Jara and Winter *International Journal of Food Contamination* 2014, **1**:3
http://www.foodcontaminationjournal.com/content/1/1/3

International Journal of Food Contamination
a SpringerOpen Journal

DATA ARTICLE Open Access

Dietary exposure to total and inorganic arsenic in the United States, 2006–2008

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Abstract

Background: Consumers are frequently exposed to arsenic in foods and considerable public and scientific concern exists regarding the potential health risks from dietary arsenic. Arsenic exists in both organic and inorganic forms and health effects are primarily attributed to inorganic forms. The most common analytical methods used to detect arsenic measure total arsenic, which includes both organic and inorganic forms. It is therefore necessary to make assumptions concerning the amounts of total arsenic found in food samples that represent inorganic arsenic. This work presents a new assessment of US dietary exposure to arsenic using data available from the FDA Total Diet Study from 2006-2008 and a series of scenarios developed to estimate inorganic arsenic levels.

Results: Total arsenic exposures were estimated for 16 population subgroups and ranged from 1.4 $x10^{-1}$ to 4.5 $x10^{-1}$ μg/kg/day. The population subgroup with the highest exposure to total arsenic was 2 year-old children. The major food group contributors to total arsenic exposure for the general US population were marine sources, which accounted for 69 percent of the total arsenic exposure, and grains, legumes and seeds, which accounted for 20 percent. The highest inorganic arsenic exposures occurred for 2 year-old children and ranged from to 1.1 $x10^{-1}$ μg/kg/day to 2.4 $x10^{-1}$ μg/kg/day. Inorganic arsenic exposures for the 2 year-olds were 3.3 to 4.8 times higher than inorganic arsenic exposures for the general population. Under Scenario 5, which assumed that 70 percent of total arsenic from terrestrial sources and 10 percent of total arsenic from marine sources existed as inorganic arsenic, the most important food group contributors to inorganic arsenic for 2 years-olds were grains, legumes and seeds (50 percent), beverages (14 percent), marine sources (13 percent) and snacks and breakfast cereals (12 percent).

Conclusions: The exposures estimates obtained in this work are below the EPA's established RfD of 0.3 μg/kg/day for inorganic arsenic and below EFSA's health concern level of 0.3 to 8.0 μg/kg/day. To improve the accuracy of future arsenic risk assessments, studies should incorporate specific analytical data measuring inorganic arsenic from foods contributing the most to inorganic arsenic exposure.

Keywords: Dietary exposure; Arsenic; Speciation; Food safety

Background

Arsenic is an element distributed in soil, water and air. It is frequently detected in drinking water and food and occurs in the items from natural and anthropogenic sources (EPA 2013; FDA 2013a). Arsenic exists in both organic and inorganic forms and a wide variety of arsenical species have been identified. Inorganic arsenic forms as arsenite (As^{3+}) and arsenate (As^{5+}) are the most harmful forms in health terms and public concern (ATSDR 2005).

The US Environment Protection Agency (EPA) and the International Agency for Research on Cancer (IARC) consider arsenic as a known human carcinogen (EPA 2012a; IARC 2012). In addition to carcinogenicity, arsenic also exerts a variety of acute (short-term) and chronic (long-term) toxicological effects. Acute effects from arsenic exposure have resulted principally from occupational exposure and include gastrointestinal effects such as nausea, diarrhea and abdominal pain (ATSDR 2005). Chronic exposure to arsenic, in addition to bladder, lung and liver cancer, has also been linked to skin toxicity, hyperpigmentation, anemia, peripheral neuropathy and neurocognitive problems (WHO 2001).

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Human exposure to arsenic is a complex issue because it is closely related to environmental pollution, occupation, lifestyle and dietary patterns of the exposed population. While drinking water has traditionally been considered as a principal contributor of consumer exposure to arsenic, recent studies have shown that food represents an even greater source of consumer exposure to arsenic (Georgopolous et al. 2008; Xue et al. 2010). Fruit juices and rice have been identified as key contributors to arsenic exposure in the population and the presence of arsenic in these foods has been the subject of considerable media coverage (Tavernise 2013a, b). Specific attention has been focused upon exposure to children below five years of age due to their significant consumption of these foods.

EPA has set an arsenic safety standard for drinking water at 10 parts per billion (ppb) while the US Food and Drug Administration (FDA) has set an action level of 10 ppb arsenic in apple juice (EPA 2012b; FDA 2013b). Other foods, however, are not subject to arsenic action levels and include rice and rice cereals.

According to EPA, the Reference Dose (RfD) for inorganic arsenic is 0.3 micrograms per kilogram body weight per day (μg/kg/day) (EPA 2012a). The RfD is an estimate of a daily oral exposure to the human population that is likely to be without appreciable risk of detrimental non-cancer effects during a lifetime. The RfD for inorganic arsenic is based on hyperpigmentation, keratosis, and possible vascular complications in humans including sensitive groups (EPA 2012a).

The Joint Food and Agricultural Organization/World Health Organization (FAO/WHO) Expert Committee on Food Additives (JECFA) in 2011 withdrew the provisional weekly intake of 15 μg/kg/day for inorganic arsenic exposure after concluding that more recent studies have shown that a range of adverse effects have been reported at exposures lower than those used when establishing the provisional weekly intake. Based on epidemiological studies, JEFCA considered inorganic arsenic exposures between 2.0 to 7.0 μg/kg/day as levels of concern for potential cancers of the skin, lung and bladder (FAO/WHO 2011).

Studies conducted by the FDA have resulted in dietary estimates of 1.3 to 12.5 μg inorganic arsenic per day (Tao and Bolger 1999). These exposures represent 0.026 to 0.25 μg/kg/day for a 50 kg person. A study by the EPA provided a mean inorganic arsenic exposure of 0.05 μg/kg/day for the overall population and ranges of 0.08 to 0.23 μg/kg/day for children under 5 years of age (Xue et al. 2010).

