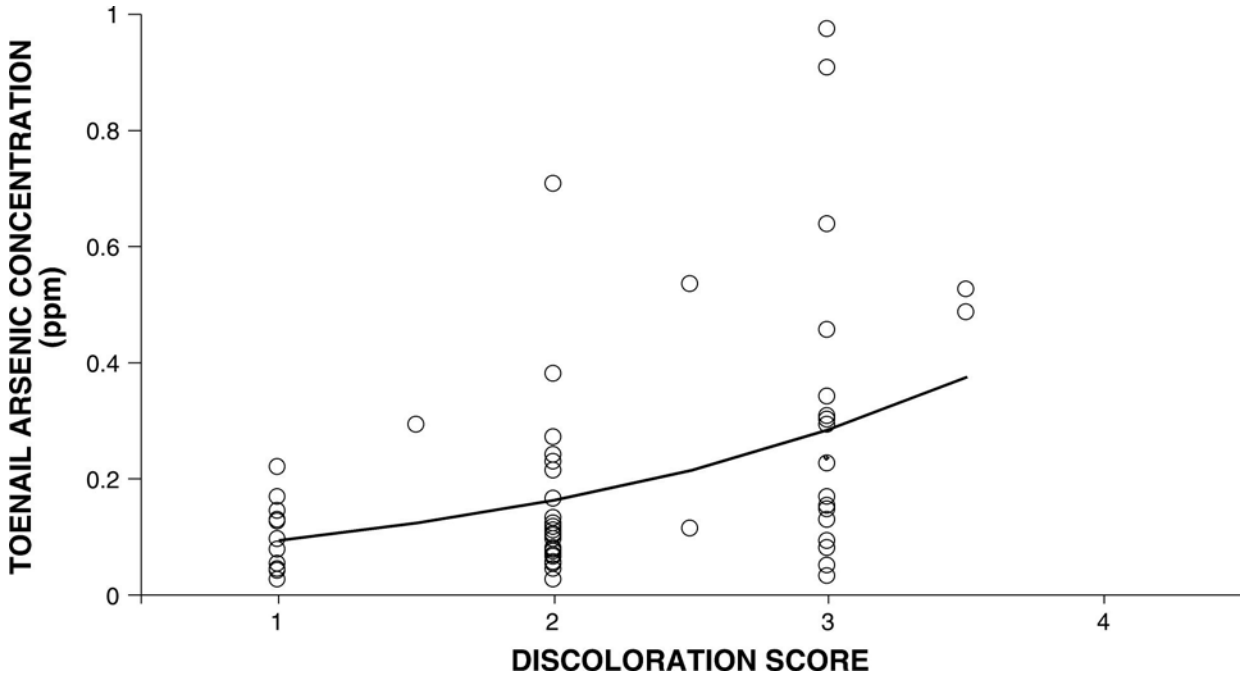


Figure 6. Toenail arsenic concentration versus discoloration of toenail samples

Comparisons among the three predominant discoloration scores, 1, 2, and 3, showed that samples with a score of 3 had significantly (α=0.05) higher arsenic concentrations than samples with a score of 1 (ANOVA based on log-transformed concentrations followed by Tukey's multiple comparison tests; p=0.0029). Non-parametric methods (Kruskal-Wallis followed by Wilcoxon tests) also showed that arsenic concentrations in samples that scored 3 are significantly higher than for those samples with a score of 2 (non-parametric p-values: 0.03 for Kruskal Wallis; non-adjusted pairwise p-values are 0.0026 for 1 vs. 3 and 0.013 for 2 vs. 3). The observed association between toenail discoloration and arsenic concentration is consistent with external contamination of toenail samples.

3.3 Environmental Sampling Results

3.3.1 Soil Sampling Results

Arsenic soil data were obtained from a total of 85 households (out of 167) participating in the biomonitoring study from at least one area on the property where these households were situated. Of these households, 29 families had at least one child less than 7 years old who participated in the study. Arsenic soil data for the study area came from two sampling investigations. Geomatrix sampled 79 properties from mid-November 2003 to January 2004 as part of this investigation (Geomatrix 2004). Of these properties, 77 households participated in the biomonitoring study. Discrete soil data were also obtained for properties of 8 families that were among the 14 properties remediated in summer and fall of 2003 (Lachell 2004, pers. comm.).

Geomatrix previously collected background soil data from the Gasport area in orchard lands, wooded-agricultural land, commercial/industrial land, and residential/public land and found that the soil sample results from Middleport (excluding the remediated properties) are consistent with background data collected from Gasport (summary data reported by U.S. EPA et al. 2003; Table 1).

To evaluate potential exposure to soil, average and maximum arsenic soil concentrations were calculated from all surface soils (i.e., yard, garden, and play area samples up to 6 in. in depth) sampled in each of the 85 yards of participants (Table 9). Average and maximum soil concentrations are displayed on the study area map to provide a visual evaluation of the geographic distribution of concentrations and participating families (Appendix E).

Table 9. Summary of arsenic concentration in soil samples (mg/kg, or ppm)

	Area Sampled				
	Yard	Garden (0–6 in.)	Play Area	Average All Samples	Maximum All Samples
All Households					
Number of yards sampled	84	23	28	85	85
Arithmetic mean (±SD)	28.0 (±37.4)	19.4 (±12.3)	21.7 (±16.3)	27.5 (±37.2)	42.4 (±120)
Geometric mean (±GSD)	21.1 (±1.9)	16.0 (±1.9)	16.9 (±2.1)	20.6 (±2.0)	24.7 (±2.2)
Median	19.7	15.9	13.2	19.5	22.5
Range	5.2–340	4.6–50.2	4.3–60.5	4.6–340	6.2–1,124
Households with Children Under 7					
Number of yards sampled	28	8	21	29	29
Arithmetic mean (±SD)	22.5 (±12.0)	22.5 (±11.0)	22.3 (±15.2)	22.5 (±11.7)	27.2 (±14.5)
Geometric mean (±GSD)	19.8 (±1.7)	20.3 (±1.6)	18.2 (±1.9)	19.9 (±1.6)	23.8 (±1.7)
Median	18.9	19.5	13.2	19.1	22.8
Range	8.2–57.7	11.3–39.7	6.9–58.8	10.4–46.4	10.4–58.8

Note: GSD - geometric standard deviation
SD - standard deviation

The average yard (not including garden or play area samples) concentration was 28 mg/kg, with a range of 5.2 to 340 mg/kg. The maximum average arsenic concentration for a yard of 340 mg/kg was much higher than average values from the other properties with soil samples (the next highest average value for any yard was 69 mg/kg). This maximum value is the average of discrete samples from this yard, which was sampled during the previous investigation of the 14 properties near the FMC facility that were remediated in 2003. This particular property had the maximum of all soil samples (1,124 ppm) available from biomonitoring study participants. This sample is a discrete sample rather than a composite sample and prior to remediation was located near the FMC facility property line. The next highest maximum yard value, a composite sample, was 103 ppm. Samples from the 0–6 in. and 0–12 in. depths were taken from 23 gardens throughout the study area. There was a close correlation between arsenic concentration of garden soil samples taken at the 0–6 in. interval and that of the 0–12 in. depth in the same garden (soil arsenic at 0–12 in. = 0.9 × soil arsenic at 0–6 in. + 2.58; $R^2 = 0.92$). Garden soil concentrations were generally lower than yard soil concentrations (Table 9). Of the 23 garden soil samples, 8 were collected from families with children in the target age group.

Twenty-eight samples were collected from play areas of yards in the study area, mostly from families with children less than 7 years old (see Appendix E for the geographic distribution of play area soil samples). Sample concentrations were generally lower than yard soil concentrations (Table 9).

3.3.2 House Dust Arsenic Results

Arsenic in house dust was quantified in two ways (Table 10)—as concentration (mg arsenic/kg dust, or ppm) in the dust and as surface loading on floors or the mass of arsenic per surface area sampled (μg/100 cm^2). The geographic distribution of the house dust data in the community shows little similarity to the distribution of soil arsenic concentrations (Appendix E).

Table 10. Summary of arsenic concentration in house dust samples

	All Households		Households with Children Under 7	
	Arsenic Concentration (mg As/kg dust)	Surface Loading of Arsenic (μg As/100 cm^2)	Arsenic Concentration (mg As/kg dust)	Surface Loading of Arsenic (μg As/100 cm^2)
Number homes sampled	96	111	36	37
Arithmetic mean (±SD)	20.3 (±29.0)	0.202 (±0.384)	21.8 (±33.2)	0.14 (±0.19)
Geometric mean (±GSD)	10.8 (±3.0)	0.071 (±4.4)	11.2 (±3.1)	0.058 (±4.0)
Median	10.4	0.072	9.8	0.53
Range	1.0–172	0.004–2.970	1.7–172	0.004–0.77

Note: GSD - geometric standard deviation
SD - standard deviation

3.3.3 Homegrown Produce Results

Homegrown produce was sampled from 42 gardens throughout the study area (see Appendix E for geographic distribution). Arsenic concentrations in all of the vegetable samples were below 0.6 ppm and much lower than background levels of arsenic in soil (Table 11). Tomatoes were the most prevalent vegetable among the gardens sampled and had consistently low arsenic concentrations near or below the limit of detection. Arsenic levels vary naturally in vegetables, and variation within a community and with multiple samples is expected. Sample arsenic concentrations are also based on the fresh weight (as sampled) and can vary depending on the vegetable water content (e.g., whether the garden had been watered recently, whether vegetables were older and/or wilted at the end of the growing season). Only a few samples from local stores were tested, which limited understanding of the variation in arsenic levels for store-bought samples. "Unwashed" vegetable samples tended to have higher arsenic concentration than "washed" vegetables samples (Table 11). All store samples were washed or carefully brushed off (onion and garlic samples).

Because of the number of variables related to arsenic concentration in vegetables (e.g., washed/ unwashed, vegetable type, soil concentration) and the limited sample size for many vegetable types and relatively few participants with urine, garden soil, and vegetable data, statistical analysis of whether vegetable concentrations were correlated with soil concentrations and higher urinary arsenic levels was deemed unproductive. The effect of eating homegrown produce on urinary arsenic levels was examined in the correlation analysis (Section 3.3.5).

3.3.4 Summary of Indirect Exposure Factors Derived from the Questionnaire

Data on potential indirect arsenic exposure factors (e.g., diet, mouthing behaviors) were derived from the exposure and family background questionnaires (Appendix D of the work plan; Exponent 2003). The summary statistics for indirect exposure factors among children less than 7 years old are presented in Table 12 (continuous variables) and Table 13 (binary and categorical variables). Summary statistics for exposure factors among children less than 13 years old and the total study population are presented in Appendix G. Because of a lack of variability and interest in several of the factors included in the questionnaires, only those exposure factors identified by footnote were included in the statistical analyses presented in Section 3.3.5.

Table 11. Summary of vegetable sampling results

		Homegrown Produce		Purchased Produce	
Vegetable Type	Sample Preparation[a]	Number of Samples	Arsenic Concentration[b] (ppm)	Number of Samples	Arsenic Concentration[b] (ppm)
Basil	Washed	2	0.038–0.107	1	0.028
Beans, green	Washed	1	*<0.013*	1	*<0.006*
	Unwashed	2	*<0.006*–0.020	--	--
Beans, yellow	Washed	0	--	1	*<0.009*
	Unwashed	1	*<0.006*	--	--
Beet, greens	Washed	0	--	1	0.028
	Unwashed	3	0.013–0.028	--	--
Beet, bulb	Washed	0	--	2	*<0.006*
	Unwashed	3	0.007–0.016	--	--
Broccoli	Washed	2	*<0.007–<0.009*	1	*<0.008*
	Unwashed	1	*<0.009*	--	--
Carrot (unpeeled)	Washed	1	0.173	1	*<0.008*
Carrot (peeled)	Washed	0	--	1	*<0.006*
Cauliflower	Washed	0	--	1	*<0.006*
	Unwashed	2	0.009–0.050	--	--
Chard	Washed	1	0.054	1	*<0.007*
Collard greens	Washed	1	0.125	1	0.008
Cucumber	Washed	3	0.010–0.044	2	*<0.002–<0.003*
	Unwashed	3	0.019–0.103	--	--
Garlic	Unwashed	1	*<0.024*	1	*<0.023*[c]
Lettuce	Washed	3	0.020–0.051	2	*<0.002*–0.004
	Unwashed	3	0.148–0.341	--	--
Mint	Washed	2	0.065–0.087	1	0.024
	Unwashed	1	0.028	--	--
Onion	Unwashed	4	*<0.006*–0.199	1	0.008[c]
Onion, green	Washed	1	0.018	1	0.010
	Unwashed	1	0.022	--	--
Pepper, banana	Washed	1	0.006	1	*<0.005*
Pepper, green	Washed	0	--	1	*<0.003*
	Unwashed	1	*<0.004*	--	--
Radish	Washed	0	--	1	0.007
	Unwashed	1	0.094	--	--
Sage	Washed	2	0.053–0.345	1	0.027
	Unwashed	1	0.574	--	--
Squash, acorn	Washed	1	0.006	1	*<0.006*
Squash, yellow	Washed	5	*<0.005*–0.063	1	*<0.005*
	Unwashed	3	0.004–0.015	--	--

Table 11. (cont.)

Vegetable Type	Sample Preparation[a]	Homegrown Produce: Number of Samples	Homegrown Produce: Arsenic Concentration[b] (ppm)	Purchased Produce: Number of Samples	Purchased Produce: Arsenic Concentration[b] (ppm)
Squash, zucchini	Washed	3	*<0.003–<0.004*	1	*<0.004*
	Unwashed	2	0.006–0.008	--	--
Tomato	Washed	14	*<0.003–<0.005*	1	*<0.004*
	Unwashed	6	*<0.004*–0.010	--	--
Tomato, cherry	Washed	2	*<0.005–<0.008*	1	*<0.007*
	Unwashed	1	0.008	--	--
Tomato, grape	Washed	0	--	1	*<0.004*
	Unwashed	1	0.007	--	--
Tomato, green	Washed	3	0.004–*<0.007*	0	--
	Unwashed	8	*<0.004*–0.009	--	--
Tomato, plum	Washed	2	*<0.004*–0.006	1	*<0.003*
Turnip (unpeeled)	Washed	1	0.013	1	*<0.005*

Note: Sample concentration values in italics and preceded with "<" had no detectable arsenic at the limits shown.

[a] "Washed" samples were thoroughly washed with tap water and rinsed with distilled water. "Unwashed" samples were not as thoroughly cleaned.

[b] Arsenic concentration is based on fresh weight of the sample, which can vary depending on water content.

[c] Purchased garlic and onion were brushed off before analyses.

Table 12. Numerical questionnaire responses for children less than 7 years old used in statistical analyses

	N	Mean (±SD)	Median	Minimum	Maximum
Participant Characteristics					
Age of child when urine sample was taken (years)[a]	77	4.3 (±2)	4.7	0.1	7
Weight (lb)[a]	75	40.4 (±14.1)	40	11	78
Height (in.)	73	40.2 (±7.5)	41	21	56
Body mass index[a]	73	17.4 (±3.8)	16.7	10.0	30.0
Number in household[a]	77	4.7 (±1.5)	4	2	10
Behavioral Characteristics (over past 7 days)					
Time playing in outdoor area (days)[a]	70	5.2 (±1.7)	5	1	7
Time playing in outdoor area (hours/day)[a]	68	3.7 (±2.6)	3	0.5	15
Time playing in other outdoor area (days)	29	3.8 (±1.9)	3	2	7
Time playing in other outdoor area (hours/day)	29	2.6 (±2.2)	2	0.5	8
Number of days attended school	23	3.7 (±1.4)	4	1	6
Number of hours attended school	23	5.9 (±2.6)	7	1	10
Time playing in or near creeks, streams, etc. (days)[a]	10	4 (±2.5)	4	1	7
Time playing in orchards, etc. (days)[a]	3	1.7 (±0.6)	2	1	2
Washed hands (times/day)[a]	77	4.4 (±3.1)	3	0	20
Number of times showered/bathed [a]	77	5.7 (±2.3)	6	1	14
Number of times taking food, drinks, pacifier outside to play[a]	74	3 (±3.9)	2	0	25
Dietary Characteristics (over past 7 days)					
Number of times eating homegrown vegetables/fruit[a]	77	1.3 (±2.7)	0	0	14
Number of servings of seafood[a]	77	0.2 (±0.5)	0	0	2
Number of servings of rice/rice products[a]	77	0.9 (±1.4)	0	0	5
Number of servings of organ meats	77	0.01 (±0.1)	0	0	1
Number of cups of tap water or drinks with tap water per day[a]	77	3.5 (±3.7)	3	0	20
Number of servings of grapes	74	0.5 (±0.9)	0	0	4

Note: SD - standard deviation

[a] Included in inferential statistical analysis.

Table 13. Categorical questionnaire responses and associated urinary arsenic levels for children less than 7 years old

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Gender[b]	Female	40	51.9	0.85 (±1.51)	3.80 (±2.46)	5.30 (±2.11)
	Male	37	48.1	0.76 (±1.42)	4.25 (±2)	5.14 (±1.67)
Location of outdoor area where child played most often?	Your yard	55	71.4	0.82 (±1.43)	4.3 (±2.15)	4.81 (±1.76)
	A neighbor's yard	1	1.3	1.71 (--)	11.17 (--)	13.92 (--)
	School Yard	0	0.0	-- (--)	-- (--)	-- (--)
	Elsewhere	8	10.4	0.82 (±1.8)	4.25 (±2.54)	5.84 (±2.2)
	Did not play outside	7	9.1	0.68 (±1.41)	1.84 (±2.41)	7.4 (±2.8)
	Multiple Locations	6	7.8	0.71 (±1.29)	4.13 (±1.69)	5.37 (±1.53)
Description of ground where child played?[b]	Dirt/uncovered soil	1	1.4	1.08 (--)	1.53 (--)	1.91 (--)
	Pavement	1	1.4	1.20 (--)	9.15 (--)	7.43 (--)
	Grassy lawn	21	30.0	0.91 (±1.42)	4.43 (±2.1)	4.87 (±1.72)
	Lawn with bare areas	36	51.4	0.76 (±1.52)	4.05 (±2.2)	4.77 (±1.79)
	Gravel	1	1.4	0.77 (--)	3.47 (--)	2.59 (--)
	Other	10	14.3	0.83 (±1.43)	5.55 (±2.05)	7.47 (±1.76)
Child visited a house/building with ongoing renovations?[b,c]	Yes	6	8.0	0.86 (±1.27)	7.93 (±1.62)	6.14 (±1.39)
	No	68	90.7	0.80 (±1.47)	3.76 (±2.21)	5.17 (±1.95)
	Don't know	1	1.3	1.14 (--)	±5.75 (--)	3.13 (--)
Play with family or neighbor's outdoor pet?[b]	Yes	50	64.9	0.78 (±1.51)	4.13 (±2.13)	5.33 (±1.79)
	No	27	35.1	0.86 (±1.4)	3.80 (±2.46)	5.02 (±2.1)

Table 13. (cont.)

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Attend daycare, preschool, etc?	Yes	23	29.9	0.80 (±1.48)	4.67 (±1.94)	5.50 (±1.6)
	No	54	70.1	0.81 (±1.47)	3.76 (±2.36)	5.11 (±2.02)
Limit child's exposure to soil or dust?[b]	Yes	5	6.6	0.83 (±1.47)	2.18 (±2.76)	3.9 (±2.2)
	No	71	93.4	0.79 (±1.46)	4.12 (±2.17)	5.28 (±1.88)
Play near creeks, streams, or tributaries?[b]	Yes	10	13.0	0.75 (±1.53)	4.23 (±2.46)	4.87 (±1.65)
	No	67	87.0	0.81 (±1.47)	3.98 (±2.22)	5.28 (±1.94)
Spend time in local orchard or produce farm?[b,c]	Yes	3	3.9	0.82 (±1.21)	5.43 (±1.04)	4.88 (±1.35)
	No	73	96.1	0.81 (±1.49)	3.96 (±2.28)	5.22 (±1.93)
Exposed to treated wood?[b]	Yes	53	68.8	0.81 (±1.54)	4.44 (±2.31)	5.36 (±1.88)
	No	24	31.2	0.8 (1±.32)	3.19 (±1.99)	4.93 (±1.95)
Child near project where treated wood was sanded, etc.?	Yes	3	3.9	0.85 (±1.12)	2.55 (±1.87)	3.93 (±1.65)
	No	74	96.1	0.8 (±1.48)	4.08 (±2.25)	5.28 (±1.91)
Treated wood used as firewood?	Yes	1	1.3	0.91 (--)	1.89 (--)	2.72 (--)
	No	73	94.8	0.81 (±1.47)	4.1 (±2.23)	5.32 (±1.9)
	Don't know	3	3.9	0.73 (±1.68)	3.02 (±2.73)	4.16 (±1.94)
Use pacifier?	Yes	7	9.1	0.56 (±1.45)	1.42 (±1.62)	5.23 (±1.94)
	No	70	90.9	0.83 (±1.45)	4.45 (±2.13)	5.22 (±1.9)
How often did child suck thumb/fingers or chew fingernails?[b]	>1x/hour	10	13.0	0.84 (±1.44)	3.25 (±2.47)	5.48 (±2.1)
	>1x/day	15	19.5	0.89 (±1.43)	4.7 (±2.47)	6.48 (±2.11)
	>1x/week	14	18.2	0.79 (±1.55)	3.21 (±2.35)	5.69 (±2.13)
	Not at all	31	40.3	0.76 (±1.5)	4.18 (±2)	4.40 (±1.62)
	Don't know	7	9.1	0.84 (±1.38)	4.99 (±2.39)	5.49 (±1.83)

Table 13. (cont.)

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
How often put objects (other than food) into mouth[b]	>1x/hour	10	13.0	0.86 (±1.44)	3.64 (±2.31)	6.53 (±1.67)
	>1x/day	16	20.8	0.84 (±1.48)	3.72 (±2.52)	5.58 (±2.23)
	>1x/week	17	22.1	0.73 (±1.57)	3.55 (±2.59)	5.30 (±2.24)
	Not at all	24	31.2	0.74 (±1.39)	3.92 (±1.83)	4.43 (±1.58)
	Don't know	10	13.0	1.02 (±1.41)	6.46 (±2.04)	5.43 (±1.71)
Breastfeeding status	Table fed only	63	87.5	0.82 (±1.5)	4.48 (±2.2)	5.01 (±1.83)
	Breast fed only	3	4.2	0.55 (±1.35)	1.53 (±2.19)	4.35 (±2.55)
	Bottle fed only	2	2.8	0.67 (±1.03)	1.23 (±1.08)	18.65 (±2.87)
	Combination	4	5.6	0.89 (±1.33)	3.36 (±2.15)	7.65 (±1.99)
Eaten vegetables or fruits from a home garden?[b]	No	55	71.4	0.80 (±1.5)	3.96 (±2.31)	5.19 (±1.97)
	Yes	22	28.6	0.83 (±1.42)	4.12 (±2.1)	5.30 (±1.71)
Eaten seafood?[b]	No	61	79.2	0.79 (±1.48)	3.84 (±2.27)	5.14 (±1.93)
	Yes	16	20.8	0.87 (±1.43)	4.73 (±2.12)	5.52 (±1.81)
Eaten rice or rice products?[b]	No	47	61.0	0.76 (±1.38)	3.59 (±2.25)	5.05 (±1.98)
	Yes	30	39.0	0.88 (±1.58)	4.76 (±2.18)	5.50 (±1.78)
Taken herbal, traditional, folk, or imported remedies, etc?	Yes	5	6.6	0.76 (±1.14)	3.85 (±1.65)	4.53 (±1.45)
	No	71	93.4	0.81 (±1.49)	4.02 (±2.29)	5.28 (±1.93)
Family income for 2002[b]	≤$40,000	24	32.4	0.89 (±1.47)	4.1 (±2.33)	4.84 (±1.91)
	>$40,000	50	67.6	0.76 (±1.45)	3.8 (±2.21)	5.36 (±1.92)

Table 13. (cont.)

