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Estimation of Dietary Intake of Inorganic Arsenic in U.S. Children

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ABSTRACT

Arsenic is a natural component of the environment and is ubiquitous in soils, water, and the diet. Because dietary intake can be a significant source of background exposure to inorganic arsenic (the most toxicologically significant form), accurate intake estimates are needed to provide a context for risk management of arsenic exposure. Intake of inorganic arsenic by adults is fairly well characterized, but previous estimates of childhood intake were based on inorganic arsenic analyses in a limited number of foods (13 food types). This article estimates dietary intake for U.S. children (1 to 6 years of age) based on reported inorganic arsenic concentrations in 38 foods and in water used in cooking those foods (inorganic arsenic concentration of 0.8 μg/L), and U.S. Department of Agriculture food consumption data. This information is combined using a probabilistic software model to extract food consumption patterns and compute exposure distributions. The mean childhood dietary intake estimate for inorganic arsenic was 3.2 μg/day with a range of 1.6 to 6.2 μg/day for the 10th and 95th percentiles, respectively. For both the mean and 95th percentile inorganic arsenic intake rates, intake was predominantly contributed by grain and grain products, fruits and fruit juices, rice and rice products, and milk.

Key Words: inorganic arsenic, children, diet, exposure.

BACKGROUND

Arsenic is a natural component of the environment and is ubiquitous in soils, water, and the diet. Dietary intake can be a significant source of background exposure

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to inorganic arsenic (the most toxicologically significant form) for most individuals, with drinking water estimated as the next most important source in U.S. populations (Meacher *et al.* 2002). Thus, accurate dietary intake estimates are needed to provide a context for risk management decisions regarding arsenic exposure. Specifically, where a given arsenic source results in exposures within the background dietary range, any mitigation measures should be evaluated carefully to determine whether the mitigation would result in a significant change in total exposures to inorganic arsenic and be a public health benefit.

Arsenic has been detected in most foods tested and may be present in foods in a variety of organic as well as inorganic forms, although most studies have reported only total arsenic concentrations in food (Dabeka *et al.* 1993; Gunderson 1995; Tsuda *et al.* 1995; FDA 2003; Tao and Bolger 1999; Egan *et al.* 2002). Estimates of total arsenic intake from food, however, do not permit an understanding of exposure to the forms of arsenic that are believed to be most toxicologically significant—specifically, inorganic arsenic.

Previous estimates of inorganic arsenic intake were based on data of speciated arsenic in only a few foods (Yost *et al.* 1998). Specifically, estimated average inorganic arsenic intakes for adults and children (6 months to 2 years) were 14.0 and 9.4 μg/day, respectively. These estimates were based on 13 foods analyzed for inorganic arsenic together with U.S. Department of Agriculture (USDA) food consumption data for years 1982 through 1990 (Yost *et al.* 1998). Later, based on a comprehensive market basket survey that analyzed inorganic arsenic content in 40 food commodities expected to provide at least 90 percent of dietary inorganic arsenic intake for adults (Schoof *et al.* 1999a), adult intake of inorganic arsenic was estimated to range from 1 to 19.5 μg/day with a mean of 3.2 μg/day (Schoof *et al.* 1999b). Thus, although there was a previous estimate of childhood intake, later refinements in the estimation method suggested that an updated estimate would provide useful information.

This investigation refines previous estimates for children in Yost *et al.* (1998) by coupling the arsenic speciation data from 38 foods and water used in cooking (all results from Schoof *et al.* [1999a] except for beer data) with a comprehensive means of integrating food consumption data to derive a more precise estimate of inorganic arsenic intake in children 1 to 6 years of age. Dietary intake of inorganic arsenic was evaluated for young children because this age group is typically considered a sensitive subpopulation evaluated in investigations of environmental or other exposures to contaminants. For example, young children are often considered more vulnerable to arsenic exposures from soil because they have more hand-to-mouth activity and higher exposure on a body weight basis when compared to adults. Accurate estimates of childhood dietary intake of inorganic arsenic can provide a context for use in evaluating the relative significance of other means of exposure to arsenic.