Estimates of consumer exposure to inorganic arsenic from foods have also been made by the European Food Safety Authority (EFSA). In 2009, inorganic arsenic exposure estimates ranged from 0.13 to 0.56 μg/kg/day for average consumers and from 0.37 to 1.22 μg/kg/day for 95th percentile consumers; the dietary exposure to inorganic arsenic for children under three years of age was estimated to be from 2 to 3 times that of adults (EFSA 2009). In 2014, using different methods to consider arsenic speciation, EFSA reported that mean inorganic arsenic dietary exposure estimates ranged from 0.20 to 1.37 μg/kg/day for infants, toddlers, and other children and from 0.09 to 0.38 μg/kg/day for the adult population. The 95th percentile dietary exposure estimate ranged from 0.36 to 2.09 μg/kg/day for infants, toddlers, and other children. For adults, the 95th percentile dietary exposure ranged from 0.14 to 0.64 μg/kg/day (EFSA 2014).

The toxicity of arsenic to humans is directly related to the form (inorganic vs. organic) that the arsenic exists in, with inorganic forms of arsenic presenting the greatest concern. Unfortunately, due to the high cost and the lack of validated analytical methods for arsenic speciation, most arsenic exposure assessments have relied upon measurements of total arsenic from food items that do not differentiate between organic and inorganic arsenic.

In some cases, differences between total and inorganic arsenic measured from foods can be significant. As an example, foods of marine origin (finfish, shellfish, algae) represent significant sources of dietary arsenic exposure although US studies have indicated that less than 10 percent of the total arsenic in finfish and approximately 30 percent of the total arsenic in shellfish from uncontaminated waters represented inorganic arsenic (Valette-Silver et al. 1999; Lorenzana et al. 2009). Worldwide literature shows similar results with 7.3 percent of total arsenic for finfish and 25 percent of total arsenic for shellfish present in the inorganic form (Mohri et al. 1990; Suñer et al. 1999 2002; Muñoz et al. 2000).

Accurate human exposure assessments for dietary inorganic arsenic require that speciation differences among inorganic and organic forms of arsenic be appropriately considered which presents a significant challenge due to the lack of speciation data. The 1999 FDA inorganic arsenic exposure assessment was performed using the best arsenic contamination data available at the time. In the absence of specific inorganic arsenic data, it was assumed that 10 percent of the total arsenic from all marine sources was considered to be present as inorganic arsenic while 100 percent of the total arsenic from all other sources was in the inorganic arsenic form (Tao and Bolger 1999).

The 2009 EFSA report considered various scenarios relating to the proportion of total arsenic representing inorganic arsenic based upon reasonable assumptions concerning arsenic speciation of various foods obtained from EFSA monitoring and in the scientific literature. For fish and other seafood, levels of inorganic arsenic were obtained using either food contamination data, fixed upper bound estimates, or fixed lower bound estimates. For all other food categories scenarios were developed to reflect either 100 percent, 70 percent, or 50 percent of the total arsenic representing inorganic arsenic. The cross combination of these assumptions produced nine scenarios characterizing inorganic arsenic in marine and terrestrial food sources (EFSA 2009). The recent 2014 EFSA exposure assessment, in contrast, used only data specifically measuring inorganic arsenic levels from fish and other seafood while assuming 70 percent of the total arsenic found in all other foods represented inorganic arsenic (EFSA 2014).

Using a traditional probabilistic exposure assessment method combined with urinary biomarker data examining various forms of inorganic arsenic, EPA researchers concluded that approximately 10 percent of arsenic exposure from foods was in the toxic inorganic form (Xue et al. 2010).

FDA recently published data concerning levels of inorganic arsenic detected from fruit juices and from rice products, including rice, rice cereals, snacks, pasta, grain-based bars and bakery mixtures (FDA 2013c). Unfortunately, arsenic has also been found in numerous other foods using analytical techniques incapable of differentiating organic and inorganic arsenic. As a result, dietary human inorganic arsenic exposure assessments still require assumptions to be made about the relative prevalence of inorganic arsenic compared with total arsenic. The most recent FDA assessment of total dietary arsenic exposure was published in 1999 and reflected arsenic measurements from food taken between 1991 and 1996 (Tao and Bolger 1999).

This paper provides an assessment of US dietary exposure to inorganic arsenic by incorporating the most recently available data (2006–2008) obtained from the FDA's Total Diet Study (TDS). A series of scenarios have been developed to estimate the percentage of total arsenic detected that represents inorganic arsenic and to identify key drivers of inorganic arsenic exposure. Exposure estimates are compared with those in the published literature and with human health criteria to determine the significance of such exposures.

Methods

Dietary intake of arsenic in the US population was estimated using results from the FDA Total Diet Study (TDS) Market Baskets 2006–1 through 2008–4. Food consumption estimates for foods analyzed in the TDS were provided by FDA and were derived from the 1994–1996 Continuing Survey of Food Intakes for Individuals (CSFII) developed by the US Department of Agriculture. Each year, FDA inspectors collect four Market Baskets from retail outlets (each from a different geographical location) containing approximately 280 different food items. The food items are prepared for consumption and then analyzed for pesticide residues, industrial chemicals, radionuclides, and toxic and nutrient elements, including arsenic (FDA 2008).

Arsenic levels detected in the TDS were reported as the total arsenic concentration in each food item (FDA 2010). Dietary exposures to arsenic were calculated by multiplying median food consumption levels by the arsenic concentrations means and divided by body weights (EPA 2011) for each population subgroup. The arsenic concentration means reported from the TDS assumed that no arsenic was present at levels below the limit of detection. Sixteen population subgroups were considered, including all males, all females, infants 6–11 months, children 2 years, children 6 years, children 6–10 years, males (14–16 years, 25–30 years, 40–45 years, 60–65 years, 70 years) and females (14–16 years, 25–30 years, 40–45 years, 60–65 years, 70 years). Total exposure to arsenic for each population subgroup was calculated by combining the arsenic contributions from each food item.