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Year home built[b]	Before 1940	40	51.9	0.81 (±1.46)	4.16 (±2.18)	5.34 (±2.05)
	1940–1980	22	28.6	0.76 (±1.46)	3.58 (±2.27)	4.81 (±1.63)
	After 1980	5	6.5	0.72 (±1.71)	3.56 (±2.9)	7.16 (±2.41)
	Don't know	10	13.0	0.94 (±1.45)	4.69 (±2.32)	4.88 (±1.66)
Street paved near home?	Yes	76	100	0.81 (±1.48)	3.99 (±2.25)	5.24 (±1.90)
Large projects involving digging, moving or adding soil?[b]	Yes	34	44.2	0.88 (±1.4)	4.78 (±2.07)	5.62 (±1.82)
	No	41	53.2	0.74 (±1.52)	3.45 (±2.37)	5.06 (±1.97)
	Don't know	2	2.6	0.94 (±1.32)	4.47 (±1.43)	2.85 (±1.14)
Family of Hispanic origin?	No	77	100	0.81 (±1.47)	4.01 (±2.24)	5.22 (±1.90)
Which group describes your family?	White	72	93.5	0.81 (±1.49)	3.96 (±2.27)	5.21 (±1.94)
	African American	3	3.9	0.72 (±1.27)	6.84 (±1.26)	4.88 (±1.03)
	Other	2	2.6	0.7 (±1.17)	2.9 (±2.17)	6.26 (±1.16)
Participate in WIC?	Yes	5	6.5	0.83 (±1.68)	3.34 (±3.15)	5.07 (±3.29)
	No	72	93.5	0.80 (±1.46)	4.06 (±2.19)	5.23 (±1.82)
Exposure to smoke in past 7 days[b]	Yes	24	31.2	0.76 (±1.50)	3.46 (±2.19)	4.25 (±1.94)
	No	39	50.6	0.81 (±1.44)	4.15 (±2.32)	5.66 (±1.82)

[a] Total number of subjects that responded to each question varied.

[b] Included in inferential statistical analyses.

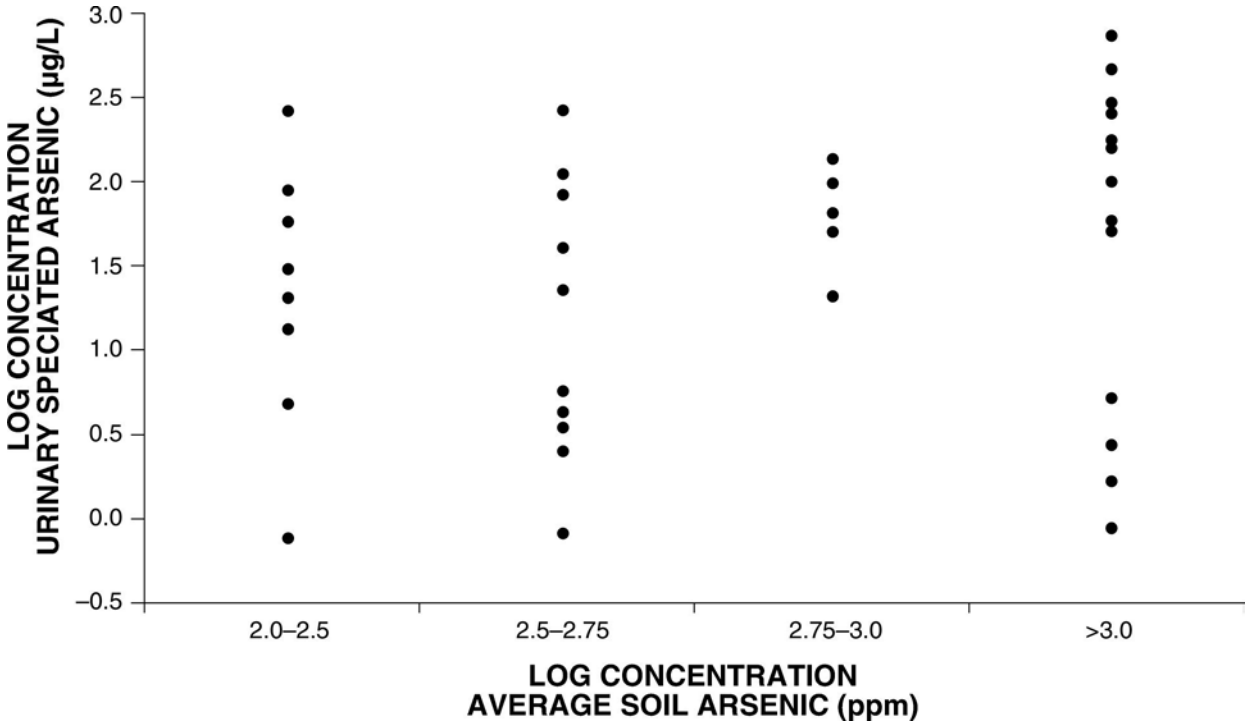
[c] Significant difference in speciated urinary arsenic levels between "Yes" and "No" responses (*t*-test; $p < 0.05$).

3.3.5 Correlation among Arsenic in Urine, Soil, House Dust and Indirect Exposure Indicators

Results and description of the examination of the relationship between urinary arsenic levels and environmental arsenic concentrations in the target population of children less than 7 years old are presented below. As a secondary analysis, we also evaluated this relationship in children less than 13 years old and in the total study population. A brief description of our results for these two populations is presented in Sections 3.3.5.2 and 3.3.5.3, respectively; complete tabular results are presented in Appendix G.

3.3.5.1 Children Less Than 7 Years Old

Visual examination of a series of scatter plots showing a dependent variable on the y-axis (e.g., arsenic in urine) and arsenic in soil or house dust on the x-axis indicated a random pattern of association for the variables. An example of this randomness is illustrated in Figure 7, which displays speciated urinary arsenic levels by average arsenic concentration ranges in soil. Even though the scatter plots did not indicate an association between the primary dependent and independent variables, we continued with the inferential analyses discussed in Section 2.8.2.



Note: Soil concentration data are displayed as categories to protect the identity of the study participants. Correlations and regression analyses were calculated on actual soil concentration and urine data.

Figure 7. Speciated urinary arsenic levels in children less than 7 years old versus the average arsenic concentration in soil

The results of the bivariate Pearson correlation analysis are presented in Table 14. Correlations between either primary outcome variable (speciated or inorganic arsenic in urine) and the level of arsenic in soil or house dust were not significantly different from zero. Speciated urinary arsenic levels had the highest correlation with mean soil arsenic level, although still weakly correlated and not statistically significant (r=0.137; p=0.392). When speciated arsenic was corrected for creatinine, the only significant correlation was with the arsenic concentration in house dust (r=0.301; p=0.030). Arsenic in house dust (concentration or loading) was not correlated with measures of arsenic in soil (average or maximum composite soil concentration) (p=0.12 to 0.96). Of the numerical or continuous exposure factors derived from questionnaires, only age (r=0.331; p=0.003) and body weight (r=0.253; p=0.029) were significantly associated with speciated urinary arsenic levels. Both of these factors were positively correlated with speciated urinary arsenic levels but negatively associated with speciated arsenic levels corrected for creatinine. The significant correlation with number of days spent in outdoor areas was in the opposite direction from that expected for soil exposure: Children who spent more time in an outdoor area had lower inorganic arsenic in urine (r=–0.24, p<0.05).

Analysis of the categorical exposure variables derived from the questionnaires showed few significant associations between urinary arsenic levels and variables indicative of potential exposure to arsenic (Table 13). Speciated urinary arsenic levels were significantly higher among those children who had been reported to spend any time in a local orchard or produce farm in the past 7 days (t-test; p=0.002). Speciated urinary arsenic levels were also significantly higher (t-test; p=0.027) among children who reported having "…visited a home or building with ongoing renovations that generated a lot of dust in the home or building" over the past 7 days. However, it is important to note that a "yes" response to spending time in an orchard was reported for only three children, whereas only six children were reported to have visited a building with ongoing renovations. Inorganic arsenic and speciated arsenic corrected for creatinine in urine were not significantly associated with any of the categorical exposure variables.

Based on the criteria described previously, age-adjusted regression models failed to indicate an association between each of the dependent variables and the environmental variables. A "base" model could not be established and therefore further multiple regression models were not run.

3.3.5.2 Children Less Than 13 Years Old

The results of the inferential analysis of children less than 13 years old were very similar to those seen in children less than 7 years old (see Appendix G for statistical details of results). Because of differences in questionnaires between the young children (less than 7 years) and the older children/adults (7 years and older), several exposure factors were not included in the inferential analysis (e.g., weight, thumb-sucking). The results of the bivariate Pearson correlation analysis showed that correlations between either primary outcome variable (speciated or inorganic arsenic in urine) and the level of arsenic in soil or house dust were not significantly different from zero. Similar to children less than 7 years old, speciated urinary arsenic levels in children less than 13 years old had the highest correlation with mean soil arsenic level, but was still weakly correlated and not significant (r=0.201; p=0.081). Creatinine-corrected speciated urinary arsenic levels were not significantly correlated with any direct measures of environmental arsenic levels. Of the indirect exposure factors derived from the questionnaire, only age (r=0.294; p<0.001) was significantly associated with speciated urinary arsenic levels.

Table 14. Correlation coefficient matrix of urinary arsenic levels, environmental arsenic levels, and numerical exposure factors for children less than 7 years old

	Urine Measurements			Environmental Measurements			
Exposure Factor	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)	Average of Yard, Play, and Garden Area (ppm)	Max of Yard, Play, and Garden Area (ppm)	Arsenic Concentration in House Dust (mg As/kg dust)	Surface Loading of Arsenic into House Dust (μg As/100 cm^2)
Speciated arsenic (μg/L)	0.736** (77)						
Speciated arsenic (creatinine corrected) (μg/g)	0.403** (77)	0.559** (77)					
Average of yard, play, and garden area (ppm)	0.230 (41)	0.137 (41)	-0.019 (41)				
Max of yard, play, and garden area (ppm)	0.062 (41)	0.045 (41)	-0.132 (41)	0.865** (41)			
Arsenic concentration in house dust (mg As/kg dust)	-0.060 (52)	0.049 (52)	0.301* (52)	-0.009 (34)	-0.056 (34)		
Surface loading of arsenic into house dust (μg As/100 cm^2)	0.081 (53)	0.090 (53)	0.232 (53)	-0.271 (35)	-0.233 (35)	0.607** (52)	
Age of child when urine was taken (years)	0.223 (77)	0.331** (77)	-0.263* (77)	0.248 (41)	0.198 (41)	-0.009 (52)	-0.045 (53)
Weight (lb)	0.136 (75)	0.253* (75)	-0.317** (75)	0.213 (41)	0.183 (41)	-0.095 (51)	-0.117 (52)
Body mass index	-0.070 (73)	-0.101 (73)	-0.103 (73)	-0.096 (41)	-0.167 (41)	-0.087 (51)	0.035 (52)
Number in household	0.020 (77)	0.010 (77)	-0.112 (77)	-0.173 (41)	-0.182 (41)	-0.201 (52)	-0.072 (53)
Time playing in outdoor area (days)	-0.240* (70)	-0.150 (70)	0.003 (70)	-0.165 (38)	-0.036 (38)	0.059 (48)	-0.053 (48)
Time playing in outdoor area (hours/day)	-0.009 (68)	-0.006 (68)	-0.110 (68)	0.072 (37)	0.201 (37)	0.124 (47)	0.019 (47)
Time spent in or near creeks, streams, etc. (days)	0.057 (10)	0.160 (10)	0.152 (10)	-0.134 (7)	0.075 (7)	-0.654 (8)	-0.527 (8)
Time spent in orchards, etc. (days)	-0.853 (3)	-0.484 (3)	-0.868 (3)	-- (1)	-- (1)	-- (1)	-- (1)
Washed hands (times/day)	-0.076 (77)	-0.052 (77)	-0.275* (77)	-0.289 (41)	-0.009 (41)	-0.035 (52)	0.113 (53)
Number of times showered/bathed in past 7 days	-0.025 (77)	-0.025 (77)	-0.175 (77)	0.12 (41)	0.241 (41)	-0.011 (52)	0.109 (53)
Number of times eating homegrown vegetables or fruits	-0.004 (77)	0.002 (77)	0.028 (77)	-0.177 (41)	-0.073 (41)	-0.405** (52)	-0.146 (53)
Number of servings of seafood	0.088 (77)	0.115 (77)	0.081 (77)	-0.169 (41)	-0.216 (41)	0.111 (52)	0.173 (53)
Number of servings of rice/rice products	0.157 (77)	0.190 (77)	0.173 (77)	0.197 (41)	0.345* (41)	-0.166 (52)	-0.311* (53)
Number of cups of tap water or drinks with tap water per day	0.023 (77)	0.094 (77)	-0.058 (77)	0.125 (41)	0.208 (41)	-0.055 (52)	0.354** (53)
Number of times taking food, etc., outside to play	0.101 (74)	-0.016 (74)	-0.081 (74)	0.036 (39)	0.125 (39)	0.101 (51)	0.077 (52)

Note: Numbers in parentheses indicate the sample sizes. Urinary and environmental variables were log-transformed before analysis.

* - $p < 0.05$
** - $p < 0.01$

Analysis of the categorical exposure variables derived from the questionnaires showed few significant associations of urinary arsenic levels with potential exposure to arsenic. As seen among the children less than 7 years old, speciated urinary arsenic levels were significantly higher among those children who had been reported to spend any time in a local orchard or produce farm in the past 7 days (geometric mean = 5.43 μg/L; N=3) compared to levels in children who did not spend any time in an orchard (geometric mean = 4.51 μg/L; N=135) (*t*-test; p=0.009). Inorganic, but not speciated, urinary arsenic levels were slightly higher among those children who lived in residences that underwent large projects that involved digging or adding soil to their yard in the past 12 months (geometric mean = 0.89 μg/L; N=68) compared to inorganic urinary arsenic levels in children whose residences had no reported digging projects (geometric mean = 0.78 μg/L; N=72) (*t*-test; p=0.028). This exposure factor was "borderline" significantly associated with inorganic urinary arsenic levels (p=0.061) among children less than 7 years old. Visiting a house with ongoing renovations was no longer significantly associated with urinary arsenic levels among children less than 13 years (*t*-test; p=0.639).

Age-adjusted regression models failed to indicate an association between each of the dependent variables and the environmental variables and therefore further multiple regression models were not run.

3.3.5.3 Total Study Population

An evaluation of the relationship between urinary arsenic levels and environmental measures in the total study population resulted in a similar lack of association as seen in the younger populations (see Appendix G for details of statistical results). The results of the bivariate Pearson correlation analysis showed that correlations between either primary outcome variable (speciated or inorganic arsenic in urine) and the level of arsenic in soil or house dust were not significantly different from zero. Speciated urinary arsenic levels in the total study population had the highest correlation (although very weak and still not significant) with arsenic levels in house dust (ppm) (r=0.110; p=0.068).

Among the indirect exposure factors derived from the questionnaires, some were found to be significantly correlated with urinary arsenic levels. However, many of these significant findings were associated with negative correlations that were not meaningful and/or estimated correlation coefficients of a smaller magnitude compared to the younger children, indicating the effect of sample size on detecting significant but small differences and also the effect of including less exposed individuals. For example, the number of reported times eating vegetables or fruits from a home garden was negatively correlated with speciated urinary arsenic levels (r=–0.097; p=0.043). Age was negatively and more weakly correlated with speciated urinary arsenic levels (r=–0.158; p<0.001) when compared to the results seen among the children less than 7 years old. Inorganic urinary arsenic levels were significantly although weakly associated with the number of people in the household (r=0.163; p<0.001) and age (r=–0.209; p<0.001).

In the total study population, males had slightly higher inorganic urinary arsenic (*t*-test; p=0.029) and speciated urinary arsenic levels (*t*-test; p=0.027). The mean inorganic urinary arsenic level in males was 0.81 μg/L compared to 0.76 μg/L in females while the speciated urinary arsenic level in males was 4.17 μg/L and 3.63 μg/L in females. Similar to children less than 13 years old, inorganic urinary arsenic levels in the total study population were slightly

higher among those subjects whose families reported large projects that involved digging or adding soil to their yards in the past 12 months (geometric mean = 0.81 μg/L; N=180) compared to inorganic urinary arsenic levels in subjects whose families did not report digging (geometric mean = 0.76 μg/L; N=252) (*t*-test; *p*=0.043). There was a significant difference in the speciated urinary arsenic levels between those subjects who reported smoking cigarettes, cigars, or a pipe over the past 7 days (or were exposed to smoke through a household member who reported smoking) and those who did not; however, this relationship was not in the expected direction. Those subjects who reported not smoking or had no household members who reported smoking over the past 7 days had significantly higher speciated urinary arsenic levels (geometric mean = 4.02 μg/L; N=309) compared to those who did smoke or were exposed to smoke (geometric mean = 3.45 μg/L; N=111) (*t*-test; *p*=0.036). This relationship was similar among children less than 7 years old; however, the difference in young children's speciated urinary arsenic levels was slightly larger and not significant.

Similar to the other population groups, age-adjusted regression models failed to indicate an association between each of the dependent variables and the environmental variables and therefore further multiple regression models were not run.

4 Discussion

This environmental exposure investigation provided individual participants with information on biomarker levels of arsenic and, if elected, arsenic concentrations in soil, house dust, and garden vegetables. Statistical analyses were also conducted to determine correlations between urinary arsenic levels and arsenic levels in soil and house dust and associations with survey responses that might affect arsenic exposure, including those that were a concern to the community based on questions at community meetings (e.g., eating vegetables, playing in creeks). Interpretation of biomarker levels and statistical analyses, including uncertainties, is discussed below.

4.1 Biomarker Arsenic Levels

The biomarker results for all study participants showed low levels of arsenic in urine that were well below reference levels used to screen for elevated inorganic arsenic exposure and a need for case follow-up. This study was able to examine inorganic arsenic exposure more specifically than the previous biomonitoring study of students of Royalton-Hartland School because it measured more than just the total amount of arsenic in urine. The current investigation also analyzed the specific forms of arsenic that result in the urine from ingestion of inorganic arsenic, the type found in soil and water.

The urinary profile of total arsenic forms and arsenic species indicated ingestion of organic arsenic forms from foods such as seafood, but no evidence of excess inorganic arsenic exposure such as from soil. Because inorganic arsenic also occurs naturally in food and water (ATSDR 2000b; Schoof et al. 1999; Yost et al. 2004), detectable low levels of speciated arsenic are expected in urine. Ingestion of some types of seafood containing arsenosugars (e.g., mussels, clams, oysters, and seaweed, as well as freshwater shellfish and commercial food products containing algae) can also increase DMA levels in urine (Le et al. 1999), although the overall effect of seafood ingestion on speciated arsenic levels is far less than on total arsenic levels in urine. Polissar et al. (1990) report that consumption of shellfish and, to a lesser extent, finfish, significantly elevated methylated (e.g., DMA) and inorganic arsenic species in urine. Many of the arsenic compounds found in marine organisms have also been identified in terrestrial organisms (Irgolic et al. 1999).

Among all participants, those with higher total arsenic levels in urine often had higher DMA levels as well but relatively low inorganic arsenic levels, which indicates that seafood or some other dietary source likely increased DMA levels for these individuals. Those who reported ingesting seafood within the last week before sampling had overall higher total arsenic levels in urine, although speciated urinary arsenic was not elevated for those who reported consumption of seafood in any of the three age groups evaluated. Not all individuals whose urinary profiles (i.e., levels of total arsenic and arsenic species) reflected organic arsenic from dietary sources reported consumption of seafood, however.

Arsenic in urine is generally considered the most reliable biomarker of recent arsenic exposure and thus has been the measure of choice in arsenic exposure studies (ATSDR 2000a,b). Most of

the absorbed arsenic is eliminated (mainly through urinary excretion) in the first 2 days (ATSDR 2000b), although urinary arsenic levels may be elevated over baseline up to a week after exposure. Nevertheless, urinary arsenic levels are unlikely to significantly underrepresent exposure to soil because sampling was conducted in 77 young children (approximately half the population of young children in the community) during the time of year when exposure to soil is expected to be greatest.

Although urinary arsenic is considered the most reliable biomarker for recent arsenic exposure, it is subject to considerable natural individual variation due to diet, water, or other sources. Thus, this biomarker represents a screening for potential excess exposure, and correlations with source exposure (e.g., soil) generally require population studies involving a number of individuals.

Urinary arsenic levels are considered most accurate when using 24-hour urine collection. However, programs involving more than a few individuals generally use first-morning void samples because 24-hour urine collection particularly from children is extremely inconvenient for participant families and the likelihood of missing samples is high (Hwang et al. 1997b). A study of young children conducted at Anaconda, Montana, used two consecutive, first-morning void urine samples for all subjects and 24-hour urine collection for a subset of subjects (Hwang et al. 1997a,b). Although the authors do not specifically report on the comparison between first-morning void samples and 24-hour collection, their use of the first-morning void data in the statistical analysis of exposure indicates confidence in these results as the best measure of urinary arsenic concentration. The authors of this study reported no differences in the statistical analysis of exposure between using the average of the two first-morning void samples or the highest of these samples.

Toenails were selected with reservation for biomonitoring in this investigation. This biomarker may provide a measure of longer-term exposure than urinary arsenic, although it is less easy to relate to a daily dose and therefore quantify exposure, and toenail samples may be contaminated by external adherence of arsenic that cannot be removed. Only a few biomonitoring studies for soil arsenic exposure have used toenails as a biomarker (e.g., Hewitt et al. 1995; Hinwood et al. 2003a), and these studies note these drawbacks. Toenail samples in the Middleport study also showed evidence of external contamination from arsenic in dust or soil based on the association of higher sample concentrations with higher discoloration score (Figure 6). Thus, this measure, although not elevated above the CDC reference level, is not considered a reliable indicator of ingested arsenic for this community. Another logistical drawback to toenail collection, particularly for children, was the amount of sample required for accurate analysis (0.5 g), which required several weeks to months of clipping. Of the six samples received from young children, none were of sufficient mass.

Like nails, hair arsenic has also been used in biomonitoring for arsenic exposure (ATSDR 2000b; Samanta et al. 2004) because it reflects a much longer period of exposure (e.g., the past month or two). However, hair is less sensitive to low-level arsenic exposure than is urine as noted in drinking water exposure studies (Valentine 1994), and few exposure studies of arsenic in soil have used this biomarker as compared to urine. Hair arsenic levels are also difficult to quantify in terms of a daily dose and are subject to external contamination (e.g., shampoos, dust; Hinwood et al. 2003a; Harkins and Susten 2003) that may be difficult to remove entirely by

washing without extracting some of the internally deposited arsenic (Hindmarsh et al. 1999). A panel of experts convened by ATSDR to evaluate the utility of hair analysis concluded that with the exception of methyl mercury, "hair is not a reliable indicator of environmental exposure or internal body burden" and that "hair analysis, if conducted, should be viewed only as a supportive tool and the results put into perspective with other more reliable data (e.g., blood and urine concentrations)" (Harkins and Susten 2003). For these reasons, and because a similar measure, toenail analysis, was used, hair collection was not performed in this study.

4.2 Comparison of Middleport Urinary Arsenic Levels to Other Sites

Speciated urinary arsenic levels of young children (i.e., less than 7 years of age) in Middleport were low in comparison with such levels reported for children at other sites involving elevated soil arsenic exposures as well as for a population without such exposures (e.g., Bellingham, Washington) (Table 15). Some of the sites in other studies had soil concentrations that are considerably greater than in Middleport. Urinary arsenic levels from the study conducted in 1985–1986 at the Ruston/North Tacoma, Washington, smelter site also reflect an ongoing high air emission source of fine particulate arsenic in airborne and settled dust in the community. This smelter was closed while the 1985–1986 study was being conducted (Polissar et al. 1987). Urinary results collected later in 1987 in the Ruston/North Tacoma community are considerably lower despite no change in soil concentrations (TPCHD 1988; Table 15). Biomonitoring at the Anaconda, Montana, smelter site was conducted more than a decade after the smelter closed and urinary results for children at that site are also lower than for children in Ruston/North Tacoma in 1985–1986.