MATERIALS AND METHODS

Prior Analyses of Inorganic Arsenic in Foods

Schoof *et al.* (1999a) used food consumption data from USDA's 1989–92 *Continuing Surveys of Food Intakes by Individuals* (CSFII; USDA 1992, 1993, 1994) and preliminary data of inorganic arsenic in 13 food types reported in Yost *et al.* (1998)

to identify or select the foods that make up at least 90 percent of the U.S. dietary intake of inorganic arsenic for adults. Specifically, dietary intakes of inorganic arsenic from all foods with either high consumption and/or expected highest levels of inorganic arsenic were estimated, and these intakes (consumption rate × inorganic arsenic concentration) were ordered from highest to lowest to identify the foods representing 90 percent of adult intake of inorganic arsenic. Schoof *et al.* (1999a) then analyzed those 39 foods and water used in cooking for speciated arsenic, including analyses of total arsenic, total inorganic arsenic, and inorganic arsenic as As^{3+}. Four samples of each food were collected from two towns and foods were prepared to be representative of foods as consumed (*e.g.*, meats were cooked—see Schoof *et al.* [1999a] for a detailed description of food collection, preparation, and analytical methods). The inorganic arsenic concentration in tap water used in cooking foods was also reported in Schoof *et al.* (1999a). Analyses of inorganic arsenic were conducted at Battelle Marine Sciences in Sequim, Washington, using preparation techniques and methods reported in detail in Schoof *et al.* (1999a).

Total and Inorganic Arsenic Concentrations

Schoof *et al.* (1999a) detected total arsenic in two or more samples of 35 of the 40 commodities (*i.e.*, all of the commodities except butter, soybean/vegetable oil, salt, whole milk, and green beans). Total inorganic arsenic was detected in two or more samples of 34 of the 40 commodities (*i.e.*, all commodities except soybean/vegetable oil, whole milk, skim milk, chicken, tuna, and orange juice). Inorganic arsenic as As^{3+} was also detected in milk. With the exception of beer, which is not consumed by children, Table 1 shows mean concentrations of total arsenic and total inorganic arsenic concentrations for the 38 foods and tap water used in cooking these commodities reported in Schoof *et al.* (1999a). These inorganic arsenic concentrations were used to derive dietary intake estimates described next.

Dietary Exposure Estimates

Dietary intake of total arsenic by children (1 to 6 years of age) was estimated using Exponent's Foods and Residue Evaluation (FARE™) model and data from USDA's CSFII for years 1994 through 1996 (for the total population) and for 1998 (in the Supplemental Children's Survey) (USDA 2000). FARE™ is a multi-objective probabilistic model for extracting food consumption patterns and computing exposure distributions using demographic and food consumption data from CSFII. FARE™ allows analysis of CSFII consumption data at the commodity level rather than as single ingredients or foods as analyzed in Schoof *et al.* (1999a). For multi-ingredient foods, FARE™ uses "recipes" to translate foods reported in the survey "as eaten" into their component ingredients. The recipes in FARE™ are based on recipes derived by USDA for nutrient analyses, but have been modified for use in additional kinds of intake analyses by including ingredients, food additives, or contaminants.

In order to use available inorganic arsenic concentration data for 38 foods and cooking water to estimate intake from the entire diet, the concentrations of arsenic in analyzed foods were applied to similar foods reported in the survey. For example, the average inorganic arsenic concentration for all fruits combined was applied to estimates for pineapple, mango, and raspberries. In addition, data for individual

Table 1. Mean total and total inorganic arsenic concentrations in foods from Schoof *et al.* (1999a).