To identify the key food contributors to dietary arsenic exposure, food items were classified into core groups generally consistent with the FDA food list core groups but with some modifications (Pennington 1992; Egan et al. 2007).

Using urinary biomarkers of inorganic arsenic exposure combined with probabilistic arsenic exposure estimates derived from food consumption databases and TDS sampling of foods for total arsenic, it has been estimated that approximately 10 percent of total arsenic in foods is present in the inorganic form (Xue et al. 2010). This approach, while elegant, provides only crude estimates as to the arsenic speciation among specific food groups.

Historically, conservative approaches have been used to assess dietary exposure to inorganic arsenic in food and the majority of them have assumed that 100 percent of terrestrial arsenic is consumed as inorganic arsenic. This assumption may overestimate dietary exposure to inorganic arsenic. On the other hand, some exposure estimates assume all arsenic detected from marine sources exists as organic arsenic, so such approaches could underestimate dietary exposure to inorganic arsenic (Yost et al. 1998; Tao and Bolger 1999).

This work presents additional options to assess arsenic inorganic exposure in food based on the classification of

Table 1 Scenario assumptions: percentage of total arsenic occurring as inorganic arsenic from marine and terrestrial sources

Inorganic arsenic	100% terrestrial	70% terrestrial	50% terrestrial
0% marine sources	**Scenario 1**	**Scenario 4**	**Scenario 7**
10% marine sources	**Scenario 2**	**Scenario 5**	**Scenario 8**
25% marine sources	**Scenario 3**	**Scenario 6**	**Scenario 9**

Table 2 Total arsenic exposure for 16 population subgroups (μg/kg/day)

Core Food groups	A. Milk and cheese	B. Meat and poultry	C. Grains, legumes and seeds	D. Fruits and vegetables	E. Desserts	F. Snacks and breakfast cereals	G. Condiments, sugars and sweeteners	H. Beverages	I. Marine sources	Total exposure
Total US F	1.7E-04	5.5E-03	3.5E-02	3.3E-03	1.3E-03	4.4E-03	6.5E-06	3.5E-03	1.2E-01	1.7E-01
Total US M	1.5E-04	4.7E-03	3.1E-02	2.8E-03	1.1E-03	3.8E-03	5.6E-06	3.0E-03	1.0E-01	1.5E-01
6-11 mo.	1.7E-04	3.6E-03	3.9E-02	2.6E-03	3.8E-03	4.5E-02	7.6E-06	5.3E-02	5.2E-02	2.0E-01
M/F 2 yr	7.1E-04	1.4E-02	1.3E-01	8.5E-03	3.4E-03	3.1E-02	1.2E-05	3.5E-02	2.3E-01	4.5E-01
M/F 6 yr	5.0E-04	1.1E-02	7.8E-02	6.3E-03	5.0E-03	3.0E-02	8.6E-06	9.5E-03	1.7E-01	3.1E-01
M/F 10 yr	3.7E-04	6.5E-03	4.8E-02	3.1E-03	3.5E-03	2.3E-02	9.4E-07	2.0E-03	1.6E-01	2.4E-01
F 14–16 yr	2.0E-04	5.3E-03	3.3E-02	2.2E-03	1.3E-03	5.8E-03	3.0E-06	2.3E-03	1.1E-01	1.6E-01
M 14–16 yr	3.0E-04	6.0E-03	4.1E-02	2.5E-03	2.1E-03	9.5E-03	1.1E-05	1.9E-03	8.5E-02	1.5E-01
F 25–30 yr	1.7E-04	5.0E-03	4.1E-02	3.5E-03	1.2E-03	3.2E-03	3.5E-06	3.5E-03	1.1E-01	1.7E-01
M 25–30 yr	2.3E-04	6.9E-03	3.9E-02	2.4E-03	1.0E-03	2.3E-03	3.9E-06	2.4E-03	9.5E-02	1.5E-01
F 40–45 yr	1.0E-04	4.6E-03	3.1E-02	3.4E-03	1.1E-03	2.7E-03	9.2E-06	3.2E-03	9.7E-02	1.4E-01
M 40–45 yr	1.5E-04	6.0E-03	3.2E-02	2.7E-03	1.2E-03	2.8E-03	1.8E-06	3.2E-03	8.8E-02	1.4E-01
F 60–65 yr	1.0E-04	3.2E-03	2.0E-02	3.5E-03	8.5E-04	3.1E-03	1.5E-05	3.1E-03	1.5E-01	1.9E-01
M 60–65 yr	1.2E-04	4.3E-03	2.7E-02	3.5E-03	1.1E-03	2.8E-03	5.7E-06	3.1E-03	1.3E-01	1.7E-01
F 70 yr	7.8E-05	3.7E-03	1.9E-02	4.1E-03	1.0E-03	3.4E-03	7.6E-06	2.2E-03	1.2E-01	1.5E-01
M 70 yr	9.2E-05	4.1E-03	1.7E-02	3.5E-03	9.6E-04	3.1E-03	1.5E-05	2.7E-03	1.3E-01	1.6E-01