Speciated urinary arsenic levels were considerably higher for young children than for adults in the 1985–1986 Ruston/North Tacoma study (Polissar et al. 1987). By contrast, speciated levels were similar between young children and older ages in Middleport (*t*-test; p=0.665), thereby reflecting little exposure to arsenic in soil and dust in Middleport.

Middleport urinary arsenic levels for all ages combined are also low and consistent with results reported for unexposed populations. Speciated urinary arsenic levels for 61 persons, all ages combined, from Bellingham, Washington (median = 9.5 μg/L, range 0.9–33 μg/L; Polissar et al. 1987, 1990) were higher than for all Middleport participants (median = 4.2 μg/L, range 0.89–19.9 μg/L). Two studies in Australia of exposure to arsenic in soil (Hinwood et al. 2004) and in drinking water (Hinwood et al. 2003b) report inorganic arsenic levels in urine for adults and children combined. The two studies appear to have used the same control, or "unexposed," group of 52 people. Average inorganic arsenic levels in five first-morning void samples collected in four seasons for the unexposed Australian group (geometric mean = 1.18 μg/L, range = <2 to 2.81 μg/L) were similar to inorganic arsenic levels of participants (all ages) in the Middleport study (geometric mean = 0.78 μg/L, range = 0.31–2.7 μg/L). Reported levels of speciated urinary arsenic for a randomly selected subset of 10 samples for this unexposed Australian group (geometric mean = 31.8 μg/L; range = 13.9–57.9 μg/L) were considerably higher than for Middleport (geometric mean = 3.9 μg/L, range = 0.89–19.9 μg/L). Reported

Table 15. Speciated urinary arsenic and soil arsenic levels for young children less than 7 years old at various sites

	Speciated Urinary Arsenic µg/L (ppb)			Soil Arsenic Concentration mg/kg (ppm)		
	N	Geometric Mean (±GSD)	Range	N	Geometric Mean (±GSD)	Range
Middleport, NY 2003 (Exponent)						
Biomonitoring study	77	4.0 (±2.2)	0.89–17.7	29	19.9 (±1.6)	10.4–58.8
Bingham Creek, UT (UCDEH 1997)						
Residences near Bingham Creek channel	696	5.86 (±1.96)	ND–35	1,045	27 (±1.8)	4–623
Ruston/North Tacoma, WA 1985–1986 (Polissar et al. 1987)						
<0.5 miles from smelter	118	52.1 (±42.5)[a]	NR	45	352 (±410)[b]	12–2,069
0.5–1.2 miles from smelter	97	22.5 (±29.3)[a]	NR	40	125 (±109)[b]	9–1,322
1.5–8.5 miles from smelter	49	13.7 (±10.3)[a]	NR	34	29.6 (±49)[b]	2–290
Reference site (Bellingham, WA) >100 miles from smelter	4	13.3 (±3.3)[a]	NR	10	6.6 (±2.7)[b]	2–10
Ruston/North Tacoma, WA 1987 (TPCHD 1988)						
<0.5 miles from smelter	88	16.2 (±16)	NR	NR	NR	NR
Anaconda, MT (Hwang et al. 1997a,b)						
Close to smelter	177	9.5 (±1.7)	NR–11.4	873	281[c]	NR
Intermediate	62	7.5 (±1.5)	NR–9.2	405	146[c]	NR
Remote	42	7.1 (±1.8)	NR–9	302	89[c]	NR

Note: GSD - geometric standard deviation
N - number of children
ND - not detected
NR - not reported

[a] Speciated urine values for Ruston/North Tacoma and Bellingham are the weighted arithmetic average and standard deviation derived from separate results for male and female.

[b] Average yard soil arsenic concentrations for Ruston/North Tacoma and Bellingham are the arithmetic average and standard deviation.

[c] Average yard soil arsenic concentrations for Anaconda are the geometric mean calculated as the weighted average of all soil samples.

soil and dust arsenic levels for this Australian group were overall lower (geometric mean = 4.3 ppm, range = 1.7–80 ppm; geometric mean = 3.9 ppm, range = 2.2–21 ppm; respectively; Hinwood et al. 2003b) than for the Middleport study (see Table 9).

Residents monitored at the Spring Valley neighborhood site in Washington, DC, by ATSDR were found to have speciated urinary arsenic levels as high as 29 μg/L (ATSDR 2003). ATSDR (2003) concluded for the Spring Valley site that "urine arsenic levels in this exposure investigation show low levels of exposure consistent with what might be found in the general population. These levels would not be expected to cause any health problems."

Therefore, although ATSDR and CDC have not established a standard reference level for speciated urinary arsenic, the levels measured in Middleport participants appear to be within background levels found in the general population as compared to a few studies on speciated and inorganic urinary arsenic levels in unexposed populations and according to the judgment of ATSDR investigators.

4.3 Sources and Factors Potentially Affecting Arsenic Exposure

This cross-sectional study examined a number of potential factors that might affect arsenic exposure for young children exposed to varying environmental levels of arsenic in soil within the Middleport community. Several elements of the study increased the likelihood of detecting exposures from arsenic in soil: 1) the study focused on the age group with greatest soil ingestion rates (i.e., children less than 7 years old); 2) biomonitoring was conducted during summer when soil and dust exposures would be highest; 3) urinary samples were analyzed for the specific forms of arsenic related to inorganic arsenic exposure; and 4) the study design examined the statistical relationship between environmental samples (e.g., soil and house dust arsenic concentrations) and individual urinary arsenic levels, including examination of potential confounding factors such as gender, age, smoking, diet, and other characteristics or sources of arsenic exposure.

On the contrary, public awareness may have resulted in decreased arsenic exposure in the community by temporarily affecting people's behavior. If such an impact occurred, however, it was not detectable. The responses from the exposure questionnaires indicate that very few parents attempted to limit their children's exposure to soil (5 out of 76 for children less than age 7 and 8 out of 135 for children less than age 13). No significant difference was found in urinary levels between children of either age groupings (less than 7 years or less than 13 years) whose exposure to soil was limited and those whose exposure was not. Moreover, controlling behaviors related to soil and dust contact by young children is difficult. The impact of general public awareness has been examined in other larger studies with similar findings of no impact on speciated urinary arsenic levels or blood lead levels (UCDEH 1997).

The cross-sectional study design allows a more sensitive assessment of the effect of arsenic in soil on community exposure than provided by a simple comparison of mean urinary results between Middleport and a control community. Such a comparison of two communities is statistically weaker and problematic because of the potential differences in sources of arsenic

and other factors that might affect biomarker results, but are difficult to measure or control. A "control" community is also less likely to participate in the efforts required to collect sample and questionnaire data. Because the study focused on Middleport residents over a range of arsenic soil concentrations in the community, an unexposed reference population was unnecessary.

The statistical analysis of the study results was intended to examine two separate aspects of the correlation between arsenic in urine and arsenic in soil: 1) whether arsenic in urine is significantly and positively correlated with arsenic in soil and/or potentially related environmental measures (e.g., house dust); and 2) what proportion of the variation in urinary arsenic can be explained by arsenic in soil or other environmental measures and what is the slope of this relationship (i.e., how much does the urinary arsenic level increase with increase in arsenic in soil or house dust). This study also examined whether arsenic in soil was correlated with house dust concentrations of arsenic or surface loading of arsenic on interior floors. House dust is considered a more proximal source of exposure to metals than soil for young children (Succop et al. 1998).

4.3.1 Correlations between Environmental Arsenic and Urinary Arsenic Levels

Few significant correlations were found in the target population of children less than 7 years old, the age group most exposed to soil. Overall, the results of this analysis are consistent with similar detailed evaluations at other sites involving larger numbers of young children and generally higher arsenic exposures than in Middleport. Specifically, a lack of a significant correlation between arsenic in urine and environmental levels of arsenic (e.g., in soil and house dust) reflects the relatively low levels of arsenic for this community compared to other communities such as Anaconda, Montana. The relatively smaller sample size of young children (i.e., less than 7 years old) with urinary data who also had yard soil data (N=41), house dust data (N=52), or complete urine, soil, and house dust data (N=34) limited the statistical power of the Middleport analysis to detect significant correlations between urinary arsenic and environmental measures, given the weakness of the correlations. For example, based on the estimated correlation coefficient between speciated urinary arsenic levels and the average arsenic concentration in soil (r=0.137), the sample size of participants less than 7 years would have to be larger (i.e., at least 203) than the estimated total population of young children in Middleport (i.e., 164) to be able to detect a significant correlation at the $\alpha = 0.05$ level with speciated urinary arsenic at these relatively low soil concentrations. To evaluate the potential effect of an increased sample size and to take into account different sources of potential exposure in an older population (e.g., playing in creeks, eating vegetables), secondary analyses were conducted on children less than 13 years old and on all participants. No significant correlations with soil or related environmental measures were found for these groups either.

At the Anaconda, Montana, site, a significant but relatively weak correlation (r =0.12 to 0.25) between speciated urinary arsenic in young children (age 24 to 72 months) and arsenic in soil but not in house dust was reported, with the highest correlation found for bare areas of soil (226 children with urine and bare soil arsenic data; Hwang et al. 1997a,b). The slope of this relationship was small such that an increase in soil arsenic concentration from 10 to 100 ppm

would increase speciated urinary arsenic by only 0.85 μg/L. Hwang et al. (1997a) attribute this small effect of arsenic in soil on arsenic in urine to low bioavailability (i.e., low amount of ingested arsenic that is absorbed into the blood stream) of arsenic in soil. By contrast, a significant relationship between speciated urinary arsenic and soil arsenic levels was not found for 696 children in Bingham Creek, Utah, where soil arsenic levels were lower than Anaconda, Montana. Thus, although urinary arsenic has been associated with arsenic concentrations in drinking water (e.g., Valentine 1994; Hinwood et al. 2003b), the association with arsenic in soil is much weaker and often unobservable, particularly for a community with relatively low soil arsenic levels such as Middleport. Any contribution of arsenic in soil to urinary arsenic would likely be small compared to the natural variation in urinary arsenic levels due to diet and water. Diet and water are the primary sources of inorganic arsenic exposure for the general population (Schoof et al. 1999; Meacher et al. 2002; Yost et al. 2004).

4.3.2 Correction for Hydration State and Other Uncertainties

We used creatinine-corrected speciated urinary arsenic data in a secondary analysis in an attempt to control for the effect of hydration state on urinary arsenic concentration in children less than 7 years old. Normalizing urinary arsenic concentrations to creatinine concentration in the urine sample (i.e., micrograms of arsenic per gram of creatinine in urine) has been used to correct for variable dilutions among spot urine samples. Because urinary arsenic concentration is expressed as micrograms of arsenic per liter of urine, a more dilute spot urine sample will result in a lower arsenic concentration.

Creatinine correction did not increase the magnitude of the estimated correlation between urine and soil arsenic levels. In fact, the relationship between creatinine-corrected speciated urinary arsenic levels and average or maximum soil arsenic level became negative such that lower soil arsenic levels were correlated (not significantly) with higher urinary arsenic levels. By contrast, the non-significant correlation with speciated urinary arsenic and house dust arsenic concentration became significant after creatinine correction and indicated an increase in the correlation coefficient from 0.049 to 0.301. The correlation between speciated urinary arsenic and age changed from a positive correlation to a negative correlation, which is more consistent with findings of Hwang et al. (1997b). However, whether the creatinine-corrected correlation in fact represents an actual correlation between arsenic in urine and house dust or is an artifact of variation introduced by creatinine correction is unclear. Studies with much larger sample sizes of young children (e.g., Hwang et al. 1997a,b; UCDEH 1997) did not find that creatinine correction improved the correlation between urinary arsenic and soil or house dust, and speciated urinary arsenic was not significantly correlated with house dust arsenic levels at the Anaconda, Montana, site (Hwang et al. 1997a). In addition, even if such a correlation exists, it is unclear if this exposure is related to arsenic in soil because arsenic levels in soil and house dust were not significantly correlated in Middleport.

Although creatinine correction of urinary levels of metals is often used to correct for hydration state, creatinine excretion levels can vary with gender, age, diet, genetic factors, or time and thus creatinine correction introduces unknown variation to the data (Boeniger et al. 1993). Thus, creatinine correction of urinary data particularly for the group of all participants was

considered inappropriate because of these differences, particularly between older children and younger children and between adults and children.

Uncertainties in quantifying actual environmental exposure concentrations must be acknowledged. Quantifying the exact concentrations is difficult unless each individual child is followed for a period of time prior to biomonitoring. Young children, however, are more likely to be exposed to their immediate home environment and the compositing of soil samples is more likely to represent exposure over a yard than a few discrete point samples. This study also examined correlations between urinary arsenic levels and the average of play area, garden, or yard samples as well as the maximum among these samples. Other indirect indicators of soil exposure (e.g., geographic distribution of urinary arsenic results, mouthing behavior, washing/bathing, taking food outside, indoor/outdoor pets, time spent in the area, digging, eating garden vegetables, correlation of house dust with soil) were also examined to evaluate whether urinary arsenic levels were elevated by soil exposure.

For several participants, house dust samples were not collected as planned soon after urinary sampling because of logistical and scheduling difficulties. Collection of house dust more than a month after urinary sampling is not expected to affect the arsenic concentration in house dust as much as it would for arsenic loading, unless some increased source of arsenic to indoor dust occurred (e.g., burning treated wood in fall and winter). Because arsenic loading is a function of arsenic concentration in the dust and the amount of dust on the floor, loading is more sensitive to changes such as cleaning or reduced transfer of dust from outdoors to indoors. The house dust loading data are thus likely to have considerable uncertainty for representing conditions at the time of urinary sampling.

4.3.3 Indirect Indicators of Potential Arsenic Exposure

Although a number of factors that potentially affect exposure to arsenic in soil or from diet or water were examined (Tables 12 and 13), few were found to significantly affect urinary arsenic levels in children less than 7 years old. Indirect indicators of soil exposure from the geographic distribution of urinary data (Appendix E) and the questionnaire responses also did not provide evidence of increased arsenic exposure. Unlike the direct correlations with soil data, these indirect analyses included data from nearly all 77 participants less than age 7. Positive responses to questions on whether participants consumed vegetables from their garden, played in creeks, spent more time outdoors, or frequently put hands or objects in the mouth were not found to be associated with higher urinary arsenic levels. Visiting an orchard within the week prior to sampling was found to be significantly associated with increased speciated urinary arsenic concentration in young children; however, very few children visited orchards. Orchards are a possible source of arsenic exposure in the area because of historical use of arsenic-containing pesticides. Consumption of garden vegetables has not been found to increase urinary arsenic levels at other sites (Polissar et al. 1987; Hwang et al. 1997a; UCDEH 1997), although increased mouthing frequency in children has been associated with increased urinary arsenic levels (Hwang et al. 1997a; UCDEH 1997).

Because of the number of factors examined for association with various urinary arsenic levels (i.e., inorganic, speciated, creatinine-corrected speciated arsenic), the likelihood that significant

associations may be found by chance alone is high, and a significant correlation is not equivalent to causation. For example, in children less than 7 years old, the number of servings of rice products consumed was significantly correlated with the maximum composite arsenic concentration in soil. One of the highest correlations for young children was between a higher consumption rate of homegrown vegetables and fruit and lower arsenic concentration in house dust. A few other associations were significant but in the opposite direction than expected: when all participants were considered, consumption of garden vegetables was significantly associated with a lower speciated urinary arsenic level. Fewer days spent outdoors by young children was correlated with higher inorganic arsenic levels in urine. Thus, with the large number of correlations and associations examined, isolated significant results must be interpreted with caution and do not necessarily indicate meaningful evidence of exposure. Overall, consistency in results that are indicative of soil exposure, for example, would be compelling evidence, but this was not observed.

Results of the statistical analyses including older children (i.e., less than 13 years old) and the total study population were overall consistent with findings for the target population of children less than age 7 (see Appendix G). The association between inorganic arsenic (but not speciated arsenic) and soil digging projects over the past year became significant for these analyses, which included older age groups; however, as noted in the results, the amount of increase in urinary inorganic arsenic was slight. In addition, soil digging projects that occurred over the past year would have had little effect on a short-term measure like urinary arsenic levels. The significant result for this question was included for completeness; however, this question was mainly asked in case of the need for individual follow-up. These analyses, which included a larger sample size of participants who played in creeks or ate garden vegetables, also showed no increase in urinary arsenic level for these activities. Age was positively correlated with speciated arsenic for the two age groups of children (i.e., less than age 7 and less than age 13) but negatively correlated for all participants. This change from a positive to negative correlation may reflect a nonlinear relationship between urinary arsenic levels and age over a wider age range, which could be a function of changes in behavior (affecting exposure), diet, or physiology with age. As described previously, the results that include participants age 7 and older must be interpreted with caution because of the potential for selection bias (i.e., lower participation rate than for young children) among the older participants.

5 Conclusions

Arsenic levels in urine and toenails for the Middleport study area were low and below program reference levels for the upper end of expected background levels in U.S. populations and not indicative of elevated exposure to inorganic arsenic of concern for case follow-up. Speciated and inorganic urinary arsenic levels were also low in comparison to other populations reported in the literature. Overall, no correlation was found in our target population (i.e., children less than 7 years old) between speciated or inorganic arsenic in urine and soil or house dust arsenic levels. There was also no evidence found from other indirect indicators of soil exposure such as geographic distribution of urinary arsenic levels in the community, garden vegetable consumption, or a variety of behaviors related to increased soil exposure, including playing in creeks. Expanding the age range to children less than 13 years old or to all participants did not materially change these findings.

The results of this study are consistent with studies at other sites involving much larger populations and ranges in arsenic soil concentrations. At these sites, the relationship of soil arsenic levels with speciated urinary arsenic levels is at best small (i.e., very small increase in urinary arsenic level with increase in soil arsenic level) and weak (i.e., most of the variation in urinary arsenic levels is not associated with soil arsenic levels). The lack of a strong relationship between arsenic in soil and arsenic in urine indicates that sources of inorganic arsenic other than soil (likely background levels in water and diet) are the primary determinants of speciated urinary arsenic levels.

6 References

ADHS and ATSDR. 2002. Public health assessment, ASARCO Hayden smelter site, Hayden, Arizona. Arizona Department of Health Services, Office of Environmental Health, Phoenix, AZ, and Agency for Toxic Substances and Disease Registry, Atlanta, GA.

APHA. 1995. Standard methods for the examination of water and wastewater. 19th Edition. American Public Health Association, Washington, DC.

ATSDR. 1998. Exposure investigation, Blackbird mine (a/k/a Panther Creek), Cobalt, Lemhi County, Idaho. CERCLIS #IDD980725832. www.atsdr.cdc.gov/HAC/pha/bird2/bla_toc.html. Last updated September 10, 1998. Accessed on June 26, 2003. Agency for Toxic Substances and Disease Registry, Exposure Investigation and Consultation Branch, Division of Health Assessment and Consultation.

ATSDR. 2000a. Case studies in environmental medicine: Arsenic toxicity. ATSDR Publication No. ATSDR-HE-CS-2002-0003. Agency for Toxic Substances and Disease Registry, Division of Health Education and Promotion.

ATSDR. 2000b. Toxicological profile for arsenic (update) 2000. U.S. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry.

ATSDR. 2002. Spring Valley neighborhood exposure investigation summary for the Science Advisory Board. www.atsdr.cdc.gov/NEWS/springvalley_06122002.html. Agency for Toxic Substances and Disease Registry.

ATSDR. 2003. Health consultation (exposure investigation—phase II) Spring Valley neighborhood (a.k.a. Spring Valley chemical munitions/American University), Washington, District of Columbia. June 11. Agency for Toxic Substances and Disease Registry.

Boeniger, M.F., L.K. Lowry, and J. Rosenberg. 1993. Interpretation of urine results used to assess chemical exposure with emphasis on creatinine adjustments: A review. Am. Ind. Hyg. Assoc. J. 54:615–627.

CDC. 2003a. Churchill County (Fallon), Nevada exposure assessment: Frequently asked questions about arsenic. www.cdc.gov/nceh/clusters/Fallon/faq-arsenic.htm. Last updated June 11, 2003. Accessed on June 26, 2003. Centers for Disease Control and Prevention.

CDC. 2003b. Cross-sectional exposure assessment of environmental contaminants in Churchill County, Nevada. Final Report. February 6, 2003. Centers for Disease Control and Prevention.

Conestoga-Rovers. 1999. FMC Middleport plant site, facility description and history. Prepared for FMC Corporation, Middleport, NY. Conestoga-Rovers & Associates.

Conestoga-Rovers. 2002. Private water well study report. Prepared for FMC Corporation, Middleport, NY. Ref. No. 2788-95. Conestoga-Rovers & Associates.

Exponent. 2003. Draft work plan for the Middleport environmental exposure investigation. Prepared for FMC Corporation, Philadelphia, PA. Exponent, Bellevue, WA.

Geomatrix. 2004. Draft Middleport biomonitoring soil sampling report, FMC Corporation, Middleport, New York. Project No. 7498. Geomatrix Consultants, Inc., Amherst, NY.

Harkins, D.K., and A.S. Susten. 2003. Hair analysis: Exploring the state of the science. Environ. Health Perspect. 11(4):576–578.

Hewitt, D.J., G.C. Millner, A.C. Nye, and H.F. Simmons. 1995. Investigation of arsenic exposure from soil at a Superfund site. Environ. Res. 68:73–81.

Hindmarsh, J.T., D. Dekerkhove, G. Grime, and J. Powell. 1999. Hair arsenic as an index of toxicity. pp. 41–49. In: Arsenic Exposure and Health Effects. W.R. Chappell, C.O. Abernathy, and R.L. Calderon (eds). Elsevier, Amsterdam.

Hinwood, A.L., M.R. Sim, D. Jolley, N. de Klerk, E.B. Bastone, J. Gerostamoulos, and O.H. Drummer. 2003. Hair and toenail arsenic concentrations of residents living in areas with high environmental arsenic concentrations. Environ. Health Perspect. 111(2):187–193.

Hinwood, A.L., M.R. Sim, D. Jolley, N. de Klerk, E.B. Bastone, J. Gerostamoulos, and O.H. Drummer. 2003b. Risk factors for increased urinary inorganic arsenic concentrations from low arsenic concentrations in drinking water. Environmental Health Research 13(3):271–284.

Hinwood, A.L., M.R. Sim, D. Jolley, N. de Klerk, E.B. Bastone, J. Gerostamoulos, and O.H. Drummer. 2004. Exposure to inorganic arsenic in soil increases urinary inorganic arsenic concentrations of residents living in older mining areas. Environmental Geochemistry and Health. 26:27–36.

Holland, J.P. 2002. Fifth annual report for the North Tacoma/Ruston medical monitoring program, January 2001 through December 2001. Occupational and Environmental Medicine Consulting, Seattle, WA.

Hwang, Y.H., R.L. Bornschein, J. Grote, W. Menrath, and S. Roda. 1997a. Environmental arsenic exposure of children around a former copper smelter site. Environ. Res. 72:72–81.

Hwang, Y.H., R.L. Bornschein, J. Grote, W. Menrath, and S. Roda. 1997b. Urinary arsenic excretion as a biomarker of arsenic exposure in children. Arch. Environ. Health. 52:139–147.

Irgolic, K.J., W. Goessler, and D. Kuehnelt. 1999. Arsenic compounds in terrestrial biota. pp. 61–68. In: Arsenic Exposure and Health Effects. W.R. Chappell, C.O. Abernathy, and R.L. Calderon (eds). Elsevier, Amsterdam.