Food	Mean total arsenic[a]	Mean total inorganic arsenic[b]
Meat and poultry		
Beef	51.5	0.39 *J*
Chicken	86.4	0.89 *J*
Pork	13.5	0.67 *J*
Fish and shellfish		
Freshwater finfish	160	1.0 *J*
Saltwater finfish	2,356	0.55 *J*
Shrimp	1,890	1.9 *J*
Tuna	512	1.0 *U*
Dairy products		
Milk (both whole and skim milk were analyzed; results combined and applied to all milk products) (As^{3+} concentration was 0.18 ng/g *J*)	2.2	1.0 *U*
Eggs	20	0.98 *J*
Legumes, nuts, and seeds		
Peanut butter (applied to legumes, nuts, and seeds)	43.7	4.7
Grain and grain products (excluding rice)		
Corn (meal)	38.6	4.4
Flour	39.2	10.9
Rice and rice products	303	73.7
Fruits and fruit juices		
Orange	1.6	2.5
Orange juice	4.8	1.0 *U*
Apple, raw	4.8	1.8 *J*
Apple, juice	7.6	2.8
Grape juice[c]	14.1	9.3
Banana	2.3	0.65 *J*
Grape	10.2	3.7
Peach	3.4	2.3
Watermelon[c]	6.7	2.1
Potatoes	2.8	0.82 *J*
Vegetables and vegetable products (excluding potatoes)		
Tomato	9.9	0.92 *J*
Green bean	2.1	1.2 *J*
Lettuce	1.4	1.5 *J*
Pea	4.3	4.5
Spinach	5.1	6.1
Carrot	7.3	3.91
Corn (kernel)	1.6	1.1 *J*
Cucumber	9.6	4.12
Onion	9.6	3.3

(Continued on next page)

Table 1. Mean total and total inorganic arsenic concentrations in foods from Schoof *et al.* (1999a). *(Continued)*

Food	Mean total arsenic[a]	Mean total inorganic arsenic[b]
Condiments, fats, and oils		
Butter	1.8	1.17 *J*
Soybean oil	1.5	0.81 *J*
Salt	4.8	0.84 *J*
Sugars and adjuncts		
Beet sugar	12.2	3.5
Cane sugar	23.8	4.44
Corn syrup	6.0	0.44 *J*
Tap water used in cooking[d]		0.8 *J*

[a]Data analyzed by Battelle Marine Sciences Laboratory, 1529 W. Sequim Bay Rd., Sequim, WA 98382-9099. Concentrations in ng/g in wet weight. Each food type represents an average concentration of four samples, with one of the four samples in each food category analyzed in triplicate (Schoof *et al.* 1999a). Beer was also analyzed by Schoof *et al.* (1999a) (*i.e.*, resulting in 40 analyses), but data for beer was not used in analyses reported here as it is not consumed by children. [b]Where no arsenic was detected (after blank-correcting), one-half the value of the method detection limit was given with a *U* designation. When the concentration of arsenic in food (after blank-correcting) was detected above the blank concentration but below the method detection limit, the value was given a *J* designation. Undetected samples have been included at one-half of the detection limits. All averaged values were computed as follows: (1) If one or more, but not all, values to be averaged were non-detected, 50% of the detection limit(s) was used in calculating the average concentration and (2) Mean values have a *U* or a *J* qualifier if all values used to calculate the mean were *U* or *J* qualified, respectively. [c]Values corrected from original reported in Schoof *et al.* (1999a). Corrections have negligible impact on findings of Schoof *et al.* (1999a) estimate. [d]Total inorganic arsenic for tap water that was used to prepare the cooked commodities reported in Schoof *et al.* (1999a). This water concentration was included in dietary estimates derived here. This estimate does not include inorganic arsenic intake from water as drinking water.

food items such as tomatoes were applied to derive an intake amount for somewhat broader foods such as tomato paste and puree.

Dietary intake of inorganic arsenic was calculated by multiplying the inorganic arsenic concentrations in each food, based on values provided in Table 1, by the amount of food consumed, based on individual dietary estimates from the 1994–96, 1998 CSFII database. Specifically, for each individual reporting 2 days of intake in the CSFII survey, an exposure estimate for each survey day was calculated, and a 2-day average estimate was derived. The following steps outline the general algorithm used by FARE™:

1. The consumption of food 1 by individual 1 at each eating period of day 1 of the survey period is multiplied by the corresponding inorganic arsenic concentration for food 1.