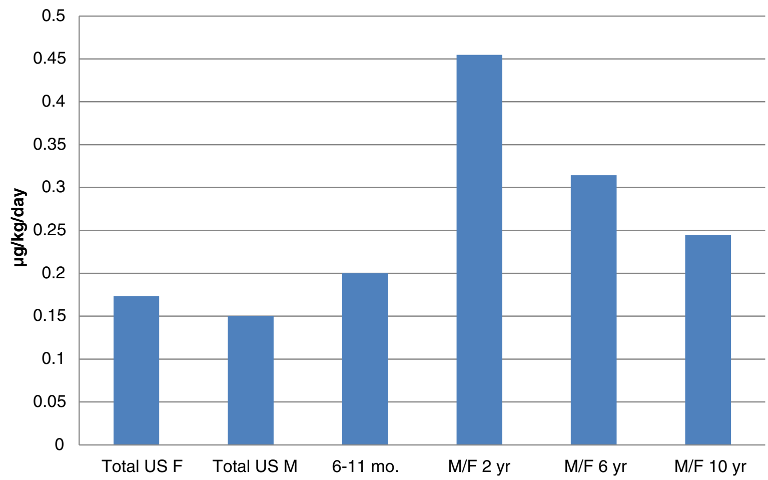

Figure 1 Total arsenic exposure for selected population subgroups.

food in marine and terrestrial sources. We postulate that marine sources contribute to inorganic arsenic exposure and could represent as much as 25 percent of the total arsenic found from marine sources (Muñoz et. al. 2000; Súñer et al. 2002; De Gieter et al. 2002; Lorenzana et al. 2009). Large variability is also seen in the arsenic speciation from food of terrestrial sources. In this study, we estimate that at least 50 percent of total arsenic from terrestrial sources exists as inorganic arsenic (Carbonell-Barrachina et al. 2012; Somella et al. 2013).

In this study, a series of nine scenarios was developed with each scenario making different assumptions of the percentages of total arsenic from marine and terrestrial sources present as inorganic arsenic. The assumptions behind each scenario are presented in Table 1. Scenario 3 presents the worst-case exposure scenario while Scenario 7 presents considers conditions that would result in the lowest exposures. Intermediate exposures are expected from Scenario 5.

Results and discussion

Total arsenic

The total arsenic exposures for the 16 age/sex groups are reported in Table 2. The total exposure estimates ranged from 1.4×10^{-1} to 4.5×10^{-1} µg/kg/day. US female exposure to total arsenic was estimated to be 1.7×10^{-1} µg/kg/day while US male exposure to total arsenic was estimated to be 1.5×10^{-1} µg/kg/day.

The population subgroup with the greatest exposure to total arsenic was 2 year-old children, who were exposed to

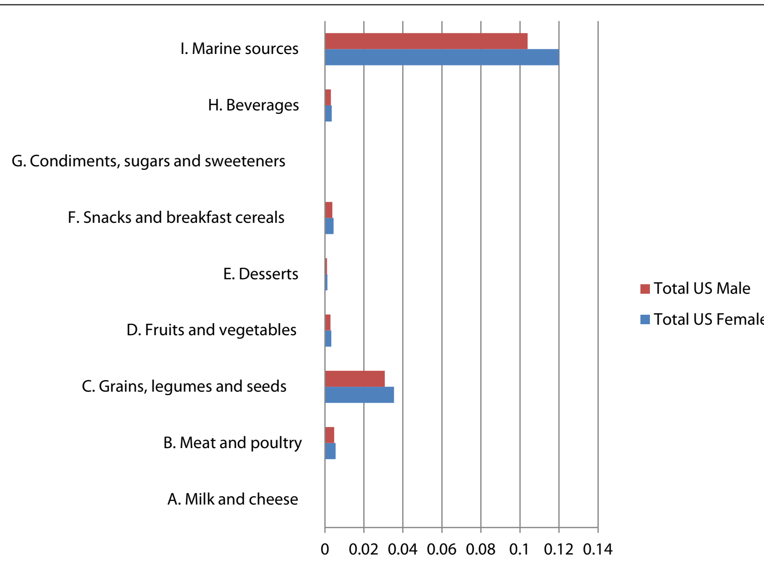

Figure 2 Core food groups and their contributions to total arsenic exposure for US males and females.

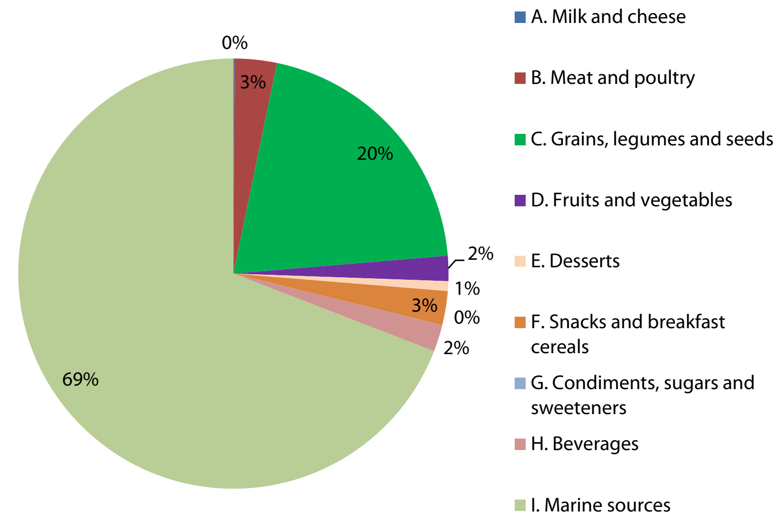

Figure 3 Core food groups and their relative contribution to total arsenic exposure.

4.5×10^{-1} μg/kg/day. Other population subgroups with exposures above at least 2.0×10^{-1} μg/kg/day included 6–11 month infants (2.0×10^{-1} μg/kg/day), 6 year-old children (3.1×10^{-1} μg/kg/day), and 10 year-old children (2.4×10^{-1} μg/kg/day). The lowest total arsenic exposures occurred in the 40–45 year old male and female groups at 1.4×10^{-1} μg/kg/day.

A graph of total arsenic exposure for US males, US females, and the most exposed population subgroups is provided in Figure 1.