Karagas, M.R., T.D. Tosteson, J. Blum, B. Klaue, J.E. Weiss, V. Stannard, V. Spate, and J.S. Morris. 2000. Measurement of low levels of arsenic exposure: a comparison of water and toenail concentrations. Am. J. Epidemiol. 152:84–90.

Lachell, W.C. 2004. Personal communication (e-mail to J. Tsuji, Exponent, Bellevue, WA, on March 30, 2004, regarding soil data for 14 Middleport, New York, properties remediated in 2003). Geomatrix Consultants, Inc., Amherst, NY.

Le, X.C., M. Ma, and V.W.-M. Lai. 1999. Exposure to arsenosugars from seafood ingestion and speciation of urinary arsenic metabolites. pp. 69–79. In: Arsenic Exposure and Health Effects. W.R. Chappell, C.O. Abernathy, and R.L. Calderon (eds). Elsevier, Amsterdam.

Meacher, D.M., D.B. Menzel, M.D. Dillencourt, L.F. Bic, R.A. Schoof, L.J. Yost, J.C. Eickhoff, and C.H. Farr. 2002. Estimation of multimedia inorganic arsenic intake in the U.S. population. Hum. Ecol. Risk Assess. 8(7):1697–1721.

NCWD. 2004. Annual drinking water quality report for 2003. Niagara County Water District. Lockport, NY.

NSDH and CBDHA. 2001. Lead and arsenic biological testing program in residential areas near the coke ovens site. Nova Scotia Department of Health and Cape Breton District Health Authority.

NYSDOH. 1987a. Incidence of cancer in the Village of Middleport (Niagara County) New York. Prepared by H.L. Howe, Ph.D., Director, Cancer Surveillance Program, Bureau of Cancer Epidemiology, New York State Department of Health.

NYSDOH. 1987b. Results of biological monitoring program for arsenic and lead: Middleport Elementary and Roy-Hart Jr/Sr High Schools. New York State Department of Health.

Polissar, L., D. Bolgiano, T.M. Burbacher, D.S. Covert, J.P. Hughes, D.A. Kalman, K.A. Lowry, N.K. Mottet, and G. van Belle. 1987. Ruston/Vashon arsenic exposure pathways study. Prepared for Washington State Department of Ecology, Olympia, WA. University of Washington, School of Public Health and Community Medicine, Seattle, WA.

Polissar, L., K. Lowry-Coble, D.A. Kalman, J.P. Hughes, G. van Belle, D.S. Covert, T.M. Burbacher, D. Bolgiano, and N. Karle Mottet. 1990. Pathways of human exposure to arsenic in a community surrounding a copper smelter. Environ. Res. 53:29–47.

Que Hee, S.S., B. Peace, C.S. Clark, J.D. Boyle, R.L. Bornschein, and P.B. Hammond. 1985. Evolution of efficient methods to sample lead sources such as house dust and hand dust in homes of children. Environ. Res. 38:77–97.

Samanta, G., R. Sharma, T. Roychowdhury, and D. Chakraborti. 2004. Arsenic and other elements in hair, nails, and skin-scales of arsenic victims in West Bengal, India. Sci. Total Environ. 326(1–3):33–47.

Sanderson, L.M., and S. Kess. 1995. Memorandum to G. Thomas, ATSDR Senior Regional Representative, U.S. EPA Region 10, dated March 23, 1995, regarding health consultation: exposure investigation Everett Smelter Site Everett, Washington. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry, Atlanta, GA.

Schoof, R.A., J. Eickhoff, L.J. Yost, E.A. Crecelius, D.W. Cragin, D.M. Meacher, and D.B. Menzel. 1999. Dietary exposure to inorganic arsenic. pp. 81–88. In: Arsenic Exposure and Health Effects. W.R. Chappell, C.O. Abernathy, and R.L. Calderon (eds). Elsevier, Amsterdam.

Succop, P., R. Bornschein, K. Brown, and C.Y. Tseng. 1998. An empirical comparison of lead exposure pathway models. Environ. Health Perspect. 106(suppl 6):1577–1583.

TPCHD. 1988. Urinary arsenic survey, North Tacoma, Washington. Prepared for Washington State Department of Ecology. Tacoma-Pierce County Health Department, Tacoma, WA.

UCDEH. 1997. Bingham Creek environmental health lead and arsenic exposure study. Final Report. University of Cincinnati, Department of Environmental Health, Cincinnati, OH.

U.S. Census. 2000. Table DP-1: Profile of general demographic characteristics, 2000. www.factfinder.census.gov. Accessed May 2003.

U.S. EPA. 1986. Test methods for evaluating solid waste: Physical/chemical methods (SW-846). U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, DC.

U.S. EPA, NYSDEC, and NYSDOH. 2003. FMC–Middleport, New York facility environmental investigations. EPA ID No. NYD002126845. Governmental Agencies' fact sheet. U.S. Environmental Protection Agency, New York State Department of Environmental Conservation, and New York State Department of Health.

Valentine, J.L. 1994. Review of health assessments for US/Canada populations exposed to arsenic in drinking water. pp. 139–152. In: Arsenic Exposure and Health. W.R. Chappell, C.O. Abernathy, and C.R. Cothern (eds). Science and Technology Letters, Northwood, UK.

WGI. 2001. Remedial investigation report-Vasquez Boulevard/I-70 Site, Operable Unit I. Washington Group International, Englewood, CO.

WHO. 2001. United Nations synthesis report on arsenic in drinking water. Chapter 3—Exposure and health effects. www.who.int/water_sanitation_health/Arsenic/ArsenicUNRep3.htm. Last updated in January 2001. Accessed June 23, 2003. World Health Organization.

Yost, L.J., S.-H. Tao, S.K. Egan L.M. Barraj, K.M. Smith, J.S. Tsuji, Y.W. Lowney, R.A. Schoof, and N.J. Rachman. 2004. Estimation of dietary intake of inorganic arsenic in U.S. children. Hum. Ecol. Risk. Assess. 10:473–483.

Appendix A

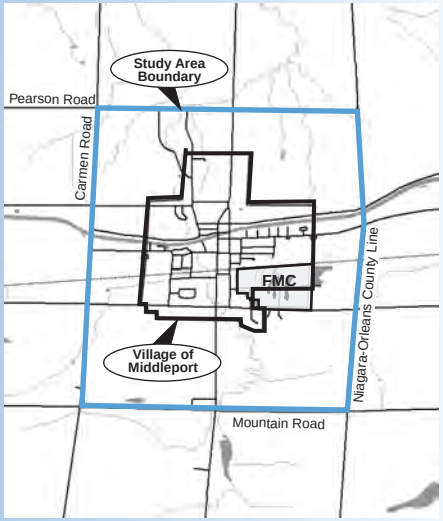
Program Brochure

Study Objectives

1. To quantify the relationship between arsenic in soil and arsenic in urine of children 6 years and younger. Arsenic in urine is a biomaker of recent arsenic exposure.

2. To provide Middleport residents with information on their biomarker levels of arsenic relative to reference levels that indicate a potential for excess exposure and a need for follow-up.

3. To provide the Middleport community and interested health agencies with information regarding residential exposure to arsenic from soil.



Middleport Environmental Exposure Study

You are invited to participate in an environmental **arsenic exposure study** planned for your area. The focus of this study is on **young children** because this age group typically has the most hand-to-mouth **contact with soil**. People of other ages may also **participate** in the sampling if they desire. The study will be conducted this **summer** by a team of **scientists** and **health professionals** from **Exponent**, an independent scientific consulting firm.

Exponent®

For more information, please call 1-800-326-7102

www.exponent.com

The Nature of Arsenic and How It Is Used

Arsenic is an element that is widely distributed in the earth's crust. Two main forms of arsenic are inorganic arsenic and **organic arsenic**. Organic forms are usually less harmful than the inorganic forms. **Inorganic arsenic occurs naturally** in water, soil, and in many kinds of rock, especially in minerals and ores that contain copper or lead. In the past, **arsenic** was primarily used as a pesticide. Most inorganic arsenic pesticides are no longer used; however, organic arsenic pesticides are still used. Almost 90% of all arsenic produced in the recent past was used as a **preservative** for wood to make it resistant to rotting and decay. Arsenic is also used in electronics, in alloys for car batteries, and has limited medicinal uses.

Everyone normally is exposed to small amounts of arsenic through the air we breathe, the water we drink, and the food we eat. Of these, **food** is usually the largest source of arsenic. Fish and **seafood** contain the greatest amounts of **arsenic**, but this arsenic is mostly in the organic form, the form least harmful. Young children are likely to eat small amounts of dust or dirt each day, which is another way they may be exposed to arsenic. Arsenic in water is more efficiently absorbed by the body than arsenic in soil.

If you are exposed to arsenic, many factors determine whether you will be harmed. These factors include the **dose** (how much), the duration (how long), the frequency (how often), the form of arsenic, and how you come in **contact** with it. Other factors may include your age, sex, diet, family traits, lifestyle, and state of health.

Questions & Answers





What is the biomonitoring study about?

Biomonitoring refers to testing a population for clinical measures of possible exposure to a particular substance, in this case arsenic. This study will investigate whether residents of the Middleport community have been exposed to excess levels of arsenic from soil.

Why is this study being conducted?

The community advisory panel of Middleport asked FMC to conduct a biomontoring study of the Middleport community.

Who is paying for the project?

FMC Corporation is funding this study. The study will be conducted by Exponent, with outside review by a scientific expert panel and involved agencies.

Who is eligible?

Anyone who is concerned about arsenic exposure may participate in the study; however, the focus of this study is on young children because this age group typically has the most hand-to-mouth contact with soil. Participation in the study is voluntary and will be encouraged in order to represent the Middleport community and include a range of soil concentrations.

What will I have to do?

If you agree to participate, you will be asked to complete questionnaires on your family activities, diet, health, and other factors that could influence arsenic exposure. We will schedule a convenient time with you to visit your home and collect soil, house dust, and homegrown vegetable samples.

We will also make arrangements with you to collect urine and possibly toenail samples (all collection materials will be supplied).

How will samples be collected?

Participants will be asked to provide two urine samples collected in the morning on two consecutive days. Arsenic in toenails will be analyzed only upon request.

Professional field staff will collect the following samples from each residential property:

Soil samples will be taken at or near residences, with preference given to areas frequently used by children.

Dust samples will be collected from the interior of homes by vacuum technique.

Samples of homegrown fruits or vegetables will be collected from yards where homeowners desire sampling.

What will be done with the samples and the data?

Samples will be shipped to federally accredited laboratories where they will be measured for arsenic levels. From the sample results, study investigators will compare arsenic levels in urine to arsenic levels in soil, house dust, and homegrown produce. Careful attention will also be given to other exposure factors such as diet, individual activities, and other sources of arsenic. If any results are obtained that indicate unusual levels of arsenic in urine or toenails, the individuals providing the samples will be contacted for follow-up. The identity of individual results will be kept confidential.

Exponent®
20 Main Street
Middleport, New York 14105
1-800-326-7102

Appendix B

Letters Reporting Individual Results

January 15, 2004

«Final_Mailing_Name»
«Address»
«City», «State» «Zip_Code»

Subject: Middleport Environmental Exposure Investigation
Urinary Arsenic Results

Dear «Final_Mailing_Name»:

We have received the final results of your urine samples collected last summer. The attached table presents these results. If you submitted toenail samples or had vegetables sampled from your garden, the results will be sent to you separately. Your urine results are within expected levels from diet and other background sources.

According to the Centers for Disease Control and Prevention and the Agency for Toxic Substances and Disease Registry, levels of total arsenic in urine less than 50 micrograms per liter[a] (μg/L) are considered normal (i.e., within levels expected in the general population without elevated arsenic exposure), levels between 50 and 200 μg/L may require monitoring by a health professional but do not necessarily represent a health risk, and levels above 200 μg/L are considered abnormal. **These levels are based on the assumption that the person has not recently eaten fish, seafood, or other foods that are relatively high in total arsenic.** For example, eating some such foods can result in a total urinary arsenic level of more than 1,000 μg/L.

To reduce this effect from diet, a more specific test for exposure to inorganic arsenic, the form of arsenic generally found in soil, was also performed. This test measures what is referred to as "speciated arsenic." Because various foods also contain small amounts of inorganic arsenic, people are expected to have low levels of speciated arsenic in their urine. The reference level used in this program for speciated urinary arsenic is 40 μg/L. Thus, a total arsenic level that exceeds 50 μg/L is still considered normal as long as the speciated arsenic level is below 40 μg/L. Speciated arsenic levels reported by the laboratory for all participants in this program were below 40 μg/L and do not indicate elevated inorganic arsenic exposure.

[a] Micrograms per liter are also called parts per billion (ppb).

Also provided with this letter is a copy of your signed consent form(s). If you have any questions, please call 1-800-326-7102.

Sincerely,

Maria Van Kerkhove, M.S.	Joyce Tsuji, Ph.D., DABT	Michael Goodman, M.D., M.P.H.
Epidemiologist	Toxicologist	Physician, Epidemiologist

Attachments

January 28, 2004

«Final_Mailing_Name»
«Address»
«City», «State» «Zip_Code»

Subject: Middleport Environmental Exposure Investigation
Toenail Sample Arsenic Results

Dear «Final_Mailing_Name»:

Thank you for participating in the Middleport Environmental Exposure Investigation. The attached table presents the laboratory findings for your toenail samples. For participants who provided a sufficient amount of toenail sample, arsenic levels are below the program reference level of 1 part per million (ppm) and therefore do not indicate elevated arsenic exposure.

The Centers for Disease Control and Prevention and the Agency for Toxic Substances and Disease Registry consider normal levels (i.e., expected levels in people without elevated arsenic exposure) for total arsenic in toenails to be less than 1 ppm. Toenails were selected for biomonitoring in this investigation with some hesitancy, because problems have been associated with this type of sampling in previous studies. Although toenail arsenic levels may provide a longer-term measure of exposure, it is difficult to relate these levels to a daily dose and amount of exposure. In addition, soil or dust coming in contact with toenails can result in arsenic binding to the toenail surface. Even at background levels of arsenic in soil, arsenic binding to the surface can yield toenail results that do not reflect ingested arsenic and may indicate higher than actual exposure.

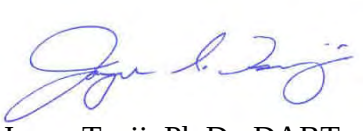
Because of possible external contamination, participants were asked to not expose their feet to soil or dust for at least a month prior to toenail sampling. Toenail samples for some participants showed signs of dirt or discoloration. Results for these samples may not reflect true internal levels of arsenic. Toenails with higher arsenic levels tend to be those with more dirt or discoloration. However, because none of the toenail sample results exceeded the program reference level of 1 ppm, such external contamination was not enough to result in anyone exceeding the reference level.

For 20 percent of participants, not enough sample was received to measure the arsenic content and therefore the laboratory could not report results for these participants. Ideally, 0.5 grams, or at least 0.05 grams, of toenails was needed to measure arsenic levels.

In summary, because none of the samples that could be quantified by the laboratory had total arsenic levels that exceeded 1 ppm, we did not find evidence of elevated arsenic exposure among participants of this part of the study.

If you have any questions, please call 1-800-326-7102.

Sincerely,


Maria Van Kerkhove, M.S.
Epidemiologist

Joyce Tsuji, Ph.D., DABT
Toxicologist

Michael Goodman, M.D., M.P.H.
Physician, Epidemiologist

Attachments

February 13, 2004

«Final_Mailing_Name»
«Address»
«City», «State» «Zip_Code»

Subject: Middleport Environmental Exposure Investigation
Garden Vegetable Sample Results

Dear «Final_Mailing_Name»:

Thank you for participating in the Middleport Environmental Exposure Investigation. The attached table presents the laboratory findings for the vegetable samples taken from your garden last summer. A summary table of community results including a few samples from local stores is also attached.

In reviewing these results, several points should be kept in mind:

- All vegetable samples were below 0.6 ppm fresh weight and much lower than background levels of arsenic in soil.
- None of the federal agencies (e.g., CDC, FDA, EPA, USDA) have set health-based limits for arsenic in vegetables.
- Arsenic levels vary naturally in vegetables and variation within a community and with multiple samples is expected.
- Sample arsenic concentrations are also based on the fresh weight (as sampled) and thus can vary depending on the vegetable water content (e.g., whether the garden had been watered recently, whether vegetables were older and wilted).
- Only a few samples from stores or produce stands were tested, which limits our understanding of the variation in arsenic levels for purchased samples.
- Some community vegetable samples were thoroughly washed of adhering soil and others were not. All purchased samples were washed or carefully brushed off (onion and garlic samples). The “unwashed” vegetables tend to have higher arsenic concentrations.

- The arsenic levels in the vegetable samples are within reported ranges for arsenic in non-meat foods according to recent FDA market basket data.
- Garden vegetables were sampled in this study along with garden soil and biomonitoring samples (e.g., urine) to examine whether arsenic levels in vegetables in the community are related to arsenic levels in soil and whether eating garden vegetables could contribute to people's arsenic exposure.
- Our evaluation of these data is ongoing and we have only just received soil results for some residences.

We are therefore providing you these individual results in advance of our complete analysis and report on this study. If you have any questions, please call us at 1-800-326-7102.

Sincerely,

Maria Van Kerkhove, M.S.
Epidemiologist

Joyce Tsuji, Ph.D., DABT
Toxicologist

Michael Goodman, M.D., M.P.H.
Physician, Epidemiologist

Attachments

Summary of vegetable sampling results

Vegetable Type	Sample Preparation[a]	Homegrown Produce		Purchased Produce	
		Number of Samples	Arsenic Concentration[b] (ppm)	Number of Samples	Arsenic Concentration[b] (ppm)
Basil	Washed	2	0.038–0.107	1	0.028
Beans, green	Washed	1	*<0.013*	1	*<0.006*
	Unwashed	2	*<0.006*– 0.020	--	--
Beans, yellow	Washed	0	--	1	*<0.009*
	Unwashed	1	*<0.006*	--	--
Beet, greens	Washed	0	--	1	0.028
	Unwashed	3	0.013–0.028	--	--
Beet, bulb	Washed	0	--	2	*<0.006*
	Unwashed	3	0.007–0.016	--	--
Broccoli	Washed	2	*<0.007–<0.009*	1	*<0.008*
	Unwashed	1	*<0.009*	--	--
Carrot (unpeeled)	Washed	1	0.173	1	*<0.008*
Carrot (peeled)	Washed	0	--	1	*<0.006*
Cauliflower	Washed	0	--	1	*<0.006*
	Unwashed	2	0.009–0.050	--	--
Chard	Washed	1	0.054	1	*<0.007*
Collard greens	Washed	1	0.125	1	0.008
Cucumber	Washed	3	0.010–0.044	2	*<0.002–<0.003*
	Unwashed	3	0.019–0.103	--	--
Garlic	Unwashed	1	*<0.024*	1	*<0.023*[c]
Lettuce	Washed	3	0.020–0.051	2	*<0.002*–0.004
	Unwashed	3	0.148–0.341	--	--
Mint	Washed	2	0.065–0.087	1	0.024
	Unwashed	1	0.028	--	--
Onion	Unwashed	4	*<0.006*– 0.199	1	0.008[c]
Onion, green	Washed	1	0.018	1	0.010
	Unwashed	1	0.022	--	--
Pepper, banana	Washed	1	0.006	1	*<0.005*
Pepper, green	Washed	0	--	1	*<0.003*
	Unwashed	1	*<0.004*	--	--
Radish	Washed	0	--	1	0.007
	Unwashed	1	0.094	--	--

Summary of vegetable sampling results (cont.)

Vegetable Type	Sample Preparation[a]	Homegrown Produce: Number of Samples	Homegrown Produce: Arsenic Concentration[b] (ppm)	Purchased Produce: Number of Samples	Purchased Produce: Arsenic Concentration[b] (ppm)
Sage	Washed	2	0.053–0.345	1	0.027
	Unwashed	1	0.574	--	--
Squash, acorn	Washed	1	0.006	1	*<0.006*
Squash, yellow	Washed	5	*<0.005*– 0.063	1	*<0.005*
	Unwashed	3	0.004–0.015	--	--
Squash, zucchini	Washed	3	*<0.003–<0.004*	1	*<0.004*
	Unwashed	2	0.006–0.008	--	--
Tomato	Washed	14	*<0.003–<0.005*	1	*<0.004*
	Unwashed	6	*<0.004*– 0.010	--	--
Tomato, cherry	Washed	2	*<0.005–<0.008*	1	*<0.007*
	Unwashed	1	0.008	--	--
Tomato, grape	Washed	0	--	1	*<0.004*
	Unwashed	1	0.007	--	--
Tomato, green	Washed	3	0.004–*<0.007*	0	--
	Unwashed	8	*<0.004*– 0.009	--	--
Tomato, plum	Washed	2	*<0.004*– 0.006	1	*<0.003*
	Unwashed	0	--	--	--
Turnip (unpeeled)	Washed	1	0.013	1	*<0.005*
	Unwashed	0	--	--	--

Note: Sample concentration values in italics and preceded with "<" had no detectable arsenic at the limits shown.

[a] "Washed" samples were thoroughly washed with tap water and rinsed with distilled water. "Unwashed" samples were not as thoroughly cleaned.

[b] Arsenic concentration is based on fresh weight of the sample, which can vary depending on water content.

[c] Purchased garlic and onion were brushed off before analyses.

April 12, 2004

«Final_Mailing_Name»
«Address»
«City», «State» «Zip_Code»

Subject: Middleport Environmental Exposure Investigation
House Dust Sample Results

Dear «Final_Mailing_Name»:

We have received the results of the dust samples from your home that were collected by SOMA (Sandler Occupational Medicine Associates, Inc.). The arsenic concentration in your house dust sample was reported as «Arsenic_ppm» ppm.

Please keep in mind that the environmental sample results (i.e., soil, dust, vegetables) are not a direct measure of arsenic exposure as are the biomonitoring results (i.e., urine) and that arsenic levels in house dust may arise from various sources including tracked in or windblown soil, smoking, and use of various consumer products. The purpose of house dust and soil sampling during this investigation was to find out how arsenic levels in these environmental samples are related to arsenic levels in urine (i.e., exposure).

We are providing your individual house dust results in advance of our complete analysis and report on this study. In our complete analysis, we will compare urine arsenic levels with arsenic levels in soil and house dust. Such a comparison helps indicate the amount of arsenic exposure that might come from soil and house dust. House dust levels will also be compared to soil levels in the community and assessed as part of risk assessment efforts that are separate from the biomonitoring study.

As was stated in the letter reporting your urinary arsenic results, the results for all participants sampled were within expected levels from diet and other background sources, and showed no evidence of elevated inorganic arsenic exposure. Thus, exposures related to soil and dust are likely to be very low.

If you have any questions, please call us at 1-800-326-7102.

Sincerely,

Maria Van Kerkhove, M.S.
Epidemiologist

Joyce Tsuji, Ph.D., DABT
Toxicologist

Michael Goodman, M.D., M.P.H.
Physician, Epidemiologist

Appendix C

Quality Assurance Review of Biological Samples

Quality Assurance Review for Biological Samples

Introduction

In support of the Middleport environmental exposure investigation (Exponent 2003), Exponent completed a quality assurance review for the following analyses: 1) total arsenic, speciated arsenic (i.e., inorganic arsenic, dimethylarsinic acid [DMA], and monomethylarsonic acid [MMA]), creatinine, and specific gravity in urine samples, 2) total arsenic in toenail samples, and 3) total arsenic and speciated arsenic (i.e., inorganic arsenic, DMA, and MMA) in vegetable samples. The quality assurance review was conducted to verify that quality assurance and quality control procedures were completed and documented as required during sample collection and analysis and that the quality of the data is sufficiently high to support its intended uses.

The quality of the data was generally very good. Data that did not meet control limits for quality control measurements were qualified as estimated (*J*) during the review. All data that were qualified as estimated (*J*) have an acceptable degree of uncertainty and represent data of good quality and reasonable confidence (U.S. EPA 1989, 1996a). No data were rejected (*R*) as unusable for this investigation. All of the qualified and unqualified data are of sufficiently high quality for their intended use. A summary of the qualified data is presented in Table C-1.