2. Step 1 is repeated for all foods identified in the assessment that were consumed by individual 1 on day 1 of the survey.
3. Steps 1 and 2 are repeated for individual 1 using his/her consumption data for day 2 of the survey.
4. An estimate of the average exposure for all pertinent foods for individual 1 for the 2-day period is obtained by taking the sum of the exposure estimates for all the foods and dividing by the number of days of the survey (*i.e.*, 2 days):

$$\frac{\sum_{d=1}^{2}\sum_{f=1}^{n}\left(C_{fd}\times R_f\right)}{2}$$

where:

d = Day of survey
f = Food number indicator
C_{fd} = Consumption of food f on day d in kg food/kg bw-day
R_f = Inorganic arsenic content in food f in mg/kg

5. Steps 1 to 4 are repeated for all individuals in the population.
6. The frequency distribution of the 2-day average exposure estimates for all individuals is derived by aggregating the exposure estimates calculated in Steps 1 through 5. This methodology was used to derive the average and the percentile estimates of dietary consumption of inorganic arsenic.

In this way, individual variability in food intake and variability in the degree of consumption of foods with various levels of arsenic are incorporated into the overall estimates of exposure.

Calculations were also made to identify what food groups were primary contributors to dietary intake of inorganic arsenic. Specifically, the mean and 95th percentile intake estimates derived using the aforementioned methods were evaluated to determine the percentage of dietary intake of inorganic arsenic related to the following food groups: meat and poultry; fish and shellfish; dairy products; eggs; legumes, nuts, and seeds; grain and grain products (except rice); rice and rice products; fruits and fruit juices; potatoes; vegetables and vegetable products (except potatoes); condiments, fats, and oils; sugars and adjuncts; and water used in cooking (Table 1).

RESULTS

The dietary intake of inorganic arsenic in children 1 to 6 years of age was estimated to have a mean of 3.2 μg/day, with a range of 1.6 to 6.2 μg/day, at the 10th and 95th percentiles, respectively (Figure 1). The total inorganic arsenic intake for individuals at any percentile level depends mainly on the degree of consumption of foods that are relatively high in inorganic arsenic (Table 1) and the total overall intake of food.

This evaluation indicates that four food groups are the primary contributors to inorganic arsenic intake by children, with the relative contributions of these food groups differing for the mean and 95th percentile inorganic arsenic intake rates (Figure 2). Specifically, grain and grain products (excluding rice) (27.5%), fruits and fruit juices (20.9%), rice and rice products (19.8%), and dairy products (14.0%) were the predominant groups contributing to intake of inorganic arsenic in the