The contributions of all of the core food groups to total arsenic exposure in the US population are shown in Figure 2. The top core food group contributing to total arsenic exposure was marine sources, which was responsible for exposures of 1.2×10^{-1} μg/kg/day for females and 1.0×10^{-1} μg/kg/day for males. Grains, legumes and seeds contributed 3.5×10^{-2} μg/kg/day for females and 3.1×10^{-2} μg/kg/day for males. Meat and poultry contributed 5.5×10^{-3} μg/kg/day (female) and 4.7×10^{-3} μg/kg/day (male) while snacks and breakfast cereals contributed 4.4×10^{-3} μg/kg/day and 3.8×10^{-3} μg/kg/day for females and males, respectively.

Figure 3 shows the percentage contribution of the nine core food groups to the total arsenic exposure in the general US population. No significant gender differences were observed. Marine sources were responsible for 69 percent of the total arsenic exposure, followed by grains, legumes and seeds (20 percent). Another three percent of the total arsenic exposure was contributed by both meat and poultry and snacks and breakfast cereals. These top four core food groups were responsible for 95 percent of the total arsenic exposure.

Rice has been identified as a major contributor to dietary arsenic exposure and steps were taken to determine

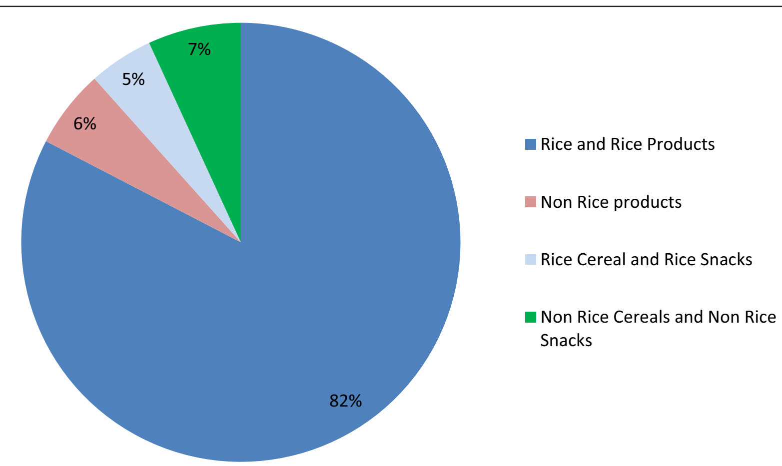

Figure 4 Exposure contribution of rice and rice derivatives from core food groups C and F.

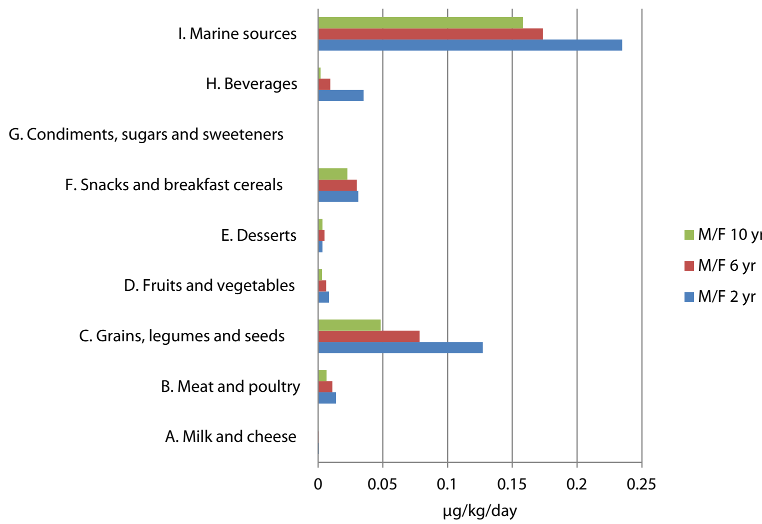

Figure 5 Core food groups and their contributions to total arsenic exposure for US children.

the specific contributions of rice and rice-based foods to total arsenic exposure. Consumption of arsenic from rice and rice-based foods was evident in the grains, legumes, and seeds (rice and rice products) and the snacks and breakfast cereals (rice cereals and rice snacks) core food groups that contributed 23 percent of the total arsenic exposure. To determine the extent by which rice and rice-based foods were responsible for arsenic exposures in these two core food groups, those groups were organized into subcategories including rice and rice products, non-rice products, rice cereals and rice snacks, and non-rice cereals and non-rice snacks. Foods containing rice contributed to 87 percent of the total arsenic exposure from those two core food groups (Figure 4), and the overall contribution of rice and rice-based foods to total arsenic exposure was 20 percent.

Among the three population subgroups receiving the highest total arsenic exposures (2 year-olds, 6 year-olds, 10 year-olds), the relative contributions of the core food groups were similar to those of the total population. Highest exposures (Figure 5) for 6 year-olds and 10 year-olds were from marine sources, followed by grains, legumes and seeds, snacks and breakfast cereals, and beverages. For 2 year-olds, highest total arsenic exposures were also from marine sources and from grains, legumes and seeds, but exposures to total arsenic from consumption of beverages were greater than exposure to total arsenic from snacks and breakfast cereals.

Inorganic arsenic

Inorganic arsenic exposure was estimated for the 16 population subgroups using nine different scenarios. Each scenario considered different assumptions regarding the fraction of total arsenic that was present as inorganic arsenic. Results of the nine scenarios are shown in Table 3.

The highest inorganic arsenic exposure (2.4×10^{-1} µg/kg/day) was observed for the 2 year-old population subgroup under Scenario 3, which assumed that 100 percent of the total arsenic from terrestrial sources and 25 percent of the total arsenic from marine sources was present as inorganic arsenic. Inorganic arsenic exposure for 2 year-olds drops to 1.8×10^{-1} µg/kg/day under Scenario 5 (70 percent terrestrial, 10 percent marine) and to 1.1×10^{-1} µg/kg/day under Scenario 7 (50 percent terrestrial, 0 percent marine). Estimated exposures to inorganic arsenic for the general population were much lower, with male and female exposures ranging from 2.3 to 2.7×10^{-2} µg/kg/day under Scenario 7, 4.3 to 5.0×10^{-2} µg/kg/day under Scenario 5, and 7.2 to 8.4×10^{-2} µg/kg/day under Scenario 3.