Samples and Analyses

The sample type, number of samples, and the target analytes completed for the Middleport environmental exposure investigation (Exponent 2003) are presented in Table C-2.

Lockport Memorial Hospital (LMH) analyzed the specific gravity and creatinine levels of urine samples. Battelle Marine Sciences Laboratory (Battelle) performed arsenic analyses on urine, toenail, and vegetable samples. LMH created duplicate samples by splitting 1 out of every 20 urine samples prior to sending the samples to Battelle. Battelle created a composite of 2 urine samples from each participant. Bottle blanks (sample bottles containing deionized or distilled water) were not created in the field as specified in the work plan, but rather created at Battelle.

Data Validation Procedures

Data validation procedures included evaluating the sample results and applicable quality control results reported by the laboratory. The laboratory was responsible for review and verification of analyte identifications, calculations, and transcriptions. The data were validated following procedures and guidance specified by the *USEPA Contract Laboratory Program, National Functional Guidelines for Inorganic Data Review* (U.S. EPA 2002), the referenced analytical methods, laboratory-specific quality control criteria, and in the context of the data quality

objectives and data quality indicators established for this project. Data validation procedures for specific gravity and creatinine were modified to accommodate quality control requirements for these methods, which are not specifically addressed by the functional guidelines.

The following items, as applicable to the analysis completed, were evaluated during quality assurance review:

- The case narrative discussing analytical problems (if any) and procedures
- Chain-of-custody documentation to verify completeness of data set
- Sample preparation logs or laboratory summary result forms to verify analytical holding times
- Results for mass spectrometer tuning, instrument calibration, and continuing calibration to assess instrument performance
- Results for method blanks and bottle blanks to check for laboratory contamination and to monitor sample collection activities, respectively
- Results for matrix spike, laboratory control sample (LCS) (i.e., a blank spike), and standard reference material (SRM) recoveries to assess analytical accuracy
- Results for laboratory duplicate sample analyses to assess analytical precision
- Results of quantification including method detection limits (MDLs) and method reporting limits (MRLs)
- Results for field quality control samples (split samples) to monitor sample collection activities
- Laboratory summaries of analytical results.

In addition, all electronic data imported or hand-entered into the Exponent database were verified against the laboratory data packages or field logs, and all discrepancies were resolved. Data qualifiers were assigned during the quality assurance review when control limits were not met, in accordance with U.S. EPA (2002).

Data Quality and Usability

The quality of the data was generally very good. Selected data were qualified as estimated (*J*) when control limits were exceeded for one or more quality control samples or procedures. All data qualified as estimated (*J*) have an acceptable degree of uncertainty and represent data of good quality and reasonable confidence (U.S. EPA 1989, 1996a). No data were rejected (*R*). All of the qualified and unqualified data are of sufficiently high quality for their intended use.

The data reported were evaluated in terms of completeness, holding times, instrument performance, laboratory and field blanks, bias, precision, and analyte identification and quantification. The results for the quality control procedures used during sample analyses are discussed in the sections below.

Completeness

Results reported by the laboratory were 100 percent complete. No data were rejected (*R*) during the quality assurance review.

Of the 84 samples submitted, analyses of 17 toenail samples and 2 duplicate toenail samples could not be completed because sample weights were below the minimum of 0.05 g required for analysis.

Holding Times and Sample Preservation

Holding time constraints, as specified by the analytical methods and standard operating procedures, were met for all samples.

Analytical Methods

Analyses were completed in accordance with the methods listed in Table C-3.

Composite urine samples were diluted with laboratory grade water and total arsenic was analyzed using inductively coupled plasma-mass spectrometry (ICP-MS). For the analysis of speciated arsenic (i.e., inorganic arsenic, DMA, and MMA), the diluted samples were acidified with hydrochloric acid to pH less than 2 and the digestate was reduced with sodium borohydride. The volatilized target compound was cryogenically trapped on a chromatography column, and analysis was completed with hydride generation atomic absorption (HGAA) spectroscopy.

After toenail polish was removed, if necessary, approximately 0.05 to 0.5 g of sample was weighed, cleaned in an ultrasonic bath, dried, digested in hot nitric acid, and analyzed for total arsenic using ICP-MS.

Vegetable samples (using the edible portion only) were cut into small pieces with a ceramic knife and placed in a clear plastic jar until approximately 30 g (wet weight basis) was obtained. Each of the samples was then freeze-dried to obtain a percent dry weight. Next, each of the freeze-dried samples was ball milled. Approximately 0.5 g of each of the freeze-dried and ball milled vegetable samples was digested in 2M sodium hydroxide at 80°C for about 16 hours. Analyses of the digestates were then completed using ICP-MS for total arsenic and HGAA for speciated arsenic.

Instrument Performance

The performance of the analytical instruments, as documented by the laboratory, was acceptable. No changes in instrument performance that would have resulted in the degradation of data quality were indicated during any analysis sequence.

Mass Spectrometer Tuning

Mass spectrometer tuning checks were completed as required, and results met control limits. Mass spectrometer tuning checks were required for analyses completed by ICP-MS.

Initial and Continuing Calibration

Initial and continuing calibrations were completed for all applicable target analytes. These calibrations met the laboratory's and validation criteria for acceptable performance and frequency of analysis, with one exception.

The laboratory-established upper control limit for inorganic arsenic for continuing calibrations (i.e., 120 percent) during the vegetable analyses was exceeded (i.e., 125 percent). Five of the results reported for inorganic arsenic in selected vegetable samples were assigned an *E*-flag by the laboratory and, subsequently, were qualified as estimated (*J*) during the quality assurance review. The bias of these qualified results is not significant because the recovery of the continuing calibration was only slightly above the upper control limit.

Method Blank and Bottle Blank Analyses

Total arsenic and inorganic arsenic were detected in the method blanks and bottle blanks associated with the analysis of urine samples. Total arsenic was reported in method blanks associated with the analysis of toenail samples. Details are discussed below.

Urine Sample Analyses

Total arsenic was detected in 20 of 25 method blanks at an average concentration of 0.088 μg/L and was detected in 11 of 16 bottle blanks at an average concentration of 0.31 μg/L. The presence of arsenic in the method blanks and the bottle blanks was due to low levels of arsenic in the reagents that were used to process the samples for analysis. Total arsenic was present in the urine samples at concentrations ranging from 2.1 to 773 μg/L. Total arsenic was present in the urine samples at concentrations that were above 5 times the data validation control limit (U.S. EPA 2002); therefore, no data required qualification.

Inorganic arsenic was present in the urine samples at a concentration ranging from 0.25 to 2.7 μg/L. Inorganic arsenic was detected in 22 of 25 method blanks at an average concentration of 0.43 μg/L and was detected in all of the 16 bottle blanks at an average concentration of 0.47 μg/L. Inorganic arsenic was present in the urine samples at concentrations that are less than 5 times the concentration in the associated blanks; therefore, these samples are most

probably reported as false positives (i.e., the analyte was not actually present in the samples, but rather in the reagents used to the process the sample).

When blank contamination issues are encountered, data validation procedures (U.S. EPA 2002) require that results reported as detected are to be restated as undetected (*U*) at the concentration reported or reported as undetected at the concentration found in the associated blank if the concentration in the sample is less than the concentration in the associated blank. However, in this case, given the low detected concentration, none of the data affected by the blank contamination were restated as undetected, but rather all 462 inorganic arsenic results reported as detected in the urine samples were qualified as estimated (*J*); these qualified results exhibit a positive bias.

Toenail Sample Analyses

Total arsenic was detected in four of five method blanks at an average concentration of 0.01 μg/g. As stated above for urine, the presence of arsenic in the method blanks and the bottle blanks was due to low levels of arsenic in the reagents that were used to process the samples for analysis.

Total arsenic concentration in toenail samples ranged from 0.02 to 0.97 μg/g. Of the 67 total arsenic results reported for toenail samples, 11 results were reported at a concentration less than 5 times the concentration in the associated blanks. These 11 results were most likely reported biased high (i.e., the analyte was not actually present in the sample, but rather in the reagents used to the process the sample). Using the approach discussed above, none of the data affected by the blank contamination were restated as undetected (*U*), but were qualified as estimated (*J*); these qualified results exhibit a positive bias.

Accuracy

The accuracy (i.e., bias) of the analytical results was evaluated by the recoveries of matrix spike, LCS, and SRM analyses. Results for these quality control procedures are described below.

Matrix Spike Recoveries

Matrix spikes are completed on field samples to determine the analytical accuracy for samples associated with the investigation. Matrix spike results are not meaningful when the native concentration of an analyte in the sample is greater than approximately 4 times the concentration of the added spike because the variability (i.e., precision) of the analysis may bias the matrix spike recovery. In these cases, the accuracy of the analysis is based on results of other quality control procedures for accuracy, such as LCS and SRM recoveries (discussed below).

The recoveries reported by the laboratory for all matrix spike analyses, and the frequency of analysis, met the laboratory criteria for acceptable performance (i.e., 75–125 percent for total

arsenic, 50–150 percent for inorganic arsenic, 60–140 percent for MMA, and 40–160 percent for DMA), with the exceptions described below.

Recoveries of 134 and 129 percent were reported for total arsenic in 2 of 25 matrix spike analyses completed on urine samples. These recoveries are above the upper laboratory-established control limit of 125 percent. No action was taken because the exceedances of these matrix spike recoveries were isolated occurrences and were not an indication of a systematic bias.

Matrix spike recoveries were not completed on toenail samples because limited sample mass was available; therefore, the LCS and SRM recoveries were used to assess the accuracy of the toenail sample results.

A recovery of 127 percent was reported for total arsenic in one of the matrix spikes completed on the vegetable samples. This recovery is slightly above the upper laboratory-established control limit of 125 percent. No action was taken because this exceedance was an isolated occurrence and not an indication of systematic bias.

Laboratory Control Sample Recoveries

LCS recoveries provide a control for the entire analytical procedure, including sample preparation as well as instrumental analysis. LCS analyses were completed for total arsenic and were associated with the toenail sample and vegetable sample analyses. LCS recoveries were not performed on urine samples (see SRM recovery discussion below).

The recoveries reported by the laboratory for all LCS analyses and their frequency of analysis, met the laboratory criteria for acceptable performance (i.e., 75–125 percent for total arsenic, 50–150 percent for inorganic arsenic, 60–140 percent for MMA, and 40–160 percent for DMA.

Standard Reference Material Recoveries

SRM recoveries provide a control for the entire analytical procedure, including sample preparation as well as instrumental analysis. The recoveries reported by the laboratory for all SRM analyses and their frequency of analysis met the Battelle criteria for acceptable performance (i.e., 75–125 percent for total arsenic), and frequency of analysis, with two exceptions discussed below.

Recoveries of 177 and 174 percent were reported for two SRM recoveries associated with the urine sample analyses. These two abnormally high recoveries were not explained by Battelle; however, it appears an incorrect aliquot of the SRM was used in the preparation. These recoveries appear to be outliers; therefore, no sample results were qualified.

The SRM analyzed along with the samples from the National Institute for Standards and Technology (NIST) included the following:

- SRM NIST 2670 (toxic substances in human urine, powder form) certified at concentrations of 480 µg/L (elevated level) and 60 µg/L (normal level)
- SRM NIST 1640 (water) certified at a concentration of 26.67 µg/L and processed along the urine samples
- SRM NIST 2976 (clam mussel) certified at a concentration of 13.3 µg/g (dry weight) and processed along with the toenail samples
- SRM NIST 568a (rice) certified at a concentration of 0.29 µg/g (dry weight) and processed along with the vegetable samples.

Precision

Results for all duplicate sample analyses and the frequency of analysis, met the laboratory criteria for acceptable performance (i.e., relative percent differences of ±25 total arsenic, ±35 for inorganic arsenic, ±25 for MMA, and ±40 for DMA), with the one exception discussed below.

A relative percent difference of 27 was reported for total arsenic in one vegetable sample and one duplicate vegetable sample. No data were qualified because the ±25 percent control limit was only slightly exceeded and there was no indication of a systematic bias.

Quantification: Method Detection Limits and Method Reporting Limits

Quantification of analyte concentration involves calculation of concentration with respect to standards, correction for sample mass or volume, dilution factor, and moisture content (or total solids content) in the samples. The correct calculation of the MRL and MDL for each analyte in a sample type and dilution level is determined by and is the responsibility of Battelle.

The case narratives from Battelle stated that no problems were encountered with respect to analyte quantification, with the exception of very low sample weights used to process 17 toenail samples and 2 duplicate toenail samples. The minimum target sample weight of 0.05 g was not attained and, therefore, an accurate quantification of total arsenic that may be present was not achieved.

The MDLs and MRLs provided by the laboratory for all sample types were acceptable for urine and toenail samples and generally met the objectives of the investigation. The MDL and MRL for each matrix in this investigation are summarized in Table C-4. A total of 165 results were reported by LMH as having a specific gravity of greater than 1.030, which was the upper MRL for specific gravity. Because the true specific gravity is likely greater, these 165 results were qualified as estimated (*J*) during the quality assurance review. Likewise, urine samples with a specific gravity of less than 1.005 were qualified as undetected (*U*); however, the true specific gravity is likely measurable.

Results detected at a concentration above the MDL, but less than the MRL were assigned a *J*-flag by the laboratory and were subsequently qualified as estimated (*J*) during the quality

assurance review. A total of 35 inorganic arsenic, 12 DMA, and 165 MMA results for urine samples were qualified as estimated (*J*) during the quality assurance review for this reason. Twelve results reported for total arsenic in toenail samples, and 35 results reported for total arsenic in vegetable samples were qualified for this reason.

Field Quality Control Samples

Field quality control samples were created by LMH and consisted of splits of 46 urine samples. Field duplicates of toenail and vegetable samples were not performed. Results for the field duplicate sample analyses were acceptable. In addition, 24 composite urine samples were sent to the Centers for Disease Control and Prevention for speciated arsenic analysis. A trip blank was not performed, although empty bottles were sent directly to the laboratory from the field and were considered bottle blanks.

References

Exponent. 2003. Work plan for the Middleport environmental exposure investigation. Draft. Prepared for FMC Corporation, Philadelphia, PA. Exponent, Bellevue, WA.

U.S. EPA. 1989. Memorandum from H.M. Fribush, Technical Project Officer, Analytical Operations Branch, to S. Wells, Chief, NPL Criteria Section, Site Assessment Branch, regarding J-qualified CLP data and recommendations for its use. U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, DC.

U.S. EPA. 1996a. Using qualified data to document an observed release and observed contamination. EPA/540/F-94/028. U.S. Environmental Protection Agency, Office of Solid Waste and Emergency Response, Washington, DC.

U.S. EPA 1996b. Method 1638. Determination of trace elements in ambient waters by inductively coupled plasma-mass spectrometry. U.S. Environmental Protection Agency, Office of Water, Engineering and Analysis Division, Washington, DC.

U.S. EPA 1996c. Method 1632. Inorganic arsenic in water by hydride generation quartz furnace atomic absorption. U.S. Environmental Protection Agency, Office of Water, Engineering and Analysis Division, Washington, DC.

U.S. EPA. 2002. USEPA contract laboratory program national functional guidelines for inorganic data review. Final. OSWER 9240.1-35. EPA 540-R-01-008. U.S. Environmental Protection Agency, Office of Emergency and Remedial Response, Washington, DC.

Tables

Table C-1. Summary of qualified data

Matrix/Analyte	Total Results Reported	Total Results Qualified Estimated (*J*)	Total Results Restated Undetected (*U*)	Total Results Rejected (*R*)
Urine				
Total arsenic	462	0	0	0
Inorganic arsenic	462	462	0	0
DMA	462	12	0	0
MMA	462	165	0	0
Creatinine	926	0	0	0
Specific gravity	926	165	0	0
Toenail				
Total arsenic	67	12	2	0
Homegrown Vegetable				
Total arsenic	100	28	0	0
Inorganic arsenic	17	5	0	0
DMA	17	0	0	0
MMA	17	0	0	0
Total solids	100	0	0	0
Control Vegetable				
Total arsenic	32	7	0	0
Total solids	32	0	0	0

Note: DMA - dimethylarsinic acid
MMA - monomethylarsonic acid

Table C-2. Summary of samples and target analytes

Sample Type	Number of Samples Analyzed	Number of Bottle Blanks	Target Analytes	Analytical Laboratory
Urine				
	462	16	Total arsenic	Battelle
	462	16	Inorganic arsenic	Battelle
	462	16	DMA	Battelle
	462	16	MMA	Battelle
	926	--	Creatinine	LMH
	926	--	Specific gravity	LMH
Toenail				
	67	--	Total arsenic	Battelle
Homegrown Vegetable				
	100	--	Total arsenic	Battelle
	17	--	Inorganic arsenic	Battelle
	17	--	DMA	Battelle
	17	--	MMA	Battelle
	100	--	Total solids	Battelle
Control Vegetable				
	32	--	Total arsenic	Battelle
	32	--	Total solids	Battelle

Note: Battelle - Battelle Marine Sciences Laboratory
DMA - dimethylarsinic acid
LMH - Lockport Memorial Hospital
MMA - monomethylarsonic acid

Table C-3. Analytical methods

Constituent	Method	Analytical Laboratory
Total arsenic	Digestion and analysis by ICP-MS according to Battelle SOP MSL-I-022-05, which is equivalent to EPA Method 1638 (U.S. EPA 1996b)	Battelle
Inorganic arsenic	Digestion and analysis by HGAA spectroscopy according to Battelle SOP MSL-I-021-00, which is equivalent to EPA Method 1632 (U.S. EPA 1996c)	Battelle
DMA	Digestion and analysis by HGAA spectroscopy according to Battelle SOP MSL-I-021-00, which is equivalent to EPA Method 1632 (U.S. EPA 1996c)	Battelle
MMA	Digestion and analysis by HGAA spectroscopy according to Battelle SOP MSL-I-021-00, which is equivalent to EPA Method 1632 (U.S. EPA 1996c)	Battelle
Total solids	Desiccation by freeze drying and gravimetric measurement	Battelle
Creatinine	LMH SOP	LMH
Specific gravity	LMH SOP	LMH

Note:
DMA - dimethylarsinic acid
EPA - U.S. Environmental Protection Agency
HGAA - hydride generation atomic absorption
ICP-MS - inductively coupled plasma-mass spectrometry
LMH - Lockport Memorial Hospital
MMA - monomethylarsonic acid
SOP - standard operating procedure

Table C-4. Summary of MDLs and MRLs

Matrix	MDL	MRL
Urine		
Total arsenic	0.2 µg/L	0.5 µg/L
Inorganic arsenic	0.06 µg/L	0.5 µg/L
DMA	0.4 µg/L	0.5 µg/L
MMA	0.08µg/L	0.5 µg/L
Creatinine	NA	NA
Specific gravity	1.005	1.030
Toenail		
Total arsenic	0.02 µg/g	0.62 µg/g
Vegetable (dry weight)		
Total arsenic	0.062 µg/g	0.2 µg/g
Inorganic arsenic	0.03 µg/g	0.2 µg/g
DMA	0.04 µg/g	0.2 µg/g
MMA	0.01 µg/g	0.2 µg/g

Note: DMA - dimethylarsinic acid
MDL - method detection limit
MMA - monomethylarsonic acid
MRL - method reporting limit
NA - not applicable

Appendix D

SOMA Report of Surface Sampling for Arsenic, Middleport, New York

SOMA
SANDLER OCCUPATIONAL
MEDICINE ASSOCIATES, INC.

April 7, 2004

VIA ELECTRONIC MAIL AND FIRST CLASS MAIL

Dr. Joyce Tsuji
Exponent
15375 SE 30th Place, Suite 250
Bellevue, WA 98007

Subject: **Report of Surface Sampling for Arsenic**
Middleport, New York

Dear Dr. Tsuji:

Sandler Occupational Medicine Associates Inc. (SOMA) is pleased to provide this report of our results of the surface sampling for arsenic in residences in Middleport, New York. This evaluation was performed at your request, as described in SOMA's proposal to FMC Corporation for Surface Sampling for Arsenic, dated August 4, 2003. This report is divided into sections containing the project background, scope of services, quality assurance, and observations and results.

Project Background

An FMC Corporation (FMC) agricultural facility located in Middleport, NY previously used arsenic compounds. The facility is located between Vernon Street on the west, Countyline Road on the east and Maple Ridge Road on the south. The study has been undertaken due to public concerns regarding arsenic exposures in the Middleport community. The purpose of our study is to quantify residual arsenic on floor surfaces in residences in Middleport. The information presented by SOMA will be incorporated into a larger study performed by Exponent. Exponent conducted outreach activities to identify residents that were willing to allow surface (dust) samples to be collected in their residences. They provided names and telephone numbers of willing residents to SOMA. The initial information provided by Exponent included 166 residences.

Scope of Services

After receiving the list of participants, SOMA attempted to schedule visits to residences over a series of 3-4 day periods. Our initial plan was to visit between 20 and 40 residences during the sampling periods. Of the initial 166, SOMA was able to conduct sampling at 97 homes. Table 1 below, describes the sampling status of the initial 166 residences. (Table 2, attached, provides additional details for residences that were not sampled).

Table 1: Summary of Sampling

Status	Number of Residences
Sampling completed	97
Incorrect sampling protocol	1
Initially sampled, unable to "re-sample"*	13
Declined	25
Scheduled but not present for visit	8
Unable to contact	22
Total	**166**

*These were households where samples were collected in October 2003, but the residents were not willing to be re-sampled in December. The request was made for re-sampling following a laboratory analysis error.

Based on the response, the sampling was conducted by SOMA teams on the following dates:

- September 3 to 5, 2003: 30 residences;
- October 8 to 10, 2003: 31 residences. The samples collected during this visit were analyzed incorrectly by the lab. Re-sampling of these residences was attempted during the December visits;
- October 15 to 17, 2003: 17 residences;
- November 12 to 14, 2003: 18 residences;
- December 3 to 5, 2003: 17 residences; and;
- December 9 to 11, 2003: 15 residences.

At the individual residences, interior surfaces were sampled for arsenic as per the protocol outlined in the Work Plan for the Middleport Environmental Exposure Investigation, prepared by Exponent, dated August 2003, and the discussions that later followed between SOMA, Exponent and FMC. The sampling was conducted as follows:

- The samples were collected and analyzed based on a modified version of a lead-dust sampling protocol described by Que Hee et al. (in Que Hee, et al. 1985. Evolution of efficient methods to sample lead sources such as house dust and hand dust in homes, of children. Environ. Res. 38:77–97).
- The samples were collected using a personal air sampling pump. A small portion of tubing was attached to the sample filter to act like a vacuum cleaner hose. The dust was collected on a filter within the sample cassette. The filters were pre-weighed and provided by H2M Labs, Inc., of Melville, New York's QA officer (H2M). The tubing connected to the sample apparatus was discarded and not included in the sample.

- A composite sample was obtained from at least three representative floor surface areas (625 cm^2 each) in each home. The three surfaces generally included a floor area directly inside the entrance most often used by the residents, a floor area in the most frequently occupied room (living room, kitchen or family room) and a floor area in the residents' bedroom (child's bedroom if any).

- The intended weight of the sample collected was approximately 500 milligrams of dust. Additional floor surface areas were sampled when the sample weight of the first three surfaces was less than 60-80% of target mass (i.e. < 300 – 400 mg). The details of the additional sampling sites were noted in the field notes.

- The samples were submitted to H2M for analysis of arsenic and total weight of dust. One blank sample was prepared and submitted once per day or for ten percent of the samples collected.

Quality Assurance

During the course of the project SOMA conducted internal quality assurance (QA) procedures and reviewed the QA process followed by H2M as described in their reports.

SOMA QA Procedures

SOMA conducted the following internal QA procedures:

- Pre and post calibration of sampling pumps.