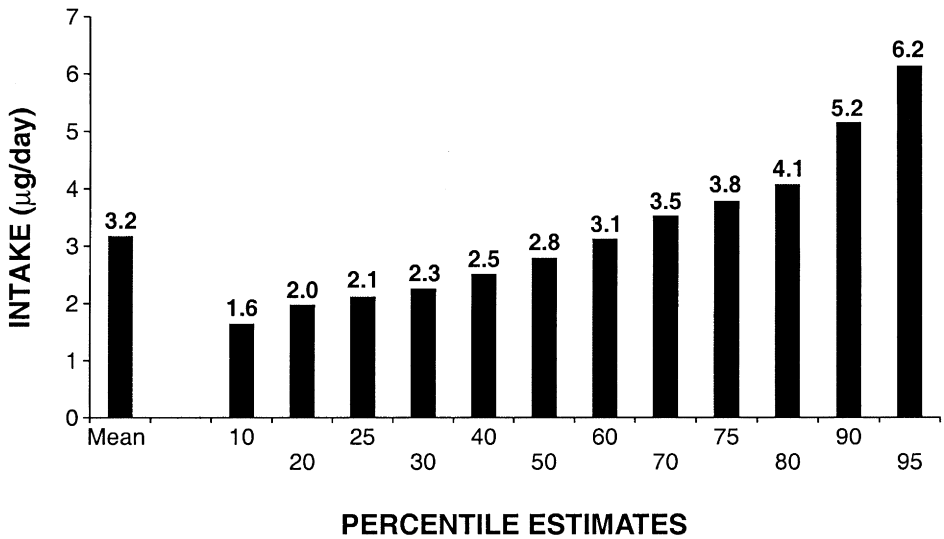

Figure 1. Percentile estimates of inorganic arsenic intake from the diet of children 1 to 6 years of age.

mean estimate. For the 95th percentile inorganic arsenic intake estimate, the same four groups contributed the following percentages to the total intake of inorganic arsenic: rice and rice products (49.9%), fruits and fruit juices (19.8%), grain and grain products (12.1%), and dairy products (9.1%). The next highest contribution was from vegetables and vegetable products (excluding potatoes), with contributions of 5.4% and 2.9% at the mean and 95th percentiles, respectively. The percentage of inorganic arsenic intake associated with all remaining food groups including fish and shellfish that have high total arsenic contents and tap water used in cooking was low (*i.e.*, 0.1% to 4.3%) at both the mean and 95th percentiles. Intake of inorganic arsenic from fish and shellfish was the lowest contribution, making up 0.1% at both the mean and 95th percentiles.

DISCUSSION

The basis for the dietary intake estimates derived here is more robust than that for the previously reported analyses for children (Yost *et al.* 1998) due to the application of inorganic arsenic data for a larger range of foods (*i.e.*, 38 foods and water used in cooking) and the application of the FARE™ model, which is a more accurate and multifaceted analysis of food intake based on U.S. population surveys. The average dietary intake estimate of inorganic arsenic derived (3.2 μg/day for children 1–6 years of age) is similar to that reported in Tao and Bolger (1999) for 2- and 6-year olds (*i.e.*, 4.41–4.64 μg/day) derived through application of an assumed percent inorganic to total arsenic content in seafood of 100% and in all other foods of 10%. The current estimate does have some uncertainties because 38 foods and water used in cooking were used to represent the entire diet. Estimates of inorganic arsenic