Inorganic arsenic exposure estimates for total male, total female, 6–11 month, 2 year-old, 6 year-old, and 10 year-old population subgroups using Scenarios 3, 5, and 7 are presented in Figure 6. Inorganic arsenic exposures for the 2 year-olds ranged from 3.3 to 4.8 times higher than the general population while exposures to the 6 year-olds were 2.7 to 3.0 times higher than the general population and exposures to 10 year-olds were 1.5 to 1.9 times higher than the general population.

The influence of assumptions concerning the percentage of total arsenic present as inorganic arsenic is considered for the 6–11 month, 2 year-old, and 6 year-old population subgroups in Table 4. Under Scenario 3, which considers all total arsenic from terrestrial sources to represent inorganic arsenic and 25 percent of all total arsenic from marine sources to represent inorganic arsenic, inorganic arsenic represents 61 percent of the total arsenic exposure. For Scenario 5 (70 percent terrestrial, 10 percent marine), inorganic arsenic represents 39 percent of the total arsenic exposure while for Scenario

Table 3 Inorganic arsenic exposure for population subgroups based on speciation assumptions (µg/kg/day)

Age/Sex groups	Scenario 1 *100% T 0% MS*	Scenario 2 *100% T 10% MS*	Scenario 3 *100% T 25% MS*	Scenario 4 *70% T 0% MS*	Scenario 5 *70% T 10% MS*	Scenario 6 *70% T 25% MS*	Scenario 7 *50% T 0% MS*	Scenario 8 *50% T 10% MS*	Scenario 9 *50% T 25% MS*
Total US F	5.4E-02	6.6E-02	8.4E-02	3.8E-02	5.0E-02	6.8E-02	2.7E-02	3.9E-02	5.7E-02
Total US M	4.6E-02	5.7E-02	7.2E-02	3.3E-02	4.3E-02	5.9E-02	2.3E-02	3.4E-02	4.9E-02
6-11 mo.	1.5E-01	1.5E-01	1.6E-01	1.0E-01	1.1E-01	1.2E-01	7.4E-02	7.9E-02	8.7E-02
M/F 2 yr	2.2E-01	2.4E-01	2.8E-01	1.5E-01	1.8E-01	2.1E-01	1.1E-01	1.3E-01	1.7E-01
M/F 6 yr	1.4E-01	1.6E-01	1.8E-01	9.8E-02	1.2E-01	1.4E-01	7.0E-02	8.8E-02	1.1E-01
M/F 10 yr	8.7E-02	1.0E-01	1.3E-01	6.1E-02	7.6E-02	1.0E-01	4.3E-02	5.9E-02	8.3E-02
F 14–16 yr	5.0E-02	6.1E-02	7.8E-02	3.5E-02	4.6E-02	6.3E-02	2.5E-02	3.6E-02	5.3E-02
M 14–16 yr	6.3E-02	7.2E-02	8.4E-02	4.4E-02	5.3E-02	6.5E-02	3.2E-02	4.0E-02	5.3E-02
F 25–30 yr	5.7E-02	6.8E-02	8.5E-02	4.0E-02	5.1E-02	6.7E-02	2.9E-02	4.0E-02	5.6E-02
M 25–30 yr	5.4E-02	6.4E-02	7.8E-02	3.8E-02	4.8E-02	6.2E-02	2.7E-02	3.7E-02	5.1E-02
F 40–45 yr	4.6E-02	5.6E-02	7.1E-02	3.3E-02	4.2E-02	5.7E-02	2.3E-02	3.3E-02	4.7E-02
M 40–45 yr	4.8E-02	5.7E-02	7.0E-02	3.4E-02	4.3E-02	5.6E-02	2.4E-02	3.3E-02	4.6E-02
F 60–65 yr	3.4E-02	4.9E-02	7.2E-02	2.4E-02	3.9E-02	6.2E-02	1.7E-02	3.2E-02	5.5E-02
M 60–65 yr	4.2E-02	5.5E-02	7.4E-02	2.9E-02	4.2E-02	6.1E-02	2.1E-02	3.4E-02	5.3E-02
F 70 yr	3.3E-02	4.5E-02	6.3E-02	2.3E-02	3.5E-02	5.3E-02	1.7E-02	2.8E-02	4.6E-02
M 70 yr	3.2E-02	4.5E-02	6.4E-02	2.2E-02	3.5E-02	5.4E-02	1.6E-02	2.9E-02	4.8E-02

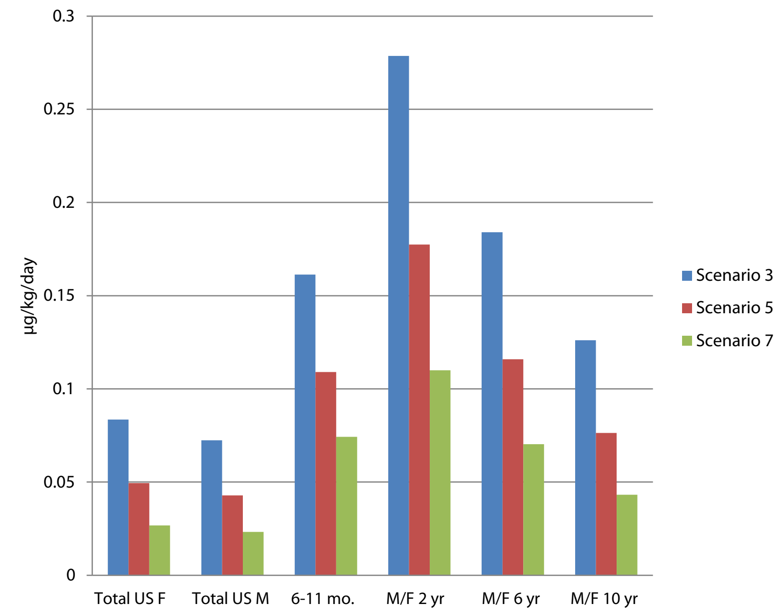

Figure 6 Inorganic arsenic exposure for US children under Scenarios 3, 5 and 7.