- Internal training of field sampling personnel.

- Use of blank filters (1 blank per ten samples). The blank filters were sent to H2M with the samples collected for analysis of arsenic and total weight of dust.

- Field measurement of house dust weights. Using a factory-calibrated field scale, we recorded field tare weights of samples collected during the sampling visit of December 3, 2003. The field tare weights on an average were within approximately 7% of laboratory reported weights. The field tare weights were not used for calculation of dust or arsenic concentration. The measurements were only used for comparison of total dust weights reported by H2M.

Review of H2M QA Procedures

For the laboratory results, we evaluated methods used by H2M, provided blanks samples at the rate they requested (2 per 20 samples) and evaluated the results based on our field notes. Specifically, H2M performed the following:

- The samples were analyzed for weight of dust in general accordance with the *Total*

Solids Method from the EPA SOW for Inorganics (ILM04.0) and as per the protocol outlined in the Work Plan for the Middleport Environmental Exposure Investigation, prepared by Exponent, dated August 2003. The method required use of pre-weighed filters (provided by H2M) and subsequent post sample weight measurements. The minimum absolute weight that could be measured with this procedure and the laboratory scales was 0.001 g.

- The reported dry dust weight of the samples was "blank" corrected by H2M. The value for blank correction ranged from 0.003 g to 0.04 g (mean=0.01 g). The weight of the blank samples was lower on the post sample weights than on pre-sample weights. The laboratory reports (SDG No. SOMA001 and SOMA002) suggest the difference in weights to be related to moisture.

- The dried sample was then digested and analyzed for arsenic by the EPA Method 6010B with TJA61E Trace ICP instrument. The minimum limit of analytical detection was 0.11 µg/filter.

- The blank filters sent by SOMA were used as Laboratory Control Sample (LCS) and Method Blank (MB) for quality assurance of the digestion process. The blank filters were analyzed for arsenic and laboratory results showed that arsenic was not detected on the blank filters.

- Duplicate and post spike analyses were conducted as part of the lab's QA process to check the validity of the results of the arsenic analysis. The recovery acceptance range used for the post spike analysis was 75% to 125%. The post spike analyses of four of the five batches of samples sent to H2M for the weight and arsenic analysis were within the acceptance range. The laboratory report, SDG No. SOMA007 stated that the post spike analysis for that batch of samples did not recover arsenic levels within the acceptance range. However, H2M reported that the batch was accepted based on LCS, Initial Calibration Verification (ICV) and Continuing Calibration Verification (CCV) standard recoveries. The validity parameter used for the duplicate analysis was 20% Relative Percentage Difference (RDP) or a deviation of ± Contract Required Detection Limit (CRDL). The detailed results of these analyses have been presented in the laboratory reports.

- H2M holds the National Environmental Laboratory Approval Program (NELAP) accreditation, with primacy in New York State for the performance of this sampling.

Observations and Results

During the sampling at Middleport the following observations were made:

- Remediation work was in progress at Vernon Street during September and October.

- The residents were instructed not to sweep/vacuum their houses for a week prior to

the sampling. However there were instances where the resident had cleaned the house in the week leading up to sampling.

- Remodeling or renovation work had been completed or was in process at some of the residential sites.

The laboratory results for individual sample locations have been provided in Table 3 (attached). Table 3 provides the last name of the resident, address, sample date, weight of dust, weight of arsenic, a calculated weight-based concentration (milligrams of arsenic per kilogram of collected dust) and a calculated area-based concentration. Additional compiled data was provided to Exponent in electronic form which provided additional information from field data forms and included sampling ID, total area of sampling, number of days since the floors were last cleaned, occurrence of any unusual activities, number of pets if any, route of entrance/exit of pets if applicable, method of heating in the house, existence of fireplace/wooden stove in the house, use of pressurized wood inside and outside the house and use of screens on the doors and windows. Copies of the field data forms have been included as Appendix A.

The weight of dust ranged from 0.004g to 0.833g (mean=0.226g). The weight of arsenic ranged from 0.11µg/filter to 60µg/filter (mean=4.7µg/filter). The weight-based concentration ranged from 1mg/kg to 173 mg/kg (mean=20mg/kg) and the area-based concentration ranged from 4.4×10^{-5}µg/cm^2 to 3.2×10^{-2}µg/cm^2 (mean=2.39×10^{-3}µg/cm^2). Copies of the laboratory results have been included as Appendix B.

Closing

We appreciate the opportunity to provide these services to Exponent, and look forward to working with you in the future. Please do not hesitate to contact us should you have any questions or comments.

Sincerely,

Priya Nagarajan, M. Eng
Industrial Hygienist

Dennis C. Ertel
by P.N.

Dennis C. Ertel, Jr., CIH, REM
Manager, Industrial Hygiene

Appendix A and B (Only included in original sent by First Class mail)

CC: Ms. Maria Van Kerkhove, Exponent, 15375 SE 30th Place, Suite 250, Bellevue, WA 98007
Dr. Martin J. Reape, FMC Corporation, 1735 Market St, Philadelphia, PA 19103

Appendix E

Geographic Distribution of Biological and Environmental Data

Contents

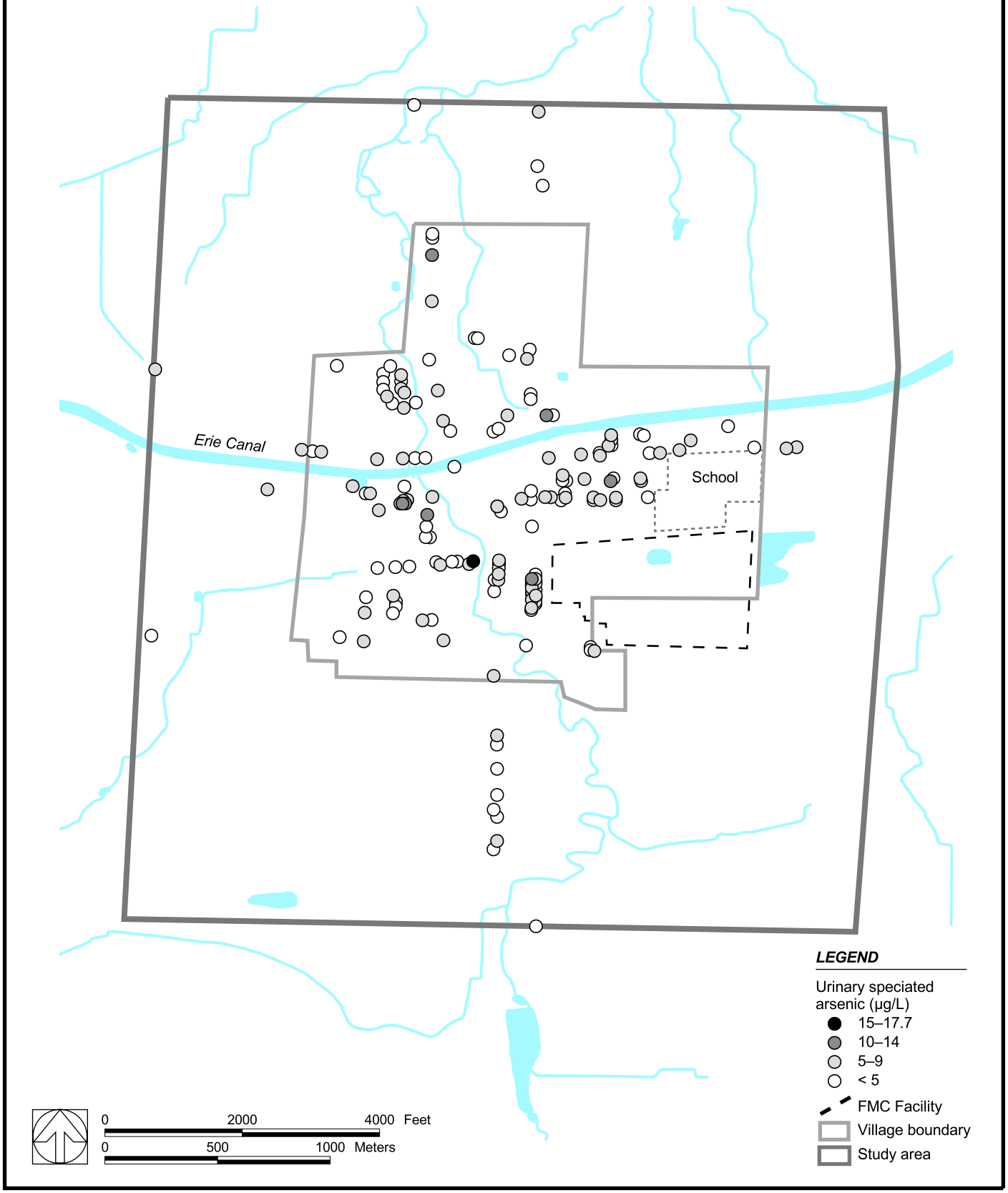


Figure E-1. Average value of speciated arsenic in urine per family—all participants

Exponent®

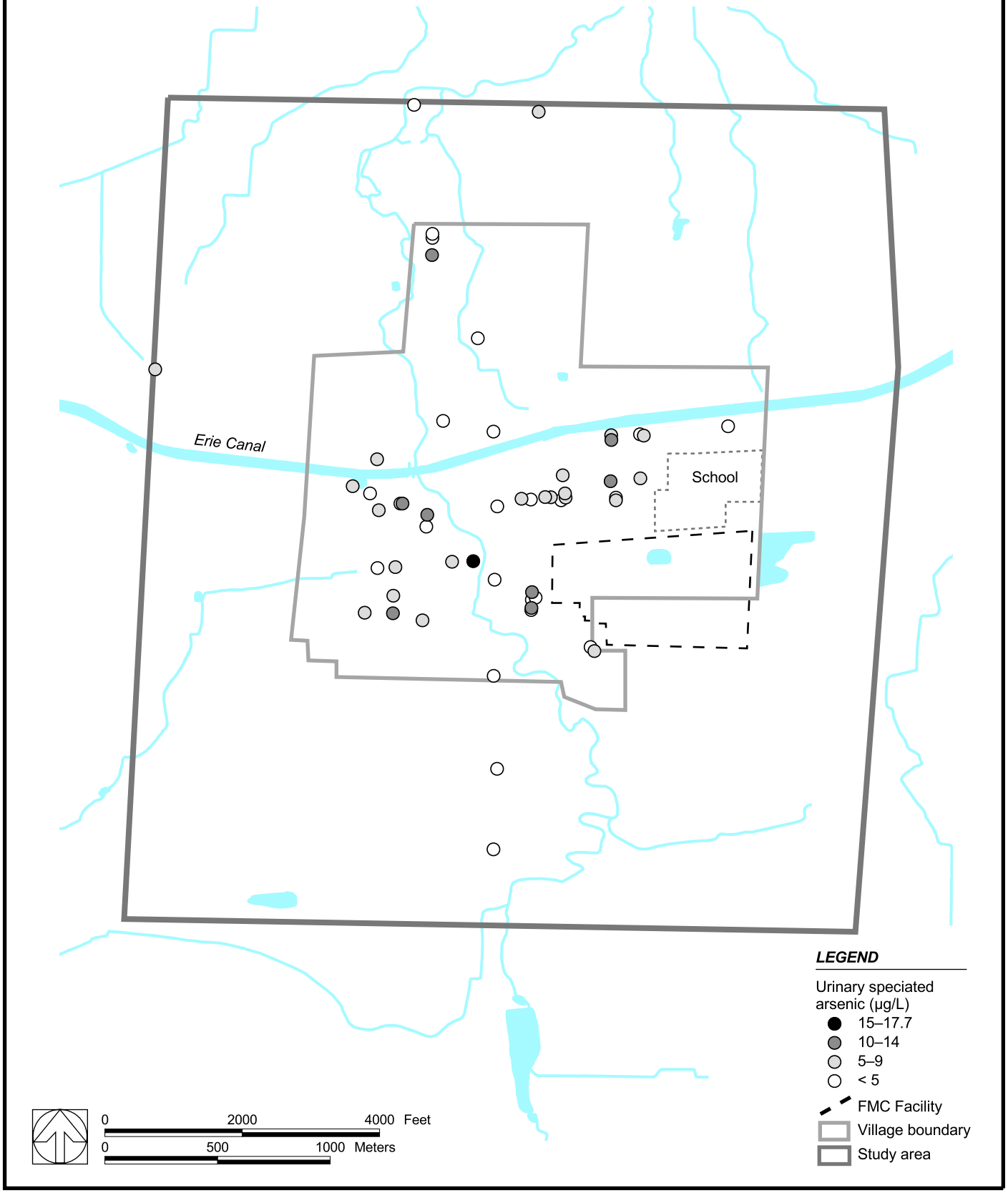


Figure E-2. Average value of speciated arsenic in urine per family—children (less than 7 years old)

Exponent®

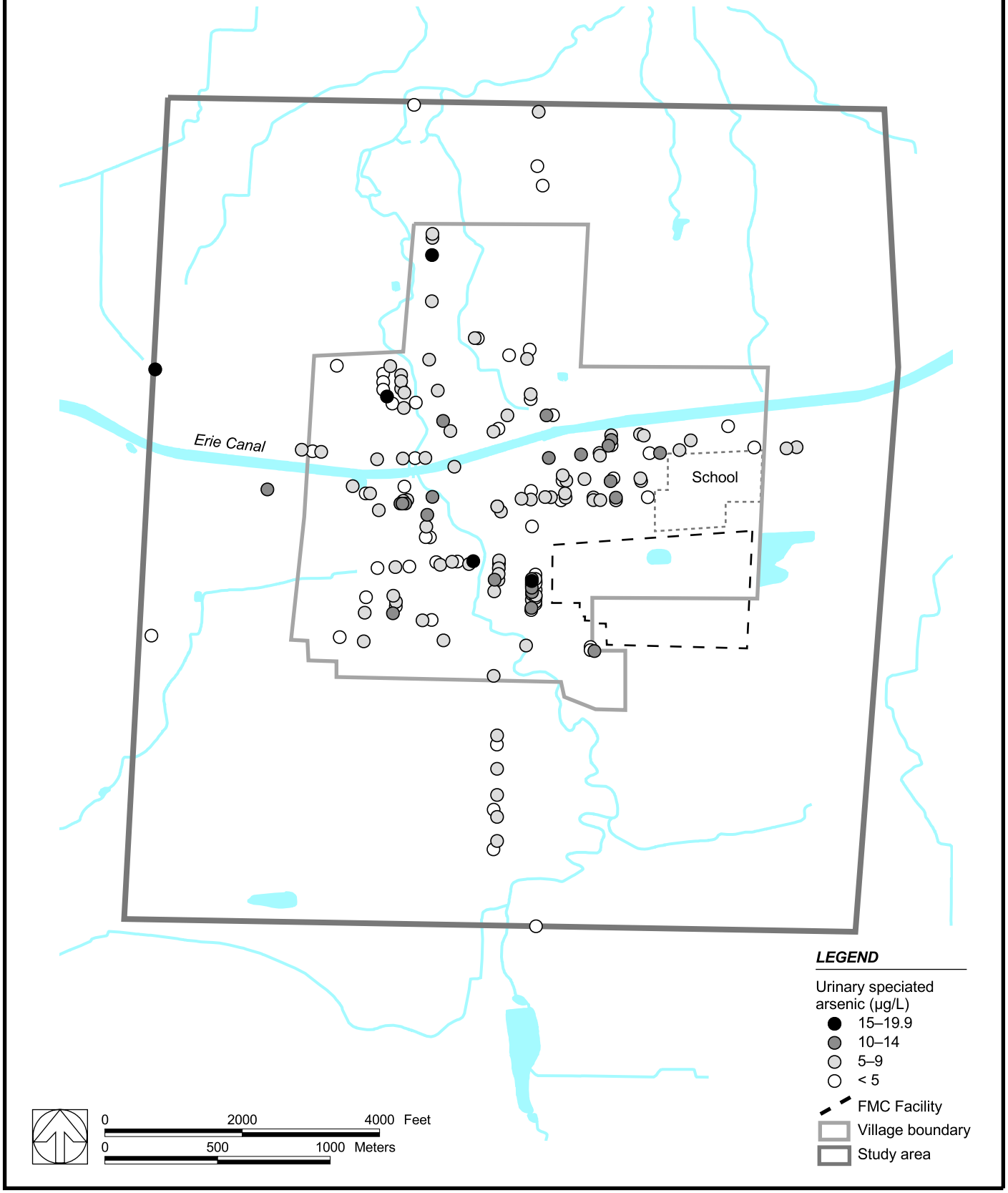


Figure E-3. Highest individual value of speciated arsenic in urine per family—all participants

Exponent®

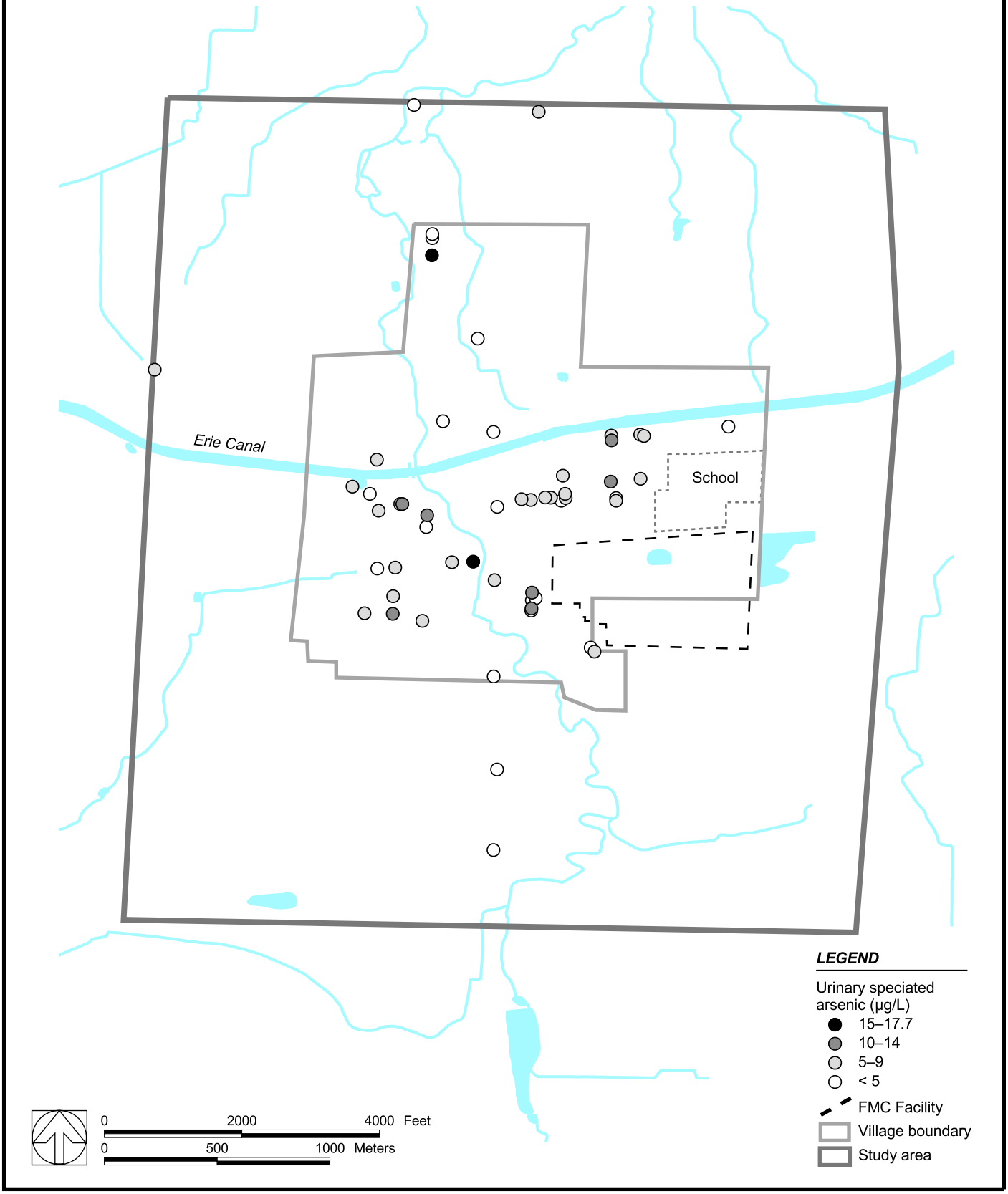


Figure E-4. Highest individual value of speciated arsenic in urine per family—children (less than 7 years old)

Exponent®

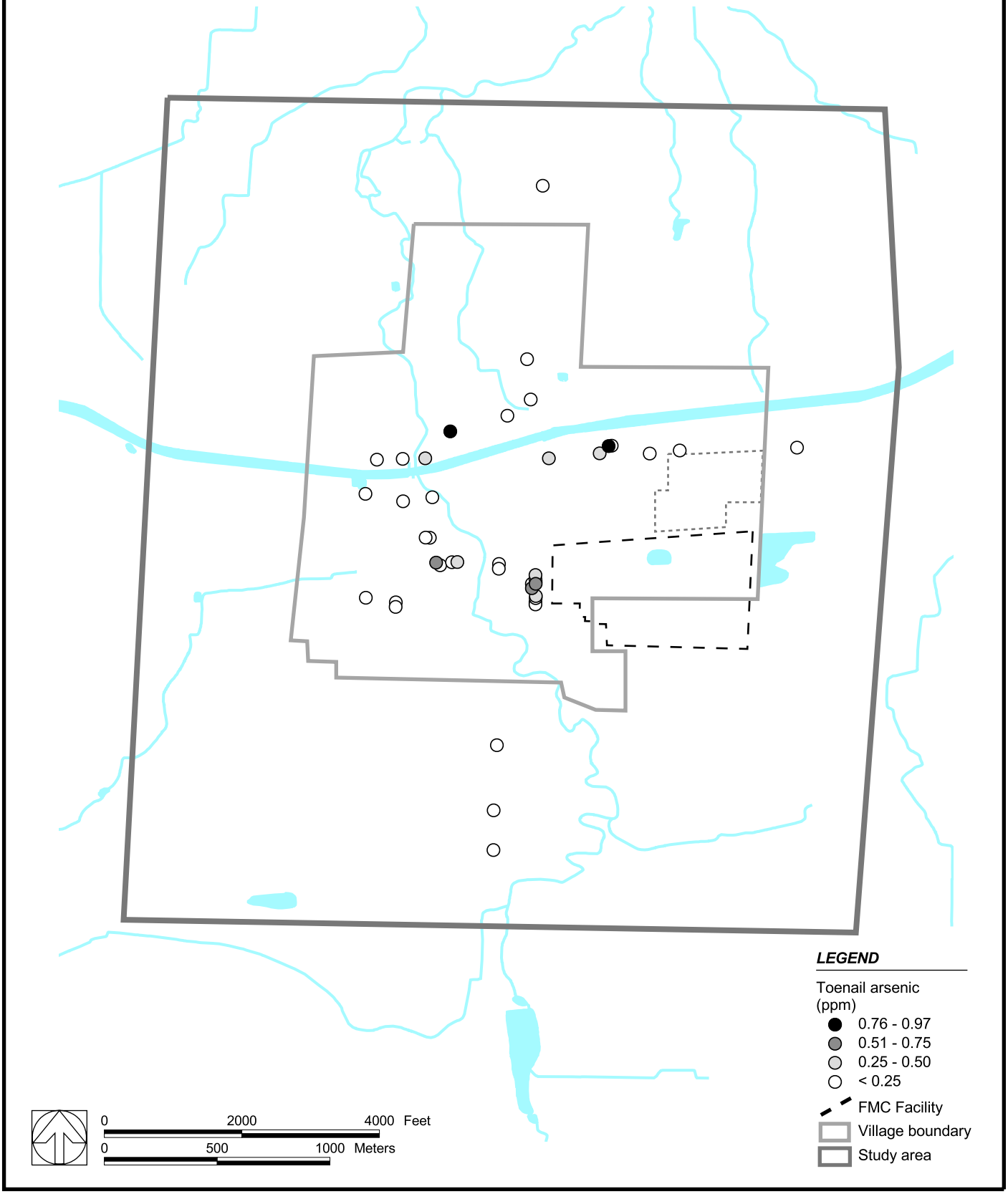


Figure E-5. Highest individual value of total arsenic in toenail samples per family