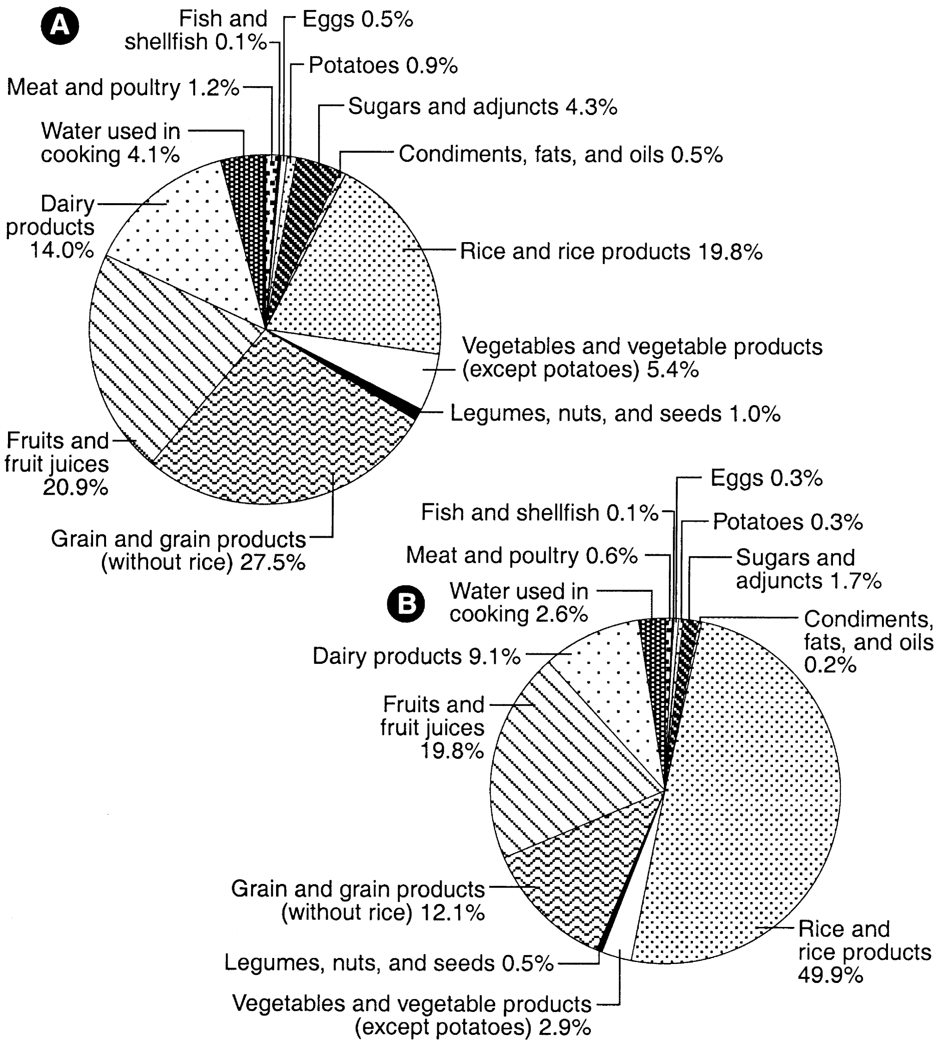

Figure 2. Contribution from specific food groups to dietary intake of inorganic arsenic for children 1 to 6 years of age. To generate the figure, data for the 38 foods and water used in cooking evaluated were first grouped into the 13 general food groups identified in Table 1. (A) Mean dietary inorganic arsenic intake rate (total intake 3.2 μg/day). (B) 95th percentile dietary inorganic arsenic intake rates (total intake 6.2 μg/day).

intake from foods for which specific arsenic data are not available were based on application of data from foods thought to be most representative of those foods or food groups. Moreover, because these foods were collected only from two towns, the inorganic arsenic data may not be entirely representative of the national food supply.

The concentration of inorganic arsenic in milk products was previously identified as an area of uncertainty in estimating children's intake of inorganic arsenic from the diet (Yost *et al.* 1998). In Schoof *et al.* (1999a), inorganic arsenic was analyzed as total inorganic arsenic (including As^{3+} and As^{5+}) and as As^{3+}, with detection limits of 2 ng/g and 1 ng/g, respectively, due to slightly greater precision in the latter analysis. Total inorganic arsenic was undetected in milk at a detection limit of 2 ng/g (Table 1), but trivalent inorganic arsenic [As^{3+}] was detected in milk at 0.18 ng/g. In the total dietary inorganic arsenic intake estimates presented in Figures 1 and 2, estimated intakes of inorganic arsenic from the consumption of milk were calculated using one-half the detection limit for total inorganic arsenic (*i.e.*, an assumed concentration of 1 ng/g). To evaluate the uncertainty in this approach, the concentration of detected trivalent inorganic arsenic (As^{3+}) (*i.e.*, 0.18 ng/g) was also applied. Application of this concentration for As^{3+} in milk rather than one-half the detection limit for total inorganic arsenic had little effect on the overall intake, yielding a mean dietary inorganic arsenic estimate of 2.9 μg/day with 1.4 and 5.8 μg/day at the 10th and 95th percentiles, respectively. Use of the As^{3+} concentration in the estimate did, however, result in a lower proportion of intake related to dairy products as a group (*i.e.*, 3.8% and 4% for As^{3+} in contrast with 14.0% and 9.1%, based on the estimate derived with one-half the detection limit for total inorganic arsenic as shown in Figure 2). Thus, this previously identified uncertainty appears to have little influence on the overall calculated intake, but has some influence on the proportion of intake related to specific food groups.