7 (50 percent terrestrial, 0 percent marine), inorganic arsenic represents 24 percent of the total arsenic exposure.

Food group contributions to inorganic arsenic exposure under Scenario 5 are presented in Figures 7, 8, and 9 for the general US population, 2 year-olds, and 6–11 month infants, respectively. For the general US population (Figure 7), 74 percent of the inorganic arsenic exposure came from just two food groups: grains, legumes and seeds (50 percent) and marine sources (24 percent). Grains, legumes and seeds also contributed 50 percent of the inorganic arsenic exposure for the 2 year-old population subgroup (Figure 8), but significant contributions were also seen from beverages (14 percent), marine sources (13 percent), and snacks and breakfast cereals (12 percent). Food group contributions for the 6–11 month infant population subgroup (Figure 9) were dramatically different than for the 2 year-olds; the major contributor for 6–11 month infants was beverages (34 percent), followed by snacks and breakfast cereals (29 percent), and grains, legumes and seeds (25 percent).

Table 4 Percentage of total arsenic represented as inorganic arsenic for selected population subgroups under all scenarios

	Inorganic arsenic contribution percentage (%)		
Age/Sex subgroups	**6-11 mo.**	**M/F 2 yr.**	**M/F 6 yr.**
Scenario 1 ***100% T + 0% MS***	74	48	45
Scenario 2 ***100% T + 10% MS***	77	54	50
Scenario 3 ***100% T + 25% MS***	81	61	59
Scenario 4 ***70% T + 0% MS***	52	34	31
Scenario 5 ***70% T +10% MS***	55	39	37
Scenario 6 ***70% T + 25% MS***	58	47	45
Scenario 7 ***50% T + 0% MS***	37	24	22
Scenario 8 ***50% T + 10% MS***	40	29	28
Scenario 9 ***50% T + 25% MS***	44	37	36

Results obtained in this study can be compared to those found by researchers using different food consumption and food contamination databases. In terms of total arsenic exposure, our findings of a general US population average exposure of 0.16 μg/kg/day was considerably lower than the EPA estimate of 0.36 μg/kg/day (Xue et al. 2010) although our estimate for total arsenic exposure for the most exposed population subgroup (2 year-olds, 0.45 μg/kg/day) was fairly close to the EPA estimate of 0.54 μg/kg/day for ages 1 to 2. Differences in the findings are likely due to differences in the food consumption databases; our study used FDA TDS estimates for consumption of the various food items analyzed in the market basket survey using data derived from the 1994–1996 CSFII while the EPA used results from the 2003–2004 National Health Assessment and Nutritional Evaluation Survey. Both studies relied on analytical results obtained from the TDS although our study considered only data obtained between 2006 and 2008 while the EPA study used data collected from 1991 to 2004.

With respect to estimated exposures to inorganic arsenic, our findings are quite consistent with other studies conducted in the US. We estimated that inorganic arsenic levels ranged from 0.023 to 0.084 μg/kg/day for the general population, depending upon which scenario was used, and from 0.11 to 0.28 μg/kg/day for two year-

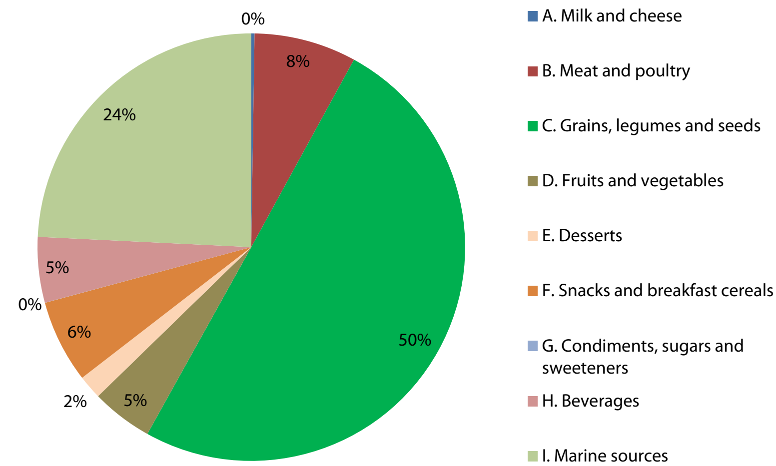

Figure 7 **Core food groups contributing to inorganic arsenic exposure for the general US population (Scenario 5).**

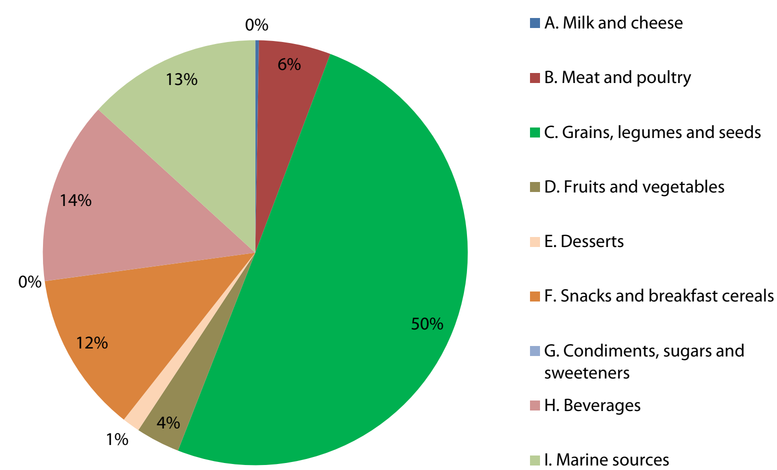

Figure 8 **Core food groups contributing to inorganic arsenic exposure for 2 year-olds (Scenario 5).**