Exponent®

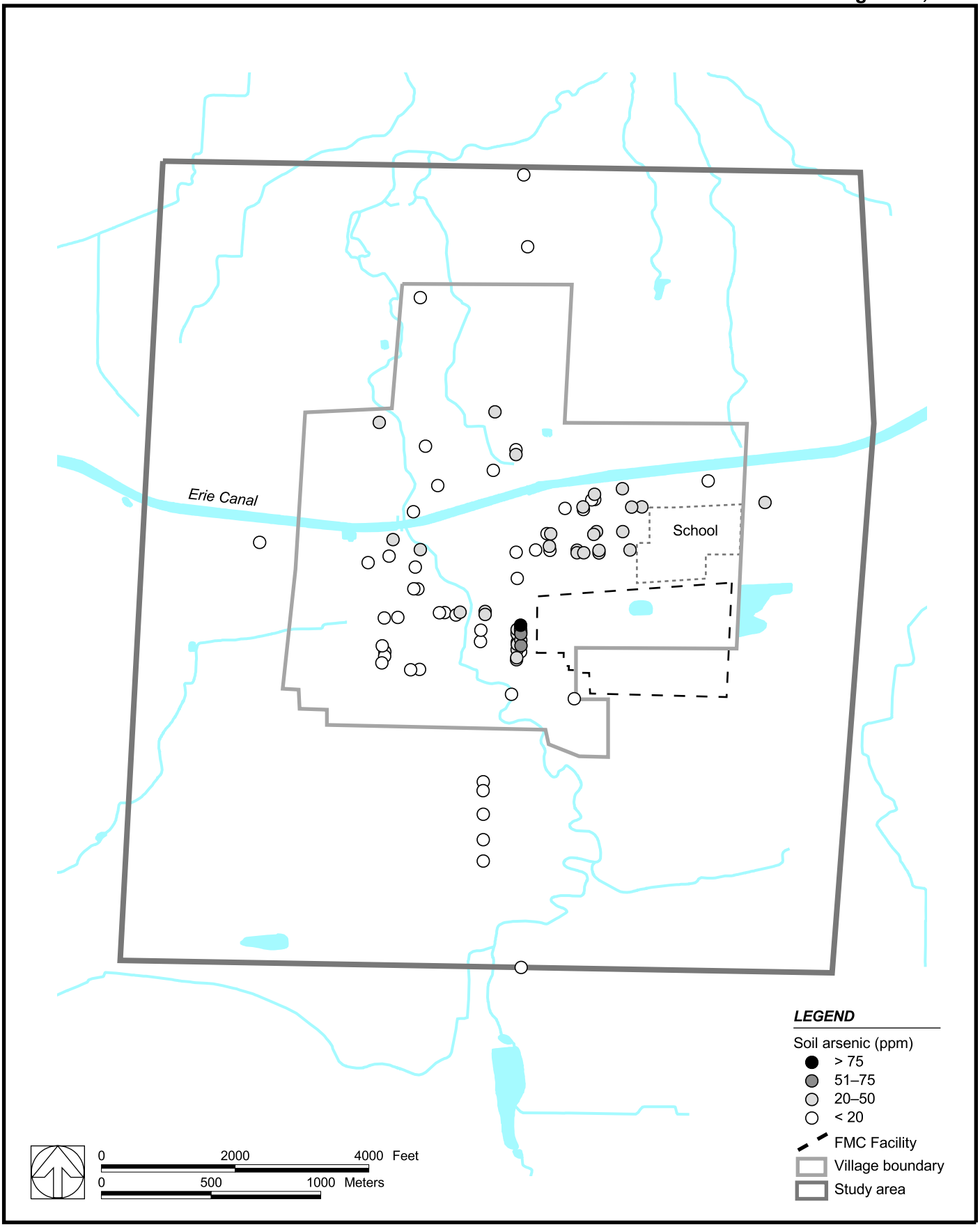


Figure E-6. Average soil arsenic data for play, yard, and garden areas of participant properties

Exponent®

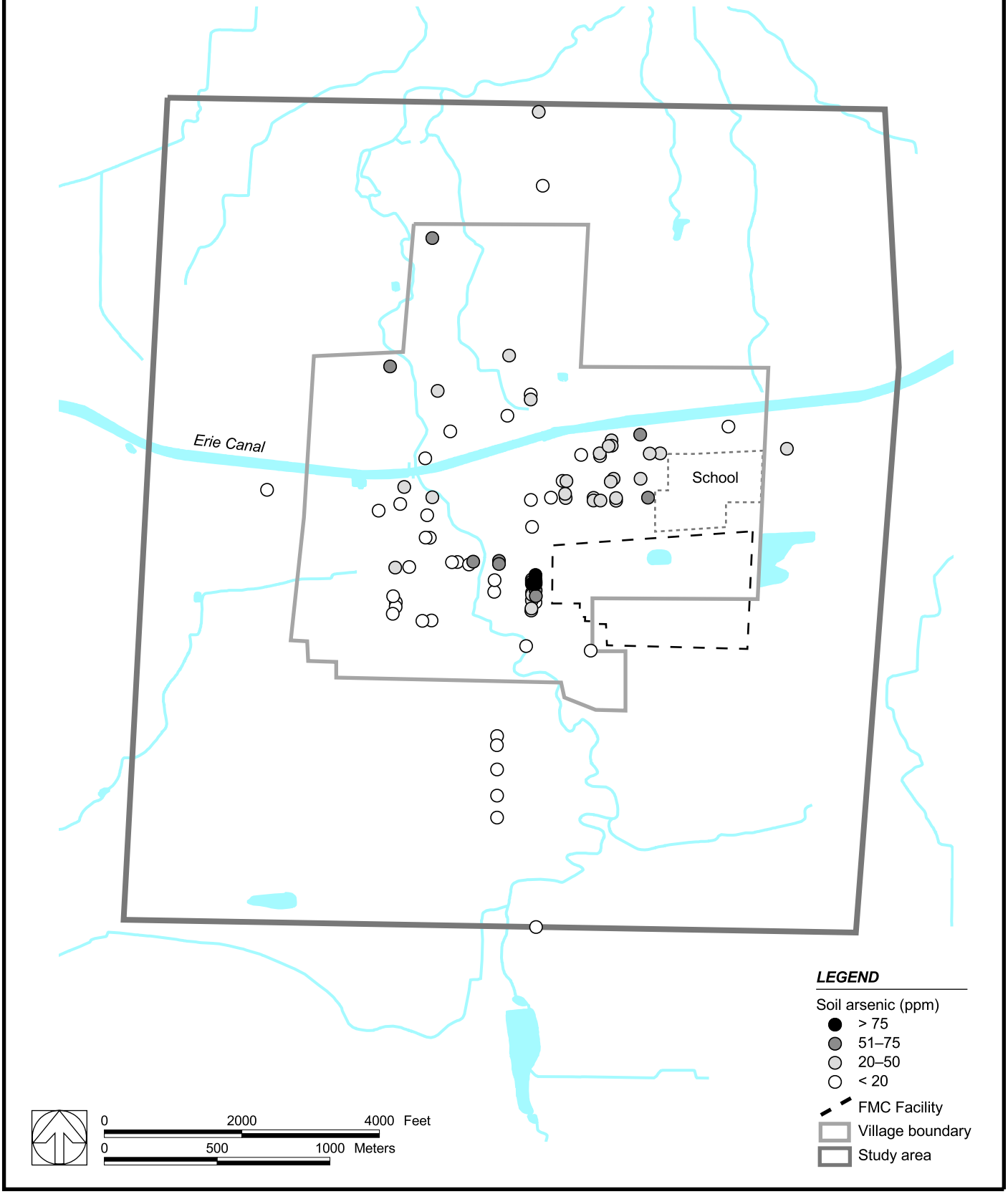


Figure E-7. Maximum soil arsenic data for play, yard, and garden areas of participant properties

Exponent®

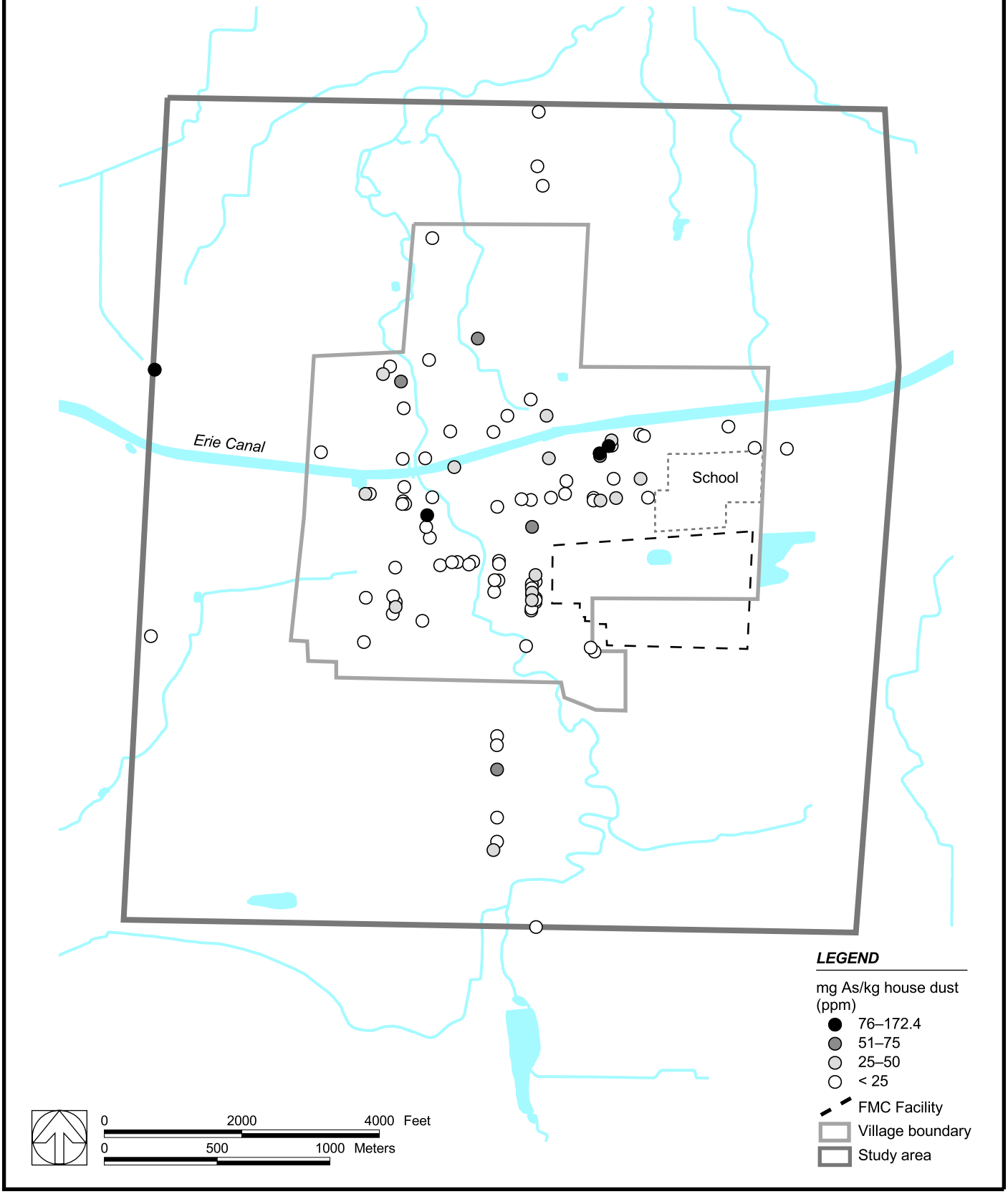


Figure E-8. House dust arsenic concentration (ppm)

Exponent®

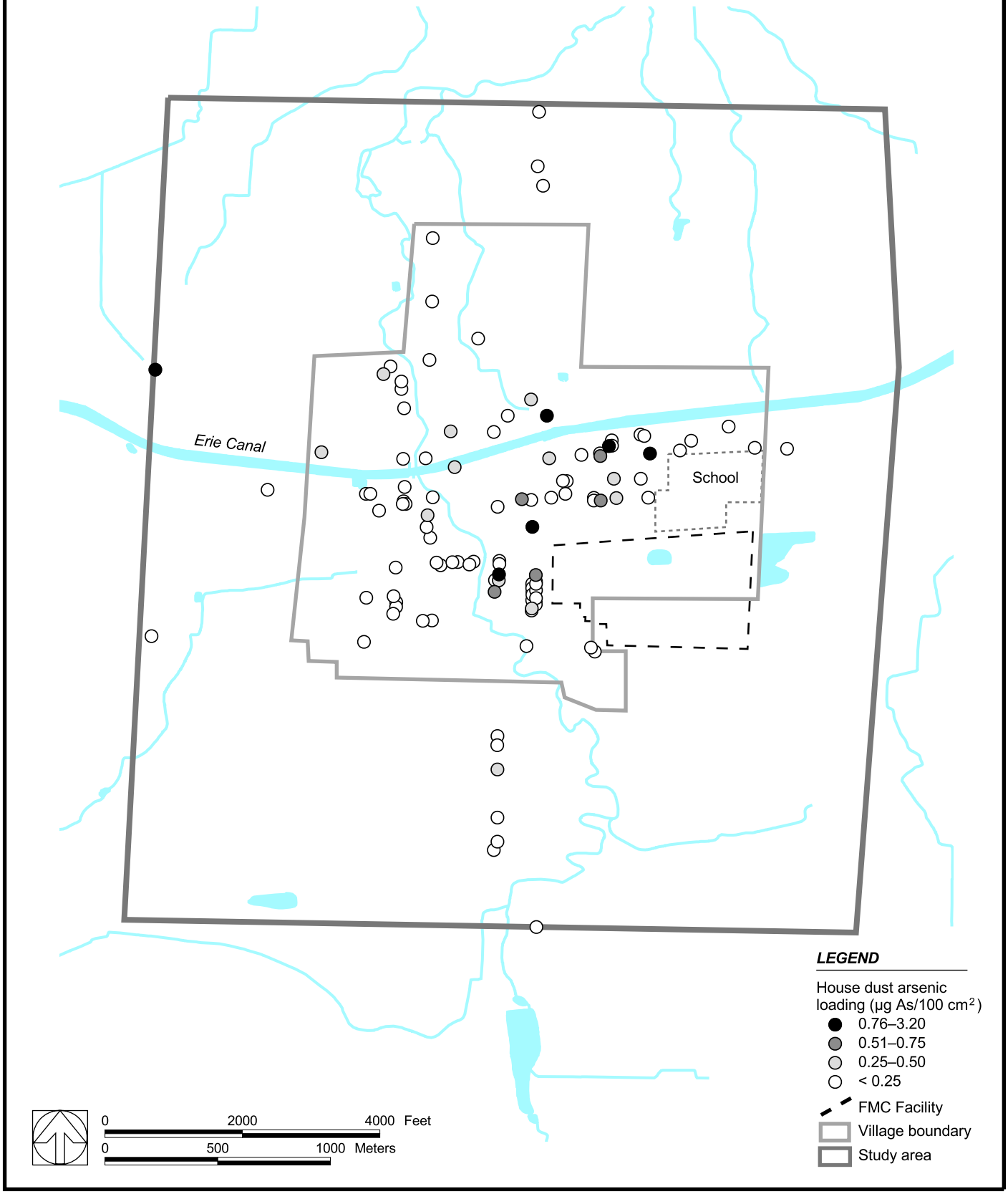


Figure E-9. House dust arsenic surface loading (µg As/100 cm²)

Exponent®

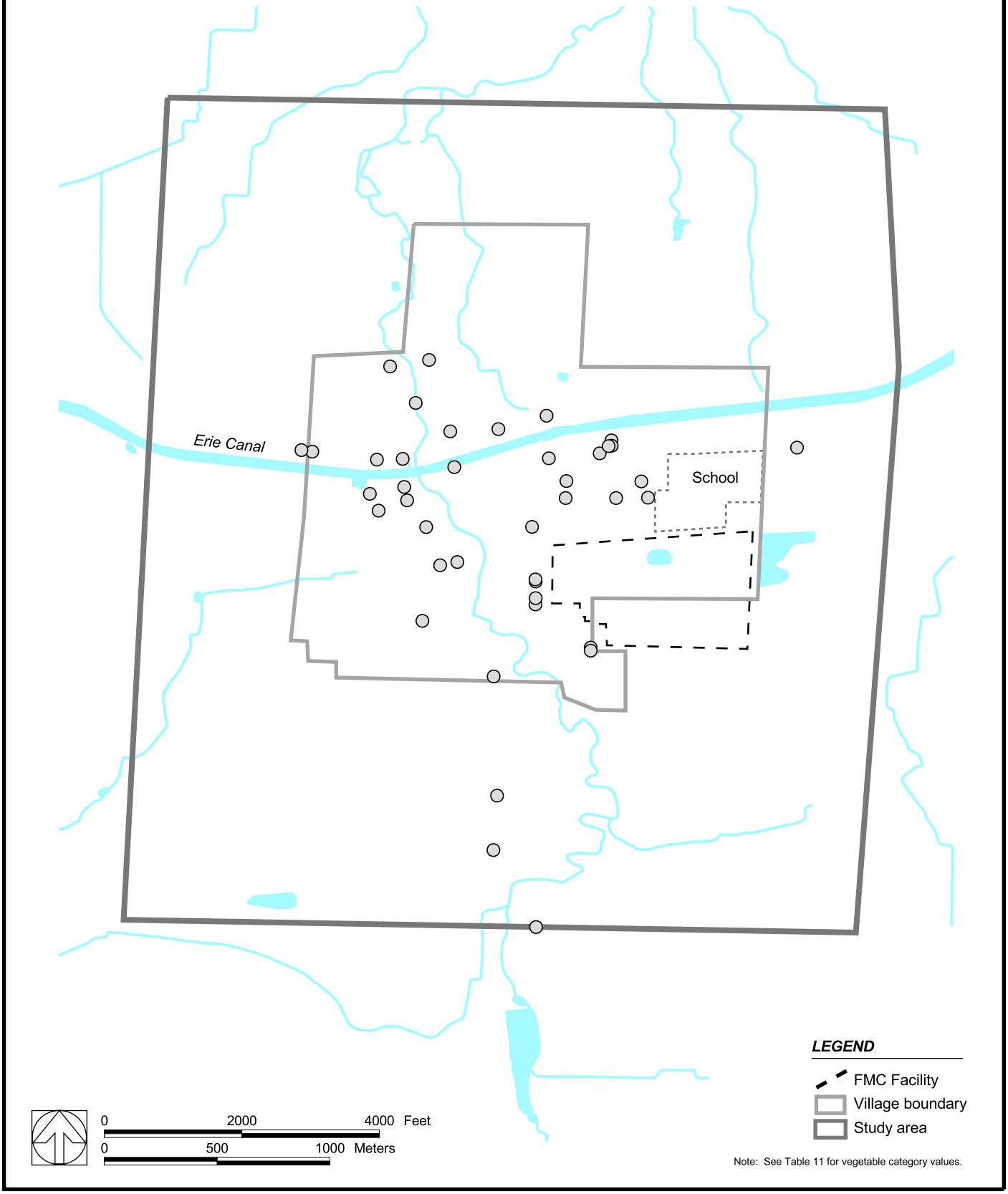


Figure E-10. Location of garden produce sampling

Appendix F

Comparison of Battelle and CDC Results for Urinary Arsenic Analyses

Table F-1. Comparison of Battelle and CDC results for total and speciated arsenic

Total Arsenic (μg/L)		Speciated Arsenic (μg/L)[a]	
Battelle	CDC	Battelle MMA, DMA, and Inorganic Arsenic	CDC MMA, DMA, As(III), and As(V)
5.38	3.4	2.17	3.35
9.24	5.8	1.21	5.25
9.40	7.8	3.30	3.35
9.41	6.1	3.75	5.05
10.3	7.5	3.27	4.95
12.0	7.3	5.17	5.75
12.1	7.6	3.12	4.75
12.6	6.8	5.61	5.35
13.2	9.6	4.61	4.45
16.3	14.2	7.64	8.95
16.5	13.9	9.19	10.5
17.0	10.8	5.59	6.80
17.7	13.3	6.13	8.00
17.9	12.1	8.71	8.65
18.4	8.8	4.79	4.80
19.1	11.0	5.55	4.75
19.2	13.4	10.9	9.30
20.1	15.1	1.05	6.65
21.0	16.4	11.2	10.5
23.6	19.0	9.60	11.4
32.8	27.3	3.70	7.75
39.9	43.9	5.94	7.55
48.2	50.4	4.91	5.45
167	176	7.99	9.40

Note: DMA - dimethylarsinic acid
MMA - monomethylarsonic acid

[a] Speciated arsenic is the sum of species indicated. Half the detection limit is used for samples in which the analyte was not detected.