The available food consumption data within CSFII also contributes some uncertainty for deriving long-term intake estimates. Ideally, consumption data for a longer term (*e.g.*, 14 days) are considered more accurate for evaluating chronic intake, particularly where the concentration of the chemical evaluated differs greatly between foods consumed (*e.g.*, rice in comparison with fruits in this analysis). The 2-day average consumption rates applied here may produce a broader distribution than a longer survey, resulting in overestimates in upper-end percentiles and underestimates of lower percentiles (Buck *et al.* 1997; Chaisson *et al.* 1999). However, only 2 nonconsecutive days' worth of food consumption data are available in the most recent CSFII survey (1994–96, 1998) database. Although the earlier CSFII survey (1989–1991) included food consumption diaries on 3 consecutive days, which might better support estimation of chronic daily intake, rapidly evolving trends in diet and the pace of introduction of new foods support the use of newer data to best represent today's consumers. This assessment, therefore, was based on the most recent consumption data. It should be noted, however, that a number of researchers have proposed or implemented alternative methods that may improve estimates of long-term food consumption patterns (Carrington and Bolger 2002; Nusser *et al.* 1996; Carriquiry 2003; Dwyer *et al.* 2003; Tran *et al.* 2004). The resulting distribution of intakes in the present assessment is narrow, ranging from 1.6 to 6.2 μg/day for the 10th and 95th percentiles, respectively. This may result from the widespread distribution of arsenic in foods.

The intake estimates derived here for the general population may not be fully representative of dietary intake of inorganic arsenic in all subpopulations. For example, the dietary intake estimate reported here also includes water used in food preparation based on an arsenic concentration of 0.8 μg/L. Due to the relatively high use of water in the preparation of food, individuals in regions with higher arsenic concentrations in water likely also have higher dietary intake of inorganic arsenic.

This estimate also does not include intake from water as drinking water. Meacher *et al.* (2002) estimated a mean inorganic arsenic intake from drinking water of 2.35 μg/day for adult females and 2.66 μg/day for adult males based on data collected in the National Arsenic Occurrence Survey (NAOS), which provided arsenic concentrations in drinking water sources for about 20% of the U.S. population. Background arsenic in drinking water in the United States is variable. In the final rule for arsenic in drinking water, the U.S. Environmental Protection Agency (USEPA) reviewed studies of arsenic in drinking water including the NAOS survey and determined that 19.9% of groundwater-based drinking water sources and 5.6% of surface water-based drinking water sources had average arsenic concentrations greater than 3 μg/L. Arsenic concentrations in drinking water also have a wide range, with 0.43% of groundwater resources and 0.10% of surface water resources having average arsenic concentrations greater than 50 μg/L (USEPA 2001). Thus, background arsenic in drinking water is another potential source of exposure beyond that in the diet.

An additional consideration that was beyond the scope of this evaluation is the degree to which ingested inorganic arsenic from food is absorbed through the gastrointestinal tract and enters systemic circulation. This article focuses solely on rates of dietary arsenic intake, and does not attempt to evaluate the absorbed dose of arsenic from the diet. However, other available literature indicates that inorganic arsenic from food is assumed to be fairly well absorbed (*i.e.*, 80–100%; Schoof *et al.* 1999b).

CONCLUSIONS

Analyses were conducted to estimate dietary intake of inorganic arsenic in children in order to provide context for evaluation of arsenic exposures from other sources. Analyses conducted here improve on previous estimates of children's dietary exposure to inorganic arsenic and indicate a mean childhood dietary intake estimate for inorganic arsenic of 3.2 μg/day with a range of 1.6 to 6.2 μg/day for the 10th and 95th percentiles, respectively. Dietary exposure to inorganic arsenic occurs naturally and is unavoidable. These intake estimates provide useful information for estimating background exposures, which are an important consideration in managing risks associated with arsenic exposure. Where a given arsenic source results in exposures within the background dietary range, any mitigation measures should be considered carefully to determine whether the mitigation would result in an actual reduction in total exposure to inorganic arsenic and be a public health benefit.

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