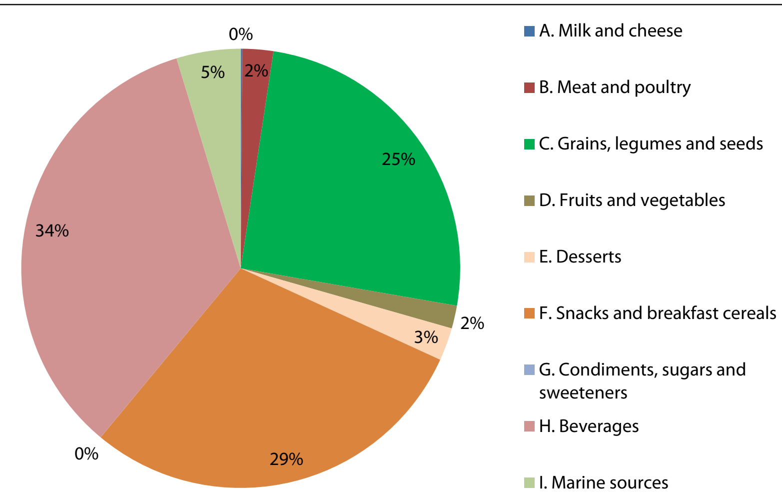

Figure 9 **Core food groups contributing to inorganic arsenic exposure for 6–11 month infants (Scenario 5).**

olds. The EPA's analysis, in which inorganic arsenic was considered to represent approximately 10 percent of the total arsenic, showed exposures ranging from 0.03 to 0.04 μg/kg/day for the general population and 0.08 to 0.23 μg/kg/day for children under five (Xue et al. 2010). The FDA's analysis, in which 10 percent of the total arsenic from marine sources and 100 percent of the total arsenic from terrestrial sources was considered to exist as inorganic arsenic, yielded inorganic arsenic exposures in the range of 0.026 to 0.25 μg/kg/day (Tao and Bolger 1999). All of the inorganic arsenic exposure estimates discussed above are significantly lower than those derived from EFSA, which range from 0.13 to 0.56 μg/kg/day for 2009 and 0.09 to 0.38 μg/kg/day for 2014 (EFSA 2009, 2014).

Conclusions

Using a variety of scenarios to estimate exposure to inorganic arsenic from foods for the general US population as well as for population subgroups, it was determined that inorganic arsenic exposure estimates for all population subgroups under all scenarios were at least slightly below the EPA's established RfD of 0.3 μg/kg/day and below the JEFCA health concern level of 2.0 to 7.0 μg/kg/day. Results compared closely with those from other US studies that used different food consumption databases, food contamination databases, and other methods to extrapolate inorganic arsenic levels from results of total arsenic analyses.

It is clear that accurate assessments of exposure to inorganic arsenic in the food supply require careful consideration of arsenic speciation among foods. Expressing arsenic exposure in terms of total arsenic and comparing such findings with the RfD for inorganic arsenic may lead to exaggerated exposures that imply a much greater risk than is actually present. In this paper, for example, the total arsenic exposure estimates for 2 year-olds (0.45 μg/kg/day) and for 6 year-olds (0.31 μg/kg/day) exceed the EPA's RfD for inorganic arsenic. Applying the speciation assumptions of Scenario 5 (10 percent of marine total arsenic is present as inorganic arsenic and 70 percent of terrestrial total arsenic is present as inorganic arsenic), for 2 year-olds and for 6 year-olds reduces inorganic arsenic exposure to 0.18 and 0.12 μg/kg/day, respectively.

While analysis of food samples for inorganic arsenic is generally much more expensive and difficult to perform than analysis for total arsenic, it may be possible to generate more accurate exposure assessments by focusing inorganic arsenic analyses on the primary food groups contributing to inorganic arsenic exposure. Grains, legumes and seeds, for example, contributed 50 percent of the estimated inorganic arsenic exposure under Scenario 5 for both the total US population and 2 year-olds, and 25 percent of the estimated inorganic exposure for 6–11 month infants. Focusing analytical approaches to detect inorganic arsenic from this food group could serve to reduce uncertainty in the exposure assessment by precluding the need to make assumptions concerning how much of the total arsenic from these foods is present in the inorganic form.

Abbreviations

EPA: U.S. Environment Protection Agency; IARC: International Agency for Research on Cancer; FDA: U.S Food and Drug Administration; RfD: Reference Dose; EFSA: European Food Safety Authority; JECFA: Joint FAO/WHO Expert Committee on Food Additives; PTWI: Provisional Tolerable Weekly Intake; TDS: Total Diet Study; CSFII: Continuing Survey of Food Intakes for Individuals; ppbm: Parts per billion.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

EJ was responsible for data collection and exposure assessment and assisted with preparation of the manuscript. CW assisted with the design of the analysis and with preparation of the manuscript. Both authors read and approved the final manuscript.

Acknowledgements

The authors thank to the Chilean Government and the National Commission for Scientific and Technological Research (CONICYT) for supporting this work through the Becas Chile Scholarships.

Received: 9 April 2014 Accepted: 25 May 2014
Published online: 29 July 2014

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doi:10.1186/s40550-014-0003-x
Cite this article as: Jara and Winter: **Dietary exposure to total and inorganic arsenic in the United States, 2006–2008.** *International Journal of Food Contamination* 2014 **1**:3.