Table F-2. Comparison of Battelle and CDC results for total, inorganic, and organic arsenic

Battelle Results				CDC Results							
Total Arsenic	Inorganic Arsenic	Organic Arsenic		Total Arsenic	Inorganic Arsenic		Organic Arsenic				
		MMA	DMA		As(III)	As(V)	MMA	DMA	TMAO	AsB	AsC
5.38	0.480 *J*	0.500 *U*	1.44	3.4	1.2 *U*	1.0 *U*	0.90 *U*	1.8	1.0 *U*	1.3	0.6 *U*
9.24	0.709 *J*	0.500 *U*	0.500 *U*	5.8	1.2 *U*	1.0 *U*	0.90 *U*	3.7	1.0 *U*	1.4	0.6 *U*
9.40	0.608 *J*	0.225 *J*	2.47	7.8	1.2 *U*	1.0 *U*	0.90 *U*	1.8	1.0 *U*	3.2	0.6 *U*
9.41	0.798 *J*	0.423 *J*	2.53 *J*	6.1	1.2 *U*	1.0 *U*	0.90 *U*	3.5	1.0 *U*	0.40 *U*	0.6 *U*
10.3	0.733 *J*	0.610	1.93	7.5	1.2 *U*	1.0 *U*	0.90 *U*	3.4	1.0 *U*	0.70	0.6 *U*
12.0	0.731 *J*	0.298 *J*	4.14	7.3	1.2 *U*	1.0 *U*	0.90 *U*	4.2	1.0 *U*	0.40 *U*	0.6 *U*
12.1	1.17 *J*	0.451 *J*	1.50	7.6	1.2 *U*	1.0 *U*	0.90 *U*	3.2	1.0 *U*	0.80	0.6 *U*
12.6	0.905 *J*	0.632	4.07	6.8	1.2 *U*	1.0 *U*	0.90 *U*	3.8	1.0 *U*	0.40 *U*	0.6 *U*
13.2	0.844 *J*	0.557	3.21	9.6	1.2 *U*	1.0 *U*	0.90 *U*	2.9	1.0 *U*	5.4	0.6 *U*
16.3	1.53 *J*	0.891	5.22	14.2	1.2 *U*	1.7	0.90 *U*	6.2	1.0 *U*	1.9	0.6 *U*
16.5	1.28 *J*	0.520	7.39	13.9	1.2 *U*	1.0 *U*	0.90 *U*	8.9	1.0 *U*	1.0	0.6 *U*
17.0	0.899 *J*	0.772	3.92	10.8	1.2 *U*	1.0 *U*	1.0	4.7	1.0 *U*	2.5	0.6 *U*
17.7	1.02 *J*	0.89	4.22	13.3	1.2 *U*	1.0 *U*	1.5	5.4	1.0 *U*	1.9	0.6 *U*
17.9	1.89 *J*	1.17	5.65	12.1	1.2 *U*	1.2	0.90 *U*	6.4	1.0 *U*	0.40 *U*	0.6 *U*
18.4	0.99 *J*	0.387 *J*	3.41	8.8	1.2 *U*	1.0 *U*	1.0	2.7	1.0 *U*	1.4	0.6 *U*
19.1	1.38 *J*	1.04	3.13	11.0	1.2 *U*	1.0 *U*	0.90 *U*	3.2	1.0 *U*	3.9	0.6 *U*
19.2	1.11 *J*	0.738	9.02	13.4	1.2 *U*	1.0 *U*	1.2	7.0	1.0 *U*	0.40 *U*	0.6 *U*
20.1	0.550 *J*	0.500 *U*	0.500 *U*	15.1	1.2 *U*	1.0 *U*	0.90 *U*	5.1	1.0 *U*	5.2	0.6 *U*
21.0	0.958 *J*	1.23	9.06	16.4	1.2 *U*	1.2	1.6	7.1	1.0 *U*	3.4	0.6 *U*
23.6	1.12 *J*	1.13	7.35	19.0	1.2 *U*	1.0	1.5	8.3	1.0 *U*	5.0	0.6 *U*
32.8	1.20 *J*	0.338 *J*	2.16	27.3	1.2 *U*	1.1	0.90 *U*	5.6	1.0 *U*	16.9	0.6 *U*
39.9	1.21 *J*	0.648	4.08	43.9	1.2 *U*	1.4	0.90 *U*	5.1	1.0 *U*	21.3	0.6 *U*
48.2	0.879 *J*	0.338 *J*	3.69	50.4	1.2 *U*	1.0 *U*	0.90 *U*	3.9	1.0 *U*	33.3	0.6 *U*
167	0.886 *J*	0.641	6.46	176	1.2 *U*	1.0 *U*	1.0	7.3	1.0 *U*	130.1	0.6 *U*

Note:
As(III) - arsenous acid (III)
As(V) - arsinic acid (V)
AsB - arsenobetaine
AsC - arsenocholine
DMA - dimethylarsinic acid
J - estimated at concentration shown
MMA - monomethylarsonic acid
TMAO - trimethyl arsine oxide
U - undetected at detection limit shown

Appendix G

Results of Statistical Analyses—Children Less Than 13 Years Old and Total Study Population

Contents

Table G-1. Numerical questionnaire responses for children less than 13 years old used in statistical analyses

	N	Mean (±SD)	Median	Minimum	Maximum
Age when urine was taken (years)	142	6.8 (±3.3)	6.8	0.1	12.8
Number of hours played in area/day	132	2.6 (±2.5)	2	0	15
Number of times washed hands/day	141	5.1 (±4.2)	4	0	30
Number of times showered/bathed	141	5.6 (±2.3)	6	1	14
Number of times eating homegrown vegetables or fruits	141	2.7 (±5.8)	0	0	28
Number of servings of seafood	141	0.2 (±0.6)	0	0	4
Number of servings of rice or rice products	141	2.1 (±4.7)	0	0	28
Number of servings of organ meats	141	0.01 (±0.12)	0	0	1
Number of cups of tap water or drinks with tap water	141	4.7 (±6.5)	3	0	42
Number of servings of grapes	137	0.4 (±0.9)	0	0	5
Number of times take food, drinks, etc. outside	138	3.0 (±3.9)	2	0	25
Number in household	142	4.7 (±1.3)	4	2	10

Note: SD - standard deviation

Table G-2. Categorical questionnaire responses and associated urinary arsenic levels for children less than 13 years old

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Gender[b]	Female	74	52.1	0.86 (±1.43)	4.58 (±2.29)	4.64 (±2.00)
	Male	68	47.9	0.80 (±1.42)	4.53 (±1.90)	4.46 (±1.70)
Visited a house/building with ongoing renovations?[b]	Yes	11	7.9	0.80 (±1.29)	5.66 (±2.01)	4.36 (±1.85)
	No	127	91.4	0.84 (±1.43)	4.46 (±2.1)	4.57 (±1.88)
	Don't know	1	0.7	1.14 (--)	5.75 (--)	3.13 (--)
Play with family or neighbor's outdoor pet?[b]	Yes	97	68.8	0.83 (±1.47)	4.54 (±2.09)	4.46 (±1.84)
	No	44	31.2	0.83 (±1.34)	4.56 (±2.17)	4.78 (±1.92)
Limit exposure to soil or dust?[b]	Yes	8	5.9	0.84 (±1.37)	2.98 (±2.59)	3.79 (±1.94)
	No	127	94.1	0.83 (±1.43)	4.59 (±2.09)	4.61 (±1.87)
Visit creeks, streams, or tributaries?[b]	Yes	26	18.4	0.83 (±1.39)	4.96 (±1.92)	4.41 (±1.47)
	No	115	81.6	0.83 (±1.44)	4.46 (±2.15)	4.59 (±1.95)
Spend time in local orchard or produce farm?[b,c]	Yes	3	2.2	0.82 (±1.21)	5.43 (±1.04)	4.88 (±1.35)
	No	135	97.8	0.83 (±1.44)	4.51 (±2.14)	4.56 (±1.88)
Exposed to treated wood?[b]	Yes	93	66.0	0.83 (±1.47)	4.62 (±2.23)	4.71 (±1.89)
	No	48	34.0	0.84 (±1.35)	4.40 (±1.87)	4.28 (±1.81)
Near project where treated wood was sanded, etc.?	Yes	4	2.8	0.82 (±1.13)	3.05 (±1.86)	3.45 (±1.62)
	No	137	97.2	0.83 (±1.43)	4.60 (±2.11)	4.60 (±1.87)
Treated wood used as firewood?	Yes	1	0.7	0.91 (--)	1.89 (--)	2.72 (--)
	No	131	93.6	0.83 (±1.43)	4.61 (±2.10)	4.66 (±1.86)
	Don't know	8	5.7	0.87 (±1.39)	4.03 (±2.26)	3.60 (±1.92)
How often put objects (other than food) into mouth[b]	Yes	48	34.0	0.86 (±1.42)	4.37 (±2.16)	5.09 (±1.85)
	No	74	52.5	0.8 (±1.45)	4.38 (±2.11)	4.32 (±1.89)
	Don't know	19	13.5	0.89 (±1.37)	5.77 (±1.94)	4.27 (±1.78)
Eaten vegetables or fruits from a home garden?[b]	No	98	69.5	0.83 (±1.47)	4.65 (±2.18)	4.61 (±1.92)
	Yes	43	30.5	0.83 (±1.32)	4.30 (±1.94)	4.44 (±1.74)
Eaten seafood?[b]	No	113	79.6	0.83 (±1.44)	4.44 (±2.12)	4.51 (±1.85)
	Yes	29	20.4	0.84 (±1.40)	5.02 (±2.04)	4.74 (±1.93)
Eaten rice or rice products?[b]	No	91	64.5	0.81 (±1.41)	4.27 (±2.15)	4.39 (±1.95)
	Yes	50	35.5	0.87 (±1.46)	5.07 (±2.02)	4.87 (±1.70)

Table G-2. (cont.)

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Family income for 2002[b]	≤$40,000	53	39.6	0.86 (±1.36)	4.73 (±2.08)	4.28 (±1.77)
	>$40,000	81	60.4	0.81 (±1.47)	4.44 (±2.15)	4.88 (±1.89)
Year home built[b]	Before 1940	83	58.5	0.86 (±1.43)	4.73 (±2.05)	4.48 (±1.92)
	1940–1980	41	28.9	0.79 (±1.37)	4.40 (±2.08)	4.50 (±1.66)
	After 1980	5	3.5	0.72 (±1.71)	3.56 (±2.9)	7.16 (±2.41)
	Don't know	13	9.2	0.86 (±1.46)	4.40 (±2.42)	4.40 (±1.91)
Street paved near home?	Yes	139	100.0	0.83 (±1.43)	4.55 (±2.11)	4.58 (±1.87)
Large projects or activites involving digging, moving or adding soil?[b,d]	Yes	68	47.9	0.89 (±1.41)	5.06 (±2.03)	4.72 (±1.88)
	No	72	50.7	0.78 (±1.44)	4.13 (±2.17)	4.45 (±1.85)
	Don't know	2	1.4	0.94 (±1.32)	4.47 (±1.43)	2.85 (±1.14)
Family of hispanic origin?	Yes	4	2.9	0.93 (±1.24)	6.30 (±1.85)	4.07 (±1.23)
	No	136	97.1	0.83 (±1.43)	4.48 (±2.11)	4.58 (±1.88)
Which group describes your family?	White	129	92.1	0.83 (±1.44)	4.50 (±2.13)	4.60 (±1.88)
	African American	5	3.6	0.73 (±1.19)	5.87 (±1.3)	4.50 (±1.15)
	Native American	4	2.9	0.96 (±1.18)	4.61 (±2.36)	3.07 (±2.21)
	Other	2	1.4	0.70 (±1.17)	2.90 (±2.17)	6.26 (±1.16)
Participate in WIC?	Yes	7	5.0	0.92 (±1.71)	3.46 (±3.11)	4.73 (±2.79)
	No	134	95.0	0.83 (±1.41)	4.62 (±2.06)	4.57 (±1.82)
Exposure to smoke in past 7 days[b]	Yes	25	19.8	0.77 (±1.51)	3.71 (±2.32)	4.39 (±1.95)
	No	101	80.2	0.84 (±1.40)	4.82 (±2.03)	4.47 (±1.82)

Note: GSD - geometric standard deviation

[a] Total number of subjects that responded to each question varied.

[b] Included in inferential analyses.

[c] Signficant difference in speciated urinary arsenic levels between "Yes" and "No" responses (*t*-test; $p=0.009$).

[d] Signficant difference in inorganic urinary arsenic levels between "Yes" and "No" responses (*t*-test; $p=0.028$).

Table G-3. Correlation coefficient matrix of urinary arsenic levels, environmental arsenic levels, and numerical exposure factors for children less than 13 years old

	Urine Measurements			Environmental Measurements			
Exposure Factor	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)	Average of Yard, Play, and Garden Area (ppm)	Max of Yard, Play, and Garden Area (ppm)	Arsenic Concentration in House Dust (mg As/kg dust)	Surface Loading of Arsenic into House Dust (μg As/100 cm^2)
Speciated arsenic (μg/L)	0.716** (142)						
Speciated arsenic (creatinine corrected) (μg/g)	0.415** (142)	0.593** (142)					
Average of yard, play, and garden area (ppm)	0.118 (76)	0.201 (76)	0.078 (76)				
Max of yard, play and garden area (ppm)	0.024 (76)	0.131 (76)	0.027 (76)	0.923** (76)			
Arsenic concentration in house dust (mg As/kg dust)	-0.100 (88)	0.088 (88)	0.204 (88)	0.155 (57)	0.062 (57)		
Surface loading of arsenic into house dust (μg As/100 cm^2)	0.096 (95)	0.134 (95)	0.123 (95)	-0.157 (63)	-0.165 (63)	0.628** (88)	
Time playing in outdoor area (hours/day)	-0.068 (132)	-0.072 (132)	0.087 (132)	0.072 (72)	0.179 (72)	-0.002 (83)	-0.068 (89)
Washed hands (times/day)	-0.068 (141)	0.029 (141)	-0.107 (141)	-0.220 (76)	-0.132 (76)	-0.036 (88)	0.045 (95)
Number of times showered/bathed	-0.093 (141)	-0.063 (141)	-0.120 (141)	-0.068 (76)	-0.015 (76)	0.107 (88)	0.166 (95)
Number of times eating homegrown vegetables or fruits	-0.017 (141)	-0.057 (141)	-0.082 (141)	-0.090 (76)	0.012 (76)	-0.301* (88)	-0.044 (95)
Number of servings of seafood	-0.050 (141)	0.034 (141)	0.067 (141)	-0.113 (76)	-0.151 (76)	0.241* (88)	0.185 (95)
Number of servings of rice/rice products	0.019 (141)	0.040 (141)	-0.040 (141)	-0.139 (76)	-0.102 (76)	-0.058 (88)	0.061 (95)
Number of cups of tap water or drinks with tap water per day	0.009 (141)	0.081 (141)	0.008 (141)	0.015 (76)	0.052 (76)	-0.195 (88)	0.050 (95)
Number of times take food, drinks outside	0.069 (138)	0.035 (138)	0.018 (138)	0.221 (74)	0.251* (74)	0.074 (87)	-0.002 (94)
Number in household	0.003 (142)	-0.024 (142)	-0.106 (142)	-0.179 (76)	-0.164 (76)	-0.185 (88)	-0.065 (95)
Age of child when urine was taken (years)	0.166* (142)	0.294** (142)	-0.310** (142)	0.036 (76)	-0.017 (76)	0.073 (88)	0.138 (95)

Note: Numbers in parentheses indicate the sample sizes.

Urinary and environmental variables were log-transformed before analysis.

* - $p<0.05$

** - $p<0.01$

Table G-4. Numerical questionnaire responses for the total study population used in statistical analyses

	N	Mean (±SD)	Median	Minimum	Maximum
Age when urine was taken (years)	439	30.9 (±22)	31.1	0.1	92.8
Number of hours played in area/day	423	1.2 (±1.8)	0.4	0	15
Number of times washed hands/day	428	8.1 (±9.9)	6	0	100
Number of times showered/bathed	433	6.4 (±2.7)	7	1	21
Number of times eating homegrown vegetables or fruits	435	6.4 (±16.7)	0	0	196
Number of servings of seafood	434	0.4 (±0.8)	0	0	6
Number of servings of rice or rice products	435	2.7 (±6.1)	0	0	70
Number of servings of organ meats	434	0.02 (±0.1)	0	0	2
Number of cups of tap water or drinks with tap water	433	5.0 (±6.3)	3	0	49
Number of servings of grapes?	430	0.5 (±1.2)	0	0	7
Number of times take food, drinks, etc. outside	429	1.8 (±3)	0	0	25
Number of people in household	437	3.9 (±1.7)	4	1	10

Note: SD - standard deviation

Table G-5. Categorical questionnaire responses and associated urinary arsenic levels for the total study population

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Gender[b,c,d]	Female	233	53.1	0.76 (±1.39)	3.63 (±2.05)	3.41 (±1.99)
	Male	206	46.9	0.81 (±1.43)	4.17 (±1.81)	3.29 (±1.76)
Visited a house/building with ongoing renovations?[b]	Yes	35	8.1	0.79 (±1.28)	4.20 (±1.99)	3.32 (±2.06)
	No	395	91.2	0.78 (±1.41)	3.83 (±1.94)	3.34 (±1.88)
	Don't know	3	0.7	0.94 (±1.19)	5.71 (±1.08)	3.54 (±1.11)
Play with family or neighbor's outdoor pet?[b]	Yes	278	63.9	0.79 (±1.42)	3.96 (±1.95)	3.33 (±1.87)
	No	157	36.1	0.77 (±1.38)	3.73 (±1.97)	3.38 (±1.92)
Limit exposure to soil or dust?[b]	Yes	28	6.6	0.72 (±1.35)	3.26 (±2.16)	3.36 (±2.23)
	No	397	93.4	0.79 (±1.41)	3.90 (±1.93)	3.35 (±1.86)
Visit creeks, streams, or tributaries?[b]	Yes	68	15.7	0.79 (±1.39)	4.08 (±1.90)	3.26 (±1.69)
	No	366	84.3	0.78 (±1.41)	3.84 (±1.96)	3.36 (±1.92)
Spend time in local orchard or produce farm?[b,c]	Yes	16	3.7	0.74 (±1.32)	3.66 (±1.93)	3.48 (±2.07)
	No	414	96.3	0.79 (±1.41)	3.87 (±1.96)	3.34 (±1.88)
Exposed to treated wood?[b]	Yes	163	37.5	0.77 (±1.42)	3.77 (±1.99)	3.21 (±1.89)
	No	272	62.5	0.77 (±1.38)	3.77 (±1.88)	3.21 (±1.88)
Near project where treated wood was sanded, etc.?	Yes	28	6.5	0.78 (±1.33)	3.27 (±1.95)	2.51 (±1.91)
	No	402	92.6	0.78 (±1.41)	3.90 (±1.95)	3.43 (±1.88)
	Don't know	4	0.9	0.88 (±1.20)	5.43 (±1.31)	2.57 (±1.54)
Treated wood used as firewood?	Yes	13	3.0	0.75 (±1.36)	3.02 (±1.71)	2.32 (±1.45)
	No	402	92.6	0.78 (±1.40)	3.90 (±1.96)	3.39 (±1.90)
	Don't know	19	4.4	0.87 (±1.47)	4.03 (±2.01)	3.29 (±1.70)
How often put objects (other than food) into mouth?[b,e]	Yes	119	27.4	0.79 (±1.37)	4.00 (±1.94)	3.68 (±1.87)
	No	285	65.5	0.77 (±1.42)	3.71 (±1.95)	3.20 (±1.90)
	Don't know	31	7.1	0.91 (±1.39)	5.13 (±1.86)	3.59 (±1.82)
Eaten vegetables or fruits from a home garden?[b]	No	286	66.1	0.78 (±1.42)	3.98 (±1.96)	3.37 (±1.90)
	Yes	147	33.9	0.78 (±1.39)	3.70 (±1.92)	3.33 (±1.86)
Eaten seafood?[b]	No	310	71.3	0.79 (±1.41)	3.80 (±1.96)	3.25 (±1.87)
	Yes	125	28.7	0.77 (±1.40)	4.05 (±1.91)	3.60 (±1.89)
Eaten rice or rice products?[b,d,e]	No	308	70.8	0.77 (±1.40)	3.65 (±1.95)	3.14 (±1.91)
	Yes	127	29.2	0.80 (±1.42)	4.46 (±1.91)	3.91 (±1.77)

Table G-5. (cont.)

	Response	N[a]	Percentage	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)
				Geometric Mean (±GSD)		
Family income in 2002[b]	≤$40,000	174	42.8	0.79 (±1.40)	3.74 (±1.95)	3.16 (±1.85)
	>$40,000	233	57.2	0.77 (±1.41)	4.07 (±1.94)	3.60 (±1.89)
Year home built[b]	Before 1940	276	63.2	0.79 (±1.43)	3.98 (±1.93)	3.40 (±1.91)
	1940–1980	113	25.9	0.76 (±1.34)	3.69 (±2.00)	3.16 (±1.77)
	After 1980	20	4.6	0.72 (±1.40)	3.62 (±1.93)	4.17 (±1.88)
	Don't know	28	6.4	0.82 (±1.42)	3.86 (±2.02)	3.18 (±2.04)
Street paved near home?	Yes	430	99.5	0.78 (±1.41)	3.87 (±1.96)	3.36 (±1.89)
	No	2	0.5	0.54 (±1.21)	3.55 (±1.16)	3.72 (±1.17)
Large projects or activites involving digging, moving or addin	Yes	180	41.3	0.81 (±1.39)	4.09 (±1.96)	3.47 (±1.90)
	No	252	57.8	0.76 (±1.41)	3.71 (±1.94)	3.25 (±1.88)
	Don't know	4	0.9	0.99 (±1.23)	5.21 (±1.37)	4.47 (±1.73)
Family of Hispanic origin?	Yes	9	2.1	0.82 (±1.25)	5.02 (±1.88)	3.55 (±1.45)
	No	421	97.9	0.78 (±1.41)	3.84 (±1.95)	3.35 (±1.90)
Which group describes your family?	White	400	93.0	0.78 (±1.41)	3.88 (±1.95)	3.35 (±1.91)
	African American	9	2.1	0.71 (±1.32)	4.78 (±1.48)	3.67 (±1.45)
	Native American	13	3.0	0.79 (±1.34)	3.31 (±2.00)	3.01 (±1.66)
	Other	8	1.9	0.74 (±1.33)	2.94 (±2.22)	3.49 (±1.82)
Participate in WIC?	Yes	12	2.8	0.89 (±1.57)	3.27 (±2.73)	3.85 (±2.56)
	No	421	97.2	0.78 (±1.40)	3.91 (±1.92)	3.36 (±1.86)
Exposure to smoke in past 7 days?[b,d]	Yes	111	26.4	0.79 (±1.44)	3.45 (±1.94)	3.14 (±1.77)
	No	309	73.6	0.78 (±1.39)	4.02 (±1.94)	3.34 (±1.90)

Note: GSD - geometric standard deviation

[a] Total number of subjects that responded to each question varied.

[b] Included in inferential analyses.

[c] Signficant difference in inorganic urinary arsenic levels (*t*-test; $p < 0.05$).

[d] Signficant difference in speciated urinary arsenic levels (*t*-test; $p < 0.05$).

[e] Signficant difference in creatinine-corrected speciated urinary arsenic levels between "Yes" and "No" responses (*t*-test; $p < 0.05$).

Table G-6. Correlation coefficient matrix of urinary arsenic levels, environmental arsenic levels, and numerical exposure factors for the total study population

	Urine Measurements			Environmental Measurements			
Exposure Factor	Inorganic Arsenic (μg/L)	Speciated Arsenic (μg/L)	Creatinine-Corrected Speciated Arsenic (μg/g)	Average of Yard, Play, and Garden Area (ppm)	Max for Yard, Play, and Garden Area (ppm)	Arsenic Concentration in House Dust (mg As/kg dust)	Surface Loading of Arsenic into House Dust (μg As/100 cm^2)
Speciated arsenic (μg/L)	0.624** (439)						
Speciated arsenic (creatinine corrected) (μg/g)	0.290** (439)	0.634** (439)					
Average of yard, play, and garden area (ppm)	-0.009 (249)	0.090 (249)	0.091 (249)				
Max of yard, play and garden area (ppm)	-0.027 (249)	0.088 (249)	0.107 (249)	0.939** (249)			
Arsenic concentration in house dust (mg As/kg dust)	-0.074 (278)	0.110 (278)	0.122* (278)	0.149* (185)	0.044 (185)		
Surface loading of arsenic into house dust (μg As/100 cm^2)	0.022 (305)	0.070 (305)	0.042 (305)	0.045 (203)	-0.022 (203)	0.713** (278)	
Time playing in outdoor area (hours/day)	0.010 (423)	0.078 (423)	0.182** (423)	-0.011 (242)	0.022 (242)	0.017 (267)	0.012 (293)
Washed hands (times/day)	-0.082 (428)	-0.086 (428)	-0.043 (428)	0.009 (243)	0.023 (243)	-0.014 (268)	-0.008 (295)
Number of times showered/bathed	-0.045 (433)	0.004 (433)	-0.092 (433)	-0.114 (246)	-0.113 (246)	0.155* (274)	0.137* (300)
Number of times eating homegrown vegetables or fruits	-0.046 (435)	-0.097* (435)	-0.038 (435)	-0.106 (248)	-0.075 (248)	-0.066 (275)	-0.018 (302)
Number of servings of seafood	-0.059 (434)	0.064 (434)	0.098* (434)	-0.081 (248)	-0.036 (248)	0.102 (274)	0.053 (301)
Number of servings of rice/rice products	-0.0004 (435)	0.080 (435)	0.073 (435)	-0.087 (248)	-0.042 (248)	0.038 (275)	0.052 (302)
Number of cups of tap water or drinks with tap water per day	-0.023 (433)	0.018 (433)	0.042 (433)	0.023 (248)	0.036 (248)	0.014 (275)	0.082 (302)
Number of times take food, drinks outside	0.054 (429)	0.092 (429)	0.118* (429)	0.062 (245)	0.066 (245)	0.088 (272)	0.022 (299)
Number in household	0.163** (439)	0.092 (439)	-0.020 (439)	-0.166** (249)	-0.152* (249)	-0.078 (278)	-0.041 (305)
Age when urine was taken (years)	-0.209** (439)	-0.158** (439)	-0.127** (439)	0.100 (249)	0.104 (249)	-0.032 (278)	-0.007 (305)

Note: Numbers in parentheses indicate the sample sizes.

Urinary and environmental variables were log-transformed before analysis.

* - $p < 0.05$

** - $p < 0.01$